Geophysical Research Letters

RESEARCH LETTER
10.1029/2022GL101954

Long-Term Slowdown of Ocean Carbon Uptake by Alkalinity Dynamics

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Key Points:

- Oceanic uptake of carbon could slow in upcoming centuries through previously unidentified alkalinity-climate feedback
- Reduced upwelling and carbonate buffer enhance the influence of alkalinity on the increase in surface ocean carbon dioxide
- Reductions in surface alkalinity will reduce the rate of carbon uptake on multi-century timescales

Supporting Information:

Supporting Information may be found in the online version of this article.

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Citation:

Chikamoto, M. O., DiNezio, P., & Lovenduski, N. (2023). Long-term slowdown of ocean carbon uptake by alkalinity dynamics. *Geophysical Research Letters*, *50*, e2022GL101954. https://doi.org/10.1029/2022GL101954

Received 31 OCT 2022
Accepted 3 JAN 2023

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Abstract Oceanic absorption of atmospheric carbon dioxide (CO_2) is expected to slow down under increasing anthropogenic emissions; however, the driving mechanisms and rates of change remain uncertain, limiting our ability to project long-term changes in climate. Using an Earth system simulation, we show that the uptake of anthropogenic carbon will slow in the next three centuries via reductions in surface alkalinity. Warming and associated changes in precipitation and evaporation intensify density stratification of the upper ocean, inhibiting the transport of alkaline water from the deep. The effect of these changes is amplified threefold by reduced carbonate buffering, making alkalinity a dominant control on CO_2 uptake on multi-century timescales. Our simulation reveals a previously unknown alkalinity-climate feedback loop, amplifying multi-century warming under high emission trajectories.

Plain Language Summary Over the past century, humans have been burning fossil fuels and adding extra carbon dioxide to the atmosphere. The ocean has been doing us a big favor by absorbing some of this carbon dioxide, lowering the amount of global warming that occurs. Our study shows that the ocean will begin to lose its ability to absorb carbon dioxide beyond the year 2100, leaving more fossil-derived carbon in the atmosphere and leading to additional global warming. Our study describes a previously undiscovered mechanism for the slowdown in ocean carbon absorption, where changes in rainfall and warming affect ocean currents that, in turn, change the chemistry of the ocean surface.

1. Introduction

Oceanic uptake of anthropogenic carbon—currently responsible for absorbing 30% of carbon dioxide (CO_2) fossil fuel-based emissions (Ciais et al., 2013; Friedlingstein et al., 2020; Sabine & Tanhua, 2010; Watson et al., 2020)—is predicted to continue to increase through the end of this century under higher atmospheric CO_2 concentrations (Tjiputra et al., 2014; Wang et al., 2016). Its evolution in subsequent centuries, in contrast, remains largely unknown (Frölicher & Joos, 2010; Koven et al., 2022; Matsumoto et al., 2010; Plattner et al., 2008; Randerson et al., 2015; Tokarska et al., 2016). Understanding this multi-century response is essential for projecting future climate change, particularly the evolution of slower Earth system components, such as the cryosphere, with significant implications for long-term sea-level rise (Archer & Brovkin, 2008; Charbit et al., 2008; Joos et al., 2013; Lord et al., 2016).

Throughout this century, the absorption of anthropogenic carbon is expected to continue to increase via enhanced uptake of CO_2 over the Southern and the North Atlantic oceans (Landschützer et al., 2015; Tjiputra et al., 2014; Wang et al., 2016). This process could become less efficient as the ocean becomes more acidic, reducing the carbonate buffering (Chikamoto & DiNezio, 2021; Doney et al., 2009; Egleston et al., 2010; Sabine et al., 2004). In the ocean carbon system, which is fundamentally governed by changes in dissolved inorganic carbon (DIC), alkalinity, temperature, and salinity (Lovenduski et al., 2007; Sarmiento & Gruber, 2006), the changes in uptake under massive CO_2 emissions become susceptible to DIC, which increases significantly with CO_2 dissolution. However, when the ocean reduces the carbonate buffer and weakens its capacity to uptake CO_2, the change in DIC becomes smaller, making ocean carbon chemistry relatively sensitive to other variables than DIC, such as alkalinity and temperature (Riebesell et al., 2009). As a result, the rate at which carbon is stored as DIC may no longer be the primary factor controlling the air-sea difference in CO_2 partial pressure driving CO_2 uptake. Other factors, such as warming or changes in alkalinity, could become equally important drivers of CO_2 uptake on these longer timescales (Chikamoto & DiNezio, 2021; Egleston et al., 2010; Matsumoto et al., 2010; Riebesell et al., 2009).

