Quantifying the drivers of ocean-atmosphere CO$_2$ fluxes
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Key Points:

1.) We have developed a quantitative framework for diagnosing regional drivers of the air-sea CO$_2$ flux.

2.) Components can be evaluated in a model or can be derived from operational data, climatologies and ocean state estimates.

3.) CO2 fluxes result from balance between air-sea heat fluxes, biological activity and disequilibrium.

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Abstract. A mechanistic framework for quantitatively mapping the regional drivers of air–sea CO$_2$ fluxes at a global scale is developed. The framework evaluates the interplay between: (1) surface heat and freshwater fluxes that influence the potential saturated carbon concentration, which depends on changes in sea surface temperature, salinity and alkalinity, (2) a residual, disequilibrium flux influenced by upwelling and entrainment of remineralized carbon- and nutrient-rich waters from the ocean interior, as well as rapid subduction of surface waters, (3) carbon uptake and export by biological activity as both soft tissue and carbonate, and (4) the effect on surface carbon concentrations due to freshwater precipitation or evaporation. In a steady state simulation of a coarse resolution ocean circulation and biogeochemistry model, the sum of the individually determined components is close to the
known total flux of the simulation. The leading order balance, identified in
different dynamical regimes, is between the CO$_2$ fluxes driven by surface heat
fluxes and a combination of biologically-driven carbon uptake and disequilibrium-
driven carbon outgassing. The framework is still able to reconstruct simu-
lated fluxes when evaluated using monthly-averaged data and takes a form
that can be applied consistently in models of different complexity and ob-
servations of the ocean. In this way, the framework may reveal differences
in the balance of drivers acting across an ensemble of climate model simu-
lations or be applied to an analysis and interpretation of the observed, real-
world air–sea flux of CO$_2$. 
1. Introduction

The atmospheric inventory of carbon is regulated on inter-annual and longer timescales by the ocean carbon reservoir, mediated by the exchanges of carbon dioxide across the air–sea interface. This exchange (1) is proportional to the difference in ocean and atmosphere partial pressures of CO$_2$, and its efficiency is a function of temperature, salinity and wind speed, encapsulated in the gas transfer coefficient, $K$:

$$F_{CO_2} = K(pCO_{2}^{\text{ocean}} - pCO_{2}^{\text{atm}})$$  \hspace{1cm} (1)

In recent decades, coordinated observation campaigns and the compilation of local data sets have provided an unprecedented global view of the regional and seasonal changes in air–sea exchange of CO$_2$ (as illustrated in Figure 1a) based on measurements of atmospheric and surface ocean pCO$_2$, along with estimates of $K$ [Wanninkhof, 1992; Takahashi et al., 2002, 2009; Wanninkhof et al., 2013]. This growing database provides a clear qualitative and quantitative view of the patterns and rates of the air–sea flux at regional and global scales.

Since atmospheric pCO$_2$ is globally mixed on timescales of a year or so, regional and seasonal variations reflect oceanic processes that drive surface pCO$_2$ levels away from equilibrium with the overlying atmosphere. These include changes in solubility due to surface heat fluxes and temperature variations, the biotic fixation of carbon, as well as the upwelling of carbon-rich deep waters [e.g. Murnane et al., 1999; Sarmiento et al., 2000; Takahashi et al., 2002, 2009; Ito and Follows, 2013].

Though the total flux can be derived from information on the pCO$_2$ gradient, quantification of the relative contributions of each of these drivers of CO$_2$ flux is difficult at
regional and global scales. At a few sites around the globe, notably the Hawaii Ocean
Time-series (HOT) and the Bermuda Atlantic Time-series Station (BATS), comprehen-
sive biogeochemical observations provide an explicit view of the contributions of physical
and biological drivers to seasonal and annual air–sea carbon fluxes [Bates, 2001; Gruber
et al., 2002; Bates et al., 2002; Dore et al., 2003, 2009; Quay and Stutsman, 2003; Keeling
et al., 2004]. Models can be tested and constrained by data from the time-series sites
and global surveys to reveal the contributions from biological and physical drivers [e.g.
Murnane et al., 1999; Sarmiento et al., 2000; Takahashi et al., 2002, 2009; Schuster and
Watson, 2007; Lovenduski et al., 2007; Lovenduski and Ito, 2009; Corbiére et al., 2011;
Manizza et al., 2013].

