



Assessing possible consequences of ocean liming on ocean pH, atmospheric CO₂ concentration and associated costs



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ABSTRACT

More than 380 Pg of carbon have been released by human activities from fossil fuel combustion and cement manufacturing since the beginning of the Industrial Revolution. This carbon release has caused atmospheric CO₂ levels to increase by ~100 ppmv compared to the highest interglacial value of the last ~800,000 years. Net uptake of carbon by the oceans has caused the surface ocean pH to drop by ~0.1 pH units. Evaluating carbon sequestration options is now timely. We use the carbon-cycle model LOSCAR to evaluate the effects of artificially enhancing ocean alkalinity from year 2020 to year 2400 on ocean pH and atmospheric pCO₂. Ten different carbon emission scenarios with scaled alkalinity input are simulated. Results show that for ocean pH to be maintained above 8.0, on the order of $2-10 \times 10^{14}$ moles of alkalinity/year for C emissions of 1500–5000 Pg C, respectively, is required. Atmospheric pCO₂ remains high (500–600 ppmv) in nine of the carbon emission scenarios. Ocean alkalization, if ever implemented at such a large scale, could allow ocean pH to be stabilized but pCO₂ would not return to pre-industrial levels. Cost estimates of the ocean alkalization operation, using quicklime (CaO) was estimated to range from 0.5 to 2.8 \$US trillion/year, depending on the target pH selected to avoid damage to marine organisms and ecosystems. For comparison, this cost is ~0.8% and ~4.6% of the 2011 global Gross Domestic Product for total anthropogenic carbon emissions of 1500 and 5000 Pg C, respectively. The cost of 1 ton of CO₂ sequestered ranges from 103 to 144 \$US for total emissions of 1500–5000 Pg C, respectively. Producing large amounts of quicklime (without carbon capture and storage) would cause substantial additional CO₂ release. This reduces the fossil fuel emissions allowed to maintain pH above 8.0 by a factor of 1.3–2.5 (for total emissions of 1000 and 5000 Pg C, respectively).

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1. Introduction

According to the Intergovernmental Panel for Climate Change, business-as-usual carbon emissions might lead to atmospheric CO₂ levels of ~700 ppmv and a 1.7–4.4 °C global temperature rise by year 2100 (IPCC, 2007). This differs from the Copenhagen Accord in which countries have pledged to limit the CO₂ concentration to below 450 ppmv, considered likely to cause a rise of global temperature by ~2 °C (UNFCCC, 2009). The delay in the implementation of a new global treaty on carbon emissions (“The Durban Platform”) which will not be in effect until 2020 (Tollefson, 2012) increases the likelihood that CO₂ levels will exceed 450 ppmv, and highlights the urgent need for serious evaluation of techniques to sequester carbon.

The current rate of carbon emissions to the atmosphere resulting from the burning of fossil fuels, cement production and land-use change is likely unprecedented during the last 56 million years

(Zeebe et al., 2009). Present-day pCO₂ is ~395 ppmv (Tans and Keeling, 2012), a 40% increase compared to the ~280 ppmv value before the Industrial Revolution. In 2011, about 9.5 ± 0.5 Pg C ($1 \text{ Pg C} = 10^{15} \text{ g C}$) were emitted to the atmosphere (Peters et al., 2013) of which about one-third was absorbed into the surface of the open oceans (Sabine et al., 2004, 2011). For over a century, theoretical studies have pointed out that CO₂ affects the radiative forcing of the atmosphere which results in increasing temperatures at Earth's surface if concentrations rise (Arrhenius, 1896). Global surface temperature has already increased by 0.74 °C when comparing the time periods 1850–1899 and 2005–2011, with a significant contribution from human activities (IPCC, 2007).