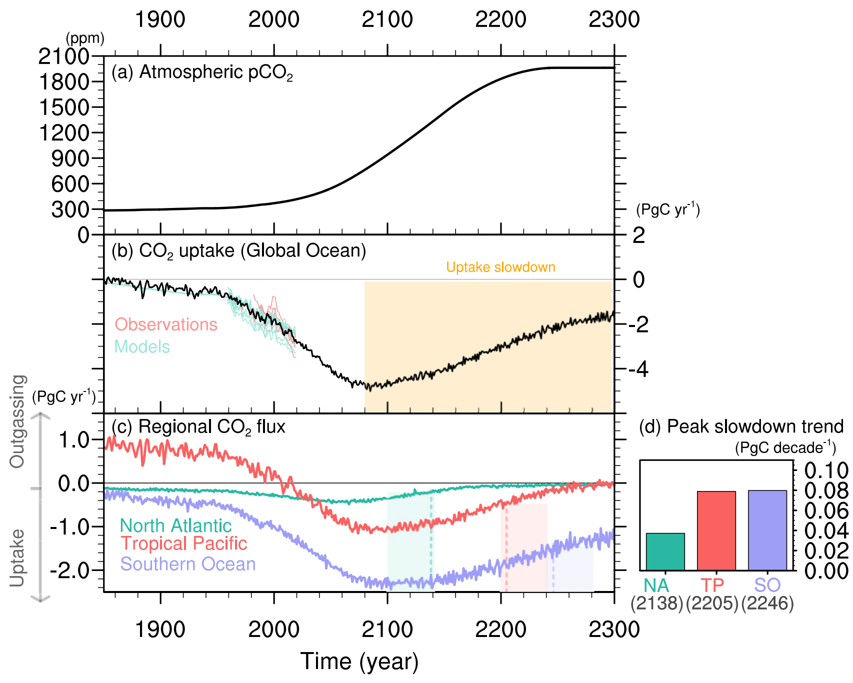

Figure 1. Acceleration and slowdown of oceanic CO_2 uptake. (a) Prescribed increase in atmospheric CO_2 concentrations (ppm), (b, c) Globally and regionally integrated sea-air CO_2 flux (PgC yr^{-1}), and (d) peak slowdown trend, that is, a slowdown in uptake in sea-air CO_2 flux (PgC $decade^{-1}$) in the North Atlantic (40°–70°N), the tropical Pacific (30°S–30°N), and the Southern Ocean (>30°S). Atmospheric CO_2 concentration is prescribed from historical observations, the RCP8.5 emission scenario, and its extension through 2300 (Lindsay et al., 2014; Meinshausen et al., 2011). Negative CO_2 flux corresponds to oceanic uptake. Gray and light pink lines in (b) derive from interpolated observations and ocean biogeochemical model hindcast simulations (Friedlingstein et al., 2020). The dash lines in panel c show the timing of the peak slowdown trend, defined by the highest trend of the sea-air CO_2 flux for 40 consecutive years since 2000 (Figure S1 in Supporting Information S1). The bar in panel d shows the peak slowdown trend, which is the average of the sea-air CO_2 flux trend over the 40 years when the trend peaks (colored shading in panel c). Numbers on the *x*-axis in (d) are peak slowdown years (dashed colored lines in (c)).

However, little is known about the relative importance of these processes and how they interact with a changing climate despite recent advances simulating these processes and the global ocean carbon cycle using numerical models. Here we explore this question using a simulation of future changes in the global climate and carbon cycle performed with the Community Earth System Model (CESM1). This model simulates a carbon cycle in the atmosphere, ocean, and terrestrial biosphere, along with the increasing oceanic carbon uptake in agreement with historical trends (Figure 1b).