A common starting point is to consider the rate of change of the concentration of
dissolved inorganic carbon ($C_T$) in the surface ocean, which can be described as the
balance of physical and biological processes:

$$\frac{\partial C_T}{\partial t} = -\nabla \cdot (\bar{u}C_T) + \nabla \cdot (\kappa \nabla C_T) - R_{C_T,P}S_{bio} - S_{CaCO_3} - \frac{F_{wC_T}}{\rho_w h} - \frac{F_{CO_2}}{h}, \quad (2)$$

Here the terms on the right hand side describe the divergence of physical transport by
advection and diffusion, including entrainment (terms one and two), biological sources
and sinks of organic soft-tissue and calcium carbonate (third and fourth terms), dilution
by freshwater fluxes (fifth term) and, last but not least, the air–sea gas exchange of CO$_2$.
Symbols are defined in detail in Table 1.

In cases where atmospheric CO$_2$ can be assumed constant, averaging over the seasonal
cycle, $\partial C_T/\partial t$ approaches zero. Further integrating over the layer depth, $h$, we can re-
arrange (2) to explicitly relate the time-averaged air–sea exchange of CO$_2$ to the driving
processes:

\[ F_{CO_2} = -\nabla \cdot (\bar{u}C_T)h + \nabla \cdot (\kappa \nabla C_T)h - R_{C_T:P}S_{bio}h - S_{CaCO_3}h - \frac{F_w C_T}{\rho_{fw}} \]  

(3)

The individual terms can be evaluated in numerical models, where complete four-dimensional diagnostic information is available [e.g. Lovenduski et al., 2013] (see also example in appendix A). While such a breakdown provides an explicit quantification of each of the processes, its application is restricted to numerical simulations since the required inputs are only sparsely covered by observations on the global scale. However, we can extend this approach to relate the driving terms in (3) to environmental fluxes and characteristics which are available in global climatologies and climate model diagnostics.

The purpose of this paper is to describe this extension and to test its validity and utility by diagnosing the drivers of CO\(_2\) fluxes in a global numerical simulation.

Section 2 outlines the framework which links climatological physical forcing and ocean carbon reservoirs. In Section 3 we test the framework by interpreting the contributions of different forcing processes in a numerical model simulation (described in Section 2.2). We quantify the errors introduced in the interpretation when input data is limited to monthly-averaged values, as we might encounter in applications to climatological observations or model comparison databases. Finally, in Section 4, we discuss the prospects for wider application of the framework.


Here we develop the extended diagnostic framework which quantitatively links physical and biological drivers to air-sea fluxes of CO\(_2\). We also describe the global ocean carbon
cycle simulation used to test the new diagnostic framework and its robustness to some underlying assumptions.

2.1. Relating air–sea CO$_2$ flux to surface drivers and ocean carbon reservoirs.

Expressions relating carbon fluxes and drivers (3) can be combined with parameterizations linking the individual terms to fluxes and state variables that are available in observed climatologies and standard climate model products. We also leverage on biogeochemical diagnostic approaches typically used to understand the controls on deep ocean carbon reservoirs [or carbon “pumps”, Brewer, 1978; Gruber et al., 1996; Ito and Follows, 2005; Williams and Follows, 2011].

At the sea surface, the total dissolved inorganic carbon concentration, $C_T$, can be separated into the saturated component, $C_{sat}$, the concentration a water mass would have if at equilibrium with atmospheric pCO$_2$, and a residual concentration, $C_{res}$, reflecting the disequilibrium due to a variety of physical and biological processes [Ito and Follows, 2013]:

$$C_T = C_{sat} + C_{res}.$$  \hspace{1cm} (4)

$C_{sat}$ depends on atmospheric pCO$_2$ and sea surface temperature, salinity and alkalinity. These relationships can be described assuming a linearization of carbonate chemistry [e.g. Williams and Follows, 2011]:

$$\delta C_{sat} \simeq \gamma_\theta \delta \theta + \gamma_S \delta S + \gamma_{A_T} \delta A_T + \delta pCO_2 \frac{\partial C_{sat}}{\partial pCO_2}$$  \hspace{1cm} (5)

where $\gamma_\theta$, $\gamma_S$ and $\gamma_{A_T}$ are solubility constants which reflect the close to linear relationships between $C_{sat}$ with temperature, salinity and alkalinity. While the relationship with pCO$_2$
is highly non-linear, for the purposes of this study we will be considering a fixed, uniform atmospheric pCO$_2$, so $\delta pCO_2 = 0$.

