Assessing the potential of calcium-based artificial ocean alkalinization to mitigate rising atmospheric CO_2 and ocean acidification

Tatiana Ilyina,[1] Dieter Wolf-Gladrow,[2] Guy Munhoven,[3] and Christoph Heinze[4,5,6]

[1] Enhancement of ocean alkalinity using calcium compounds, e.g., lime has been proposed to mitigate further increase of atmospheric CO_2 and ocean acidification due to anthropogenic CO_2 emissions. Using a global model, we show that such alkalinization has the potential to preserve pH and the saturation state of carbonate minerals at close to today's values. Effects of alkalinization persist after termination: Atmospheric CO_2 and pH do not return to unmitigated levels. Only scenarios in which large amounts of alkalinity (i.e., in a ratio of 2:1 with respect to emitted CO_2) are added over large ocean areas can boost oceanic CO_2 uptake sufficiently to avoid further ocean acidification on the global scale, thereby elevating some key biogeochemical parameters, e.g., pH significantly above preindustrial levels. Smaller-scale alkalinization could counteract ocean acidification on a subregional or even local scale, e.g., in upwelling systems. The decrease of atmospheric CO_2 would then be a small side effect. **Citation:** Ilyina, T., D. Wolf-Gladrow, G. Munhoven, and C. Heinze (2013), Assessing the potential of calcium-based artificial ocean alkalinization to mitigate rising atmospheric CO_2 and ocean acidification, *Geophys. Res. Lett.*, *40*, doi:10.1002/2013GL057981.

1. Introduction

[2] Various geoengineering schemes of carbon dioxide removal (CDR) from the atmosphere have been proposed as additional means to keep growing greenhouse gas concentrations at bay until substantial emission reductions are realized [*Royal Society*, 2009; *Intergovernmental Panel on Climate Change* (*IPCC*), 2005]. One of the CDR methods is enhanced weathering—a process in which large amounts of silicate [*Köhler et al.*, 2010; *Schuiling and Krijgsman*, 2006] or carbonate rocks [*Caldeira and Rau*, 2000; *Harvey*, 2008; *Kheshgi*, 1995; *Rau*, 2011; *Rau and Caldeira*, 1999;] are processed, grinded, and dispersed on land or ocean. In the ocean, enhanced weathering would increase its total alkalinity (TA) characterized by the excess of proton acceptors (mainly bicarbonate and carbonate ions) over proton donors [*Wolf-Gladrow et al.*, 2007]. The net result of such artificial ocean alkalinization will be an increase of the seawater capacity to absorb and store carbon. This mimics naturally occurring reactions and accelerates processes that would otherwise remove CO_2 only on time scales of tens to hundreds of thousands of years [*Archer*, 2005]. An increase in TA would also alter seawater constituents so that its acidity decreases. Thus, in contrast to direct sequestration of carbon into the deep ocean [*Ridgwell et al.*, 2011] alkalinity enhancement, along with a drawdown of atmospheric CO_2, would also drive ocean pH and the saturation state of $CaCO_3$ (referred to as Ω) to higher values thereby counteracting ongoing ocean acidification. Unmitigated, ocean acidification is likely to have diverse negative impacts on marine biodiversity, food webs, and ecosystems including decrease in calcification rates or changes in reproduction and physiology observed in some marine organisms [*Gattuso and Hansson*, 2011].

