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UNIVERSITY OF CALIFORNIA
SANTA CRUZ

**THE EVOLUTION OF OCEAN CARBON CYCLE FEEDBACKS IN
OBSERVATIONS AND MODELS**

A dissertation submitted in partial satisfaction
of the requirements for the degree of

DOCTOR OF PHILOSOPHY

in

OCEAN SCIENCES

by

Mar C. Arroyo

September 2025

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Abstract

The Evolution of Ocean Carbon Cycle Feedbacks in Observations and Models

by

Mar C. Arroyo

Since the Industrial Revolution, the ocean has absorbed a cumulative ~40% of the anthropogenic carbon (C_{ant}) released into the atmosphere by fossil fuel emissions. C_{ant} accumulation in the upper ocean has driven an increase in the partial pressure of carbon dioxide gas ($p\text{CO}_2$) and associated declines in pH and carbonate ion concentration. These chemical changes, collectively referred to as ocean acidification (OA), progressively weaken the ocean's buffer capacity and reflect the evolution of a positive marine carbon cycle feedback that reduces the efficiency of future C_{ant} uptake and amplifies the influence of natural variability on the carbonate system. This dissertation investigates the spatial and temporal changes in the ocean carbon cycle caused by C_{ant} using a combination of in situ observations, data synthesis products, and output from regional and global ocean models to improve our understanding of the processes governing the ocean carbon sink and its evolving feedbacks. Chapter 1 evaluates the impact of C_{ant} accumulation on multiple OA metrics throughout the water column in the North Pacific Ocean and California Current Large Marine Ecosystem using ship-based observations. Results indicate that the greatest increases in $p\text{CO}_2$ occur subsurface, where C_{ant} content is moderate and $p\text{CO}_2$ change can exceed overlying surface change by $\geq 100\%$. Amplified $p\text{CO}_2$ responses in the interior ocean are related to background ocean carbonate chemistry, with the greatest subsurface

changes associated with poorly buffered waters that have experienced substantial organic matter remineralization. Chapter 2 evaluates the impact of C_{ant} on the seasonal variability of $p\text{CO}_2$ in the surface ocean using output from global ocean biogeochemical models (GOBMs) used by global carbon budgeting efforts to estimate the historical ocean carbon sink strength. Results indicate that dissimilar model representations of surface ocean $p\text{CO}_2$ seasonality, particularly during winter, lead to increasing disagreement in annual ocean carbon sink strength estimates over time. Chapter 3 examines how differences in representations of interior ocean C_{ant} and natural carbon influence patterns of amplified subsurface $p\text{CO}_2$ change using the same set of GOBMs, in addition to observation-based data products. Results indicate that GOBMs dissimilarly simulate subsurface C_{ant} -induced $p\text{CO}_2$ changes, particularly at the depth of maximum winter mixing, when these signals can re-emerge at the surface and bias estimates of the annual ocean carbon sink strength. This research contributes to ongoing international efforts to better constrain the global ocean carbon sink. Discrepancies between observation- and model-based estimates of the modern ocean carbon sink have grown over time, with across-model disagreements compounding in future climate projections. This points to an outstanding need to constrain sources of model discrepancies. This work helps to address this by clarifying: (1) a model's projected end-of-century ocean carbon sink magnitude is highly dependent on its post-spin-up seasonal and annual mean-state; (2) a more realistic representation of interior ocean carbon distributions and ecosystem processes is needed to achieve a more realistic representation of ocean carbon cycle change and the evolution of its feedbacks.

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The co-author A. J. Fassbender listed in this publication supervised the research which forms the basis of the dissertation. All listed co-authors provided feedback and reviewed the respective manuscript.

Introduction

The ocean serves as Earth's largest active reservoir of carbon, holding roughly 60 times more carbon than the atmosphere and exerting a dominant control over the global carbon cycle. Carbon is readily exchanged between the ocean and atmosphere primarily through fluxes of carbon dioxide (CO_2) gas, as the sea surface and overlying atmosphere continuously strive to reach equilibrium between their respective partial pressures of CO_2 ($p\text{CO}_2$) following Henry's Law, mediated by temperature, salinity, and pressure. Once dissolved, aqueous CO_2 reacts with seawater to form carbonic acid (H_2CO_3), a weak acid that dissociates in two steps to bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) ions, releasing hydrogen ions (H^+). This “peculiar buffer mechanism of seawater” (Revelle & Seuss, 1957) facilitates the molecular transformation of dissolved CO_2 into multiple carbon species and supports the ocean's large capacity to store dissolved inorganic carbon (DIC) with only modest changes in pH ($-\log_{10}(\text{H}^+)$).

Moving beyond equilibrium partitioning, interacting physical, chemical, and biological processes generate strong vertical gradients of DIC throughout the ocean (e.g., DeVries, 2022; Siegel et al., 2023). These processes, referred to as ocean carbon “pumps”, include the solubility pump and the biological carbon pumps, which together reduce DIC concentrations in the surface ocean and maintain higher concentrations at depth. The solubility pump enhances CO_2 uptake in cold, high-latitude surface waters, where dense, DIC-rich waters are transported into the interior ocean by large-scale circulation. The solubility pump accounts for roughly $\frac{1}{3}$ of the modern DIC gradient. The remaining $\frac{2}{3}$ is attributed to biological carbon pump processes, a collection of

processes by which DIC is converted into organic matter by phytoplankton productivity in the sunlit surface ocean and is then exported to depth, where it is subsequently consumed by heterotrophs and remineralized back into dissolved forms. These ocean carbon pumps work in tandem to sequester carbon from exchange with the atmosphere on timescales of months to millennia.

Since the Industrial Revolution, fossil fuel combustion and land-use change have driven a rapid rise in atmospheric CO₂ levels, greatly exceeding concentrations observed over the last 800,000 years (Lüthi et al., 2008). The ocean has absorbed approximately 25% of the fossil fuel emissions each year, amounting to nearly 40% of cumulative emissions to date (Friedlingstein et al., 2024), thereby modulating the pace of atmospheric CO₂ accumulation. However, the resulting accumulation of anthropogenic carbon (C_{ant}) in the upper ocean increases the concentration of DIC, manifesting as the rise in ocean $p\text{CO}_2$ and H^+ concentrations and declines in CO_3^- and pH. These chemical changes, collectively referred to as ocean acidification (OA), have widespread implications for the health of marine organisms and ecosystems (Doney et al., 2020). Moreover, C_{ant} accumulation progressively weakens the ocean's buffer capacity and increases the sensitivity of the carbonate system to natural physical, chemical, and biological processes (Riebesell et al., 2009). As OA progresses, a growing body of research is dedicated to understanding how anthropogenic perturbations are altering ocean carbon cycle feedbacks, in efforts to better predict the long-term rates and consequences of climate change.

The overarching goal of this dissertation is to elucidate how the ocean carbon

cycle is evolving in response to sustained anthropogenic perturbations, leveraging a combination of in situ observations, data synthesis products, and output from regional and global ocean models. More specifically, this work focuses on changes to the spatial and temporal variability in carbon cycling modulated by natural and anthropogenic carbon interactions. By moving beyond surface-only, annual mean-state perspectives, this research aims to provide a more comprehensive understanding of marine carbon cycle feedbacks by investigating interior ocean changes caused by C_{ant} and their connections to seasonal surface dynamics in a rapidly changing ocean. This dissertation consists of three research chapters and concludes with a synthesis chapter that summarizes the results and assesses the broader significance of the findings.

Chapter 1 quantifies changes in OA metrics caused by C_{ant} accumulation in the open North Pacific Ocean and coastal California Current Large Marine Ecosystem (CCLME) using ship-based observations. While previous studies have primarily focused on surface ocean changes in one or two OA metrics, this chapter expands upon those works by characterizing the vertical structure of C_{ant} -induced changes across a broader suite of metrics. Further, this chapter explores the implications of these changes on the habitability of marine organisms in the coastal zone. To assess modern conditions, output from a high-resolution regional ocean model of the CCLME is used to estimate the frequency, intensity, and duration of high- $p\text{CO}_2$ events ($>1000 \mu\text{atm}$) at the continental shelf break. This chapter is published in *Geophysical Research Letters*.

Chapter 2 evaluates the changes in surface ocean $p\text{CO}_2$ seasonal variability using simulations from seven global ocean biogeochemical models (GOBMs) recently

evaluated by the REgional Carbon Cycle Assessment and Processes Phase 2 (RECCAP2) project. This chapter quantifies how C_{ant} accumulation and climate change independently modify the thermal and biophysical drivers of $p\text{CO}_2$ seasonality over the historical period from 1980 to 2018. As the seasonal cycle is the dominant mode of air-sea CO_2 flux variability in most open ocean regions, this chapter emphasizes how seasonality biases inherent to GOBMs lead to substantial differences in cumulative annual ocean carbon uptake over time. This chapter has been submitted to *Global Biogeochemical Cycles*.

Chapter 3 builds on the previous two chapters by examining how GOBMs represent interior ocean $p\text{CO}_2$ changes caused by C_{ant} accumulation, in comparison to a data-based synthesis product. This chapter focuses on the vertical distributions of natural and anthropogenic carbon pools and how their interaction drives amplified nonlinear $p\text{CO}_2$ responses below the surface ocean. Across-model differences in C_{ant} -induced $p\text{CO}_2$ changes are assessed at the annual maximum mixed layer depth, which serves as an estimate of the subsurface signals that can re-emerge at the surface during winter mixing. Such re-emergence can modulate surface ocean $p\text{CO}_2$ conditions and potentially bias estimates of the annual ocean carbon sink. Trends at this depth are used to explore how interior ocean carbon cycle feedbacks may intensify in the near future as the ocean continues to absorb C_{ant} .

A final synthesis chapter summarizes the main findings and reflects on the broader implications.

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Chapter 1: Dissimilar sensitivities of ocean acidification metrics to anthropogenic carbon accumulation in the Central North Pacific Ocean and California Current Large Marine Ecosystem

Mar C. Arroyo, Andrea J. Fassbender, Brendan R. Carter, Christopher A. Edwards, Jerome Fiechter, Addie Norgaard, and Richard R. Feely

1.1 Abstract

We analyze and compare changes to ocean acidification metrics caused by anthropogenic carbon (C_{anth}) accumulation in the North Pacific Ocean and California Large Marine Ecosystem (CCLME). The greatest declines in pH and carbonate mineral saturation state occur near the surface, coincident with the highest C_{anth} concentrations. However, maximal increases in the partial pressure of carbon dioxide ($p\text{CO}_2$) and hydrogen ion concentration occur subsurface where C_{anth} values are lower. We attribute dissimilar sensitivities of these metrics to the background ocean chemistry, which has naturally high $p\text{CO}_2$ and low buffering capacity in subsurface waters due to accumulated byproducts of organic matter respiration, which interacts with C_{anth} . In the CCLME, rising subsurface $p\text{CO}_2$ has increased the frequency, duration, and intensity of hypercapnia ($p\text{CO}_2 \geq 1000 \mu\text{atm}$) on the continental shelf. Our findings suggest that hypercapnia induced by C_{anth} accumulation can co-occur with hypoxia in the CCLME and is an additional modern stressor for marine organisms.

1.2 Introduction

The ocean has absorbed ~25% of the carbon dioxide released to the atmosphere by anthropogenic activities since the preindustrial (PI) era, leading to measurable changes in ocean chemistry (Friedlingstein et al., 2022). Increases in surface ocean

carbon dioxide partial pressure ($p\text{CO}_2$) cause associated increases in hydrogen ion concentration ($[\text{H}^+]$) and declines in pH ($-\log_{10}([\text{H}^+])$) and carbonate ion concentration ($[\text{CO}_3^{2-}]$; Caldeira & Wickett, 2003; Feely et al., 2004, 2008). Such chemical changes due to anthropogenic carbon (C_{anth}) accumulation are collectively referred to as ocean acidification (OA) and have widespread implications for the health of marine organisms and ecosystems (Doney et al., 2020; Gattuso et al., 2015; Orr et al., 2005).

Two common metrics used to assess the progression and potential impact of OA on marine organisms are pH and the saturation state of calcium carbonate minerals, such as aragonite (Ω_{Ar}). Numerous prior works have shown that these metrics, and others, vary distinctly in their response to rising surface ocean $p\text{CO}_2$ (Bates et al., 2014; Cai et al., 2020; Feely et al., 2009; Jiang et al., 2019; Lauvset et al., 2015), due in part to differing sensitivities of each parameter to environmental conditions (Bittig et al., 2018; Egleston et al., 2010; Fassbender et al., 2017; Hagens & Middelburg, 2016; Riebesell et al., 2009). Fewer studies have considered how OA metrics evolve below the air-sea interface in response to carbon accumulation (Cai et al., 2011; Carter et al., 2019; Fassbender et al., 2021; Feely et al., 2018; Jiang et al., 2015; Lauvset et al., 2020; Resplandy et al., 2013; Ríos et al., 2015), and the findings suggest dissimilar carbonate system property perturbations with depth.

A less-emphasized OA metric of growing concern is the volume of water with in situ $p\text{CO}_2$ exceeding $1000 \mu\text{atm}$ (hypercapnia; McNeil & Sasse, 2016). Hypercapnia has been shown to negatively interfere with physiological and neurological functions in fishes and other marine organism by hindering organismal ability to excrete CO_2

during respiration and by disrupting internal acid-base regulation (Nilsson et al., 2012; Perry & Gilmour, 2006). Hypercapnic conditions can already be found in modern ocean environments, including upwelling regions like the California Current Large Marine Ecosystem (CCLME) in the Northeast Pacific Ocean (Feely et al., 2018). In this region, seasonal, wind-driven upwelling along the US West Coast transports subsurface waters, naturally low in pH, Ω_{Ar} , and dissolved oxygen (O_2) and elevated in pCO_2 due to prior organic matter remineralization, to the continental shelf environment (Feely et al., 2008; Gruber et al., 2012; Hauri et al., 2009, 2013). Subsurface remineralization inherently links high pCO_2 and low pH values to low O_2 values, which can be intensified on the continental shelf by locally-driven processes (Chan et al., 2017, 2019; Feely et al., 2016, 2018). During the upwelling season, certain coastal regions in the CCLME can experience severe hypoxia ($[O_2] \leq 60 \mu\text{mol kg}^{-1}$; Connolly et al., 2010; Grantham et al., 2004; Peterson et al., 2013). The potential expansion and intensification of hypercapnic zones in the CCLME with continued C_{anth} accumulation poses a risk to organisms already exposed to spatially nonuniform stressors of acidification and hypoxia (Cheresh & Fiechter, 2020; Feely et al., 2018; McNeil & Sasse, 2016), in combination with ocean warming (Kroeker et al., 2013).

In this study, we evaluate and compare changes in OA metrics caused by C_{anth} accumulation in the Central North Pacific Ocean (CNPO) and the CCLME since industrialization. Our study builds on the prior efforts of Feely et al. (2016) and Carter et al. (2019) to extend North Pacific Ocean C_{anth} estimates to the US West Coast. We focus on the vertical structure of C_{anth} -sensitivity for pH, Ω_{Ar} , pCO_2 , and $[H^+]$ and

consider the impact of subsurface changes on the habitable zones of marine organisms in the CCLME. We then use output from a regional ocean model to examine the frequency, intensity, and duration at which the modern-day shelf ecosystem experiences conditions of hypercapnia.

1.3 Methods

1.3.1 Cruise Data and Derived Carbonate System Parameters

We use observational data from the 2015 CLIVAR/GO-SHIP P16N Leg 2 cruise (Cross et al., 2017) in the CNPO and from the 2016 West Coast Ocean Acidification (WCOA2016) cruise (Alin et al., 2017; Figure 1.1) in the CCLME. Discrete measurements of the dissolved inorganic carbon (DIC), total alkalinity (TA), O₂, and inorganic nutrients (phosphate and silicate) were collected from each station, in addition to vertical conductivity-temperature-depth profiles. P16N stations between 22.5°N and 56°N and WCOA transects between 25°N and 54°N are used herein. DIC and TA concentrations were measured on board each cruise by coulometric titration (Johnson et al., 1993) and open-cell potentiometric titration (Millero et al., 1993), respectively, following standard procedures (Dickson et al., 2007). Routine analysis of Certified Reference Materials (Dickson et al., 2003) and replicate samples ensured DIC and TA uncertainties were better than $\pm 0.1\%$. O₂ concentration was determined by a modified Winkler titration (Carpenter, 1965) with an uncertainty of $\pm 1.4 \mu\text{mol kg}^{-1}$.

Additional carbonate system parameters were calculated from DIC, TA, temperature, salinity, phosphate, and silicate using CO2SYSv1.1 for MATLAB (van Heuven et al., 2011; Lewis & Wallace, 1998) with the dissociation constants for

carbonic acid by Lueker et al. (2000), bisulfate ion by Dickson et al. (1990), and total boron by Uppström (1974). Calculated parameters include in situ pH on the total scale, Ω_{Ar} , pCO_2 , $[H^+]$, and Revelle Factor (RF).

1.3.2 Estimating C_{anth} in the CNPO and CCLME

C_{anth} distributions were estimated in the CCLME using a variant on the approach described by Carter et al. (2019). This method utilizes ensembles of extended Multiple Linear Regressions (eMLR) from fourteen repeat hydrographic sections in the Pacific Ocean between 1991 and 2017 to estimate decadal C_{anth} accumulation along these various sections, including those in the CCLME. These estimates were then added to the 1994 C_{anth} estimates determined from the ΔC^* technique (Sabine et al., 2004), as regridded from the Global Ocean Data Analysis Project (GLODAP; Key et al., 2004), to yield modern C_{anth} values. Finally, the C_{anth} estimates are mapped temporally and spatially within the Pacific Ocean using regressions based on in situ temperature, salinity, and year. In contrast to the Carter et al. (2019) approach to map C_{anth} across the entire Pacific basin, depth was not used as a regression predictor for the US West Coast C_{anth} reconstructions due to seasonal coastal upwelling that links seawater properties more closely to density than to depth. We estimate an uncertainty of $\sim 8 \mu\text{mol kg}^{-1}$ for C_{anth} in the CCLME (Text S1.1).

C_{anth} estimates for the CNPO during the 2015 P16N cruise were obtained directly from the gridded product by Carter et al. (2017) and linearly interpolated onto the discrete sampling positions. Gridded C_{anth} values at 25m were extrapolated to the surface. Unlike the approach used for the CCLME more broadly, this approach assumes

that surface C_{anth} keeps up with the expectations from complete equilibrium with atmospheric changes. Note that the C_{anth} estimated in the CCLME is offset from values estimated in the CNPO by 1 year due to different cruise years.

1.3.3 Determining the Impact of C_{anth} Accumulation

C_{anth} estimates were subtracted from modern, observed DIC concentrations to yield a PI DIC estimate. PI estimates of pH, Ω_{Ar} , $p\text{CO}_2$, and $[\text{H}^+]$ were computed from PI DIC and modern TA using CO2SYS as described above. The impacts of C_{anth} accumulation on the carbonate system were evaluated by computing the change of pH, Ω_{Ar} , $p\text{CO}_2$, and $[\text{H}^+]$ between the modern and PI values (e.g., $\Delta\text{pH} = \text{pH}_{\text{modern}} - \text{pH}_{\text{PI}}$). This method only accounts for the influence of C_{anth} and does not consider physically or biologically induced changes in ocean biogeochemistry or changes in heat and freshwater fluxes since industrialization.

1.3.4 Tracking Hypercapnic Events in the CCLME with a Regional Ocean Model

Observations in the CCLME provide a snapshot of the spatial extent of hypercapnia ($p\text{CO}_2 \geq 1000 \mu\text{atm}$). To estimate the modern spatiotemporal variability of hypercapnic events in the CCLME, we use a near real-time (NRT), 4-dimensional variational data assimilative model of the CCLME implemented using the Region Ocean Modelling System (ROMS; Moore, et al., 2011ab; 2013). The model domain extends from 30°N–48°N and 134°W–115.5°W (Figure 1.1b) and has a horizontal resolution of 1/10° with 42 terrain-following vertical layers (Figure S1.1a; Veneziani et al., 2009). The model reproduced the observed density field along each CCLME Line

with high fidelity ($r \geq 0.81$; Figure S1.2) and was used to explore conditions over 10 simulation years (2011-2020).

The subsurface upper boundary for hypercapnic water in the observations is aligned with an average density of $1027.5 \pm 0.7 \text{ kg m}^{-3}$ (Figure S1.3). The depth of this density surface was tracked daily over the 10-year model period to estimate the frequency, intensity, and duration of hypercapnic events at the 200 m isobath (roughly the continental shelf break; Figure S1.1; Text S1.2). If this density surface shoals onto the continental shelf during seasonal upwelling events, or if respiration processes increase $p\text{CO}_2$ in the local subsurface waters, nearshore communities could be at risk for hypercapnia. Event frequency is defined as the number of days in a year that the hypercapnic density surface is shallower than 200 m. Event duration is defined as the number of consecutive days that the hypercapnic density surface is shallower than 200 m. Event intensity is defined as the local volume of hypercapnic water above 200 m. Events within 0.5° of latitude of the model boundaries were neglected to account for biases attributed to boundary conditions (Text S1.2).

1.4 Results and Discussion

1.4.1 Anthropogenic Changes in CNPO and CCLME Carbonate Chemistry

Depth-resolved changes in carbonate system parameters caused by C_{anth} accumulation in the CNPO are shown in Figure 1.1 (left). The highest C_{anth} values of $\sim 58 \pm 8 \text{ } \mu\text{mol kg}^{-1}$ are found at the surface where C_{anth} is absorbed, with peaks in the subtropics where waters are most efficient at absorbing C_{anth} (Sabine et al., 2004). In the high latitudes, absorbed C_{anth} gets transported to the ocean interior for storage,

primarily in the midlatitude thermocline, with modern concentrations of $\sim 50 \mu\text{mol kg}^{-1}$ (Carter et al., 2019; Gruber et al., 2019; Khatiwala et al., 2013).

C_{anth} accumulation in the ocean leads to the formation of carbonic acid, a weak acid that dissociates and is partially neutralized by reaction with $[\text{CO}_3^{2-}]$, producing $[\text{H}^+]$ and bicarbonate. The effects of these chemical processes manifest as reductions in pH and Ω_{Ar} , predominantly in the near-surface ocean where C_{anth} values are greatest. As a result, the patterns of ΔpH and $\Delta\Omega_{\text{Ar}}$ are similar to the distribution of C_{anth} , with near-surface (upper 30m) values ranging from -0.125 to -0.095 and from -0.70 to -0.40, respectively, in the CNPO (Figures 1.1b and 1.1d). Similar upper ocean declines of pH and Ω_{Ar} due to C_{anth} accumulation were found in the coastal CCLME (Figure 1.1; right). Surface ocean C_{anth} estimates in the CCLME ranged from 45 to $75 \mu\text{mol kg}^{-1}$ in 2016, with the highest values observed south of 30°N (Figure S1.4). Lower surface C_{anth} values were found adjacent to the coast near Cape Mendocino (40°N) and Pt. St. George (42°N), as seen in 2013 by Feely et al. (2016). This heterogeneity in surface C_{anth} values, which is also found in the vertical C_{anth} gradient, can be explained by seasonal, nonuniform coastal upwelling in the CCLME that lifts subsurface water carrying lower C_{anth} to shallower depths along the coast. In the CCLME, near-surface (upper 30 m) values of ΔpH and $\Delta\Omega_{\text{Ar}}$ ranged between -0.160 to -0.116 and -0.72 to -0.33, respectively, along Line 11 (Figures 1.1c and 1.1e), which is within the ranges found for the entire CCLME domain (ΔpH : -0.167 to -0.095; $\Delta\Omega_{\text{Ar}}$: -0.74 to -0.29).

Unlike $\Delta\Omega_{\text{Ar}}$, the maximum change in pH is not always at the surface, but sometimes in the shallow subsurface in both the open (~ 200 m; Carter et al., 2019) and

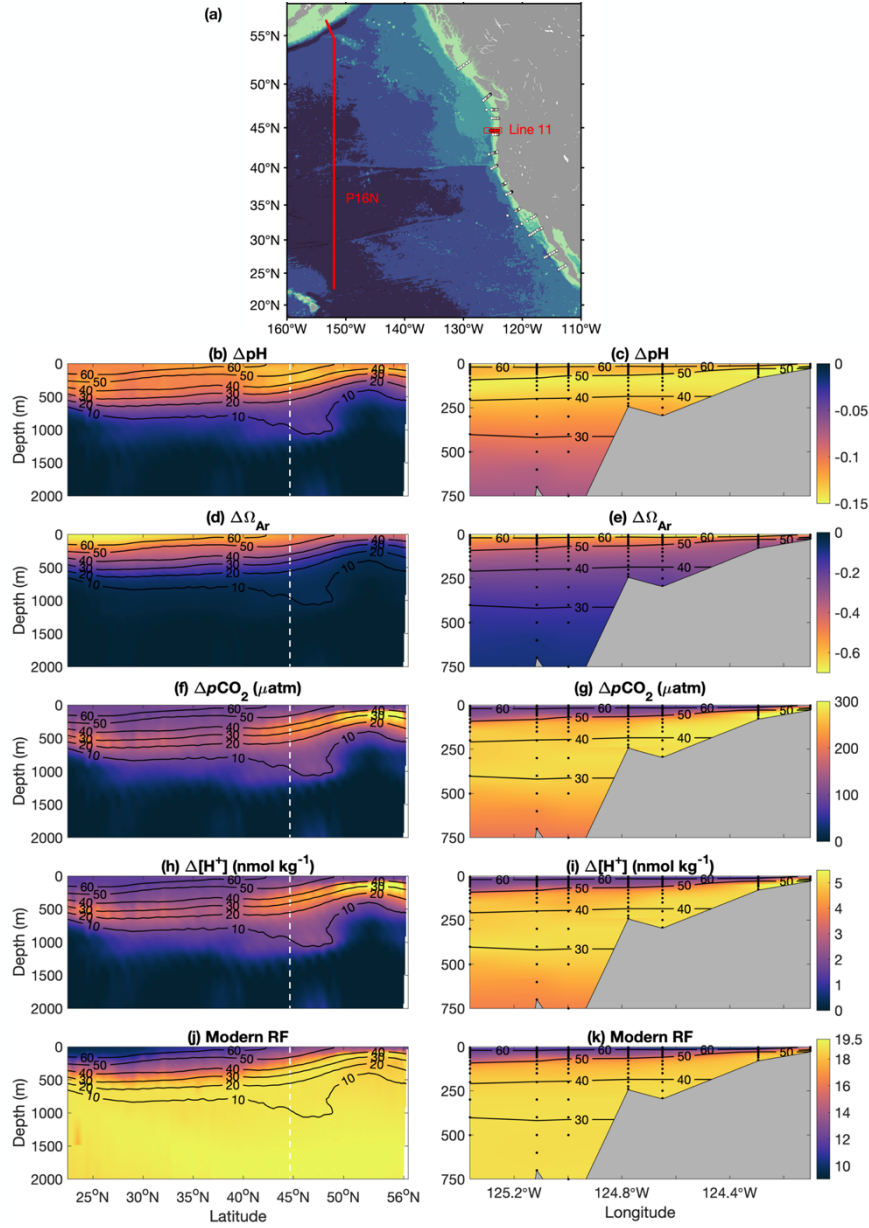


Figure 1.1. Changes (Δ) caused by C_{anth} ($\mu\text{mol kg}^{-1}$; contours) accumulation along (a; map) $\sim 152^\circ\text{N}$ in the CNPO (left) and along Line 11 ($\sim 45^\circ\text{N}$) in the CCLME (right) for (b, c) pH, (d, e) Ω_{Ar} , (f, g) $p\text{CO}_2$, and (h, i) $[\text{H}^+]$. Panels (j) and (k) show modern RFs. Dashed white lines in the left panels mark the CNPO data at 45°N shown in Figure 1.2.

coastal (~ 100 m) oceans. This result agrees with findings of Lauvset et al. (2020) who performed a similar analysis for interior ΔpH and $\Delta\Omega_{\text{Ar}}$ using the GLODAPv2 gridded climatology with C_{anth} values for 2002 (Lauvset et al., 2016). A larger subsurface ΔpH

reflects the enhanced sensitivity of pH to changes in C_{anth} (i.e., DIC) at depth, where the ratio of DIC to TA is higher than that at the surface (see Figure 7 of Lauvset et al., 2020). However, these larger changes in shallow, subsurface pH are of very similar magnitude to surface pH changes.

The vertical patterns of $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ are distinct from those exhibited by ΔpH and $\Delta\Omega_{\text{Ar}}$ (Figure 1.1 f-i). The largest changes in $p\text{CO}_2$ and $[\text{H}^+]$ in the open ocean typically occur well below the surface (~200–600 m) coincident with lower C_{anth} (30–45 $\mu\text{mol kg}^{-1}$), a feature that extends to the subsurface CCLME (~240–400 m). The greatest $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ along ~152°W exceed 275 μatm and 4.5 nmol kg^{-1} , respectively, in the CNPO (See Figure 4 of Fassbender et al., 2021). Lower maxima values of 175 μatm and 3.5 nmol kg^{-1} are found in the northern mid-latitudes (20–40°N). In the CCLME along Line 11, increases in $p\text{CO}_2$ and $[\text{H}^+]$ ranged between 70–200 μatm and 1.3–3.5 nmol kg^{-1} , respectively, within the upper 80 m where $C_{\text{anth}} \geq 50$ $\mu\text{mol kg}^{-1}$ (excluding the station closest to shore). On this transect, maximal values for $\Delta p\text{CO}_2$ of ~250 μatm and for $\Delta[\text{H}^+]$ of 5.2 nmol kg^{-1} were found below this depth (~240 m; excluding the station closest to shore). These observed subsurface $p\text{CO}_2$ changes are larger than the atmospheric $p\text{CO}_2$ increase caused by human activities (from ~280 μatm to ~420 μatm = ~140 μatm). In both the open and coastal oceans, the subsurface maximum in $\Delta[\text{H}^+]$ is not coincident with a subsurface maximum in ΔpH . This is because ΔpH reflects a relative, rather than absolute, change in $[\text{H}^+]$ and thus depends on the initial $[\text{H}^+]$ conditions (Fassbender et al., 2021).

The reason that $p\text{CO}_2$ and $[\text{H}^+]$ are more sensitive to smaller, subsurface C_{anth}

changes in the open and coastal North Pacific is related to the background chemistry of these waters. The ocean's ability to buffer the chemical changes imposed by C_{anth} addition is often quantified using the Revelle sensitivity Factor (Broecker et al., 1979; Egleston et al., 2010; Fassbender et al., 2017; Middelburg et al., 2020; Revelle & Suess, 1957; Sabine et al., 2004). RF is defined as the ratio between a fractional change in pCO_2 and a fractional change in DIC for a given carbonate system perturbation, assuming constant alkalinity:

$$RF = \frac{\partial pCO_2 / pCO_2}{\partial DIC / DIC} \quad \text{Eq 1.}$$

A low RF value indicates a larger relative DIC change will occur for a given, relative pCO_2 perturbation, and signifies a greater capacity of the ocean to absorb anthropogenic CO_2 from the atmosphere. In contrast, waters with high RF values will be less efficient at absorbing anthropogenic atmospheric CO_2 . Under the same construct, this also means that waters with different RF values that experience the same relative DIC perturbation will exhibit different relative pCO_2 changes (Fassbender et al., 2017, 2018).

Large gradients in RF have been identified across the global surface ocean (Bittig et al., 2018; Fassbender et al., 2017; Jiang et al., 2019; Sabine et al., 2004). Surface RFs in the CNPO are low in the warm tropics (~ 10) and increase toward the cold poles (~ 16), reflecting regional variability in carbon uptake efficiency (Figure 1.1j). RF values similarly increase with depth and toward the poles below the main thermocline, reaching a maximum of ~ 19 below 1000 m in the CNPO. These high subsurface RF values of 18 to 19 extend to the coastal CCLME (Figure 1.1k). High RF

values are analogous to higher ratios of DIC to TA (Broecker et al., 1979; Egleston et al., 2010), as there is a relatively lower proportion of $[\text{CO}_3^{2-}]$ and higher proportion of aqueous CO_2 among the species in the DIC pool. However, waters with similar RF values may exhibit dissimilar $p\text{CO}_2$ responses to identical relative changes in DIC. This is because the magnitude of $\Delta p\text{CO}_2$ for a given increase in C_{anth} also depends on the background $p\text{CO}_2$ level, which can vary significantly depending on the aqueous CO_2 fraction of DIC (Fassbender et al., 2017; Feely et al., 2018).

In the context of C_{anth} accumulation in the CNPO, the largest absolute changes in $p\text{CO}_2$ since industrialization are found between 200 and 600 m, despite lower C_{anth} values (20–30 $\mu\text{mol kg}^{-1}$) at these depths relative to the surface. Within this depth range, the CNPO has naturally elevated subsurface RF and $p\text{CO}_2$ values due to accumulated byproducts of organic matter remineralization, reflected by substantial O_2 utilization (Figure S1.5). Background RF and $p\text{CO}_2$ values remain high below this depth range; however, the magnitude of $\Delta p\text{CO}_2$ decreases with depth in concert with declining C_{anth} . Like the open CNPO, CCLME subsurface waters are naturally elevated in RF and $p\text{CO}_2$ due to net respiration (Figure S1.6), and $\Delta p\text{CO}_2$ values exhibit similar subsurface maxima. These findings suggest that opposing vertical gradients in the accumulation of C_{anth} and respiration byproducts create a particular depth range in which the $p\text{CO}_2$ sensitivity to DIC is elevated and the C_{anth} concentration is moderate, causing a subsurface $\Delta p\text{CO}_2$ maximum in the open and coastal North Pacific. Since the $[\text{H}^+]$ and $p\text{CO}_2$ are tightly linked parameters, the patterns of $\Delta[\text{H}^+]$ are like those of $\Delta p\text{CO}_2$ (Figure 1.2).

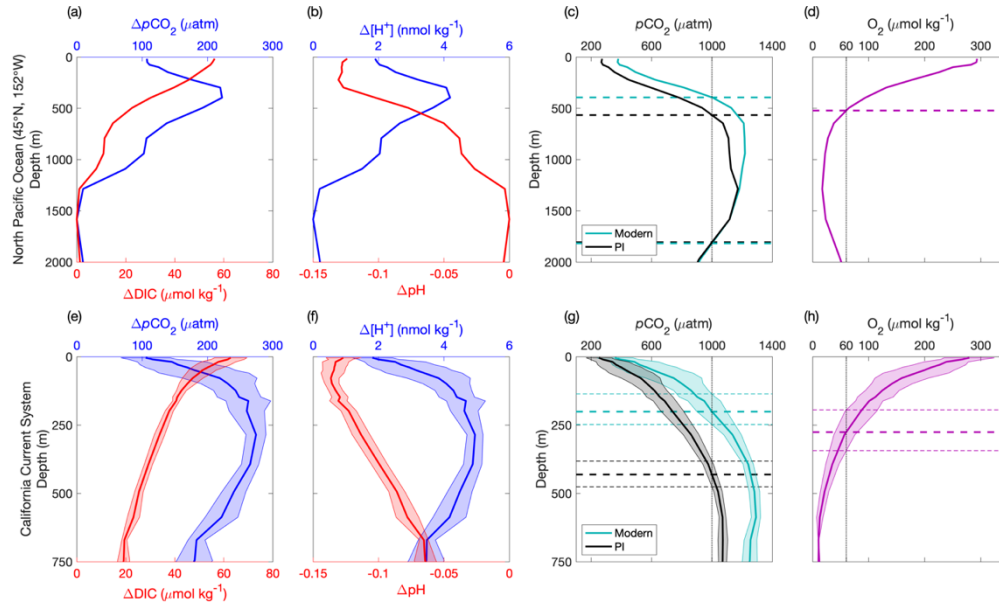


Figure 1.2. Profiles from the (top) upper 2000m in the CNPO at 45°N, 152°W and (bottom) upper 750m in the CCLME. The bold lines in lower panels indicates the average profile from all CCLME stations and shading represents ± 1 standard deviation. The changes (Δ) due to C_{anth} accumulation are shown for (a, e) $p\text{CO}_2$ and DIC and (b, f) $[\text{H}^+]$ and pH. (c, g). Modern (teal) and preindustrial (black) $p\text{CO}_2$ and corresponding depth ranges (dashed lines) for hypercapnia ($p\text{CO}_2 \geq 1000 \mu\text{atm}$, vertical line). (d, h) Modern O_2 and corresponding depth range (dashed lines) for hypoxia ($[\text{O}_2] \leq 60 \mu\text{mol kg}^{-1}$, vertical line).

The patterns of chemical change in OA metrics are notably coherent in the CCLME, with similar magnitudes of subsurface $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ increases throughout the domain (Figure 1.2 e-f). Compared to the open CNPO at 45°N, maxima in $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ in the CCLME appear shallower (~ 280 vs. ~ 400 m) and larger in magnitude ($\Delta p\text{CO}_2$: 270 vs. 220 μatm ; $\Delta[\text{H}^+]$: 4.9 vs. 3.8 nmol kg^{-1} ; Figure 1.2). The average C_{anth} values at the depths of maximal $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ in the open ocean and CCLME are similar (30–34 $\mu\text{mol kg}^{-1}$); however, the associated density surfaces carrying elevated background RF and $p\text{CO}_2$ are shallower nearshore due to the natural ocean physics (Figure S1.6). Though regional differences between the depth and magnitude of $\Delta p\text{CO}_2$ and $\Delta[\text{H}^+]$ maxima are largely the result of circulation and upwelling, local processes in the CCLME may intensify the subsurface chemical

response. Enhanced local organic matter remineralization caused by high levels of overlying primary production and a benthic boundary layer preventing export to depth can further increase subsurface $p\text{CO}_2$ on the continental shelf (Fassbender et al., 2011), leading to more pronounced responses to subsurface C_{anth} accumulation (Figure 1.3; Figure S1.7).

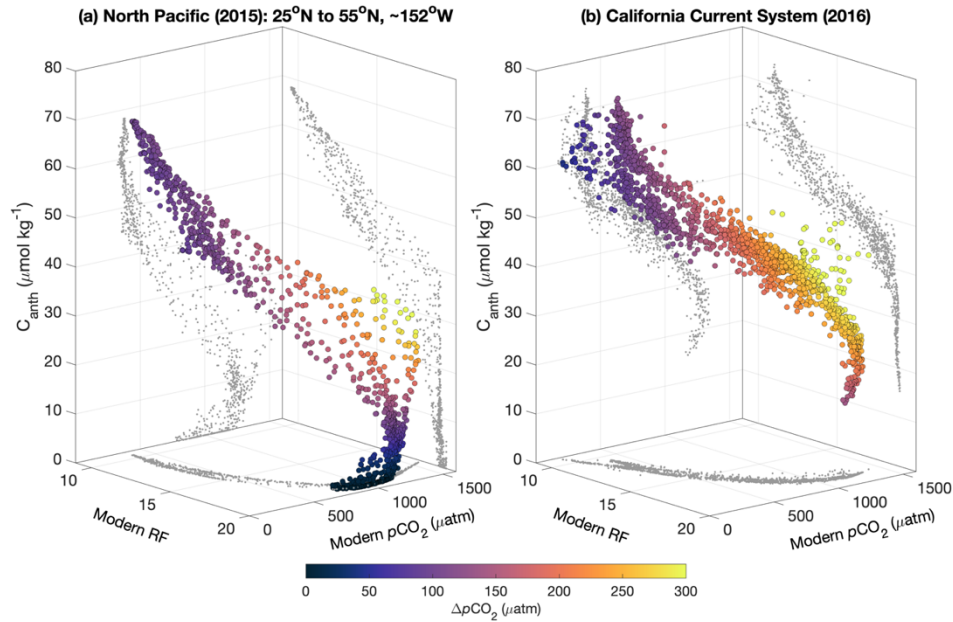


Figure 1.3. The change in $p\text{CO}_2$ ($\Delta p\text{CO}_2$) due to C_{anth} accumulation in relationship to modern RF and $p\text{CO}_2$ in the (a) CNPO between 25°N and 55°N, ~152°W from P16N and (b) the CCLME from WCOA2016.

1.4.2 Expansion of the CCLME Hypercapnic Domain due to C_{anth}

Accumulation

The changes in seawater chemistry due to C_{anth} accumulation have driven an increase in hypercapnic volume in the CNPO and CCLME. The minimum hypercapnic depth shoaled by 175 m (from 565 m to 390 m) in the open ocean at 45°N, 152°W (Figure 1.2c) and by 185 to 300 m (from 435 m to 200 m on average) in the CCLME

(Figure 1.2g) compared to the PI mean minimum hypercapnic depths. This shoaling in the coastal region corresponds to a ~73% average increase (range: 58–94%) in the hypercapnic volume above 750 m, creating a modern coastal environment where 64% to 100% of the hypercapnic water above 750 m is simultaneously hypoxic (Figure 1.2h). This expansion in the CCLME presently leaves, on average, ~27% of the water column above 750 m as suitable habitat for marine organisms susceptible to hypercapnia.

By tracking the shallowest density layer on which hypercapnia was observed during WCOA2016 in model simulations, we find that hypercapnic events at the 200 m isobath occur throughout most of the domain between 2011 and 2020 (Figure 1.4). On average, events are most frequent in the northern region (north of Cape Mendocino), where hypercapnic conditions may occur up to 180 days per year (Figure 1.4a) and individual events may last for 3–4 months (Figure 1.4b). During intense events, up to 40% of the local water column (120–200 m) may experience hypercapnic conditions (Figure 1.4c). In contrast, events are less frequent and of shorter duration in the southern CCLME (south of Point Conception), occurring less than 20 days per year and lasting up to 3 days, on average. However, these short-lived events can still be intense, with up to 25% of the water column being hypercapnic (150–200 m). Less intense (<10% of the water column) and relatively short-lived (<5 days) events occur, on average, in the central region (between Cape Mendocino and Point Conception), where hypercapnia was experienced for up to 40 days per year.

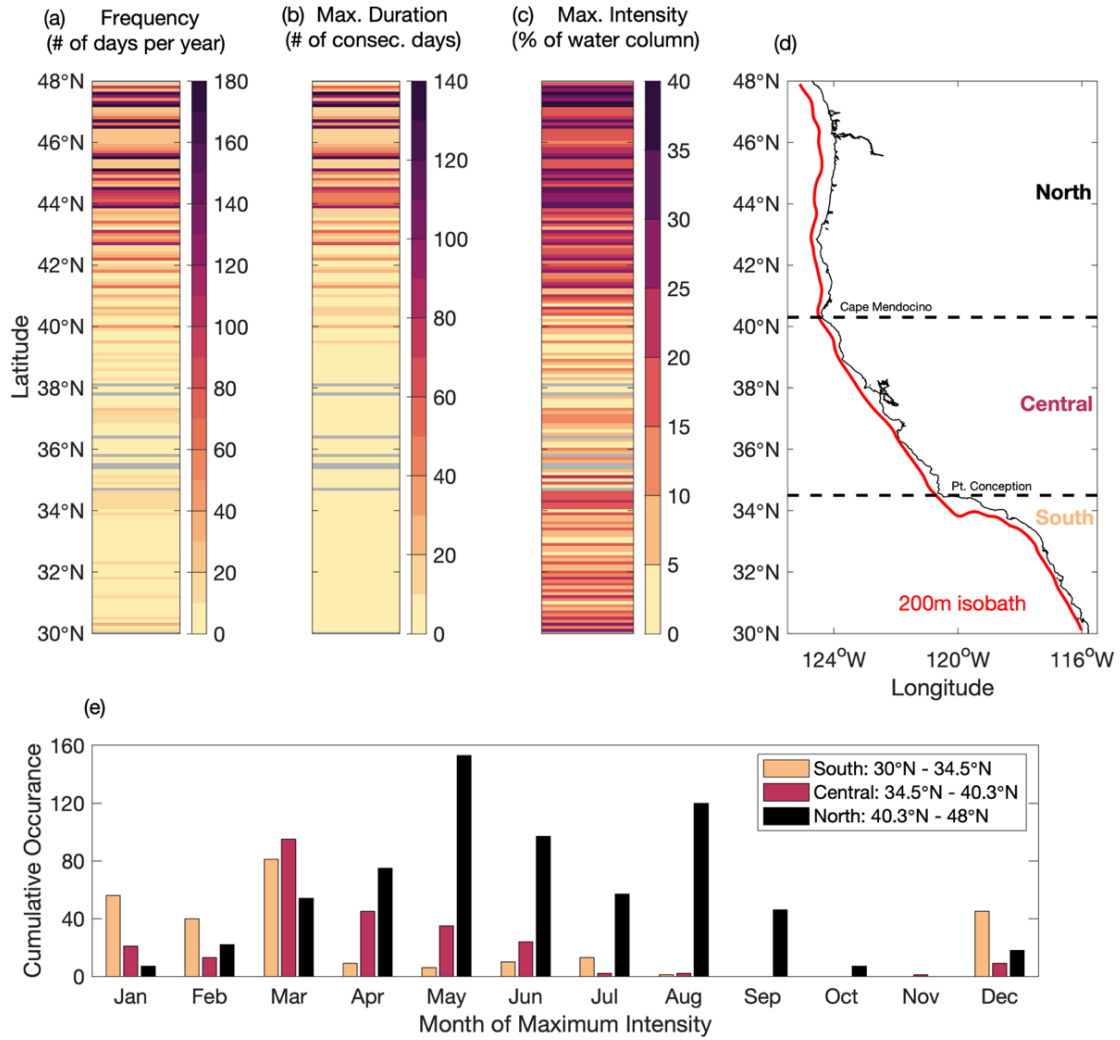


Figure 1.4. Hypercapnic events at the 200m isobath in the CCLME estimated from daily near real-time model physics. Metrics of (a) frequency, (b) maximum duration, and (c) maximum intensity were averaged from 2011 to 2020. Gray indicates no hypercapnic event occurred at the 200m isobath between 2011 and 2020. (d) Map of the 200m isobath (red) and CCLME region delineation following that by Checkley & Barth (2009): southern (30°N – 34.5°N); central (34.5°N – 40.3°N); and northern (40.3°N – 48°N). (e) Cumulative month of maximum intensity between 2011 and 2020 for each 0.1° of latitude.

The timing of the most intense hypercapnic events in the CCLME may be influenced by seasonal variations of the hypercapnic density surface depth in response to the alongshore pressure gradient, modulated by wind stress forcing (Figure 1.4e; Figures S1.10 to S1.14). Between 2011 and 2020, the month of maximum hypercapnia intensity was tracked at each model grid along the 200 m isobath ($n = 1,112$; Figure

1.4e). During winter and early spring (December–March), the hypercapnic density surface may reside above the 200 m isobath in certain locations, preconditioning the local water column with hypercapnic water at depth (Figures S1.10 to S1.12). During these months in the central and southern domains, intermittent upwelling events induced by weak equatorward winds (or the relaxation of downwelling-favorable winds; Figures S1.13 and S1.14), generated the most intense hypercapnic events. In contrast, downwelling-favorable alongshore wind stress during the winter months in the northern domain may suppress hypercapnic events at the 200 m isobath. In this northern region, the most intense hypercapnic events occurred in March–August, when winds shift predominantly equatorward along the coast and uplift subsurface isopycnals to shallower depths. The resulting alongshore pressure gradient in response to upwelling favorable conditions progressively deepened the isopycnal structure at the 200 m isobath from north to south in the central and southern domains, suppressing the occurrence of hypercapnic events at the 200 m isobath in spring and summer.

The metrics describing hypercapnic events in the CCLME consider high $p\text{CO}_2$ waters delivered only from offshore, physical sources by determining the shallowest density layer on which hypercapnia is observed. Though this density typically was not found inshore of the 200 m isobath in CCLME NRT simulations, hypercapnia was observed inshore of the 200 m isobath during WCOA2016 (Figure S1.8). Assuming that the data-assimilative model simulations capture the physical dynamics of upwelling, this suggests that observed nearshore hypercapnia is further intensified by

local organic matter respiration on the continental shelf that contributes to elevated subsurface $p\text{CO}_2$ (Fassbender et al., 2011; Feely et al., 2018; Figure S1.7).

1.5 Conclusion

While many studies focus on changes in pH and Ω_{Ar} to assess the progression of OA, our findings illustrate that these and other OA metrics (i.e., $p\text{CO}_2$ and $[\text{H}^+]$) display different vertical sensitivities to C_{anth} accumulation within the water column. The CNPO and CCLME are particularly chemically sensitive to anthropogenic perturbations at depth, leading to the quantifiable intensification of subsurface $p\text{CO}_2$ and $[\text{H}^+]$ levels. The enhanced sensitivities are due to the age of the waters at mid-depths and the associated accumulation of byproducts from organic matter remineralization, which elevate subsurface $p\text{CO}_2$ and reduce the ocean buffering capacity. Subsurface $p\text{CO}_2$ intensification is relevant to the CCLME where pervasive hypercapnia may be introduced from offshore sources and subsequently enhanced by local remineralization processes. We find that the minimum hypercapnic depth has shoaled by an average of 73% (to 200 m) in the upper 750 m of the CCLME since the PI era between 20°N and 50°N. Hypercapnia is predicted to increase in the CCLME (Feely et al., 2018) and globally (McNeil & Sasse, 2016) with continued C_{anth} accumulation, and thus will become an increasingly important stressor to susceptible pelagic and benthic communities. Consideration of multiple OA metrics, including $p\text{CO}_2$ and $[\text{H}^+]$, supports a more complete characterization of the extent of subsurface anthropogenic chemical change.

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1.7 Supplementary Information

Text S1.1: Anthropogenic Carbon Estimates in the California Current Large Marine Ecosystem

The CCLME anthropogenic carbon (C_{anth}) estimates reported in this study are an update to those in the upper 200 m by Feely et al. (2016). We employ the approach by Carter et al. (2019) – which was focused on basin-scale estimates – with mapping modifications for the coastal environment. Our approach maps reconstructed C_{anth} estimates in the Pacific Ocean by Carter et al. (2019) to the location (longitude, latitude, and depth) of interest in the CCLME using regressions with in situ ocean temperature, salinity, and year as predictors. In contrast to mapping C_{anth} in the open ocean as done by Carter et al. (2019), depth is excluded as a predictor for mapping in the CCLME as seawater properties are more closely related to density (with temperature and salinity collectively serving as a proxy for this property) than water column depth.