Substituting for $C_{sat}$ from (4) and (5) into (3) we express the drivers of $F_{CO2}$ in terms of changes in environmental characteristics.

$$F_{CO2} = \gamma_\theta \left( -\nabla \cdot (\vec{u}\theta) + \nabla \cdot (\kappa \nabla \theta) \right) h$$

$$+ \gamma_S \left( -\nabla \cdot (\vec{u}S) + \nabla \cdot (\kappa \nabla S) \right) h$$

$$+ \gamma_{AT} \left( -\nabla \cdot (\vec{u}AT) + \nabla \cdot (\kappa \nabla AT) \right) h$$

$$+ \left( -\nabla \cdot (\vec{u}C_{res}) + \nabla \cdot (\kappa \nabla C_{res}) \right) h$$

$$- R_{CT:pS_{bio}}h - S_{CaCO_3}h$$

$$- \frac{F_wC_T}{\rho_{fw}}$$

(6)

The first three terms on the right couch convergences of physical and biogeochemical characteristics as drivers of CO$_2$ fluxes. We can, in turn, relate these convergences in terms of air–sea fluxes of heat and freshwater. Consider, for example, the governing equation for surface temperature:

$$\frac{\partial \theta}{\partial t} = -\nabla \cdot (\vec{u}\theta) + \nabla \cdot (\kappa \nabla \theta) - \frac{F_{heat}}{\rho C_p h}$$

(7)

Assuming steady state we can substitute $-\nabla \cdot (\vec{u}\theta) + \nabla \cdot (\kappa \nabla \theta) = (F_{heat}/\rho C_p h)$ into (6).

Similar substitutions can be made, including reframing biological sources and sinks in
terms of the divergence of the nutrient transport (see appendix B for a detailed derivation):

\[
F_{CO_2} = \gamma_{\theta} \frac{F_{heat}}{\rho C_p} + \frac{F_W}{\rho_{fw}} (\gamma S \overline{\Theta} + \gamma_{AT} \overline{AT} - \overline{CT})
\]

\[
- R_{CT:P} (\nabla \cdot (\vec{u}P) + \nabla \cdot (\kappa \nabla P)) h
\]

\[
- \frac{1}{2} R_{CaCO_3} R_{CT:P} (\nabla \cdot (\vec{u}P) + \nabla \cdot (\kappa \nabla P)) h
\]

\[
+ (\nabla \cdot (\vec{u}C_{res}) + \nabla \cdot (\kappa \nabla C_{res})) h
\]

(8)

We have now expressed the air–sea flux as a set of components related to fluxes that are
either evaluated operationally (such as surface heat fluxes) or which can be derived from
climatologies and ocean state estimates (such as nutrient transports). Each term in (8)
quantifies a specific process that drives a proportion of the air–sea flux of CO₂.

In (8), the first term on the right (first row) is the “potential” CO₂ flux due to heat
fluxes [Murnane et al., 1999; Takahashi et al., 2002, 2009], which drive changes in \( C_{sat} \),
the solubility of CO₂. The second row of terms reflect the influence of surface evaporation
and precipitation in three ways: the dilution of salinity and alkalinity, both affecting \( C_{sat} \),
and the direct dilution of dissolved inorganic carbon, \( C_T \). Notice that the dilution of \( C_T \)
and alkalinity compensate: one acts to reduce pCO₂ solubility and the other to increase it.
Both heat and freshwater fluxes, \( F_{heat} \) and \( F_W \), are standard products of climate models
and climatologies based on operational weather products [Kistler et al., 2001; Dee et al.,
2011]. These drivers are quantified and reasonably well characterized.
The third row describes the production or remineralization of organic soft tissue with a carbon to phosphorus ratio $R_{C,P}$, integrated to depth $h$. Using the governing equation for phosphate (see appendix B), we have linked biological sources and sinks to the divergence of the phosphate transport field. Advective and eddy-diffusive phosphate fluxes are easily obtained from ocean models and may be reconstructed offline (with some introduced error) if the flow field and phosphate concentration is known. Estimates may also be based on climatological distribution of phosphate [e.g. the World Ocean Atlas, Garcia et al., 2014] with flow field and mixing parameters based on ocean state estimates [Wunsch and Heimbach, 2007; Forget, 2010; Forget et al., 2015]. We note that nitrate might equally well be the basis of the biological flux divergence estimate, depending on the currency of the model or data available. However, nitrogen fixation and denitrification make the interpretation of the flux divergences a little more uncertain.

