CO₂ Mitigation via Capture and Chemical Conversion in Seawater

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A lab-scale seawater/mineral carbonate gas scrubber was found to remove up to 97% of CO_2 in a simulated flue gas stream at ambient temperature and pressure, with a large fraction of this carbon ultimately converted to dissolved calcium bicarbonate. After full equilibration with air, up to 85% of the captured carbon was retained in solution, that is, it did not degas or precipitate. Thus, above-ground CO_2 hydration and mineral carbonate scrubbing may provide a relatively simple point-source CO_2 capture and storage scheme at coastal locations. Such low-tech CO_2 mitigation could be especially relevant for retrofitting to existing power plants and for deployment in the developing world, the primary source of future CO_2 emissions. Addition of the resulting alkaline solution to the ocean may benefit marine ecosystems that are currently threatened by acidification, while also allowing the utilization of the vast potential of the sea to safely sequester anthropogenic carbon. This approach in essence hastens Nature's own very effective but slow CO_2 mitigation process; carbonate mineral weathering is a major consumer of excess atmospheric CO_2 and ocean acidity on geologic times scales.

1. Introduction

Because of impacts to climate (*1*) and ocean biogeochemistry (*2*), it is necessary to stabilize if not reduce atmospheric CO_2 concentrations. Current efforts are failing at this task (*3*), and an expanded search for cost-effective CO_2 mitigation options and implementation policies is needed. Avoiding CO_2 emissions by transitioning to non-fossil energy use is an obvious solution, but the pace of this transition is not keeping up with energy demand; human consumption of coal, natural gas, and oil is projected to increase rather than decrease in coming decades, especially in the developing world (*4*). This dictates that effective methods be found for reducing the carbon footprint of these fuels (including increasing the efficiency of their use) until alternative energy sources can be adequately exploited. As a first step, considerable effort has focused on actively removing CO_2 from point sources such as electric power, fertilizer, and cement plants representing ~50% of total fossil fuel emissions (*4*). While various chemically or biologically mediated schemes for such CO_2 mitigation have been proposed, the majority of the research to date has focused on carbon capture and storage, CCS —the capture, purification, and underground storage of molecular CO_2 (*5, 6*). However, deployment of CCS may

ultimately be limited by cost (*7, 8*), capacity (*9*), and public acceptance (*10, 11*). It is therefore unclear whether currently favored technologies alone will be of sufficient scale and timeliness to significantly contribute to the very large and rapid CO_2 emissions reduction required to stabilize atmospheric CO_2 concentrations (*12*).

Building on previous proposals to employ wet limestone scrubbing as a point source CO_2 mitigation tool (*13–17*), a series of laboratory experiments was conducted to determine the effectiveness of this approach. This method uses the following chemical reaction to effect CO_2 capture and conversion to calcium bicarbonate in solution:

$$CO_{2(g)} + H_2O_{(l)} + CaCO_{3(s)} \longleftrightarrow Ca(HCO_3)_{2(aq)} + (CO_{2(aq)} + CO_3^{2-}) \quad (1)$$
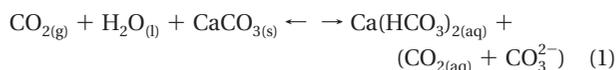
Note that while $Ca(HCO_3)_{2(aq)}$ will be the dominant compound formed, equilibrium reactions require that $CO_{2(aq)}$ and CO_3^{2-} also be present, though in much smaller quantities within typical pH ranges. The reaction can be spontaneously driven to the right (CO_2 capture and conversion achieved) under elevated CO_2 concentrations, as found in flue gas and other waste gas emissions. Using seawater for the reaction is less effective in dissolving $CaCO_3$ per unit of added CO_2 because seawater already contains significant quantities of HCO_3^- and CO_3^{2-} that act to buffer pH and $CaCO_3$ saturation state depression that drives reaction 1. Nevertheless, calcium carbonate undersaturation and $CaCO_3$ dissolution can be easily achieved when seawater is equilibrated with CO_2 in concentrations typical of flue gas, 5–15% by gas volume. Such a process in effect mimics and speeds up Nature's own use of reaction 1; carbonate mineral (e.g., limestone) weathering is a major, natural consumer of excess atmospheric CO_2 on geologic time scales (*18*). An electrochemically driven version of reaction 1 has also been described (*19*).