The “other CO₂ problem”: ocean acidification – results from the storage of 150 Pg of anthropogenic carbon in the oceans (Sabine et al., 2011) and has already led to a global decline of surface ocean pH by ~0.1 units, a 16% decline in the carbonate ion concentration and decrease in the saturation state of carbonate minerals (Doney et al., 2009; Dore et al., 2009). As these parameters are the controlling variables in the calcification process of corals and other calcifying organisms (Rodolfo-Metalpa et al., 2011), the stability of carbonate minerals will be reduced with potentially negative

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effects on marine life. Immediate effects from surface waters undersaturated with respect to aragonite may already occur within the next decade in the Arctic Ocean (Orr et al., 2005) and by 2050 in the Southern Ocean, if not already seasonally important (Yamamoto-Kawai et al., 2009; Byrne et al., 2010; Mathis et al., 2011).

As there is no indication that CO₂ emissions will slow down anytime soon, evaluating the full range of carbon sequestration options is increasingly urgent. Various methods to capture and/or sequester CO₂ have been proposed. They include capture of CO₂ from the air on solvents (Lackner, 2003; Lackner et al., 2012; House et al., 2011), direct disposal into underground geological reservoirs (House et al., 2006; Kelemen et al., 2011), deep sea disposal (Ridgwell et al., 2011) and more options (IPCC, 2005; Shepherd et al., 2009). Increasing ocean alkalinity (addition of soluble minerals to the surface oceans) or reacting CO₂-rich flue gas from fossil-fuel power plants with seawater and alkaline rocks has also been suggested (Kheshgi, 1995; Rau and Caldeira, 1999; Rau et al., 2007; Köhler et al., 2010, 2013; Renforth and Kruger, 2013). In theory, increasing the ocean's alkalinity inventory should allow more anthropogenic carbon to be absorbed by the ocean while at the same time buffering the acidifying effect of CO₂ on ocean chemistry. Enhancing ocean CO₂ uptake would also reduce the rise in global temperature.

The objective of the present study is to investigate open ocean alkalization as a mean to sequester anthropogenic carbon and mitigate the decrease in surface ocean pH. The results of this study quantify the required increase in ocean alkalinity in order to decrease atmospheric pCO₂ and to maintain ocean pH at a specified value. To determine these two parameters, we use the carbon cycle model LOSCAR (Zeebe, 2012) (Section 2.1).

We also estimate the approximate amount of quicklime (CaO) and the cost of one ton of CO₂ sequestered using quicklime as the source of alkalinity (Sections 2.2 and 2.3). Finally, we calculate the allowed carbon emissions to maintain a specific pH level of 8.0, accounting for additional CO₂ from increased quicklime production (Section 2.4).

2. Methods

2.1. Determining the amount of alkalinity

We employ the Long-term Ocean-atmosphere-Sediment Carbon cycle Reservoir Model (LOSCAR) (Zeebe, 2012) to investigate the effects of increasing ocean alkalinity ("ocean alkalization") on ocean pH and atmospheric pCO₂. The model has been previously applied to the problem of ocean acidification and climate change in the future and during the Paleocene-Eocene Thermal Maximum (Zeebe et al., 2008, 2009; Komar and Zeebe, 2011). The model is divided into ten oceanic boxes. It includes surface, intermediate and deep water of the Atlantic, Indian, Pacific Oceans, and a high-latitude box. It also includes a sediment module and the carbonate and silicate weathering feedbacks. The full model structure is described in Zeebe (2012).

We manipulated the alkalinity in the upper 100 m of the water column in the Atlantic, Indian and Pacific Oceans. In each simulation, alkalinity was added proportionally to carbon emissions. We chose to use ten carbon emissions scenarios, which have previously shown their impact on surface ocean pH (Zeebe et al., 2008). This previous study has also shown how the decline in ocean pH is sensitive to the carbon release time (Zeebe et al., 2008). In the present study, we use a carbon release time of 500 years for all emissions scenarios. The scenarios are based on (1) historic emissions data (Marland et al., 2008) and (2) future emissions are based on a Gaussian function (Zeebe et al., 2008) that match total emissions and release time of carbon. The start year of the carbon emissions is year 1750.