2. Experimental Design and Methods

CESM1, an Earth system model that includes a mathematical representation of the atmosphere, ocean, and terrestrial biosphere, can simulate how rising atmospheric CO_2 affects ocean carbon uptake through changes in atmospheric and ocean dynamics (Long et al., 2013). Here, we analyzed a simulation of the CESM run under a "business-as-usual" emission scenario (Moore et al., 2018; Randerson et al., 2015). The emission scenario is the RCP8.5 high emission scenario for the 21st century, followed by a smooth transition to stabilized concentration after 2250 achieved via linear adjustment of emissions after 2150 (Meinshausen et al., 2011). Under this worst-case scenario, atmospheric CO_2 concentrations increase to 1962 ppm by the year 2250, remaining stable at that level until the year 2300 (Figure 1a). This simulation, therefore, allows the study of the influence of the response of the ocean carbon cycle to a significant and sustained increase in atmospheric CO_2 over the course of many centuries. We analyzed changes in ocean uptake in the significant regions of the Southern Ocean (>30°S), the tropical Pacific (30°S–30°N), and the North Atlantic Ocean (40°–70°N). For each region, we decomposed the changes in sea-air CO_2 flux into the contributions from changes in gas exchange (i.e., wind speed), sea-ice extent, temperature-dependent solubility, and the sea-air difference in partial pressure of CO_2 (ΔpCO_2)

(Wanninkhof, 2014; Wanninkhof et al., 2013). In addition, to fully understand the drivers of simulated changes in oceanic carbon uptake, we quantified the influence of changes in sea surface temperature, salinity, DIC, and alkalinity on ΔpCO_2, the main driver of long-term changes in sea-air CO_2 flux.

3. Results

The simulation predicts that oceanic uptake of carbon will peak toward the year 2080 and decline in subsequent centuries, stabilizing at about half the peak magnitude by the year 2300 (Figure 1b). This decline occurs despite atmospheric CO_2 concentrations continuing to increase at similar rates for at least a century after the peak in CO_2 uptake (Figure 1a). The slowdown in CO_2 uptake occurs in all main regions where the ocean is currently absorbing anthropogenic carbon: the Southern Ocean, the tropical Pacific Ocean, and the North Atlantic (Figure 1c and Figure S1 in Supporting Information S1). The most significant slowdown occurs in the Southern Ocean, where the CO_2 uptake decreases by 58% from the peak by the year 2300. The tropical Pacific will shift from historical outgassing to peak absorption in the year 2100, with no flux by 2300, meaning zero area-averaged sea-air CO_2 flux. The North Atlantic is projected to exhibit negligible uptake by the mid-22nd century, no longer the dominant region absorbing anthropogenic carbon as it currently is (Takahashi et al., 2009). Together, the changes over the three areas account for over 80% of the global ocean uptake reduction in subsequent centuries. The changes in sea-air CO_2 flux associated with these variations in uptake are controlled primarily by changes in ΔpCO_2 globally and in each of these sensitive regions (Figure S2 in Supporting Information S1). Other factors influencing air-sea gas exchange, such as temperature, wind speed, or sea-ice coverage, play a secondary role (Text S1 and Figure S2 in Supporting Information S1).