[3] Variants of increasing TA via ocean-based enhanced weathering using calcium compounds can be realized by adding quick lime (CaO), lime (calcium hydroxide, $Ca(OH)_2$), or calcium carbonate ($CaCO_3$). Quick lime will react with water and form calcium hydroxide that will readily dissociate into Ca^{2+} and $2\,OH^-$, i.e., yielding the same ions as if adding lime. $CaCO_3$ can dissociate into Ca^{2+} and CO_3^{2-} if the aqueous solution is undersaturated (i.e., $\Omega < 1$) with respect to the mineral phase (calcite, aragonite) of the added material. The surface ocean is currently supersaturated with respect to aragonite and calcite ($\Omega > 1$) except for a few locations (some upwelling regions or close to strong CO_2 sources near volcanically active sites) [e.g., *Feely et al.*, 2004]. Thus, $CaCO_3$ has to be dissolved in an undersaturated solution before addition to the ocean. Whereas the resulting Ca^{2+} stays in solution (fully dissociated), the other dissociation products, namely OH^- and CO_3^{2-}, further react with H^+ to form water or HCO_3^- and thereby increasing pH (lowering $[H^+]$) and $[HCO_3^-]$ and decreasing $[CO_2]$. The addition of quick lime and lime has the same impact on TA: 1 mol of (quick) lime will add 2 mol to TA, while dissolved inorganic carbon (TCO_2) is not changing. When adding 1 mol Ca^{2+} and 1 mol CO_3^{2-} (i.e., when using $CaCO_3$), TA increases by 2 mol and TCO_2 by 1 mol, and thus, this method has lower effectiveness for ocean CO_2 sequestration than addition of (quick) lime. The addition of Ca^{2+} will result in small changes of the (high, about 10 mmol L^{-1}) concentration of Ca^{2+} in the ocean and thus will have little effects on Ω via $[Ca^{2+}]$; Ca^{2+} addition has an impact on Ω via changes in TA and thus $[CO_3^{2-}]$ [*Zeebe and Wolf-Gladrow*, 2001].

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[1]Max Planck Institute for Meteorology, Hamburg, Germany.
[2]Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research, Bremerhaven, Germany.
[3]Department of Astrophysics, Geophysics and Oceanography, University of Liège, Liège, Belgium.
[4]Geophysical Institute, University of Bergen, Bergen, Norway.
[5]Bjerknes Centre for Climate Research, Bergen, Norway.
[6]Uni Climate, Uni Research AS, Bergen, Norway.

Corresponding author: T. Ilyina, Max Planck Institute for Meteorology, Bundesstraße 53, DE-20146 Hamburg, Germany. (tatiana.ilyina@mpimet.mpg.de)
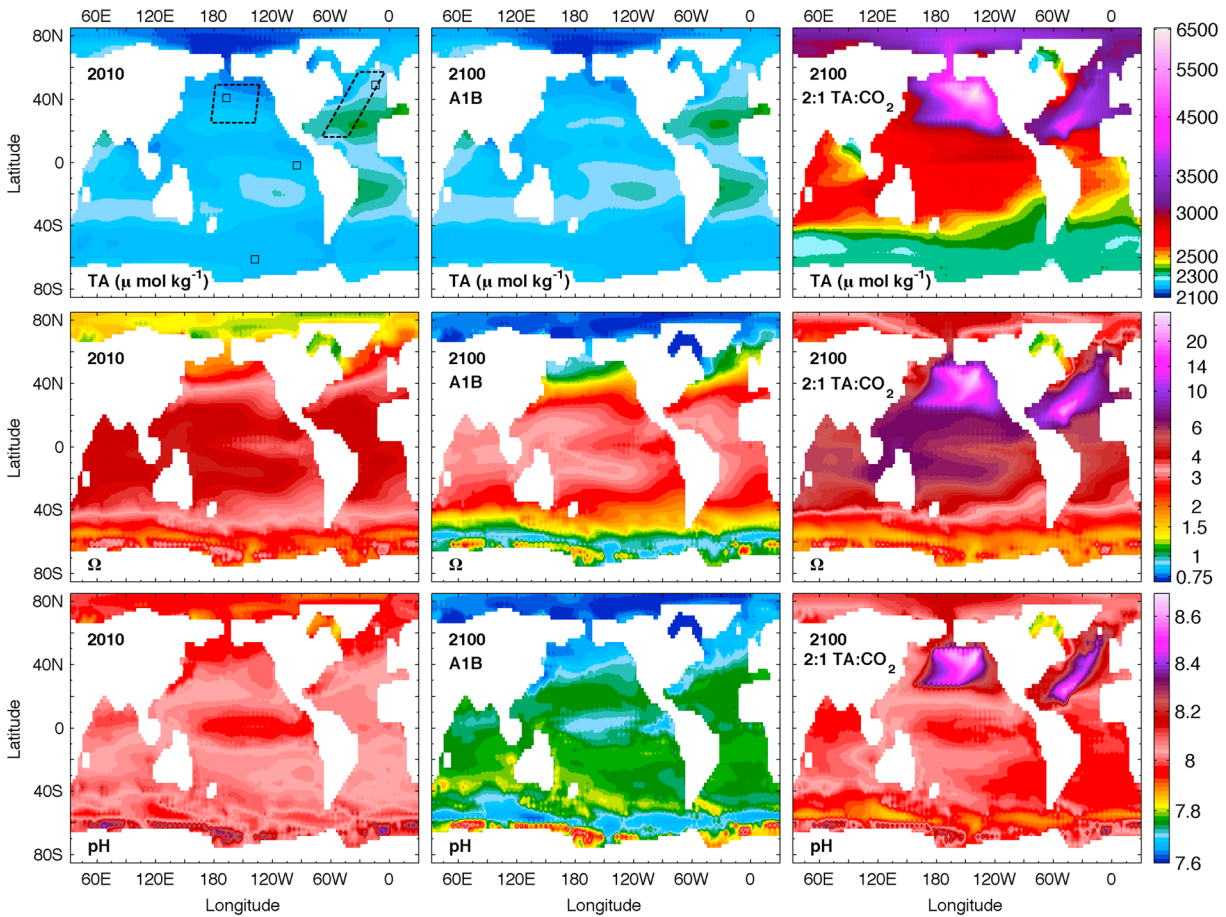
Figure 1. Surface maps of (top row) TA, (middle row) Ω, and (bottom row) pH in the years 2010 (left column) and 2100 without artificial ocean alkalinization (middle column), and calculated in the large-scale alkalinization scenario with the molar ratio (of $TA:CO_2$) 2:1 (right column). Sites of large-scale (dotted parallelograms) and subregional (solid squares) scenarios of alkalinity enhancement are shown in the upper left panel.

