Ocean Carbon Uptake Under Aggressive Emission Mitigation

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Abstract. Nearly every nation has signed the UNFCC Paris Agreement, committing to mitigate global anthropogenic carbon (Cant) emissions and limit global mean temperature increase to 1.5°C. A consequence of emission mitigation is reduced efficiency of ocean Cant uptake, which is driven by mechanisms that have not been studied in detail. The historical pattern of continual increase in atmospheric CO2 has resulted in a proportional increase in Cant uptake. Here, we explore how this proportionality will weaken and find significant effects related to changes in the vertical transfer of Cant from the surface to the deep ocean, and also ocean chemistry. We define ocean uptake growth consistent with an exact proportionality to the atmospheric growth rate, i.e. the historical scaling, to be 100% efficient. Using a model hierarchy consisting of a commonly used one-dimensional ocean carbon cycle model and a complex Earth System Model (ESM), we find that declines in the efficiency of ocean uptake are greatest under aggressive emission mitigation. To understand the drivers of efficiency declines, we use the ESM to compare scenarios with aggressive emission mitigation (1.5°C), intermediate emission mitigation (RCP4.5), and no emission mitigation (RCP8.5). Using the one-dimensional ocean carbon cycle model, we demonstrate how growth of ocean Cant uptake is a balance between enhancement due to a positive atmospheric CO2 growth rate, and decreases due to the positive growth rate of dissolved CO2 in the surface ocean. Without emission mitigation (RCP8.5), changes in efficiency are almost entirely the result of changes in the buffer capacity of the ocean, which accelerates the growth rate of dissolved CO2 in the surface ocean. Under the declining CO2 regime of the 1.5°C scenario, the dominant driver of efficiency decline is the carbon gradient effect, wherein Cant in the ocean interior slows the removal of Cant from the surface. Although the carbon gradient effect is an unavoidable consequence of emission mitigation, it can be reduced by hastily pursuing emission mitigation.

1 Introduction

The ocean has absorbed 39% of the CO2 from industrial era fossil fuel combustion and cement production (Friedlingstein et al., 2019). The rest of the CO2 remains in the atmosphere where it acts as the primary driver of climate change. At the global scale, the partial pressure of CO2 in the atmosphere (pCO2atm) is greater than the partial pressure of CO2 in the surface ocean (pCO2ocn), thus there is a net ocean sink. The difference in partial pressures has grown over time, therefore ocean uptake of atmospheric CO2 has increased over the industrial era (Khatiwala et al., 2009; DeVries, 2014). The carbon that has been added to the ocean and atmosphere as the result of anthropogenic CO2 emissions is referred to as anthropogenic carbon, Cant.
The rate of ocean $C_{ant}$ uptake is controlled by the rate of physical removal of $C_{ant}$ from the surface ocean into the ocean interior. Various processes set the rate of physical $C_{ant}$ removal, with significant contributions coming from vertical diapycnal diffusion and isopycnal eddy diffusion (Bopp et al., 2015; Gnanadesikan et al., 2015). Advection dominates regional patterns $C_{ant}$ fluxes into (reeemergence) and out of (subduction) the seasonal mixed layer (Bopp et al., 2015; Toyama et al., 2017). The large positive and negative signs of the advective flux mostly cancel when globally integrated (Bopp et al., 2015), thus advection does not play a dominant role in setting the globally integrated $C_{ant}$ air-sea flux. In density space, $C_{ant}$ is primarily absorbed in lighter subtropical waters, and then transferred to denser mode and intermediate waters by diapycnal fluxes associated with watermass transformation (Iudicone et al. 2016). Using an effective surface diffusivity ($K_{z,eff}$), i.e. summarizing the net removal by these processes as a single diffusive process, one-dimensional diffusion models have been shown to be consistent with observations and complex models (Gnanadesikan et al., 2015; Oeschger et al., 1975).

Growth of the natural sinks (land biosphere and ocean) has been outpaced by the growth of atmospheric CO$_2$ and thus sink efficiency has declined (Canadell et al., 2007; Raupach et al., 2014). Efficiency of land and ocean sinks is described by the CO$_2$ sink rate ($k_S$; Raupach et al., 2014), which is the combined ocean-land CO$_2$ uptake per unit atmospheric CO$_2$ above preindustrial levels ($C_{ant,A}$; Pg C):

\[
k_S(t) = \frac{F_{ant,L}(t) + F_{ant,M}(t)}{C_{ant,A}(t)}
\]

Where $F_{ant,L}$ (Pg C yr$^{-1}$) is the anthropogenic land sink and $F_{ant,M}$ (Pg C yr$^{-1}$) is the anthropogenic ocean sink. Observations of $k_S$ from 1959-2012 indicate a robust declining trend, and thus the rate of increase in the natural sinks was slower than the accumulation of carbon in the atmosphere. Using a simple climate model, Raupach et al. (2014) attribute the declining trend to slower-than-exponential CO$_2$ emissions growth (~35% of the trend), a decline in the size of major volcanic eruptions, which cause brief periods of global cooling (~25%), response of the natural sinks to a warming climate (~20%), and nonlinear responses to increasing atmospheric CO$_2$ (mostly attributable to ocean chemistry; ~20%).

Slowing of the emissions growth rate, and thus the $pCO_{2}^{atm}$ growth rate, reduces the efficiency of ocean $C_{ant}$ uptake (McKinley et al., 2020; Raupach et al., 2014); this response is related to the timescales of $C_{ant}$ transfer to the ocean interior (Raupach et al. 2014). A reduced $pCO_{2}^{atm}$ growth rate is inevitable, due either to climate policy, or by the eventual exhaustion of fossil fuel reservoirs. Nearly every nation has signed the Paris Agreement, which requires participating governments to pledge to mitigate future greenhouse gas emissions in an attempt to limit the global mean temperature increase to 1.5$^\circ$C. While countries’ emissions pledges are incompatible with the 1.5$^\circ$C goal (UNEP, 2019), continued commitment to these emissions pledges will inevitably end the historical pattern of exponential $pCO_{2}^{atm}$ growth in coming decades.

The reductions to efficiency that are attributable to a slowing $pCO_{2}^{atm}$ growth rate will be at least partially compensated by a decrease in the strength of the climate-carbon feedbacks that reduce efficiency of ocean $C_{ant}$ uptake (Friedlingstein et al., 2013; Raupach et al., 2014). Past studies have separated carbon cycle feedbacks into CO$_2$ concentration effects and climate driven effects (Friedlingstein et al., 2013). The CO$_2$ concentration driven effect is the net result of two effects: increased flux driven by increasing $pCO_{2}^{atm}$ and reduced flux driven by a declining buffer capacity of the ocean. The buffering capacity of
the ocean refers to the transfer of absorbed CO$_2$ via chemical reactions into chemical species that do not exchange with the atmosphere. As more CO$_2$ is added to the ocean, buffer capacity decreases (Fassbender et al., 2017). When buffer capacity is reduced, more of the CO$_2$ remains in a form that can exchange with the atmosphere and the efficiency of ocean C$_{ant}$ uptake declines.

Climate driven effects stem from the warming of the surface ocean, which impacts gas solubility and ocean circulation. Projected warming reduces ocean uptake (Friedlingstein et al., 2013) and thus reduces the efficiency of ocean uptake. This impact of warming on future carbon uptake has been quantified using a climate feedback framework (Friedlingstein et al., 2013; Randerson et al., 2015; Schwinger and Tjiputra, 2018). However, the idealized simulations used in these studies have not allowed for quantification of the additional contribution of reduced buffering capacity to reduced ocean carbon uptake.

This work expands upon previous work that has quantified future change in ocean C$_{ant}$ uptake by separately accounting for changes due to buffering. We will compare a future scenario with moderate levels of mitigation (RCP4.5; Meinshausen et al., 2011), and an aggressive mitigation scenario where the 1.5°C target is met (1.5°C; Sanderson et al., 2017) to RCP8.5 using an Earth System model (ESM). In the RCP8.5 scenario (Meinshausen et al., 2011), $pCO_2^{atm}$ increases exponentially and represents our no mitigation baseline. With our set of ocean carbon cycle simulations, we will calculate how warming and reduced buffering, referred to here as chemical capacity, affect ocean C$_{ant}$ uptake.

The contribution of ocean C$_{ant}$ uptake to $k_S$ is referred to as $k_M$:

$$k_M(t) = \frac{F_{ant,M}(t)}{C_{ant,A}(t)}$$  \hspace{1cm} (2)

In the past, $k_M$ has been influenced by a slowing of the CO$_2$ emissions growth rate (McKinley et al., 2020), volcanic aerosol induced cooling of the surface ocean (McKinley et al., 2020), changing ocean chemistry, and changes to physical climate (Friedlingstein et al., 2013).

Under exponentially increasing $pCO_2^{atm}$, constant gas solubility, and constant chemical capacity, $k_M$, would remain constant, and thus by definition, the proportionality between increases in atmospheric CO$_2$ and increases in ocean C$_{ant}$ uptake would also remain constant. Because these conditions approximately describe the historical conditions of the ocean carbon cycle, constant proportionality for ocean C$_{ant}$ uptake has been used as a null hypothesis in studies of the drivers of historical regional and global scale changes in the ocean carbon cycle (Lovenduski et al., 2008; Gruber et al., 2019a). Here we refer to this constant proportionality (i.e. $k_M =$ constant) as the "historical scaling", instead of the seemingly contradictory original term, the "transient steady state assumption" (Gammon et al., 1982; Tanhua et al., 2007; Lovenduski et al., 2008; Gruber et al., 2019a). With constant $k_M$, the evolution of ocean C$_{ant}$ concentration at all points in space also follows the historical scaling. This is because the exponential shape of $pCO_2^{atm}$ is passed on to $pCO_2^{cen}$ by the C$_{ant}$ air-sea flux. The amplifying effect that approximately exponentially increasing $pCO_2^{atm}$ has on the ocean C$_{ant}$ concentration and ocean C$_{ant}$ uptake can be removed using the historical scaling, and in previous work, deviations from the historical scaling for ocean C$_{ant}$ uptake and C$_{ant}$ concentrations have been attributed entirely to internal ocean mechanisms. However, we illustrate that previous work has likely overestimated of the impacts of internal variability on the ocean carbon cycle, given the close relationship between the histor-
ical scaling and $k_M$, which is sensitive to external mechanisms such as volcanic forcings and $pCO_{2}^{atm}$ growth rate (McKinley et al., 2020).