However, since reaction 1 is reversible, one could expect that any CO_2 mitigation initially attained would be lost when the resulting solution was re-equilibrated with low-CO_2 ambient air, degassing the excess CO_2 from solution, elevating pH, and saturating and reprecipitating $CaCO_3$. While this scenario can be avoided by storing the produced solution away from the atmosphere (subterranean injection or subsurface ocean dilution and storage ref *14, 20*), chemical precipitation of $CaCO_{3(s)}$ from seawater is in fact difficult to achieve, requiring calcium carbonate ion concentrations in excess of 18× saturation (*21*). Indeed, the surface ocean is already 4–6× supersaturated in $CaCO_{3(aq)}$ because nucleation and precipitation is chemically hindered by the presence of Mg^{2+}, PO_4^{4-}, and other ions (*22*).

On the other hand, $CaCO_{3(s)}$ can be biologically precipitated during carbonate shell formation, providing an additional mechanism to reverse reaction 1. Relative to the available pool of dissolved inorganic carbon in seawater, however, biocalcification is an inefficient, slow process. For example, with ~6 × 10¹¹ kgs of Ca annually added to the ocean via hydrologic and hydrothermal inputs (*23*), and 6 × 10¹⁷ kgs of dissolved Ca residing in the ocean (*24*), a mean seawater Ca (and by inference, $CaCO_{3(aq)}$) residence time of 1 million years is implied if steady state is assumed. Shorter residence times can be anticipated under non-steady state conditions, for example during the significant carbonate dissolution event that followed Paleocene-Eocene ocean acidification (*25*). Here as much as 100 kyrs were required to reprecipitate the excess dissolved $CaCO_3$ and $Ca(HCO_3)_2$ from seawater. Considering that this ocean alkalinity transient was probably vastly larger (*26*) than that conceivable by any

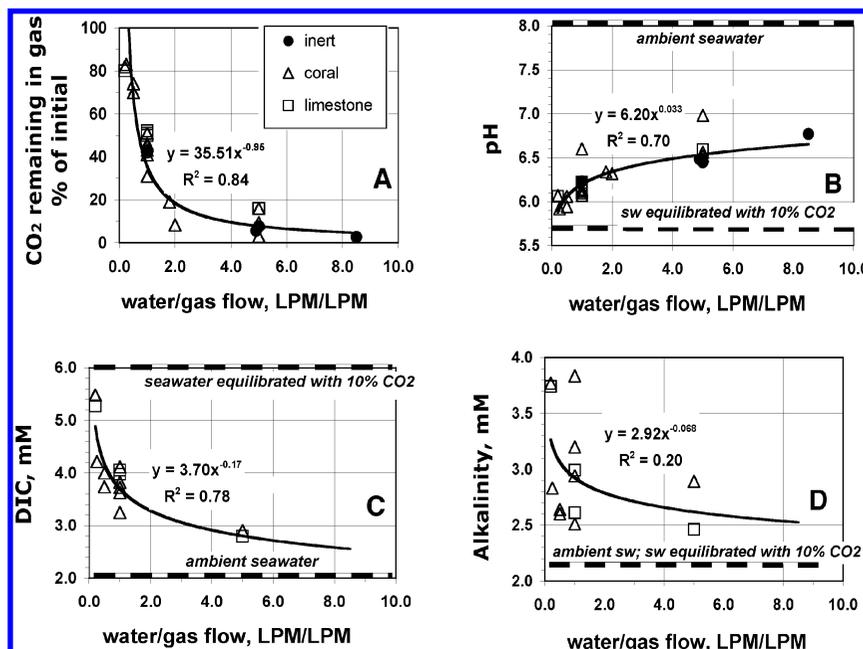* Corresponding author phone: 1 925 423 7990; fax 1 925 422 7438; e-mail: rau4@llnl.gov.