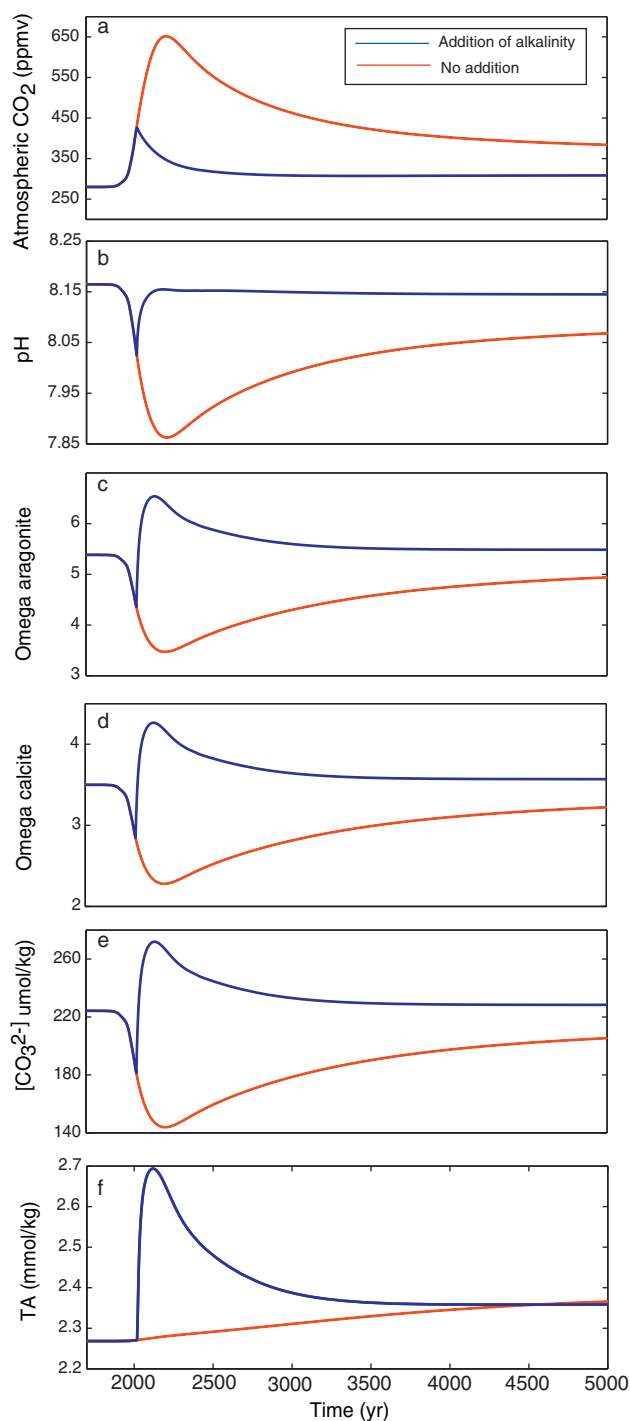


Fig. 1. Comparison of the variations of various carbonate chemistry parameters during addition of alkalinity (blue lines, scaled to carbon emissions and for 380 years) and without alkalinity addition (red lines) in carbon emissions scenarios of 1500 Pg C over 500 years. Letters from a to f label plots of atmospheric CO₂ (ppmv), pH, surface omega aragonite, surface omega calcite, carbonate ion concentration ($\mu\text{mol kg}^{-1}$) and total alkalinity (mmol kg^{-1}), respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The ten carbon emission scenarios are represented by total emissions of 600, 1000, 1500, 2000, 2500, 3000, 3500, 4000, 4500 and 5000 Pg C over the course of 500 years (Zeebe et al., 2008; Uchikawa and Zeebe, 2008). The lower C emissions scenarios imply constraints on greenhouse gas emissions. We added the alkalinity for 380 years from year 2020 until year 2400 and ran the model until year 5000 (Figs. 1 and 2).