2. Model Scenarios of Alkalinity Enhancement

[4] We relate the amount of TA injected into the ocean to anthropogenic CO_2 emissions. In all scenarios, for every mole of emitted CO_2, we add a proportional flux of TA to the surface ocean in different locations (marked on Figure 1) or globally. We examine alkalinization scenarios bracketing the range of molar ratios of 0.2:1 mol mol^{-1} (mole TA added per mole emitted fossil fuel CO_2; later on referred to as TA: CO_2) at the lower end and 2:1 at the higher end.

[5] We start ocean alkalinization in the year 2020, continuing a preindustrial model run (beginning in 1750) forced with historic emissions and the IPCC's A1B scenario until the year 2100 [*IPCC*, 2007] and gradually declining thereafter, until the end of the simulation period (being 3050 years). Alkalinity enhancement is assumed to continue for as long as fossil fuel CO_2 is emitted in all model scenarios, except the one in which the effect of termination after 10 years is explored. Note that production of lime from $CaCO_3$ liberates CO_2 which should be compensated for [*Kheshgi*, 1995]. In order to reflect an increasing fossil fuel energy demand and therefore a decreasing effectiveness of CO_2 sequestration, we also ran scenarios in which TCO_2 is increased alongside with increasing TA.

[6] Projections of alkalinity enhancement have been calculated with the Hamburg Ocean Carbon Cycle model [*Six and*

Maier-Reimer, 1996; *Ilyina et al.*, 2009]. The model includes the inorganic carbon cycle chemistry following *Maier-Reimer and Hasselmann* [1987] where TA is a prognostic variable expressed as the sum of carbonate and borate alkalinities, as well as water dissociation products. Simple ecosystem dynamics is based on the explicit parameterization of nutrients, phytoplankton and zooplankton, detritus, and dissolved organic matter in the water column. There is a diagenetic model for the bioturbated sediment zone (following *Maier-Reimer et al.* [2005]). The model is forced by prescribed and globally uniform weathering fluxes and uses an averaged flow field representative of the modern ocean (i.e., without climate feedback and changes in riverine fluxes). The model runs with a time step of 1 month and has a horizontal resolution of 3.5°. It has 22 vertical layers with thicknesses varying between 25 m at the surface and 700 m in the deep ocean.