The focus of this work is to determine the primary mechanisms in projections of future climate that will drive reductions to the efficiency of ocean $C_{ant}$ uptake under emission mitigation and, thus, a reduced growth rate of atmospheric CO$_2$. Here we consider efficiency of ocean $C_{ant}$ uptake as a measure of how effectively the input, $pCO_{2}^{atm}$, is converted into the desired output, ocean $C_{ant}$ uptake. More efficient ocean $C_{ant}$ uptake would result in the ocean absorbing more carbon at a given $pCO_{2}^{atm}$. In the following sections, we use a one-dimensional ocean carbon cycle model to diagnose the mechanisms of efficiency decline in climate projections from a complex ESM, with the goal of understanding future changes in the efficiency of ocean $C_{ant}$ uptake.

2 Methods

In this section, we develop equations for ocean sink efficiency and introduce our one-dimensional ocean carbon cycle model. We identify the mechanisms driving changes in efficiency by comparing the behavior of a one-dimensional ocean carbon cycle model to an ESM, the Community Earth System Model (CESM). CESM simulations are publicly available, provided by the National Center for Atmospheric Research (NCAR). The one-dimensional ocean carbon cycle model emulates the ocean circulation and carbon cycling of the ESM, but allows for rapid integration to facilitate mechanistic exploration.

2.1 Efficiency Metric and Historical Scaling

Ocean sink rate ($k_M$; Equation 2) represents the efficiency of ocean $C_{ant}$ uptake. The efficiency metric used here, $\eta$, is $k_M$ referenced to the year 1990, so that efficiency may be expressed as a percentage:

$$\eta(t) = \frac{k_M(t)}{k_M(1990)} \times 100$$

Referencing $k_M$ to 1990 values maximizes the time ocean $C_{ant}$ uptake is at 100% efficiency during the historical period, 1920-2006 (Figure S1). The historical scaling for ocean $C_{ant}$ uptake ($F_{ant}$) is closely related to $k_M$:

$$\dot{F}_{ant}(t) = k_M(1990)C_{ant,A}(t) = F_{ant}(1990)\frac{C_{ant,A}(t)}{C_{ant,A}(1990)}$$

The rightmost expression in Equation 4 is the $F_{ant}$ historical scaling, and is based on the assumption of constant efficiency, and thus mathematically equivalent to extrapolating $F_{ant}$ using a fixed $k_M$ (Equation 2.4). The overset “*” notation indicates the variable that has been extrapolated with the historical scaling. When using the historical scaling, $F_{ant}(1990)$ is diagnosed from the CESM, and $\dot{F}_{ant}(t)$ is obtained mathematically by extrapolating $F_{ant}(1990)$ based on the relative increase in atmospheric $C_{ant}$ from 1990 values ($\frac{C_{ant,A}(t)}{C_{ant,A}(1990)}$). For example, $F_{ant}(1990)$ simulated by CESM was 1.7 Pg $C_{ant}$ yr$^{-1}$, and the atmospheric perturbation in 1990 was 74 ppm ($\sim$157 Pg C). In the future, if the atmospheric perturbation is doubled from 1990...
values to 148 ppm (∼314 Pg C), $F_{\text{ant}}$ would also double to 3.4 Pg $C_{\text{ant}}$ yr$^{-1}$. Expressing ocean carbon sink efficiency ($\eta$) as Equation 5 illustrates the link to the historical scaling:

$$\eta(t) = \frac{F_{\text{ant}}(t)}{F_{\text{ant}}(t)} \times 100$$  \hspace{1cm} (5)$$

Equation 5 is equivalent to Equation 3, and is obtained by expanding Equation 3 into terms of $F_{\text{ant}}$ and $C_{\text{ant}, A}$. Under the approximately exponential $pCO_{2}^{\text{atm}}$ increase of the historical period, $k_M$ is relatively constant, thus $F_{\text{ant}}(t) \approx \frac{\dot{F}_{\text{ant}}(t)}{F_{\text{ant}}(t)}$ and historical period efficiency is $\sim 100\%$. Because it is approximately equal to $F_{\text{ant}}$, $\dot{F}_{\text{ant}}$ has been used by biogeochemical oceanographers to estimate historical $F_{\text{ant}}(t)$ (Lovenduski et al., 2008). In the future, as $k_M$ declines from 1990 values, $F_{\text{ant}}$ will be less than $\frac{\dot{F}_{\text{ant}}(t)}{F_{\text{ant}}(t)}$, and efficiency will decline. Here $\frac{\dot{F}_{\text{ant}}(t)}{F_{\text{ant}}(t)}$, extrapolated into the future with projected $pCO_{2}^{\text{atm}}$, will be used to represent an upper bound for future ocean $C_{\text{ant}}$ uptake.

While $k_M$ remains constant, the global mean ocean $C_{\text{ant}}$ profile ($C_{\text{ant}}(z,t)$) can also be estimated ($\dot{C}_{\text{ant}}(z,t)$) using the historical scaling (Tanhua et al., 2007; Gruber et al., 2019a):

$$\dot{C}_{\text{ant}}(z,t) = C_{\text{ant}}(z,1990) \frac{C_{\text{ant}, A}(t)}{C_{\text{ant}, A}(1990)}$$  \hspace{1cm} (6)$$

The $C_{\text{ant}}$ historical scaling exists because the $C_{\text{ant}}$ air-sea flux is effectively "pulling" $pCO_{2}^{\text{ocn}}$ towards $pCO_{2}^{\text{atm}}$, so under the observed exponentially increasing $pCO_{2}^{\text{atm}}$, $pCO_{2}^{\text{ocn}}$ and $pCO_{2}^{\text{atm}}$ are both exponential curves. Because ocean chemistry has remained relatively constant over the historical period, we have been able to assume that surface ocean $C_{\text{ant}}$ concentration is linearly related to $pCO_{2}^{\text{ocn}}$. Mathematically, surface ocean $C_{\text{ant}}$ concentration is closely related to the time integral of the $C_{\text{ant}}$ air-sea flux (Methods 2.3). Therefore, because the integral of an exponential is also an exponential, surface $C_{\text{ant}}$ concentration has also increased exponentially, and that exponential is then propagated by ocean circulation to deeper layers. However, looking forward, the linear relationship between $C_{\text{ant}}$ concentration and $pCO_{2}^{\text{ocn}}$ will end due to a decreasing chemical capacity for CO$_2$. Also, the propagation of the surface exponential signal to depth by ocean circulation is not instantaneous, thus when emissions are mitigated, waters towards the surface will be changing in proportion to flattening atmospheric CO$_2$, but deeper in the water column they will be changing in proportion to the exponential atmospheric CO$_2$ signal from decades prior. Future $C_{\text{ant}}(z)$, will deviate from $\dot{C}_{\text{ant}}(z)$.

### 2.2 Ocean Component of the Earth System Model

The CESM provides a realistic simulation of the response of the ocean carbon cycle to climate change. The CESM’s ocean component model, POP2, provides the ocean biogeochemistry output analyzed in sections 3.1 and 3.2. POP2 output is from publicly available CESM climate simulations provided by NCAR. POP2 features 60 vertical levels and a nominal 1° x 1° horizontal resolution. Surface boundary layer physics are parameterized using the K-Profile Parameterization (KPP) of Large et al. (1994). Unresolved advection by eddies is parameterized with the Gent-McWilliams parameterization (1990). Isopyc- nal mixing is parameterized with the Redi (1982) diffusion operator. The biogeochemical output comes from the embedded
Biogeochemical Elemental Cycle (BEC) model (Moore et al., 2004). \( C_{\text{ant}} \) concentration is calculated in the model as the difference between natural carbon \( (C_{\text{nat}}) \), a tracer that experiences a fixed preindustrial \( pCO_2^{\text{atm}} \), and contemporary carbon \( (C_{\text{con}}) \), a tracer that experiences time evolving \( pCO_2^{\text{atm}} \).

All of the climate simulations used here are forced with \( pCO_2^{\text{atm}} \) from the Representative Concentration Pathways (RCPs) or a 1.5°C scenario (Sanderson et al., 2017) from 2006-2080. Over the historical period, 1920-2005, these simulations are forced with observations of \( pCO_2^{\text{atm}} \). For the 1.5°C scenario, a concentration pathway was designed that limited warming the CESM to 1.5°C, for the purpose of investigating avoided climate impacts (Sanderson et al., 2017). This scenario features the same forcing as RCP8.5 until 2017, except for CO\(_2\). The projected CO\(_2\) forcing was not smoothly joined to the historical CO\(_2\) forcing, creating a period of low \( C_{\text{ant}} \) flux, which the ocean \( C_{\text{ant}} \) sink recovers from by 2017 (Figure S2).

Multiple simulations are run with the same \( pCO_2^{\text{atm}} \) forcing to generate single model ensembles for each scenario. The ensemble approach allows for separation of internal variability from the forced signals, with the latter being the focus of this study. NCAR has run multiple ensembles with different forcings including CESM Large Ensemble (40 members, RCP8.5; Kay et al. 2014), CESM Medium Ensemble (15 members, RCP4.5), and the CESM Low-Warming Ensemble (10 members, 1.5°C; Sanderson et al., 2017). Ocean biogeochemistry output is limited to 9 members for the medium ensemble and the 3 for the low warming ensemble, thus we also only use 9 ensemble members for the RCP8.5 experiment.