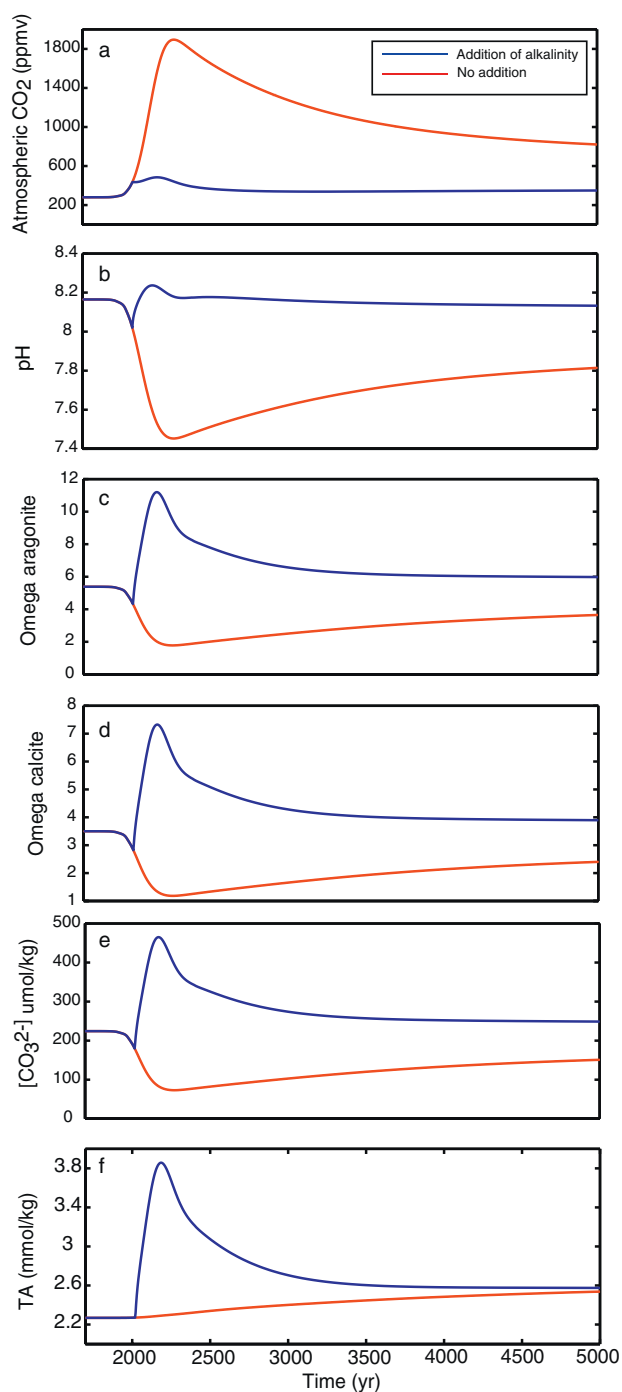


Fig. 2. Comparison of the variations of various carbonate chemistry parameters during addition of alkalinity (blue lines, scaled to carbon emissions and for 380 years) and without alkalinity addition (red lines) in carbon emissions scenarios of 5000 Pg C over 500 years. This figure shows that high alkalinity inputs cause ocean pH (b) to become more alkaline than pre-industrial values. Resulting pCO₂ and ocean pH from this example were not used in the plot of Fig. 3. Letters from a to f label plots of atmospheric CO₂ (ppmv), pH, omega aragonite, omega calcite, carbonate ion concentration ($\mu\text{mol kg}^{-1}$) and total alkalinity (mmol kg^{-1}), respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In the LOSCAR model, the alkalinity added was increased incrementally. The incremental factor was first determined by trial and error (the “X” factor). Then a range of values was determined so that surface pH is restored to pre-industrial values (~ 8.2) in each simulation. This “X” factor is included in the model and scales carbon emissions to alkalinity input, ranging from 0 to $3.5 \cdot 10^{-3} \text{ mol/m}^3/\text{Pg}$

C. The “X” factor was then multiplied by the rate of carbon emission per year and a factor “ $12/10^{15}$ ” to convert the units into Pg C/year.

The average total alkalinity input (in mol/year) was calculated by multiplying carbon emissions times the “X” factor times the total volume of input, and then dividing by the number of years of alkalinity input (380 years). The total volume was calculated as the combined volume of the upper 100 m of the Atlantic, Indian and Pacific Oceans. It is calculated by multiplying the combined surface area of the three oceans ($3.14 \times 10^{14} \text{ m}^2$) with the total depth over which alkalinity is released in the model ($=100 \text{ m}$). The total volume to which alkalinity was added is $3.14 \times 10^{16} \text{ m}^3$.