The fourth row represents the production of calcium carbonate in the surface ocean linked simply to soft tissue production in the surface ocean by a simple constant rain ratio, $R_{CaCO_3}$ and integrated to depth $h$. This simple scaling holds in the upper ocean, although the full governing equation for alkalinity could be used (again, see appendix B), to take into account the different dissolution profiles of organic and inorganic particles.

The fifth and final row describes the divergence of the transport of the disequilibrium carbon reservoir, $C_{res}$, defined here as $C_{res} = C_T - C_{sat}$. Below the surface mixed layer $C_{res}$, as defined, includes contributions from regenerated organic matter whereas, when modeling the carbon reservoirs in the deep ocean, the regenerated pool is explicitly diagnosed [Brewer, 1978; Gruber et al., 1996; Williams and Follows, 2011; Lauderdale et al., 2013]. Thus, here the divergence of $C_{res}$ transport includes three important effects:
(i) the entrainment of subsurface waters rich in regenerated $C_T$ that will drive outgassing, (ii) changes in surface fluxes of heat and freshwater that affect CO$_2$ solubility. For example, while cooling implies a positive change in the saturated carbon and therefore ocean CO$_2$ uptake, in a limited time period this potential flux is not achieved so the CO$_2$ flux due to $C_{res}$ is negative (outgassing) to compensate, and (iii) the time-delay of air–sea carbon flux response which results in accumulation of significant disequilibrium and shifts the air–sea flux response downstream of the previous two influences.

$C_{res}$ can be diagnosed in models or from climatological data [the GLODAP atlas, Key et al., 2004] by estimating $C_{sat}$ and subtracting it from $C_T$. It is not currently possible to cleanly separate three factors that affect the $C_{res}$ fluxes. This component will also accumulate small errors associated with linearization of the carbonate chemistry in (5).

Each of the terms has a clear, mechanistic interpretation and we suggest that (8) is more informative than (3). Additionally, all of the terms are formulated so that they can be quantitatively mapped from climatological databases and standard output from appropriate carbon cycle models.

As a proof-of-concept, we interpret the relative role of physical and biological drivers on patterns of air–sea carbon flux in a coarse-resolution ocean circulation and biogeochemistry model simulation. This provides a testbed where the air–sea flux of CO$_2$ is calculated prognostically at every timestep by the model (using equation 1). We test the sensitivity of the framework to the input data density: climatological ocean data which might be used as inputs typically resolves monthly variations. Using the numerical simulations we ask whether a useful diagnosis could be made with inputs at such temporal resolution.
In the next subsection we briefly describe the numerical simulation which forms the testbed. The application is discussed in Section 3.

### 2.2. Testbed numerical simulation

The testbed simulation is a coarse resolution (2.8° × 2.8°, 15 vertical levels) version of the MITgcm \cite{Marshall1997} with online biogeochemistry \cite{Dutkiewicz2006, Parekh2006}. The configuration is similar to the control experiment in Lauderdale et al. \cite{Lauderdale2013}, using the Gent and McWilliams \cite{Gent1990} scheme with a constant thickness diffusivity of 1000 m² s⁻¹ but with the addition of the KPP non-local boundary layer mixing scheme \cite{Large1994} that enhances diapycnal mixing in regions where the water column is convectively unstable (above the constant value of 5×10⁻⁵ m²s⁻¹), including entrainment due to mixed layer deepening.