In coupled climate models, historical climate variability of the carbon sink is not expected to match observations because the phasing of ENSO or other internal climate variability is different in each ensemble member. Averaging across an ensemble removes the imprint of internal variability leaving only the climate system response to external forcing. With only a single coupled climate simulation, decadal means are used to smooth internal climate variability, but with an ensemble, a single year of the ensemble mean provides a snapshot of the climate response to external forcing. All output analyzed here is the ensemble mean because we are focused on the externally forced signal. CESM is used for maps and sections, and we tune a one-dimensional model to replicate its global mean behavior to elucidate the underlying mechanisms.

### 2.3 One-Dimensional Ocean Carbon Cycle Model

The behavior of the one-dimensional ocean carbon cycle model is more easily interpreted than the behavior of the complex ESM. Here we employ an established one-dimensional ocean carbon cycle model that is based on impulse response functions. This model is easily interpretable and has been used for decades to emulate ocean carbon uptake simulated by complex ESMs (Joos et al., 1996; Meinshausen et al., 2011; Raupach et al., 2014). The impulse response function form of our model has been used to convert emissions projections into the CO\(_2\) concentration pathways (RCP4.5, RCP8.5) that are used to force the CESM, as well as others in the CMIP5 suite of ESMs (Meinshausen et al., 2011) used in IPCC AR5 (Cias et al., 2013).

Impulse response functions can be used to characterize the response of dynamical systems to small perturbations around a steady state. In our case, the small perturbation is the \( C_{\text{ant}} \) perturbation to the preindustrial carbon cycle. With this method, the full response of the system is considered to be the sum of the system’s response to infinitely many discrete pulses. For the ocean carbon cycle, a pulse is the \( C_{\text{ant}} \) added to the surface ocean by air-sea exchange for a time frame specified by the one-dimensional model’s time step. The impulse response function describes how long that \( C_{\text{ant}} \) pulse remains in surface ocean.
Air-sea flux and surface concentration are calculated at each time step, while the response function is fixed. The surface ocean is mathematically expressed as the convolution integral of the impulse (air-sea flux) and the impulse response function (lifetime of that pulse). A conceptual version of this approach is illustrated in Appendix A.

The one-dimensional model is forced with the same historical and projected \( pCO_2^{atm} \) that is used to force the ocean component of the ESM, and historical and projected SST change simulated by the ocean component of ESM. With this one-dimensional model we can calculate the annual mean surface concentration and the globally integrated air-sea flux. The one-dimensional model consists of two equations (Equation 7,10) that are solved at each timestep to obtain the air-sea flux of \( C_{ant} \):

\[
C_{ant}(t) = \frac{1}{h} \int_{t_i}^{t} F_{ant}(u) r(t - u) du
\]

This is the convolution integral of \( C_{ant} \) air-sea flux \( (F_{ant}) \) and the mixed-layer impulse response function, \( r(t) \). A convolution integral calculates the concentration at time \( t \) by calculating the fraction of previous pulses \( (F_{ant}(u)) \), that entered the ocean at times \( u \) (from \( t_i = 0 \) to \( t \)), that remain in the surface ocean at time \( t \). The effective mixed layer depth, \( h \), converts the output of the convolution integral into from \( \text{mmol m}^{-2} \) to units of \( \text{mmol m}^{-3} \). We use \( h \) to tune the historical \( C_{ant} \) air-sea flux of the one-dimensional model to the historical \( C_{ant} \) air-sea flux of the ocean component of the ESM. In our case, the optimal \( h \) to match the behavior of the CESM ocean component model is 109 m.

The mixed-layer impulse response function \( (r(t)) \) used here is from Joos et al. (1996), and was diagnosed by those authors from the box-diffusion model, HILDA (HIgh Latitude-exchange/interior Diffusion-Advection). With this method, the response function is fixed throughout time, which is equivalent to the assumption that ocean circulation is constant.

The convolution integral (Equation 7) represents the time integral of a box-diffusion model’s surface \( C_{ant} \) tendency equation:

\[
\frac{\partial C_{ant}}{\partial t} = \frac{F_{ant}}{h} + \frac{K_{z,eff}}{h} \frac{\partial C_{ant}}{\partial z}
\]

\[
C_{ant}(t) = \frac{1}{h} \int_{0}^{t} \left( F_{ant} + K_{z,eff} \frac{\partial C_{ant}}{\partial z} \right) dt
\]

Where \( h \) again, is the effective mixed layer depth (same as Equation 7) and \( K_{z,eff} \) is the effective vertical diffusivity of the one-dimensional model. The one-dimensional model’s \( K_{z,eff} \) must match that of the ocean component of the ESM in order for the growth of \( C_{ant} \) uptake of the one-dimensional model to match that of the ocean component of the ESM (Gnanadesikan et al., 2015). Diagnosing an ocean model’s mixed layer impulse response function diagnoses the net \( C_{ant} \) removal by simulated physical processes, and thus the \( K_{z,eff} \) of the ocean model. However, diagnosing the mixed layer impulse response function requires special simulations (Joos et al., 1996), and is unnecessary if one instead uses \( h \) as tuning parameter (Meinshausen et al., 2011). Experiments with one-dimensional carbon cycle models show that \( K_{z,eff} \) is indirectly tuned by adjusting \( h \) (Oeschger et al., 1975). Thus by changing \( h \) of the one-dimensional model, we can match the \( K_{z,eff} \) of the CESM and therefore the one-dimensional model replicates CESM ocean \( C_{ant} \) uptake over the historical period.
\(C_{\text{ant}}(t)\) and \(F_{\text{ant}}\) are calculated explicitly by the one-dimensional model, and the second term on inside the integral in Equation 9, the diffusive \(C_{\text{ant}}\) flux, is not modeled explicitly by the one-dimensional model, but can be determined exactly by residual. In the one-dimensional model, the \(C_{\text{ant}}\) air-sea flux is calculated as follows:

\[
F_{\text{ant}} = c k_g (pCO_{2\text{atm}}^2 - pCO_{2\text{ocn}}^{\text{cum}})
\]  

(10)

\(F_{\text{ant}}\) is the ocean flux of anthropogenic carbon, which is dependent on the air-sea partial pressure gradient and the gas exchange coefficient (m\(^{-2}\) yr\(^{-1}\)). A conversion factor, \(c\), converts the flux units from ppm m\(^{-2}\) yr\(^{-1}\) to mmol m\(^{-2}\) yr\(^{-1}\). We calculate \(pCO_{2\text{ocn}}^{\text{cum}}\) as the preindustrial \(pCO_{2\text{ocn}}^{\text{cum}}\) (\(pCO_{2\text{ocn},\text{PI}}\)) plus an anthropogenic perturbation (\(\delta pCO_{2\text{ocn}}^{\text{cum}}\)). Model calculation of \(\delta pCO_{2\text{ocn}}^{\text{cum}}\) is based on a parameterization that includes effects of changing buffer capacity and temperature (Appendix B).

### 2.4 Process Decomposition Using One-Dimensional Ocean Carbon Cycle Model Simulations

| Experiment Name                                      | Description                  | Symbol | Scenarios                  |
|------------------------------------------------------|------------------------------|--------|---------------------------|
| All Effects (Control)                                | full chemistry               | \(\Delta C_{\text{total}}\) | RCP8.5, RCP4.5, 1.5°C     |
| Constant Chemical Capacity                           | constant buffer factor       | \(\Delta C_{\text{ccc}}\)  | RCP8.5, RCP4.5, 1.5°C     |
| Historical Scaling                                  | constant efficiency          | \(\Delta C_{\text{hs}}\)   | RCP8.5, RCP4.5, 1.5°C     |

| Effect Name                                           | Effect Symbol                | Equation                       |
|-------------------------------------------------------|------------------------------|--------------------------------|
| Carbon Gradient Effect                                | \(\Delta C_{\text{cgrad}}\)  | \(\Delta C_{\text{ccc}} - \Delta C_{\text{hs}}\) |
| Chemical Capacity Effect                              | \(\Delta C_{\text{chem}}\)   | \(\Delta C_{\text{total}} - \Delta C_{\text{ccc}}\) |

1. \(\Delta C_{\text{hs}}\) is calculated directly from \(pCO_{2\text{atm}}^2\) (\(\dot{C}_{\text{hs}} = \int F_{\text{ant}} \, dt\); Equation 4)

We analyze two additional idealized one-dimensional model experiments for each scenario, for a total of 6 complementary simulations that are used to diagnose the changes seen in the CESM simulations. The naming convention for these experiments and a brief description are listed in Table 1. Cumulative change is denoted as \(\Delta C_X\). In addition to these simulations, we calculate cumulative \(C_{\text{ant}}\) uptake consistent with the historical scaling, \(\dot{C}_{\text{hs}}\), for each scenario. \(\dot{C}_{\text{hs}}\) is calculated directly from the prescribed \(pCO_{2\text{atm}}^2\) (Table 1; Equation 4). With these experiments, we quantify how two negative effects, \(\Delta C_{\text{chem}}\) and \(\Delta C_{\text{cgrad}}\), combine to make \(\Delta C_{\text{total}}\) lower than the historical scaling (\(\Delta \dot{C}_{\text{hs}}\)):

\[
\Delta C_{\text{total}} = \Delta \dot{C}_{\text{hs}} + \Delta C_{\text{chem}} + \Delta C_{\text{cgrad}}
\]  

(11)

Changes in ocean chemical capacity, \(\Delta C_{\text{chem}}\), are the change in \(C_{\text{ant}}\) in a one-dimensional model simulation with all effects, \(\Delta C_{\text{total}}\), minus the change in \(C_{\text{ant}}\) in a one-dimensional model simulation with constant chemical capacity, \(\Delta C_{\text{ccc}}\) (Table 1). The carbon gradient effect, \(\Delta C_{\text{cgrad}}\), is the difference between \(\Delta C_{\text{ccc}}\) and the time integral of \(F_{\text{ant}}\), \(\Delta \dot{C}_{\text{hs}}\). The carbon
gradient effect is due to interactions between the transport of $C_{\text{ant}}$ out of the surface ocean and the growth rate of $pCO_{2}^{\text{atm}}$. Because circulation is fixed in the one-dimensional ocean carbon cycle model, changes to the carbon gradient effect are not due to changes in circulation. In the results, we show that ocean $C_{\text{ant}}$ uptake has a low sensitivity to simulated changes in circulation from 1920-2080.