3. Mitigation Potential of Large-Scale Alkalinity Enhancement

[7] Increasing seawater TA enhances oceanic uptake of CO_2 from the atmosphere. In order to increase the contact area of the alkalized ocean with the air above, we first tested a model scenario in which alkalinity was distributed uniformly over the entire surface ocean (Figure S2 in the supporting

Table 1. Summary of Modeled Alkalinization Scenarios and Their Resulting Effects on the Sea Surface pH and Atmospheric pCO_2 Compared to the A1B Scenario Without Alkalinity Enhancement

Scenario	Global Ocean	Atlantic and Pacific					North Atlantic and Pacific	North Atlantic	North Pacific	Eastern Equatorial Pacific	Southern Ocean
Area (km^2)	360×10^6	47.10×10^6					435×10^3	203×10^3	232×10^3	306×10^3	160×10^3
Molar ratio of TA:CO_2	2:1	2:1	1.5:1	1:1	0.5:1	0.2:1	2:1	1:1	1:1	1:1	1:1
Max CO_2 drawdown (ppm)	450	450	402	287	150	72	369	168	267	285	265
Max regional pH increase	-	0.31	0.23	0.15	0.075	0.029	0.21	0.10	0.23	0.23	0.27
Max global pH increase	0.37	0.29	0.22	0.14	0.073	0.028	0.18	0.06	0.11	0.13	0.13

information and Table 1). In this global alkalinity enhancement scenario following the 2:1 (TA:CO_2) molar ratio, atmospheric CO_2 does not exceed 490 ppm (compared to 1000 ppm unmitigated) when fossil fuel CO_2 emissions follow the IPCC A1B scenario. Global seawater pH and Ω even slightly increase compared to today's values, approaching their preindustrial state. Large application area results in a uniform increase in seawater TA, pH, and Ω over the global ocean surface. Thus, the addition of large amounts of alkalinity globally (corresponding to about 2.4 Pmol yr^{-1} during the years 2020–2030) would substantially mitigate atmospheric CO_2 and ocean acidification without environmental stress generated by elevating biogeochemical properties significantly beyond naturally occurring levels. Global natural silicate weathering contributes about 0.012 Pmol yr^{-1} [*Munhoven*, 2002]. Hence, an alkalinization scenario with the molar ratio 2:1 TA:CO_2 is equivalent to accelerating the natural weathering flux by 2 orders of magnitude. Annual world bulk production of all varieties of lime is estimated at 0.3 Pg yr^{-1} on average between the years 2006 and 2010 [*The United States Geological Survey*, 2011]. Assuming this is all lime, this is equivalent to 0.005 Pmol Ca(OH)$_2$ yr^{-1}. The lime production would have to be increased by much more than 2 orders of magnitude worldwide to fulfill an efficient alkalinization scenario.

[8] In further model experiments, we focus on a set of large-scale regional scenarios in which TA was added over an area of about 47 million km^2 in the Pacific and Atlantic Ocean covering approximately one seventh of the World Ocean (Figure 1). Cumulative fossil fuel CO_2 emissions (since the year 1750) of about 1600 Pg C by the year 2100 are projected to elevate the atmospheric CO_2 concentration above 900 ppm and decrease global mean surface pH by about 0.4 compared to preindustrial values (Figure 2). Adding alkalinity in relatively small amounts, i.e., in proportion 0.2:1 mol mol^{-1} (TA:CO_2), has only little effects on atmospheric CO_2 (max −72 ppm; Table 1) and pH (<+0.03). Adding alkalinity in a 2:1 ratio would allow to keep atmospheric CO_2 below 520 ppm by the end of this century. Global mean surface pH would fall only by <0.1 (instead of 0.4 if no alkalinization is undertaken) compared to today's values, Ω would stay above preindustrial for about 100 years if TA is increased by 2.8 Pmol yr^{-1}.