This approach represents an improvement over the approach used previously by Feely et al. (2016) due to the larger quantities of C_{anth} estimates used to fit the relationships used to quantify C_{anth} : Feely et al. (2016) fit 2nd order polynomials to C_{anth} estimates from a single zonal (P02) and a single meridional (P16) section intersecting the North American Coast within the CCLME and the Gulf of Alaska, respectively, whereas the new approach uses these sections alongside many additional sections and directly incorporates anthropogenic change estimates obtained from the 2007 to 2016 WCOA cruise occupations (Carter et al., 2019). Despite the change in the methods, the new approach does not yield significantly different C_{anth} estimates on average (Table S1.1) than either Feely et al.'s (2016) polynomial approach or a direct application of the unmodified regressions outlined by Carter et al. (2019). All approaches seem comparable to within the stated standard uncertainties of $\sim 8 \mu\text{mol } C_{\text{anth}} \text{ kg}^{-1}$. Depth-dependent uncertainties in the anthropogenic changes in pH, Ω_{Ar} , $p\text{CO}_2$ and $[\text{H}^+]$ are shown in Figure S1.9. These uncertainties are calculated by first propagating the $\pm 8 \mu\text{mol kg}^{-1} C_{\text{anth}}$ uncertainty in DIC through CO2SYS to estimate the uncertainties in preindustrial pH, Ω_{Ar} , $p\text{CO}_2$ and $[\text{H}^+]$. We then compute the difference between the modern, observed parameter and the preindustrial estimated values calculated with and without the $\pm 8 \mu\text{mol kg}^{-1} C_{\text{anth}}$ uncertainty in DIC. The uncertainties in the anthropogenic changes in pH, Ω_{Ar} , $p\text{CO}_2$ and $[\text{H}^+]$ are reported using the root-mean-square (RMS) difference between values of all parameters calculated normally (with no C_{anth} uncertainty) and after adding the $\pm 8 \mu\text{mol kg}^{-1} C_{\text{anth}}$ uncertainty, with the standard deviation of the mean profile each parameter calculated.

Table S1.1. Average and root mean squared (RMS) differences between the C_{anth} estimation approach used in this study (using WCOA 2016 data) and the approaches used by Feely et al. (2016) polynomial and by the unmodified Carter et al. (2019) for WCOA cruises. (“This study” minus the indicated method, all units are $\mu\text{mol kg}^{-1}$)

Dataset	Average vs. Feely et al. 2016	RMS vs. Feely et al. 2016	Average vs. Carter et al. 2019	RMS vs. Carter et al. 2019
WCOA 2007	-1.3	4.9	-5.6	6.6
WCOA 2011	-3.2	6.1	-5.6	6.3
WCOA 2012	-3.9	6	-5.5	6.2
WCOA 2013	-4.1	6.4	-6.7	7.4
WCOA 2016	-4.9	7.3	-6.1	6.9

Table S1.2. The root-mean-squared (RMS) difference between the anthropogenic changes (Δ) in pH, Ω_{Ar} , $p\text{CO}_2$, and $[\text{H}^+]$ calculated with and without the $\pm 8 \mu\text{mol kg}^{-1}$ uncertainty in C_{anth} and the standard deviation of the mean profile of parameters with and assuming no C_{anth} uncertainty in the upper 750 m.

Parameter	RMS vs. $-8 \mu\text{mol kg}^{-1}$ C_{anth}	RMS vs. $+8 \mu\text{mol kg}^{-1}$ C_{anth}
ΔpH	0.021	0.021
$\Delta\Omega_{\text{Ar}}$	0.067	0.068
$\Delta p\text{CO}_2$	37 μatm	34 μatm
$\Delta[\text{H}^+]$	0.66 nmol kg^{-1}	0.62 nmol kg^{-1}

Parameter	Standard Deviation ($-8 \mu\text{mol kg}^{-1} C_{\text{anth}}$)	Standard Deviation ($+8 \mu\text{mol kg}^{-1} C_{\text{anth}}$)	Standard Deviation (no C_{anth} uncertainty)
ΔpH	0.020	0.017	0.018
$\Delta\Omega_{\text{Ar}}$	0.157	0.181	0.169
$\Delta p\text{CO}_2$	55 μatm	85 μatm	70 μatm
$\Delta[\text{H}^+]$	0.99 nmol kg^{-1}	1.55 nmol kg^{-1}	1.26 mol kg^{-1}

normally and with the $\pm 8 \mu\text{mol kg}^{-1} C_{\text{anth}}$ uncertainty across all depths in the upper 750 m as a point of reference (Table S1.2; Figure S1.9). When the $\pm 8 \mu\text{mol kg}^{-1}$ uncertainty in individual C_{anth} estimates is propagated through calculations, the depth-dependent uncertainties in ΔpH , $\Delta\Omega_{\text{Ar}}$, $\Delta p\text{CO}_2$, and $\Delta[\text{H}^+]$ do not change the primary findings of

this study (Figure S1.9). However, the corresponding uncertainty in $\Delta p\text{CO}_2$ would influence the volume of water deemed hypercapnic.

Text S1.2 Tracking hypercapnic events in the CCLME

In this study, we define hypercapnic conditions in waters with in situ $p\text{CO}_2$ values greater than or equal to 1000 μatm , following thresholds set by the prior studies of McNeil & Sasse (2016) and Feely et al., (2018). However, many other definitions for the hypercapnic thresholds exist in current literature. For example, Esbaugh et al. (2012) found hypercapnia effects on the acid-base balance of adult Gulf toadfish with $p\text{CO}_2$ levels as low as 750 μatm . Other studies find detrimental effects occur in fishes when $p\text{CO}_2$ values exceed a concentration threshold over a certain exposure time. Nilsson et al. (2012) found loss of behavior lateralization (e.g., preferred direction of turning) occurred in larval coral reef fishes exposed to $p\text{CO}_2$ levels of ~ 900 μatm for 4 days. Though the exact definition of hypercapnia may vary, a review by Heuer & Grosell (2014) discuss that many sublethal effects on marine fishes may occur when $p\text{CO}_2$ values range from 500 to 25,000+ μatm at different life stages, suggesting that 1000 μatm is a realistic (while somewhat arbitrary) threshold to contextualize ecological harm. These effects of elevated $p\text{CO}_2$ levels include impacts to neurosensory behavior (e.g., activity levels) as well as the growth and development of certain species (see their Figure 2). We acknowledge that our definition of hypercapnia ($p\text{CO}_2 \geq 1000$ μatm) may not apply to all marine organisms. A key point of our study is to emphasize that the largest C_{anth} -driven changes in $p\text{CO}_2$ (and $[\text{H}^+]$) occur well below the surface in the mesopelagic, while the largest C_{anth} -driven changes in pH and Ω_{Ar} are more

localized to the upper ~150 m of the water column in the CCLME. We build off previous research from this region to discuss hypercapnia as an additional modern stressor to mid-water organisms. Additionally, the mesopelagic zone is where dissolved oxygen levels are naturally low, leading to the compounding effect of low dissolved oxygen and high $p\text{CO}_2$ that make it difficult for organisms to breathe and respire.

To estimate hypercapnic events in the CCLME, we tracked the average observed minimum hypercapnic density surface during WCOA 2016 (Figure S1.3) in daily output from a near real-time model in the CCLME between 2011 and 2020. We define an event when this average density surface is shallower than the 200 m isobath (approximate shelf break) on a given day. For example, if the density surface oscillates around the 200 m isobath during the transition from non-upwelling to upwelling conditions over the course of several days, each daily crossing of the 200 m isobath will be categorized as a distinct daily event instead of as oscillations of the same event. Though this may lead to a slight overestimation on the number of hypercapnic events each year, we report the average maximum duration when events occur at a given latitude (Figure 1.4b) to show how long events may persist at the 200 m isobath. Additionally, we report regional averages for northern, central, and southern subregions of the CCLME. However, regional averaging might not be representative for every location within each subregion because there are certain locations that are hotspots for hypercapnic events (see Figures S1.10 and S1.11). In the real coastal ocean, the presence of subsurface hypercapnic water on the continental shelf will depend on the local bathymetry and coastal topography of the region. These spatial

features may not be fully resolved in the relatively coarse CCLME NRT model. In this study, we use the CCLME NRT model bathymetry to determine the location (latitude and longitude) of the 200 m isobath for internal consistency within the modelling framework, despite differences between this model and the higher resolution gridded bathymetric data from the General Bathymetric Chart of the Oceans (GEBCO; Figure S1.1b). Further, model results within ~ 0.5 to 1° of the CCLME NRT domain boundary may be subjected to uncertainties attributed to boundary conditions. We neglect hypercapnic events that occur within 0.5° of the CCLME NRT latitudinal bounds in the event analysis to account for these biases.

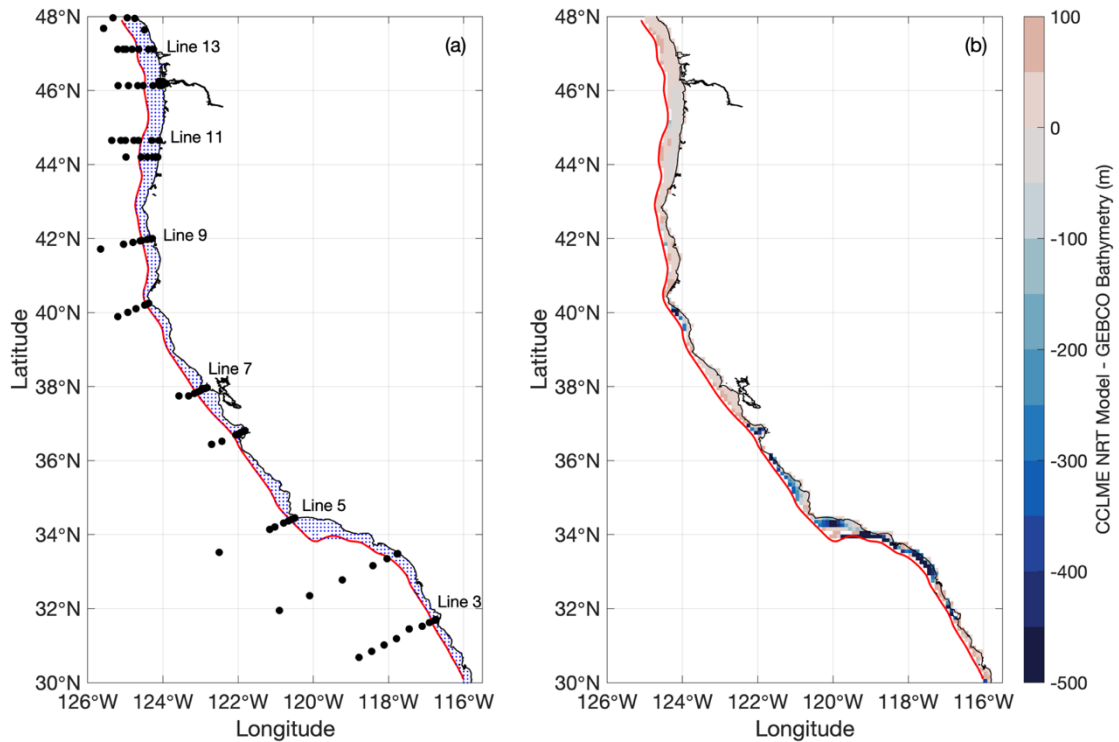


Figure S1.1. (a) Observational and near real time (NRT) model domain in the CCLME. Black dots indicate the sampling stations from various cross-shelf transects during WCOA 2016. The red line indicates the continental shelf boundary at the 200 m isobath and blue dots indicate the model grid points on the continental shelf. (b) Differences between CCLME NRT model and GEBCO bathymetry (m).

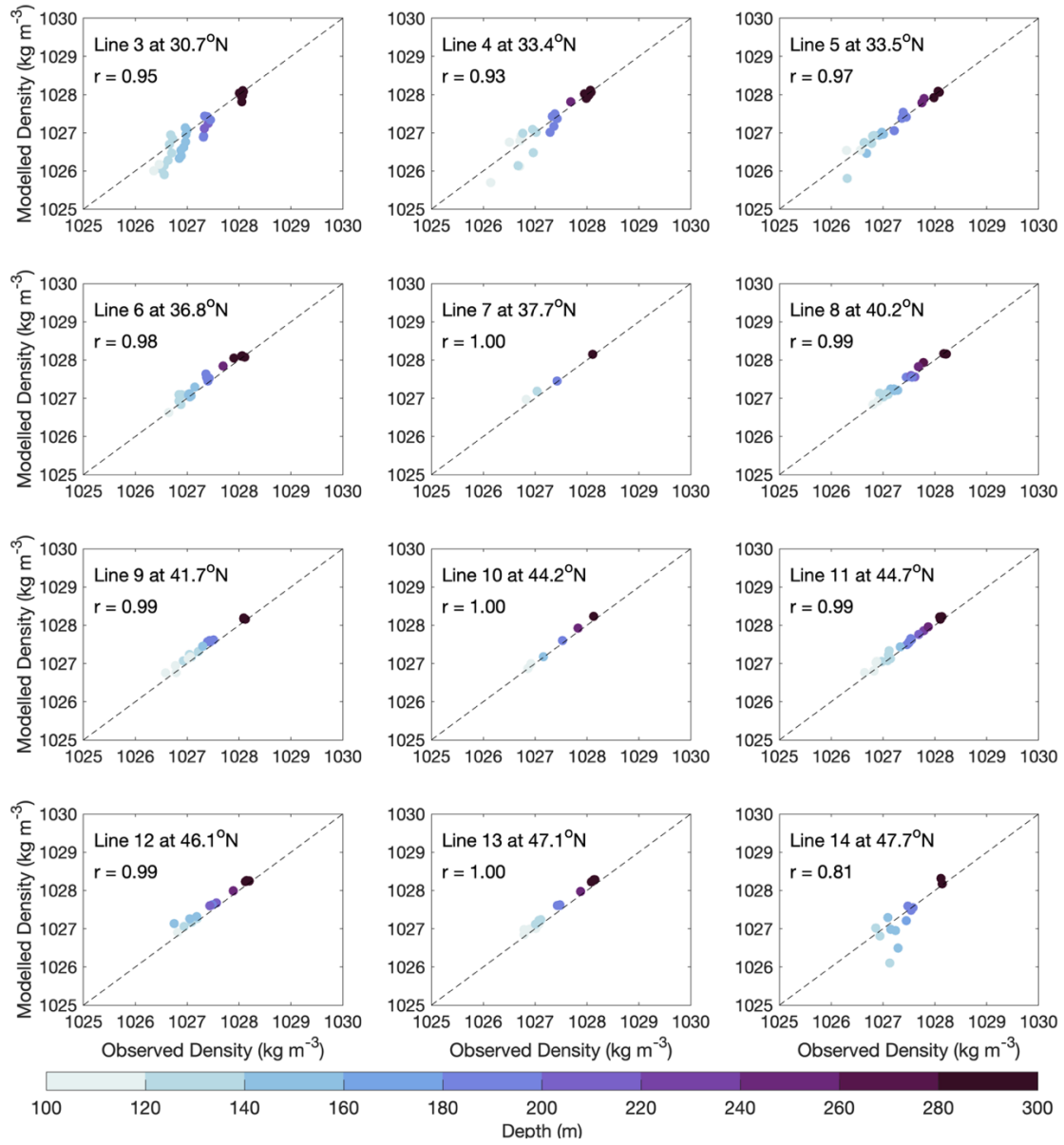


Figure S1.2. Comparison of WCOA 2016 observations and simultaneous (by day) CCLME NRT 2016 simulations of density (kg m^{-3}) at stations sampled along each cruise transect (Lines 3 to 14; see Figure S1.1a) between 100 and 300 m (colors), enveloping the depth range of the observed upper density of hypercapnia (see Figure S1.3). In each panel, the 1:1 line (dashed) is shown, and the Pearson's correlation coefficient (r) for each cruise transect is indicated in the upper left corner.

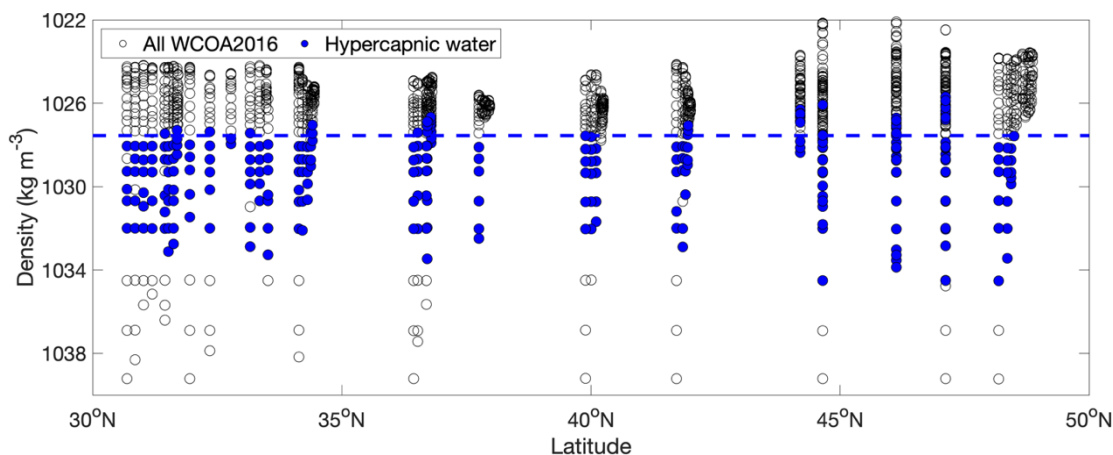


Figure S1.3. Observed densities (kg m^{-3}) on which hypercapnia (blue; $p\text{CO}_2 \geq 1000 \mu\text{atm}$) occurred during the WCOA 2016 cruise (all circles) from samples collected within the CCLME NRT model domain (30°N - 48°N). The blue dashed line represents the observed average minimum density for hypercapnia (1027.5 kg m^{-3}) that was tracked in the CCLME NRT physical model simulations to estimate occurrence of hypercapnic events.

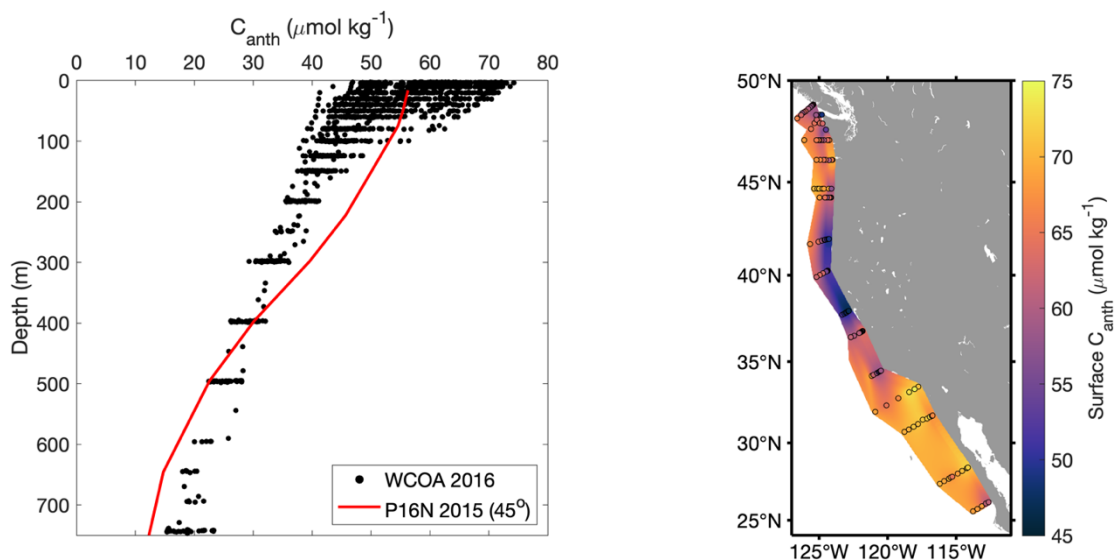


Figure S1.4. (a) Estimates of anthropogenic carbon (C_{anth} , $\mu\text{mol kg}^{-1}$) in the upper 750 m at WCOA 2016 cruise stations in the CCLME (black dots) and at 45°N , $\sim 152^\circ\text{W}$ in the North Pacific Ocean (red) from Carter et al., 2017. (b) Surface distribution of C_{anth} during WCOA 2016, interpolated from surface observations (circles).

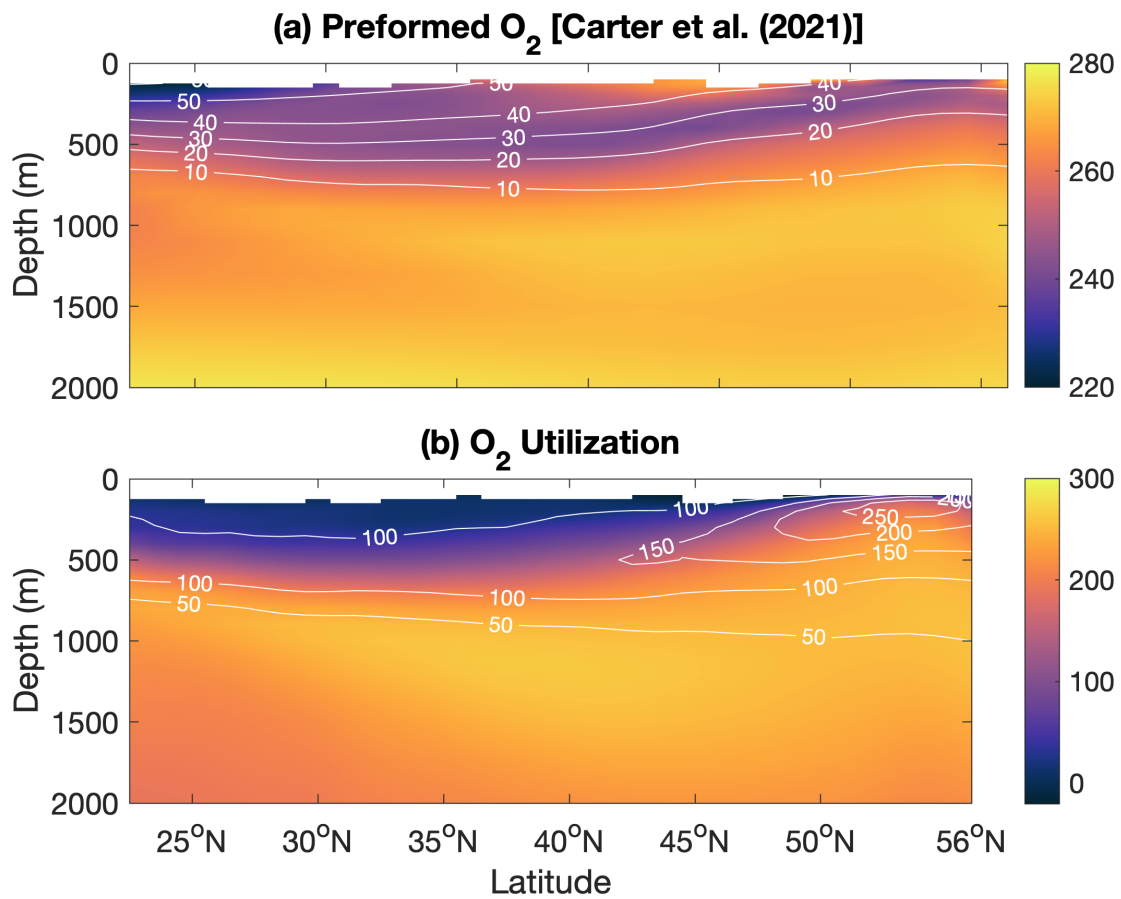


Figure S1.5. (a) Preformed dissolved oxygen ($\mu\text{mol kg}^{-1}$) from Carter et al. (2020) and (b) the utilization of dissolved oxygen ($\mu\text{mol kg}^{-1}$) based on the difference between preformed O_2 from Carter et al. (2020) and observed O_2 from GLODAPv2 2016 (Lauvset et al. 2016) in the Central North Pacific Ocean along $\sim 152^\circ\text{N}$ between 22.5°N and 56°N . White contours in (a) show anthropogenic carbon ($\mu\text{mol kg}^{-1}$) values through the year 2002 from GLODAPv2, and white contours in (b) show the change in $p\text{CO}_2$ (modern $p\text{CO}_2$ – preindustrial $p\text{CO}_2$; μatm) due to anthropogenic carbon accumulation.

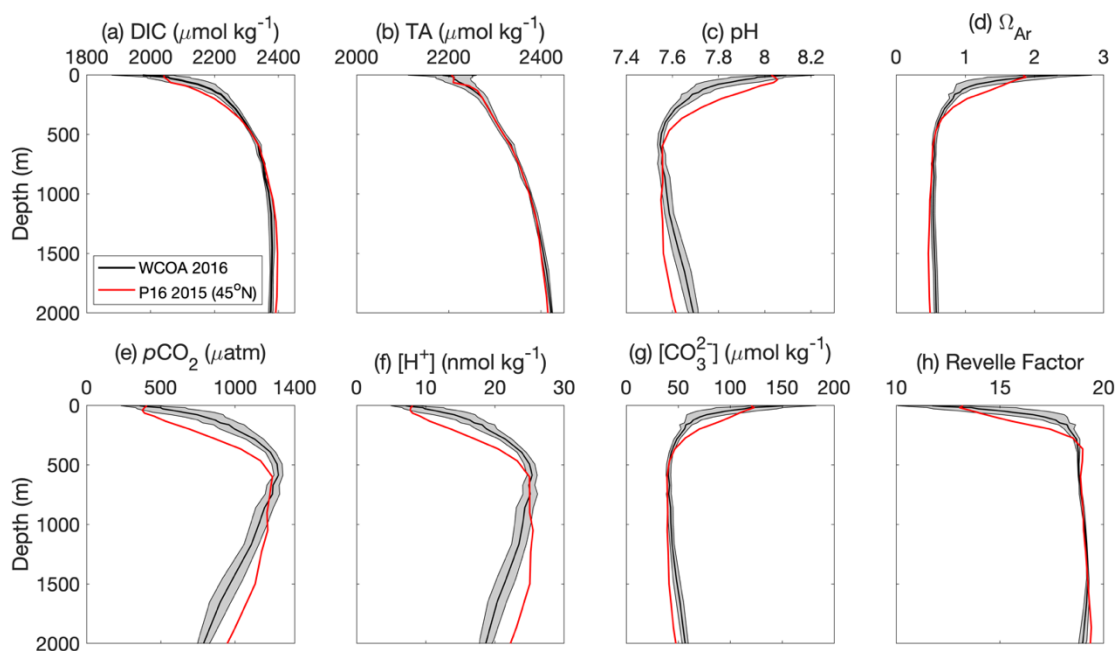


Figure S1.6. Profiles of carbonate system parameters in the open and coastal North Pacific Ocean. Mean (black) and one standard deviation (gray) profiles during the WCOA 2016 cruise in the CCLME and at 45°N, ~152°W from CLIVAR/GO-SHIP P16N in 2015 (red) for: (a) DIC ($\mu\text{mol kg}^{-1}$), (b), TA ($\mu\text{mol kg}^{-1}$), (c) pH on the total scale, (d) aragonite saturation state (Ω_{Ar}), (e) $p\text{CO}_2$ (μatm), (f) $[\text{H}^+]$ (nmol kg^{-1}), (g) $[\text{CO}_3^{2-}]$ ($\mu\text{mol kg}^{-1}$), and (h) Revelle Factor.

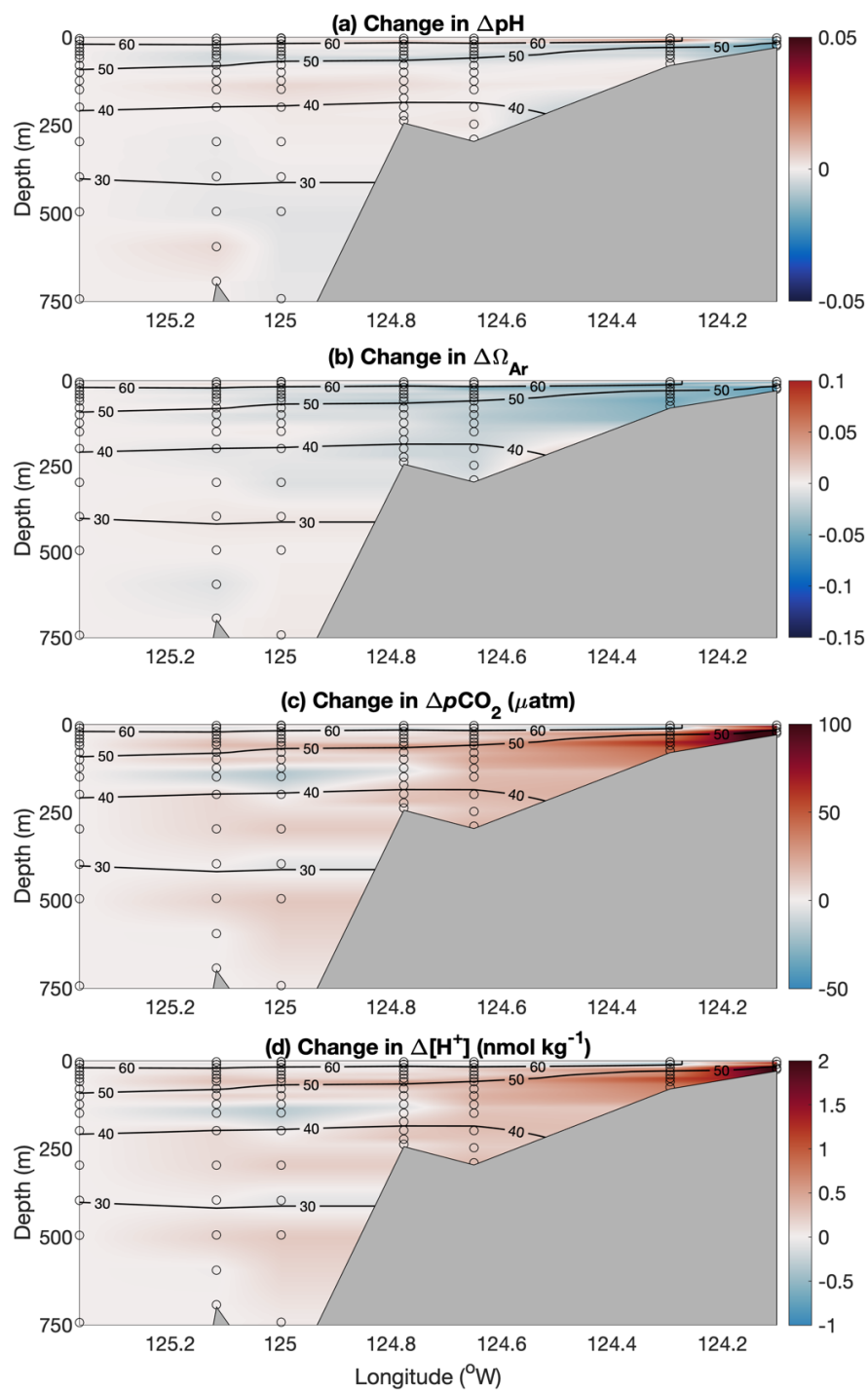


Figure S1.7. Changes in carbonate system parameters along density surfaces from offshore to onshore at Line 11 (~45°N; Figure S1a) in the CCLME as a metric for enhanced remineralization over the continental shelf. The furthest offshore value was subtracted from all values on the same density surface for (a) ΔpH , (b) $\Delta \Omega_{Ar}$, (c) ΔpCO_2 (μatm), and (d) $\Delta [H^+]$ ($nmol\ kg^{-1}$) where Δ = modern - preindustrial. Contours show concentrations of anthropogenic carbon (C_{anth} ; $\mu mol\ kg^{-1}$).

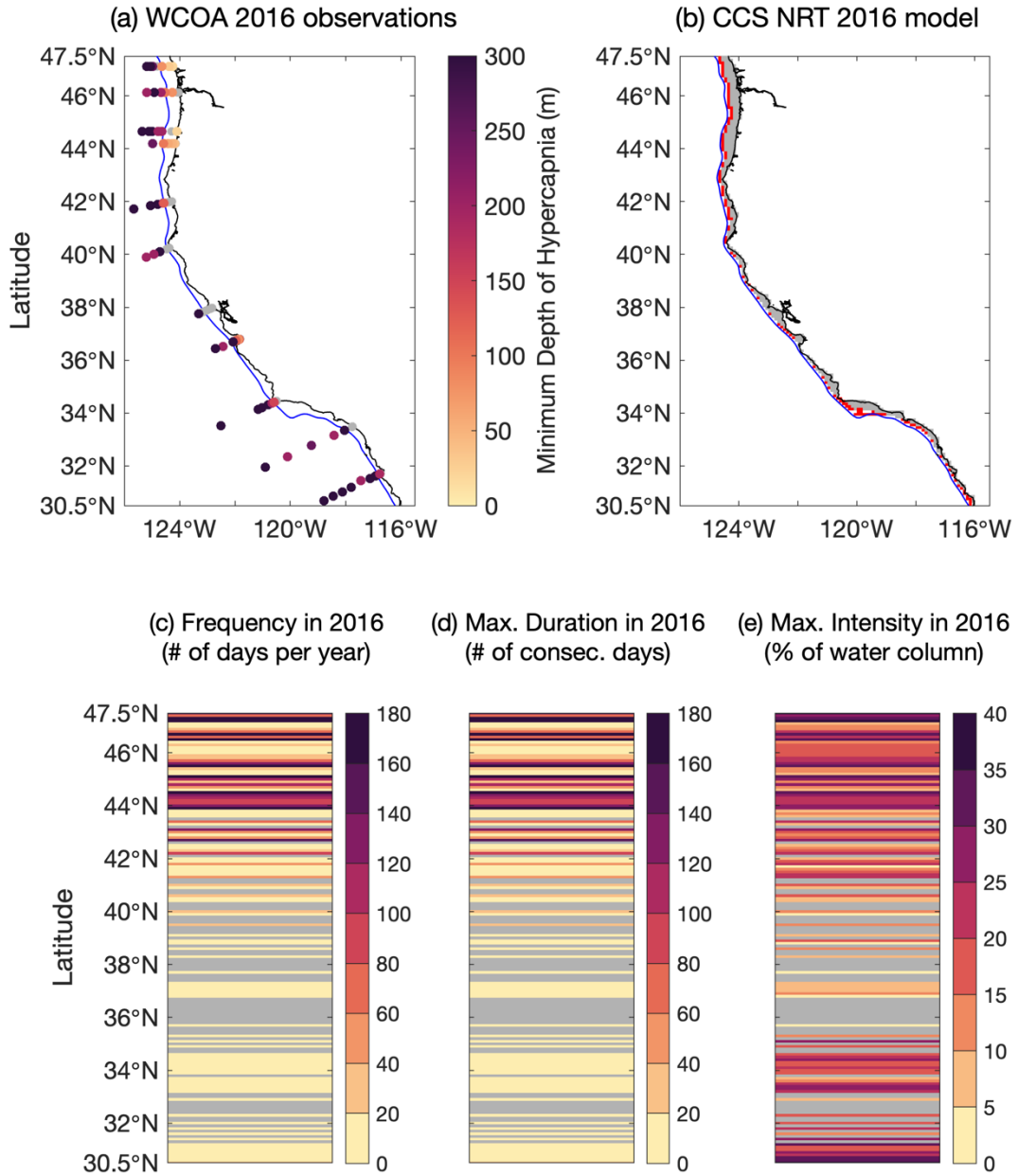


Figure S1.8. Hypercapnia in the CCLME in 2016. (a) Observed minimum depth (m) of hypercapnia at sampling stations during WCOA 2016. (b) The simulated locations (red) of hypercapnia on the shelf in 2016 estimated by the CCLME NRT model, determined by tracking the minimum hypercapnic density (see methods). In (a) and (b), gray indicates no hypercapnia occurrence (observed or simulated), and the blue line delineates the 200 m isobath. The simulated (c) frequency, (d) maximum duration, and (e) maximum intensity of hypercapnic events at the 200 m isobath in 2016. Gray indicates no hypercapnic event occurred. See main text for metric definitions.

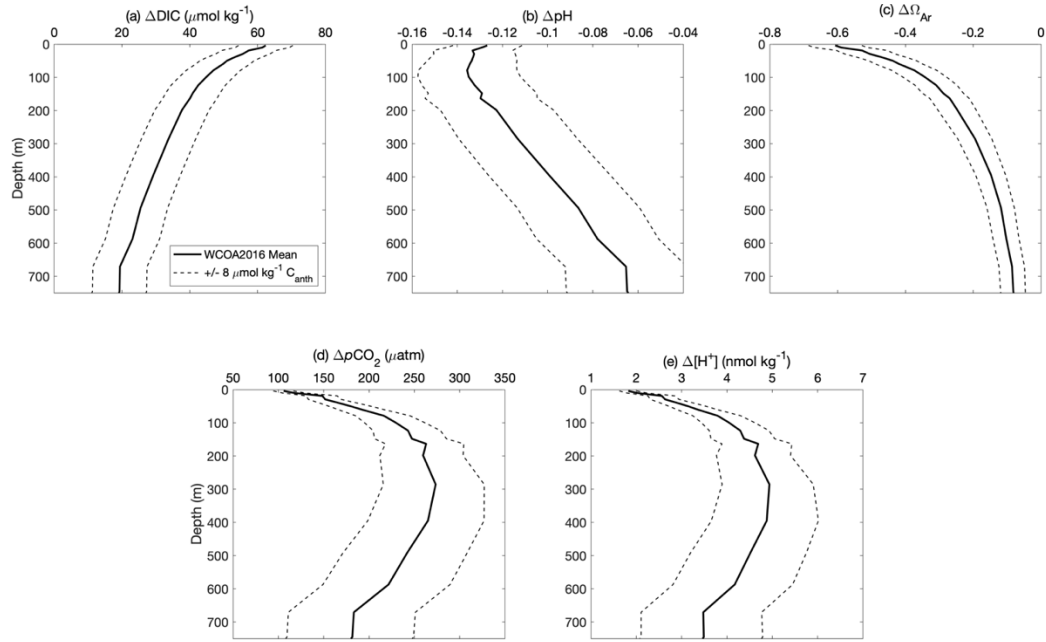


Figure S1.9. Depth-dependent uncertainties in (a) ΔDIC , (b) ΔpH , (c) $\Delta\Omega_{\text{Ar}}$, (d) $\Delta p\text{CO}_2$, and (e) $\Delta[\text{H}^+]$ in the upper 750 m in the CCLME. The bold line in each figure represents the average profile from all CCLME stations, as in Figure 1.2 of the main text. The dashed lines represent the average profiles of parameters when the $\pm 8 \mu\text{mol kg}^{-1}$ uncertainty in individual C_{anth} estimates is propagated through calculations (see Text S1.2 and Table S1.2).

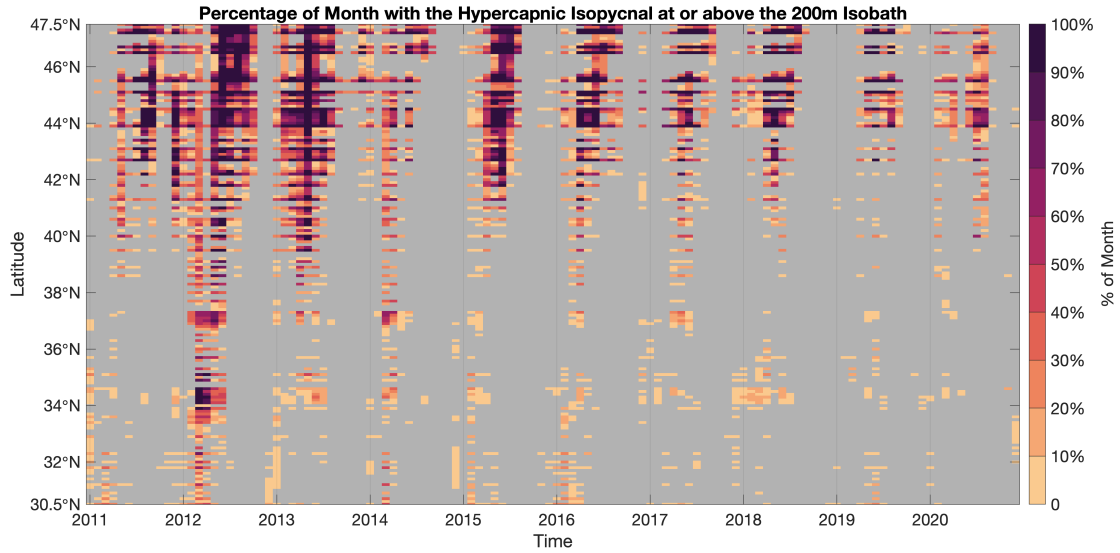


Figure S1.10. Percentage of the month when the hypercapnic isopycnal was at or above the 200 m isobath for months between 2011 and 2020 in the CCLME NRT model. Gray indicates no hypercapnic event occurred at the 200 m isobath.

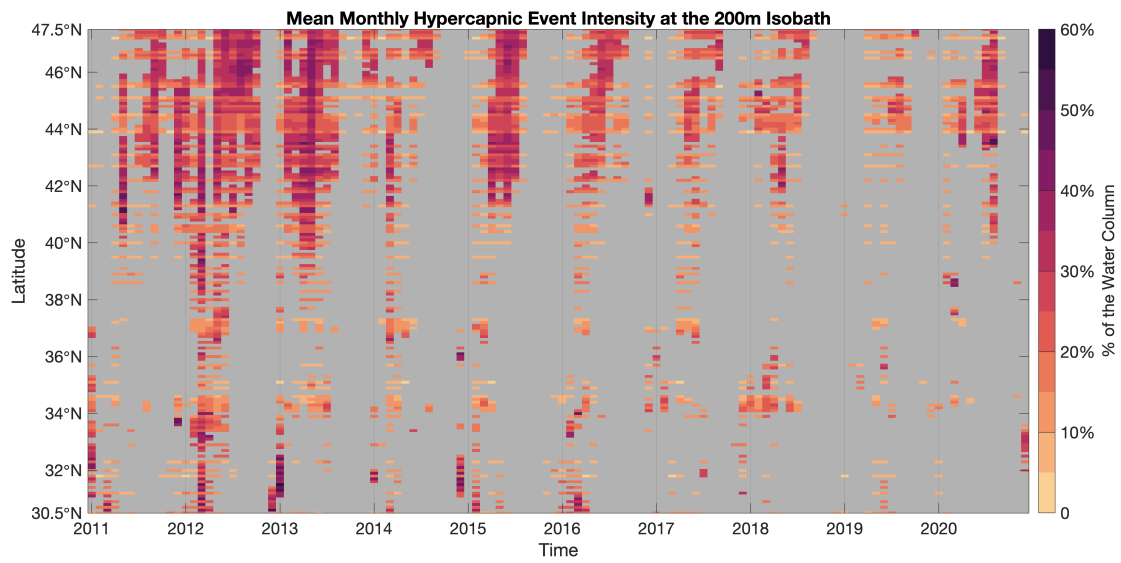


Figure S1.11. Mean monthly hypercapnic intensity at the 200 m isobath for events that occurred during between 2011 and 2020 in the CCLME NRT model. Gray indicates no hypercapnic event occurred at the 200 m isobath.

Climatological Depth of the Hypercapnic Isopycnal at the 200m Isobath

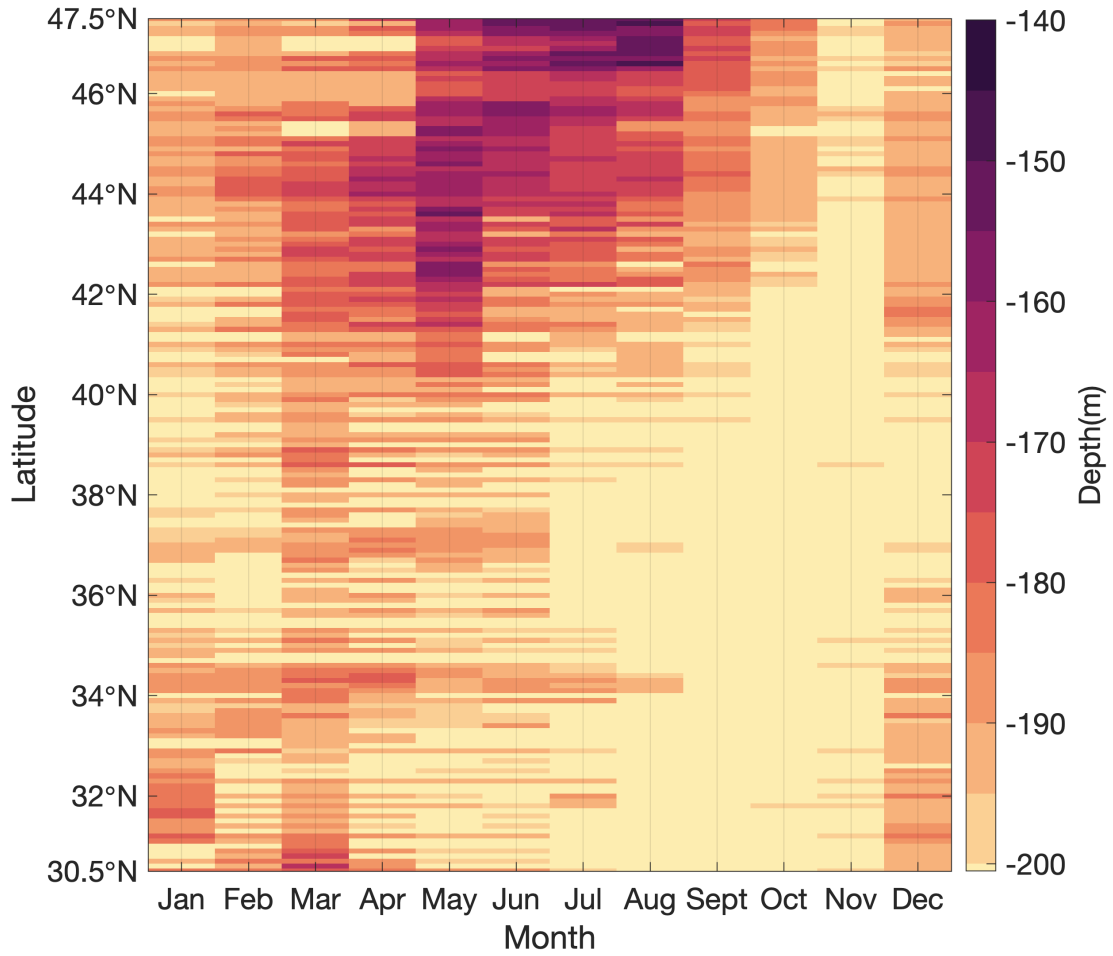


Figure S1.12. Climatological mean depth of the hypercapnic isopycnal at the 200 m isobath. The climatology was calculated by taking the average across months of the mean monthly hypercapnic isopycnal depth between 2011 and 2020 in the CCLME NRT model and assuming a mean monthly isopycnal depth of 200 m when no event occurred.

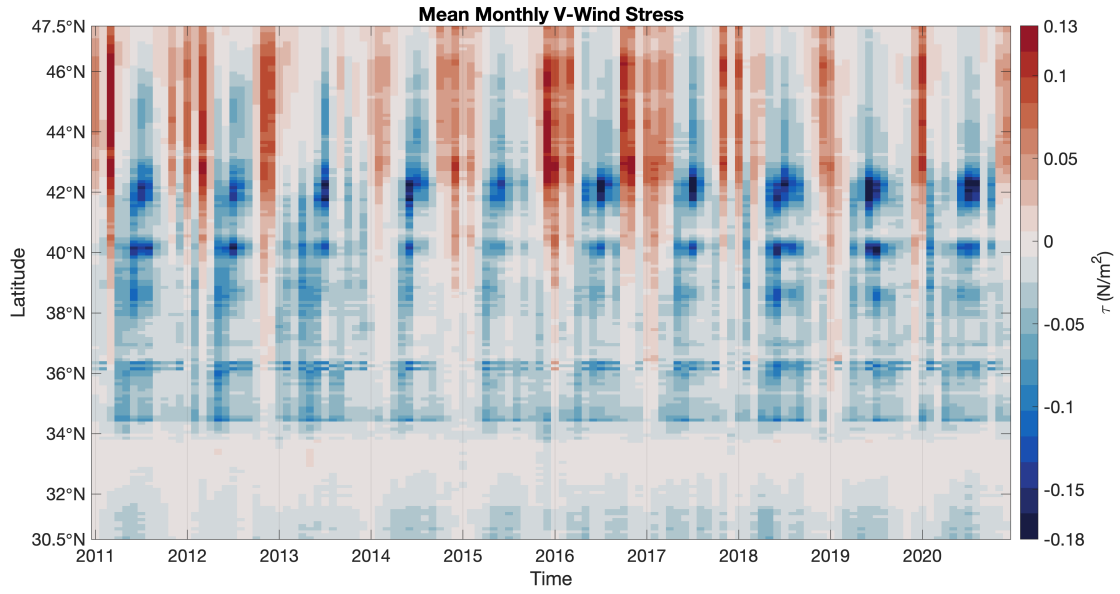


Figure S1.13. Mean monthly v-wind stress (τ ; N/m^2) averaged longitudinally between the coast and the 200 m isobath for months between 2011 and 2020 in the CCLME NRT model.

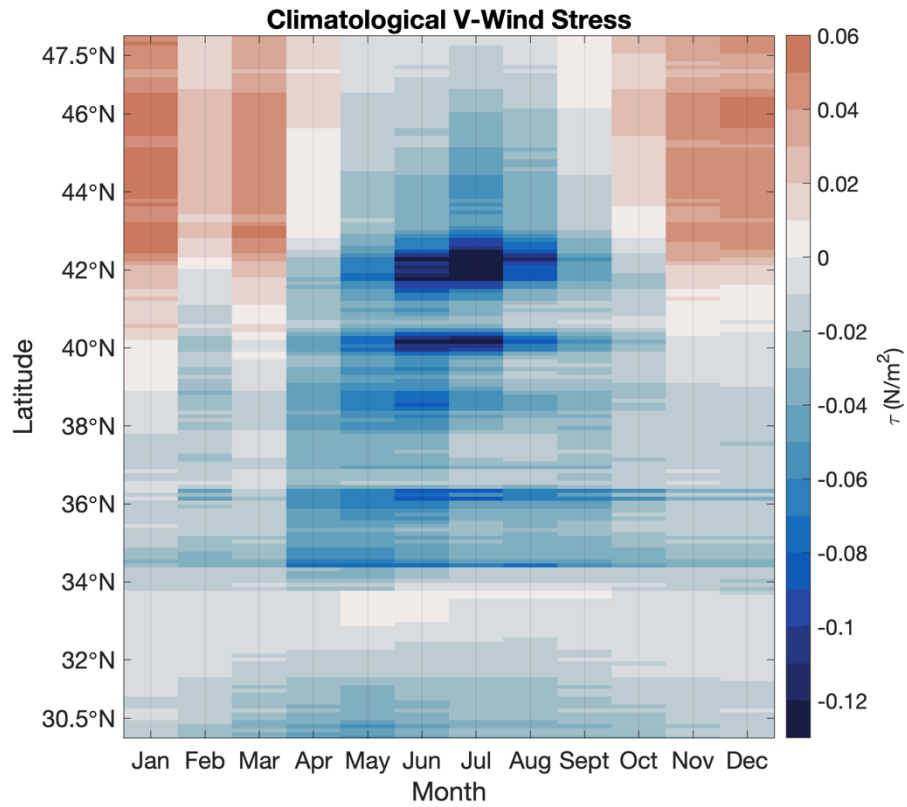


Figure S1.14. Climatological monthly mean v-wind stress (τ ; N/m^2) averaged longitudinally between the coast and the 200 m isobath for months between 2011 and 2020 in the CCLME NRT model.