2.5 $C_{\text{ant}}$ Air-Sea Flux Decomposition

We use the one-dimensional model to identify the dominant controls on ocean $C_{\text{ant}}$ uptake, by decomposing the $C_{\text{ant}}$ air-sea flux into multiple terms. In order to perform this decomposition, surface ocean $pCO_{2}^{\text{ocn}}$ must be represented by a single value (Equation 10). For the ocean component of the ESM, defining an effective $pCO_{2}^{\text{ocn}}$, representing the entire surface ocean, is not straightforward due to the spatial variability of $pCO_{2}^{\text{ocn}}$. Therefore, this decomposition is only feasible in the one-dimensional ocean carbon cycle model.

The $C_{\text{ant}}$ air-sea flux can be considered to be a function of three indirectly related variables: $F_{\text{ant}}(pCO_{2}^{\text{atm}}, C_{\text{ant}}, T)$. Because these variables are indirectly related, we can decompose the total derivative of the $C_{\text{ant}}$ air-sea flux ($\frac{dF_{\text{ant}}}{dt}$) into its partial derivatives:

$$\frac{dF_{\text{ant}}}{dt} = \frac{\partial F_{\text{ant}}}{\partial pCO_{2}^{\text{atm}}} \frac{\partial pCO_{2}^{\text{atm}}}{\partial t} + \frac{\partial F_{\text{ant}}}{\partial pCO_{2}^{\text{ocn}}} \frac{\partial pCO_{2}^{\text{ocn}}}{\partial t}$$

The first term on the right hand side is the impact of the atmospheric CO$_2$ growth rate on the flux (atmosphere component) and the second term is the impact of the ocean CO$_2$ growth rate on the flux (ocean component). In Equation 12, $\frac{\partial F_{\text{ant}}}{\partial pCO_{2}^{\text{atm}}} = 0$, and is constant, thus variations in $F_{\text{ant}}$ are solely due to variations in the atmospheric CO$_2$ growth rate and the ocean CO$_2$ growth rate. The $pCO_{2}^{\text{ocn}}$ closely follows $pCO_{2}^{\text{atm}}$, and the sign of their growth rates is the same. When the atmospheric CO$_2$ growth rate is positive, the atmospheric CO$_2$ growth rate acts to enhance $F_{\text{ant}}$, and the ocean CO$_2$ growth rate acts to decrease $F_{\text{ant}}$ (Equation 10). The atmospheric CO$_2$ growth rate is prescribed, and cannot be separated further in this framework, while the ocean component is expanded into the following terms:

$$\frac{\partial pCO_{2}^{\text{ocn}}}{\partial t} = \frac{\partial \delta F}{\partial \delta C_{\text{ant}}} + \frac{\partial T}{\partial \delta C_{\text{ant}}}$$

The two ocean terms are the product of the change in $C_{\text{ant}}$ times the buffer factor, and the change in temperature times the sensitivity of $pCO_{2}^{\text{ocn}}$ to warming. Substituting Equation 8 we arrive at three terms controlling the ocean component:

$$\frac{\partial pCO_{2}^{\text{ocn}}}{\partial t} = F_{\text{ant}} \frac{\partial C_{\text{ant}}}{\partial t} + K_{\text{eff}} \frac{\partial C_{\text{ant}}}{\partial z} + \frac{\partial T}{\partial \delta C_{\text{ant}}}$$

$$\frac{\partial pCO_{2}^{\text{ocn}}}{\partial t} = F_{\text{ant}} \frac{\partial C_{\text{ant}}}{\partial t} + K_{\text{eff}} \frac{\partial C_{\text{ant}}}{\partial z} + \frac{\partial T}{\partial \delta C_{\text{ant}}}$$

$$\frac{\partial pCO_{2}^{\text{ocn}}}{\partial t} = F_{\text{ant}} \frac{\partial C_{\text{ant}}}{\partial t} + K_{\text{eff}} \frac{\partial C_{\text{ant}}}{\partial z} + \frac{\partial T}{\partial \delta C_{\text{ant}}}$$
We refer to the three terms on the right hand side, from left to right, as the impact of the air-sea flux on \( pCO_{2}^{\text{ocn}} \), the impact of ocean circulation on \( pCO_{2}^{\text{ocn}} \), and the impact of warming on \( pCO_{2}^{\text{ocn}} \). Because the impact of warming is small, the ocean CO\(_2\) growth rate is a balance between the impact of the air-sea flux on \( pCO_{2}^{\text{ocn}} \) and the impact of ocean circulation on \( pCO_{2}^{\text{ocn}} \). The sign of \( F_{\text{ant}} \) is always positive in all scenarios, thus the impact of the air-sea flux always acts to increase \( pCO_{2}^{\text{ocn}} \). The sign of the vertical gradient (\( \frac{\partial C_{\text{ant}}}{\partial z} \)) is always negative, thus the impact of ocean circulation always acts to decrease \( pCO_{2}^{\text{ocn}} \). In the \( \Delta C_{\text{total}} \) experiment (Table 1), \( F_{\text{ant}} \), the vertical \( C_{\text{ant}} \) gradient, the buffer factor, and sensitivity to warming, are freely evolving (Equation 14). In the \( \Delta C_{\text{ccc}} \) experiment, the buffer factor is fixed at preindustrial values. The \( \Delta C_{\text{hs}} \) experiment is equivalent to a constant buffer factor, a warming sensitivity of 0, and, as shown in the results, setting the \( C_{\text{ant}} \) profile to that predicted by historical scaling. The vertical profile alters \( pCO_{2}^{\text{ocn}} \) through the vertical gradient (Equation 14). In the results we will use Equation 14 to illustrate how changes to the impact of ocean circulation result in reduced efficiency of ocean \( C_{\text{ant}} \) uptake in response to emission mitigation.

3 Results

In the following sections, we use ocean output from the ocean component of the ESM to calculate the efficiency of ocean \( C_{\text{ant}} \) uptake (Results 3.1) and determine if the evolution of \( C_{\text{ant}} \) concentration along meridional sections is consistent with historical scaling (Results 3.2). We then use the one-dimensional model to attribute changes in the efficiency of \( C_{\text{ant}} \) uptake to physical and/or chemical mechanisms (Results 3.3). Changes to the air-sea flux arising from changes to the atmospheric CO\(_2\) growth rate and ocean CO\(_2\) growth rate are also diagnosed (Results 3.4). Our analysis includes scenarios with aggressive emission mitigation (1.5°C), intermediate emission mitigation (RCP4.5), and no emission mitigation (RCP8.5). See Figure 2c for the \( pCO_{\text{atm}}^{2} \) forcing for these scenarios.

3.1 Projected Spatial Redistribution of the \( C_{\text{ant}} \) Air-Sea Flux

Using output from the ocean component of the ESM, we diagnose \( C_{\text{ant}} \) air-sea flux for three future scenarios: 1.5°C, RCP4.5, RCP8.5. Here we focus on the projected spatial distribution of \( C_{\text{ant}} \) air-sea flux from 2020-2080.

In the 1.5°C scenario the spatial pattern of the \( C_{\text{ant}} \) air-sea flux changes significantly from 2020-2080. While most of the ocean is a sink in 2020, in 2050 and 2080 there are large regions of \( C_{\text{ant}} \) outgassing (Figure 1, bottom row). Most pronounced is the emergence of \( C_{\text{ant}} \) outgassing in the equatorial Pacific. The outcrop region of Sub-Antarctic Mode Water (SAMW) at about 50°S also experiences outgassing by 2080. In 2020, the Kuroshio and subpolar North Atlantic are some of the most intense sinks of \( C_{\text{ant}} \), but by 2080, these regions are sources. The Equatorial Pacific, SAMW outcrop region, Kuroshio, and subpolar North Atlantic are broad regions that are sources of \( C_{\text{ant}} \) in 2080 under the 1.5°C scenario. Contrastingly, Southern Ocean \( C_{\text{ant}} \) uptake persists throughout the simulation.

In the RCP4.5 scenario, changes to the spatial pattern lie somewhere between RCP8.5 and the 1.5°C scenario. Equatorial Pacific outgassing of \( C_{\text{ant}} \) grows over time, but is less widespread and intense than in the 1.5°C scenario (Figure 1, middle row). \( C_{\text{ant}} \) air-sea flux intensity decreases over time for the subpolar and mid-latitude Atlantic and Kuroshio region. Beyond
the equatorial Pacific, the spatial pattern of \( C_{\text{ant}} \) air-sea flux is similar to the RCP8.5 scenario, but the amplitude of uptake is reduced.

Relative to the scenarios with emission mitigation (1.5°C and RCP4.5) the RCP8.5 scenario features a consistent spatial pattern of the \( C_{\text{ant}} \) air-sea flux (Figure 1, top row). The primary change over time is an amplification of magnitude, with the highest flux intensity occurring in 2080.

Ocean \( C_{\text{ant}} \) uptake is greatest in RCP8.5, and is the lowest in 1.5°C (Figure 2a). In all scenarios ocean \( C_{\text{ant}} \) inventory increases throughout the period (Figure 2d). In the RCP4.5 scenario, \( C_{\text{ant}} \) air sea flux peaks in 2050, and then gradually declines. In the 1.5°C scenario, ocean \( C_{\text{ant}} \) uptake peaks in 2020, and is almost zero by 2080. In the RCP4.5 scenario ocean \( C_{\text{ant}} \) uptake initially increases and then returns to 2020 values by 2080.