Two examples of the total amount of alkalinity calculated above are given in the following. In these examples, model simulations show that surface seawater pH is maintained above 8.0. First, in a carbon emission scenario of 1500 Pg C/500 years, 1054 Pg will be emitted from year 2020 to year 2400; if the “X” factor is $2.0 \times 10^{-3} \text{ mol/m}^3/\text{Pg C}$, the calculations yield an average total alkalinity input of $1.74 \times 10^{14} \text{ mol/year}$. Second, in the case that anthropogenic emissions result in total carbon emissions of 5000 Pg/500 years (4559 Pg C will be emitted from year 2020 to year 2400); and at a “X” factor of $2.5 \times 10^{-3} \text{ mol/m}^3/\text{Pg C}$, this results in an average total alkalinity input of $9.49 \times 10^{14} \text{ mol/year}$. To each increment of the “X” factor and for each carbon emission scenario, the minimum pH of the surface water of the Atlantic, Indian and Pacific oceans was recorded from the simulation output and averaged. The maximum atmospheric pCO₂ was also recorded at each “X” factor increment and for each carbon emission scenario. The simulations provide a total of 80 minimum pH- and maximum pCO₂ values.

2.2. Determining the amount of quicklime and cost of alkalization using quicklime

How does this alkalinity input translate into the amount of specific alkaline minerals? Many alkaline minerals are present at the surface of the Earth. A non-exhaustive list is given in (Lackner, 2002). We decided to focus our attention on the use of quicklime (CaO) because the deposits of limestone, from which quicklime is derived, cover most continents in large quantities (Morse and Mackenzie, 1990) and because it is easily soluble in seawater. Quicklime is formed by heating of limestone at 1000°C . Anthropogenic CO₂ would be sequestered as dissolved bicarbonate ions after reaction with quicklime and seawater, which may be represented by the reaction:



However, note that the actual overall stoichiometry of this process is more complex (calculated in our model simulations). The total amount of quicklime (QL, units in tons/year) is calculated by dividing total alkalinity values by 2 and multiplying by 56 (molar mass of quicklime) and then divide by 10^6 to convert grams to tons. The 2 mole equivalent increase in total alkalinity results from the dissolution of one mole of quicklime as shown by reaction (1) where calcium ions increase by two-mole equivalent (cf. Zeebe and Wolf-Gladrow, 2001).

In contrast, invasion of anthropogenic CO₂ into the surface ocean without the addition of quicklime:



keeps alkalinity constant but raises seawater acidity ($[\text{H}^+]$)—in other words, lowers seawater pH. Comparison of reactions (1) and (2) shows that H⁺ is neutralized by adding CaO, which increases alkalinity and stabilizes pH values.

In the two examples discussed in the paragraph above for total emissions of 1500 and 5000 Pg C, the alkalinity input is translated