[9] Effects of alkalinity enhancement propagate into the ocean interior. In the present day ocean, waters shallower than about 4500 m depth on average are supersaturated with respect to calcite. Ocean acidification reduces Ω and by the end of the 21st century, the global ocean below 300 m is projected to become undersaturated and thus corrosive for calcite (Figure 3). In our scenario with the highest mitigation effectiveness (with a molar ratio 2:1 TA:CO_2), the saturation horizon (depth at which $\Omega = 1$) deepens by a few hundred meters during the first few centuries after the onset of alkalinization.

[10] Some technologies considered for alkalinity enhancement, such as production of Ca(OH)$_2$ from quick lime obtained by decarbonation of $CaCO_3$ have large energy and fossil fuel CO_2 footprints. For instance, the heat production required to decarbonate $CaCO_3$ to Ca(OH)$_2$ would add extra CO_2 emissions, e.g., leading to a doubling of global CO_2 emissions in a 2:1 scenario. If the required heat could be produced from renewables, e.g., using a solar calcination technique [*Nikulshina et al.*, 2006], only a marginal extra CO_2 emission would arise in addition to the CO_2 released from $CaCO_3$ during calcination (or decarbonization). In our scenarios, CO_2 emitted during production of Ca(OH)$_2$ and due to the processing plus transport of materials is not considered explicitly. Instead, we ran scenarios in which TCO_2 is increased alongside with increasing TA in a ratio 2:1 (TA:TCO_2; Figure S4). Potential of these scenarios to mitigate atmospheric CO_2 is roughly half of that calculated for scenarios in which only TA is altered. This range of model scenarios also reflects on the different effectiveness of other Ca-based alkalinity enhancement methods which inevitably boost the uptake of atmospheric CO_2 and increase seawater pH and Ω, for instance, when using $CaCO_3$ instead of Ca(OH)$_2$ or replacing half of the Ca(OH)$_2$ by Ca(HCO$_3$)$_2$ (see section 1).