Extrapolation of the ocean \( C_{\text{ant}} \) uptake based on the historical scaling (\( \hat{F}_{\text{ant}} \)) is dependent solely on \( pCO_{2}^{\text{atm}} \). Lower \( pCO_{2}^{\text{atm}} \) results in a lower estimate of ocean \( C_{\text{ant}} \) uptake, and higher \( pCO_{2}^{\text{atm}} \) results in greater uptake. In all scenarios, simulated air-sea \( C_{\text{ant}} \) uptake is far less than \( \hat{F}_{\text{ant}} \) (Figure 2a). Reduced uptake relative to \( \hat{F}_{\text{ant}} \) indicates that in the future, ocean \( C_{\text{ant}} \) uptake will be less efficient (Figure 2b). Efficiency is the ratio of the solid lines (\( F_{\text{ant}} \)) to the respective dashed lines (\( \hat{F}_{\text{ant}} \)) in Figure 2a, with this ratio plotted in Figure 2b. Efficiency (Figure 2b) remains greater than 90% from 1990 through 2010, but then declines under all future scenarios, with greater efficiency declines as emission mitigation increases.
The efficiency decrease is linear in RCP8.5 and RCP4.5, but exponential in the 1.5°C scenario. The 1.5°C scenario is the only scenario with negative $pCO_2^{atm}$ growth rates, which substantially modifies the ocean carbon cycle response, as shown below.

### 3.2 Projected Changes in the Ocean Interior

Here we analyze the evolution of the $C_{ant}$ vertical gradient by applying the historical scaling to the vertical profile of $C_{ant}$ (Equation 6). A weakening of the vertical gradient of $C_{ant}$ would reduce the ability of physical removal of $C_{ant}$ to maintain the $C_{ant}$ air-sea flux ($F_{ant}$, Equation 14). Thus, via the vertical $C_{ant}$ gradient, interior $C_{ant}$ can alter the air-sea flux. Deviations of globally averaged $C_{ant}$ profiles ($C_{ant}(z)$) from the $C_{ant}$ historical scaling ($\ast C_{ant}(z)$) are defined as $C_{ant}(z) - \ast C_{ant}(z)$. Wherever $C_{ant}(z) > \ast C_{ant}(z)$, more carbon is stored at that location than predicted by the $C_{ant}$ historical scaling (Equation 6) and the deviation is positive. Assuming ocean circulation remains constant, if deviations are less at the surface relative to the interior, the vertical gradient would be weaker than expected by the historical scaling, and thus ocean $C_{ant}$ uptake would be less efficient. Therefore the historical scaling may be used to identify how the pattern of changing interior $C_{ant}(z)$ deviates from the historical scaling (Figure 3) and thus reduces the efficiency of the $C_{ant}$ air-sea flux (Figure 2b). With more rapid emission mitigation, globally average profiles reveal a pattern of increasingly positive deviations from the historical scaling at depth (Figure 3).

In the RCP8.5 and RCP4.5 scenarios, $C_{ant}(z)$ increases from 2020-2080 at all depths, but at the surface, $C_{ant}(z)$ increase is less than $\ast C_{ant}(z)$ increase (Figure 3a). In the RCP4.5 scenario, the $C_{ant}$ at depth is greater than $\ast C_{ant}(z)$, while in the RCP8.5 scenario it is less than $\ast C_{ant}(z)$ (Figure 3b). In both the RCP8.5 and RCP4.5 scenarios, the increase in $C_{ant}$ is surface...
intensified, which enhances the vertical gradient. The enhanced vertical gradient allows for increased vertical diffusion of \( C_{ant} \), and thus increased ocean \( C_{ant} \) uptake. However, in RCP4.5 and RCP8.5 the enhancement of the vertical gradient is not as strong as the historical scaling would suggest (Figure 3b). In the 1.5°C scenario, the largest change in \( C_{ant}(z) \) is at depth, and at the surface \( C_{ant} \) decreases. This results in a much weaker vertical gradient, weaker vertical diffusion, and thus a reduced ocean \( C_{ant} \) uptake. The surface loss of \( C_{ant} \) is a short term response to declines in \( pCO_{2}^{atm} \) that began in 2036, while the increase in \( C_{ant}(z) \) at depth is from the long-term increase in \( pCO_{2}^{atm} \) relative to preindustrial times (Figure 3c).

The signals found in \( C_{ant}(z) \) are found throughout the ocean (Figure 4). In the RCP8.5 scenario (Figure 4, top row), the surface layer exhibits the strongest negative deviation, but there is no positive deviation in the interior. The negative deviation is seen in deep waters between 25°N and 60°N, and also in the bowls of the northern and southern subtropical gyres. The negative deviation grows from 2020-2080, and appears to propagate into the ocean interior with NADW. The historical scaling alone cannot identify whether buffering or solubility is the driver of lower \( C_{ant}(z) \) than \( C_{ant}(z) \) in the interior.

In the RCP4.5 scenario, the surface layer exhibits a growing negative deviation (Figure 4, middle). The negative surface deviation spans from the southern to the northern end of the zonal mean section. In the interior, however, there is a growing positive deviation. The magnitude and growth rate of the interior positive deviation is less than in the 1.5°C scenario. The positive deviation is due to the lagged response of the ocean interior to the intermediate rate of emission mitigation in RCP4.5, in which \( pCO_{2}^{atm} \) slowly plateaus (Figure 2c).

The 1.5°C scenario features the largest positive \( C_{ant} \) deviations from the historical scaling (Figure 4, bottom row). The positive deviation is found throughout the thermocline. These are waters that outcrop in the equatorial Pacific and mid to high latitudes, consistent with these regions being a source of \( C_{ant} \) by 2080 (Figure 1). In the next section, we will evaluate the relative role of buffering for all scenarios.

### 3.3 Drivers of Simulated Changes in Efficiency

We utilize projections of ocean \( C_{ant} \) uptake from the one-dimensional model to provide mechanistic understanding of the changes in ocean carbon uptake efficiency simulated by the full model. Changes in ocean \( C_{ant} \) are examined to determine what drives projected changes in efficiency. These changes that are quantified with the one-dimensional model are separated into the carbon gradient effect, \( \Delta C_{cgrad} \) (Table 1), and effects related to the ocean’s chemical capacity to absorb carbon, \( \Delta C_{chem} \) (Table 1). The one-dimensional model can accurately recreate the change in \( C_{ant} \) inventory for all scenarios (Figure 2d), indicating that the assumption of constant circulation and parameterized chemistry are reasonable through 2080. For the RCP8.5 scenario, the difference between the one-dimensional model cumulative flux and ocean component model is the largest. The cumulative error is 4 Pg \( C_{ant} \), only 3% of the 2080 cumulative flux, again indicating that the one-dimensional model is a useful diagnostic tool for quantifying changes in the efficiency of ocean uptake.

Over the historical forcing period (1920-2006) \( \Delta C_{cgrad} \) drives \( \Delta C_{total} \) to be slightly lower than the historical scaling (Figure 5). Because there is no internal ocean variability in the one-dimensional model (constant ocean circulation), \( \Delta C_{cgrad} \) in this period is due to only the integrated effects of short term variations in the growth rate of the \( pCO_{2}^{atm} \) forcing, which drive a slight reduction in total uptake (McKinley et al., 2020).
Figure 3. Output from the ocean component of the CESM of the global mean $C_{ant}$ profiles ($C_{ant}(z)$) (orange, solid), and profiles of $\tilde{C}_{ant}(z)$ (gray, dashed), for the (a) RCP8.5 scenario, (b) RCP4.5 scenario, and (c) 1.5°C scenario. The shaded region between the dashed and solid lines indicates the deviation from the historical scaling. Light lines are for 2020 and dark lines are for 2080. The shaded region between the lines is shown for zonal mean sections in Figure 4.
Figure 4. Ocean component model output of the global zonal mean deviation of $C_{ant}$ concentration (mmol m$^{-3}$) from the historical scaling of $C_{ant}$ ($C_{ant} - C_{ant}^*$. Rows and columns same as Figure 1. Positive regions indicate faster carbon accumulation than historical scaling, negative regions indicate slower accumulation. Contour lines are surfaces (kg m$^{-3}$).

In RCP8.5, the ocean cumulatively absorbs 393 Pg $C_{ant}$ (Figure 5) by 2080, approximately 2.5 times the present-day $C_{ant}$ inventory (160-166 Pg $C_{ant}$; Devries, 2014). In RCP8.5, the historical scaling tracks the simulation with constant chemical capacity (Figure 5). This indicates that if ocean chemical capacity remains constant, the ocean would absorb an additional 158 Pg $C_{ant}$. The positive $\Delta C_{cgrad}$ is attributable to fitting CESM $C_{ant}$ uptake to a not-quite exponential $pCO_{2}^{atm}$ in the historical period. Changes in ocean $C_{ant}$ uptake due to warming were calculated, but are not shown as warming effects are small relative to $\Delta C_{chem}$, making up <5% of the total efficiency decline. This small contribution is consistent with the change due to warming calculated in studies of climate-carbon feedbacks (Randerson et al., 2015, Schwinger et al., 2018).

In RCP4.5, the ocean absorbs 307 Pg $C_{ant}$ (Figure 5) by 2080. Cumulative uptake predicted by the historical scaling tracks the constant chemical capacity simulation until around 2040 (Figure 5). After 2040, the historical scaling and the constant chemical capacity simulation diverge, indicating the increasing carbon gradient effect is related to the decreasing growth rate of the $pCO_{2}^{atm}$ forcing in this time frame. The $\Delta C_{cgrad}$ effect amounts to -52 Pg $C_{ant}$ in 2080, however, the $\Delta C_{chem}$ effect is stronger, amounting to -84 Pg $C_{ant}$ by 2080.

In the 1.5°C scenario, the ocean absorbs 213 Pg $C_{ant}$ (Figure 5) by 2080. The $\Delta C_{chem}$ effect is the weakest in this scenario, -37 Pg C in 2080. The weak $\Delta C_{chem}$ effect is consistent with this scenario taking up the least $C_{ant}$ because chemical capacity decreases as $C_{ant}$ uptake increases. $\Delta C_{cgrad}$ is the dominant change in this scenario, accounting for -90 Pg $C_{ant}$. The strongly reduced vertical gradient of $C_{ant}$ (Figure 3,4) results in reduced vertical $C_{ant}$ removal (Equation 8).
3.4 Decomposition of the \( C_{\text{ant}} \) Air-Sea Flux

Here we diagnose the mechanisms controlling the \( C_{\text{ant}} \) air-sea flux \( (F_{\text{ant}}) \) in the one-dimensional model \( \Delta C_{\text{total}} \) simulations. The one-dimensional model has been tuned so the \( \Delta C_{\text{total}} \) simulation matches the CESM (Figure 2d). The one-dimensional form of the model allows for the decomposition of \( C_{\text{ant}} \) air-sea flux (Methods 2.5).