In our experimental setup, atmospheric pCO_2 (pCO_2^{atm}) is prescribed to increase continuously and stabilize approaching the year 2300; therefore, the simulated surface ocean partial pressure of CO_2 (pCO_2^{ocn}) is the main controlling factor of the temporal evolution of ΔpCO_2. Indeed, the slowdown of ocean uptake begins around 2080 when the rate of increase in pCO_2^{ocn} is faster than the rate of increase of pCO_2^{atm}, regardless of the continued increase in pCO_2^{atm}. Furthermore, the decline in ΔpCO_2 in the 22nd century is driven by an acceleration in the rate of increase of pCO_2^{ocn}, which is faster than the increase rate for pCO_2^{atm}. This pCO_2^{ocn} increase reduces ΔpCO_2, leading to a slowdown in uptake. The pCO_2^{ocn} trend is dominated by changes in DIC and alkalinity (Figure 2). In the Southern Ocean, DIC is not yet in equilibrium and contributes to the reduction in pCO_2^{ocn} (Figure 2b). Instead, alkalinity and temperature increase pCO_2^{ocn}, slowing the uptake of CO_2. All changes in the tropical Pacific Ocean, that is, DIC, alkalinity, and temperature, contribute to the pCO_2^{ocn} increase that slows the uptake (Figure 2c). In contrast, the pCO_2^{ocn} trend in the North Atlantic Ocean is explained by the increase in DIC (Figure 2d). The global ocean reflects a combination of regional responses (Figure 2a), highlighting the relative importance of non-DIC as well as DIC factors on the pCO_2^{ocn} trend. In addition, the impacts of DIC and alkalinity on the pCO_2^{ocn} trend will increase after this century. Once anthropogenic CO_2 accumulation reduces the carbonate buffer of the ocean, the ocean's capacity to absorb CO_2 from the atmosphere will diminish (Doney et al., 2009; Egleston et al., 2010). This condition will increase the sensitivity of pCO_2^{ocn} to DIC and alkalinity, amplifying the DIC and alkalinity effects on the uptake up to three times in 2300 (Figures 2e and 2f, Figures S3 and S4 in Supporting Information S1). That is, the efficiency of the ocean for carbon uptake will be more affected by the variability of ocean surface DIC and alkalinity over the next two centuries.

Anthropogenic warming will stratify the upper ocean, altering the exchange of DIC and alkalinity between the deep ocean and the surface. We find that the Southern Ocean will become more thermally stratified, preventing the upwelling of high alkaline waters and reducing surface alkalinity (Figure 3 and Figure S5 in Supporting Information S1). In the future, ocean pCO_2 will become more sensitive to changes in DIC and alkalinity due to reduced carbonate buffer (Figure 3). This amplifies the influence of the reductions in surface alkalinity, which will be just a small fraction (less than 3%) of the current conditions but will have an outsized influence on surface pCO_2 due to reduced carbonate buffering. Together, these effects contribute to the slowdown of carbon uptake in key regions of the world ocean. In the tropical Pacific, decreased upwelling due to thermal stratification and weakened trade winds results in less transport of high alkalinity waters to the surface, accelerating the increase in surface pCO_2 and reducing CO_2 uptake. In the North Atlantic Ocean, ocean surface freshening with less evaporation and the collapse of the Atlantic meridional overturning circulation (Figures S5 and S6 in Supporting Information S1) play a significant role in reducing the downward transport of carbon into the deep ocean (DeVries et al., 2017; Fontela et al., 2016) and accumulating anthropogenic CO_2 in the surface (Figure 3a). This excess surface carbon pool will end the current significant uptake of CO_2 in this region after 2100. This positive