FIGURE 1. A, Percent of initial CO_2 remaining in a 1v/9v CO_2/air gas stream after passing through a seawater reactor with the seawater/gas flow ratio as indicated in liters per minute, LPM. Reactor aggregate packing as indicated. **B,** Corresponding reactor effluent pH versus seawater/gas flow rate. **C,** Corresponding total dissolve inorganic carbon concentration, [DIC], in reactor effluent versus seawater/gas flow rate. **D,** Corresponding alkalinity in reactor effluent versus seawater/gas flow rate. Best fit power equation to data shown on each graph, together with r^2. Ambient seawater values and predicted values (*34*) at equilibrium with a 1v/9v CO_2/air gas stream are indicated by horizontal dashed lines.

human, proactive use of reaction 1, it appears safe to assume that point-source CO_2 mitigation by wet limestone scrubbing could allow carbon storage in the ocean for many tens of thousands of years if not substantially longer. CO_2 emissions offsets are currently being credited for carbon sequestration in terrestrial biomass whose residence time may be <100 yrs (*6*).

2. Experimental Section

To test the preceding ideas, A hollow, cylindrical Plexiglas carbonate reactor (Precision Marine model PM3311; 10.2 cm ID, 45.7 cm tall) was modified to allow seawater and gas streams to enter, longitudinally flow through, and exit the reactor chamber that had been filled with 2.2–2.5 kgs of mineral carbonate aggregate of a specific size class. Size classes ranged from a minimum dimension of <2.8 mm to 34 mm. Two types of mineral carbonate aggregate were tested, one being aragonitic coral fragments and the other calcitic limestone from a local quarry owned by Cemex, Inc. in Davenport, CA.

The effect of both concurrent and countercurrent gas/water flow regimes was explored, as were gas and water flow rates (<1 LPM), carbonate particle size and source, and gas/water residence time. In all cases the upstream and downstream gas % CO_2 concentration was monitored using a Bacharach model 2820 gas analyzer with upstream water trap (precision = ±0.1%). The gas source was a tank of 10.0% (by volume) CO_2 in air as supplied by Praxair, Inc. The upstream and downstream water pH and temperature were monitored using a daily calibrated IQ Scientific model 170 meter with pH and temperature probe (±0.01 units and ±0.1 °C, respectively), and water samples were routinely taken and frozen for later analysis of total dissolved inorganic carbon (DIC) and alkalinity. The DIC was analyzed using a UIC, Inc. CM5012 carbon analyzer with a CM5130 acidification module. Precision of replicate analyses was ±0.01 mM. Alkalinity measurements were performed on an Orion 950 FASTQC Titrator system using a Thermo/Orion 9206BN pH electrode, precision = ±0.02 mM. The instrument was

calibrated using a seawater standard (Batch 94) from Prof. A. Dickson's laboratory at Scripps Institution of Oceanography, La Jolla, CA. The relative proportion of alkalinity increase to DIC increase provided an approximate measure of the fraction of added CO_2 that was converted to calcium bicarbonate; HCO_3^- ions are the primary contributor to seawater alkalinity. As a measure of permanent storage of the absorbed carbon, the DIC and alkalinity were also measured on selected reactor solution samples (<400 mL) that were fully equilibrated with air via vigorous bubbling of the solution samples using an air pump until pH stabilized, usually 0.5–1 h.

3. Results and Discussion

In an initial configuration, the reactor effectively removed up to 97% of the CO_2 in the incoming gas stream, the percentage being inversely proportional to the water/gas flow rate ratio (Figure 1a). This relationship was relatively insensitive to variation in the carbonate particle types (coral or limestone), particle size, or direction of water and gas flow. The total dissolved inorganic carbon concentration (DIC) in the downstream water increased by up to 2.6× over ambient seawater in these initial experiments (Figure 1c).