Chapter 2: The increasing impact of seasonality biases on model-based estimates of the ocean carbon sink

Mar C. Arroyo, Andrea J. Fassbender, and Keith B. Rodgers

2.1 Abstract

Global ocean carbon cycle syntheses combine observation-based surface ocean $p\text{CO}_2$ products and global ocean biogeochemical models (GOBMs) to estimate air-sea CO_2 fluxes. These efforts have shown that GOBMs systematically underestimate the ocean anthropogenic CO_2 sink relative to observational products, though the causes remain uncertain. Here, we test the hypothesis that systematic biases in model representations of surface ocean $p\text{CO}_2$ seasonality contribute to long-term biases in annual ocean carbon uptake. Using simulations from seven GOBMs, we evaluate changes in the surface $p\text{CO}_2$ seasonal cycle amplitudes ($A\text{-}p\text{CO}_2$) and seasonal $p\text{CO}_2$ drivers between 1980-2018, isolating the roles of anthropogenic carbon (C_{ant}) invasion and climate change. Within each model, historical $A\text{-}p\text{CO}_2$ growth is primarily caused by C_{ant} -induced increases in the mean-state sensitivity of $p\text{CO}_2$ to seasonal thermal, physical, and biological processes. In contrast, across-model differences in C_{ant} -driven $p\text{CO}_2$ amplification are primarily tied to differences in the seasonal amplitudes of sea surface temperature and dissolved inorganic carbon. We find that models with larger 1980-1984 $A\text{-}p\text{CO}_2$ exhibit proportionally larger C_{ant} -driven $A\text{-}p\text{CO}_2$ increases, widening the across-model $A\text{-}p\text{CO}_2$ spread over time. As a result, cumulative annual ocean carbon sink estimates increasingly diverge over time due to a growing model disagreement in wintertime sea-air $p\text{CO}_2$ disequilibrium. These findings reveal that biases in the

seasonal $p\text{CO}_2$ cycle cause biases in the cumulative annual sea-air CO_2 flux, underscoring the importance for community efforts to improve our understanding of seasonal carbon cycle processes to increase confidence in simulations of the ocean carbon sink.

2.2 Introduction

The ocean is a major sink for anthropogenic carbon dioxide (CO_2) emissions generated by human activities (Friedlingstein et al., 2024), serving as a key regulator of Earth's climate. The flux of anthropogenic carbon (C_{ant}) into the surface ocean increases the inventory of dissolved inorganic carbon (DIC) (DeVries et al., 2023; Gruber et al., 2019; Khatiwala et al., 2013; Müller et al., 2023) and causes shifts in seawater chemistry that reduce the efficiency of further ocean carbon uptake (Broecker et al., 1979; Middelburg et al., 2020; Revelle & Suess, 1957; Rodgers et al., 2020; Toyama et al., 2017). C_{ant} accumulation increases the partial pressure of CO_2 ($p\text{CO}_2$) and diminishes carbonate ion concentrations, progressively eroding the ocean's buffering capacity and increasing the sensitivity of the marine carbonate system to change (Riebesell et al., 2009). These secular alterations in mean-state ocean conditions represent a long-term, positive carbon cycle feedback on the global climate system, with a weakening ocean carbon sink leading to a larger remaining fraction of emitted CO_2 gas in the atmosphere over time. Here and throughout, we use the term “mean state” to signify average conditions on annual to multi-annual timescales. Rodgers et al. (2020) use idealized model perturbations to estimate that reductions in mean-state ocean buffering may cause up to a ~60% decline in the global ocean carbon sink by

2100 under high emission scenarios. It is therefore critical to understand how the ocean carbon cycle responds to rising atmospheric CO₂ levels and climate change to constrain the changing ocean carbon sink.

Synthesis efforts to track the historical evolution of the ocean carbon sink such as the Global Carbon Project (Friedlingstein et al., 2024) and the recent REgional Carbon Cycle Assessment and Processes Phase 2 (RECCAP2) initiative (DeVries et al., 2023; Terhaar et al., 2024) use air-sea CO₂ flux estimates from global ocean biogeochemical model (GOBM) simulations (Hauck et al., 2020) and observation-based *p*CO₂ data products (Chau et al., 2022; Landschützer et al., 2014; Rödenbeck et al., 2015). GOBMs are hindcast models forced by reanalysis data to simulate physical, chemical, and biological processes influencing ocean carbon cycling under transient rising atmospheric CO₂ and climate variability. Because these models represent both natural and anthropogenic carbon fluxes, they are implicitly used in the Global Carbon Project to quantify anthropogenic perturbations to the ocean carbon cycle. These models, however, are known to contain systematic biases that contribute to uncertainties in air-sea CO₂ flux estimates (DeVries et al., 2023; Fay & McKinley, 2021; Khatiwala et al., 2013; Terhaar et al., 2024; Wanninkhof et al., 2013). Notably, GOBMs generally underestimate the global ocean carbon sink strength relative to data-product-based estimates (particularly since ~2002; Friedlingstein et al., 2024), with substantial inter-model differences in CO₂ flux variability (Hauck et al., 2020; Terhaar et al., 2024). Discrepancies in GOBM-based CO₂ flux estimates arise in part due to biases in the mean-state surface ocean carbonate chemistry, which dictates the

buffering capacity for C_{ant} uptake, and in ocean circulation, which controls C_{ant} transport from the surface mixed layer into the interior ocean (Terhaar et al., 2024). These biases are known to cause projected air-sea CO_2 flux discrepancies in structurally similar Earth System Models (ESMs; Fu et al., 2022; Goris et al., 2018; Terhaar et al., 2022; Vaithinada Ayar et al., 2022). The RECCAP2 assessment by Terhaar et al. (2024) further finds that the across-model differences in historical ocean carbon uptake in GOBMs are tightly linked to structural differences in preindustrial spin-up procedures (S  f  rian et al., 2016). Additional sources of model-product discrepancy come from biases in the observation-based $p\text{CO}_2$ data products themselves due to the spatiotemporal sparsity in data sampling and inherent uncertainties in statistical or machine-learning gap filling methodologies (Gloege et al., 2021; Gregor et al., 2019; Hauck et al., 2023; Mayot et al., 2024), particularly in the Southern Ocean where data-based CO_2 flux estimates are highly uncertain (Bushinsky et al., 2019).

Despite these known carbon cycle biases, GOBMs simulate the seasonal variability of air-sea CO_2 fluxes, which allows us to evaluate the role of seasonal carbon cycle representations in modulating model-based estimates of the ocean carbon sink (Rodgers et al., 2023). In recent decades, there has been a documented amplification of surface ocean $p\text{CO}_2$ seasonal cycles across much of the open ocean. Such seasonal $p\text{CO}_2$ amplifications have been recorded by longstanding surface $p\text{CO}_2$ measurements (Landsch  tzer et al., 2018) and reconstructed in GOBMs used in synthesis efforts (Rodgers et al., 2023). On a seasonal level, C_{ant} accumulation magnifies how natural processes influence carbon cycle variability (Fassbender et al., 2018; Gallego et al.,

2018; Hauck & Völker, 2015; Kwiatkowski & Orr, 2018; Riebesell et al., 2009). Because seasonal $p\text{CO}_2$ variations strongly regulate air-sea CO_2 fluxes, changes in the amplitude, structure, or phase of surface $p\text{CO}_2$ seasonality may alter the cumulative annual ocean carbon sink (Fassbender et al., 2018, 2022; Gorgues et al., 2010; Hauck & Völker, 2015; Kessler & Tjiputra, 2016; Nevison et al., 2016; Riebesell et al., 2009; Rodgers et al., 2008). Using large-ensemble simulations of one ESM, Fassbender et al. (2022) find that accounting for the projected amplification in surface ocean $p\text{CO}_2$ seasonality caused directly by C_{ant} leads to a modest increase in the cumulative global ocean carbon sink over the 21st century (by $\sim 8\%$). Therefore, C_{ant} -induced changes in seasonal carbon cycle variations may act as a negative carbon cycle feedback on seasonal timescales, working to enhance ocean carbon uptake and partially offset the dominant, positive annual mean-state feedback attenuating the ocean carbon sink (e.g., Rodgers et al., 2020).

Motivated by this link between $p\text{CO}_2$ seasonal cycle alterations and changing ocean carbon uptake, our study investigates the historical changes in surface $p\text{CO}_2$ seasonal cycle amplitudes as represented in multiple GOBMs used for carbon budgeting efforts. Previous studies have identified biases in ESM representations of seasonal $p\text{CO}_2$ variations, specifically in prominent C_{ant} uptake regions of the Southern Ocean (Kessler & Tjiputra, 2016; Mongwe et al., 2018; Nevison et al., 2016) and subpolar North Atlantic Ocean (Goris et al., 2018). How these simulated seasonal biases work alongside secular changes in annual mean-state ocean conditions to influence ocean carbon sink estimates across multiple GOBMs remains to be studied.

Although it is often assumed in a “mean state” view that timescales of variability are independent from one another and thus linear, changes in the seasonal $p\text{CO}_2$ cycle may in turn influence annual secular $p\text{CO}_2$ trends by modulating air-sea CO_2 fluxes through underlying nonlinear carbonate system interactions (Fassbender et al., 2018, 2022). Here, we test the hypothesis that an important contribution to systematic annual mean-state carbon cycle biases in GOBMs stems in part from systematic biases in the representations of the surface ocean $p\text{CO}_2$ seasonality. Accurate model representations of $p\text{CO}_2$ seasonal cycle change may be essential for constraining modern air-sea CO_2 fluxes for carbon budgeting efforts and reducing uncertainties in future ocean carbon sink projections.

In this study, we evaluate and compare the simulated changes in $p\text{CO}_2$ seasonal cycle amplitudes across different GOBMs used by the Global Carbon Project and in RECCAP2 assessments. We build on the work of Rodgers et al. (2023) to examine the drivers of $p\text{CO}_2$ seasonality change over the historical period from 1980 to 2018. Specifically, we isolate the impacts of C_{ant} accumulation and evolving climate states on the total change in $p\text{CO}_2$ seasonality over the open ocean. We aim to demonstrate how biases in seasonal carbon cycle drivers inherent to GOBMs influence the simulated differences in $p\text{CO}_2$ seasonal amplitude change. Lastly, we illustrate how these seasonality biases drive across-model differences in the cumulative annual ocean carbon sink strength over time.

2.3 Methods

2.3.1 Global Ocean Biogeochemical Models

We used simulations from a subset of global ocean biogeochemical models provided by the ocean section of the RECCAP2 effort (Müller, 2023) to evaluate the changes in surface ocean $p\text{CO}_2$ seasonal cycles between 1980 and 2018 (Table 2.1). GOBM output is provided on monthly timescales at $1^\circ \times 1^\circ$ horizontal resolution. All models were forced with historical levels of atmospheric CO_2 and observational-based reanalysis climate products (e.g., wind, precipitation, temperature, humidity) to resolve the source, sink, and transport processes of the ocean carbon cycle. For all simulations, the majority of models first performed a pseudo-preindustrial spin-up procedure, forced with a constant atmospheric CO_2 fixed at preindustrial levels and a repeated normal-year (climatological) climate forcing representative of either 1948 or 1958 (depending on the choice of reanalysis product; see DeVries et al., 2024 supplement). The goal of this initial spin-up phase is for the model to reach a steady state where ocean carbon fluxes become time-invariant on multi-year timescales and globally sum to zero before industrialization. After industrialization, models continue to use normal-year climate forcing until transient historical reanalysis fields become available (either 1948 or 1958). Incomplete spin-ups, in which a model has not fully reached a steady state with respect to carbon fluxes at industrialization, introduce biases (constant in time) and drifts (time evolving) that differ between models, and are retained through the simulations (Hauck et al., 2020). Model biases may arise from differences in model configuration, including the choice of reanalysis forcing product, the length of spin-up phases, and the starting year of the historical simulation (DeVries et al., 2023; Hauck

et al., 2020; Terhaar et al., 2024). Forcing fields used by these GOBMs include CORE (Large & Yeager, 2009), JRA-55 (Kobayashi et al.,

Table 2.1. Global ocean biogeochemical models used in this study. WOA18 = World Ocean Atlas 2018.

Model	Reference	Notes
CCSM-WHOI (CCSM 3.1)	Doney et al., 2009	Ends in 2017
CESM-ETHZ	Lindsay et al., 2014; Yang & Gruber, 2016	
CNRM-ESM-1	Berthet et al., 2019; Séférian, Delire, et al., 2016; Séférian et al., 2019	
EC-Earth3	Aumont et al., 2015; Döscher et al., 2022	
FESOM-REcoM-LR	Hauck et al., 2020	No preindustrial spin-up Used WOA18 phosphate
NorESM-OC1.2	Schwinger et al., 2016	
ORCA025-GEOMAR	Chien et al., 2022; Kriest & Oschlies, 2015	No preindustrial spin-up Used WOA18 silicate

2015), and NCEP/NCAR reanalysis products (Kalnay et al., 1996). Additionally, while some models simulate carbon transport from land by rivers or carbon burial in sediments, others do not. Detailed information about model performance and comparisons of carbon cycle processes against observation-based data products can be found in recent RECCAP2 chapters (DeVries et al., 2023; Rodgers et al., 2023; Terhaar et al., 2024). For our analysis, we excluded sea ice-covered regions in the Arctic Ocean and high-latitude Southern Ocean due to the paucity of seasonal $p\text{CO}_2$ observations available to validate model performance.

2.3.2 Quantifying Mechanisms of Surface Ocean $p\text{CO}_2$ Seasonal Cycle Amplitude Change

We use seven RECCAP2 GOBMs (Table 2.1), each performing four historical simulations (Table 2.2) that allow us to independently quantify the effects of increasing atmospheric CO₂ and changes in climate state on the reconstructed surface ocean *p*CO₂ seasonal cycle amplitudes between 1980 and 2018. For each model, seasonal cycle amplitudes of *p*CO₂ (*A-p*CO₂) are computed as the local maximum minus minimum value in each simulation year. Since we are interested in multi-decadal changes, the change in *p*CO₂ seasonal cycle amplitude ($\Delta A-pCO_2$) is computed by subtracting the mean *A-p*CO₂ between 1980-1984 from the mean *A-p*CO₂ between 2014-2018 ($\Delta A-pCO_2 = 2018-2014 \text{ } A-pCO_2 - 1980-1984 \text{ } A-pCO_2$). Seasonal cycle amplitudes are averaged over the 5-year periods to smooth for interannual variability before computing the amplitude change. Output from the CCSM-WHOI model (Doney et al. 2009; Table 2.1) is only available through 2017, so we consider climatologies and averages over 2014-2017 for this model.

Simulation A (Sim A) is a contemporary simulation using transient, rising atmospheric CO₂ and time-evolving climate forcing, capturing the net effect of anthropogenic and natural carbon cycle processes. The total change in *A-p*CO₂ ($\Delta_{\text{total}} A-pCO_2$) is evaluated over this simulation as the combined impact of C_{ant} accumulation and changes in climate variability, which includes the nonlinear interaction between the rising atmospheric CO₂ and changing physical climate state.

$$\Delta_{\text{total}} A-pCO_2 = 2014-2018 \text{ } Sim A \text{ } A-pCO_2 - 1980-1984 \text{ } Sim A \text{ } A-pCO_2 \quad \text{Eq 1.}$$

Simulation B serves as a control run, forced with constant, preindustrial atmospheric CO₂ levels and repeated normal-year climate forcing. We use *Simulation*

B to estimate the models' drift over time. Because drifts in $p\text{CO}_2$ seasonal cycle amplitude are extremely small between 1980 and 2018 in each model (Figure S2.1), we do not correct for model drift as many RECCAP2 studies do.

In *Simulation C (Sim C)*, atmospheric CO_2 levels increase at historical rates, while climate variability is held constant under repeated normal-year forcing. This simulation is used to isolate the steady-state impact of C_{ant} accumulation on seasonal $p\text{CO}_2$ amplitudes ($\Delta_{\text{Cant-SS A-}p\text{CO}_2}$) in the absence of changes in climate variability:

$$\Delta_{\text{Cant-SS A-}p\text{CO}_2} = 2014\text{-}2018 \text{ Sim C A-}p\text{CO}_2 - 1980\text{-}1984 \text{ Sim C A-}p\text{CO}_2 \quad \text{Eq 2.}$$

Lastly, *Simulation D (Sim D)* is forced with constant, preindustrial atmospheric CO_2 and time-evolving climate forcing (i.e., with radiative warming) to isolate the effect of changing climate variability. We will simply refer to this as the impact of “climate change” on seasonal $p\text{CO}_2$ amplitudes ($\Delta_{\text{clim A-}p\text{CO}_2}$):

$$\Delta_{\text{clim A-}p\text{CO}_2} = 2014\text{-}2018 \text{ Sim D A-}p\text{CO}_2 - 1980\text{-}1984 \text{ Sim D A-}p\text{CO}_2 \quad \text{Eq 3.}$$

In the real world, climate change works to further modify the accumulated C_{ant} pool in the upper ocean by altering CO_2 gas solubility (via heat and freshwater exchange) and/or by modifying water column density structure (via shifts in wind patterns or ocean circulation). This non-steady-state C_{ant} component arises due to the nonlinear interaction between rising atmospheric CO_2 and climate change and represents the difference in perturbation between the fully-forced contemporary simulation (*Sim A*) and the sum of the simulations with rising atmospheric CO_2 alone (*Sim C*) and with climate change alone (*Sim D*). Non-steady-state C_{ant} fluxes simulated

by the RECCAP2 GOBMs are generally smaller than both the C_{ant} fluxes driven by rising atmospheric CO_2 and climate-change-driven CO_2 fluxes over the study period (DeVries et al., 2023). Because non-steady-state C_{ant} fluxes are not captured in *Simulation C* under constant climate forcing, we subtract *Simulation D* from *Simulation A* over the two time periods to estimate the total (steady state and non-steady state) C_{ant} perturbation on $p\text{CO}_2$ amplitude changes ($\Delta_{\text{Cant}} \text{A-}p\text{CO}_2$).

$$\Delta_{\text{Cant}} \text{A-}p\text{CO}_2 = 2014\text{-}2018 (\text{Sim A} - \text{Sim D}) \text{A-}p\text{CO}_2 - 1980\text{-}1984 (\text{Sim A} - \text{Sim D}) \text{A-}p\text{CO}_2 \quad \text{Eq 4.}$$

Table 2.2. Simulations performed by each global ocean biogeochemical model used in this study. Flux components follow RECCAP2 nomenclature and are abbreviated as: Nat = natural, C_{ant} = anthropogenic, SS = steady-state, NS = non-steady-state.

Simulation	Atmospheric CO_2	Climate Forcing	Flux Components
Simulation A	Increasing, historical	time-varying	Nat-SS + Nat-NS + C_{ant} -SS + C_{ant} -NS + bias + drift
Simulation B	Constant, preindustrial	Constant, repeated normal-year	Nat-SS + bias + drift
Simulation C	Increasing, historical	Constant, repeated normal-year	Nat-SS + C_{ant} -SS + bias + drift
Simulation D	Constant, preindustrial	time-varying	Nat-SS + Nat-NS + bias + drift

Our approach to isolate and quantify the anthropogenic changes in $p\text{CO}_2$ seasonal cycle amplitudes by subtracting the initial 1980-1984 period from the ending 2014-2018 period removes the effect of model biases and drifts (until 1980), in addition to the steady-state fluxes prior to industrialization. This approach for model bias and

drift correction differs slightly from the method used in the Global Carbon Budget and other RECCAP2 studies. The Global Carbon Budget estimates the anthropogenic air-sea CO₂ flux by subtracting the control steady-state *Simulation B* from the fully-forced contemporary *Simulation A* to account for flux changes since industrialization (Friedlingstein et al., 2024). In this way, model biases and drift over time are also removed. Because GOBMs have different degrees of model biases, we do not calculate changes in seasonal $p\text{CO}_2$ amplitude since industrialization. Instead, we use 1980-1984 as a reference period for the mean-state conditions and focus on more recent decadal changes. Further, we do not correct for models that simulate riverine carbon exchange or carbon burial processes, as we assume these processes do not substantially influence open ocean $p\text{CO}_2$ seasonal cycles and their anthropogenic changes on these timescales.

2.3.3 Thermal and Biophysical Drivers of $p\text{CO}_2$ Seasonality and Carbonate System Calculations

Surface ocean $p\text{CO}_2$ seasonality is controlled by regionally varying and often competing thermal, physical, and biological processes. We employ the methodology of Fassbender et al. (2022) to decompose simulated $p\text{CO}_2$ seasonal cycles into the thermal (T) and biological and physical (biophysical; BP) components to better understand how C_{ant} and climate perturbations alter the drivers independently to yield the simulated overall $p\text{CO}_2$ response in each model. The $p\text{CO}_2$ decomposition method by Fassbender et al. (2022) was originally applied to output from an ESM and builds upon the empirical framework initially put forth by Takahashi et al. (1993, 2002). Although we are primarily focused on the changes in $p\text{CO}_2$ seasonal cycle amplitudes (as we

compute the annual maximum minus annual minimum values regardless of the timing), this methodology preserves asymmetric seasonal changes in $p\text{CO}_2$ and its components that may alter the phenology or phasing of seasonal $p\text{CO}_2$ variations.

Within the framework of Fassbender et al. (2022), it is first necessary to define a neutral state about which seasonal variations can be identified. For each model, simulation year, and grid cell, we first compute the annual mean (AM) $p\text{CO}_2$ using the annual means of its drivers: sea surface temperature (SST), sea surface salinity (SSS), DIC, total alkalinity (TA), and inorganic nutrients (phosphate, PO_4 ; silicate, Si).

$$\text{AM } p\text{CO}_2 = f(\overline{\text{SST}}, \overline{\text{SSS}}, \overline{\text{DIC}}, \overline{\text{TA}}, \overline{\text{PO}_4}, \overline{\text{Si}}) \quad \text{Eq 5.}$$

Here, f signifies a carbonate system calculator, and overbars denote annual mean values. Climatological estimates of seasonal sea surface PO_4 and Si values were obtained from the World Ocean Atlas 2018 (WOA18) for models that did not provide those output fields (Table 2.1). Computing the annual mean $p\text{CO}_2$ from its drivers rather than an average of monthly $p\text{CO}_2$ values produces a value that is less influenced by seasonal $p\text{CO}_2$ extrema and thermodynamically consistent with the annual means of its drivers.

The thermal $p\text{CO}_2$ component ($p\text{CO}_2 \text{ T}$) captures the changes in $p\text{CO}_2$ caused by seasonal SST variations. Seasonal warming elevates surface $p\text{CO}_2$ whereas seasonal cooling reduces surface $p\text{CO}_2$ values due to changes in CO_2 gas solubility. The thermal $p\text{CO}_2$ component is computed in a similar fashion to AM $p\text{CO}_2$, but instead using the transient model fields of monthly SST and annual mean values of all other input parameters to isolate the effect of seasonal temperature changes on $p\text{CO}_2$ variability.

$$p\text{CO}_2 \text{ T} = f(\text{SST}, \overline{\text{SSS}}, \overline{\text{DIC}}, \overline{\text{TA}}, \overline{\text{PO}_4}, \bar{S}_i) \quad \text{Eq 6.}$$

This yields the simulated time series of the thermal $p\text{CO}_2$ component at monthly resolution. The efficacy at which seasonal SST variability alters the $p\text{CO}_2$ T seasonal cycle depends on the mean-state AM $p\text{CO}_2$ value, whereby $p\text{CO}_2$ values change by $\sim 4.23\%$ for each 1°C change (Takahashi et al., 1993). In other words, the AM $p\text{CO}_2$ governs the sensitivity of $p\text{CO}_2$ to seasonal SST variations: higher AM $p\text{CO}_2$ values allow for the same seasonal SST variation to exert a larger influence on seasonal $p\text{CO}_2$ variability.

The residual, biophysical $p\text{CO}_2$ component ($p\text{CO}_2$ BP) accounts for all other nonthermal carbon cycle processes, including air-sea CO_2 exchange, organic matter production and remineralization, calcium carbonate cycling, water mass mixing, and freshwater exchange. In the analysis by Rodgers et al. (2023), open-ocean biophysical $p\text{CO}_2$ seasonality in the GOBMs was found to be largely driven by seasonal variations of DIC, with seasonal surface salinity and total alkalinity variations exerting minor controls (their Figure 7). For this reason, we conceptualize the structure and phasing of the $p\text{CO}_2$ BP seasonal cycle as primarily driven by that of the DIC seasonal cycle. Increased levels of primary production in spring and summer coincident with mixed layers shoaling reduce surface $p\text{CO}_2$ and DIC, while deep winter mixing introduces subsurface waters with high $p\text{CO}_2$ and respired DIC into the upper ocean. To ensure internal consistency with the simulated $p\text{CO}_2$ seasonal cycle within each model, we follow the procedure used in Rodgers et al. (2023) to compute the time series of $p\text{CO}_2$

BP as the difference between the model $p\text{CO}_2$ time series and the seasonal $p\text{CO}_2$ T anomaly, computed using the $p\text{CO}_2$ T time series and AM $p\text{CO}_2$.

$$p\text{CO}_2 \text{ BP} = p\text{CO}_2 - (p\text{CO}_2 \text{ T} - \text{AM } p\text{CO}_2) \quad \text{Eq 7.}$$

As seasonal DIC variations predominantly control $p\text{CO}_2$ BP seasonal cycles, we describe the efficiency of the $p\text{CO}_2$ response to seasonal biophysical processes using the annual mean Revelle Factor (RF), which dictates the sensitivity of $p\text{CO}_2$ to seasonal changes in DIC (Broecker et al., 1979; Egleston et al., 2010; Fassbender et al., 2017; Revelle & Suess, 1957). The Revelle Factor is often discussed in the context of the surface ocean buffering capacity for C_{ant} accumulation, where waters with low RF values can more efficiently absorb anthropogenic CO_2 from the atmosphere, resulting in relatively smaller $p\text{CO}_2$ changes. This same concept of buffering efficiency also applies to seasonal carbon cycling, where waters with lower AM RF values experience smaller relative changes in seasonal $p\text{CO}_2$ variability for the same seasonal DIC variation (Fassbender et al., 2018). The reverse is true for waters characterized by higher AM RF.

While the timing of seasonal $p\text{CO}_2$ T and $p\text{CO}_2$ BP variations typically oppose each other, they are not often the same magnitude or entirely out of phase. There is a well-known geographic distribution of the relative influence of thermal and biophysical processes on $p\text{CO}_2$ such that seasonal SST changes dominate $p\text{CO}_2$ seasonality in mid-latitude regions, in contrast to high-latitude subpolar regions and low-latitude equatorial regions that are more influenced by seasonal DIC changes (Fassbender et

al., 2018; Takahashi et al., 2002). The seasonal phasing of the overall $p\text{CO}_2$ cycle will align with that of its dominating driver.

We compute the seasonal cycle amplitudes of SST, $p\text{CO}_2$ T, DIC, and $p\text{CO}_2$ BP and their associated changes due to C_{ant} accumulation and climate change following equations 1-4 to inform the mechanisms of overall $A\text{-}p\text{CO}_2$ change. The seasonal $p\text{CO}_2$ decomposition and the Revelle Factor computations were performed using CO2SYS Version 3.1.2 for MATLAB (van Heuven et al., 2011; Lewis & Wallace, 1998; Sharp et al., 2020). We use the dissociation constants for the carbonate system (K_1 and K_2) by Lueker et al. (2000), hydrogen sulfate by Dickson et al. (1990), and hydrogen fluoride by Perez & Fraga (1987) and the total boron-to-chlorinity ratio of Lee et al. (2010). Colormaps for figures are sourced from the cmocean MATLAB package (Thyng et al., 2016).

2.3.4 Interpreting mechanisms of $p\text{CO}_2$ seasonal amplitude change

The transient changes in $A\text{-}p\text{CO}_2$ are governed by alterations of the season variability drivers as well as changes in the annual mean-state surface conditions. Expansions or contractions in the seasonal cycle amplitudes of the thermal ($A\text{-}p\text{CO}_2$ T) and biophysical ($A\text{-}p\text{CO}_2$ BP) $p\text{CO}_2$ components could occur if there are changes to the magnitude of seasonal SST and/or DIC variability (x axes of Figure 2.1), or to the sensitivity of $p\text{CO}_2$ to seasonal SST and/or DIC variations, as dictated by mean-state AM $p\text{CO}_2$ and AM RF, respectively (y axes of Figure 2.1). The direction of the sloping contours in each panel of Figure 2.1 emphasizes that altering either the variability or the sensitivity driver results in nonlinear changes in the $p\text{CO}_2$ component seasonal

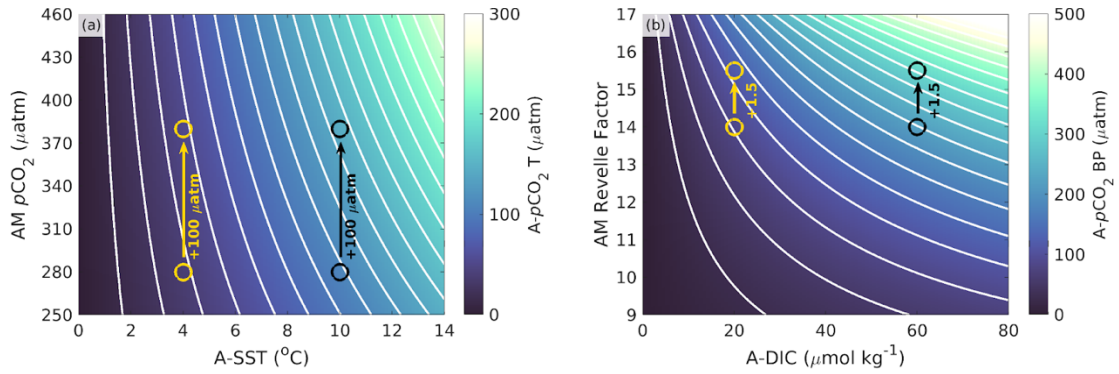


Figure 2.1. The seasonal cycle amplitudes (A) of (a) the thermal $p\text{CO}_2$ component ($A\text{-}p\text{CO}_2\text{ T}$) as a function of sea surface temperature seasonal cycle amplitude (A-SST) and the annual mean (AM) $p\text{CO}_2$ value, and (b) the biophysical $p\text{CO}_2$ component ($A\text{-}p\text{CO}_2\text{ BP}$) as a function of dissolved inorganic carbon seasonal cycle amplitude (A-DIC) and the annual mean Revelle Factor (AM RF). Changes in $A\text{-}p\text{CO}_2\text{ T}$ and $A\text{-}p\text{CO}_2\text{ BP}$ can be caused by altering the variability driver seasonality (x axes), the sensitivity of $p\text{CO}_2$ to that variability driver (y axes), or both. Yellow and black annotations represent theoretical changes in sensitivity terms at different initial variability driver conditions (see text). Contours were generated using CO2SYS with values of mean values of total alkalinity = 2350 $\mu\text{mol/kg}$, temperature = 18 $^{\circ}\text{C}$, salinity = 34.8, silicate = 15 $\mu\text{mol/kg}$, and phosphate = 1.5 $\mu\text{mol/kg}$.

cycle amplitude. In this sense, for a given magnitude of driver change, the initial driver conditions dictate the magnitude of $A\text{-}p\text{CO}_2$ component change. For example, rising ocean carbon content through C_{ant} invasion directly elevates the sensitivity of $p\text{CO}_2$ to thermal and biophysical processes by increasing the AM $p\text{CO}_2$ and AM RF (i.e., reduction in ocean buffering). This process is represented by vertical arrows in each panel of Figure 2.1. However, there will be differing magnitudes of $A\text{-}p\text{CO}_2$ component change if the initial magnitude of driver seasonal amplitude (e.g., A-SST or A-DIC; x axes) is smaller (yellow) or larger (black). Here, surface waters characterized by a larger initial A-SST or A-DIC (black) exhibit a larger initial $A\text{-}p\text{CO}_2\text{ T}$ or $A\text{-}p\text{CO}_2\text{ BP}$ and will experience a relatively larger increase in the $A\text{-}p\text{CO}_2$ component, visualized as the black vertical arrows crossing more $A\text{-}p\text{CO}_2$ component contours. In a similar way, the magnitude of $A\text{-}p\text{CO}_2$ component change will be dependent on the initial

values of the sensitivity drivers (y axes). In our analysis, we independently consider the impact of C_{ant} accumulation and climate change on both the underlying A-SST and A-DIC variability drivers as well as the AM $p\text{CO}_2$ and AM RF sensitivity terms to elucidate the combined role of seasonal and annual mean-state biases in causing differing A- $p\text{CO}_2$ changes between GOBMs.

2.4 Results and Discussion

2.4.1 Mechanisms of $p\text{CO}_2$ seasonal cycle amplitude change

We present the changes in A- $p\text{CO}_2$ and its thermal and biophysical components between 1980-1984 and 2014-2018 for one GOBM (CESM-ETHZ; Lindsay et al., 2014; Yang & Gruber, 2016) to illustrate the mechanisms of change. In this model (Figure 2.2), the total change in $p\text{CO}_2$ seasonal cycle amplitude (Δ_{total} A- $p\text{CO}_2$; Eq 1.) ranged from -26 to 55 μatm , with the largest increases over much of the subtropical and northern subpolar regions (Figure 2.2b). The Δ_{total} A- $p\text{CO}_2$ is near zero or negative in low latitudes and some areas of the Southern Ocean. Using the different forcing scenarios for this model, we can attribute Δ_{total} A- $p\text{CO}_2$ to two perturbations: (1) C_{ant} accumulation, accounting for steady-state and nonlinear non-steady-state contributions, (Eq. 4), and (2) climate change (Eq 3.). Much of the total A- $p\text{CO}_2$ increase can be ascribed to C_{ant} accumulation in the water column (Figure 2.2c). The spatial structure of C_{ant} -induced A- $p\text{CO}_2$ change aligns with the 1980-1984 pattern of A- $p\text{CO}_2$ such that larger changes occur in northern subtropical and subpolar areas that experience naturally large seasonal swings in $p\text{CO}_2$. In contrast to the effect of C_{ant} , the impact of climate change is spatially heterogeneous and can locally enhance (when

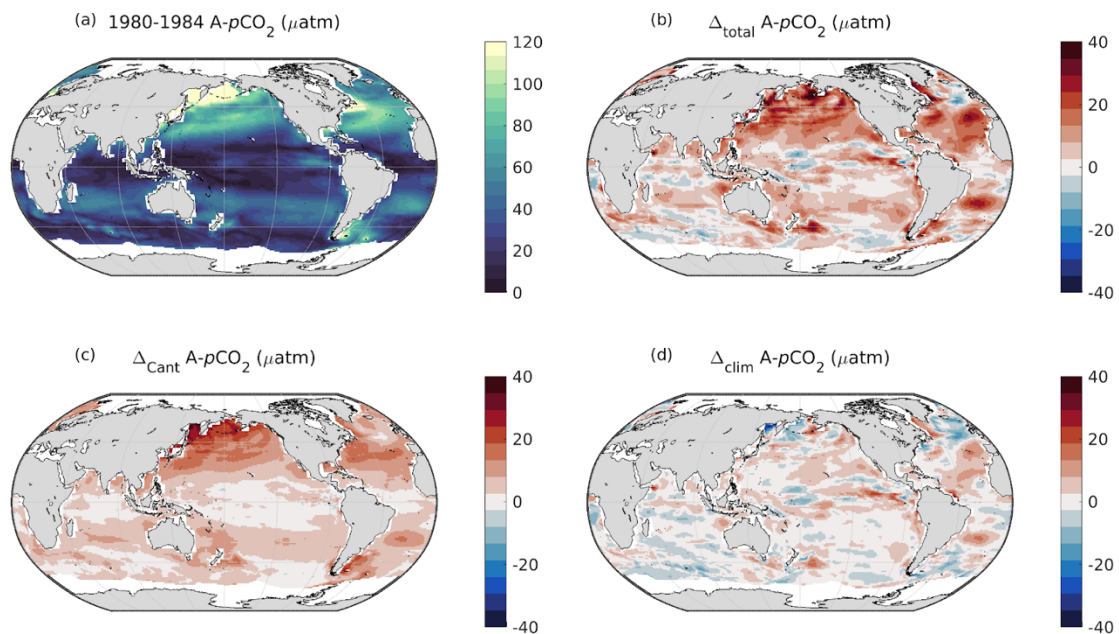


Figure 2.2 (a) The 1980-1984 mean $p\text{CO}_2$ seasonal cycle amplitude ($A\text{-}p\text{CO}_2$) in a representative global ocean biogeochemical model (CESM-ETHZ; Lindsay et al., 2014; Yang & Gruber, 2016) and (b) the total change in $A\text{-}p\text{CO}_2$ between 1980-1984 and 2014-2018. The total change is attributed to the changes caused by (c) anthropogenic carbon accumulation ($\Delta_{\text{cant}} A\text{-}p\text{CO}_2$) and (d) climate change ($\Delta_{\text{clim}} A\text{-}p\text{CO}_2$). Seasonal cycle amplitudes are computed as the local maximum value minus the local minimum value each year, then averaged over each five-year period.

positive) or dampen (when negative) the C_{ant} -induced $A\text{-}p\text{CO}_2$ increase (Figure 2.2d).

Reductions in total $A\text{-}p\text{CO}_2$ over this time period are found where the effect of climate change is large enough to counteract the C_{ant} -induced $A\text{-}p\text{CO}_2$ increase.

The changes in $A\text{-}p\text{CO}_2$ are further decomposed into the respective changes in the thermal ($p\text{CO}_2 \text{ T}$; Figure 2.3) and biophysical ($p\text{CO}_2 \text{ BP}$; Figure 2.4) $p\text{CO}_2$ seasonal cycle amplitudes in this same model. In a similar approach, we isolate the drivers of C_{ant} - and climate-driven $A\text{-}p\text{CO}_2 \text{ T}$ and $A\text{-}p\text{CO}_2 \text{ BP}$ changes by examining the impacts on the respective seasonal variability and sensitivity drivers. As seen with $A\text{-}p\text{CO}_2$, local changes in $A\text{-}p\text{CO}_2 \text{ T}$ and $A\text{-}p\text{CO}_2 \text{ BP}$ caused by C_{ant} invasion are often larger than climate-driven $A\text{-}p\text{CO}_2 \text{ T}$ and $A\text{-}p\text{CO}_2 \text{ BP}$ changes.

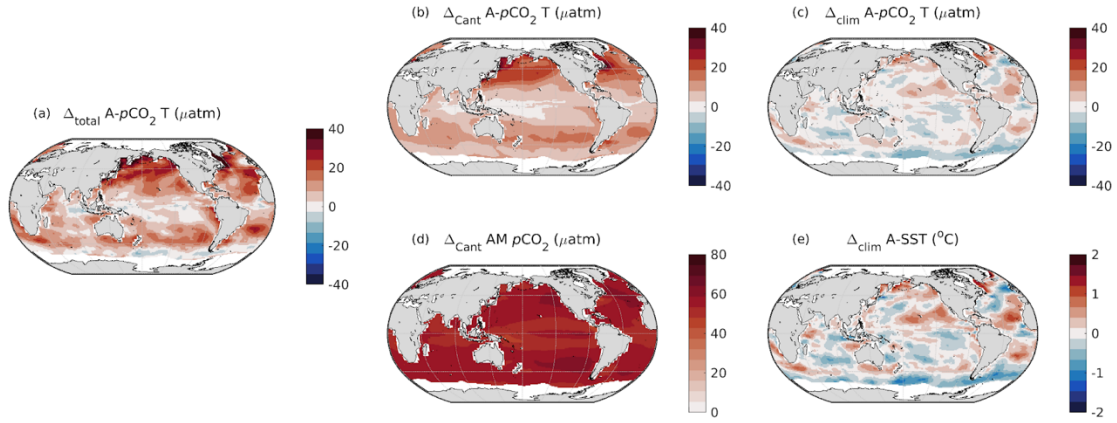


Figure 2.3. (a) The total change in the seasonal cycle amplitude of the thermal $p\text{CO}_2$ component ($\Delta_{\text{total}} \text{A-}p\text{CO}_2 \text{ T}$) between 1980-1984 and 2014-2018 as simulated by CESM-ETHZ and the attribution of change to (b) anthropogenic carbon accumulation ($\Delta_{\text{Cant}} \text{A-}p\text{CO}_2 \text{ T}$) and (c) climate change ($\Delta_{\text{clim}} \text{A-}p\text{CO}_2 \text{ T}$). (d) The change in the annual mean surface ocean $p\text{CO}_2$ (thermal sensitivity driver) caused anthropogenic carbon accumulation ($\Delta_{\text{Cant}} \text{AM } p\text{CO}_2$). (e) The change in the seasonal cycle amplitude of sea surface temperature (thermal variability driver) caused by climate change ($\Delta_{\text{clim}} \text{A-SST}$).

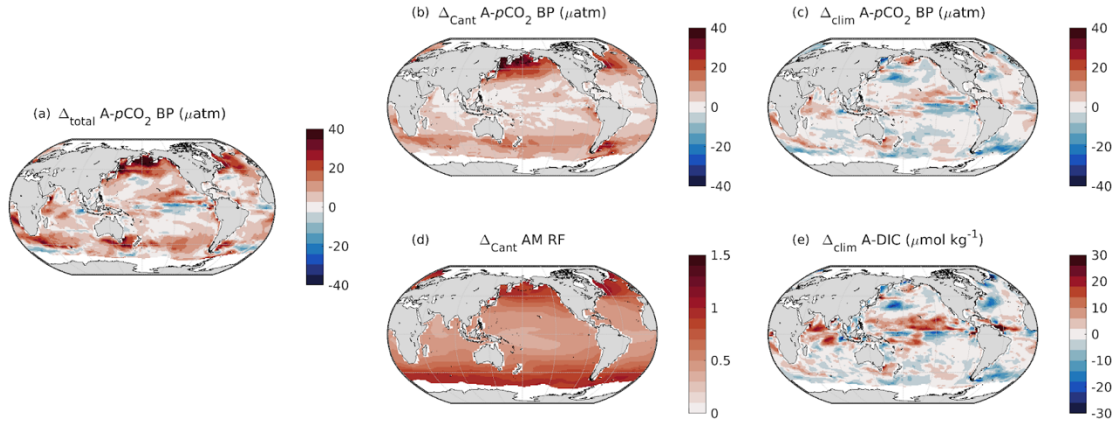


Figure 2.4. (a-c) The changes in the seasonal cycle amplitude of the biophysical $p\text{CO}_2$ component ($\text{A-}p\text{CO}_2 \text{ BP}$) between 1980-1984 and 2014-2018 as simulated by CESM-ETHZ, following Figure 3 (a-c). (d) The change in the annual mean surface ocean Revelle Factor (biophysical sensitivity driver) caused anthropogenic carbon accumulation ($\Delta_{\text{Cant}} \text{AM RF}$). (e) The change in the seasonal cycle amplitude of sea surface dissolved inorganic carbon (biophysical variability driver) caused by climate change ($\Delta_{\text{clim}} \text{A-DIC}$).

As a direct consequence of C_{ant} invasion, seasonal variations of SST and DIC will exert a larger influence on $p\text{CO}_2$ component seasonal cycles due to the increase in the annual mean-state sensitivity terms. C_{ant} uptake increases the annual mean (AM) $p\text{CO}_2$ relatively uniformly (Figure 2.3d), which increases the sensitivity of $p\text{CO}_2$ to

SST variations. Because there are no direct alterations to the SST seasonal cycle under C_{ant} accumulation alone, the rise in AM $p\text{CO}_2$ simply scales the existing pattern of the 1980-1984 A- $p\text{CO}_2$ T and A-SST (Figures S2.2 and S2.3), as reflected in the large-scale spatial distribution of ΔC_{ant} A- $p\text{CO}_2$ T (Figure 2.3b). Mid-latitude subtropical regions experience a larger ΔC_{ant} A- $p\text{CO}_2$ T response where there are regionally larger seasonal SST variations. Additionally, the increase in upper-ocean DIC by C_{ant} reduces the ocean buffering capacity, translating to an increase in the surface annual mean Revelle Factor (AM RF; Figure 2.4d) and a heightened $p\text{CO}_2$ sensitivity to seasonal DIC variability. RF values in cooler, fresher high-latitude subpolar waters with higher 1980-1984 RFs increase faster than in warmer, saltier subtropical waters with lower 1980-1984 RFs (Egleston et al., 2010; Fassbender et al., 2017; Takahashi et al., 1980). As a result, the spatial pattern in the C_{ant} -driven increase in A- $p\text{CO}_2$ BP reflects the combined response to the spatial distribution of the 1980-1984 DIC seasonal cycle and ΔC_{ant} AM RF (Figure 2.4b; Figure S2.2). That is, the largest ΔC_{ant} A- $p\text{CO}_2$ BP occurs where the 1980-1984 A-DIC and AM RF as well as the ΔC_{ant} AM RF are large. This is seen in high-latitude subpolar regions, particularly in the Northern Hemisphere. C_{ant} -induced changes in A- $p\text{CO}_2$ T and A- $p\text{CO}_2$ BP are often larger than that of the overall C_{ant} -induced A- $p\text{CO}_2$ change (Figure 2.2c), which highlights the opposing nature of seasonal $p\text{CO}_2$ T and $p\text{CO}_2$ BP cycle phasing.

In addition to enhancing $p\text{CO}_2$ sensitivities, C_{ant} accumulation in the upper ocean weakens the seasonal cycle amplitude of DIC in parts of the Southern Ocean and subpolar Northern Hemisphere, regions typically characterized by persistent wind-

driven surface divergence (Figure 2.5a). In these regions, C_{ant} concentrations decrease slightly from the surface through the seasonal thermocline (above the maximum mixed layer depth), before decreasing more steeply within the permanent thermocline below (Figure 2.5c). Seasonal C_{ant} accumulation causes surface DIC concentrations to increase at different rates in winter and summer, reflecting seasonal changes in the background chemistry and mixed layer depths (Figure 2.5b). Although C_{ant} invasion primarily occurs during the windy winter months when the mixed layer reaches its

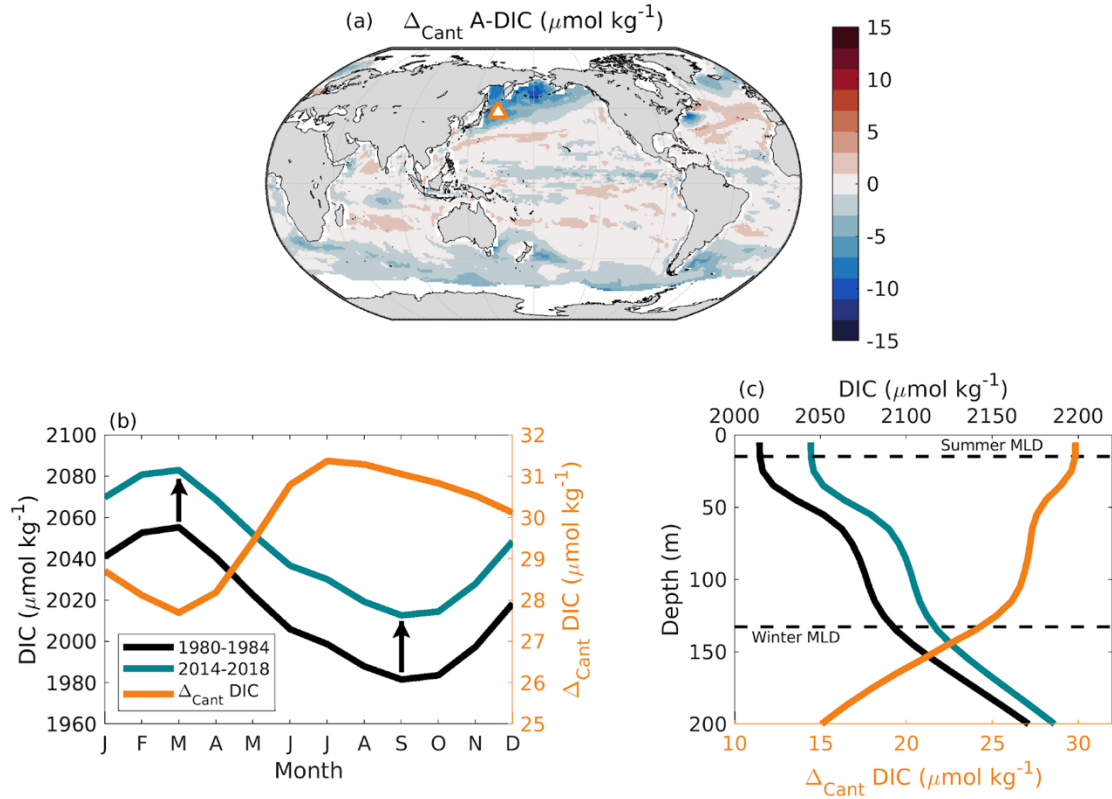


Figure 2.5. (a) The change in the surface ocean DIC seasonal cycle amplitude caused by anthropogenic carbon accumulation ($\Delta C_{\text{ant}} \text{ A-DIC}$) between 1980-1984 and 2014-2018. The (b) smoothed seasonal DIC cycles and (c) annual mean vertical DIC profiles in 1980-1984 (black), 2014-2018 (teal), and the C_{ant} accumulation between the time periods ($\Delta C_{\text{ant}} \text{ DIC}$; orange) from a representative location in northwestern North Pacific Ocean (42.5°N , 153.5°E ; orange triangle in (a)). Dashed lines in (c) mark the shallower summer (July, August, September) and deeper winter (January, February, March) mean mixed layer depths (MLDs).

annual maximum (e.g., Rodgers et al., 2008), surface DIC increases faster in summer (Figure 2.5b, orange line). This occurs because shallower summer mixed layer depths retain accumulated C_{ant} near the surface (Figure 2.5c). When destratification initiates in the fall (approximately in September in the location marked in Figure 2.5), surface waters enriched with C_{ant} are redistributed over deeper mixed layers and are diluted by subsurface waters containing a lower C_{ant} burden. This dilution causes the winter surface DIC value to increase more slowly than in summer. As a result, seasonal C_{ant} redistribution effectively reduces the surface ocean winter-to-summer (maximum-to-minimum) range in DIC and decreases the upper-ocean vertical DIC gradient in these regions, independent of climate-induced changes in ocean mixing. Reductions in A-DIC will cause reductions in A- $p\text{CO}_2$ BP; however, the C_{ant} -induced decrease in A-DIC over this time period is relatively small compared to the A-DIC in the reference period (<10%; Figure S2.4). Thus, the magnitude and phasing of A-DIC in 1980-1984 are largely preserved at the end of the simulation, in the absence of climate change (Figure 2.5b). The spatial patterns of the C_{ant} -driven increase in A- $p\text{CO}_2$ BP are therefore primarily explained by the 1980-1984 A-DIC distribution and the enhanced sensitivity of $p\text{CO}_2$ to DIC variations via AM RF changes.

In contrast to the effect of C_{ant} accumulation, the effect of climate change on thermal and biophysical $p\text{CO}_2$ seasonal cycle amplitudes can be primarily explained by changes in SST and DIC seasonal drivers (Figures 2.3c and 2.4c), with smaller changes to sensitivity drivers (Figure S2.2). As there are no changes in surface temperature seasonality caused by C_{ant} alone, all of the changes in A-SST between 1980 and 2018

are associated with climate change (Figure 2.3e). Similarly, because the C_{ant} -effect on DIC seasonality is relatively small (Figure 2.5; Figure S2.4), the Δ_{clim} A-DIC is responsible for the majority of the total A-DIC change (Figure 2.4e; Figure S2.2). As a result, the spatial variability in Δ_{clim} A- $p\text{CO}_2$ T and Δ_{clim} A- $p\text{CO}_2$ BP maps to that of Δ_{clim} A-SST and Δ_{clim} A-DIC, respectively.