Air-sea flux \( (F_{\text{ant}}) \) in the one-dimensional model is controlled by large opposing components (Figure 6), the atmosphere component, driven by the atmosphere \( \text{CO}_2 \) growth rate, and the ocean component, driven by the ocean \( \text{CO}_2 \) growth rate. Because the air-sea flux is adding \( C_{\text{ant}} \) to the surface in all simulations, the impact of the air-sea flux on \( p\text{CO}_2^{\text{ocn}} \) is to increase \( p\text{CO}_2^{\text{ocn}} \). while the impact of ocean circulation is to decrease \( p\text{CO}_2^{\text{ocn}} \) because of the downward vertical \( C_{\text{ant}} \) gradient.

Figure 5. Cumulative ocean \( C_{\text{ant}} \) uptake (Pg C) for the historical period and the three scenarios in the one-dimensional model. The cyan line is the extrapolation based on the historical scaling. The dark blue line is the one-dimensional model simulation of constant chemical capacity (PI buffer factor) with variable solubility. The black line is the one-dimensional model simulation that includes all effects (variable chemical capacity and variable solubility). Light green shading represents decreases in uptake related to the carbon gradient effect (\( \Delta C_{\text{grad}} \)), teal shading represents decreases in uptake related to chemical capacity (\( \Delta C_{\text{chem}} \)). Negative indicates loss of ocean carbon.

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(Equation 14). Thus the vertical $C_{ant}$ gradient plays an important role in sustaining $C_{ant}$ removal by ocean circulation, and thus slowing the ocean CO$_2$ growth rate.

Figure 6. (a-c) Total $C_{ant}$ air-sea flux (green line; Pg $C_{ant}$ yr$^{-1}$) and its ocean (blue line; Pg $C_{ant}$ yr$^{-1}$) and atmosphere (orange line; Pg $C_{ant}$ yr$^{-1}$) components. The values for the air-sea flux and its components are calculated by integrating the $C_{ant}$ air-sea flux growth rate ($\frac{dF_{ant}}{dt}$, units of Pg $C_{ant}$ yr$^{-2}$), and the ocean component ($\frac{\partial pCO_{ocn}}{\partial t}$ $\frac{\partial F_{ant}}{\partial pCO_{ocn}}$, units of Pg $C_{ant}$ yr$^{-2}$) and atmosphere component ($\frac{\partial pCO_{atm}}{\partial t}$ $\frac{\partial F_{ant}}{\partial pCO_{atm}}$, units of Pg $C_{ant}$ yr$^{-2}$), from left to right, starting at the beginning of the simulation in 1850. The annual air-sea flux is shown on a reduced axis in (a-c). In (d-f) the same terms (Equation 12) are integrated starting in 2040 (vertical dashed line in (a-c)) and on a log scale to highlight the changing roles of the ocean and atmosphere components under rapid mitigation scenarios. Components are non-zero at 2040 in (d-f) because of the air-sea $pCO_2$ difference in 2040.

In the RCP8.5 scenario, the atmosphere component acts to enhance $F_{ant}$, and the ocean component acts to reduce the $F_{ant}$ (Figure 6a-b). The actual $F_{ant}$ (Figure 6, green) is a small residual of these tendencies. Increasing $pCO_{atm}^2$ acts to increase the air-sea $pCO_2$ difference, while increasing $pCO_{ocn}^2$ acts to decrease $F_{ant}$ (Equation 12). If $pCO_{ocn}^2$, increased only very slightly, such as the hypothetical scenario where the ocean is well mixed from surface to deep, ocean $C_{ant}$ uptake would be the magnitude of the atmospheric component (Figure 6a-b). In fact, the ocean component is also subject to a balance between two large terms. The increase in $pCO_{ocn}^2$ is mitigated by ocean circulation, thus ocean circulation enhances $F_{ant}$ (Equation 14). $F_{ant}$ increases $pCO_{ocn}^2$ and, which in turn reduces $F_{ant}$ (Equation 14).
In the RCP4.5 scenario, the atmosphere and ocean components act similarly to the RCP8.5 scenario, with the atmosphere component enhancing \( F_{\text{ant}} \), while the ocean component acts to reduce \( F_{\text{ant}} \) (Figure 6c-d). However, due to increased emission mitigation, \( pCO_{2\text{atm}} \) plateaus (Figure 2c), and in 2040 \( F_{\text{ant}} \) begins to decline. The ocean component is acting to decrease \( F_{\text{ant}} \) and by 2040 overwhelms the atmosphere component. The \( pCO_{2\text{ocn}} \) increases slower than \( pCO_{2\text{atm}} \), which acts to enhance the flux early in the period, but the slower rate of change of \( pCO_{2\text{ocn}} \) cause \( F_{\text{ant}} \) to decline in the later period (Figure 6d).

In the 1.5°C scenario, initially the atmosphere component acts to enhance \( F_{\text{ant}} \), and the ocean component acts to reduce \( F_{\text{ant}} \). However, by 2040, the roles of \( F_{\text{ant}} \) components have switched (Figure 6a-b). Due to rapid emission mitigation, \( pCO_{2\text{atm}} \) plateaus, and then begins to decrease slightly, acting to reduce \( F_{\text{ant}} \) for the first time since the preindustrial era. After 2040, the ocean component acts to enhance \( F_{\text{ant}} \) (Figure 6f). If efficiency were constant, it follows that the ratio of atmosphere to ocean components is constant and the change in \( pCO_{2\text{atm}} \) for a a given change in \( pCO_{2\text{atm}} \) would also be constant. From 2013 to 2036, the year of \( pCO_{2\text{atm}} \) begins to decline (Figure 2c), \( pCO_{2\text{atm}} \) increases 39 ppm, and \( pCO_{2\text{ocn}} \) increases 39 ppm and \( \frac{\Delta pCO_{2\text{atm}}}{\Delta pCO_{2\text{ocn}}} = 1.00 \). From 2036 to 2080, \( pCO_{2\text{atm}} \) also declines 39 ppm and \( pCO_{2\text{ocn}} \) declines 37 ppm, thus \( \frac{\Delta pCO_{2\text{atm}}}{\Delta pCO_{2\text{ocn}}} = 0.95 \). From 2040 to 2080, the \( C_{\text{ant}}(z) \) has weakened relative to the historical scaling (Figure 4), thus also weakening the impact of ocean circulation on \( pCO_{2\text{atm}} \) (Equation 14). The weakening of the vertical \( C_{\text{ant}} \) gradient therefore plays a significant role in reducing \( \frac{\Delta pCO_{2\text{atm}}}{\Delta pCO_{2\text{ocn}}} \), and thus efficiency, on the downward \( pCO_{2\text{atm}} \) trajectory.

4 Discussion

4.1 Drivers of Future Efficiency Declines

Ocean carbon uptake will decline as a result of emission mitigation. We also show that the efficiency of ocean carbon uptake, i.e. how closely ocean carbon uptake follows the observed proportionality between uptake and atmospheric \( CO_2 \), is also reduced as mitigation becomes more rapid, consistent with the results of Raupach et al. (2014). Under exponentially increasing \( pCO_{2\text{atm}} \) (RCP8.5), reductions in efficiency of ocean \( C_{\text{ant}} \) uptake are almost entirely due to reduced buffer capacity. We find that an exponentially increasing \( pCO_{2\text{atm}} \) allows for the entire vertical \( C_{\text{ant}} \) profile to behave as a function of \( pCO_{2\text{atm}} \). Because ocean \( C_{\text{ant}} \) uptake is well characterized by one-dimensional physics, under constant chemistry and ocean circulation, if the vertical profile behaves as a function of \( pCO_{2\text{atm}} \) then air-sea flux will also behave as function of \( pCO_{2\text{atm}} \) (Equation 4, Equation 8). With rapid mitigation, the vertical \( C_{\text{ant}} \) profile, which is set by the integrated effects of past \( C_{\text{ant}} \) accumulation at depth, does not change to immediately to adjust to the trajectory of \( pCO_{2\text{atm}} \). We find that in a scenario featuring rapid emission mitigation (1.5°C), the \( C_{\text{ant}} \) concentration change from 2020-2080 is greatest in the thermocline, a behavior that has been identified in other simulations of rapid emission mitigation (Tokarska et al., 2019). The past \( C_{\text{ant}} \) accumulation at depth weakens the vertical \( C_{\text{ant}} \) gradient compared to a vertical \( C_{\text{ant}} \) profile that reaching the same \( pCO_{2\text{atm}} \) under exponential \( pCO_{2\text{atm}} \) increase, constant ocean chemistry, and constant circulation (Figure 3).

The air-sea flux is a balance between the atmospheric \( CO_2 \) growth rate and the ocean \( CO_2 \) growth rate (Equation 12), and in all scenarios the atmospheric \( CO_2 \) growth rate dominates the balance. The positive atmospheric \( CO_2 \) growth rates throughout the RCP8.5 and RCP4.5 scenarios acts to enhance the air-sea flux. In the 1.5°C scenario, ocean \( pCO_{2\text{atm}} \) declines after 2036,
and the negative atmospheric CO₂ growth rate acts to decrease the air-sea flux, while the negative ocean CO₂ growth rate acts to enhance the air-sea flux. The negative growth rates of the 1.5°C scenario occur in the only scenario where efficiency declines exponentially.

The dominant mechanisms governing the decline in efficiency are different in each scenario, due to the differing degrees of emission mitigation. Internal ocean mechanisms (reduced chemical capacity) dominate the reduction of efficiency in the RCP8.5 scenario, and external mechanisms (increasing carbon gradient effect) dominate the reduction of efficiency in the 1.5°C scenario. For the ocean, warming effects have a small impact relative to the carbon gradient and chemical capacity effects.