[11] Our results indicate that a large-scale regional alkalinization scenario (i.e., 2:1 mol mol^{-1} TA:CO_2) would significantly perturb distributions of some key biogeochemical parameters (Figure 1). In this scenario, a rise in surface TA above 5500 μmol kg^{-1} and in seawater pH up to 8.7 is projected in regions where alkalinity is injected. Surface Ω would rise above 30, significantly beyond naturally occurring levels (ranging between about 4 and 6 for calcite in the modern surface ocean). Experimental manipulation of calcium concentration [*Gattuso et al.*, 1998] showed that at aragonite saturation levels somewhat above present conditions, calcification rate in corals increased threefold; it does not increase further as aragonite saturation rises above the last glacial levels. Hence, high Ω values associated with alkalinity enhancement would imply increased biotic and abiotic precipitation of $CaCO_3$ (which is likely to start at Ω values of 20–25) [*Morse and He*, 1993]. As $CaCO_3$ formation and precipitation removes alkalinity, an increase in the upper ocean precipitation rate would decrease the oceanic capacity to take up atmospheric CO_2 and thus would set an upper limit for alkalinization. An elevated $CaCO_3$ particle export flux could enhance ballasting of particulate organic carbon with possible implications for the biological pump [*Klaas and Archer*, 2002]. Furthermore, speciation, abundance, and toxicity of metals are a function of pH and water hardness and most metals are less soluble in waters with higher pH [*Millero et al.*, 2009]. Also, biologically driven redox reactions involving nitrogen transformations in seawater are pH dependent so that large-scale changes in pH may alter the marine nitrogen cycle
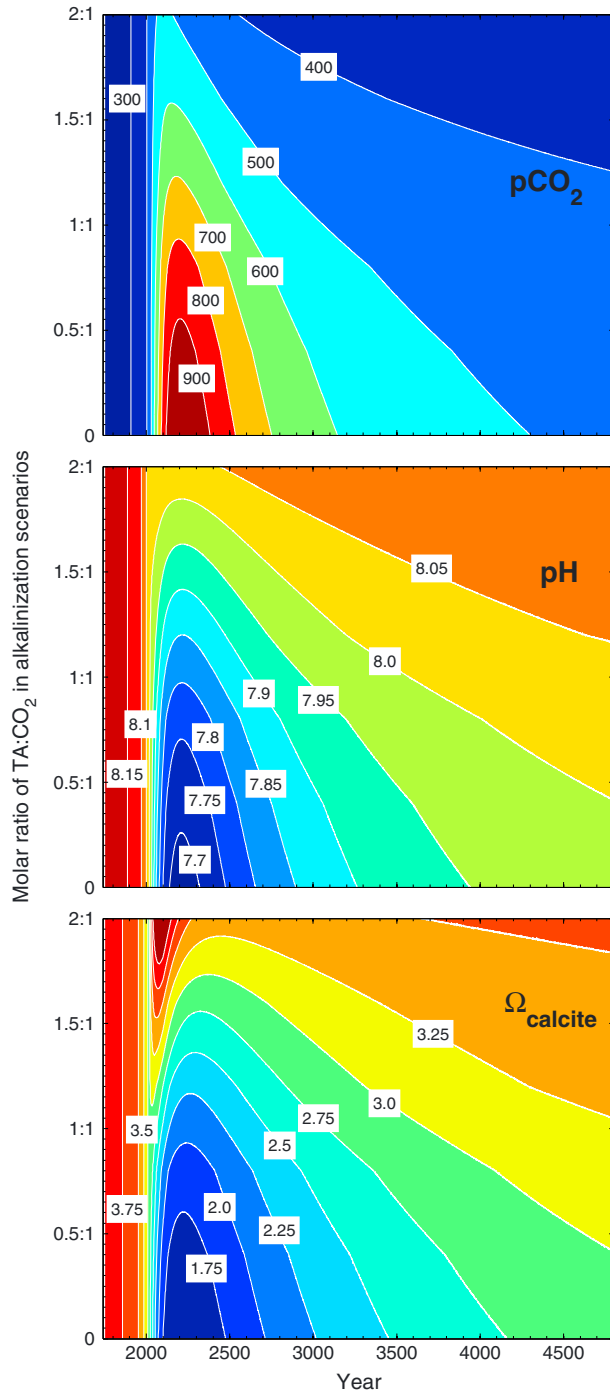
Figure 2. Mitigation potential of artificial ocean alkalinization: atmospheric CO_2 concentration (in ppm), sea surface pH, and Ω are shown by white contour lines for different alkalinization scenarios (represented in molar ratios TA:CO_2) as a function of time.

[Kranz et al., 2010]. Although the biological effects of alkalinity enhancement have not been fully assessed across the range of marine organisms, in a recent study, Cripps et al. [2013] demonstrated that liming of seawater in treatments lead to respiratory alkalosis (a disruption of acid-base homeostasis) in crustacean species already at pH 8.46 (with corresponding Ω of calcite values of 7.09). Such values (and higher) are projected in regions of alkalinity addition in our 2:1 TA:CO_2

scenario (Figure 1). With time ocean currents redistribute the alkalized water mass over a larger volume so that they resemble their today's pattern, albeit at higher TA and Ω values (Figure S2).