Over the study period, C_{ant} accumulation is responsible for much of the changes in A- $p\text{CO}_2$ T and A- $p\text{CO}_2$ BP (Figures 2.3a and 2.4a), which ultimately increases the overall A- $p\text{CO}_2$. Seasonal $p\text{CO}_2$ amplitudes increase the most in the Northern and Southern Hemisphere mid-latitudes ($\sim 25\text{--}40^\circ$; Figure 2.2a), largely reflecting the increase in the thermal component amplitude in those regions (Figure 2.3a). The enhanced $p\text{CO}_2$ sensitivity to underlying seasonal thermal, biological, and physical processes, caused by C_{ant} -driven AM $p\text{CO}_2$ and AM RF increases, gives more chemical leverage to seasonal SST and DIC variations, which themselves respond primarily to climate perturbations. In equatorial regions, where $p\text{CO}_2$ seasonality is generally small, and in subpolar high latitudes, where there are large changes in SST and DIC seasonality, the effect of climate change may outweigh the $p\text{CO}_2$ amplification caused by C_{ant} . The total A- $p\text{CO}_2$ T increase is greater than that of A- $p\text{CO}_2$ BP in much of the surface ocean, owing to a larger increase caused by C_{ant} and a smaller opposing effect of climate change when globally integrated. These findings are in agreement with Rodgers et al. (2023), which employs a complementary approach to attribute the anthropogenic changes in $p\text{CO}_2$ seasonality using the same GOBMs, and with 21st century projections of $p\text{CO}_2$ seasonality change made using ESMs (Fassbender et al.,

2022; Gallego et al., 2018). We have illustrated the spatial patterns and magnitudes of anthropogenic change using one GOBM (CESM-ETHZ); however, the mechanisms of A- $p\text{CO}_2$ change hold for all GOBMs (Figure 2.6; Figures S2.2 and S2.3).

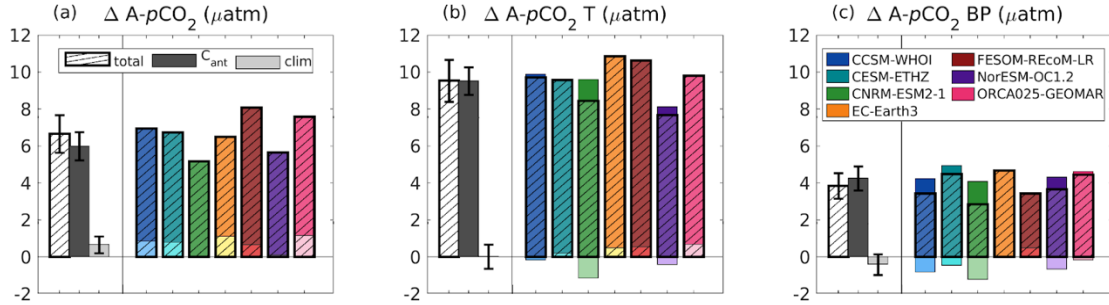


Figure 2.6. Area-normalized global mean changes in the (a) surface ocean $p\text{CO}_2$ seasonal cycle amplitude ($\Delta A-p\text{CO}_2$) from 1980-1984 to 2014-2018 and its decomposition into changes in the (b) thermal ($\Delta A-p\text{CO}_2 T$) and (c) biophysical ($\Delta A-p\text{CO}_2 BP$) $p\text{CO}_2$ seasonal cycle components, as simulated by the GOBMs. In panels (a-c), GOBM ensemble-mean ($n=7$) changes are shown in grayscale, with error bars representing ± 1 standard deviation, and individual GOBMs are shown in color. The total amplitude change (hashed) is broken down into its contributions from anthropogenic carbon accumulation (C_{ant} , dark shading) and climate change (clim, light shading). Sea ice regions (Arctic and subpolar Southern Ocean) are excluded from the global averaging.

2.4.2 Drivers of Across-Model Differences in $p\text{CO}_2$ Seasonal Cycle Amplitude

Change

On average, GOBMs simulate a global increase in the total $p\text{CO}_2$ seasonal cycle amplitude of $6.6 \pm 1.1 \mu\text{atm}$, with $5.9 \pm 0.6 \mu\text{atm}$ increase caused by C_{ant} accumulation and $0.7 \pm 0.5 \mu\text{atm}$ increase due to climate change, over the nearly 40 year period (Figure 2.6; excluding sea ice regions in the Arctic and Southern Ocean). On a global scale, the C_{ant} response is strengthened by A- $p\text{CO}_2$ changes caused by climate change over recent decades, though the fractional contribution of climate is inconsistent across models (Figure 2.6a). We consider the factors responsible for differing spatial structures and magnitudes of seasonal $p\text{CO}_2$ amplitude change between models by

examining the A- $p\text{CO}_2$ T and A- $p\text{CO}_2$ BP responses to their respective seasonality (A-SST and A-DIC) and sensitivity driver (AM $p\text{CO}_2$ and AM RF) changes.

Although globally-averaged total changes in A- $p\text{CO}_2$, A- $p\text{CO}_2$ T, and A- $p\text{CO}_2$ BP are dominated in magnitude by the C_{ant} response, the across-model differences in the spatial pattern of local Δ_{total} A- $p\text{CO}_2$ T and Δ_{total} A- $p\text{CO}_2$ BP are primarily related to the differences caused by climate-induced change (Figure 2.7). Local across-model correlation coefficients for regressions of the simulated Δ_{clim} A-SST and Δ_{clim} A-DIC with Δ_{total} A- $p\text{CO}_2$ T and Δ_{total} A- $p\text{CO}_2$ BP, respectively, are more tightly positively correlated (Figure 2.7a and 2.7b) compared to the relationships between Δ_{Cant} AM $p\text{CO}_2$ and Δ_{Cant} AM RF and Δ_{total} A- $p\text{CO}_2$ T and Δ_{total} A- $p\text{CO}_2$ BP, respectively (Figure 2.7c and 2.7d). This is because the increase in mean-state sensitivity terms caused by C_{ant} accumulation serves to coherently amplify the underlying seasonal structure of the $p\text{CO}_2$ component cycles, and models exhibit broadly similar spatial patterns of both the 1980-1984 seasonal variability drivers and C_{ant} -induced sensitivity term changes (in spite of model biases in magnitude; Figures S2.2 and S2.3). In contrast, climate change predominantly modifies seasonal SST and DIC cycle amplitudes to expand or contract local seasonal $p\text{CO}_2$ component amplitudes, independent of the amplifying effects of C_{ant} accumulation. As a result, across-model differences in the spatial patterns of Δ_{total} A- $p\text{CO}_2$ T and Δ_{total} A- $p\text{CO}_2$ BP, and thus the overall Δ_{total} A- $p\text{CO}_2$, are better explained by differences in the spatial pattern of climate-induced change that strengthen or diminish the coherent $p\text{CO}_2$ amplification caused by C_{ant} (Figures 2.3d and 2.4d). The across-model relationship between Δ_{clim} A-SST and Δ_{total} $p\text{CO}_2$ T (global mean $\pm 1\sigma$: r

$= 0.91 \pm 0.12$) is statistically more robust than that between Δ_{clim} A-DIC and Δ_{total} $p\text{CO}_2$ BP (global mean $\pm 1\sigma$: $r = 0.54 \pm 0.39$; Figure S2.5), as climate-driven changes in biophysical $p\text{CO}_2$ seasonality may also be impacted by changes in seasonal salinity or total alkalinity cycles that we do not fully explore. In contrast, while the relationships with sensitivity driver changes appear strong, albeit patchy, the local relationships between Δ_{cant} AM $p\text{CO}_2$ and Δ_{total} $p\text{CO}_2$ T and between Δ_{cant} AM RF and Δ_{total} $p\text{CO}_2$ BP are generally not statistically significant. This is unsurprising as the seasonal variability drivers exert greater control over the $p\text{CO}_2$ seasonal cycle amplitude than the sensitivity drivers, as demonstrated by the contours in Figure 2.1.

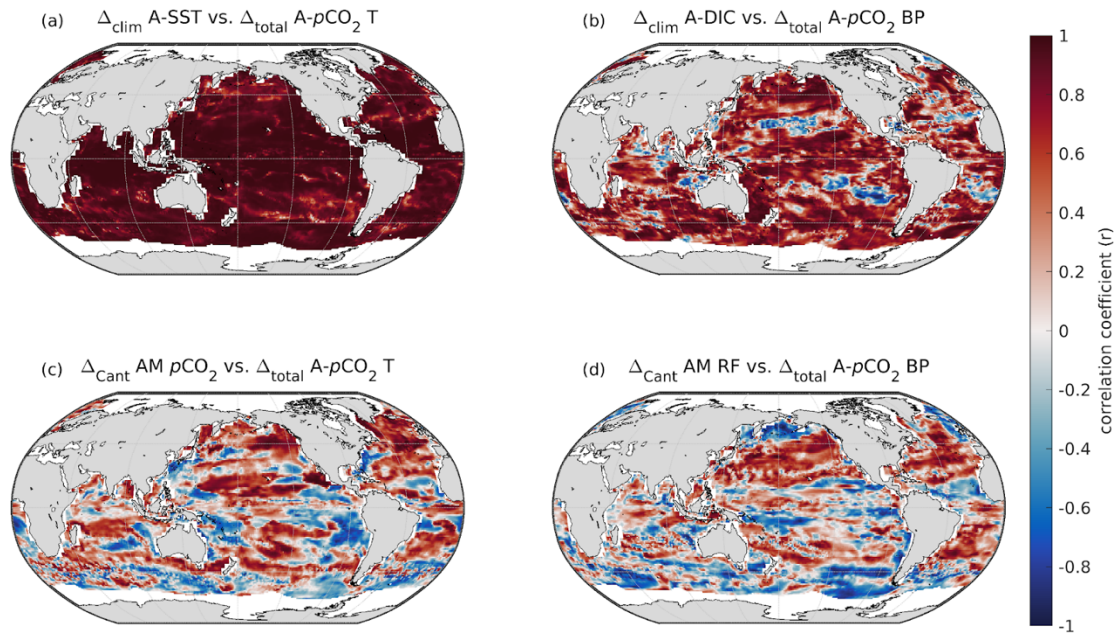


Figure 2.7. The across-model local correlation coefficient (r) for regressions of (a) Δ_{clim} A-SST vs. Δ_{total} A- $p\text{CO}_2$ T, (b) Δ_{clim} A-DIC vs. Δ_{total} A- $p\text{CO}_2$ BP, (c) Δ_{cant} AM $p\text{CO}_2$ vs. Δ_{total} A- $p\text{CO}_2$ T, and (d) Δ_{cant} AM RF vs. Δ_{total} A- $p\text{CO}_2$ BP.

The across-model differences in global mean climate-driven changes in A- $p\text{CO}_2$ T are somewhat larger than the associated A- $p\text{CO}_2$ BP changes, though not statistically different (Figure 2.6b and 2.6c; GOBM-ensemble global mean $\pm 1\sigma$: Δ_{clim}

$\Delta_{\text{A-}p\text{CO}_2 \text{ T}} = 0.02 \pm 0.65 \text{ } \mu\text{atm}$ versus $\Delta_{\text{clim A-}p\text{CO}_2 \text{ BP}} = -0.41 \pm 0.57 \text{ } \mu\text{atm}$). The global mean $\Delta_{\text{clim A-}p\text{CO}_2 \text{ T}}$ is near zero when averaged over the GOBM ensemble, with three models simulating a dampening effect and four models simulating an amplifying effect (Figure 2.6b). In contrast, $\Delta_{\text{clim A-}p\text{CO}_2 \text{ BP}}$ largely works against C_{ant} -driven changes on the global scale in many models (5 out of 7; Figure 2.6c). There are, however, substantial across-model regional differences in the sign of $\Delta_{\text{clim A-}p\text{CO}_2 \text{ BP}}$, largely caused by different representations of $\Delta_{\text{clim A-DIC}}$ (Figure S2.3). We find that the across-model differences in the spatial patterns of $\Delta_{\text{clim A-DIC}}$ are most strongly related to differences in the climate-driven changes in the seasonal surface salinity variations, particularly in low latitudes, and in seasonal biological processes, as inferred from the change in the seasonal amplitude of salinity-normalized surface nitrate; seasonal alterations in mixed layer depths between time periods appear to play a smaller role (Figure S2.6). The climate-driven changes in A-DIC tend to be largest and differ the most between models in low latitudes (e.g., Figure 2.4e; Figure S2.3), where the hydrological cycle is most active. Substantial recent decadal changes in precipitation patterns and upper-ocean circulation have developed in the Equatorial Pacific (Tuchen et al., 2024; Watanabe et al., 2024) resulting from an undetermined combination of natural climate variability and forced climate change. Net surface freshwater flux perturbations are expected to impact DIC concentrations (Hogikyan & Resplandy, 2024; Williams et al., 2007). These modeled differences in $\Delta_{\text{clim A-DIC}}$ over the study period may have important implications for differences in future ocean change. In projections of $p\text{CO}_2$ seasonality change in large-ensemble simulations of one ESM,

Fassbender et al. (2022) found that only ~50% of the surface ocean A- $p\text{CO}_2$ BP amplification by 2100 can be attributed to C_{ant} invasion, emphasizing the role of climate change in shaping the future carbonate system response to anthropogenic perturbations. Since climate change primarily drives the total A-DIC change in each model (Figure S2.2) and dictates the inter-model differences in the spatial pattern of Δ_{total} A- $p\text{CO}_2$ BP over the study period (Figure 2.7b), we expect that differing model responses to climate-induced changes in ocean physics and biological processes may become increasingly relevant. Although climate change imparts differing surface spatial changes upon the seasonal $p\text{CO}_2$ components, the influence of climate change largely works to reinforce the C_{ant} -driven amplification of the global mean, ensemble-mean A- $p\text{CO}_2$ over the study period (Figure 2.6a), reflecting the compensation of thermal and biophysical A- $p\text{CO}_2$ terms.

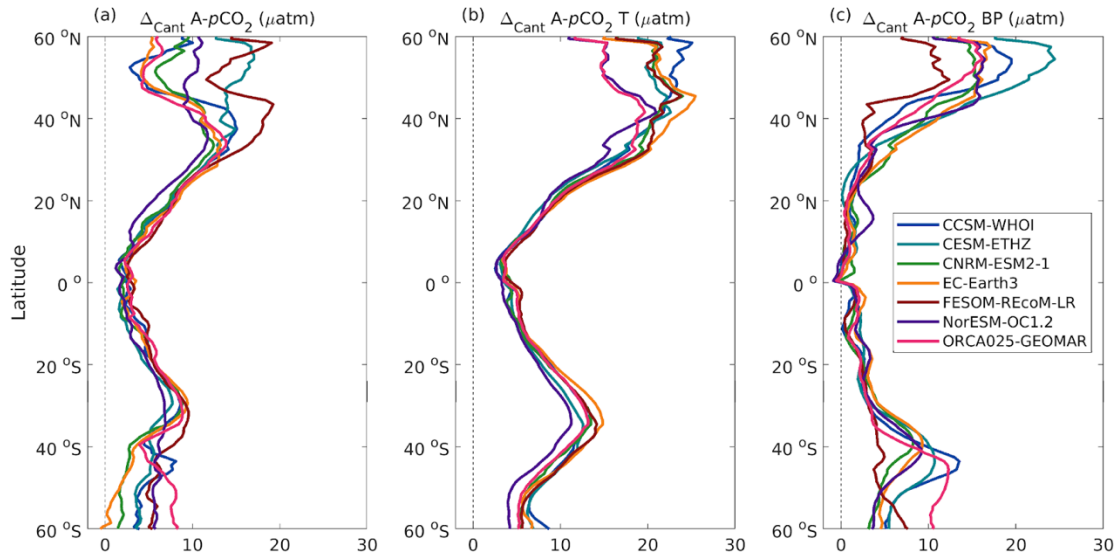


Figure 2.8. The zonally-averaged total C_{ant} -driven change in the seasonal cycle amplitudes of (a) $p\text{CO}_2$ (Δ_{Cant} A- $p\text{CO}_2$) and its (b) thermal (Δ_{Cant} A- $p\text{CO}_2$ T) and (c) biophysical (Δ_{Cant} A- $p\text{CO}_2$ BP) components for individual GOBMs. Sea ice regions (Arctic and subpolar Southern Ocean) are excluded from the zonal averaging.

While across-model discrepancies in the climate response can help explain contrasting model spatial patterns of $\Delta_{\text{total}} \text{A-}p\text{CO}_2 \text{ T}$ and $\Delta_{\text{total}} \text{A-}p\text{CO}_2 \text{ BP}$, the overall contribution of climate-forced alterations to the magnitude of $\Delta_{\text{total}} \text{A-}p\text{CO}_2$ is small on the global scale. Rather, accumulated C_{ant} and the associated increase in chemical leverage to underlying seasonal processes predominantly controls the magnitude of $\Delta_{\text{total}} \text{A-}p\text{CO}_2$. Although models generally agree on the latitudinal structure of C_{ant} -driven $p\text{CO}_2$ seasonal cycle expansions, there are outstanding regional disagreements, as reflected in the zonal mean $\Delta_{\text{Cant}} \text{A-}p\text{CO}_2$ and its components for individual GOBMs (Figure 2.8). Models generally simulate similar magnitudes of $\Delta_{\text{Cant}} \text{A-}p\text{CO}_2$ in tropical and subtropical latitudes (30°S to 30°N) where background $p\text{CO}_2$ seasonality is generally small, but this agreement breaks down moving towards temperate and subpolar latitudes, particularly for the biophysical component (Figure 2.8c). Prior work aiming to evaluate inter-model differences in annual anthropogenic air-sea CO_2 fluxes has emphasized the role of model biases in the mean-state sensitivity fields, namely the AM $p\text{CO}_2$ and AM RF (e.g., Terhaar et al., 2022). We aim to highlight the role of seasonality biases in driving across-model disagreements in C_{ant} -induced $p\text{CO}_2$ seasonal cycle amplitude change and ultimately seasonal sea-air CO_2 fluxes. To do so, we hereafter consider the steady-state effect of rising atmospheric CO_2 alone in altering $p\text{CO}_2$ seasonal cycles (denoted as $\text{C}_{\text{ant-ss}}$) and its drivers using *Simulation C* (Eq 3.; Table 2.2) and exclude the small heterogenous influence of nonlinear non-steady-state interactions between C_{ant} and climate change (Figure S2.7).

The across-model differences in C_{ant} -driven changes to $p\text{CO}_2$ component amplitudes shown in Figure 2.8 can be explained by the increased sensitivity of $p\text{CO}_2$ (via rising AM $p\text{CO}_2$ and AM RF) to its seasonal variability drivers (A-SST and A-DIC) and by differences in the magnitude of seasonal driver variations themselves (Figure 2.9). Under rising atmospheric CO_2 alone, seasonal variations in SST remain unaltered from initialized conditions, and DIC seasonal cycles experience regionally small C_{ant} -driven changes over the study period (Figure 2.5). Thus, a given model's 1980-1984 spatial structure and magnitude of A-SST and A-DIC respectively dictate the A- $p\text{CO}_2$ T and A- $p\text{CO}_2$ BP patterns over time, incorporating systematic biases from post spin-up conditions. Models that are biased high (low) with respect to the initialized A-SST or A-DIC simulate persistently larger (smaller) A- $p\text{CO}_2$ T or A- $p\text{CO}_2$ BP growth caused by C_{ant} invasion, as indicated by high, positive correlation coefficients in Figures 2.9a and 2.9b. These relationships are nearly global for both the thermal (global mean $\pm 1\sigma$: $r = 0.82 \pm 0.25$) and biophysical (global mean $\pm 1\sigma$: $r = 0.44 \pm 0.40$) $p\text{CO}_2$ components. Patchy relationships between the 1980-1984 A-DIC and $\Delta_{\text{Cant-SS}}$ A- $p\text{CO}_2$ BP near the equator and in mid-latitudes indicate where interannual variability may be large. The across-model differences in $\Delta_{\text{Cant-SS}}$ AM $p\text{CO}_2$ and $\Delta_{\text{Cant-SS}}$ AM RF appear to correlate positively with $\Delta_{\text{Cant-SS}}$ A- $p\text{CO}_2$ T and $\Delta_{\text{Cant-SS}}$ A- $p\text{CO}_2$ BP in large parts of the open ocean (Figures 2.9c and 2.9d), but are less influential than across-model differences in the 1980-1984 A-SST and A-DIC (Figure S2.8). Models exhibit a rather consistent meridional pattern in $\Delta_{\text{Cant-SS}}$ AM $p\text{CO}_2$ and $\Delta_{\text{Cant-SS}}$ AM RF and display much larger differences in their initial A-SST and A-DIC conditions (Figure

S2.9). Therefore, while sensitivity term changes are the dominant mechanism of C_{ant} -driven $p\text{CO}_2$ amplification, the initial magnitudes and spatial patterns of A-SST and A-DIC predominantly cause the across-model differences in $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ T}$ and $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ BP}$, respectively. Accordingly, as a result of initial driver conditions, models with larger 1980-1984 A- $p\text{CO}_2$ exhibit proportionally larger C_{ant} -driven A- $p\text{CO}_2$ increases (Figure S2.10). This finding indicates that the spread in A- $p\text{CO}_2$ increases over time, primarily due to across-model differences in initial A-SST and A-DIC conditions (Figure S2.11).

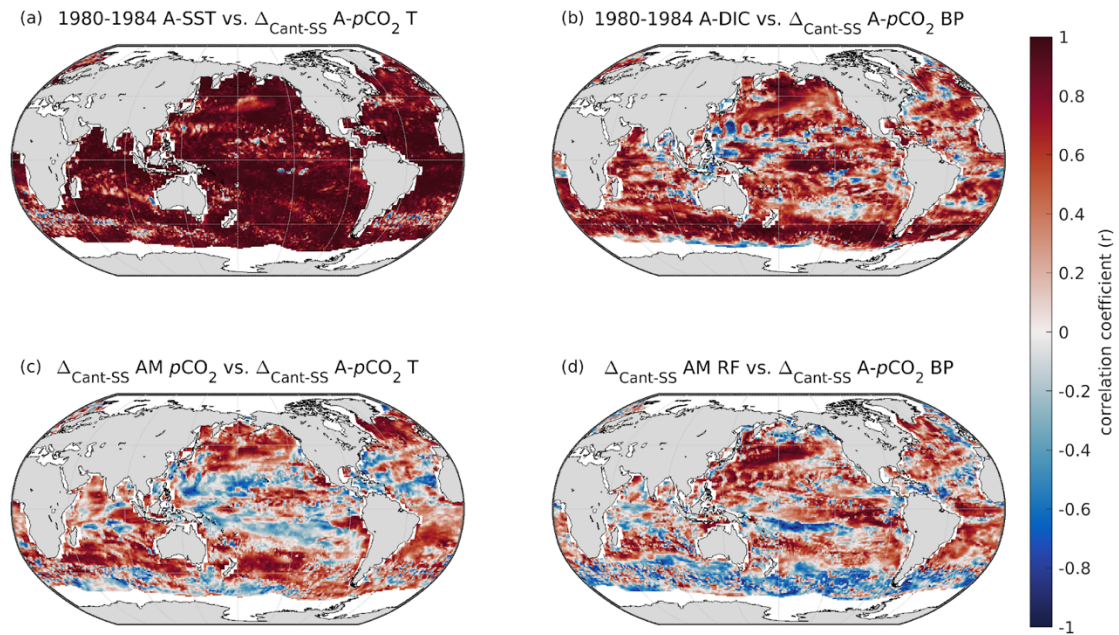


Figure 2.9. The across-model local correlation coefficient for regressions of (a) 1980-1984 A-SST vs. $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ T}$, (b) 1980-1984 A-DIC vs. $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ BP}$, (c) $\Delta_{C_{\text{ant-SS}}} \text{AM } p\text{CO}_2$ vs. $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ T}$, and (d) $\Delta_{C_{\text{ant-SS}}} \text{AM RF}$ vs. $\Delta_{C_{\text{ant-SS}}} A\text{-}p\text{CO}_2 \text{ BP}$. $C_{\text{ant-SS}}$ represents the change caused by the steady-state increase in anthropogenic carbon by the rise in atmospheric CO_2 recorded in *Simulation C* and excludes the heterogeneous, non-steady-state effect of climate change on C_{ant} . Models that are biased high (low) with respect to the starting A-SST or A-DIC render a larger (smaller) $C_{\text{ant-SS}}$ -induced change in A- $p\text{CO}_2 \text{ T}$ or A- $p\text{CO}_2 \text{ BP}$.

The local magnitude of $\Delta_{\text{Cant}} \text{A-}p\text{CO}_2$ in each model reflects the superposition of competing local C_{ant} -driven $\text{A-}p\text{CO}_2$ T and $\text{A-}p\text{CO}_2$ BP changes. Thus, the across-model differences of $\Delta_{\text{Cant}} \text{A-}p\text{CO}_2$ are additionally linked to the spatial pattern of thermal versus biophysical driver dominance. We represent the dominating variability driver by taking the difference between the $\text{A-}p\text{CO}_2$ T and $\text{A-}p\text{CO}_2$ BP (Takahashi et al., 2002), averaged over the analysis period in *Simulation C* (1980-2018; Figure 2.10a). While there is potential for the dominating driver to change over time (e.g., if the $p\text{CO}_2$ component amplitudes increase at different rates), all models maintain the same internal dominating driver over this simulation (Figure S2.12). All considered models agree on thermal driver dominance in the subtropical gyres (Figure 2.10b), where $\text{A-}p\text{CO}_2$ T is larger than $\text{A-}p\text{CO}_2$ BP (Figure 2.10a), indicating that models agree on the seasonal surface $p\text{CO}_2$ phasing (winter $p\text{CO}_2$ lows and summer $p\text{CO}_2$ highs). However, inconsistencies in the dominating driver emerge over northern subpolar gyres and in parts of the equatorial regions and Southern Ocean (yellow regions in Figure 2.10b). In these regions, biophysical processes are expected to dominate, and surface $p\text{CO}_2$ is expected to have opposite seasonal phasing to the subtropics. Model disagreement on the spatial extent of biophysical driver dominance indicates that the GOBMs represent seasonal carbon cycle processes differently, particularly in high-latitude subpolar regions. Rodgers et al. (2023) have shown that, on average, GOBMs underestimate the magnitude of mean-state A-DIC compared to observation-based estimates over much of the surface ocean. This leads to a widespread bias toward thermal driver dominance in most models (their Figure 6; Figure S2.12), resulting in a

systematically larger C_{ant} -driven thermal $p\text{CO}_2$ amplification (Figure 2.6). Discrepancies in surface seasonal DIC variability contribute to persistent inter-model discrepancies in the magnitude and spatial pattern of overall A- $p\text{CO}_2$ changes.

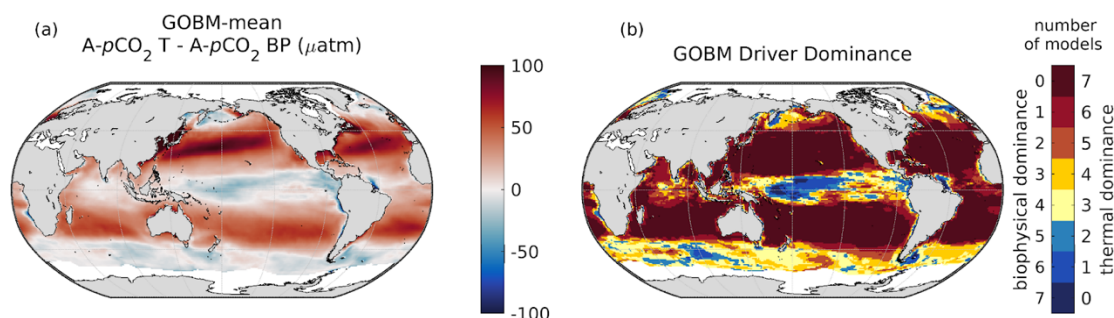


Figure 2.10. (a) GOBM ensemble-mean ($n = 7$) difference between A- $p\text{CO}_2$ T and A- $p\text{CO}_2$ BP averaged over 1980-2018 in *Simulation C*. Positive values indicate where thermal processes dominate over biophysical processes (vice versa). (b) Categorical map reflecting the number of models in agreement that thermal processes (reds, lower winter surface $p\text{CO}_2$) or biophysical processes (blues, higher winter surface $p\text{CO}_2$) dominate the local $p\text{CO}_2$ seasonal cycle. Yellow areas delineate the regions with the largest inter-model disagreement in the dominating driver.

2.4.3 Relevance to Seasonal Sea-Air CO_2 Fluxes and the Ocean Carbon Sink Strength

Alterations to $p\text{CO}_2$ seasonal cycles have the potential to impact the strength of the annual ocean carbon sink. The seasonal variability of sea-air CO_2 fluxes is primarily driven by the gradient in $p\text{CO}_2$ between the surface ocean and atmosphere. Since atmospheric $p\text{CO}_2$ is relatively well mixed and exhibits comparatively small seasonal variations, seasonal sea-air CO_2 fluxes and oceanic C_{ant} uptake are predominantly controlled by the seasonality of surface $p\text{CO}_2$. Accordingly, over the study period, we find that the C_{ant} -driven amplification of A- $p\text{CO}_2$ results in the amplification of seasonal sea-air $p\text{CO}_2$ disequilibrium ($\Delta p\text{CO}_2^{\text{sea-air}}$) over much of the middle and high latitudes ($>20^\circ\text{N}$ and $<20^\circ\text{S}$; Figure 2.11 a-c), resulting in higher highs and lower lows

annually. Near the equator and in high latitudes, $\Delta p\text{CO}_2^{\text{sea-air}}$ decreases in all seasons due to C_{ant} , suggesting that the rise in atmospheric CO_2 is larger than the rise in surface $p\text{CO}_2$. Similarly, as across-model spread in $A\text{-}p\text{CO}_2$ grows over time due to persistent seasonal driver biases (Figure S2.11), so does the spread in $A\text{-}\Delta p\text{CO}_2^{\text{sea-air}}$ (Figure 2.11 d-f), represented by the increases in standard deviation across models. The divergence between model $A\text{-}\Delta p\text{CO}_2^{\text{sea-air}}$ over time reflects that models biased toward larger 1980-1984 $A\text{-}\Delta p\text{CO}_2^{\text{sea-air}}$ exhibit a larger growth in $A\text{-}\Delta p\text{CO}_2^{\text{sea-air}}$ with C_{ant} accumulation (Figure S2.13). The largest increases in spread of seasonal $\Delta p\text{CO}_2^{\text{sea-air}}$ are found at high northern hemisphere latitudes ($>40^\circ\text{N}$), particularly in summer, where models considerably disagree on the dominating $p\text{CO}_2$ seasonal driver and surface $p\text{CO}_2$ phasing (Figure 2.10b).

It is important to note that an increase in the seasonal cycle amplitude of surface $p\text{CO}_2$ or $\Delta p\text{CO}_2^{\text{sea-air}}$ may not always lead to a change in the net annual ocean carbon sink, for example, if the changes in seasonal sea-air CO_2 fluxes compensate for each other. To evaluate the impact of C_{ant} -driven surface $p\text{CO}_2$ and $\Delta p\text{CO}_2^{\text{sea-air}}$ seasonal cycle changes on net seasonal and annual CO_2 fluxes, wind speeds must also be considered. C_{ant} -induced changes to $\Delta p\text{CO}_2^{\text{sea-air}}$ seasonality work to disproportionately increase CO_2 fluxes during the season with maximum wind speeds (Fassbender et al., 2022). Considering that the GOBMs are forced with reanalysis wind speeds constrained by real-world data, the magnitude and spatial pattern of seasonal winds are generally fixed across models. Thus, across-model differences in the seasonal phasing and magnitude of $\Delta p\text{CO}_2^{\text{sea-air}}$, the latter of which increases over time with C_{ant}

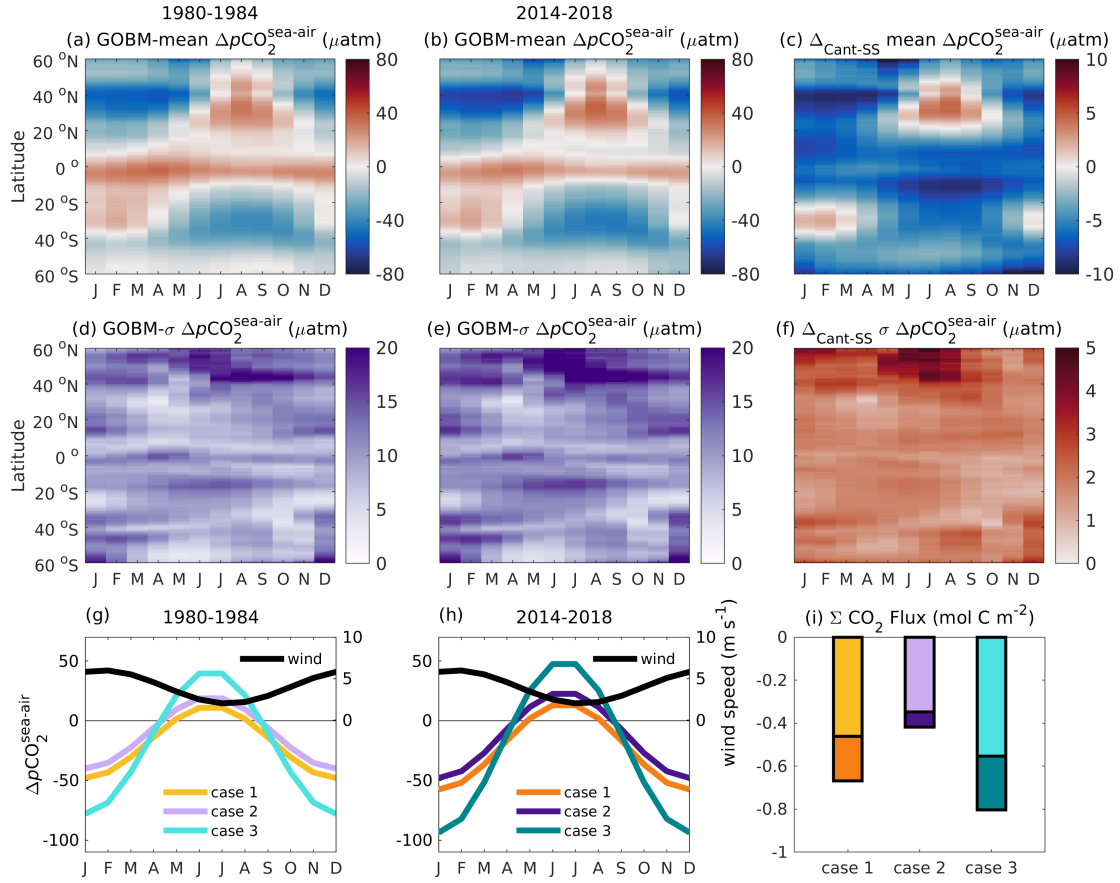


Figure 2.11. Hovmöller plots showing GOBM ensemble-mean zonal averages from 1980-1984 (left) and 2014-2018 (center) and their difference (right) for the steady-state anthropogenic-carbon-induced change ($\Delta_{\text{Cant-SS}}$; *Simulation C*) to (a-c) the sea-air disequilibrium in $p\text{CO}_2$ ($\Delta p\text{CO}_2^{\text{sea-air}}$), where negative values represent the ocean carbon sink, and (d-f) the across-model $\Delta p\text{CO}_2^{\text{sea-air}}$ standard deviation. Theoretical $\Delta p\text{CO}_2^{\text{sea-air}}$ seasonal cycles representative of a thermally-dominated region of the Northern Hemisphere for the (g) 1980-1984 (left) and (h) 2014-2018 (center) periods, with a theoretical seasonal cycle of wind speed (black line). Case 1 (oranges) and Case 2 (purples) simulate the same seasonal cycle amplitude of $\Delta p\text{CO}_2^{\text{sea-air}}$, but Case 2 has a higher annual mean value (+8 μatm from Case 1). Case 1 and Case 3 (teals) simulate the same annual mean $\Delta p\text{CO}_2^{\text{sea-air}}$, but Case 3 has a larger seasonal cycle amplitude (2x larger than Case 1). The seasonal cycle changes in $\Delta p\text{CO}_2^{\text{sea-air}}$ between (g) and (h) are fixed to 20% and represent the impact of $\text{C}_{\text{ant-SS}}$ accumulation. (i) The theoretical cumulative annual ocean CO_2 sink (ΣCO_2 flux) for each time period computed using seasonal $\Delta p\text{CO}_2^{\text{sea-air}}$ and winds from (g) and (h), with the gas transfer parameterization by Wanninkhof et al. 2014. Negative values represent the ocean carbon sink.

accumulation, should theoretically contribute to across-model differences in seasonal CO_2 fluxes. The question is then whether modeled seasonal CO_2 flux differences can help to explain historical annual ocean carbon sink differences between GOBMs. Before addressing this question, we use the following theoretical example to illustrate

how $p\text{CO}_2$ seasonality biases can work alongside annual mean-state biases to alter the annual ocean carbon sink under seasonally-varying wind speeds (Figure 2.11 g-i).

In regions where the thermal $p\text{CO}_2$ component dominates (e.g., subtropics), surface ocean $p\text{CO}_2$ reaches a seasonal low during the colder winter months. The timing of low winter $p\text{CO}_2$ coincides with seasonally high winter wind speeds, making winter the maximum ocean CO_2 sink season in subtropical regions (e.g., Rodgers et al., 2008), even though the largest $\Delta p\text{CO}_2^{\text{sea-air}}$ is often in summer (Fassbender et al. 2018; 2022). In our theoretical example, we consider three different scenarios of seasonal $p\text{CO}_2$ cycles with the same seasonal phasing in $\Delta p\text{CO}_2^{\text{sea-air}}$ but differing degrees of bias in the seasonal cycle amplitude and annual mean value (Figure 2.11 g-i). Case 1 (oranges) and Case 2 (purples) exhibit the same seasonal cycle amplitude of $\Delta p\text{CO}_2^{\text{sea-air}}$, but Case 2 has a higher annual mean value (+8 μatm from Case 1). Case 1 and Case 3 (teals) exhibit the same annual mean $\Delta p\text{CO}_2^{\text{sea-air}}$ value, but Case 3 has a larger seasonal cycle amplitude (2x larger than Case 1). In all cases, $\Delta p\text{CO}_2^{\text{sea-air}}$ reaches a seasonal low in winter and seasonal high in summer, indicative of thermal driver dominance. Here, Case 3 is theoretically characterized by a larger A-SST and consequently a larger seasonal amplitude of $\Delta p\text{CO}_2^{\text{sea-air}}$. The other cases with smaller A-SST (Case 1 and Case 2) show weaker seasonal extrema. We impose a 20% increase in the $\Delta p\text{CO}_2^{\text{sea-air}}$ seasonal cycle for each theoretical case to simulate the effect of C_{ant} accumulation and compute the change in cumulative annual CO_2 flux under the same seasonal wind speeds. Case 2, with a higher annual mean $\Delta p\text{CO}_2^{\text{sea-air}}$, exhibits the smallest cumulative annual CO_2 sink increase in this example. In contrast, Case 3, with the lowest

wintertime $\Delta p\text{CO}_2^{\text{sea-air}}$, exhibits the largest increase in cumulative annual sea-air CO_2 flux (Figure 2.11i). This is because higher winter wind speeds enhance the wintertime $\Delta p\text{CO}_2^{\text{sea-air}}$ changes, giving greater strength to the CO_2 sink season in this case.

Although this example considers the impact of C_{ant} -induced seasonal $\Delta p\text{CO}_2^{\text{sea-air}}$ changes in the annual carbon sink in thermally-dominated regions, the ocean carbon sink is expected to be enhanced globally by seasonal $p\text{CO}_2$ changes in the GOBMs. Because GOBMs are systematically biased toward thermal dominance (Rodgers et al., 2023; Figure 2.10; Figure S2.12), the thermal $p\text{CO}_2$ component amplification largely outcompetes the biophysical $p\text{CO}_2$ component amplification (Figure 2.6). Thus, in biophysically-dominated regions where the seasonal $p\text{CO}_2$ (and $\Delta p\text{CO}_2^{\text{sea-air}}$) phasing is opposite, the simulated C_{ant} -induced increase in the wintertime $p\text{CO}_2$ high is lower than it should be. This inherent GOBM tendency toward thermal dominance works to increase the annual ocean carbon sink by increasing/reducing the wintertime CO_2 influx/efflux in thermally-/biophysically-dominated regions (e.g., Fassbender et al. 2022). We thus expect that the growing disagreement in seasonal $\Delta p\text{CO}_2^{\text{sea-air}}$ between models, due in part to biases in seasonal driver magnitudes and the spatial pattern of driver dominance, is increasing the across-model discrepancy in seasonal CO_2 fluxes and ultimately the cumulative global ocean carbon sink strength.

With this theoretical framework, we now evaluate how the simulated differences in C_{ant} -induced $A\text{-}p\text{CO}_2$ and seasonal $\Delta p\text{CO}_2^{\text{sea-air}}$ changes between GOBMs relate to the across-model differences in the cumulative annual sea-air CO_2 fluxes ($\sum \text{CO}_2 \text{ Flux}$) over time (Figure 2.12). To do so, we compute the mean change in

$A-pCO_2$, $\Delta pCO_2^{sea-air}$, and cumulative annual sea-air CO_2 flux driven by C_{ant-SS} invasion over consecutive 5-year increments in each model, relative to their respective 1980-1984 mean starting values. This provides an estimate of how each parameter evolves over time within each model, smoothing for interannual variability. The local across-model correlation coefficients between $\Delta_{Cant-SS} A-pCO_2$ and $\Delta_{Cant-SS} \sum CO_2$ Flux do not show a fully coherent relationship over the analysis period (1980-1984 to 2014-2018 global mean $\pm 1\sigma$: $r = -0.10 \pm 0.45$; Figure 2.12a; Figure S2.14). However, within each model, the $\Delta_{Cant-SS} A-pCO_2$ and $\Delta_{Cant-SS} \sum CO_2$ Flux time series are significantly negatively correlated (Figure S2.14). This result directly links increases in $A-pCO_2$ to increases in the annual ocean carbon sink within each model, suggesting that the anticipated mechanism operates within each internally-consistent model framework. The model relationships exhibit decadal rates of carbon sink change ranging from -0.48 to $-0.28 \text{ mol C m}^{-2} \text{ decade}^{-1} \mu\text{atm}^{-1}$ (Table S2.1). These differences are caused by model-specific biases in annual and seasonal carbon cycle representations, which obscure coherent local across-model relationships between $\Delta_{Cant-SS} A-pCO_2$ and $\Delta_{Cant-SS} \sum CO_2$ Flux over the analysis period (Figure 2.12a).

We next evaluate the relationship between C_{ant} -driven changes in winter $\Delta pCO_2^{sea-air}$ and $\sum CO_2$ Flux to highlight the influence of wind speed seasonality in disproportionately modifying seasonal sea-air CO_2 fluxes over time. Local across-model C_{ant} -driven changes in winter $\Delta pCO_2^{sea-air}$ are tightly, positively correlated with changes in $\sum CO_2$ Flux over much of the ocean (Figure 2.12c; global mean $\pm 1\sigma$: $r = -0.44 \pm 0.37$), indicating that GOBMs simulating larger winter $\Delta pCO_2^{sea-air}$ decreases

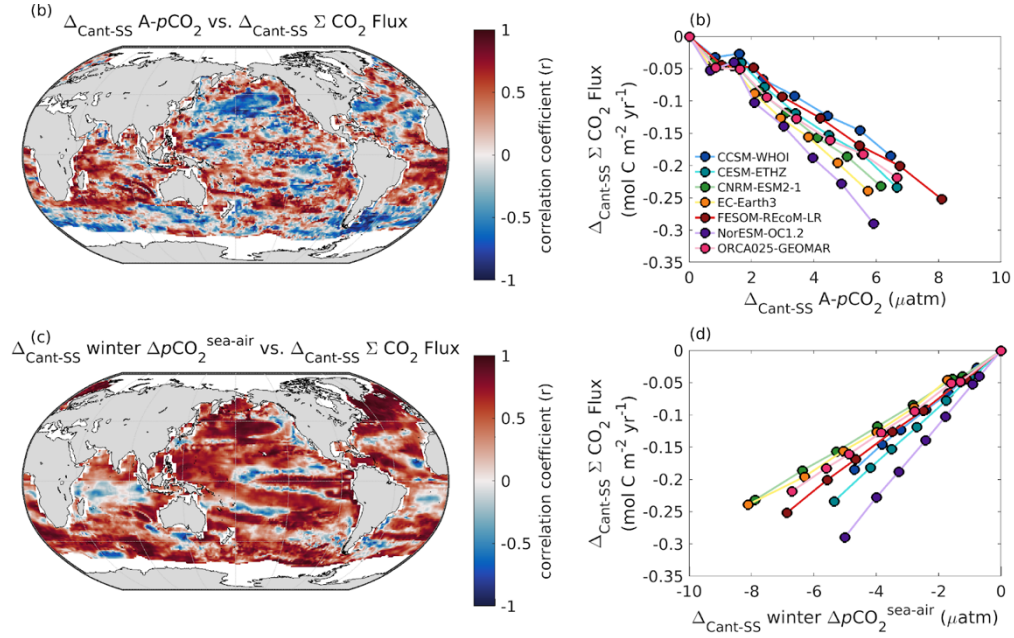


Figure 12. The across-model local correlation coefficient (r) for regressions of the C_{ant} -induced changes ($\Delta_{\text{Cant-SS}}$) to the cumulative annual CO_2 flux (ΣCO_2 flux) versus (a) $\Delta p\text{CO}_2$ or versus (c) the winter sea-air $p\text{CO}_2$ disequilibrium ($\Delta p\text{CO}_2^{\text{sea-air}}$) between 1980-1984 and 2014-2018. $\Delta_{\text{Cant-SS}}$ represents the change caused by the steady-state increase in anthropogenic carbon in *Simulation C*. Winter is defined as January, February, and March in the Northern Hemisphere and as June, July, and August in the Southern Hemisphere. Panels (b) and (d) show the area-weighted global mean relationships over time within each GOBM at five-year increments between 1980 and 2018 (colored circles). Negative values of the $\Delta_{\text{Cant-SS}}$ ΣCO_2 flux (y-axes) indicates that the cumulative carbon sink increases over time. Sea ice regions (Arctic and subpolar Southern Ocean) are excluded from the global averaging.

exhibit larger annual ocean carbon sink increases between 1980-1984 and 2014-2018. The seasonal alignment of strong winter winds with C_{ant} -driven reductions in $\Delta p\text{CO}_2^{\text{sea-air}}$ enhances the seasonal ocean CO_2 sink and the net annual ocean carbon sink strength. While the $\Delta p\text{CO}_2^{\text{sea-air}}$ tends to decrease across all seasons outside of the subtropical regions (Figure 2.11c), indicating a near year-round strengthening of the ocean carbon sink, changes in globally-mean seasonal CO_2 fluxes are largest in winter (Figure S2.15). This mechanistic relationship between winter $\Delta_{\text{Cant-SS}}$ $\Delta p\text{CO}_2^{\text{sea-air}}$ and $\Delta_{\text{Cant-SS}}$ ΣCO_2 Flux holds globally within each model, (Figure 2.12d; Figure S2.16), yielding

increases in the ocean carbon sink strength ranging from 0.29 to 0.58 mol C m⁻² decade⁻¹ μatm⁻¹ (Table S2.1). These results align with our expectation that seasonal biases in carbon cycling representation, combined with changes in mean-state conditions, cause a divergence in winter Δ*p*CO₂^{sea-air} and sea-air CO₂ fluxes between models, leading to an increasing difference in the cumulative annual ocean carbon sink over time.

2.5 Conclusions

Systematic discrepancies in ocean carbon uptake between GOBMs pose an important challenge for the carbon cycle community to resolve, as they have potential implications for the skill of climate model projections. This study aims to test the hypothesis that systematic discrepancies in the net annual ocean carbon sink estimated from GOBMs relative to observational-based products are, in part, sourced from systematic biases in model representations of surface ocean carbon cycle seasonality. To do so, we evaluate the mechanisms responsible for historical changes in the surface ocean *p*CO₂ seasonal cycle amplitude as represented by GOBMs used in synthesis efforts such as the Global Carbon Project (Friedlingstein et al. 2024) and RECCAP2 studies (Devries et al., 2024) and link the across-model differences in C_{ant}-induced *p*CO₂ seasonality change to diverging trends in cumulative annual ocean carbon uptake. Our results largely agree with Rodgers et al. (2023), finding that decadal increases in *p*CO₂ seasonal amplitudes in each model were primarily driven by the invasion of C_{ant} (GOBM-ensemble global mean ± 1σ: +5.9 ± 0.6 μatm) but were modulated by the combined, heterogeneous impact of natural climate variability and forced climate change (+0.7 ± 0.5 μatm). Our analysis demonstrates how C_{ant} accumulation and

climate change independently alter the mean-state chemical conditions (AM $p\text{CO}_2$ and AM RFs) and the underlying seasonal SST and DIC variability drivers of $p\text{CO}_2$ seasonality. The C_{ant} -driven amplification of seasonal $p\text{CO}_2$ cycles is primarily caused by an enhanced sensitivity of $p\text{CO}_2$ to seasonal thermal, biological, and physical processes via rising ocean carbon content and buffering capacity declines. In contrast, the impact of climate change on $p\text{CO}_2$ seasonal amplitudes is largely seen through the alterations of seasonal SST and DIC variations, with relatively minor changes in $p\text{CO}_2$ sensitivities. In some considered models, climate-driven changes in $p\text{CO}_2$ amplitudes over recent decades may outweigh the C_{ant} -induced amplification in equatorial and high latitude regions to cause a net decline in the overall $p\text{CO}_2$ seasonal cycle amplitude.