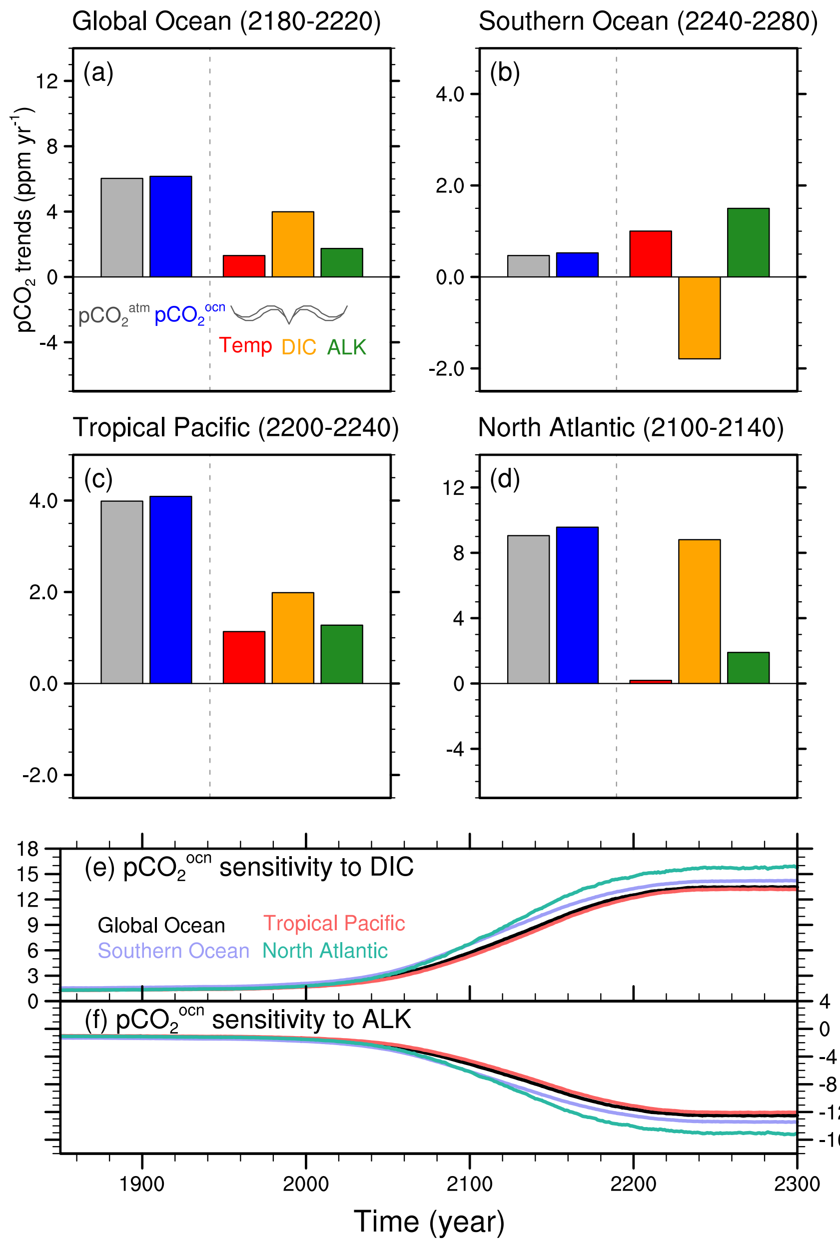

Figure 2. Drivers of oceanic CO_2 slowdown. (a–d) The atmospheric pCO_2 trend (gray), ocean surface pCO_2 trend (blue), and the contributions of changes in temperature (red), dissolved inorganic carbon (DIC) (orange), and alkalinity (green) to the oceanic pCO_2 trend (ppm yr^{-1}), and (e and f) the sensitivity of ocean pCO_2 to DIC and alkalinity. The sum of the contributions of temperature, salinity (not shown but very small), DIC, and alkalinity equals the ocean pCO_2 trends (details in Figure S3 in Supporting Information S1). Positive trends in ocean surface pCO_2 correspond to the trend toward decreasing oceanic uptake of CO_2. The DIC and ALK contributions show only the pCO_2 trends to the undiluted effect (physical and biological processes), excluding the diluted effects (Text S2 in Supporting Information S1). The analysis period is the maximum positive trend in sea-air CO_2 flux: 2100–2140 in the North Atlantic, 2200–2240 in the equatorial Pacific, and 2240–2280 in the Southern Ocean.

trend of DIC turns to a negative trend over time. This is because reduced CO_2 dissolution due to buffer reduction slows the increase rate of the surface ocean DIC, and the stratified ocean reduces the upwelling of DIC-rich water to the surface. The negative DIC trend will again promote ocean carbon uptake (Figure S3 in Supporting Information S1). Instead, the negative trend of alkalinity due to reducing the exchange of alkaline deep water with the surface and ocean warming will increase the surface ocean CO_2, slowing the carbon uptake in this region (Figure 3d and Figure S3d in Supporting Information S1).