4. Potential of Artificial Ocean Alkalinization to Address Regional Impacts of Ocean Acidification

[12] While the World Ocean is capable of taking up large amounts of CO_2 [e.g., *Maier-Reimer and Hasselmann*, 1987; *Broecker and Takahashi*, 1977; *Sabine et al.*, 2004], a combination of physical, chemical, and biological properties together with hydrodynamic conditions make certain areas of the ocean act as net sinks or net sources of atmospheric CO_2 over the year [*Key et al.*, 2004]. Colder waters can dissolve more gaseous CO_2 but have a higher Revelle factor (lower buffering capacity) than warmer waters. Analogous to salinity, surface ocean TA is mainly determined by the addition or removal of freshwater via precipitation, sea ice melting, and evaporation and has a geographical gradient of about 450 µmol kg^{-1} with higher values in the gyres in the Atlantic and Pacific Ocean (Figure 1). In further model experiments, alkalinity is added in four subregions with different hydrodynamic conditions: the sink areas in the North Pacific and the North Atlantic, and the source areas in the upwelling regions off the coast of Peru and the Southern Ocean. In these smaller-scale scenarios, alkalinity is introduced over an area of 160,000–230,000 km^2. Amounts of alkalinity necessary to increase pH and/or Ω locally would be substantially lower compared to large-scale scenarios. Whereas seawater pH rises following the declining atmospheric CO_2, waters not yet affected by alkalinization are transported by ocean flow and mixing. As a result, subsurface pH drops further and contributes to decreasing surface pH values in regions remote from the alkalinization sites. The increase in pH varies regionally (Table 1), i.e., it increases on-site and decreases in remote areas. Hence, regional alkalinity enhancement has the potential to fully mitigate ocean acidification only at the point of application. On longer time scales (>1000 years), the changes in atmospheric CO_2 and ocean pH become independent on the location of the alkalinization sites and are determined only by the amount of alkalinity added. About 1500 years after the onset of alkalinization, the global ocean surface TA approaches the TA in the unmitigated A1B scenario, whereas atmospheric CO_2 and seawater pH differ from their unmitigated counterparts throughout the whole simulation (Figure 3).

[13] The North Atlantic had the lowest effectiveness in decreasing CO_2 and increasing pH in our scenarious pH (Table 1). Even though both North Atlantic and North Pacific absorb atmospheric CO_2 (on average over a year), they have different circulation regimes. Alkalinity added in the surface waters in the North Atlantic would be transported into the deep ocean, particularly in the areas of deep water formation, thus having only a small effect on the surface pH in this region. On the contrary, adding alkalinity to surface waters which would be longer in contact with the air above showed to be more effective in lowering of atmospheric CO_2. In line with the circulation pattern and the prevailing Antarctic Circumpolar Current, alkalinization at the Southern Ocean site had the fastest and largest effect on the regional surface pH. Alkalinity addition to the net CO_2 source areas also yielded the largest mitigation effect on the global pH.
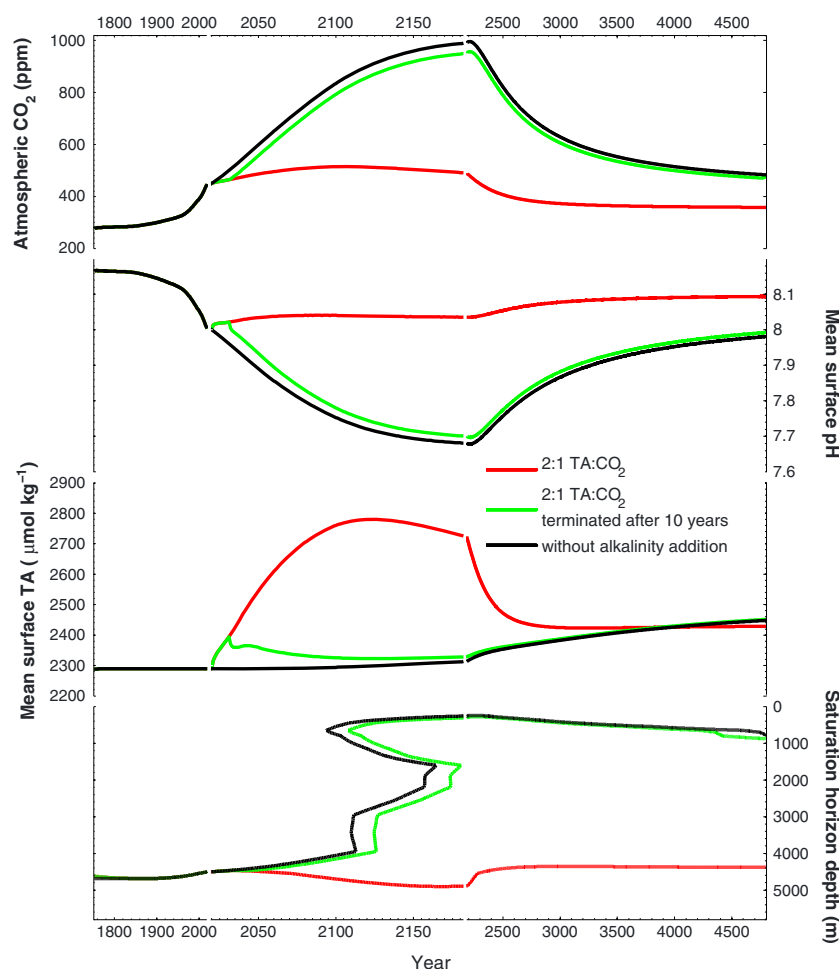
Figure 3. Temporal evolution of atmospheric CO_2, pH, TA, and the saturation horizon of calcite (depth at which $\Omega = 1$) in the unmitigated A1B scenario (black), the 2:1 mol mol^{-1} TA:CO_2 scenario (red), and the 2:1 scenario in which alkalinization terminated after 10 years (green). Note that a broken x axis has been used to zoom into the years 2020–2200.