The growing carbon gradient effect in the 1.5°C scenario is due to a weakening of the \( C_{\text{ant}} \) vertical gradient, thus a declining rate at which \( C_{\text{ant}} \) mixes and diffuses into the ocean interior. The magnitude of the \( C_{\text{ant}} \) air-sea flux is limited by the rate of surface \( C_{\text{ant}} \) removal (Graven et al., 2012), thus slower removal results in a reduced growth rate of ocean \( C_{\text{ant}} \) uptake. The vertical gradient is weaker in scenarios with slower than exponential \( pCO_{2}^{\text{atm}} \) increase, compared to the vertical gradient at the same \( pCO_{2}^{\text{atm}} \) concentration in an exponentially increasing \( pCO_{2}^{\text{atm}} \) scenario, because \( C_{\text{ant}} \) concentration is enhanced at depth relative to exponential scenarios (Figure 3,4). The \( C_{\text{ant}} \) concentration is elevated at depth because it takes longer for the slower than exponential scenarios to reach the same \( pCO_{2}^{\text{atm}} \), allowing more cumulative \( C_{\text{ant}} \) transfer to deeper waters. The waters at depth effectively push back on the changes occurring at the surface due to changing \( pCO_{2}^{\text{atm}} \), which is qualitatively similar to how back-pressure in a pipe slows the flow of fluid through the pipe. It is the \( C_{\text{ant}} \) at depth that is providing the "back-pressure", resisting the flow of \( C_{\text{ant}} \) into the interior. The faster emissions are mitigated, the more evident the back-pressure exerted by ocean interior \( C_{\text{ant}} \) becomes (Figure 3,4,5,6). However, delaying emission mitigation would act to increase the total back-pressure effect that would eventually occur. If the RCP8.5 scenario is followed into the 22nd century, future emissions would be flat from 2100 to 2150 and then decline dramatically (van Vuuren et al., 2011). As \( pCO_{2}^{\text{atm}} \) growth slows in response to the declining rate of emissions, the back-pressure effect from the ocean will appear, but at a greater magnitude due to the much greater load of \( C_{\text{ant}} \) in the interior ocean. Therefore, climate simulations extending beyond 2100 are needed to quantify the back-pressure effect in high emission scenarios.

With aggressive emission mitigation, regional patterns of the \( C_{\text{ant}} \) air-sea flux shift, with implications for regional carbon cycle monitoring (Peters et al., 2017). The surface waters of regions that trend towards \( C_{\text{ant}} \) outgassing under emission mitigation (Figure 1; bottom row) are renewed by advection with waters that are much older than the waters that renew the waters of the subtropics (Toyama et al., 2017). As emissions are mitigated from 2020-2080, there is a positive change in \( C_{\text{ant}} \) concentration in the ocean interior, with this back-pressure effect being most pronounced at ~400m (Figure 3). Regionally, advective fluxes are important drivers of \( C_{\text{ant}} \) reemergence (Bopp et al., 2015), thus the regional impacts of ocean circulation on \( pCO_{2}^{\text{ocn}} \) (Equation 14) would include the effects of advective fluxes in addition to mixing/diffusive fluxes (which dominate globally). In the outgassing regions of Figure 1 (bottom row), advective fluxes bring waters with an increasing load of \( C_{\text{ant}} \) to the surface, thus reemergence is increasing, acting to increase \( pCO_{2}^{\text{ocn}} \). The increase in reemergence ultimately overwhelms the weakening downwards diffusive \( C_{\text{ant}} \) flux that acts to decrease \( pCO_{2}^{\text{ocn}} \). Therefore, advection of \( C_{\text{ant}} \) is driving the \( C_{\text{ant}} \) air-sea flux further towards outgassing in the equatorial Pacific, supbolar and mid-latitude North Atlantic, SAMW outcrop region, and the
Kuroshio (Toyama et al., 2017). The Circumpolar Deep Water (CDW) that is upwelled into the surface of the Southern Ocean south of 50°S, is old relative to the subtropics, but it is uncontaminated by $C_{\text{ant}}$. Below $\sim400$m the magnitude of the back-pressure decreases, therefore upwelling of CDW does not result in an increasing load of $C_{\text{ant}}$ being brought to the surface. Southern Ocean $C_{\text{ant}}$ uptake persists because the positive $C_{\text{ant}}$ tendency is absent in the upwelling watermass.

The back-pressure from $C_{\text{ant}}$ at depth is an unavoidable consequence of emission mitigation. While more efficient ocean $C_{\text{ant}}$ uptake is desirable when drawing down $pCO_{2\text{atm}}$, in fact, peak sink efficiency occurs under exponential growth of $pCO_{2\text{atm}}$. How long the ocean will remain a net sink depends on the strength of the back-pressure effect, which depends on the strength of surface ocean $C_{\text{ant}}$ removal. If the back-pressure effect is stronger, due to more vigorous ocean circulation transfer of $C_{\text{ant}}$ to depth in the years prior to mitigation, the sink will disappear at a faster rate. With rapid mitigation, the ocean $C_{\text{ant}}$ uptake peaks in approximately 2030 and nearly disappears by 2080. Despite the decline in the efficiency of ocean $C_{\text{ant}}$ uptake under rapid mitigation, ESMs indicate that the ocean becomes the primary $C_{\text{ant}}$ sink in scenarios with aggressive mitigation (Jones et al., 2013) and without mitigation (Randerson et al., 2015) because the land uptake declines more rapidly than ocean uptake (Zickfeld et al., 2016). The ocean will ultimately remove most atmospheric $C_{\text{ant}}$ over tens of thousands of years (Archer, 2005, Archer et al., 2009).

### 4.2 Validity of the One-Dimensional Model Representation of Ocean Physics

Our one-dimensional ocean carbon cycle model represents multiple physical removal process as a single diffusive process (Equation 8). Parameterizing these various processes in this manner requires defining an effective vertical diffusivity of the ocean, $K_{z,\text{eff}}$ and better observational estimates of ocean mixing are required to in order to recreate the effective diffusivity of the actual ocean. In the three-dimensional ocean models used in ESMs, up to $\sim30\%$ of simulated $K_{z,\text{eff}}$ is attributable to isopycnal eddy mixing (Gnanadesikan et al., 2015). Varying a three-dimensional ocean model’s isopycnal eddy diffusivity within the range of typical model values results in a 92 Pg C range of cumulative ocean $C_{\text{ant}}$ uptake under instantaneous CO$_2$ doubling (Gnanadesikan et al., 2015), thus the sensitivity of ocean $C_{\text{ant}}$ uptake to isopycnal eddy mixing is much larger than the sensitivity to changing ocean circulation. Models with spatially varying isopycnal eddy diffusivities, such as the NCAR CESM and GFDL ESM2G, have parameterizations that produce a range of diffusivities from $\sim300$ m$^2$s$^{-1}$ in gyres, to $\sim1500$ m$^2$s$^{-1}$ in boundary currents (Dunne et al., 2012; Danabasoglu et al., 2011). Observational estimates of isopycnal eddy diffusivity from tracers (Ledwell et al., 1998) and satellite altimetry (Abernathey and Marshall, 2013) are uncertain, but consistently suggest that real world eddy diffusivities could be much higher, with ranges from $\sim1000$ m$^2$s$^{-1}$ in gyres, to $\sim10,000$ m$^2$s$^{-1}$ in boundary currents.

While the mean state of ocean circulation is most important over the never 60 years, as warming increases, the magnitude climate-carbon feedbacks increase, such as changes to ocean circulation driving changes in ocean carbon uptake (Randerson et al., 2015), which is not represented by our one-dimensional ocean carbon cycle model. The small effect of changing ocean circulation in our simulation is likely because changes due to declines in AMOC are not yet evident by 2080 (Sarmiento and LeQuéré, 1996; Randerson et al., 2015). While assuming that ocean climate-carbon feedbacks are small prior to 2080 is consistent with the behavior of the CESM (Randerson et al., 2015), this may not be hold true for the Earth System itself.
The uncertainties associated with the timing and magnitude of climate-carbon feedbacks can be avoided by mitigating climate change (Randerson et al., 2015).

### 4.3 Identification of the Impacts of Internal Ocean Variability using the Historical Scaling

Given the direct relationship between the efficiency of ocean $C_{\text{ant}}$ uptake and the deviations from the historical scaling, we suggest that future work quantify the impact of external mechanisms on observed interannual variability of ocean carbon uptake. The historical scaling relies on the assumption of fixed sink efficiency due to exponential growth of $pCO_2^{atm}$. While most of the industrial era is consistent with an exponential CO$_2$ growth, variability in emissions drives variability in the atmospheric growth rate that then results in decadal variability in the ocean carbon sink (McKinley et al., 2020). If the historical scaling is used to identify changes in observations of $C_{\text{ant}}$ concentration (Gruber et al., 2019a), and the atmospheric growth rate has recently slowed, changes due to internal variability are mixed with signals related to the carbon gradient effect, and the changes due to internal variability (Gruber et al., 2019a) would be overestimated. We also emphasize that in a future with emission mitigation, deviations from historical scaling will not be driven by changes due to internal ocean variability alone, given the dynamic response of the ocean to changes in $pCO_2^{atm}$.

### 5 Conclusion

Atmospheric CO$_2$ has grown exponentially over the industrial era, and so has ocean $C_{\text{ant}}$ concentration at all depths (DeVries, 2014; Gruber et al., 2019). Under the exponential forcing regime, ocean $C_{\text{ant}}$ uptake also grows exponentially and, over the historical era, maintains high efficiency of ocean $C_{\text{ant}}$ uptake as we have defined it here. In future scenarios, regardless of whether countries mitigate emissions, efficiency of ocean $C_{\text{ant}}$ uptake will decline. However the mechanisms differ depending on the degree of mitigation. In the RCP8.5 scenario, a scenario with no emission mitigation, reduced buffer capacity explains nearly all of the loss in efficiency of ocean $C_{\text{ant}}$ uptake by 2080, 158 Pg C. In the case of scenarios with emission mitigation, such as RCP4.5 and the 1.5°C scenario, the loss of efficiency is more due to the carbon gradient effect. The carbon gradient effect explains 38% of efficiency loss in RCP4.5 scenario, and 71% of the efficiency loss in the 1.5°C scenario. The carbon gradient effect increases with time in all scenarios with emission mitigation, and the equatorial Pacific becomes a prominent source of $C_{\text{ant}}$ in both cases. Under aggressive mitigation (1.5°C) this carbon gradient effect will result in large regions of ocean becoming a source by 2080, with the exception of the Southern Ocean.