Our analysis reveals how structural biases in SST and DIC seasonality representations systematically influence the across-model differences in the dominant C_{ant} -driven amplification of the thermal and biophysical $p\text{CO}_2$ seasonal cycles. We identify a direct link between a given model's initial 1980-1984 SST and DIC seasonal cycle amplitudes and the magnitudes of the respective thermal and biophysical $p\text{CO}_2$ component amplitude change by 2014-2018, particularly when the C_{ant} signal is not muddled by climate change. This finding indicates that, while the C_{ant} -induced seasonal $p\text{CO}_2$ amplification within each model is mechanistically driven by a change in the mean-state sensitivity conditions, the across-model differences in the magnitude of change are tied to the differing mean-state seasonal SST or DIC variations themselves (Figure 2.9). Across-model differences in mean-state sensitivity changes were less influential in causing differences in the C_{ant} -driven $p\text{CO}_2$ amplification. This result

further indicates that models that are biased toward larger seasonal cycle amplitudes of SST or DIC simulate a larger C_{ant} -induced increase of the thermal or biophysical $p\text{CO}_2$ seasonal amplitudes, such that simulated $p\text{CO}_2$ component amplitudes become increasingly divergent between models over time. Additionally, the globally-integrated thermal $p\text{CO}_2$ component amplification is larger than that of the biophysical component in all models (Figure 2.6). This in part reflects the findings of Rodgers et al. (2023) in which the same models were shown to be systematically biased toward too-weak DIC seasonality, leading to an overemphasis of temperature-driven $p\text{CO}_2$ seasonality globally. As a result, there is substantial inter-model disagreement in seasonal driver dominance in high-latitude regions where the balance is systematically tipped toward thermal dominance. In consequence, we find that the across-model disagreement in overall C_{ant} -driven $p\text{CO}_2$ amplitude change grows over time, due to (1) increasing differences in the *magnitude* of surface $p\text{CO}_2$ amplitudes caused by differences in the initialized magnitudes of variability driver seasonal cycles (SST and DIC; Figure 2.9; Figure S2.10) and (2) persistent differences in the *spatial pattern* of $p\text{CO}_2$ amplitude change caused by initialized differences in the spatial extent of driver control (Figure 2.10; Figure S2.12). We thus expect that differences in the rate of seasonal $p\text{CO}_2$ amplification between GOBMs in their current configurations will become increasingly more pronounced over time, as C_{ant} invasion will remain as the dominant anthropogenic signal under sustained fossil fuel emissions.

We demonstrate how the growing across-model spread in the transient C_{ant} -induced $p\text{CO}_2$ amplitude change leads to diverging decadal trends in the cumulative

annual global ocean carbon sink between 1980 to 2018 (Figure 2.12). This trend divergence is primarily driven by an increasing across-model disagreement in the C_{ant} -induced change to the wintertime sea-air $p\text{CO}_2$ disequilibrium, which gets disproportionately translated to wintertime CO_2 flux differences by strong winter winds. Since winter is the dominant season for ocean carbon uptake, the diverging wintertime sink enhancements directly contribute to a progressively inconsistent annual ocean carbon sink strength between models over the historical period. An important implication of this key finding is that we have identified a clear link within individual GOBMs between the seasonal carbon cycle and the annual mean-state for sea-air CO_2 fluxes (Figure S2.15). The use of multiple GOBMs was instrumental in establishing and understanding this relationship. We highlight that these timescales are not independent, and thus, persistent seasonal discrepancies in the ocean carbon cycle should be expected to rectify into longer-term annual mean-state trend discrepancies over time.

In closing, we believe that the seasonal carbon cycle may be of great value in evaluating the utility of GOBMs for trends in ocean carbon uptake. In order for GOBMs to be appropriately used for carbon cycle synthesis efforts, the sources of uncertainty (e.g., model structural uncertainty, forcing uncertainty, spin-up uncertainty) should be randomly distributed about the behavior of the real system. One of the difficulties in constraining model uncertainty is that observation-based data products also have many sources of uncertainty, and thus assessing model biases proves to be challenging, and to date, somewhat controversial or even inconclusive. Understanding how the ocean

carbon cycle responds to anthropogenic forcing is not only important for deriving a “best estimate”, in which a multi-model mean may be assumed to average across errors in individual model estimates, but also for the across-model assessment of uncertainty. In their current configurations, the multi-GOBM mean cannot be used to provide a state estimate of the seasonal surface ocean carbon cycle, because the biases are systematic. From this study, we infer that the systematic seasonality biases in these same GOBMs may be expected to play a first-order role in contributing to systematic biases in the annual mean-state conditions, thereby complicating the goal of using these models for carbon cycle synthesis efforts. The results of this study highlight the importance of incorporating more realistic representation of seasonal carbon cycle processes to further refine estimates of the global ocean carbon sink.

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2.7 Supplementary Information

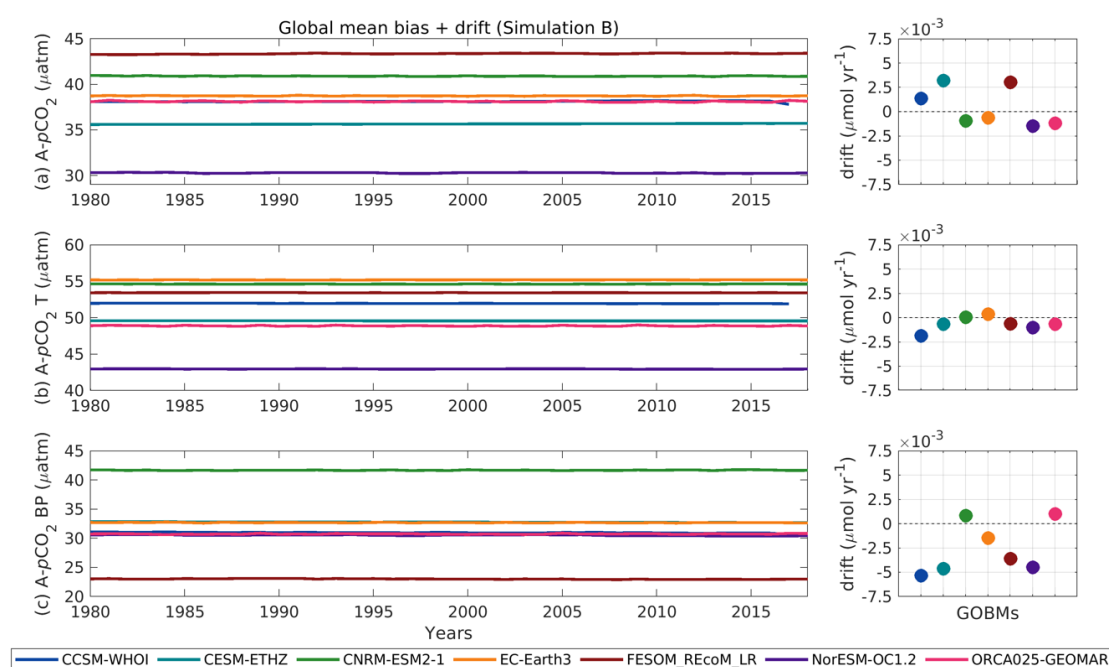


Figure S2.1. Area-weighted global mean seasonal cycle amplitude (A) of (a) $p\text{CO}_2$, (b) the thermal $p\text{CO}_2$ component ($p\text{CO}_2 \text{ T}$), and (c) the biophysical $p\text{CO}_2$ component ($p\text{CO}_2 \text{ BP}$) in control *Simulation B*. Amplitudes are computed as the maximum minus the minimum value of each year. Sea ice covered regions in the Arctic and Southern Ocean were excluded from averages.

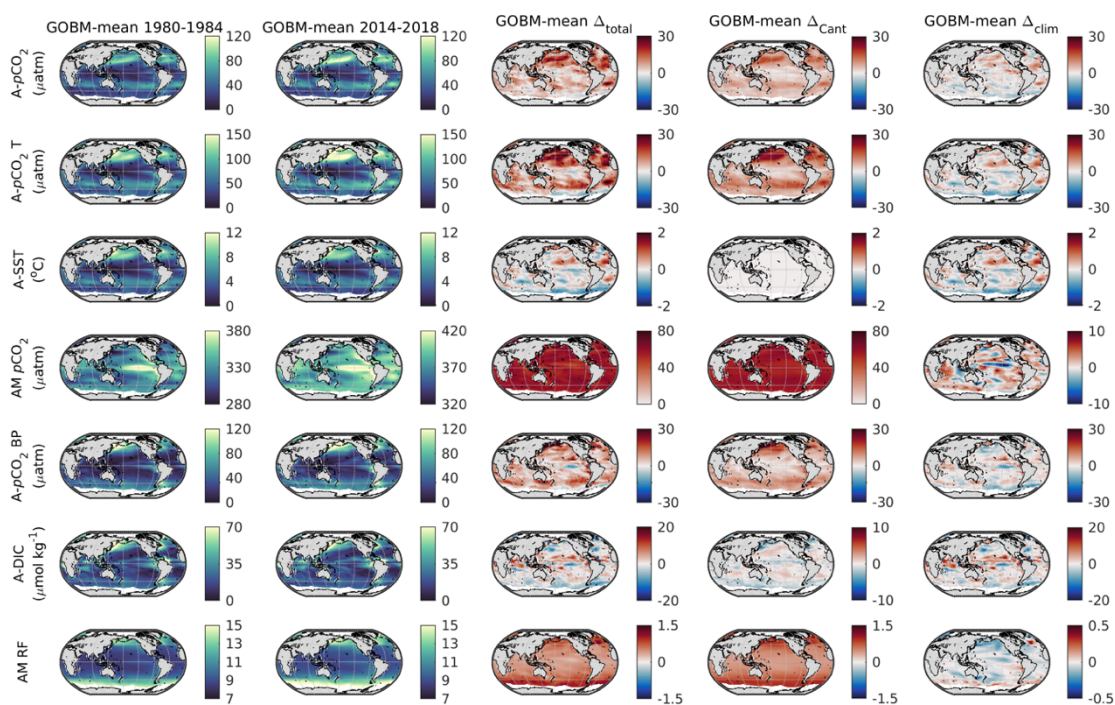


Figure S2.2. GOBM ensemble-mean values for (column 1) 1980-1984 climatology, (column 2) 2014-2018 climatology, (column 3) the total change (Δ_{total}) between the two time periods and its attribution to (column 4) anthropogenic carbon accumulation (Δ_{cant}) and (column 5) climate change (Δ_{clim}). Each row represents a different variable, from top to bottom: A- $p\text{CO}_2$, A- $p\text{CO}_2$ T, the seasonal cycle amplitude of sea surface temperature (A-SST), the annual mean $p\text{CO}_2$ (AM $p\text{CO}_2$), A- $p\text{CO}_2$ BP, the seasonal cycle amplitude of surface dissolved inorganic carbon (A-DIC), and the annual mean Revelle Factor (AM RF).

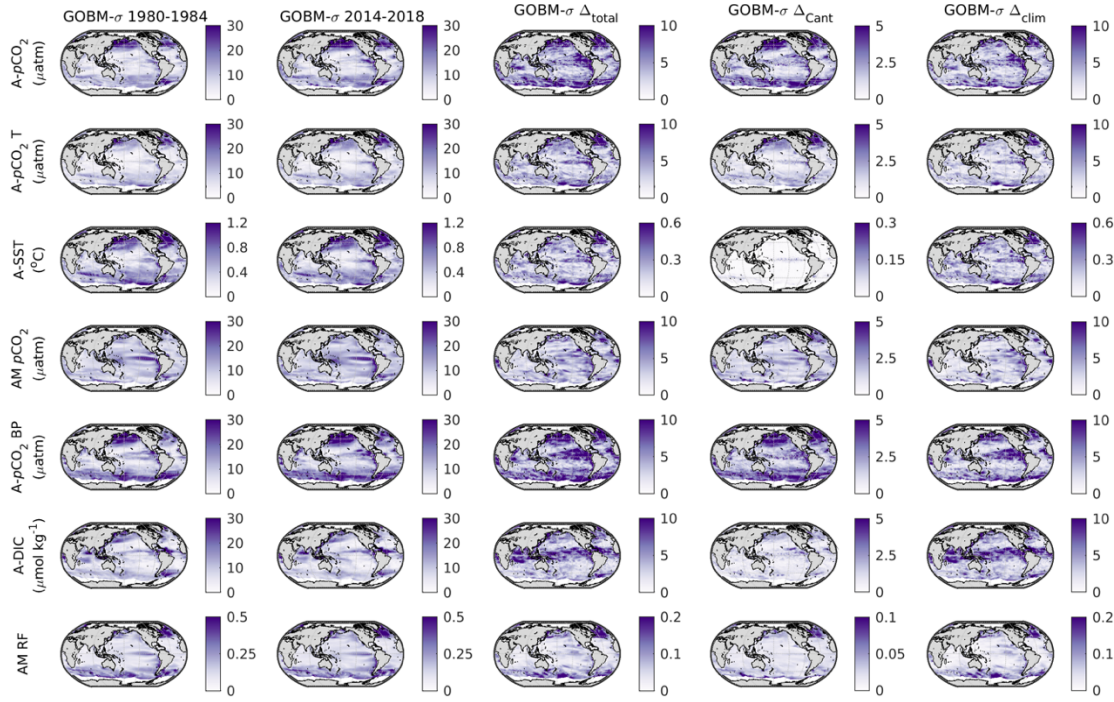


Figure S2.3. Standard deviations about the GOBM ensemble-mean values following Figure S2.2.

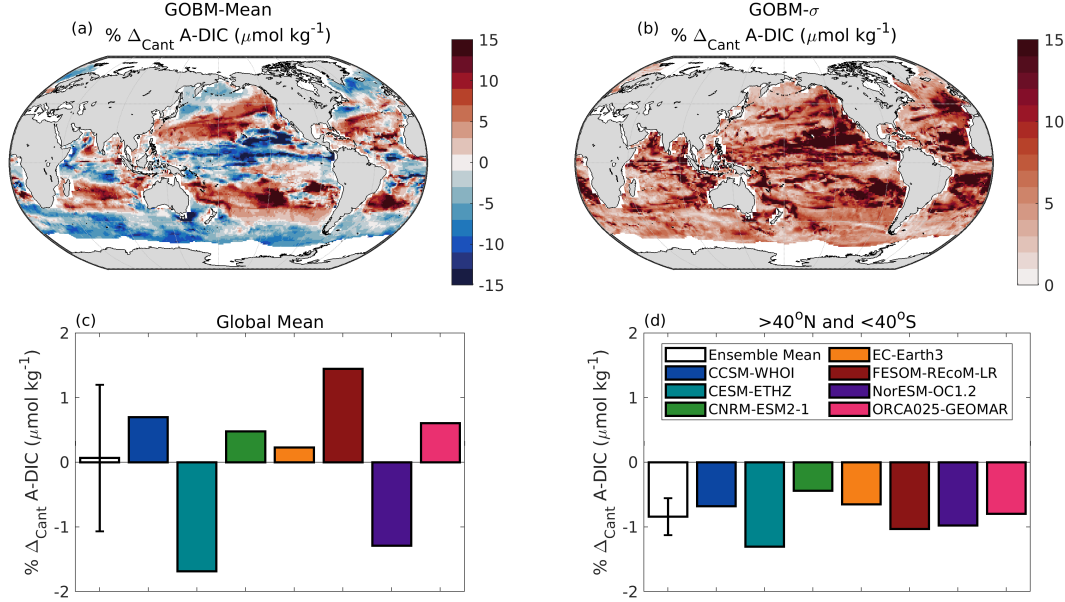


Figure S2.4. (a) GOBM-ensemble mean percent change of the surface ocean DIC seasonal cycle amplitude due to anthropogenic carbon accumulation ($\% \Delta_{\text{Cant}} \text{A-DIC}$) between 1980-1984 and 2014-2018 and (b) the standard deviation of $\% \Delta_{\text{Cant}} \text{A-DIC}$ across models. Area-weighted (c) global mean and (d) high-latitude mean (above 40°N and below 40°S) $\% \Delta_{\text{Cant}} \text{A-DIC}$, represented as the GOBM-ensemble mean and standard deviation (white with black error bar) and for individual GOBMs (colors). Sea ice covered regions in the Arctic and Southern Ocean were excluded from averages.

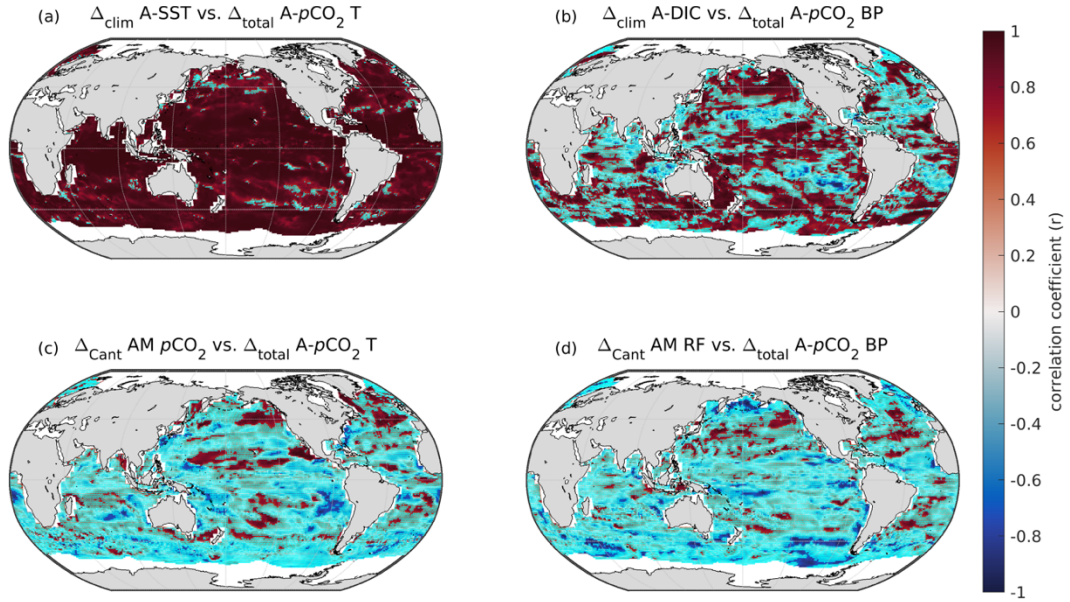


Figure S2.5. The local across-model correlation coefficient (r) for regressions of (a) Δ_{clim} A-SST vs. Δ_{total} A- $p\text{CO}_2$ T, (b) Δ_{clim} A-DIC vs. Δ_{total} A- $p\text{CO}_2$ BP, (c) Δ_{Cant} AM $p\text{CO}_2$ vs. Δ_{total} A- $p\text{CO}_2$ T, and (d) Δ_{Cant} AM RF vs. Δ_{total} A- $p\text{CO}_2$ BP, following Figure 7 in the main text. Cyan regions delineate where relationships are not statistically significant ($p = 0.1$).

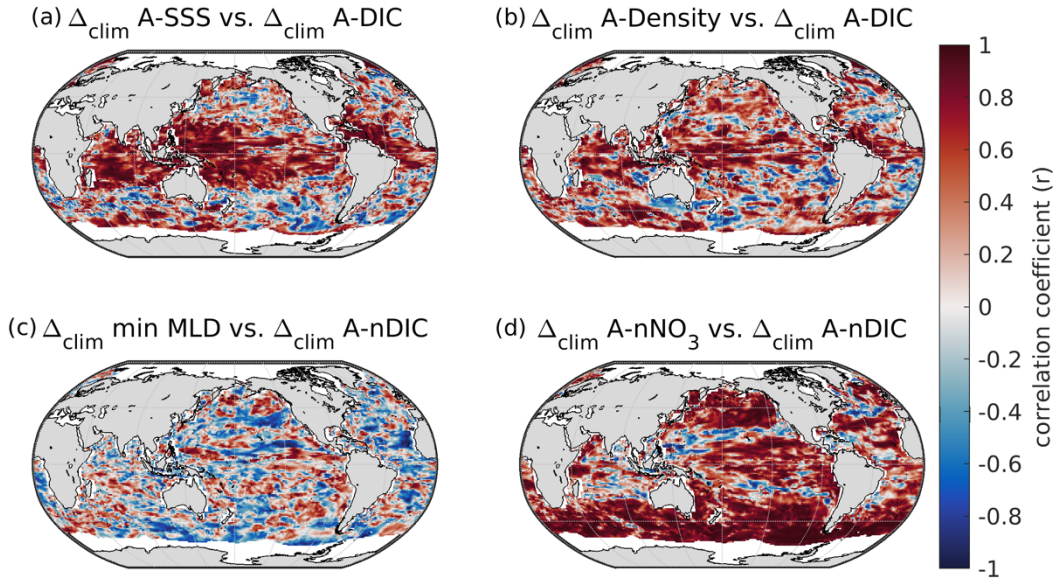


Figure S2.6. Local cross-model correlation coefficients (r) for regressions of the climate-driven changes (Δ_{clim}) of the (a) seasonal cycle amplitudes (A) of sea surface salinity vs. dissolved inorganic carbon (Δ_{clim} A-SSS vs. Δ_{clim} A-DIC), (b) surface density vs. DIC (Δ_{clim} A-Density vs. Δ_{clim} A-DIC), (c) annual minimum mixed layer depth vs. salinity-normalized dissolved inorganic carbon (Δ_{clim} min MLD vs. Δ_{clim} A-nDIC), and (d) salinity-normalized surface nitrate vs. nDIC (Δ_{clim} A-n NO_3 vs. Δ_{clim} A-nDIC) as an estimate of climate-induced change on biological processes. Salinity normalization is referenced to the GOBM-ensemble mean value ($S=34.10$) over the analysis period.

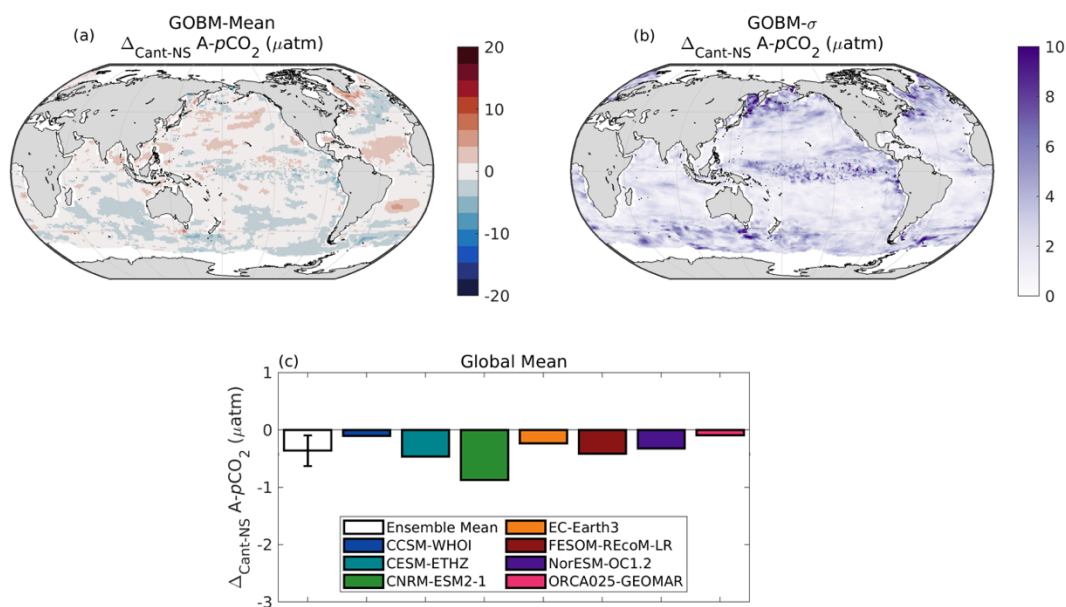


Figure S2.7. (a) GOBM-ensemble mean change in $p\text{CO}_2$ seasonal amplitude due to the non-steady-state (NS) anthropogenic carbon fluxes ($\Delta_{\text{Cant-NS}} A\text{-}p\text{CO}_2$) between 1980-1984 and 2014-2018 and (b) the standard deviation of $\Delta_{\text{Cant-NS}} A\text{-}p\text{CO}_2$ across models. (c) Area-weighted global mean $\Delta_{\text{Cant-NS}} A\text{-}p\text{CO}_2$, represented as the GOBM-ensemble mean and standard deviation (white with black error bar) and for individual GOBMs (colors). Sea ice covered regions in the Arctic and Southern Ocean were excluded from averages.

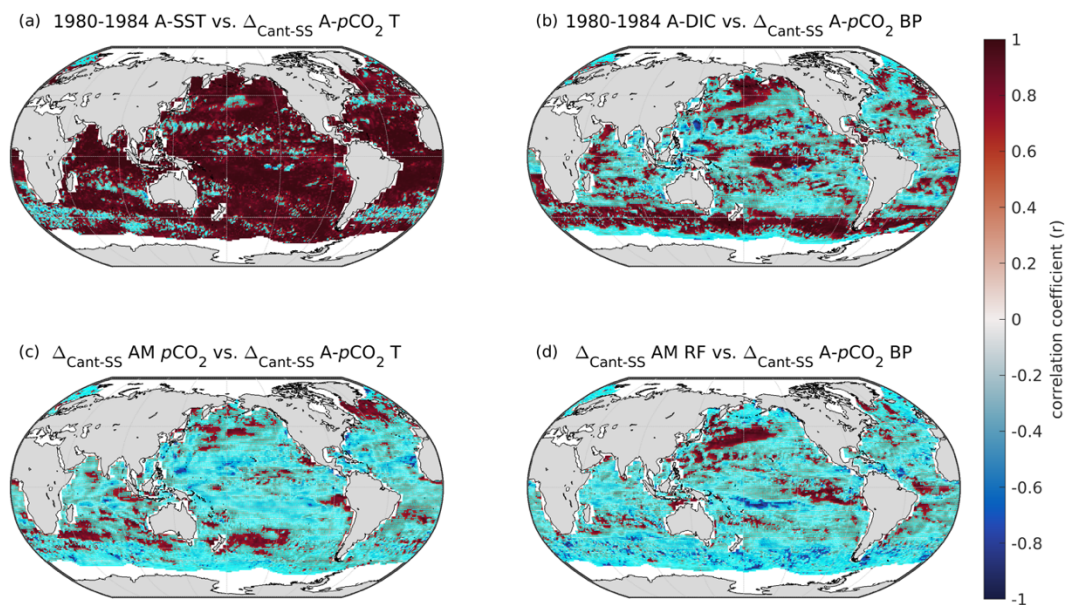


Figure S2.8. The local across-model correlation coefficient for regressions of (a) 1980-1984A-SST vs. $\Delta_{\text{Cant-SS}} A\text{-}p\text{CO}_2$ T, (b) 1980-1984 A-DIC vs. $\Delta_{\text{Cant-SS}} A\text{-}p\text{CO}_2$ BP, (c) $\Delta_{\text{Cant-SS}} \text{AM } p\text{CO}_2$ vs. $\Delta_{\text{Cant-SS}} A\text{-}p\text{CO}_2$ T, and (d) $\Delta_{\text{Cant-SS}} \text{AM RF}$ vs. $\Delta_{\text{Cant-SS}} A\text{-}p\text{CO}_2$ BP, following Figure 9 in the main text. Cant-SS represents the change caused by the steady-state increase in anthropogenic carbon recorded in *Simulation C*. Cyan regions delineate where relationships are not statistically significant ($p = 0.1$).

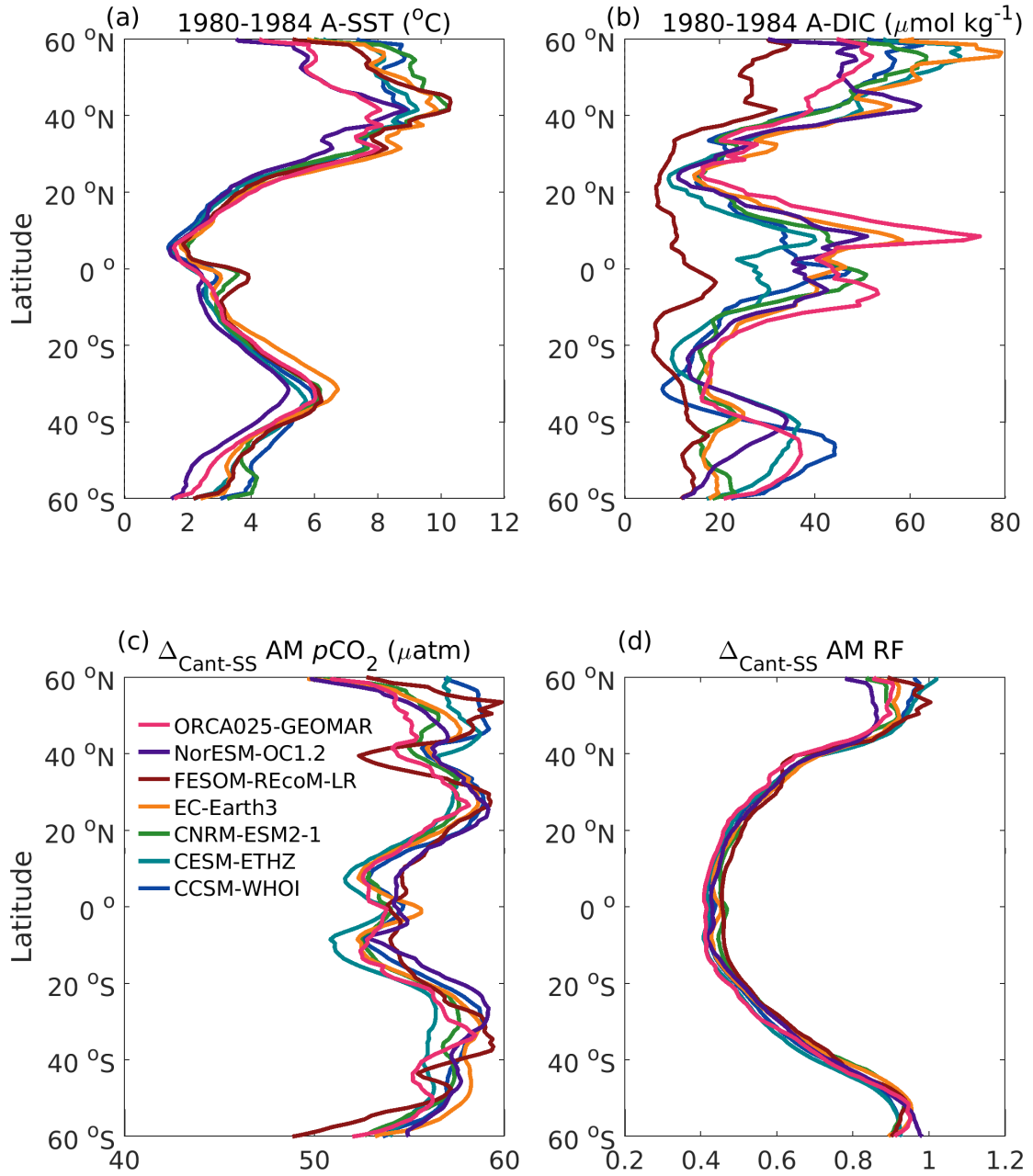


Figure S2.9. The zonally-averaged values of the 1980-1984 seasonal cycle amplitudes (A) of (a) sea surface temperature (A-SST) and (b) dissolved inorganic carbon (A-DIC) in *Simulation C* and the steady-state C_{ant} -driven change in the annual mean (AM) values of (c) $p\text{CO}_2$ ($\Delta_{C_{ant}-SS}$ AM $p\text{CO}_2$) and (b) the Revelle Factor ($\Delta_{C_{ant}-SS}$ AM RF) between 1980-1984 and 2014-2018 in *Simulation C* for individual GOBMs. Sea ice regions (Arctic and subpolar Southern Ocean) are excluded from the zonal averaging.

1980-1984 A- $p\text{CO}_2$ vs. $\Delta_{\text{Cant-SS}}$ A- $p\text{CO}_2$

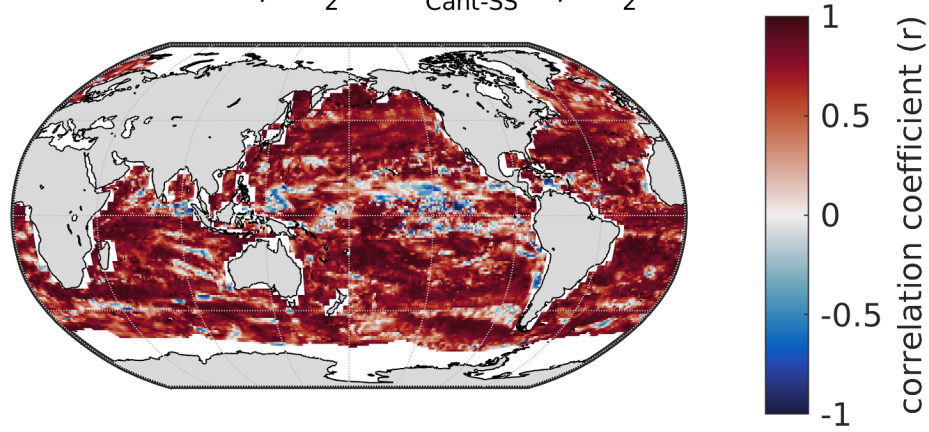


Figure S2.10. The local across-model correlation coefficient for regressions of 1980-1984 A- $p\text{CO}_2$ vs. $\Delta_{\text{Cant-SS}}$ A- $p\text{CO}_2$. $\Delta_{\text{Cant-SS}}$ represents the change caused by the steady-state increase in anthropogenic carbon recorded in *Simulation C*.

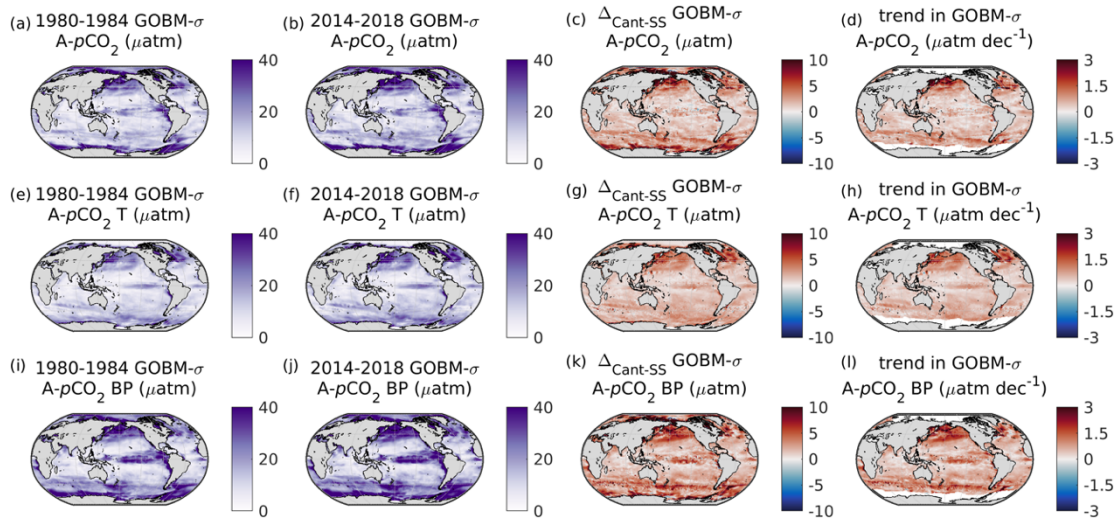


Figure S2.11. The standard deviation (σ) as a metric for across-model spread in the (a,e,i) 1980-1984 period and (b,f,j) 2014-2018 period, the change caused by steady-state C_{ant} accumulation between these two periods (c,g,k), and (d,h,l) local trends ($\mu\text{atm decade}^{-1}$) in the across-model standard deviation from 1980 to 2018 period for the seasonal cycle amplitude (A) of (top) $p\text{CO}_2$ and the (middle) thermal $p\text{CO}_2$ component (A- $p\text{CO}_2$ T) and (bottom) biophysical $p\text{CO}_2$ component (A- $p\text{CO}_2$ BP) in *Simulation C*.

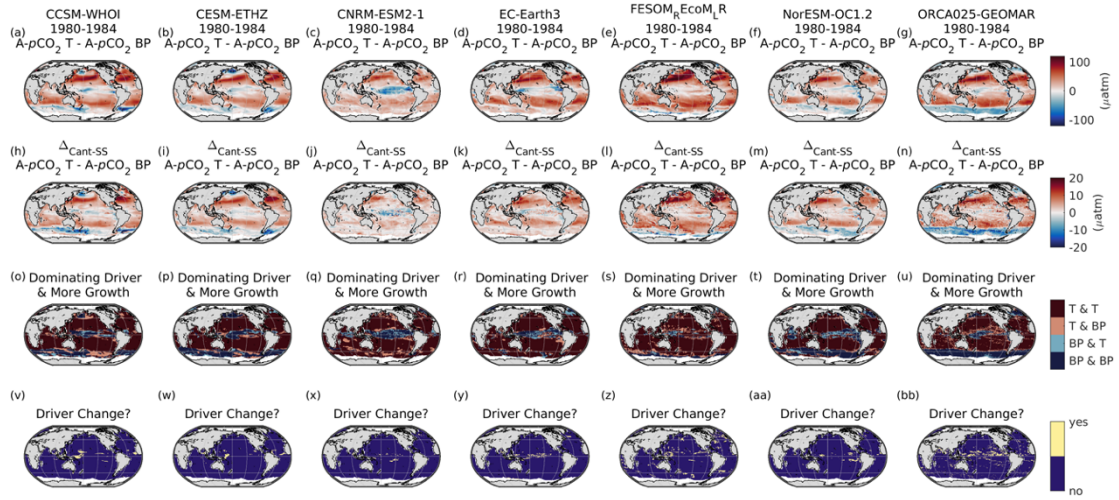


Figure S2.12. For each GOBM (columns), (row 1) the difference between $A-pCO_2$ T and $A-pCO_2$ BP averaged over 1980-1984 in *Simulation C*. Positive values indicate where thermal processes dominate over biophysical processes. The reverse is true for negative values. (row 2) The steady-state effect of anthropogenic carbon accumulation ($\Delta C_{\text{ant-ss}}$) between 1980-1984 and 2014-2018 on the balance between $A-pCO_2$ T and $A-pCO_2$ BP. Positive values indicate that the $A-pCO_2$ T grows more, and negative values indicate that $A-pCO_2$ BP grows more. (row 3) Categorical maps to delineate which driver dominated in 1980-1984 and which driver experienced more growth in seasonal cycle amplitude caused by steady-state C_{ant} between 1980-1984 and 2014-2018. This is labeled as Dominating Driver & More Growth, where T = pCO_2 T and BP = pCO_2 BP. (row 4) The change in driver dominance between pCO_2 T and pCO_2 BP between 1980-1984 and 2014-2018, where blue indicates the dominating driver is maintained and yellow indicates the dominating driver changes.

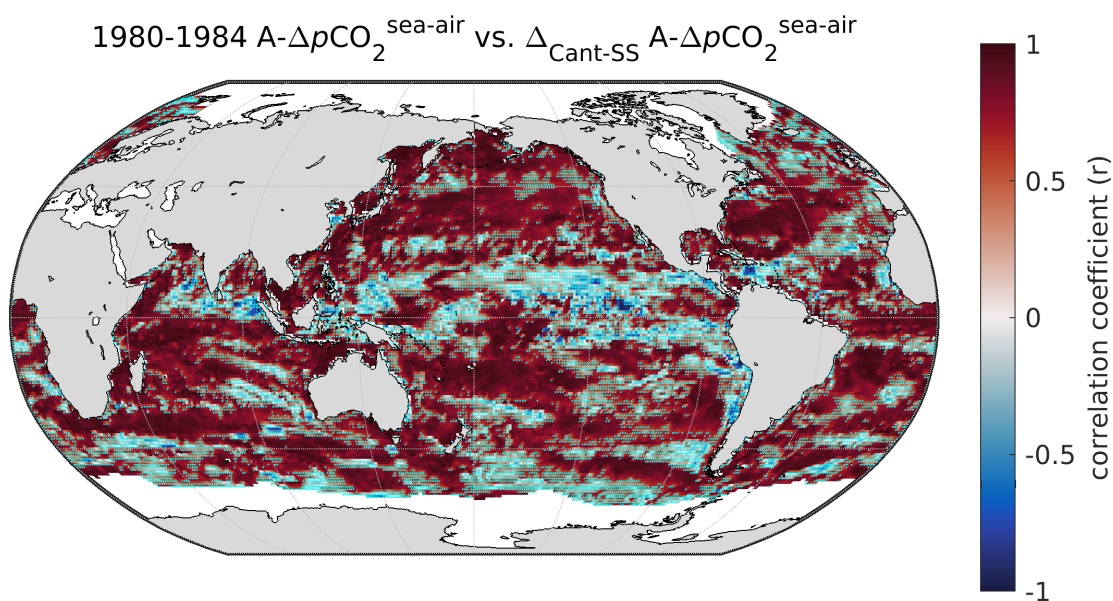


Figure S2.13. The across-model local correlation coefficient for the regressions of the 1980-1984 seasonal cycle amplitude of the disequilibrium between sea surface $p\text{CO}_2$ and atmospheric $p\text{CO}_2$ ($A-\Delta p\text{CO}_2^{\text{sea-air}}$) vs. the change in seasonal cycle amplitude of the sea-air $p\text{CO}_2$ disequilibrium ($\Delta_{\text{Cant-SS}} A-\Delta p\text{CO}_2^{\text{sea-air}}$) due to steady-state anthropogenic carbon accumulation in *Simulation C* between 1980-1984 and 2014-2018. Cyan regions delineate where relationships are not statistically significant ($p = 0.1$).

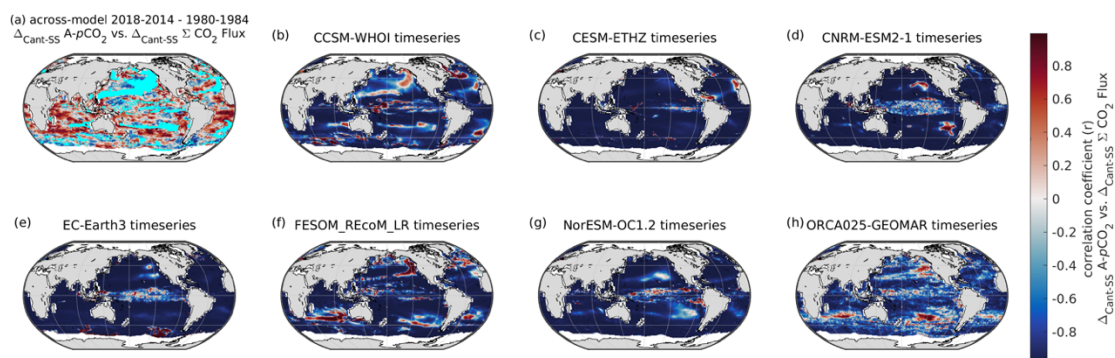


Figure S2.14. (a) The across-model local correlation coefficient for the regressions of the change in the $p\text{CO}_2$ seasonal cycle amplitude ($\Delta_{\text{Cant-SS}} A-p\text{CO}_2$) and the change in the cumulative annual CO_2 flux ($\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ flux}$) due to steady-state anthropogenic carbon accumulation in *Simulation C* between 1980-1984 and 2014-2018, following Figure 12a in the main text. Cyan regions delineate where relationships are not statistically significant ($p = 0.1$). (b-h) The local correlation coefficient for regressions of $\Delta_{\text{Cant-SS}} A-p\text{CO}_2$ vs. $\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ flux}$ time series within each GOBM in *Simulation C* at five-year increments between 1980-1984 and 2014-2018.

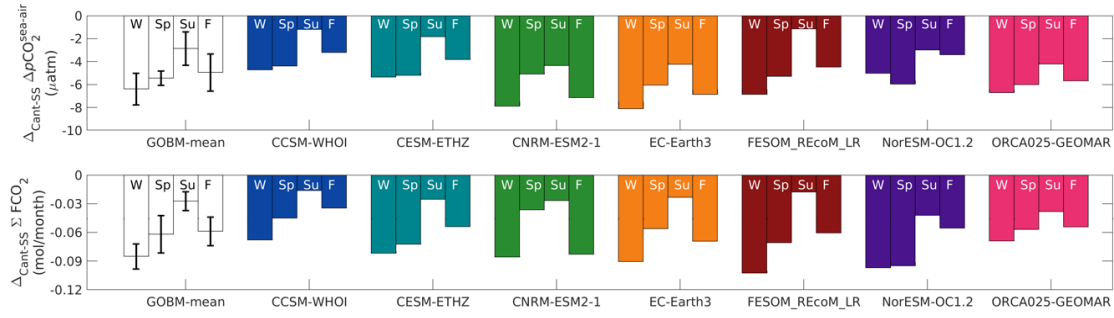


Figure S2.15. Area-weighted global mean C_{ant} -induced changes ($\Delta C_{\text{ant-ss}}$) to the (a) sea-air $p\text{CO}_2$ disequilibrium ($\Delta p\text{CO}_2^{\text{sea-air}}$) and (b) cumulative annual CO_2 flux (ΣCO_2 flux) in different seasons between 1980-1984 and 2014-2018 for the across-model mean (white with black standard deviation error bars) and for each model (colors). Seasons corresponding to the Northern Hemisphere are Winter (W; January, February, March), Spring (Sp; April, May, June), Summer (Su; July, August, September), and Fall (F; October, November, December). Seasons are shifted by six months for the Southern Hemisphere.

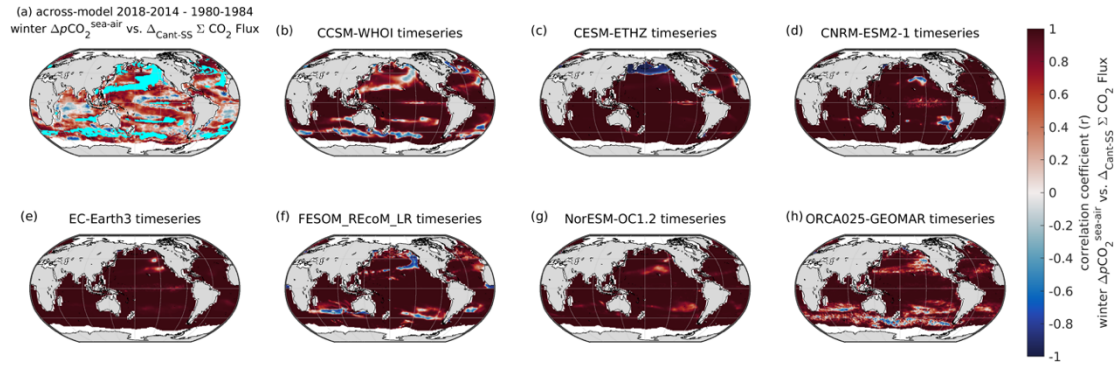


Figure S2.16. (a) The across-model local correlation coefficient for the regressions of the change in the wintertime disequilibrium between surface ocean and atmospheric $p\text{CO}_2$ ($\Delta C_{\text{ant-ss}}$ winter $\Delta p\text{CO}_2^{\text{sea-air}}$) and the change in the cumulative annual CO_2 flux ($\Delta C_{\text{ant-ss}}$ ΣCO_2 flux) due to steady-state anthropogenic carbon accumulation in *Simulation C* between 1980-1984 and 2014-2018, following Figure 12c in the main text. Cyan regions delineate where relationships are not statistically significant ($p = 0.1$). (b-h) The local correlation coefficient for regressions of $\Delta C_{\text{ant-ss}}$ winter $\Delta p\text{CO}_2^{\text{sea-air}}$ vs. $\Delta C_{\text{ant-ss}}$ ΣCO_2 flux time series within each GOBM in *Simulation C* at five-year increments between 1980-1984 and 2014-2018.

Table S2.1. The slopes and R^2 values for regressions of the area-weighted global mean time series of $\Delta_{\text{Cant-SS}} \text{A-}p\text{CO}_2$ and $\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ Flux}$ and of $\Delta_{\text{Cant-SS}} \text{winter } \Delta p\text{CO}_2^{\text{sea-air}}$ and $\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ Flux}$ for each GOBM between 1980 and 2018, as shown in Figure 12b and 12d in the main text.

Model	$\Delta_{\text{Cant-SS}} \text{A-}p\text{CO}_2$ vs. $\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ Flux}$		$\Delta_{\text{Cant-SS}} \text{winter } \Delta p\text{CO}_2^{\text{sea-air}}$ vs. $\Delta_{\text{Cant-SS}} \sum \text{CO}_2 \text{ Flux}$	
	slope (mol C m ⁻² dec ⁻¹ μatm^{-1})	R²	slope (mol C m ⁻² dec ⁻¹ μatm^{-1})	R²
CCSM-WHOI	0.28	0.98	-0.39	0.99
CESM-ETHZ	0.31	0.99	-0.40	0.99
CNRM-ESM-1	0.34	0.99	-0.27	0.99
EC-Earth3	0.38	0.99	-0.28	0.99
FESOM- REcoM-LR	0.28	0.99	-0.33	0.99
NorESM-OC1.2	0.43	0.98	-0.51	0.99
ORCA025- GEOMAR	0.29	0.99	-0.30	0.99

Chapter 3: Amplified Interior Ocean $p\text{CO}_2$ Changes and Emerging Surface Ocean Carbon Cycle Feedbacks in Global Ocean Biogeochemical Models

3.1 Introduction

The ocean plays a critical role in regulating global climate, having absorbed approximately 40% of all fossil fuel carbon dioxide gas (CO_2) emissions released to the atmosphere to date (Friedlingstein et al., 2024). The persistent uptake of anthropogenic carbon (C_{ant}) by the surface ocean is primarily facilitated by the air-sea exchange of CO_2 , where CO_2 fluxes vary regionally and seasonally in response to physically and biogeochemically induced gradients in surface ocean properties. Over the next century, the annual ocean carbon sink is predicted to grow roughly in proportion to the trend in atmospheric partial pressure of CO_2 ($p\text{CO}_2$; e.g., McKinley et al., 2017; McKinley et al., 2020; Terhaar, 2024). Studies using coupled Earth System Model (ESM) simulations have sought to quantify the strength of this so-called carbon-concentration feedback, whereby an increase in atmospheric $p\text{CO}_2$ leads to an increase in oceanic $p\text{CO}_2$ levels (Arora et al., 2013, 2020; Friedlingstein et al., 2006). In this framework, the carbon-concentration feedback is viewed as a negative feedback on atmospheric $p\text{CO}_2$ growth as ocean carbon uptake slows the accumulation of CO_2 in the atmosphere; however, the use of the term “feedback” is conceptually incomplete as the carbon-concentration feedback quantifies the ocean response to anthropogenic forcing rather than a true feedback that amplifies or dampens the initial perturbation.

To more fully capture the role of ocean chemistry in modulating ocean carbon uptake, we introduce the term “carbon-chemistry feedback” to describe how rising atmospheric $p\text{CO}_2$ leads to intensified changes in ocean $p\text{CO}_2$ via changes in the

ocean's buffering capacity. This feedback arises from the accumulation of C_{ant} in the upper ocean, which increases the inventory of dissolved inorganic carbon (DIC) and weakens the ocean's ability to buffer additional CO_2 uptake (Eggleston et al., 2010; Fassbender et al., 2017; Middelburg et al., 2020; Sabine et al., 2004). As buffering capacity declines, the efficiency of surface ocean C_{ant} uptake erodes, thereby reducing the ocean's ability to absorb additional CO_2 and weakening the ocean carbon sink. In this framework, the carbon-chemistry feedback ultimately represents a positive feedback (i.e., climate-change enhancing) that accelerates atmospheric $p\text{CO}_2$ growth by progressively diminishing the ocean carbon sink efficiency over time via changes in ocean chemistry (Broecker et al., 1979; Revelle & Suess, 1957). The buffering capacity of the ocean carbonate system is commonly described using the Revelle sensitivity Factor (RF), defined as the relative change in $p\text{CO}_2$ associated with a relative change in DIC, holding all other conditions constant.

$$\text{RF} = \frac{\partial p\text{CO}_2 / p\text{CO}_2}{\partial \text{DIC} / \text{DIC}} \quad \text{Eq 1.}$$

Higher RF values indicate that waters are more chemically sensitive to DIC perturbations, leading to proportionally larger relative changes in $p\text{CO}_2$. As RF increases with continued C_{ant} accumulation, upper ocean waters become increasingly sensitivity to DIC additions, intensifying the carbon-chemistry feedback over time.