However, it was clear from the alkalinity measurements that a substantial portion of the carbon absorbed into the water remained as molecular or hydrated CO_2 (carbonic acid); much of the added carbon had not reacted with the carbonate to produce $Ca(HCO_3)_{2(aq)}$ via reaction 1 (Figure 1d, 2a). This was further demonstrated when inert particle aggregate of similar dimensions to the carbonate mineral proved to be about as effective as the former particles in absorbing CO_2 (Figure 1a). Lack of significant conversion of CO_2 to $Ca(HCO_3)_{2(aq)}$ would mean reduced potential for long-term solution storage of the captured CO_2 if the solution were exposed to air, allowing for CO_2 escape from solution. Nevertheless, simple seawater scrubbing of flue gas in the absence of carbonate mineral, followed by subsurface ocean storage of the resulting solution has been previously con-
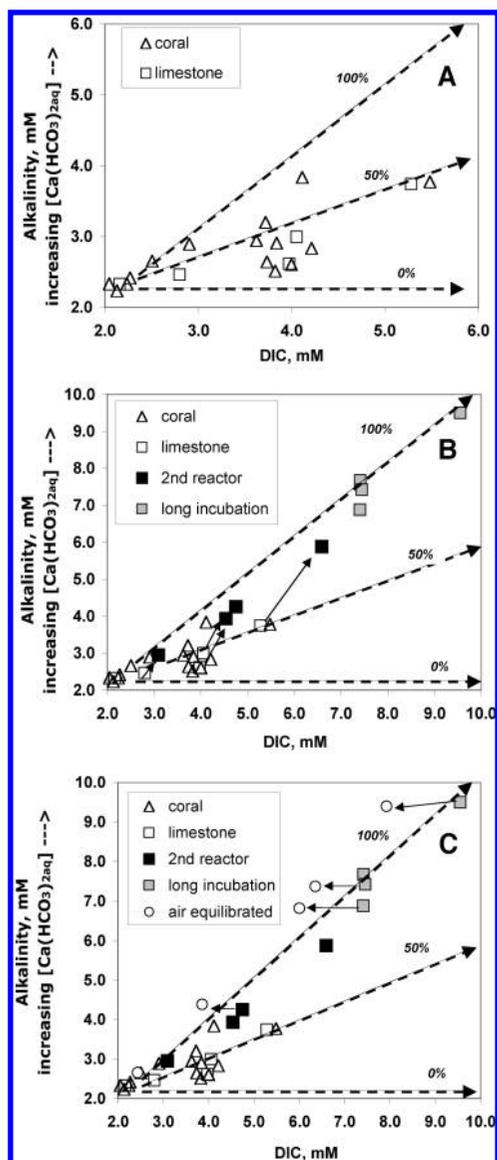
FIGURE 2. The alkalinity and total dissolved inorganic carbon concentrations, DIC, in seawater effluent from (A) a single reactor containing either coral or limestone packing as described in the text. B, data as in A in addition to effluent from a second downstream limestone reactor containing much finer limestone particles. The results from paired first and second reactors are linked by thin solid lines with arrows. Data for one to two week seawater + CO_2 incubations in a single reactor are indicated by gray squares. C, the results as shown in A and B in addition to data for selected reactor solutions that were fully equilibrated with air. Dashed lines denote the percentage of DIC contributed by alkalinity, principally calcium bicarbonate, $Ca(HCO_3)_2$. For comparison, ambient seawater values for DIC and alkalinity averaged 2.1 and 2.3 mmol/L (mM), respectively.

sidered as an effective though limited-capacity CO_2 mitigation strategy (27).