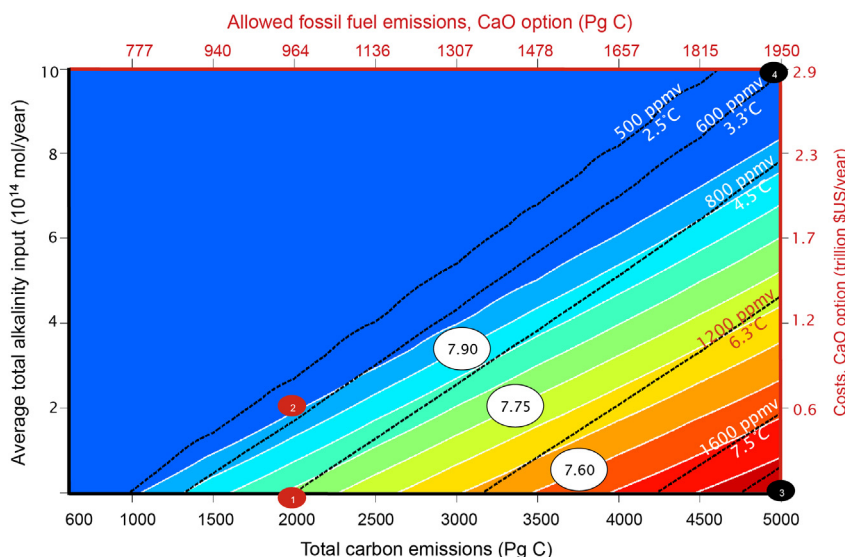


Fig. 3. Maximum concentration of atmospheric CO₂ (ppmv) and subsequent increase in global temperature (°C) (black lines) (using a climate sensitivity of 3 °C per doubling of CO₂ (IPCC, 2007) and minimum seawater pH (white lines, average values of the surface of the Indian, Pacific and Atlantic oceans), as a function of total carbon emissions (bottom x-axis) (1 Pg C = 10¹⁵ g C) and required average alkalinity input (mol/year) (left y-axis) in order to maintain seawater pH above a certain threshold. The right y-axis represents the associated costs using CaO for alkalinity supply. These costs are valid for year 2011 and do not take into account the years of inflation for 380 years. The top x-axis represents the allowed fossil fuel emissions if CaO is used in order to maintain seawater pH above 8.0. Unabated total carbon emissions is represented at total alkalinity = 0 mol/year. Simulations were performed using the LOSCAR model (Zeebe, 2012). Numbers 1–4 refer to different scenarios (see Text for an explanation).

into a quicklime input of 4.88×10^9 tons/year (1500 Pg C) and 2.66×10^{10} tons/year (5000 Pg C), respectively.

In 2011, the worldwide production of quicklime was 330×10^6 tons (USGS, 2012). Based on the previous calculations, the current production of quicklime would need to be increased by a factor of 18 and 90 for emissions scenarios of 1500 and 5000 Pg C, respectively. Note that our cost estimates are only valid for year 2011. Moreover, we are underestimating the cost over longer term because they do not take into account the inflation rate, which is uncertain in the long run and depends on a variety of socio-economic and technological changes, which are difficult to predict at present-day.

In 2011, the average price of one ton of quicklime produced from limestone and ready to be shipped was approximately 100 US\$/ton (USGS, 2012). Additionally, the cost of transport, shipment and discharge into the ocean can be estimated by comparing to the average price in year 2011 of the use of a very large crude carrier (VLCC) (~200,000 tons dead weight tonnage) for which the cost averages 56,000 US\$/day (UNCTAD, 2012). Estimating that it will take arbitrarily two weeks to discharge the quicklime, this averages 784,000\$ per VLCC, which gives ~4 US\$/ton of quicklime for the shipment cost. This is valid if the extraction of limestone and subsequent transformation into quicklime is carried out in proximity to ports. Hence, we recognize that this cost is probably a minimum estimate because implementation charges for the operation are not taken into account. We estimate the cost of releasing one ton of quicklime in the ocean at ~\$104US (2011 price standards). For example, in the extreme 5000 Pg C total emission scenario, the average cost is estimated as 2.66×10^{10} tons/year * \$104 = \$2.8 trillion US/year. This large quicklime input allows seawater pH to be maintained above 8.0 but only decreases atmospheric pCO₂ from ~1900 ppmv to 630 ppmv (Fig. 3). For total emissions of 1500 Pg C, 4.88×10^9 tons/year of quicklime is required and the cost is estimated at \$0.5 trillion US/year to lower atmospheric pCO₂ from ~650 ppmv to ~470 ppmv.

To illustrate the level of increased shipping capacity required for the quicklime operation, we can estimate how much the actual deadweight tonnage would have to be increased to supply the alkalinity/CaO. As of 2012, the Review of Maritime Transport

reported that the sum of deadweight tonnage of oil tankers, general cargo ships and bulk carriers sums up to 1.24×10^9 tons (UNCTAD, 2012). As we have shown above, in a 1500 and 5000 Pg C scenario, the amount of required quicklime is 4.88×10^9 and 2.66×10^{10} tons/year, respectively. This means that all existing carriers would have to be called at a port between 4 and 21 times a year to receive and distribute its cargo of quicklime, in a 1500–5000 Pg C scenario, respectively.