Although the largest accumulation of C_{ant} is generally in the surface ocean, recent studies have shown that the largest C_{ant} -driven increases in $p\text{CO}_2$ have occurred in the interior ocean (Arroyo et al., 2022; Fassbender et al., 2023; Knor et al., 2025; Müller & Gruber, 2024). Coherent patterns of intensified subsurface $p\text{CO}_2$ change

caused by C_{ant} have been found across broad ocean regions (Fassbender et al., 2023), with the most pronounced changes occurring in the North Pacific Ocean. This phenomenon reflects the intensification of the carbon-chemistry feedback below the surface, caused by the transport of C_{ant} along deep and intermediate water masses into the interior ocean, where waters are naturally enriched in DIC and have low buffering capacities. Just like at the surface, C_{ant} penetration into the water column reduces the subsurface buffering capacity, recorded as an increase in RF. The primary difference is that subsurface waters already have naturally elevated RF values (Figure 3.1), due to the buildup of natural carbon from organic matter remineralization and calcium carbonate dissolution, and thus a heightened sensitivity of $p\text{CO}_2$ to DIC perturbations. Differing vertical distributions of C_{ant} and natural carbon create subsurface hotspots that magnify the impact of C_{ant} perturbations on the carbonate system below the surface, causing enhanced nonlinear responses in $p\text{CO}_2$. Therefore, the magnitude and distribution of carbon pools that comprise interior ocean DIC dictate regional differences in interior C_{ant} -driven $p\text{CO}_2$ changes (e.g., Fassbender et al, 2023).

Over the historical period, global ocean biogeochemical models (GOBMs) have been used by the Global Carbon Budget to track the ocean carbon sink evolution by reconstructing annual air-sea CO_2 flux variability under the forcing of transient historical atmospheric CO_2 and observation-based reanalysis climate data (DeVries et al., 2023; Friedlingstein et al., 2024; Terhaar et al., 2024). A central challenge in constraining model-based estimates of air-sea CO_2 fluxes is identifying and accurately representing the mechanisms that dictate long-term changes in surface ocean $p\text{CO}_2$.

Recent observational and modeling studies have emphasized the importance of wintertime CO₂ fluxes in governing net annual ocean carbon uptake, as stronger winds during winter enhance gas exchange across the local air-sea $p\text{CO}_2$ disequilibrium (Arroyo et al., in review; Bushinsky et al., 2019; Fassbender et al., 2022; Rodgers et al., 2008; Rustogi et al., 2023). While most efforts to understand and constrain long-term changes in the ocean carbon sink have focused on annual, sea surface carbon cycle process representations, we hypothesize that the seasonal interactions with subsurface carbon pools may play an increasingly important role in modulating surface ocean $p\text{CO}_2$ and air-sea CO₂ fluxes, particularly in winter. This reflects the potential emergent influence of subsurface carbon-chemistry feedbacks on surface ocean carbon dynamics.

Across large parts of the ocean, subsurface $p\text{CO}_2$ changes caused by C_{ant} accumulation are outpacing the atmospheric $p\text{CO}_2$ change (Arroyo et al., 2022; Fassbender et al., 2023). Thus, we previously hypothesized that C_{ant} -driven changes to subsurface $p\text{CO}_2$ will impact surface ocean $p\text{CO}_2$ levels during water mass ventilation (Fassbender et al., 2023). Herein, we test this hypothesis, postulating that changing interior conditions may play a contributing role in modulating surface ocean $p\text{CO}_2$ conditions, particularly during the “wintertime window” (Rodgers et al. 2008) when upper-ocean mixing reaches its maximum depth and facilitates the re-emergence of amplified $p\text{CO}_2$ waters (Rodgers et al., 2020) This translation of the carbon-chemistry feedback from its more pronounced expression in subsurface waters to surface waters is likely to get more extreme in the future as the ocean absorbs more C_{ant} . Thus, we are

interested in exploring the progression of interior ocean $p\text{CO}_2$ change in GOBMs used to estimate the historical ocean carbon sink.

In this study, we evaluate how GOBMs represent interior ocean $p\text{CO}_2$ changes caused by C_{ant} accumulation in the Central North Pacific Ocean, in comparison to a data-based estimate. Building from the analysis of Fassbender et al. (2023), we focus our analysis on examining representations of the nonlinear component of $p\text{CO}_2$ change driven by C_{ant} accumulation. We show how differences in the vertical distribution of carbon pools contribute to the simulated differences in the patterns and magnitudes of amplified $p\text{CO}_2$ responses in the interior ocean. Finally, we demonstrate that subsurface conditions may serve as an emergent constraint on winter surface $p\text{CO}_2$ conditions in GOBMs by projecting how near-future carbon burdens intensify the carbon-chemistry feedback globally at the annual maximum mixed layer depth.

3.2 Data and Methods

3.2.1 Global Ocean Biogeochemical Models (GOBMs)

We evaluate the interior ocean distributions of natural and anthropogenic carbon in seven global ocean biogeochemical models (GOBMs) compiled by the second iteration of the REgional Carbon Cycle Assessment and Processes (RECCAP2; Table 3.1; Müller, 2023; Terhaar et al., 2024). GOBMs are general ocean circulation models coupled with sea ice and biogeochemical cycling components that perform hindcast simulations of both steady-state and transient carbon fluxes driven by natural and anthropogenic perturbations. Many of the models included in this study have been used by the Global Carbon Project to estimate the contemporary magnitude and

variability of the ocean carbon sink annually (Friedlingstein et al. 2024). In each model, water column biological carbon pump processes are simulated based on a nutrient-phytoplankton-zooplankton-detritus (NPZD) ecosystem framework and include the processes of net primary production parameterized by nutrients, light availability, and temperature, as well as particulate organic carbon export and remineralization at depth (Doney et al., 2024). Considered GOBMs do not include calcium carbonate mineral cycling or dissolved organic matter pools. RECCAP2 model fields are provided on a $1^\circ \times 1^\circ$ horizontal grid at monthly resolution for 2D surface fields (including mixed layer depths) and annual resolution for 3D interior ocean fields from 1980 to 2018, with differing vertical grid resolutions for each model. We exclude sea ice-dominated regions in the Arctic and Southern Oceans from our analysis.

Table 3.1. Global ocean biogeochemical models used in this study. WOA18 = World Ocean Atlas 2018.

Model	Reference	Notes
CCSM-WHOI (CCSM 3.1)	Doney et al., 2009	Ends in 2017
CESM-ETHZ	Lindsay et al., 2014; Yang & Gruber, 2016	
CNRM-ESM-1	Berthet et al., 2019; Séférian, Delire, et al., 2016; Séférian et al., 2019	
EC-Earth3	Aumont et al., 2015; Döscher et al., 2022	
FESOM-REcoM-LR	Hauck et al., 2020	No preindustrial spin-up Used WOA18 phosphate
NorESM-OC1.2	Schwinger et al., 2016	
ORCA025-GEOMAR	Chien et al., 2022; Kriest & Oschlies, 2015	No preindustrial spin-up Used WOA18 silicate

For each model, we use fields of salinity, temperature, total alkalinity (TA), DIC, and nutrients (phosphate and silicate) to compute $p\text{CO}_2$ and RF values using CO2SYS Version 3.1.2 for MATLAB (van Heuven et al., 2011; Lewis & Wallace, 1998; Sharp et al., 2020). We use the dissociation constants for the carbonate system (K_1 and K_2) by Lueker et al. (2000), hydrogen sulfate by Dickson et al. (1990), and hydrogen fluoride by Perez and Fraga (1987), and the total boron-to-chlorinity ratio of Lee (2010). We report $p\text{CO}_2$ values in the interior ocean referenced to surface sea surface pressure, rather than in situ pressure, as CO2SYS calculator calculates the equilibrium value that would exist at the surface ocean. We use surface-referenced interior ocean $p\text{CO}_2$ throughout to enable direct comparison to atmospheric $p\text{CO}_2$.

To isolate the C_{ant} -driven change (ΔC_{ant}) in interior carbonate chemistry, we consider two RECCAP2 simulations: Simulation A and Simulation D. Simulation A is a contemporary run, forced by rising atmospheric CO_2 and transient historical atmospheric reanalysis data, capturing the combined impact of C_{ant} accumulation, natural climate variability, and climate change on carbon cycling. In Simulation D, atmospheric CO_2 is fixed at preindustrial levels, while climate is forced with transient historical atmospheric reanalysis data. We compute the difference between Simulation A and Simulation D for each model as an estimate of C_{ant} -driven change since the preindustrial era, removing the influence of climate variability and climate change. For example:

$$\Delta C_{\text{ant}} p\text{CO}_2 = p\text{CO}_2^{\text{Simulation A}} - p\text{CO}_2^{\text{Simulation D}} \quad \text{Eq 2.}$$

density grid, compute the model-mean values in density space, and then finally remap the model-mean values back to depth space using model-mean density distributions. This approach avoids signal artifacts caused by different circulation structures and preserves coherent vertical patterns across models.

3.2.2 Observation-based Interior Carbon Distributions from GOBAI-O₂

We compare model-based estimates of interior carbon pools to those derived from machine learning-based and statistical algorithms applied to interior ocean observations. Many of the considered data products and algorithms were commonly trained on quality-controlled discrete shipboard observations compiled by the GLObal Data Analysis Product (GLODAP), making them predominantly internally consistent (Lauvset et al., 2024). We rely mostly on the observation-based data product Gridded Ocean Biogeochemistry from Artificial Intelligence - Oxygen version 2.1 (GOBAI-O₂; Sharp et al., 2023) to derive interior carbonate chemistry. GOBAI-O₂ provides monthly 1° x 1° mapped fields of dissolved oxygen (O₂), salinity, and temperature in the upper 2000 m. The data product was constructed using machine learning approaches trained on discrete shipboard measurements (from GLODAPv2.2022) as well as biogeochemical (BGC)-Argo float observations. The algorithm was applied to time-evolving, monthly Argo-based gridded temperature and salinity climatologies (Roemmich and Gilson, 2009; hereafter abbreviated to RG09) to produce time-evolving, 3D O₂ fields at monthly resolution from 2004 to 2024. We then use salinity, temperature, and O₂ with space-time location (latitude, longitude, depth, time) to compute time-evolving interior fields of DIC, total alkalinity (TA), and inorganic

macronutrients (phosphate and silicate) with Empirical Seawater Property Estimation Routines (ESPERs version 1; Carter et al., 2021). Similar to GOBAI-O₂, ESPERs are predictor algorithms trained on the discrete observations from GLODAPv2.2020. We use the mixed routine (ESPER_MIXED), which provides biogeochemical estimates as the average of neural network and locally interpolated regression predictions. Finally, we use the monthly salinity and temperature fields from RG09 and ESPER-derived TA, DIC, and nutrients to compute observation-based estimates of $p\text{CO}_2$ and RF using CO2SYSv3 as described above.

To estimate C_{ant} , we apply the Tracer-based Rapid Anthropogenic Carbon Estimation (TRACE) algorithm (version 1; Carter et al., 2025) to the salinity, temperature, and space-time locations (latitude, longitude, pressure, time) from RG09. The TRACE method is built on a modified transit time distribution framework and is trained on transient tracer measurements from GLODAPv2.2022, with additional constraints from water mass ideal age estimates. Notably, when applied to the RECCAP2 GOBMs, TRACEv1 reproduces the global C_{ant} inventory to within $\pm 10\%$ of the simulated inventories between 1980 and 2014 (Carter et al., 2025), giving us confidence in the skill of reconstructed C_{ant} from observation-based monthly salinity and temperature fields. TRACE-estimated C_{ant} values are then subtracted from ESPER-derived DIC values to estimate a quasi-preindustrial DIC concentration. We then use this preindustrial DIC estimate with salinity and temperature from RG09 and ESPER-derived TA and nutrients to compute quasi-preindustrial $p\text{CO}_2$ and RF using

CO2SYSv3. The impact of C_{ant} accumulation on carbonate system variables derived from GOBAI-O₂ is computed as, for example,

$$\Delta_{C_{\text{ant}}} p\text{CO}_2 = p\text{CO}_2^{\text{Modern}} - p\text{CO}_2^{\text{Preindustrial}} \quad \text{Eq 3.}$$

Observation-based mixed layer depths are computed for each month from potential density anomalies (derived from monthly salinity and temperature from RG09) as the depth at which the local potential density anomaly exceeds the overlying 10m value by 0.03 kg m⁻³ (de Boyer Montégut et al., 2004). Using these monthly mixed layer depth estimates for each horizontal grid cell, we then extract carbonate system values at the maximum mixed layer depth in each year. Although GOBAI-O₂ and derived fields are monthly resolved through 2024, we compute annual means, then the 2014-2018 mean for comparisons with interior ocean GOBMs fields.

3.2.3 Nonlinear Component of Interior Change caused by C_{ant}

Amplified interior ocean $p\text{CO}_2$ changes arise due to interactions between C_{ant} and natural carbon in naturally high-DIC waters that induce nonlinear carbonate chemistry responses. Adapting methods described in Fassbender et al. (2023), we quantify the contribution of carbonate system nonlinearities to the total $p\text{CO}_2$ change caused by C_{ant} over time in the RECCAP2 GOBMs and in our GOBAI-O₂-based observational estimate. Similar to their approach, we focus on the respiratory component of the biological carbon pump contributing to nonlinear changes due to its dominance in setting natural vertical gradients of DIC. In the modern history of a water mass, surface waters first exchange natural carbon and C_{ant} with the atmosphere (setting the preformed DIC concentration) and then subduct and accumulate natural carbon

byproducts from organic matter remineralization (and calcium carbonate mineral dissolution, which GOBMs do not simulate) along density-driven water mass circulation pathways. To isolate the nonlinear carbonate system changes associated with natural carbon additions at depth, we focus on regenerated carbon (C_{reg}) from organic matter remineralization.

For each GOBM and for GOBAI- O_2 , we derive C_{reg} throughout the water column from apparent oxygen utilization (AOU), computed as the difference between the expected O_2 concentration at saturation with the atmosphere and model O_2 or GOBAI- O_2 ($\text{AOU} = O_2^{\text{saturated}} - O_2^{\text{modern}}$). We convert the AOI to C_{reg} using the canonical O_2 :C ratio of 170/117 ($C_{\text{reg}} = 117/170 * \text{AOU}$) from Anderson and Sarmiento (1994). In the near-surface ocean, signals of biologically-mediated C_{reg} concentrations may be negative, signifying net photosynthesis, and/or may be muddled by thermodynamic processes controlling air-sea O_2 disequilibrium and gas exchange, as surface waters more rapidly exchange O_2 with the atmosphere compared to CO_2 .

To isolate the nonlinear component of the total pCO_2 change induced by carbon pool interactions, we first compute the total pCO_2 change caused by C_{ant} in the absence of C_{reg} . This is done by subtracting contemporaneous C_{reg} estimates from the total DIC concentration ($\text{DIC}^{\text{no_reg}} = \text{DIC} - C_{\text{reg}}$). We also correct TA for the small influence of nitrate produced during organic matter remineralization using the canonical O_2 :N ratio of 170/16 and the scaling factor of 1.36 ($\text{TA}^{\text{no_reg}} = \text{TA} - (1.36 * (16/170) * \text{AOU})$; Anderson and Sarmiento, 1994; Wolf-Gladrow, 2007). We use these inputs of $\text{DIC}^{\text{no_reg}}$ and $\text{TA}^{\text{no_reg}}$ in combination with salinity, temperature, and nutrients (phosphate and

silicate) in CO2SYSv3 to compute fields of hypothetical fields of $p\text{CO}_2$ that include the influence of C_{ant} without the influence of C_{reg} . The C_{ant} -driven change in the absence of C_{reg} accumulation is then computed for both the GOBMs ($\Delta_{C_{\text{ant}}}^{\text{no-reg}} p\text{CO}_2 = p\text{CO}_2^{\text{no-reg, Simulation A}} - p\text{CO}_2^{\text{no-reg, Simulation D}}$) and for GOBAI-O₂ ($\Delta_{C_{\text{ant}}} p\text{CO}_2^{\text{no-reg}} = p\text{CO}_2^{\text{no-reg, Modern}} - p\text{CO}_2^{\text{no-reg, Preindustrial}}$). Finally, we quantify the nonlinear (NL) component of C_{ant} -induced change as

$$\Delta_{C_{\text{ant}}}^{\text{NL}} p\text{CO}_2 = \Delta_{C_{\text{ant}}} p\text{CO}_2 - \Delta_{C_{\text{ant}}}^{\text{no-reg}} p\text{CO}_2 \quad \text{Eq 4.}$$

3.3 Results

3.3.1 Interior Ocean Carbon Pool Structures in RECCAP2 GOBMs

The 2014-2018 GOBM ensemble-mean DIC distribution is substantially lower than that derived from GOBAI-O₂ along 150.5°W in the Central Pacific Ocean (Figure 3.1; Figure S3.1). These DIC differences can be attributed to dissimilar representations of preindustrial ocean buffering and its effect on the solubility pump (Terhaar et al., 2024; Figure S3.2), as well as dissimilar representations of C_{ant} and C_{reg} accumulations. On average, simulated upper-ocean C_{ant} values are slightly larger than those estimated by TRACE in northern and southern subtropical gyres. In contrast, C_{ant} values are largely underestimated near the surface in the equatorial region and along coherent intermediate water density surfaces below the subtropical gyres. Recent studies by the RECCAP2 project have shown that the GOBMs largely underestimate integrated C_{ant} inventories in the upper 3000 m by 20-30% compared to observation-based C_{ant} estimation methods (Terhaar et al., 2024; Figure S3.3). GOBM biases in C_{ant} storage

have been primarily attributed to structural model biases and incomplete spin-up procedures, such as delayed and differing industrialization start dates, in addition to substantial biases in ocean circulation and surface ocean carbonate chemistry that collectively influence net sea-air CO₂ fluxes. Discrepancies in C_{reg} also appear at depth. Below ~200m, simulated C_{reg} is higher in the South Pacific and lower in the North Pacific compared to estimates from GOBAI-O₂. As a key metric of the biological carbon pump, C_{reg} reflects the integrated signals of organic matter accumulation along

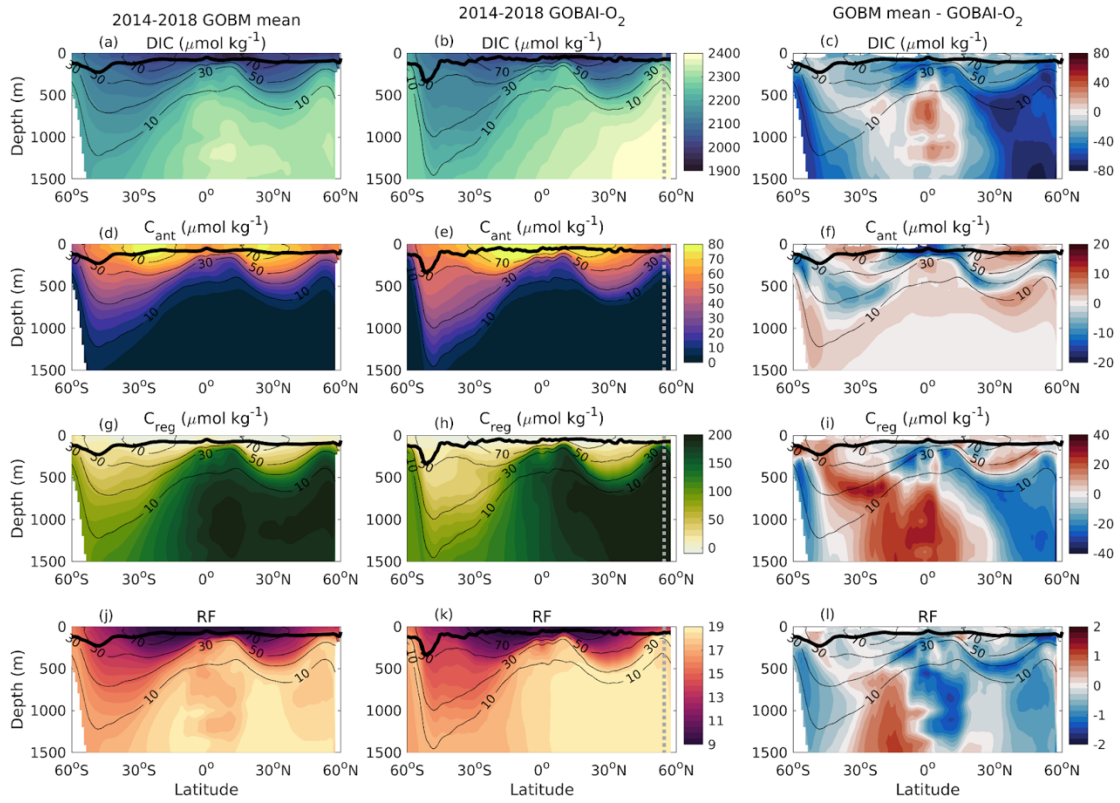


Figure 3.1. Interior carbon pool distributions along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (column 1) an ensemble-mean of global ocean biogeochemical models (GOBMs; $n = 7$) and (column 2) GOBAI-O₂, with (column 3) their differences. Sections of (a-c) dissolved inorganic carbon (DIC), (d-f) anthropogenic carbon (C_{ant}), and (g-i) regenerated carbon (C_{reg}). Meridional and vertical differences in carbon pool distributions cause differences in (j-l) the Revelle sensitivity factor (RF). In each panel, black contours represent the 2014-2018 mean (thin) C_{ant} values and (thick) maximum mixed layer depths, with model-mean C_{ant} contours plotted in column 3. The vertical gray dashed line on the GOBAI-O₂ panels marks the location of profiles in Figure 3.3.

water mass circulation pathways (Frenger et al., 2024). Therefore, C_{reg} estimates are strongly influenced by thermocline ventilation rates and the choice of ecosystem model parameterizations that supply carbon for respiration, which substantially differ between models (Doney et al., 2025; Terhaar et al., 2024; Figure S3.4). In the North Pacific, where waters are old, incomplete preindustrial spin-ups would also affect deep signals of integrated C_{reg} accumulation (as well as preformed preindustrial DIC). Discrepancies in total DIC (and in TA; Figure S3.5) ultimately drive biases in the RF (Figure S3.6). Models broadly capture the increases in RF with depth, indicating a greater sensitivity of $p\text{CO}_2$ to DIC changes, but tend to underestimate interior RF in the North Pacific, with equatorial region patchiness in the model-mean subsurface waters associated with large DIC and TA discrepancies in one model (NorESM-OC1.2).

3.3.2 Intensified subsurface $p\text{CO}_2$ change in RECCAP2 GOBMs

The 2014-2018 GOBM ensemble-mean total $p\text{CO}_2$ change caused by C_{ant} ($\Delta_{\text{Cant}} p\text{CO}_2$) generally captures the meridional patterns found in the estimates derived from GOBAI- O_2 for the same 150.5°W Pacific transect (Figure 3.2; Figure S3.7). The largest signals of interior ocean $p\text{CO}_2$ change occur between ~ 100 and 500 m in the North Pacific, a feature previously identified by studies using ship-based measurements (Arroyo et al., 2022; Fassbender et al., 2023). More moderate $\Delta_{\text{Cant}} p\text{CO}_2$ signals are seen in the South Pacific, characterized by a weaker subsurface maxima. The majority of $\Delta_{\text{Cant}} p\text{CO}_2$ increases in northern thermocline waters can be attributed to the nonlinear component of $p\text{CO}_2$ change ($\Delta_{\text{Cant}}^{\text{NL}} p\text{CO}_2$; Figure S3.8). Though models largely capture the spatial structure of $\Delta_{\text{Cant}} p\text{CO}_2$ and $\Delta_{\text{Cant}}^{\text{NL}} p\text{CO}_2$, they systematically

underestimate the nonlinear $p\text{CO}_2$ response to C_{ant} compared to the GOBAI- O_2 derived estimates within the thermocline and in intermediate to deep water masses in the Southern Ocean. This is what leads to an underestimation of the total $\Delta_{C_{\text{ant}}} p\text{CO}_2$ relative to observed estimates at these depths (Figure 3.2c). Within the upper ocean, in both the ensemble mean and GOBAI- O_2 derived estimates, $\Delta_{C_{\text{ant}}}^{\text{NL}} p\text{CO}_2$ values are small or sometimes negative, which can be linked to small or negative C_{reg} values in these waters. Negative $\Delta_{C_{\text{ant}}}^{\text{NL}} p\text{CO}_2$ values work to dampen the overall C_{ant} -driven $p\text{CO}_2$ changes.

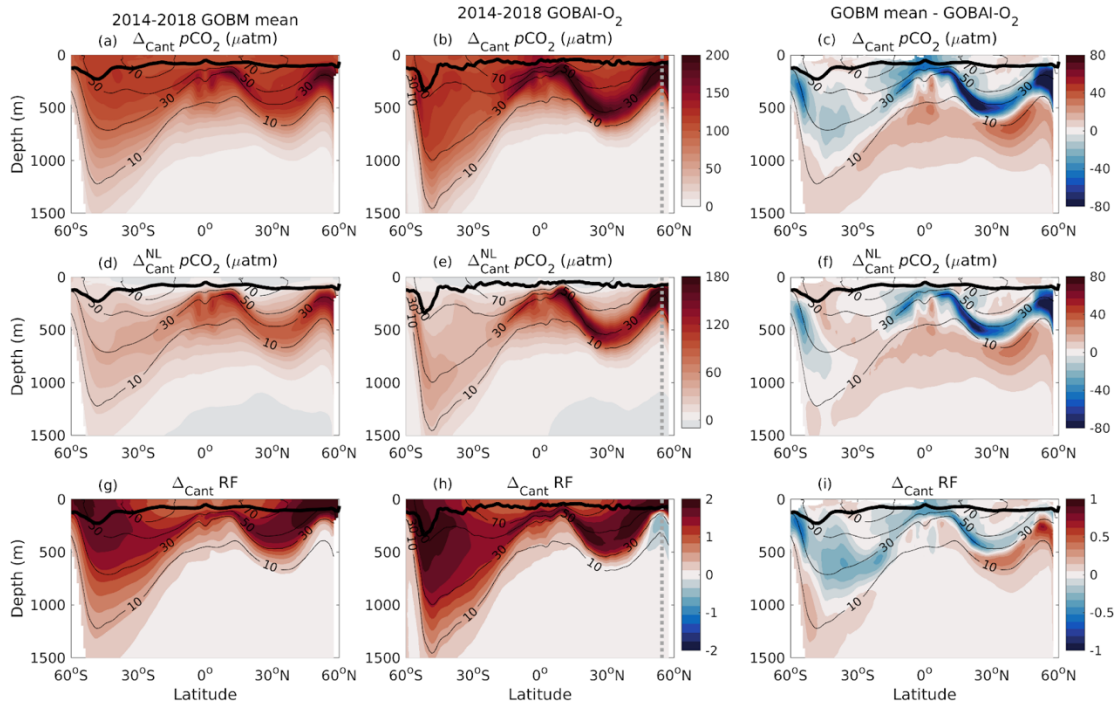


Figure 3.2. Interior ocean carbonate system changes caused by anthropogenic carbon ($\Delta_{C_{\text{ant}}}$) accumulations along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (column 1) an ensemble-mean of global ocean biogeochemical models (GOBMs; $n = 7$) and (column 2) GOBAI- O_2 , with (column 3) their differences. Sections of C_{ant} -driven changes in the (a-c) partial pressure of CO_2 ($p\text{CO}_2$), (d-f) nonlinear $p\text{CO}_2$ component, and (g-i) the Revelle Factor (RF). In each panel, black contours represent the 2014-2018 mean (thin) C_{ant} values and (thick) maximum mixed layer depths, with model-mean C_{ant} contours plotted in column 3. The vertical gray dashed line on the GOBAI- O_2 panels marks the location of profiles in Figure 3.3.

Spatial patterns of discrepancy between ensemble-mean and observation-based $\Delta_{\text{Cant}} p\text{CO}_2$ primarily map to the C_{ant} discrepancies but are driven by discrepancies in both C_{ant} and C_{reg} . In intermediate water masses, the ensemble-mean discrepancies in C_{ant} and C_{reg} are similar in magnitude ($\sim \pm 10\text{-}20 \mu\text{mol kg}^{-1}$), and both contribute to the overall DIC burden that drives the nonlinear $p\text{CO}_2$ response. Because the nonlinear $p\text{CO}_2$ response depends on the combined magnitude of DIC perturbations from its preformed condition, differences in either C_{ant} or C_{reg} work to influence differences in the total $\Delta_{\text{Cant}} p\text{CO}_2$. Further, we link weaker simulated interior $p\text{CO}_2$ changes to discrepancies in interior carbonate system buffering. Similar to $\Delta_{\text{Cant}} p\text{CO}_2$, $\Delta_{\text{Cant}} \text{RF}$ exhibits a subsurface maxima, with the largest increases regionally occurring in the upper and midwater column in the subpolar regions of the Southern and North Pacific Oceans. Increases in RF indicate an increased sensitivity of $p\text{CO}_2$ to DIC. However, on average, the models show similar spatial patterns of $\Delta_{\text{Cant}} \text{RF}$ underestimation. Notably, observation-based estimates reveal a C_{ant} -induced decline in RF in the high-latitude subpolar North Pacific ($\sim 55\text{-}60^\circ\text{N}$) between ~ 250 and 500 m (Figure 3.3), indicating that these waters have surpassed the maximum RF values and maximum $p\text{CO}_2$ sensitivity to DIC changes (Fassbender et al., 2017, 2023). This feature is not well captured in the individual GOBMs (Figure S3.9) and does not appear in the model-mean $\Delta_{\text{Cant}} \text{RF}$. Because the values of RF modulate the magnitude of nonlinear $p\text{CO}_2$ amplification, misrepresentations in RF and its changes directly lead to biases in the C_{ant} -driven $p\text{CO}_2$ changes.

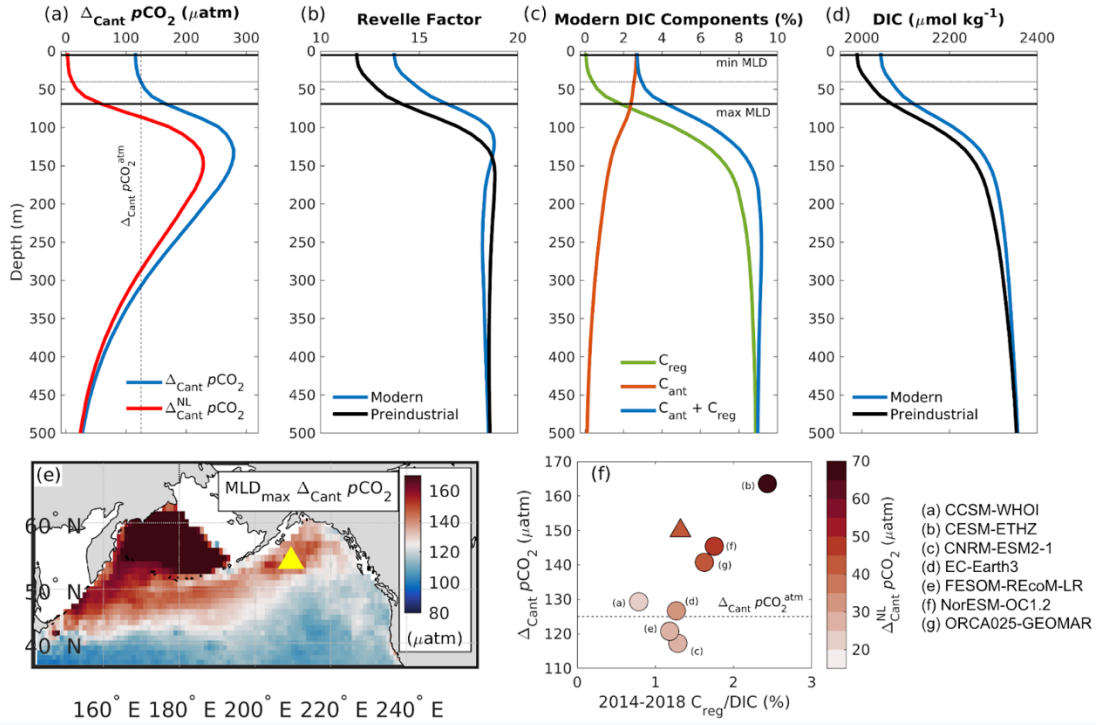


Figure 3.3. Vertical profiles from a location in the subpolar North Pacific (55.5°N, 150.5°W; yellow triangle in (e)) derived from GOBAI-O₂ for (a) C_{ant}-induced pCO₂ change (ΔC_{ant} pCO₂; blue) and the nonlinear component of change (Δ^{NL}_{Cant} pCO₂; red), (b) modern (blue) and preindustrial (black) RF, (c) C_{ant} (orange) and C_{reg} (green) components of DIC and their sum (blue), and (d) modern (blue) and preindustrial (black) DIC. Profiles represent the 2014-2018 mean conditions. Thicker horizontal lines denote the minimum and maximum annual mixed layer depths (MLDs), and thin horizontal dashed line in each marks the depth at which the ΔC_{ant} pCO₂ in the water column equals to the change in atmospheric pCO₂ since the preindustrial period (ΔC_{ant} pCO₂^{atm} = ~125 ppm in 2014-2018; vertical line in panel (a)). (e) a map of the ΔC_{ant} pCO₂ at the maximum MLD averaged between 2014-2018 from GOBAI-O₂, with the color map centered on ΔC_{ant} pCO₂^{atm}. Red values indicate where changes in ΔC_{ant} pCO₂ exceed ΔC_{ant} pCO₂^{atm}. (f) At the location marked in (a), the relationship between ΔC_{ant} pCO₂ and the percentage of C_{reg} in DIC (% C_{reg}/DIC) for GOBAI-O₂ (triangle) and GOBMs (circles, letters), colored by the Δ^{NL}_{Cant} pCO₂, with all values reflecting conditions at the maximum MLD between 2014-2018.

The spatial pattern and vertical structures of ΔC_{ant} pCO₂ and Δ^{NL}_{Cant} pCO₂ can be understood through the interactions between C_{ant} and C_{reg} within the total DIC pool (Figure 3.3). We illustrate this using profiles from the subpolar North Pacific Ocean (55.5°N, 150.5°W), as an example. In order to achieve a realistic total C_{ant}-induced change in pCO₂, there are multiple carbon cycle components to be appropriately represented in GOBMs, in addition to C_{ant} distributions. The vertical structure of C_{ant}-

induced $p\text{CO}_2$ change depends on the underlying DIC structure controlled in part by natural carbon processes, including biological carbon pump processes that sustain C_{reg} , and on the vertical gradients in RF established during preindustrial spin-up phases. The magnitude and shape of the $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ profile are caused by the magnitude and shape of the combined C_{ant} and C_{reg} contribution to the total DIC profile. Nonlinear contributions account for the majority of the total $\Delta_{\text{Cant}} p\text{CO}_2$ below the surface under moderate accumulations of C_{ant} ($\sim 10\text{-}50 \mu\text{mol kg}^{-1}$).

3.3.3 Spatial Patterns of Nonlinear $p\text{CO}_2$ Amplification at the Maximum Mixed Layer Depth

To evaluate where subsurface nonlinear $p\text{CO}_2$ increases could have a nontrivial impact on winter surface $p\text{CO}_2$ and thus seasonal air-sea CO_2 fluxes, we focus on the C_{ant} -induced changes at the maximum mixed layer depth (MLD_{max} ; Figure S3.10). At our test site in the subpolar North Pacific Ocean (55.5°N , 150.5°W), GOBAI- O_2 -based estimates indicate that nonlinear $p\text{CO}_2$ component changes have elevated $\text{MLD}_{\text{max}} \Delta_{\text{Cant}} p\text{CO}_2$ enough to exceed the atmospheric $p\text{CO}_2$ change (Figure 3.3a and 3.3e). This interior intensification of the carbon-chemistry feedback suggests that the seasonal re-emergence of subsurface $p\text{CO}_2$ may be an important mechanism modulating winter surface $p\text{CO}_2$ conditions in this region when deep mixing entrains subsurface waters, in line with our hypothesis. To compare with GOBAI- O_2 , we estimate the total and nonlinear $p\text{CO}_2$ changes at the MLD_{max} in each model by locally subsampling the annual mean profiles at the MLD_{max} for each year. We find that models with larger $\Delta_{\text{Cant}} p\text{CO}_2$ exhibit larger $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ responses at the MLD_{max} (Figure S3.11b), with

magnitudes of $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ in individual models tightly linked to the fraction of C_{reg} in total DIC (Figure 3.3e; Figure S3.11a). Similar to the observation-based estimate, elevated $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ pushes the total $\Delta_{\text{Cant}} p\text{CO}_2$ above the atmospheric $p\text{CO}_2$ change in many of the models in this location.

To more broadly assess the influence of nonlinear $p\text{CO}_2$ changes at the MLD_{max} , we look globally at the percentage of total $\Delta_{\text{Cant}} p\text{CO}_2$ attributed to $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ for each GOBM in comparison to GOBAI-O₂. At the MLD_{max} , the C_{ant} -induced nonlinear $p\text{CO}_2$ responses are shown to amplify (when positive) or dampen (when negative) the overall $p\text{CO}_2$ change (Figure 3.4), with substantial spatial heterogeneity in the fractional nonlinear $p\text{CO}_2$ contribution. The subpolar North Pacific and areas of the subpolar Southern Ocean exhibit the largest nonlinear component contributions. In those basins, RF values are naturally elevated in the upper water column (Figure 3.1; Figure S3.6), indicating a naturally heightened sensitivity of $p\text{CO}_2$ to DIC changes. Conversely, nonlinear $p\text{CO}_2$ dampening appears in the GOBAI-O₂-based estimate and in some models in parts of the subtropical regions (Figure S3.12), where the MLD_{max} tends to be shallow and within the productive euphotic zone. In these regions, C_{reg} is negative (oxygen is supersaturated) and therefore nonlinear contributions from natural carbon changes are working to dampen the overall impact of C_{ant} accumulation on $p\text{CO}_2$ (Figure S3.13).

Across models, we find that local C_{reg} concentrations at the MLD_{max} are significantly positively correlated with the magnitude of nonlinear $p\text{CO}_2$ contributions globally (Figure S3.11). The spatial patterns of the percentage of $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ at the

MLD_{max} in individual models highlight how regional differences in the vertical C_{ant} , C_{reg} , and RF distributions create regional subsurface hotspots that may re-emerge as

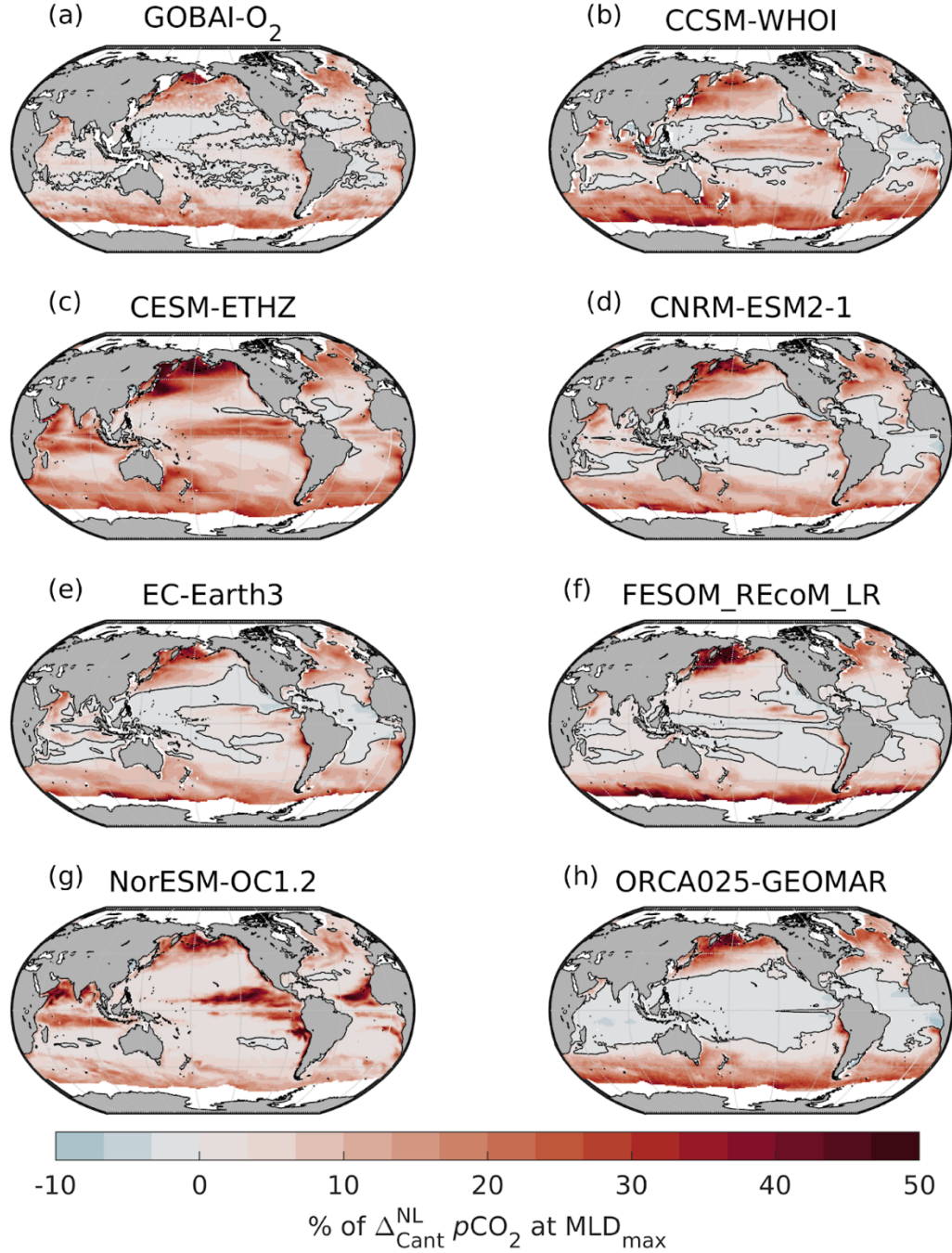


Figure 3.4. The percent of the total C_{ant} -induced pCO_2 change attributed to nonlinear pCO_2 changes (% of $\Delta_{Cant}^{NL} pCO_2$) at the maximum mixed layer depth (MLD_{max}) for (a) GOBAI- O_2 and (b-h) GOBMs, averaged over 2014-2018.

disproportionately elevated subsurface $p\text{CO}_2$ waters. Because $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ comprises a large fraction of $\Delta_{\text{Cant}} p\text{CO}_2$ (at the MLD_{max} and below), these results reinforce the importance of more accurately capturing biological carbon pump processes in GOBMs to improve the representation of interior ocean changes.

To further assess the potential connectivity between subsurface and surface expression of the carbon-chemistry feedback, we examine the total $\Delta_{\text{Cant}} p\text{CO}_2$ at the MLD_{max} (Figure 3.5). In the GOBAI- O_2 estimate, $\Delta_{\text{Cant}} p\text{CO}_2$ at the MLD_{max} has exceeded the atmosphere in regions where there are high fractions of nonlinear $p\text{CO}_2$ changes, particularly in the subpolar North Pacific, the Southern Ocean, and the northwest region of the Indian Ocean. Elevated changes in $p\text{CO}_2$ relative to the atmosphere may suggest that the wintertime reemergence of subsurface waters with amplified $p\text{CO}_2$ is already detectable in the observational record. Some of the GOBMs capture these patterns, but many largely underestimate total $\Delta_{\text{Cant}} p\text{CO}_2$ at the MLD_{max} (Figure S3.15). Notably, models that capture the elevated $\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$ (Figure 3.4) still underestimate the overall $\Delta_{\text{Cant}} p\text{CO}_2$ at the MLD_{max} (Figure 3.5). This is likely linked to the underestimation of the C_{ant} and C_{reg} pools. The interior ocean differences in total and nonlinear C_{ant} -driven $p\text{CO}_2$ changes between model and GOBAI- O_2 estimates are primarily due to discrepancies in C_{ant} (Figure 3.2). However, the across-model differences in total $\Delta_{\text{Cant}} p\text{CO}_2$ at the MLD_{max} are most strongly positively correlated with the combined inventory of C_{ant} and C_{reg} , and not with C_{ant} or C_{reg} alone (Figure S3.11d). These results highlight the importance of carbon pool interactions in modulating the magnitude of total C_{ant} -driven changes. GOBMs that inadequately

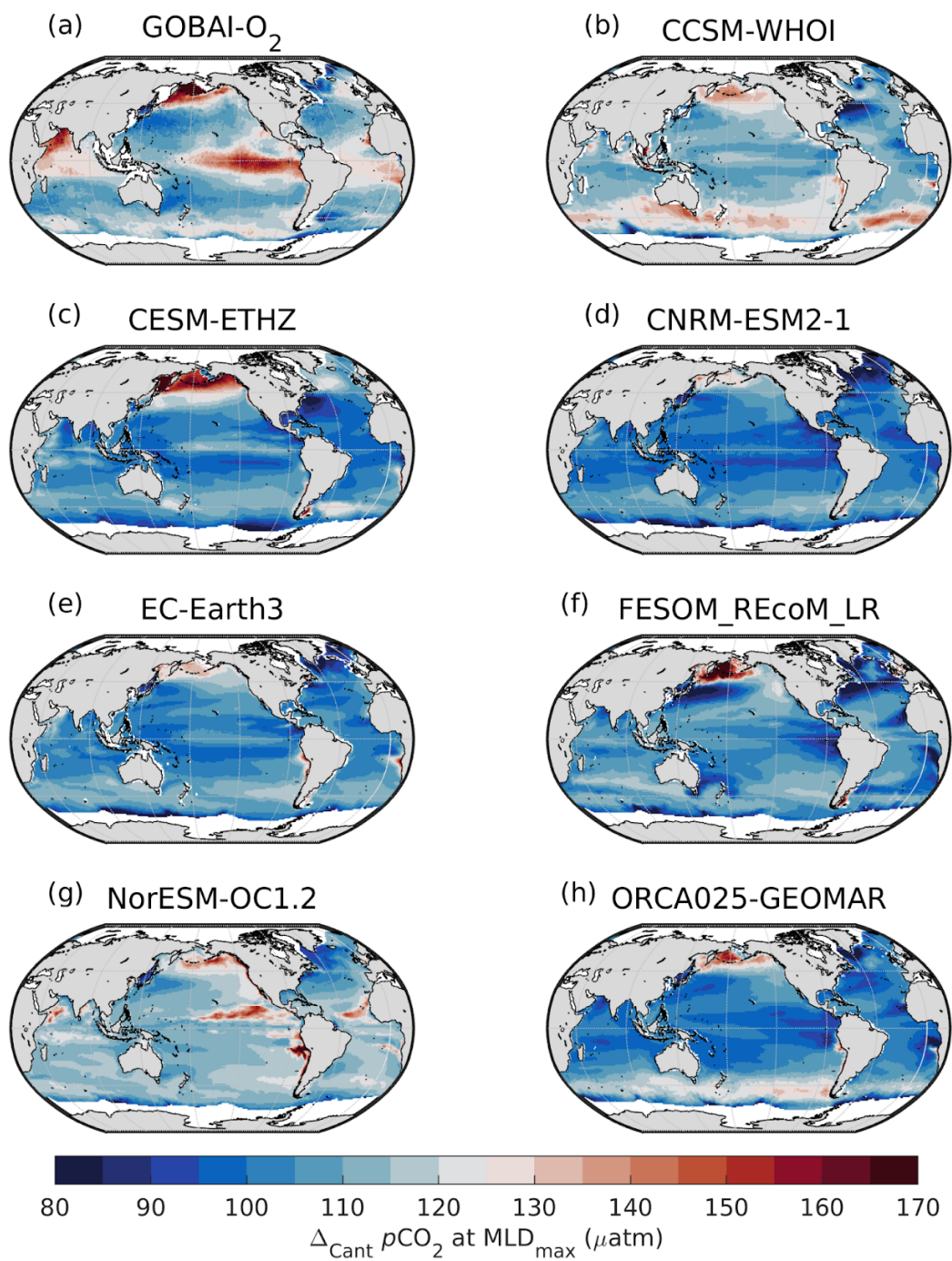


Figure 3.5. The total C_{ant} -induced $p\text{CO}_2$ change ($\Delta_{\text{Cant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) for (a) GOBAI-O₂ and (b-h) GOBMs, averaged over 2014-2018, centered on the change in atmospheric $p\text{CO}_2$ since the preindustrial period (~125 ppm). Red values indicate where changes in $p\text{CO}_2$ at the maximum mixed layer depth exceed the atmospheric $p\text{CO}_2$ change.

represent interior ocean $p\text{CO}_2$ changes at the maximum mixed layer depth may inadequately capture connectivity between subsurface and surface expression of the carbon-chemistry feedback.

3.3.4 Historical Trends in Interior Ocean $p\text{CO}_2$ Amplification and Future Outlooks

While the subsurface-to-surface translation of the carbon-chemistry feedback is muted in many of the models (Figure 3.5), we find that the trends in $\Delta C_{\text{ant}} p\text{CO}_2$ at the maximum mixed layer depth (Figure S3.16) and across sections of the interior ocean (Figure S3.17) are growing over time. Increasing trends over time reflect the compounding effect of C_{ant} accumulation, where the nonlinear response of $p\text{CO}_2$ to DIC becomes more pronounced as the background buffering capacity declines. Thus, the translation of subsurface signals to overlying surface waters during winter may become increasingly discernible in the future with C_{ant} accumulation. To anticipate future changes and their seasonal expressions at the surface, we project the most recent decadal trends in C_{ant} (from 2009-2018) at the MLD_{max} onto the 2014-2018 DIC conditions in each model for an additional 10 (to 2024-2028) and 20 years (to 2034-2038), allowing us to assess how the subsurface carbon-chemistry feedback manifests beyond the 2014-2018 mean state. This pseudo-projection approach allows us to assess whether the subsurface intensified nonlinear feedback becomes more prominent as C_{ant} continues to accumulate at a persistent rate, while keeping C_{reg} constant.