5. Effects of Termination of Artificial Ocean Alkalinization

[14] While TA of the oceans does not change when CO_2 is dissolved, it is projected to increase in a high CO_2 world as a result of decreased production [*Ilyina et al.*, 2009] and increased dissolution of $CaCO_3$ (Figure 1) [*Ilyina and Zeebe*, 2012]. Due to slow equilibration with sediments at the ocean bottom, concomitant changes in seawater carbonate chemistry go on for several tens of thousands of years [*Archer*, 2005; *Ridgwell and Hargreaves*, 2007]. Likewise, even a short-term alkalinization would have long-lasting effects on seawater carbonate chemistry. We look at the effect of termination of alkalinization after 10 years (Figure 3). In this scenario, atmospheric CO_2 decreases by 30 ppm (in the 2:1 mol mol^{-1} TA:CO_2 scenario) relative to the unmitigated A1B scenario in the 21st century. Once alkalinization has stopped, atmospheric CO_2 reverts back to rising at the rate determined by the fossil fuel CO_2 emissions growth. Unlike solar radiation management (using sulfate aerosols) [*Niemeier et al.*, 2010], the effect of alkalinization is permanent (at least on the time scale of several thousands of years) and CO_2 concentration remains about 10 ppm below the unmitigated scenario during the tail part of the simulation when fossil fuel CO_2 emission cease. Similarly, a no-penalty termination of

alkalinization holds for seawater pH so that it stays markedly above the unmitigated A1B scenario throughout the simulation period. Thus, alkalinization would not involve a long-term commitment.

6. Summary

[15] In summary, scenarios in which large amounts of alkalinity (i.e., in a ratio of 2:1 with respect to emitted CO_2) are added over large ocean areas or even globally can boost oceanic uptake of fossil fuel CO_2 from the atmosphere and avoid further ocean acidification (so that seawater pH and Ω do not fall below the values at the onset of alkalinization) on the global scale. Such alkalinity enhancement could drive some key biogeochemical parameters to values significantly above natural levels. Hence, while alkalinization has high mitigation potential if applied globally, large-scale regional applications can potentially have a number of environmental implications that are intrinsically difficult to predict. In the light of our results, reduction of greenhouse gas emissions is necessary to address both climate change and ocean acidification globally. On short time scales, alkalinity enhancement could target local ocean acidification mitigation allowing avoidance of large changes in surface pH and Ω in certain high-value ecosystems of the world ocean, e.g., in semi-enclosed basins,

near coral reefs, or upwelling regions. The global decrease of atmospheric CO_2 would then be considered a small side effect. The mitigation attained via alkalinity enhancement would remain so that climate does not immediately revert once this geoengineering method is no longer applied. While further research is needed to offer detailed information for knowledgeable policy decisions, our results provide the first assessment of ocean alkalinity enhancement and the concomitant long-term changes in ocean biogeochemistry based on a three-dimensional ocean general circulation model.

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