Two additional experimental modifications were performed in an attempt to increase CO_2 conversion to $Ca(HCO_3)_{2(aq)}$. One modification was to add a similarly sized second reactor downstream from the first reactor into which limestone with a much greater surface area per volume was placed (maximum particle dimension 0.28 cm). This had the effect of greatly increasing the water reaction surface area exposed to the carbonate, resulting in substantially increased formation of $Ca(HCO_3)_{2(aq)}$ as measured by increased alkalinity relative to increased DIC (Figure 2b). In other experi-

ments seawater in equilibrium with the CO_2/air mix was allowed to reside in the reactor of 1−2 weeks, greatly prolonging the gas, water, and carbonate contact time. Such treatments also substantially increased DIC concentration and CO_2 conversion to $Ca(HCO_3)_{2(aq)}$ (Figure 2b).

In order to test the permanence of the preceding carbon storage in seawater, selected reactor solution samples were copiously purged with air (Methods), thus stripping excess, uncombined molecular CO_2 from solution. Comparing resulting DIC and alkalinity to that of the original solutions and to ambient seawater demonstrates that 61−85% of the carbon originally added to the seawater remained in solution (Figure 2c), with little change in alkalinity and with no visual evidence of carbonate precipitation after aeration. Furthermore, during the course of aeration the pH of these solutions were returned to values near that of ambient seawater. The apparent reason for the DIC retention is that the loss of carbon via precipitation of $CaCO_3$ and the equilmolar loss of CO_2 from solution is strongly inhibited in seawater, as previously discussed. These results therefore demonstrate a method of adding significant quantities of anthropogenic carbon to the ocean that reduces or eliminates seawater pH depression, while avoiding the risks associated with concentrating, handing, and storing molecular CO_2 (e.g., CCS). It also suggests that once converted to marine $Ca(HCO_3)_{2(aq)}$, carbon storage will be long lasting, barring any uptake and precipitation of $CaCO_{3(s)}$ by marine calcifying organisms.

Indeed, it can be anticipated that the addition of $Ca(HCO_3)_{2(aq)}$ to seawater would enhance downstream bio-calcification rates as previously demonstrated by the experimental addition of Na or Ca bicarbonate to coral incubations (28, 29). Seawater/carbonate/CO_2 reactors similar to that experimentally used here are in fact routinely used to generate alkalinity and thus preserve or enhance shell formation by the marine organisms within saltwater aquaria (30). However, as evident in the geologic record (discussed above), once released into the ocean, inefficiencies in biological conversion of excess $Ca(HCO_3)_{2(aq)}$ (reversal of reaction 1) appear to allow seawater carbon residence times of many kyrs. In the meantime, the addition of $Ca(HCO_3)_{2(aq)}$ to the ocean should help preserve marine calcifiers that are currently challenged by increasing ocean acidity (2). This approach thus not only mitigates CO_2, but also potentially treats the effects of ocean acidification. Further research at larger scales and in more realistic settings is needed to prove these dual benefits.