Projected C_{ant} -induced changes in $p\text{CO}_2$ at the MLD_{max} over the next 20 years suggest that the subsurface carbon-chemistry feedback will continue to intensify under linearly increasing C_{ant} accumulation. In all of the 20-year model projections, ΔC_{ant}

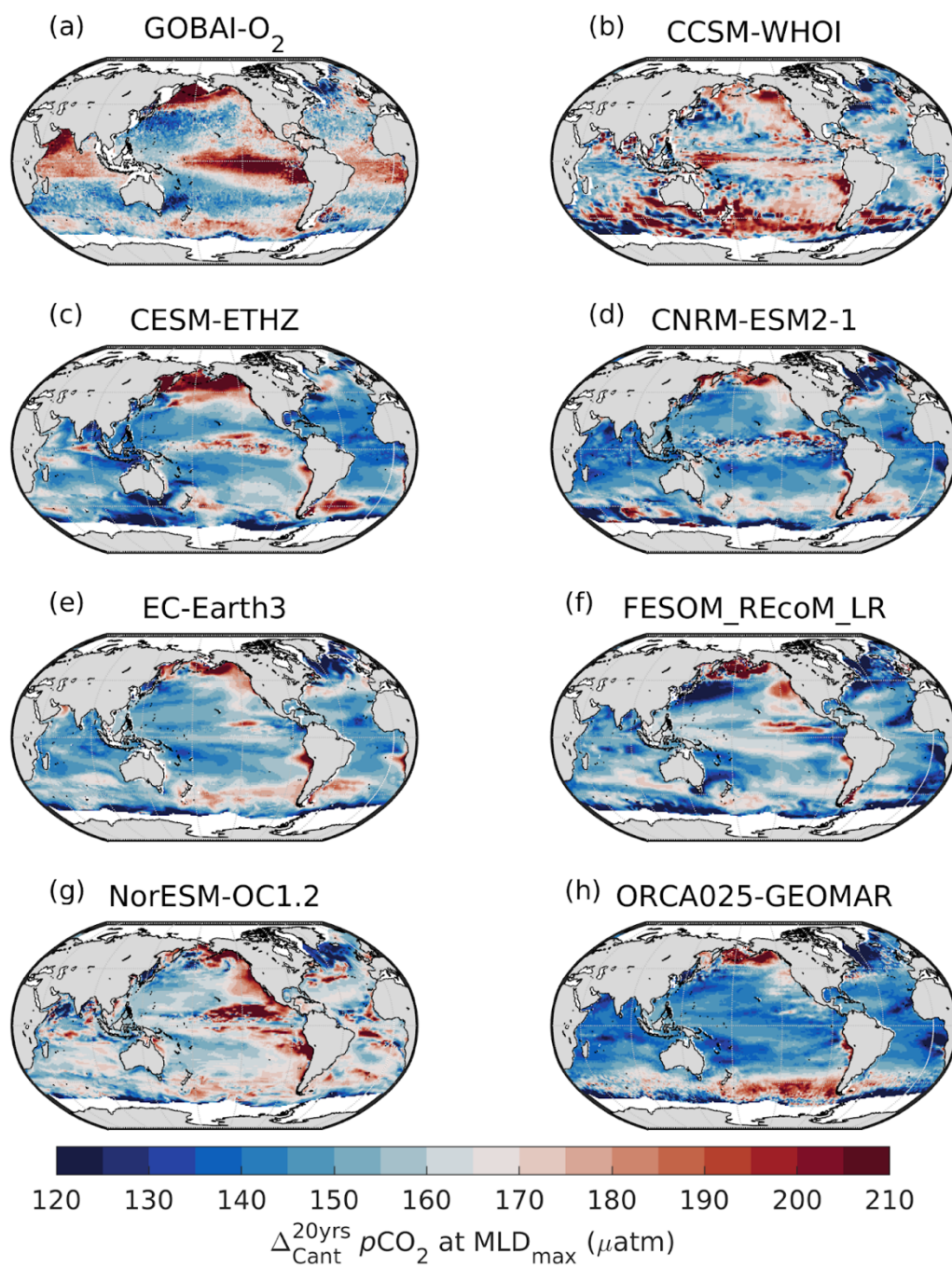


Figure 3.6. The total C_{ant} -induced $p\text{CO}_2$ change ($\Delta_{\text{Cant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) projected in 20 years (2034-2038) for (a) GOBAI- O_2 and (b-h) GOBMs based on a linear projection of the local 2009-2018 trends in anthropogenic carbon accumulation. Projections of future C_{ant} at the MLD_{max} are applied to the 2014-2018 mean conditions. $\Delta_{\text{Cant}} p\text{CO}_2$ values are centered on the projected change in atmospheric $p\text{CO}_2$ since the preindustrial period (~ 167 ppm) based on a linear projection of the 2009-2018 trend in atmospheric at Mauna Loa Observatory. Red values indicate where changes in $p\text{CO}_2$ at the maximum mixed layer depth exceed the atmospheric $p\text{CO}_2$ change.

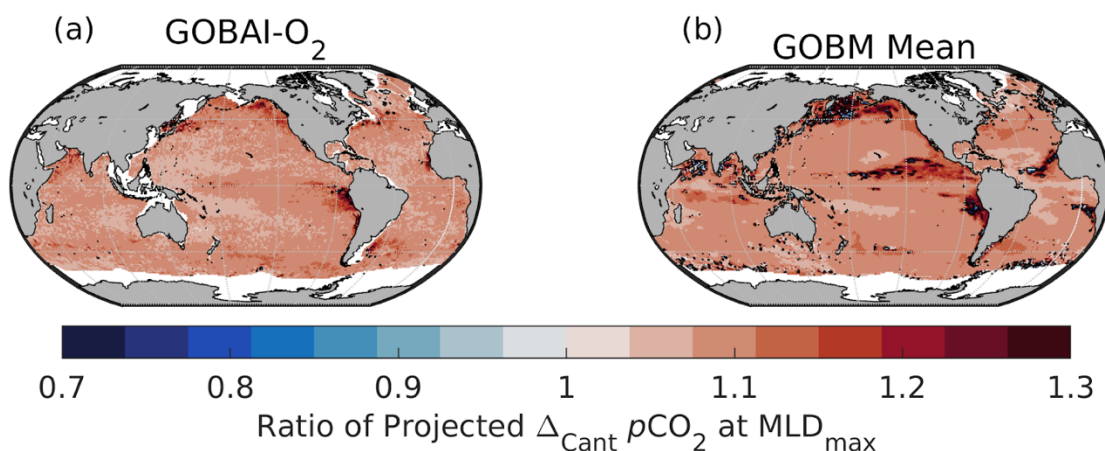


Figure 3.7. The ratio of the total C_{ant} -induced $p\text{CO}_2$ change ($\Delta C_{\text{ant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) between the projected 2024-2028 to 2034-2038 change and the 2014-2018 to the projected 2024-2028 change (ratio = $[\Delta C_{\text{ant}}^{20 \text{ yrs}} p\text{CO}_2 - \Delta C_{\text{ant}}^{10 \text{ yrs}} p\text{CO}_2] / [\Delta C_{\text{ant}}^{10 \text{ yrs}} p\text{CO}_2 - \Delta C_{\text{ant}}^{\text{modern}} p\text{CO}_2]$) for (a) GOBAI- O_2 and (b) ensemble-mean of GOBMs, based on a linear projection of the local 2009-2018 trends in anthropogenic carbon accumulation. Projections of future C_{ant} at the MLD_{max} are applied to the 2014-2018 mean conditions. Red values indicate that $\Delta C_{\text{ant}} p\text{CO}_2$ increase is larger in the latter projected decade.

$p\text{CO}_2$ signals exceed the atmospheric $p\text{CO}_2$ change in the subpolar North Pacific Ocean and expand in spatial extent across the subpolar North Pacific and Southern Oceans (Figure 3.6). In addition, some models show signal emergence in new regions characterized by surface divergence, such as the upwelling zones in the Equatorial Pacific and along eastern boundary current systems off the west coasts of North and South America. While the spatial patterns differ between models, compared to the data-based estimate, the projected $\Delta C_{\text{ant}} p\text{CO}_2$ change from 10 to 20 years into the future is larger in magnitude (by ~10-20%) than the projected $\Delta C_{\text{ant}} p\text{CO}_2$ change from now to 10 years, under the same rate of C_{ant} accumulation (Figure 3.7; Figures S3.18 and S3.19). These results suggest a growing potential for subsurface processes to modulate surface ocean $p\text{CO}_2$ trends through time.

3.4 Discussion

GOBMs generally capture the expected pattern of elevated $p\text{CO}_2$ responses to C_{ant} accumulation below the surface where waters are naturally weakly buffered due to the accumulation of biogenic carbon from organic matter remineralization. However, on average, GOBMs tend to underestimate the magnitude of these interior ocean signals (Figure 3.2), including at the maximum annual MLD (Figure 3.5). The magnitude of C_{ant} -induced $p\text{CO}_2$ change is dictated not only by C_{ant} additions but also by the simulated C_{reg} inventory, as it is the interaction between these carbon pools that leads to nonlinear amplifications of interior $p\text{CO}_2$ change (Figure 3.4; Figure S3.11).

The GOBMs evaluated herein are used by the Global Carbon Budget to estimate the evolving ocean carbon sink on annual timescales (Friedlingstein et al., 2024; Terhaar et al., 2024). In previous work, we found that wintertime surface ocean $p\text{CO}_2$ conditions largely modulate the magnitude of the cumulative annual ocean carbon sink (Arroyo et al., in review). This work aims to clarify whether differences in simulated interior ocean carbonate chemistry significantly affect overlying surface ocean $p\text{CO}_2$ dynamics, particularly on seasonal timescales. We find that GOBMs have incomplete carbon cycle representations below the surface and, as a result, are dissimilarly representing C_{ant} -induced changes at the maximum MLD. Misrepresentation of the subsurface carbon-chemistry feedback in GOBMs could be biasing simulations of surface ocean $p\text{CO}_2$ during the “wintertime window” for C_{ant} uptake (Rodgers et al., 2008) and, thus, estimates of the annual ocean carbon sink strength. While this work highlights the potential for subsurface intensified changes to shape future surface ocean

conditions, our ability to directly quantify the impact on surface ocean $p\text{CO}_2$ and air-sea CO_2 flux remains limited both by observational data availability and structural modeling constraints.

3.4.1 Limitations of Observational Products and GOBMs

One major limitation of our analysis is the lack of a seasonally resolved, observation-based 3D DIC product that accurately captures upper ocean conditions in winter. Existing observation-based DIC data products incorporate synoptic-scale ship-based carbon measurements that are biased toward the Northern Hemisphere and summer and may therefore underrepresent the seasonal carbon cycle processes we seek to characterize (e.g., MOBO-DIC; Keppler et al. 2020; 2023). The GOBAI- O_2 data product incorporates both shipboard and BGC-Argo observations, which fully capture seasonal dynamics, making it well-suited for large-scale, seasonal biogeochemical analyses for O_2 . Importantly, the statistical algorithms we use to infer the ocean carbon cycle from O_2 are derived from shipboard observations. Although we have confidence in the observation-based estimates of interior ocean $p\text{CO}_2$ and C_{ant} , we have less confidence in the seasonal surface ocean $p\text{CO}_2$ values. Surface ocean O_2 equilibrates much more rapidly with the overlying atmosphere compared to CO_2 . Such decoupling of equilibration timescales means that surface ocean $p\text{CO}_2$ predicted from surface O_2 may not reflect wintertime conditions, leading to potential over- or underestimates of sea-air $p\text{CO}_2$ disequilibrium.

The GOBMs considered by RECCAP2 also remain limited in their ability to fully simulate carbon cycle feedbacks due to biases related to model biogeochemistry,

biological processes, and physical circulation that interact in complex ways to influence carbon cycling. These discrepancies may arise due to differences in each model's structural setup and spin-up protocol, which establish the mean-state vertical distributions of DIC through natural solubility and biological carbon pump processes. The initial vertical structure of DIC sets the baseline carbonate system buffering capacity (i.e., vertical RF structure) prior to the additions of C_{ant} . Biases in interior carbon pool distributions lead to biases in the ΔC_{ant} $p\text{CO}_2$ signal throughout the water column, but particularly in thermocline waters where nonlinear contributions amplify C_{ant} -driven trends. In structurally similar ESMs, the biases in surface buffering efficiency are known to cause biases in air-sea CO_2 fluxes (Terhaar et al., 2022), often leading to an underestimation of the C_{ant} burden in the upper ocean. While the influence of biases in surface carbonate chemistry on annual natural and C_{ant} fluxes in GOBMs remains less certain and possibly obscured by spin up durations that are too short (Terhaar et al., 2024), we identify common subsurface biases in the vertical DIC distributions for both the C_{ant} and C_{reg} pools in GOBMs. Further, systematic biases in the seasonal variability of the surface ocean carbon cycle have been identified across RECCAP2 GOBMs (Arroyo et al., in review; Rodgers et al., 2023). Seasonal biases in carbon cycling are particularly relevant for the strength and timing of the surface carbon-chemistry feedback, as the seasonal re-emergence of subsurface C_{ant} (e.g., Rodgers et al., 2020) and C_{reg} couples the interior and surface carbon pools.

Additional inconsistencies in biological carbon pump processes influence natural accumulations of biogenic carbon at depth, particularly as GOBMs differ in

their choice of ecosystem components and do not incorporate calcium carbonate cycle dynamics or dissolved organic matter pools. Model-specific parameterizations of particulate organic carbon production in the surface, export below the euphotic zone, and remineralization below the surface contribute to substantial across-model disagreements in C_{reg} inventories in the interior ocean. RECCAP2 studies find that GOBMs tend to underestimate the export flux at 100 m, generally falling near the lower end of observational-based flux estimates (Doney et al., 2024). Once particles sink from the euphotic zone, they are remineralized back into C_{reg} , using parameterizations based on local temperature, O_2 concentrations, or both, depending on the model. However, progress in constraining biological carbon pump processes in models is challenged by large observational uncertainties in key mechanisms that determine the strength and variability of export pathways (e.g., Boyd et al., 2019; Henson et al., 2022; Siegel et al., 2023; Walker and Palevsky, 2025). Future improvements will likely require integrating results from both field and laboratory studies (e.g., Siegel et al., 2016).

Finally, the vertical distributions of C_{ant} and C_{reg} are sensitive to physical circulation pathways. C_{ant} transport from the surface into the interior is strongly dependent on water mass formation, while C_{reg} accumulates along the flow path of deep and intermediate currents. Across the GOBMs evaluated by RECCAP2, uncertainties in interior ocean carbon storage and fluxes have been linked to large differences in ocean circulation, particularly in the North Atlantic and Southern Oceans (Terhaar et al., 2024). Many GOBMs are commonly biased toward too weak strengths of the Atlantic Meridional Overturning Circulation (AMOC), a primary pathway of C_{ant}

storage into the interior ocean, and exhibit a wide spread in the upwelling salinity (and thus density) of circumpolar deep water in the Southern Ocean, which is transported northward to supply mode and intermediate water masses. Further, our approximations of C_{reg} are estimated by the quantity of O_2 utilized by remineralization processes, assuming that seawater was fully equilibrated with atmospheric O_2 when the parcels last left contact with the surface mixed layer. However, incomplete air-sea equilibration of O_2 at the surface may translate to important biases in C_{reg} , particularly in the Southern Ocean where there is strong vertical mixing (Carter et al., 2021; Jutras et al., 2025).

3.4.2 Future outlooks

To fully capture the evolution of subsurface carbon signals modulated by C_{ant} and C_{reg} , future work may be better suited for fully coupled ESMs that simulate future ocean change, in addition to using a methodology to explicitly track interior carbon cycle changes occurring in consistent water masses. Interior signals of amplified $p\text{CO}_2$ change have not emerged in all GOBM hindcasts at the MLD_{max} ; however, our pseudo-projection analysis using recent C_{ant} trends reveals that modeled signals may become more apparent in the next few decades. Because the historical interior GOBM output archived by RECCAP2 is limited to annual resolution with relatively coarse vertical layers in some models, ESMs may provide a path forward with their ability to simulate future C_{ant} burdens and incorporate future climate change feedbacks that can influence subsurface-to-surface connectivity. In our analysis, we estimate $\Delta_{C_{\text{ant}}} p\text{CO}_2$ at the MLD_{max} by subsampling annual mean profiles at the depth of the maximum monthly

MLD. However, monthly mean MLDs do not capture the deepest signals of mixing, which are episodic (e.g., Holte et al., 2017; Johnson & Lyman, 2022), suggesting our assessments may underestimate signals of interior $\Delta_{\text{Cant}} p\text{CO}_2$ that may potentially influence surface ocean carbon cycling.

One outstanding question is how the amplified subsurface component of the carbon-chemistry feedback interacts with effect of climate change on the ocean. As CO_2 levels rise, climate change is expected to further modify ocean circulation and physical properties of seawater, which in turn can influence ocean carbon uptake. Future climate change will modify the depth of seasonal mixing which could effectively dampen or limit the subsurface to surface interactions via enhanced stratification. Additionally, climate change may alter the biological carbon pump processes and therefore C_{reg} accumulation at depth, which will translate to alterations in the nonlinear component of C_{ant} -induced $p\text{CO}_2$ change. Enhanced stratification may cause organic matter particles to be remineralized closer to the surface due to reduced gravitational sinking and increases in upper-ocean warming could lead to an increase in remineralization rates. While the effect of climate change on the biological carbon pump is largely uncertain (e.g., Walker and Palevsky, 2025), these changes may manifest as a shoaling of the respiration horizon into surface water where C_{ant} content is high, effectively shoaling the amplifying nonlinear response of $p\text{CO}_2$ due to C_{reg} and C_{ant} interactions with potential consequences for air-sea CO_2 fluxes (e.g., Kwon et al., 2009).

3.5 Conclusion

A growing body of work has emphasized the importance of seasonal carbon cycling in the surface ocean for understanding and quantifying the evolving ocean carbon sink and associated feedbacks (Arroyo et al., in review; Fassbender et al., 2018, 2022; Gallego et al., 2018; Goris et al., 2018; Gorgues et al., 2010; Hauck and Volker, 2015; Kessler and Tjiputra, 2016; Riebesell et al., 2009; Rodgers et al., 2008, 2020, 2023). This study builds on this foundation by extending the seasonal surface perspective into the ocean interior (Arroyo et al., 2022; Fassbender et al., 2023), highlighting the role of subsurface processes in modulating seasonal surface carbon dynamics. In our analysis, we use a suite of GOBMs provided by RECCAP2 and an observation-based product (GOBAI-O₂; Sharp et al., 2023) to investigate the evolution of the marine carbon cycle feedbacks in the interior ocean. To more fully capture the role of ocean chemistry in modulating ocean carbon uptake, we introduce the term carbon-chemistry feedback to describe how rising atmospheric $p\text{CO}_2$ leads to intensified changes in ocean $p\text{CO}_2$. This positive feedback on atmospheric $p\text{CO}_2$ levels arises from the accumulation of anthropogenic carbon (C_{ant}) and associated reductions in buffering capacity that result in amplified $p\text{CO}_2$ changes, particularly in the interior ocean. We hypothesized that interior ocean conditions will become increasingly important in influencing surface conditions, particularly when upper-ocean mixing reaches its maximum depth and facilitates the re-emergence of amplified $p\text{CO}_2$ waters.

In line with prior work, the largest C_{ant} -induced increases in $p\text{CO}_2$ occur in the ocean interior, where naturally reduced buffering capacity in high-DIC waters amplifies the chemical sensitivity to added C_{ant} . These subsurface signals of $p\text{CO}_2$

change are highly variable across models and are generally underestimated relative to observational estimates, due to model biases in interior DIC distributions. Our analysis isolated the nonlinear component of C_{ant} -induced $p\text{CO}_2$ change, which arises from the interactions between C_{ant} and regenerated carbon (C_{reg}) supplied by organic matter remineralization. Model discrepancies in the total C_{ant} -induced $p\text{CO}_2$ change reflect discrepancies in the combined inventory and spatial structure of C_{ant} and C_{reg} , which together generate the nonlinear $p\text{CO}_2$ response that dominates the signal in thermocline waters of the North Pacific Ocean. At the depth of the annual maximum mixed layer depth (MLD_{max}), observation-based estimates suggest that the nonlinear component of C_{ant} -induced $p\text{CO}_2$ change pushes the total $p\text{CO}_2$ change to exceed that of the atmosphere, particularly in the North Pacific and Southern Ocean regions. However, C_{ant} -induced $p\text{CO}_2$ at the MLD_{max} are inconsistently captured across GOBMs. While the changes are caused by the combined inventory of both C_{ant} and C_{reg} , across-model differences in the nonlinear component of C_{ant} -induced $p\text{CO}_2$ change at the MLD_{max} are tightly linked to the across-model differences in the inventory of C_{reg} . Our results highlight the need for more accurate representations of biological carbon pump processes in GOBMs, in addition to consistent and sufficiently long preindustrial spin-up procedures, in order to more realistically represent the total C_{ant} -induced change of $p\text{CO}_2$.

In regions such as the subpolar North Pacific and the Southern Ocean, the re-emergence of subsurface waters carrying amplified $p\text{CO}_2$ changes caused by C_{ant} may reduce future ocean carbon uptake. We find that interior trends in C_{ant} -induced $p\text{CO}_2$

changes in GOBMs are increasing over time, suggesting that the translation of subsurface signals to overlying surface waters during winter may become increasingly discernible in the future with C_{ant} accumulation. To test this, we performed a pseudo-projection analysis by extending the most recent decadal C_{ant} trends at the MLD_{max} to 20 years into the future and find that C_{ant} -induced $p\text{CO}_2$ at the MLD_{max} becomes increasingly emergent in each GOBM.

While established frameworks for ocean carbon cycle feedbacks have primarily focused on the surface, annual mean changes (e.g., Aurora et al., 2013; 2019; Friedlingstein et al., 2003), our findings support the shift toward a seasonally resolved, three-dimensional framework of the carbon-chemistry feedback. Such reframing more fully captures the interactions between subsurface and surface carbon cycling and the resulting positive feedback on rising atmospheric CO_2 caused by amplified ocean $p\text{CO}_2$ changes induced by changes in ocean chemistry.

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3.7 Supplementary Information

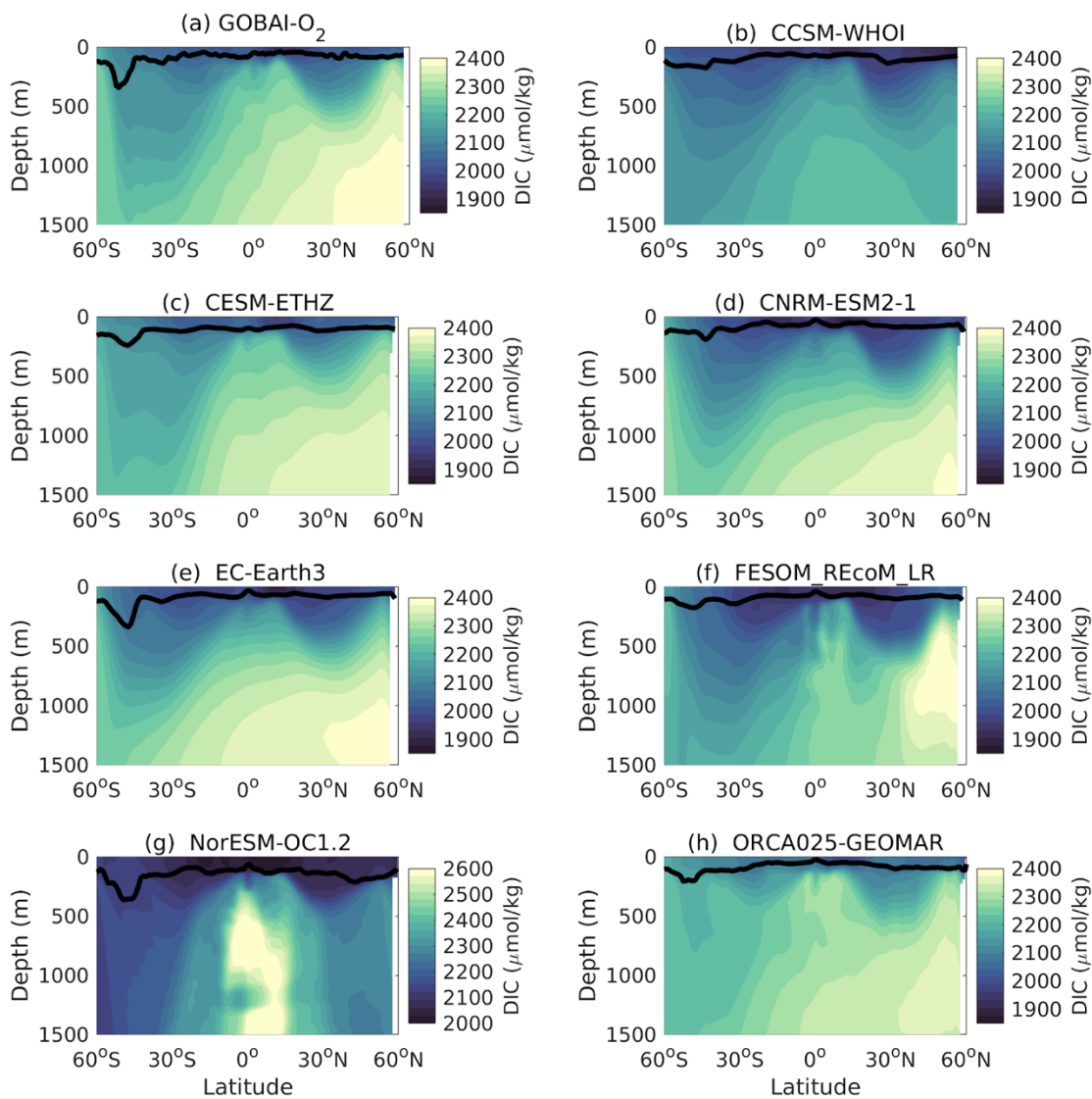


Figure S3.1. Interior distributions of DIC along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI- O_2 and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths. All subplots except (g) have the same colorbar limits.

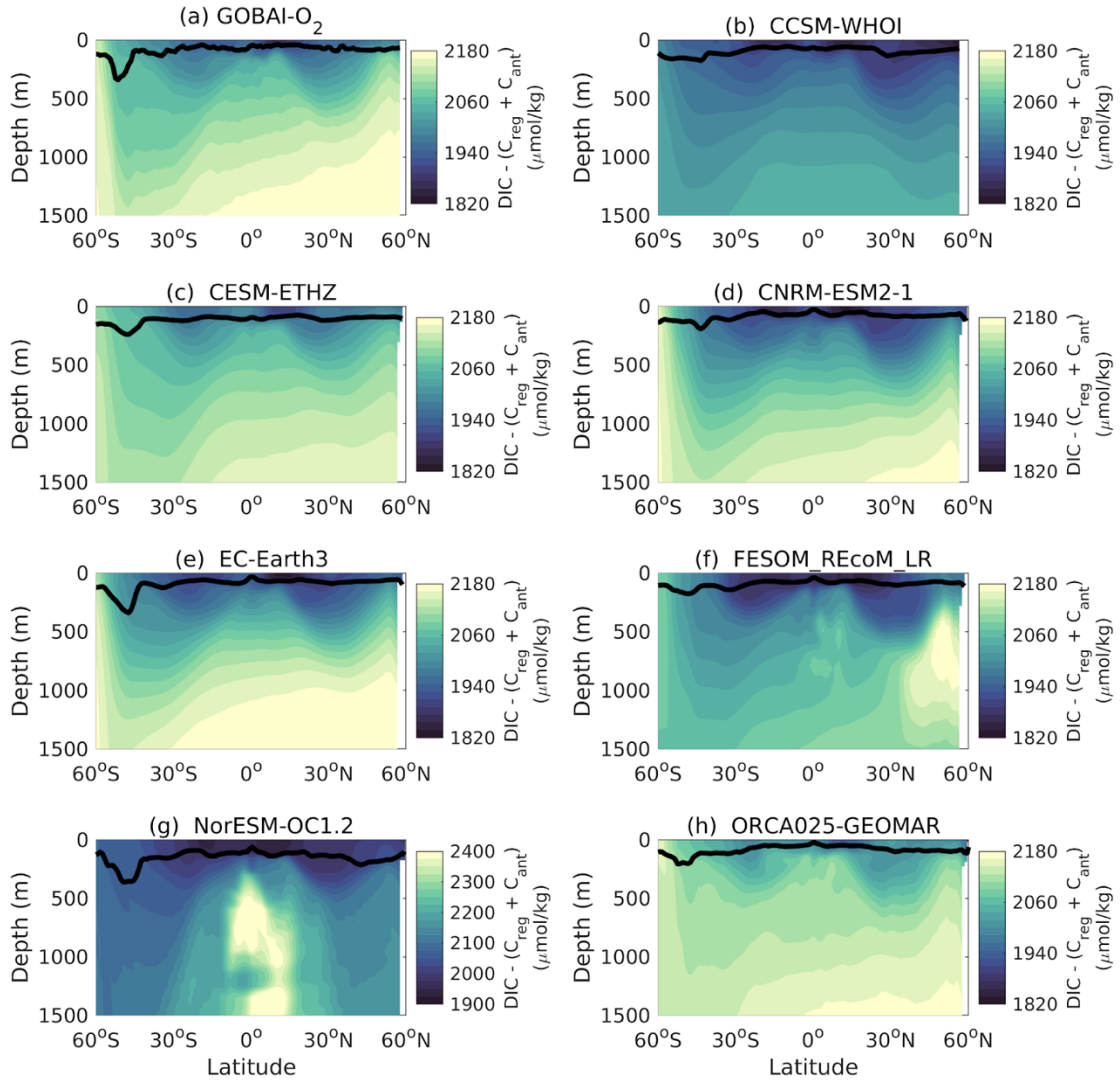


Figure S3.2. Interior distributions of DIC removing the accumulation of anthropogenic carbon (C_{ant}) and regenerated carbon (C_{reg}) from organic matter remineralization ($\text{DIC} - C_{\text{ant}} - C_{\text{reg}}$) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI- O_2 and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths. All subplots except (g) have the same colorbar limits.

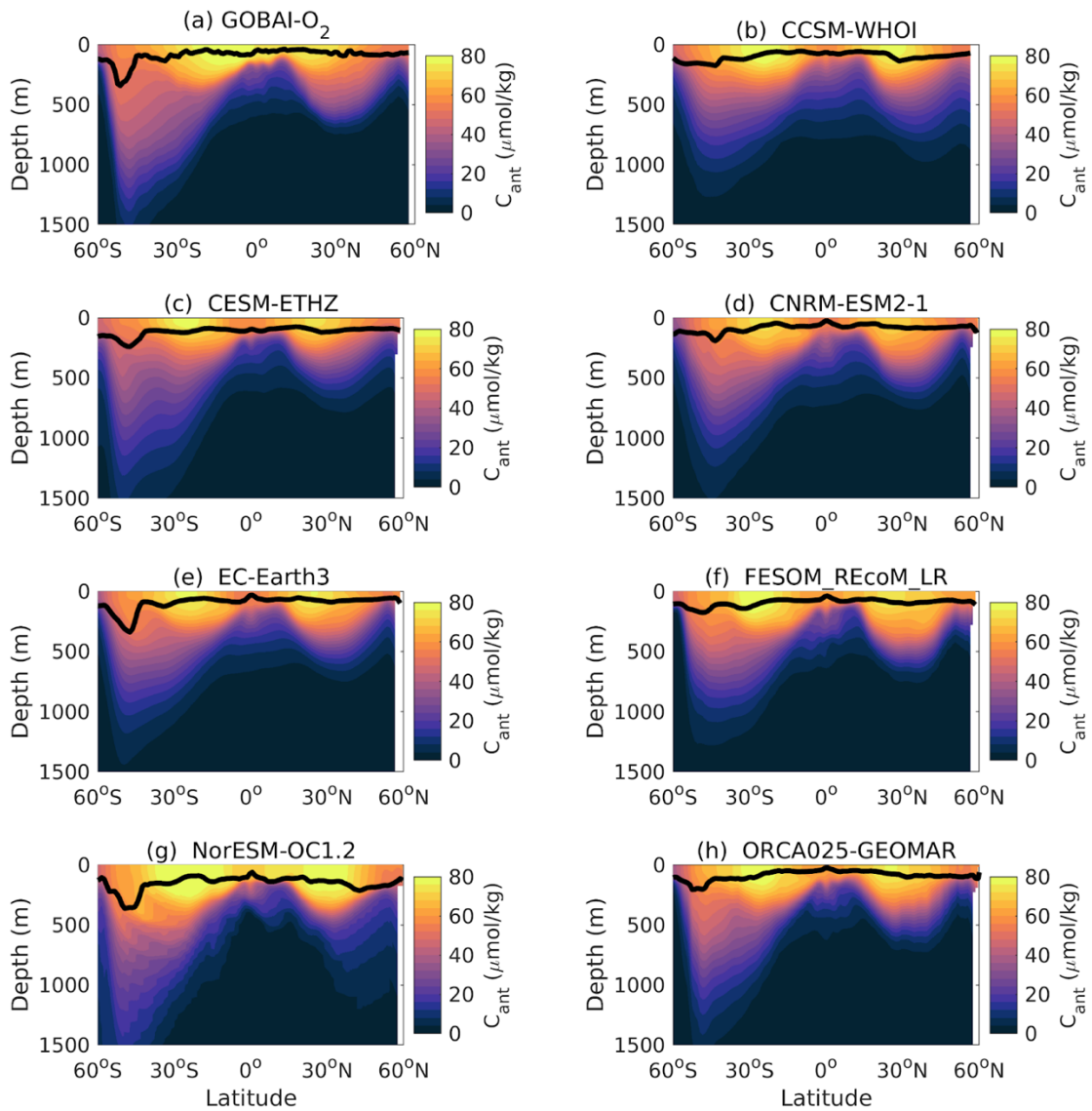


Figure S3.3. Interior distributions of anthropogenic carbon (C_{ant}) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI- O_2 and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths. All subplots have the same colorbar limits.

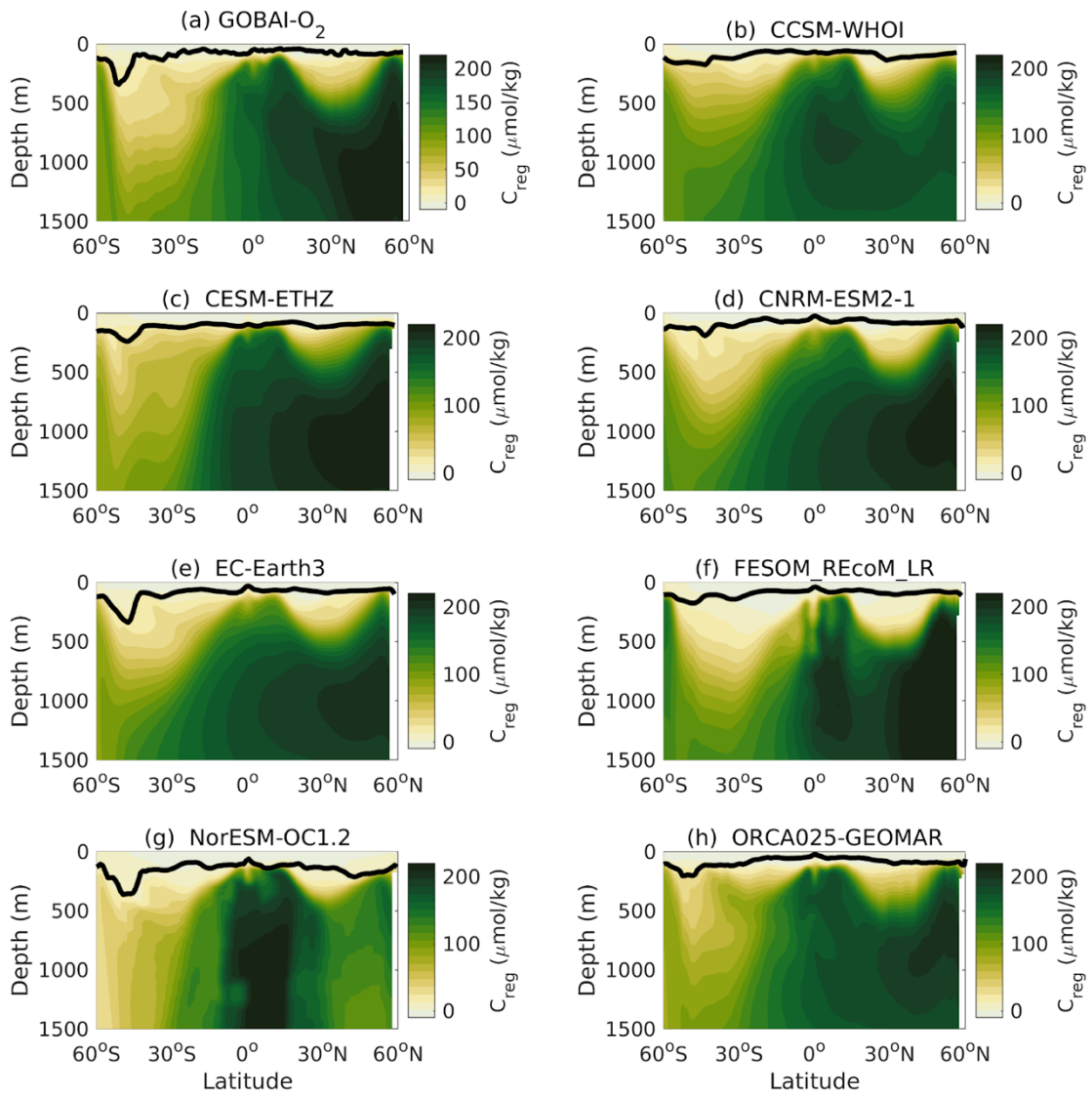


Figure S3.4. Interior distributions of regenerated carbon (C_{reg}) from organic matter remineralization along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI-O₂ and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths. All subplots have the same colorbar limits.

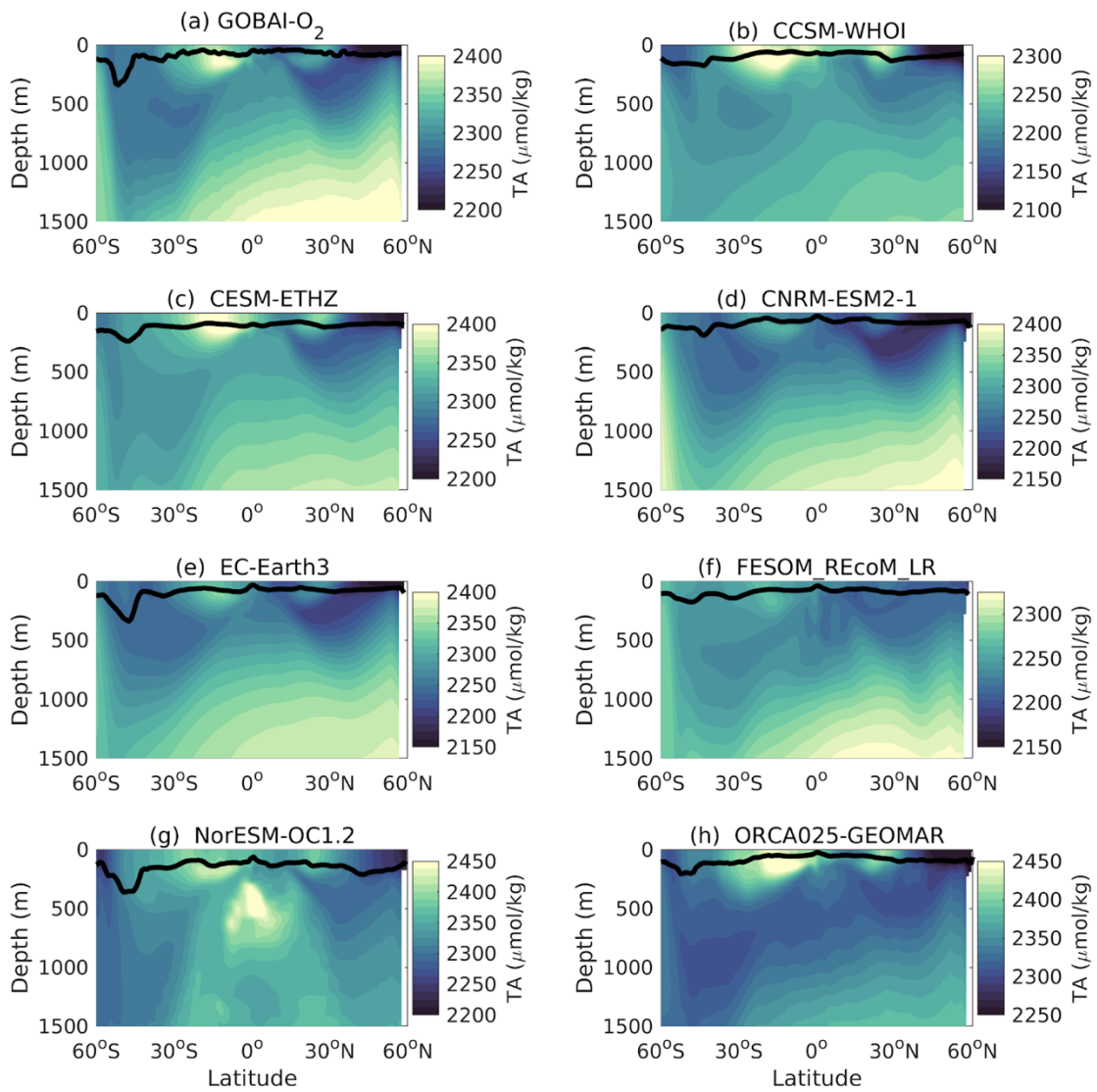


Figure S3.5. Interior distributions of total alkalinity (TA) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI-O₂ and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths.

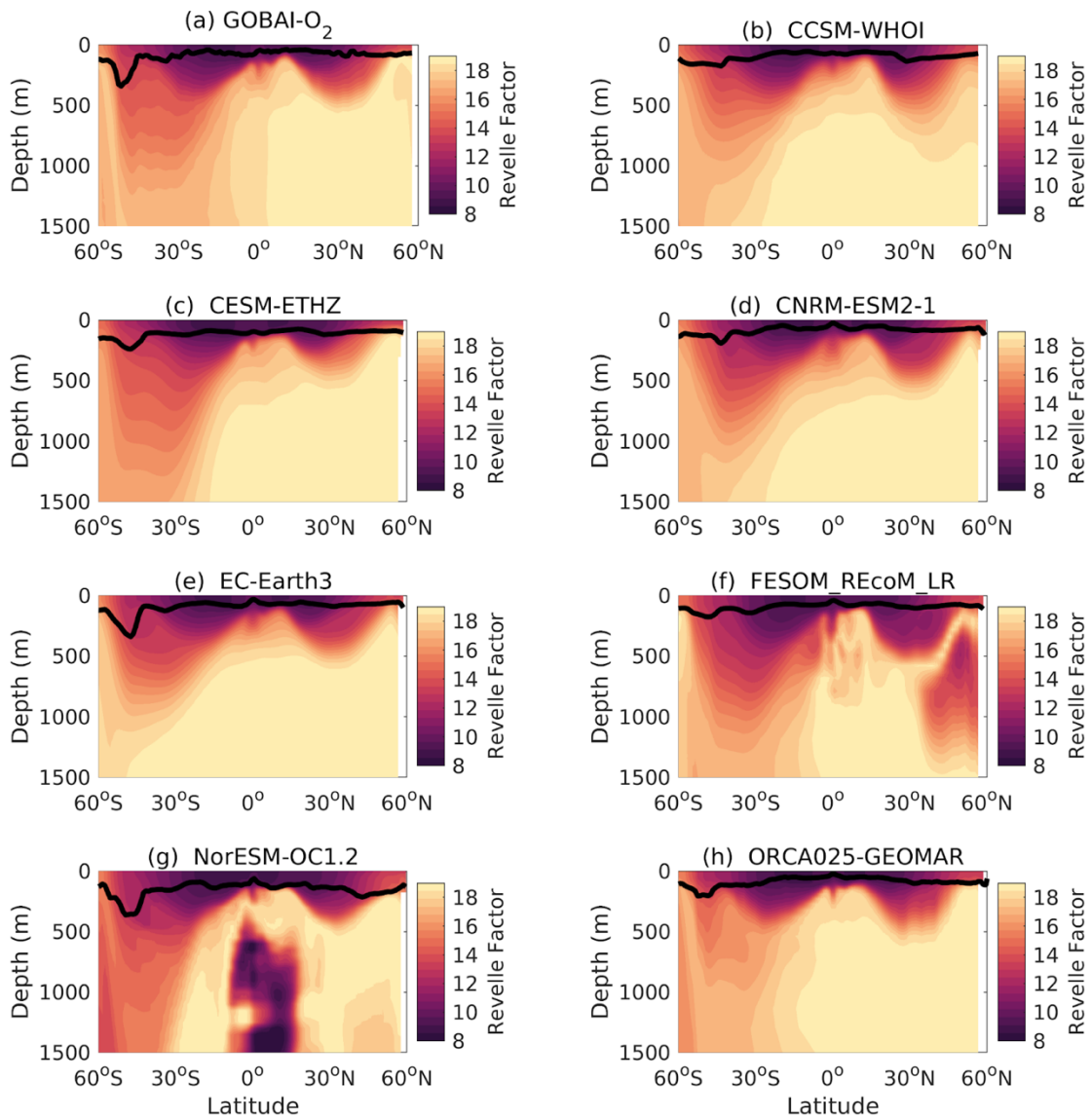


Figure S3.6. Interior distributions of Revelle Factors (RF) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI-O₂ and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths. All subplots have the same colorbar limits.

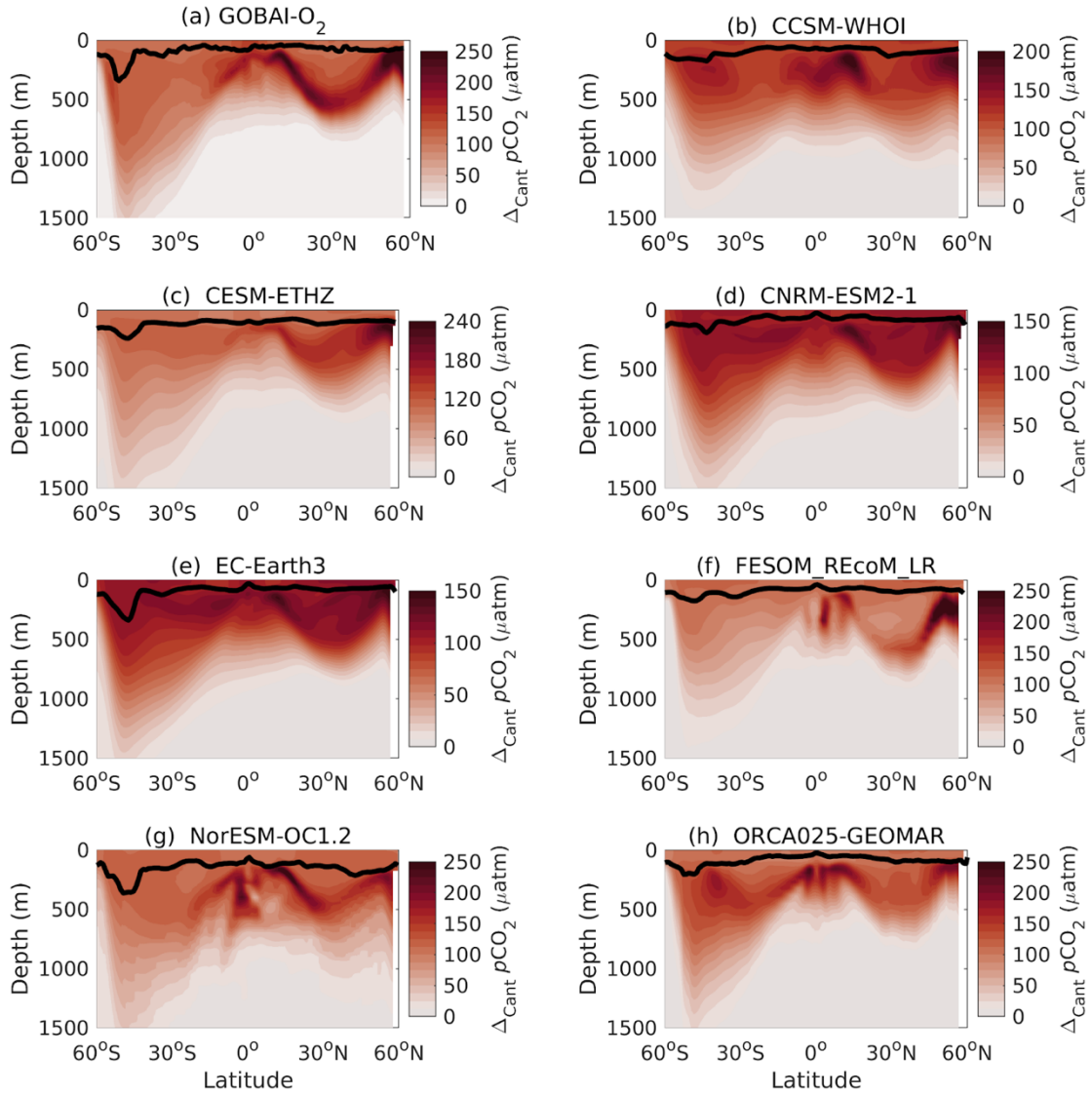


Figure S3.7. Interior distributions of $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}} p\text{CO}_2$) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI- O_2 and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths.

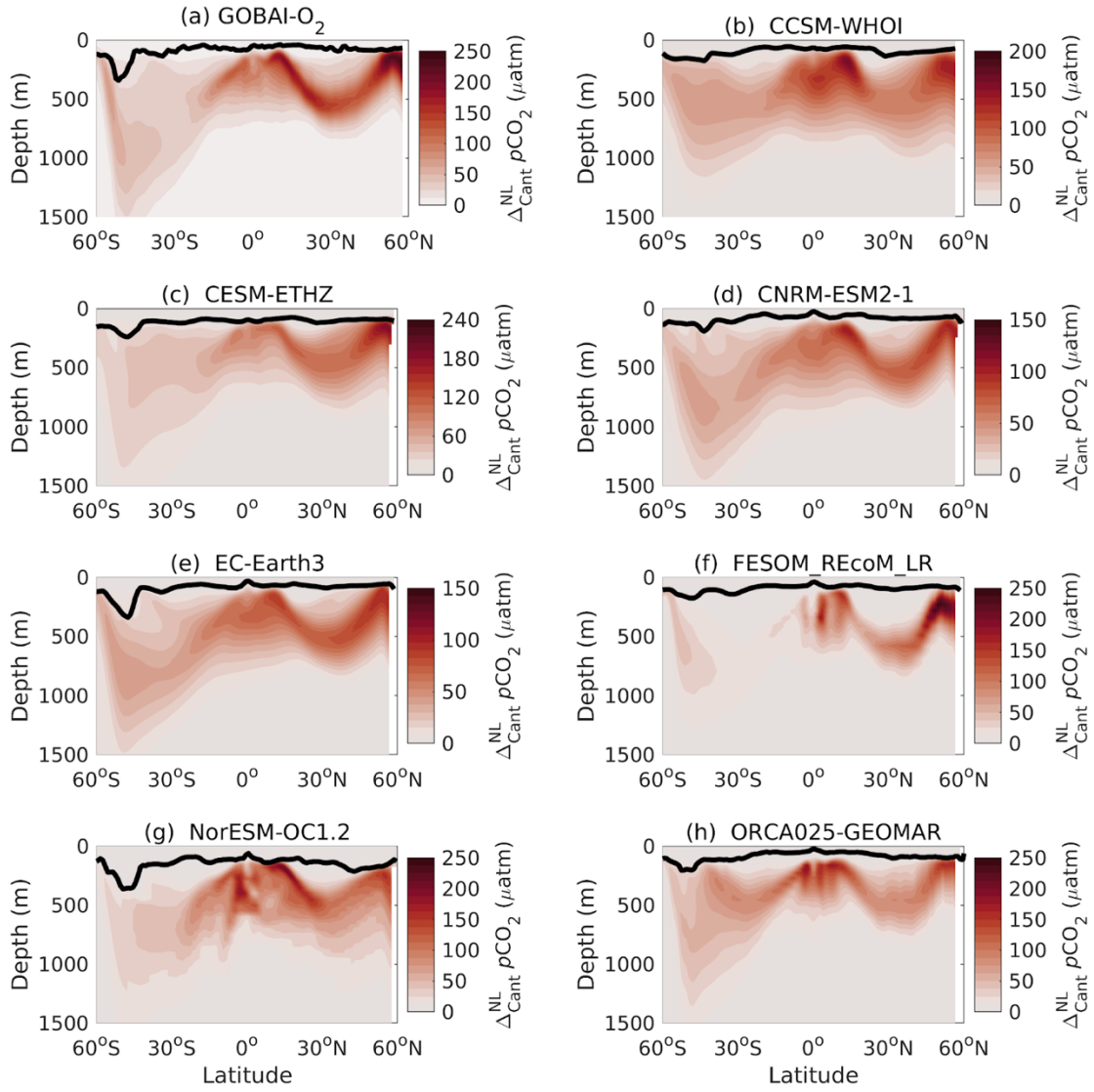


Figure S3.8. Interior distributions of the nonlinear component of $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta^{\text{NL}}_{\text{Cant}} p\text{CO}_2$) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI- O_2 and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths.