Biological transformations between carbon and nutrients are related using the fixed stoichiometric ratios of \( R_{C:N:P:O} = 117 : 16 : 1 : -170 \) \cite{Anderson1994} with a prescribed inorganic to organic rain ratio \( (R_{CaCO_3}) \) of 7% \cite{Yamanaka1997}. Air–sea fluxes of CO₂ are calculated prognostically using (1) and are dependent on the square of local wind speed \cite{Wanninkhof1992}. The annually-averaged atmospheric CO₂ is 281 µatm and the total ocean-atmosphere carbon inventory is conserved as there is no riverine carbon input, sediment carbon burial nor external CO₂ emissions (hence we neglect the accumulation of anthropogenic CO₂ at this stage). The simulations examined here have been integrated for 25,000 years, coming to a clear thermodynamic and biogeochemical steady state.

Advective and diffusive fluxes of all state variables, surface fluxes of heat and freshwater, as well as biological production and losses of dissolved and particulate organic carbon and...
calcium carbonate were evaluated at each timestep and saved as monthly averaged fields (see appendix A). This provides a complete set of inputs for application and verification of the diagnostic framework. Furthermore, we use monthly-averaged physical and biogeochemical scalars, velocity and diffusivity fields mimicking the data density of typically available climatologies, to calculate oceanic fluxes in “offline” mode and then apply the diagnostic framework to discover if a useful diagnosis could be made with inputs at such a low temporal resolution.

3. Results and Discussion: Quantifying CO₂ fluxes and drivers


The global simulation qualitatively and quantitatively captures the magnitude and variation of the climatological, observed air–sea flux of CO₂ [Figure 1a–c, Takahashi et al., 2009]. Some differences are expected since the climatology reflects the current, transient state of the carbon cycle (in the reference year 2000, atmospheric CO₂ was around 350 µatm) while the simulation is for the natural, preindustrial state (∼280 µatm). In the climatology, uptake of anthropogenic carbon by the ocean is evident in the stronger, more widespread fluxes at high latitudes, relative to the simulation, as well as the slightly weaker outgassing at the Equator. The simulation provides a plausible, self-consistent system for which all fluxes and state variables can be accurately quantified. In the following section, we infer the drivers of the simulated pattern using the framework outlined in (8).

3.2. Inferred CO₂ flux and drivers.

We evaluated each of the terms in equation (8) in the numerical simulation using fluxes from each timestep and averaged into monthly fields. Heat and freshwater fluxes were
obtained from the model’s physical boundary conditions. The saturated and disequilibrium components were evaluated following standard procedures [Williams and Follows, 2011] using $C_T$, $\theta$, $S$ and $A_T$ in the upper ocean, as well as the atmospheric pCO$_2$ of 281 µatm from the simulation. The solubility coefficients ($\gamma_{\theta}$, $\gamma_S$ and $\gamma_{A_T}$) were determined (Table 2) by calculating $C_{sat}$ over a range of values for temperature, salinity or alkalinity while holding the others (including atmospheric CO$_2$) at model surface mean values [Lewis and Wallace, 1998; Goodwin and Lenton, 2009; Ito and Follows, 2013].

We integrated the terms in (8) over the upper 50 m, in order to map the individual driver distributions in terms of an air-sea carbon flux. The framework reveals that surface heat forcing (Figure 2a), the dilution of alkalinity (Figure 2c) and $C_T$ (Figure 2d), the divergent transport of $C_{res}$ (Figure 2e) and biological sources and sinks (Figure 2f) are all of similar magnitude though with different patterns. The forcing due to dilution of salinity (Figure 2b) is much less significant. We also note that the dilution of alkalinity and $C_T$ are strongly compensatory, as anticipated.