Based on the preceding results, a quantitative model of this form of carbon capture and storage in seawater is depicted in Figure 3. Could such an approach be used to provide cost-effective CO_2 and ocean acidity mitigation at globally meaningful scales? Assuming that seawater is the only practical water source and alkalinity reservoir for the process (e.g., ignoring the use of inland saline aquifers), and assuming that optimized, large scale reactors could capture and add 6 mmol of carbon per liter of seawater (Figure 3), then 3.8×10^3 tonnes of seawater would be needed per tonne CO_2 captured. As a perspective, an average of 1.3×10^6 tonnes seawater are pumped per GWh_e generated by California coastal power plants that use once-through seawater cooling (31). Since 600 tonnes CO_2/ GWh_e are emitted by these natural gas fueled power plants, low-cost, downstream reuse of "free", once-through cooling water for reaction 1 could capture 57% of a power plant's CO_2 emissions. A higher percentage of CO_2 capture would require additional seawater pumping, at a modest energy penalty and monetary cost of $2−7/tonne CO_2 captured, highly dependent on reactor configuration and siting (13, 15, 16). Note that seawater scrubbing of flue gas is already used for SO_2 mitigation at some coastal power plants (32).
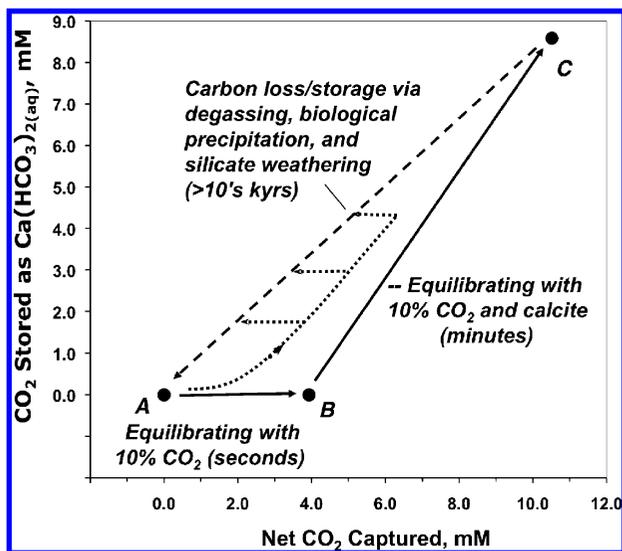
FIGURE 3. Model of seawater/limestone CO_2 capture and storage potential. Point: A, ambient seawater; B, seawater fully equilibrated with 10% CO_2; C, seawater fully equilibrated with 10% CO_2 and calcite (as contained in limestone). Note that net CO_2 stored $= \frac{1}{2}$ total carbon added during equilibration with limestone. Reactor inefficiencies in equilibrating CO_2 and limestone, and post-release CO_2 degassing mean that CO_2 capture and conversion to $Ca(HCO_3)_{2(aq)}$ will be less than the maximum theoretically possible under full equilibration (dotted line trajectories). The carbon thus released to the ocean will ultimately be lost (biological carbonate precipitation, CO_2 degassing) or stored (silicate weathering). Likely time scales are indicated for seawater carbon gain via partial equilibration in a reactor, and for post-release seawater carbon removal.

Reaction 1 consumes 2.3 tonnes of $CaCO_3$ per tonne CO_2 reacted. Use of an impure carbonate source such as limestone would likely increase the mass requirement to ~2.5 tonnes mineral/tonne CO_2 captured. This then equates to 855 tonnes limestone consumed per GWh_e if 57% of CO_2 emitted/GWh_e is to be consumed by reaction 1 as in the scenario above. The energy expenditure and cost of this limestone (extraction, processing, and transport) is is anticipated to average 80 kWh and $20 per tonne CO_2 captured, respectively, at coastal sites, but these estimates are quite sensitive to transport mode and distance (*13, 17*). Use of significant waste limestone stockpiles could significantly lower this cost, as well as reduce the environmental impact of new limestone extraction, if not remediate existing, massive waste limestone storage sites (*17*).

With capital costs estimated to be $2/tonne CO_2 (*13, 15*), a total cost of <$30/tonne CO_2 captured is then indicated. If 80% of this carbon can ultimately be converted and stored as $Ca(HCO_3)_{2(aq)}$ (Figures 2c, 3), a cost of <$38/tonne CO_2 mitigated is suggested. This compares to >$75/tonne CO_2 mitigated estimated for CCS when employed at conventional power plants (*7, 8*). The cost advantage of reaction 1 is largely achieved by avoiding CCS's expensive capture and purification of molecular CO_2. The relative technological simplicity of reaction 1 means that it could potentially be rapidly employed in the developing world where the need for CO_2 emissions reduction is greatest; analogous to widely used wet limestone or seawater flue gas scrubbing for SO_2 mitigation (*32, 33*). However, the method's large demand for carbonate mineral and water will likely limit its application to coastal sites. Further evaluation is needed of the economics, potential scale, permanence, environmental cost/benefit, and societal acceptability of this and other approaches to CO_2 emissions reduction and ocean acidity mitigation.

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