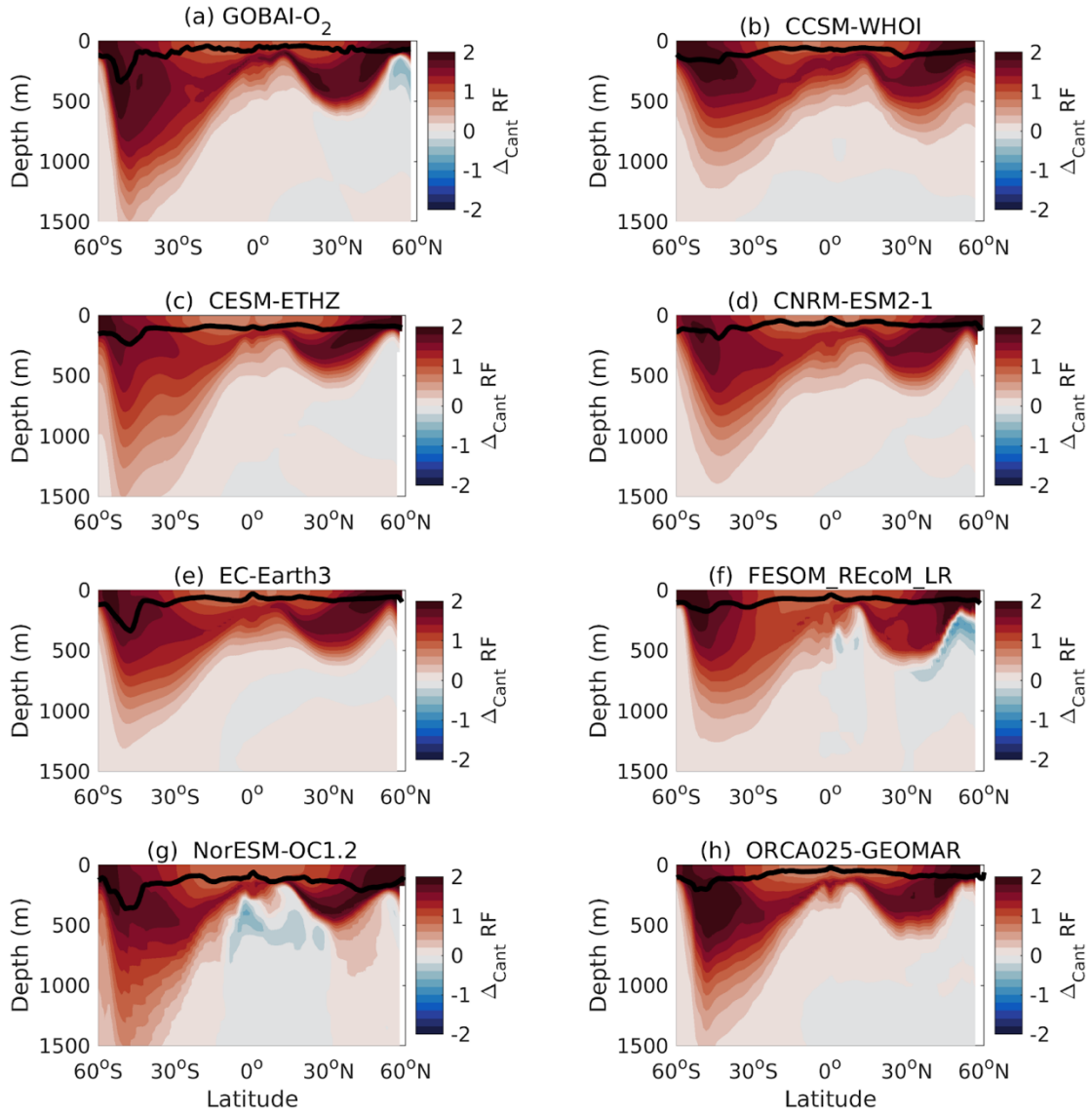


Figure S3.9. Interior distributions of the Revelle Factor change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}} \text{RF}$) along a meridional transect in the Pacific Ocean (150.5°W) from 2014-2018, derived from (a) GOBAI-O₂ and (b-h) individual GOBMs. Black contours represent the 2014-2018 mean maximum mixed layer depths.

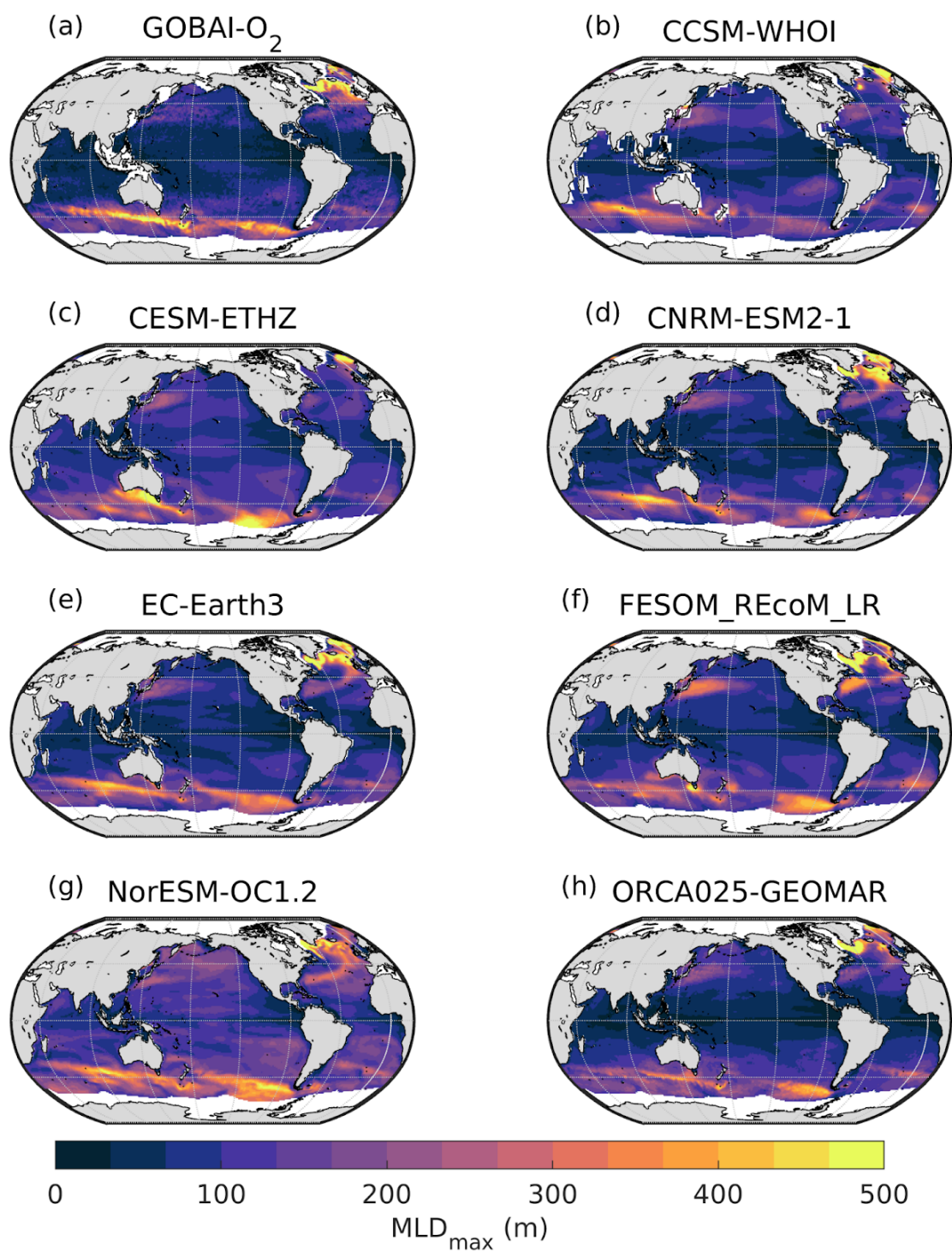


Figure S3.10. 2014-2018 mean maximum mixed layer depth (MLD_{max}) derived from (a) GOBAI-O₂ and (b-h) individual GOBMs.

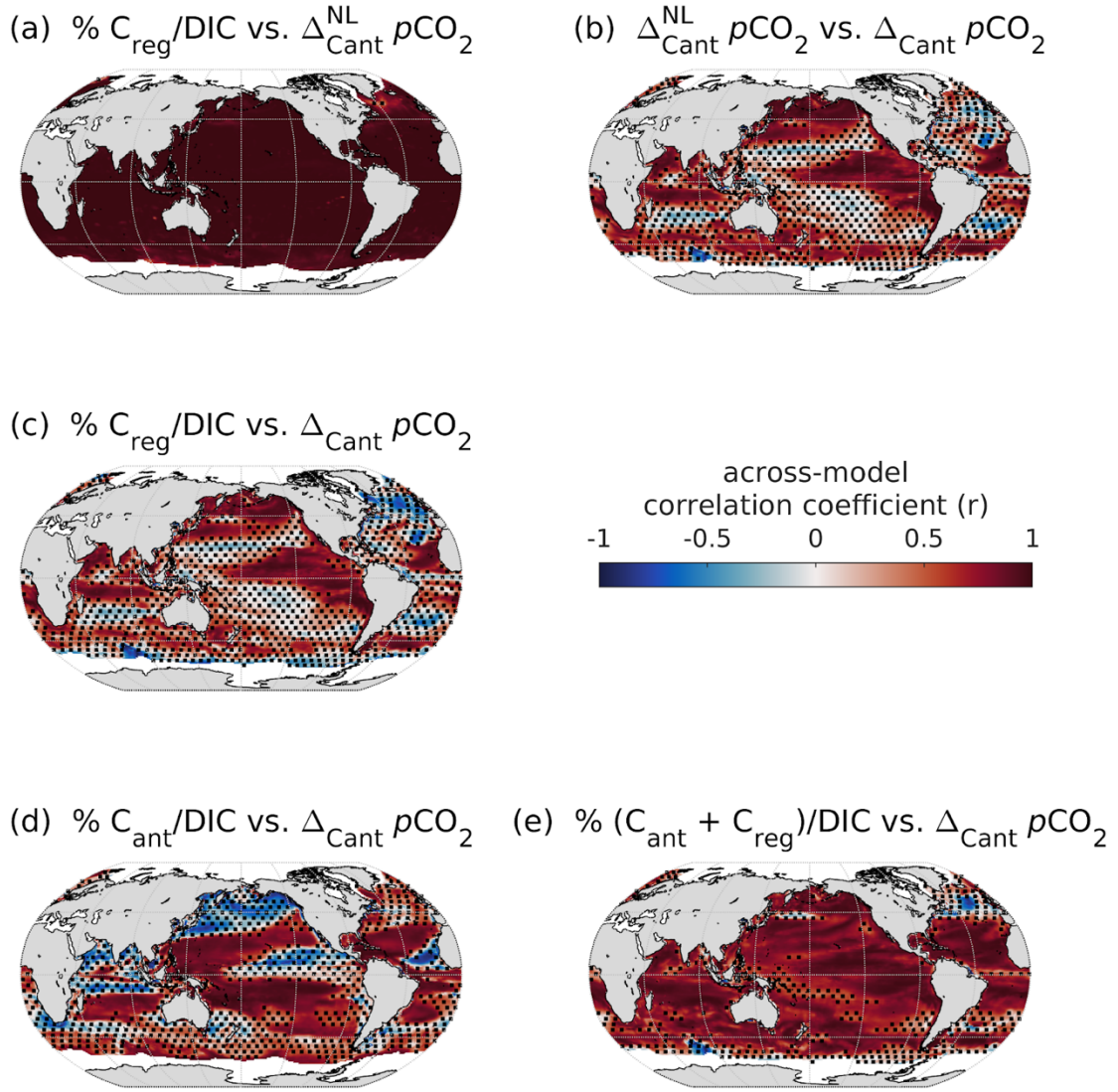


Figure S3.11. Across-model correlation coefficients for the 2014-2018 distributions at the maximum mixed layer depth for (a) percentage of regenerated carbon within the DIC pool ($\% C_{\text{reg}}/\text{DIC}$) vs the nonlinear component of $p\text{CO}_2$ change caused by anthropogenic carbon (C_{ant}) accumulation ($\Delta_{\text{Cant}}^{\text{NL}} p\text{CO}_2$), (b) $\Delta_{\text{Cant}}^{\text{NL}} p\text{CO}_2$ vs the total $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}} p\text{CO}_2$), (c) $\% C_{\text{reg}}/\text{DIC}$ vs. $\Delta_{\text{Cant}} p\text{CO}_2$, (d) $\% C_{\text{ant}}/\text{DIC}$ vs. $\Delta_{\text{Cant}} p\text{CO}_2$, and (e) $\% C_{\text{ant}} + C_{\text{reg}}/\text{DIC}$ vs. $\Delta_{\text{Cant}} p\text{CO}_2$. Stippling marks where correlation coefficients are not statistically significant ($p > 0.1$);

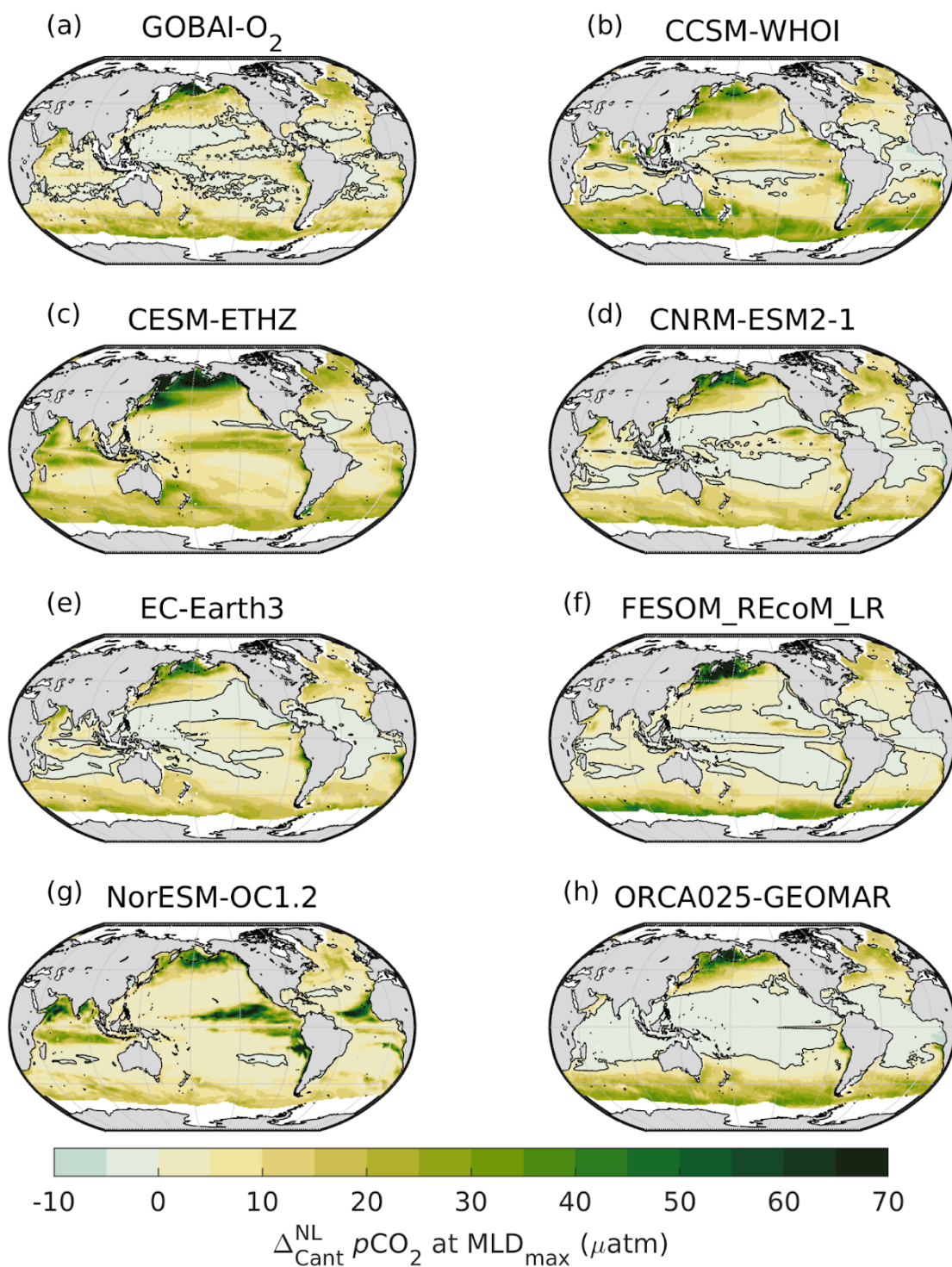


Figure S3.12. 2014-2018 mean distributions of the nonlinear component of $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}}^{\text{NL}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) derived from (a) GOBAI- O_2 and (b-h) individual GOBMs.

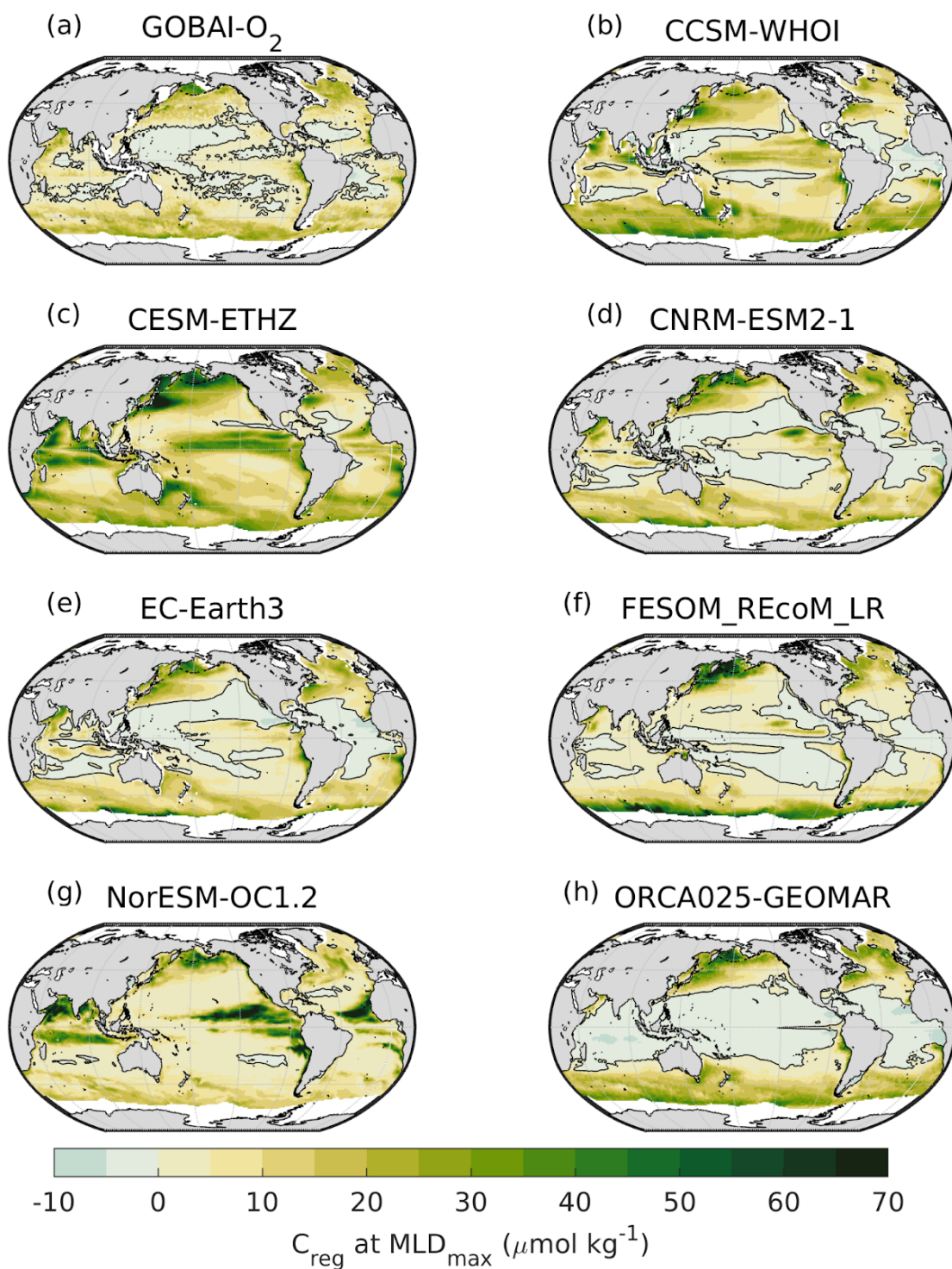


Figure S3.13. 2014-2018 mean distributions of regenerated carbon (C_{reg}) at the maximum mixed layer depth (MLD_{max}) derived from (a) GOBAI- O_2 and (b-h) individual GOBMs.

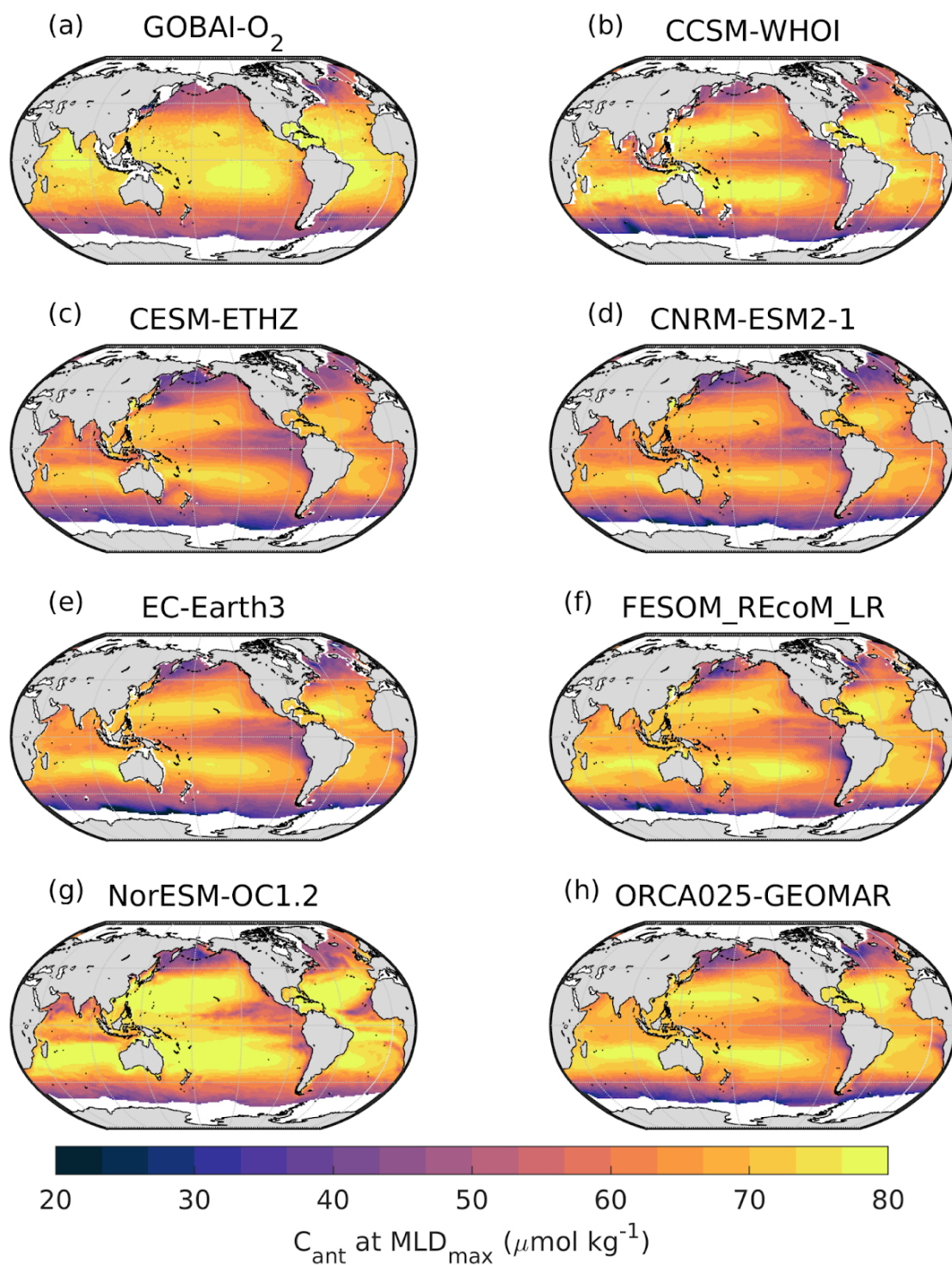


Figure S3.14. 2014-2018 mean distributions of anthropogenic carbon (C_{ant}) at the maximum mixed layer depth (MLD_{max}) derived from (a) GOBAI-O₂ and (b-h) individual GOBMs.

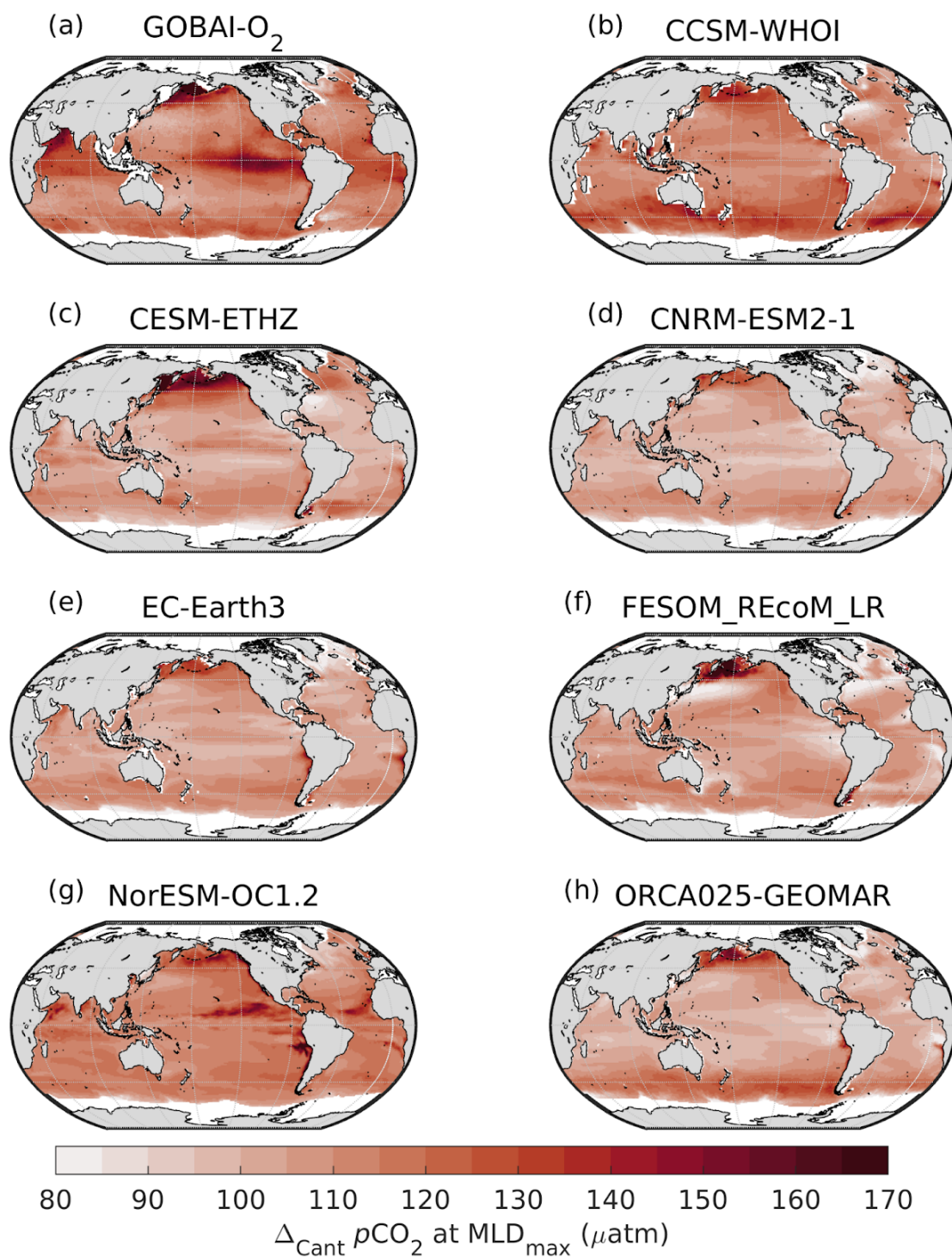
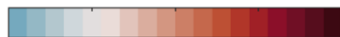


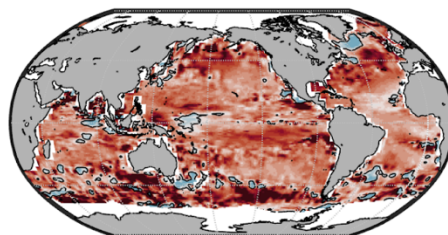
Figure S3.15. 2014-2018 mean distributions of the total $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) derived from (a) GOBAI-O₂ and (b-h) individual GOBMs. This is the same as Figure 5 in the main text but visualized with a gradient colorbar.

Ratio of 2009-2018 to 1980-1989 trend
in $\Delta p\text{CO}_2$ at the MLD_{max}

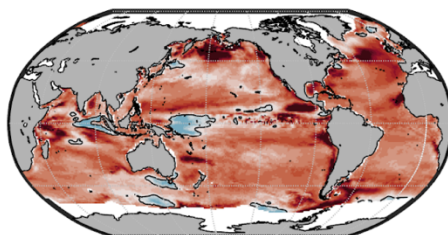
0.5 1 1.5 2 2.5



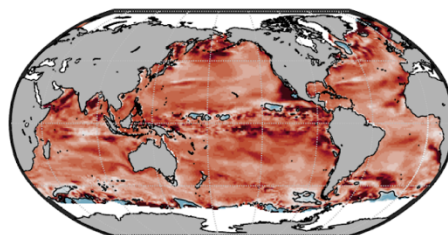
(a) CCSM-WHOI



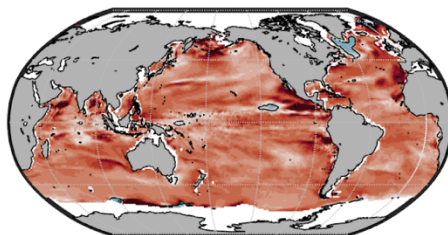
(b) CESM-ETHZ



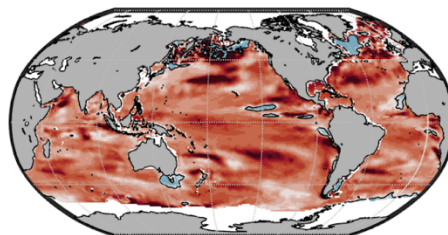
(c) CNRM-ESM2-1



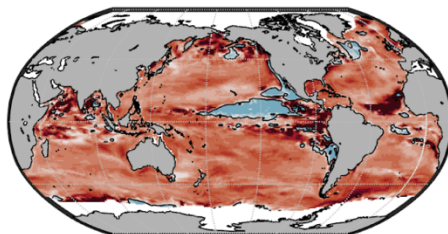
(d) EC-Earth3



(e) FESOM_REcoM_LR



(f) NorESM-OC1.2



(g) ORCA025-GEOMAR

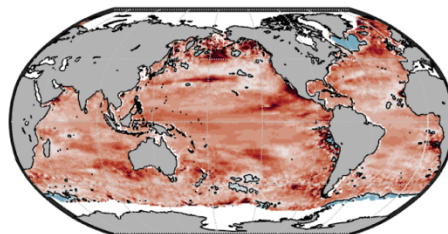


Figure S3.16. Ratio of the trends in $p\text{CO}_2$ change caused by anthropogenic carbon accumulation ($\Delta_{\text{Cant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) between 2009-2018 and 1980-1989 for individual models. Red indicates where the 2009-2018 trend is larger than the 1980-1989 trend.

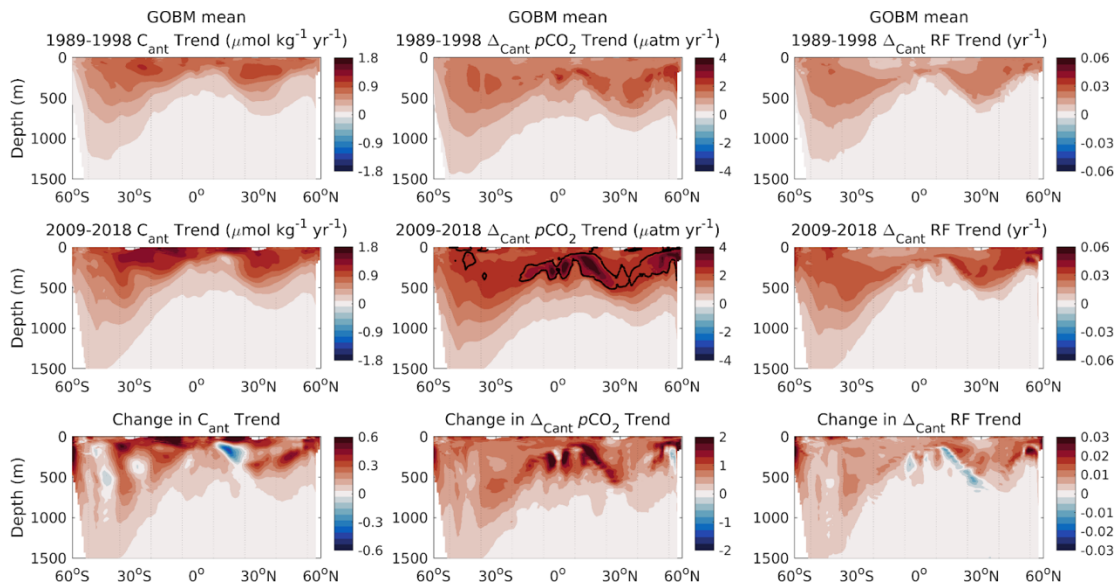


Figure S3.17. Ensemble-mean trends (a,d,g) C_{ant} , (b,e,h) $\Delta_{\text{Cant}} p\text{CO}_2$ and (c,f,h) $\Delta_{\text{Cant}} \text{RF}$ for the time periods of (row 1) 1989-1998 and (row 2) 2009-2018 and (row 3) their difference in trends between the two time periods (20 years). In panels (b) and (e), black contours outline the observed trend in atmospheric $p\text{CO}_2$ (1989-1998 trend = 1.42 $\mu\text{atm}/\text{year}$; 2009-2018 = 2.39 $\mu\text{atm}/\text{year}$) at the Mauna Loa Observatory. Stippling indicates where the spread in the ensemble mean trends exceeds 30% of the ensemble mean signal.

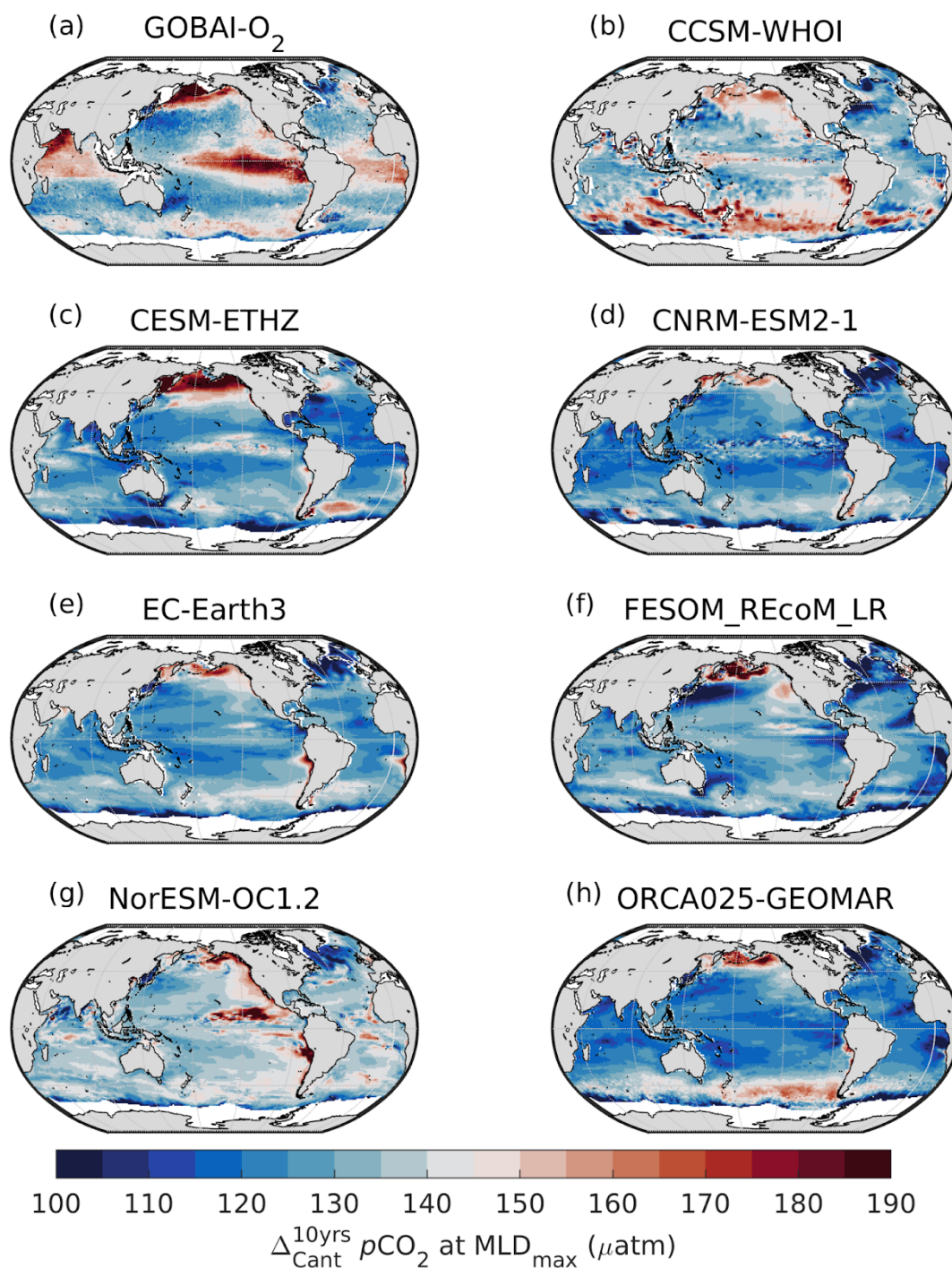


Figure S3.18. The total C_{ant} -induced $p\text{CO}_2$ change ($\Delta_{\text{Cant}} p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) projected in 10 years for (a) GOBAI-O₂ and (b-h) GOBMs, centered on the projected change in atmospheric $p\text{CO}_2$ since the preindustrial period (~145 ppm). Projections of future C_{ant} at the MLD_{max} were applied to the 2014-2018 mean conditions. Red values indicate where changes in $p\text{CO}_2$ at the maximum mixed layer depth exceed the atmospheric $p\text{CO}_2$ change

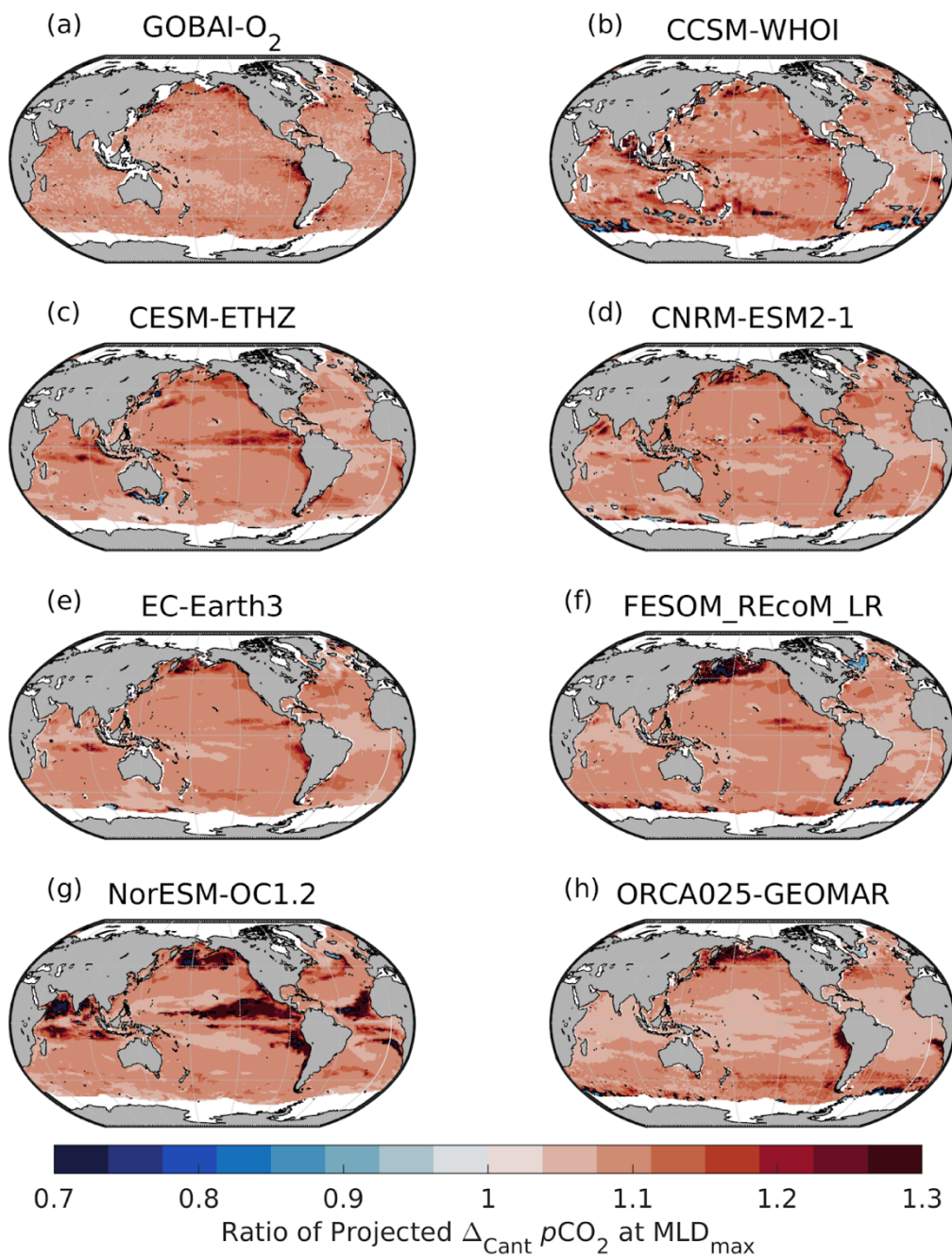


Figure S3.19. The ratio of the total Δ_{Cant} -induced $p\text{CO}_2$ change (Δ_{Cant} $p\text{CO}_2$) at the maximum mixed layer depth (MLD_{max}) between the projected 2024-2028 to 2034-2038 change and the 2014-2018 to the projected 2024-2028 change (ratio = $[\Delta_{\text{Cant}}^{20 \text{ yrs}} p\text{CO}_2 - \Delta_{\text{Cant}}^{10 \text{ yrs}} p\text{CO}_2] / [\Delta_{\text{Cant}}^{10 \text{ yrs}} p\text{CO}_2 - \Delta_{\text{Cant}}^{\text{modern}} p\text{CO}_2]$) for (a) GOBAI- O_2 and (b-h) individual GOBMs, based on a linear projection of the local 2009-2018 trends in anthropogenic carbon accumulation. Projections of future Δ_{Cant} at the MLD_{max} are applied to the 2014-2018 mean conditions. Red values indicate that Δ_{Cant} $p\text{CO}_2$ increase is larger in the latter projected decade.

Conclusions

The ocean plays a fundamental role in regulating global climate by absorbing and storing a substantial fraction of the anthropogenic carbon (C_{ant}) released to the atmosphere from human activities. Over the past several decades, much work has been dedicated to understanding how the ocean is responding to this unprecedented rise in carbon content, from both observational and modeling perspectives. While many studies have focused on surface, annual mean changes to quantify the changing ocean carbon sink, recent research has begun to emphasize the importance of seasonality and interior ocean chemistry changes caused by C_{ant} in modulating the ocean's role as a positive feedback on climate. This dissertation builds on that foundation by using a combination of in situ observations, data synthesis products, and output from regional and global ocean models to investigate how C_{ant} accumulation is altering spatial and temporal patterns of ocean carbonate chemistry.

Chapter 1 investigated the signals of interior ocean acidification in the North Pacific Ocean and California Current Large Marine Ecosystem using ship-board observations. This study was made possible by the decades of measurements collected by scientist as part of CLIVAR and GO-SHIP repeat hydrography programs, compiled by GLODAP, in addition to newly available C_{ant} estimates throughout the water column. These datasets allowed us to assess how ocean acidification is evolving in the interior ocean. We found that $p\text{CO}_2$ and $[\text{H}^+]$ exhibit the largest C_{ant} -induced changes in subsurface mesopelagic waters, in contrast to more traditionally explored metrics of Ω_{Ar} and pH that exhibit surface-intensified changes. The amplified subsurface $p\text{CO}_2$

response reflects the reduced buffering efficiency in high-DIC waters due to substantial organic matter remineralization, which amplifies the chemical sensitivity to added C_{ant} . In coastal upwelling regions like the CCLME, subsurface C_{ant} -induced $p\text{CO}_2$ increases expand the volumes of water with hypercapnic conditions ($p\text{CO}_2 \geq 1,000 \mu\text{atm}$) and increase the overlap with hypoxia stress, underscoring the vulnerability of interior ecosystems to ocean acidification. These results provided the foundation for a subsequent global analysis of amplified subsurface signals of ocean acidification (Fassbender et al., 2023).

Chapter 2 investigated how C_{ant} accumulation and climate change have altered surface ocean $p\text{CO}_2$ seasonality over the historical period from 1980 to 2018. This study was made possible by decades of work by observationalists and modelers, united by a common goal of understanding and constraining the ocean's role in the global carbon cycle. Building from prior work from RECCAP2 efforts, this study compared $p\text{CO}_2$ seasonality changes across seven global ocean biogeochemical models (GOBMs) used in the Global Carbon Budget. This chapter demonstrates how seasonality biases inherent to GOBMs lead to substantial differences the magnitude and spatial patterns of $p\text{CO}_2$ seasonality change across the open ocean. Dissimilar model representations of $p\text{CO}_2$ seasonality, particularly during winter, lead to an increasing disagreement in cumulative ocean carbon sink strength estimates between models. By leveraging multiple models, this chapter provides a systematic evaluation of how differences in seasonal carbon cycle dynamics contribute to long-term discrepancies in historical carbon sink estimates.

Building on these insights, Chapter 3 examined whether differences in simulated interior ocean carbonate chemistry in GOBMs significantly influence surface $p\text{CO}_2$ dynamics, particularly on seasonal timescales. This chapter focuses on the interior carbon pool distributions and the amplified signals of C_{ant} -induced $p\text{CO}_2$ change in the subsurface in GOBMs. To compare to GOBMs, this study was made possible by a new era of seasonally-resolved, interior observations made by BGC-Argo floats, transformed into globally gridded data products using machine learning, in addition to updated techniques for estimating interior ocean C_{ant} . Consistent with previous findings, GOBMs simulate amplified subsurface $p\text{CO}_2$ responses to C_{ant} . However, models vary widely and generally underestimate these subsurface $p\text{CO}_2$ signals compared to observations, due to biases in interior DIC distributions. By isolating the nonlinear component of C_{ant} -induced $p\text{CO}_2$ change, which arises from interactions between C_{ant} and natural additions of regenerated carbon (C_{reg}) from organic matter remineralization, we find that discrepancies in subsurface $p\text{CO}_2$ change reflect differences in the combined inventory and spatial distribution of C_{ant} and C_{reg} across models. These nonlinear responses dominate the North Pacific thermocline and strongly influence $p\text{CO}_2$ conditions at the depth of annual maximum mixing in winter, suggesting that misrepresentation of subsurface carbon chemistry feedbacks can bias seasonal and annual ocean carbon sink estimates.

Taken as a whole, this dissertation research has several important implications for the changing ocean carbon cycle. First, the amplified $p\text{CO}_2$ signals in the mesopelagic interior highlight growing concerns about ocean habitability, particularly

for marine organisms that inhabit depth layers where C_{ant} accumulation and reduced buffering capacity are most intense and overlap with additional stressors of low oxygen and increasing temperatures. Second, this work provides a path forward for better constraining the magnitude and variability of the historical ocean carbon sink in GOBM by identifying the processes that influence the subsurface and seasonal expressions of evolving ocean carbon cycle feedbacks. Our findings indicate that the signals of $p\text{CO}_2$ change are not adequately captured in current models and can be improved by constraining the physical, chemical, and biological processes that impact C_{ant} uptake and transport and natural carbon cycling via the biological carbon pump, in addition to adopting more consistent and sufficiently long model spin-up strategies. These efforts will also increase confidence in future climate projections by providing a more accurate representation of historical ocean carbon uptake. Finally, these results have relevance for marine carbon dioxide removal (mCDR) strategies, which aim to enhance ocean uptake and storage of C_{ant} from the atmosphere as a climate change mitigation strategy. These findings show that the effectiveness of mCDR strategies will depend on where and how deep carbon is sequestered, in the context of intensified subsurface carbon-concentration feedbacks that dictate long-term ocean carbon storage.

This dissertation research provides new insight into the evolving role of the ocean in the global carbon cycle and highlights the importance of marine carbon cycle feedbacks in a rapidly changing ocean.