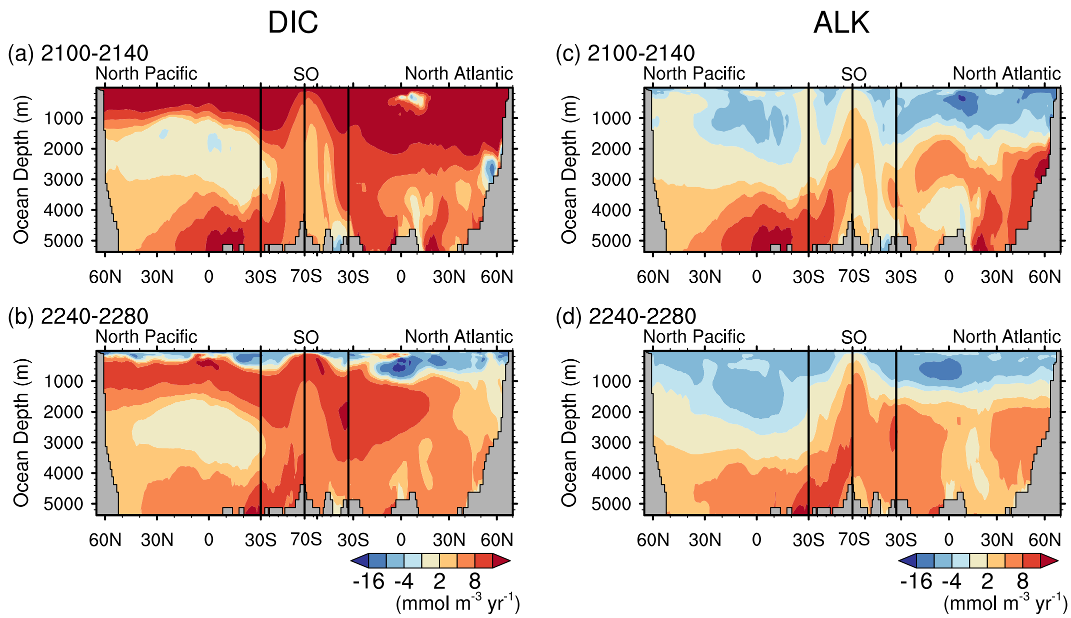

Figure 3. Spatiotemporal dissolved inorganic carbon (DIC) and alkalinity patterns. Simulated hydrographic sections of trends in (a, b) DIC and (c, d) alkalinity during 2100–2140 and 2240–2280. The depth sections are zonal averages. DIC and alkalinity are normalized by salinity (the standard salinity is 35 psu).

The responses identified here are part of a previously unknown positive alkalinity-climate feedback loop that could leave more anthropogenic CO_2 in the atmosphere and amplify long-term anthropogenic warming (Figure 4). Ocean warming and increased sea surface freshwater strengthen the thermal and salinity stratification of the upper ocean. This response increases surface DIC as long as anthropogenic carbon continues to be injected, promoting ocean acidification and reducing carbon uptake (Terhaar et al., 2021). Furthermore, ocean stratification decreases the upwelling of deep alkaline and DIC-rich waters to the surface, reducing the surface alkalinity and DIC. Compared to the 21st century, when changes in surface DIC are driven by CO_2 dissolution, these changes will be controlled by ocean dynamics. Unlike DIC lowering by stratification, which decreases pCO_2^{ocn}, decreasing alkalinity accelerates the rate of increase of pCO_2^{ocn} relative to pCO_2^{atm}, reducing the uptake