2.3. Determining the cost of one ton of CO₂ sequestered from alkalization/CaO

The cost of one ton of CO₂ sequestered from ocean alkalization/CaO can be directly compared to other mitigations options (Table 1). To estimate this cost, we use the following equation:

$$\text{Cost (\$US)}(1 \text{ ton CO}_2) = \frac{K * (56/44)}{\text{RCO}_2} \quad (3)$$

where, “K” is the total cost of discharging one ton of quicklime into the ocean, which was estimated at about \$104US per ton of quicklime (see above); RCO₂ is the ratio of mole of CO₂ sequestered in

Table 1
Various carbon capture and storage options and comparison of the cost of 1 ton of CO₂ sequestered.

Method	\$US	Reference
Seawater/limestone CO ₂ capture	38	Rau (2011)
Capture at power plants	75	MITEI (2009)
Increase alkalinity by electrochemical splitting of CaCO ₃	100	Rau (2008)
Olivine dispersal (rivers and ocean)	50	Köhler et al. (2010)
Air capture	100–1000	Keith (2009) and House et al. (2011)
Geological storage	0.5–8	IPCC (2007)
Ocean alkalization	103 (5000 Pg C)–144 (1500 Pg C)	This study

the ocean per mol of CaO added (see Supporting Information for detailed calculations); the factor “56/44” converts from mol of CaO to grams of CO₂. The calculated RCO₂ varies from 0.81 to 1.28 for total emissions of 1000 to 5000 Pg C, respectively. Using the equations described in the supporting information, we calculate a range of cost per ton of CO₂ between ~103 US\$ for 5000 Pg C and ~144 US\$ for 1500 Pg C emissions scenarios.

2.4. Determining the quicklime penalties

Without carbon capture and storage facilities, the transformation of limestone into quicklime releases significant amounts of CO₂ to the atmosphere. To obtain minimum seawater pH and maximum atmospheric pCO₂ from our model simulations, we have used carbon emissions scenarios (from 600 to 5000 Pg C) that do not take into account the emissions related to this quicklime CO₂ input. It is therefore essential to estimate the amount of carbon emitted from the mining and transformation operations and from this number estimate the amount of allowed carbon emissions if the quicklime option is chosen (Fig. 3, see top x-axis). We have calculated the allowed carbon emissions that will maintain seawater pH above 8.0. This pH value of 8.0 was chosen because of the U.S. Environmental Protection Agency (EPA)'s statement in 1976 (still in effect in 2013) on the effects of pH variations on surface marine organisms (EPA, 1976). Before the Industrial Revolution, surface-ocean pH was globally ~8.2 and has decreased by 0.1 pH units since the Industrial Revolution. The EPA stated that no more than a 0.2 pH units change outside of their normally occurring range should be tolerable by planktic marine organisms. A pH value of 8.0 units illustrates this EPA statement as quicklime is added to the oceans to stabilize pH at 8.0.

First, the carbon emissions related to the CaO option are calculated knowing the total alkalinity values, the number of moles of CO₂ (1.41) emitted during the quicklime operations (transformation and combustion of C from transport, mining operations, etc.) (Kheshgi, 1995). Secondly, the allowed carbon emissions are then calculated by subtracting the carbon emissions (CaO option) from the total carbon emissions (Fig. 3, top x-axis).

3. Results and discussion

The amount of alkalinity to be added to the world ocean required to significantly offset CO₂ emissions would be substantial. For example, without alkalization, total emissions of 2000 Pg C over the course of 500 years would cause seawater pH to drop to ~7.78 and atmospheric pCO₂ to rise to 800 ppmv (Zeebe et al., 2008) (circle 1, Fig. 3). For this carbon emission scenario, model simulations show that the average total alkalinity input would have to be $\sim 2 \times 10^{14}$ mol/year (Fig. 3) in order to maintain pH at 8.0 and limit the pCO₂ to increase to ~574 ppmv (circle 2, Fig. 3). In another scenario in which emissions remain unabated until year 2400 with total carbon input of 5000 Pg C, model simulations show that surface ocean pH would decline to pH 7.45 and pCO₂ to increase to ~1900 ppmv without alkalinity addition (circle 3, Fig. 3). In order to maintain pH above 8.0, an average of $\sim 10 \times 10^{14}$ moles of alkalinity per year would need to be added to the world oceans over 380 years. However, maximum atmospheric CO₂ levels would decrease from ~1900 ppmv (unabated, circle 3, Fig. 3) to ~600 ppmv (circle 4, Fig. 3). The reduction in atmospheric CO₂ resulting from the alkalinity input is substantial but not sufficient to limit the rise in global temperature to less than 3.3 °C if applying a temperature sensitivity of 3 °C per doubling of CO₂.