Changes in the vertical $C_{\text{ant}}$ concentration gradient are responsible for this carbon gradient effect. Under exponential $pCO_2^{atm}$ growth, with constant chemical capacity and constant solubility, the vertical $C_{\text{ant}}$ gradient behaves as a function of $pCO_2^{atm}$, thus the $C_{\text{ant}}$ air-sea flux behaves as though it is solely a function of $pCO_2^{atm}$, rather than a function of $pCO_2^{atm}$, $C_{\text{ant}}$, and temperature. When emissions are mitigated and the growth in $pCO_2^{atm}$ slows, the surface ocean can respond rapidly. However, the ocean interior $C_{\text{ant}}$ concentration response lags the surface response. Below 100m in the rapid mitigation scenario, $C_{\text{ant}}$ concentration increases from 2020-2080, while above 100m, the $C_{\text{ant}}$ concentration decreases, thus the downward $C_{\text{ant}}$ concentration gradient is greatly reduced. A reduced downward vertical gradient results in less effective downward dif-
fusion of $C_{\text{ant}}$. Ocean $C_{\text{ant}}$ uptake is limited by surface ocean $C_{\text{ant}}$ removal (Graven et al., 2012), thus this results in reduced uptake in the future, relative to ocean $C_{\text{ant}}$ uptake under the same $pCO_{2\text{atm}}$ concentration in the historical period. This reduction of the vertical gradient, an unavoidable result of emission mitigation, is the driver of efficiency declines.

Under emission mitigation, the carbon gradient effect results in a enhanced outgassing of $C_{\text{ant}}$ in the equatorial Pacific, and a transition from $C_{\text{ant}}$ uptake to $C_{\text{ant}}$ outgassing in the subpolar and mid-latitude North Atlantic, Kurshio, and SAMW outcrop region. These regions are also hotspots for reemergence (Bopp et al., 2015, Toyama et al., 2017). Reemergence of older watermasses, from depths where $C_{\text{ant}}$ continues to increase, drives a tendency towards outgassing in these regions. The waters of the subtropics are renewed with shallower waters, above where the continued $C_{\text{ant}}$ increase occurs in the ocean component of ESM, and the surface waters of the Southern Ocean are renewed with waters below the $C_{\text{ant}}$ increase. Thus, in subtropics and Southern Ocean, the air-sea $C_{\text{ant}}$ uptake continues.

Diffusive processes control the removal of $C_{\text{ant}}$ from the surface ocean, and because the diffusivity of the surface ocean is highly uncertain, it creates large uncertainties in $C_{\text{ant}}$ uptake (Gnanadesikan et al., 2015). Determining the effective vertical diffusivity of the upper ocean is essential to reducing uncertainty in future ocean $C_{\text{ant}}$ uptake, particularly under 21st century emission mitigation scenarios.

**Code and data availability.** The code used to run the one-dimensional model is provided by the authors in a GitHub repository (https://qoccm.readthedocs.io/en/latest/). Raw output from the coupled ocean model simulations can download from NCAR’s Earth System Grid (https://www.earthsystemgrid.org/).

**Appendix A: Physical Interpretation of the Impulse Response Function Based Model one-dimensional model**

Impulse response functions are a powerful tool in dynamical system analysis. With a response function, one can understand the response of a system to any pulse, as long as the response is linear. The response function used in our one-dimensional model has the same shape as the conceptual example in Figure A1. For the one-dimensional model case, the response function was derived by equilibrating HILDA to a doubling $CO_2$, and then tracking fraction of $C_{\text{ant}}$ that remained in the surface box (the mixed layer) (Joos et al. 1996).

In our conceptual example of the mixed layer response function, at $t = 0$ 100% of the tracer remains in the mixed layer, while 200 years later, only 10% of the tracer remains (Figure A1). This is a general example of response function that can be applied to any transient tracer, but the values in this example have been scaled so that it is most similar to the evolution $C_{\text{ant}}$.

Ocean circulation, with vertical diffusion playing the largest role for short lived transient tracers, sets the time that it takes to reach this value. The mixed layer pulse response function must be calculated for each transient tracer of interest because the spatial distribution of flux is tracer dependent, thus each tracer flux distribution uniquely samples the spatially variable vertical diffusivity of the ocean. The mixed layer pulse response function for $C_{\text{ant}}$ is determined by simulating the exposure of the surface ocean to a pulse of atmospheric $CO_2$. In order to use a single case as our pulse response function, the size of the
Figure A1. A conceptual example of the response function. This response function characterizes the decay of a transient tracer in the surface ocean.

pulse we give our model cannot affect the time it would take that pulse to reach 10%. In other words, the time it takes for any subsequent pulse to reach 10% in the mixed layer must also remain constant. In theory, the timescale could change as result of changes to ocean circulation, and as seen in the results this does occur, but minimally affects the response (Figure 2c).

One can use the convolution integral of the pulse and the response function to determine the surface ocean concentration of a transient tracer:

\[ C_o(t) = \int_{t_i}^{t} F_c(u)r(t-u)du \]  

(A1)

This is a slightly simplified version of the one-dimensional model equation, where \( C_o \) is the surface concentration of a tracer, \( F_c \) is the flux, and \( r \) is the response function. In our case, the pulse, \( F_c \), is the change in carbon concentration at the surface each year. By taking the convolution integral of the pulse, and its response function, we can determine the change in mixed-layer concentration (Figure A2). A convolution integral (Equation A1) calculates the concentration at time \( t \) by calculating the fraction of previous pulses \( (F_c(u)) \), that entered the ocean at times \( u \) (from \( t_i = 0 \) to \( t \)), that remain in the surface ocean at time \( t \). This is generalizable to any tracer that is initially absent in the ocean. An intuition for convolution integral can be formed by visualizing it in discrete form (Figure A2). By summing all of the discrete pulses that are present in the mixed layer at a given time, one can arrive at an approximation of the exact convolution integral (Figure A2c,d). In this case the exact solution is the ocean concentration of the transient tracer. In this generalized example, we show the effect of pulse sampling frequency (Figure A2c). With more frequent pulse sampling, the more accurate the approximation of the convolution integral (A2c,d).
Figure A2. (a,b) Time series of the flux of transient tracer (concentration yr$^{-1}$), with different sampling frequencies shown with the blue bars. The left column is a 10 year sampling frequency, 1 year frequency in the right column. The blue bars are $F_c$ in Equation A1. (c,d) Predicted transient tracer concentration in the surface ocean, the left hand-side of Equation A1. The predicted concentration (black line) at any point in time is the sum of the individual pulses at that time (colored lines).

thus with infinitely many pulses one can capture the full convolution integral (concentration of some transient tracer). In the one-dimensional model case, the pulse is sampled annually, with no benefit to sampling at sub-annual frequencies.

Interestingly, the convolution integral can be used to solve for the flux. All we need to know is the flux at $t = 0$. The flux of transient tracer can be described with the following equation:

$$ F_C = \frac{k}{h} (C_{atm} - C_{ocn}) $$

(A2)

In this equation $F_C$ is the flux of tracer in units of quantity ($\mu$mol, mol, kg, etc) per m$^3$. On the right hand side, the air-sea difference is multiplied by the gas transfer velocity, $k$ (m yr$^{-1}$), and the mixed layer depth, $h$ (m).
We can calculate the initial flux of transient tracer with knowledge of the atmospheric history of the transient tracer, and also because by definition, we know the initial ocean concentration is 0. Beginning at $t = 0$, at each time step we can make these calculations in the following order to determine the flux for the entire period we have atmospheric measurements of the tracer:

1. Calculate the air-sea flux (Equation A2)

2. Sum up the pulses still present in the mixed layer to determine concentration at the next timestep (Equation A1).

This process is repeated to calculate the next year’s air-sea flux. After many time steps, the flux is responding to the change in concentration that occurred in the previous year due to the previous year’s flux, and any pulses that remain in the mixed layer.

**Appendix B: One-Dimensional Ocean Carbon Cycle Model Chemistry**

The $pCO_{2}^{ocn}$ of the one-dimensional ocean carbon cycle model is calculated as follows:

$$pCO_{2}^{ocn} = [pCO_{2}^{ocn,PI} + \delta pCO_{2}^{ocn}(C_{ant}, T_0)] \exp(\alpha_T \delta T) \quad (B1)$$

The response of $pCO_{2}^{ocn}$ to warming is parameterized as an exponential function as in Takahashi et al. (1993), with $\alpha_T$ set to 0.0423 (Equation A24; Joos et al., 2001). The $\delta pCO_{2}^{ocn}$ is calculated using a fixed ocean alkalinity of 2300 $\mu$mol kg$^{-1}$ and the preindustrial temperature, $T_0$. The chemistry of $\delta pCO_{2}^{ocn}$ is parameterized as follows:

$$\delta pCO_{2}^{ocn}(C_{ant}, T_0) = C_{ant}[A1 + C_{ant}(A2 + C_{ant}(A3 + C_{ant}(A4 + C_{ant}A5)))] \quad (B2)$$

With coefficients:

$$A1 = (1.5568 - 1.3993 \times 10^{-2} \times T_0) \quad (B3)$$

$$A2 = (7.4706 - 0.20207 \times T_0) \times 10^{-3} \quad (B4)$$

$$A3 = -(1.2748 - 0.12015 \times T_0) \times 10^{-5} \quad (B5)$$

$$A4 = (2.4491 - 0.12639 \times T_0) \times 10^{-7} \quad (B6)$$

$$A5 = -(1.5468 - 0.15326 \times T_0) \times 10^{-10} \quad (B7)$$

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