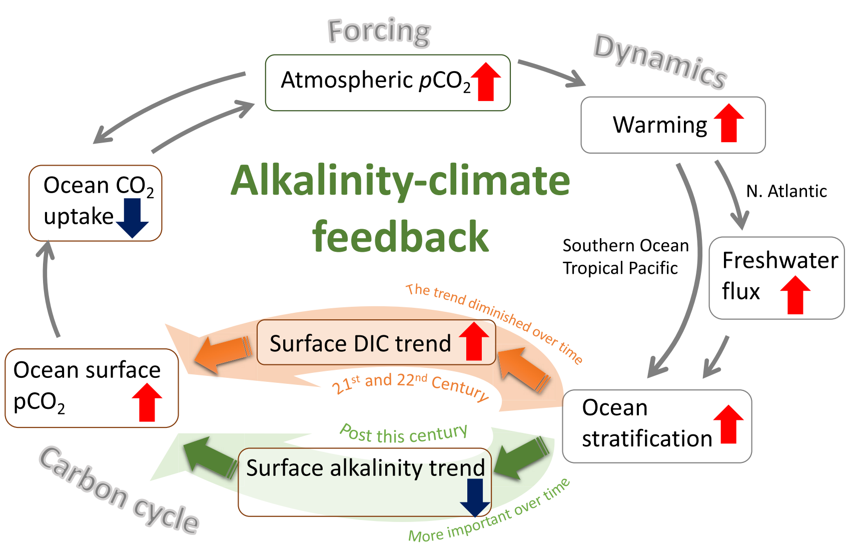

Figure 4. The alkalinity-climate feedback. This schematic illustration shows how changes in upper-ocean density stratification and the ocean carbon cycle respond to increased atmospheric CO_2 and feedback on one another to produce an amplified response on multi-century timescales. A vertical red arrow indicates an increase in that variable, while a vertical dark blue arrow indicates a decrease.

of CO_2 (Figure S3 in Supporting Information S1). This implies that uptake rates, which are currently controlled by changes in DIC, will be also influenced by changes in alkalinity after this century. The uptake response will be further amplified in the following centuries when the ocean reduces carbonate buffering associated with anthropogenic CO_2 dissolution. As CO_2 uptake becomes less effective, it accelerates the increase of pCO_2^{atm}, further amplifying the warming, thus closing the feedback loop. This positive feedback loop could play a dominant role in reducing the uptake of CO_2 alongside weakening the Atlantic meridional overturning circulation (Holliday et al., 2020; Randerson et al., 2015; Terhaar et al., 2021) and the Walker Circulation (DiNezio et al., 2009; Terada et al., 2020).

4. Conclusions

Ocean acidification and reductions in surface alkalinity slow oceanic carbon uptake as the ocean accumulates anthropogenic CO_2 and reduces the carbonate buffer. On multi-century timescales, alkalinity dynamics could become the primary driver of oceanic CO_2 uptake if high CO_2 emissions continue. Projections of ocean carbon uptake for this century are dominated by increases in DIC (Arora et al., 2013; Friedlingstein, 2015; Omta et al., 2011) that lag behind increases in pCO_2^{atm}. Therefore, uncertainties converge in numerical models (Roy et al., 2011; Wang et al., 2016). In contrast, longer-term changes, particularly under high emissions, might be more uncertain due to the multiple processes involved in the changes in CO_2 uptake, ocean circulation, precipitation, evaporation, and carbon chemistry, including the positive feedback presented here. The response of these processes to multi-century climate change is likely model-dependent. Furthermore, a freshwater flux from melting ice on Antarctica or Greenland (Perner et al., 2019) not included in our simulation may facilitate upper-ocean salinity stratification, reducing surface ocean alkalinity and slowing carbon uptake on long timescales. Other processes not included in our simulation could play a substantial role on these timescales. Alkalinity could increase, driven by the dissolution of calcified organisms due to ocean acidification (Fabry et al., 2008; Orr et al., 2005). Seafloor calcium carbonate neutralization, which acts on 1000 years timescales (Archer et al., 1997; Chikamoto et al., 2008), could also increase the alkalinity (Chikamoto et al., 2009; Paquay & Zeebe, 2013), as could enhance weathering under a wetter and warmer climate (Lord et al., 2016; Renforth & Henderson, 2017; Weyhenmeyer et al., 2019). The high sensitivity of oceanic carbon uptake to alkalinity on multi-century timescales sheds light on the effect of artificial alkalinity addition for carbon sequestration (Middelburg et al., 2020; Renforth & Henderson, 2017). The positive feedback presented here could delay the influence of these processes, extending the period required for the long-term stabilization of atmospheric CO_2.

Data Availability Statement

The analysis data of the CESM1 simulation in this study were uploaded to https://zenodo.org/record/7425089#.Y5ZGH-zMIq0 (https://doi.org/10.5281/zenodo.7425089). The observational data of sea-air CO_2 flux in Figure 1a were downloaded from https://www.ldeo.columbia.edu/res/pi/CO2/carbondioxide/pages/air_sea_flux_2010.html.

Acknowledgments

The authors are grateful to J. Randerson, who provided output from a series of historical and future projection simulations of CESM analyzed in this study. We are grateful to anonymous reviewers for their insightful comments. This research is supported by the National Science Foundation Grant OCE-1635465.

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