The use of quicklime produced without carbon capture and storage will drastically reduce the allowed carbon emissions from fossil fuel burning because emissions resulting from the

transformation of limestone into quicklime are considerable as they release ~1.4 moles of CO₂ per mole of limestone (Kheshgi, 1995). This figure includes ~0.4 moles resulting from the burning of coal during the mining operations of limestone plus 1 mole as a result of the transformation of limestone into quicklime (Kheshgi, 1995). When the calculated carbon emissions resulting from the CaO production are subtracted from total carbon emissions (Fig. 3, bottom x-axis) the amount of allowed fossil fuel emissions decreases significantly (Fig. 3, top x-axis). As examples, for the 1500 and 5000 Pg C scenarios, only 940 and 1950 Pg C are allowed to be emitted, respectively in order to keep seawater pH above 8.0. Note that from the beginning of the Industrial Revolution from year 1750 to the present, cumulative carbon emissions are already ~380 Pg C. Low-carbon technologies may increase the amount of allowed C emissions if the route of quicklime is ever taken. As shown at a small scale by Licht et al. (2012), solar energy may be used to drive quicklime production without carbon dioxide emission.

Mining, transport and discharge of quicklime at the scale required to offset CO₂ emissions would also require substantial financial efforts. The total cost that encompasses transportation and oceanic dispersal is estimated at 104 US\$/ton of quicklime at 2011 price standards. Hence, at total emissions of 1500 and 5000 Pg C, the alkalization operation would cost on average ~\$0.5 and ~\$2.8 trillion US per year, respectively in order to maintain surface ocean pH above 8.0. These numbers are roughly 0.7% and 4.0% of the global Gross Domestic Product for Fiscal Year 2011 (\$69.97 trillion).

Other options to increase ocean alkalinity have been proposed such as removing electrochemically HCl from the ocean and subsequent neutralization of the acid with silicate rocks (House et al., 2007) or dispersing fine powdered olivine in tropical areas and in the oceans (Köhler et al., 2010, 2013). However, these options are also energy-intensive and a number of biogeochemical issues have been flagged (Shepherd et al., 2009). According to our estimates, the cost to sequester one ton of CO₂ using the CaO option is ~\$144 and ~\$103 US for total carbon emissions of 1500 and 5000 Pg C, respectively. However, as explained above, the allowed fossil fuel emissions are much less for the CaO option. The cost of ocean alkalization is comparable to the range of other mitigation options (Table 1) and in the lower bound of CO₂ air capture ~\$100–1000 US/ton CO₂ (Keith, 2009; House et al., 2011).

4. Conclusions

We have examined the potential of ocean alkalization – adding alkalinity to the ocean continuously over 380 years – as a mitigation option for reducing the concentration of atmospheric CO₂ levels and to maintain ocean pH above a certain threshold, while still emitting carbon according to ten different scenarios. The amount of alkalinity required to stabilize ocean surface pH above 8.0 is large and ranges from 2.0×10^{14} to 10×10^{14} mol/year in a 1500 to 5000 Pg C emission scenario, respectively.

We also have estimated to first order the associated costs of alkalization using quicklime as the source of alkalinity and calculated it to range from one to several trillion US\$ per year, depending on which ocean pH and pCO₂ targets are chosen. The route of using quicklime as the source of alkalinity reduces considerably the allowed fossil fuel emissions if no carbon capture and storage exist at the processing facilities. Finally, there are many uncertainties associated with alkalization, including the financing, the response of marine organisms, and the logistic feasibility of such an endeavor.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.ijggc.2013.05.005>.

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