We can simplify the diagnostics, showing the leading order aggregated terms in Figure 3. The total reconstructed flux, $F_{CO2}$ (Figure 3a), is largely accounted for by heat flux forcing of solubility (Figure 3b) and the sum of direct biological forcing and the divergence of $C_{res}$ (Figure 3c). The compensation between $C_{res}$ fluxes and biological forcing (Figure 2e,f) that lead us to group the drivers in this way is, in part, because biological carbon uptake is supported by the entrainment of regenerated nutrients, which simultaneously supplies regenerated carbon from the ocean interior to drive $C_{res}$ outgassing [Ito and Follows, 2013].
The biogenic-$C_{res}$ compensation is far from complete on the local scale due to inefficiencies in biological activity, which may be restricted from full potential in some regions such as the Southern Ocean, limited by light and/or micronutrient concentrations [Dutkiewicz et al., 2006]. Nevertheless, unutilized nutrients subducted from these regions may fuel a biological activity in other parts of the ocean through entrainment into the surface layer [Sarmiento et al., 2004]. This is the case in most regions (Figure 2f), although, again, the $C_{res}$ component is also affected by changes in solubility, particularly the cooling and rapid subduction of water masses from the surface. This would reinforce $C_{res}$ outgassing at high latitudes because the potential saturated concentration determined by the heat forcing is not achieved in the limited time at the surface, so that $C_{res}$ is negative to compensate. Conversely, there are a few regions of $C_{res}$ driven uptake that correlate with patches of intense heat fluxes (warming, leading to outgassing in Figure 2a), which suggests that again, these surface waters are being rapidly advected downstream before local equilibration to the potential saturated concentration, determined by the heat forcing.

There is a negligible contribution from the sum of CO$_2$ fluxes driven by the hydrological cycle, that is, changes in salinity and alkalinity, which affect $C_{sat}$, and changes in the $C_T$ concentration (Figure 3d).

We note that the total flux evaluated as the sum of the individually quantified components accurately reflects $F_{CO2}$ as calculated in the forward simulation (compare the maps in Figures 1b and 3a, or the zonal-average in Figure 1d). This is pleasing, indicating that no key processes have been overlooked (compared to the numerical simulation).

The global, annual average model–reconstruction difference is very small: $-8 \times 10^{-10}$ mol C m$^-2$ s$^-1$, less than one per cent of typical average fluxes locally, and of the order of
0.1 PgC yr$^{-1}$ integrated globally over one year (Table 3). Over this timeframe, most of the components go to zero, which would be expected at steady state. Outgassing driven by $C_{res}$ is closely compensated by the uptake driven by biological forcing, with a small residual probably representing a proportion of $C_{res}$ linked to errors in linearization of the $C_{sat}$ equation (5).

When we use fluxes calculated “offline” using monthly-averaged physical transports along with monthly averages of physical and biogeochemical concentrations to imitate climatological databases (Figure 1d), we find a degree of error is introduced, particularly leading to overestimation of equatorial outgassing and Southern Ocean uptake. Partly, there is an offset at midlatitudes (around 40$^\circ$N/S) where the CO$_2$ flux framework slightly overestimates oceanic carbon uptake in the swift Gulf Stream, Kuroshio and Antarctic Circumpolar currents. This is not surprising since such regions will be prone to errors in the estimated flux divergence of $C_{res}$ due to swift current and sharp gradients. Furthermore, uncertainty in vertical mixing of carbon and nutrients as a result of smoothing vertical concentration gradients, leads to errors in the biological and disequilibrium terms. Nevertheless, the framework is still able to reconstruct the net air–sea CO$_2$ fluxes and their variation to a reasonable degree, which gives us confidence that careful application of (8) to climatological data and observations could warrant merit.

### 3.3. Regional variations in the drivers of air–sea CO$_2$ fluxes.

Having demonstrated that the diagnostic framework is able to capture the magnitude and regional variations in CO$_2$ fluxes, we now illustrate the application of the framework by quantifying the relative significance of different CO$_2$ flux drivers over several distinct regions of the simulated ocean. The results are summarized in Table 3.
We chose the aggregated regions according to whether they are a net source or sink or neutral with respect to atmospheric \( \text{CO}_2 \) (see the zero contour of the simulated net air–sea \( \text{CO}_2 \) flux in Figure 1b and the shaded regions in Figure 4). The regions are: (i) the uptake region of the North Atlantic and North Pacific basins, (ii) the outgassing region of the tropics and subtropics, (iii) the region of uptake in the Southern Ocean midlatitudes (north of \( \sim 60^\circ \text{S} \) to the zero \( \text{CO}_2 \) contour), and finally, (iv) the relatively neutral polar Southern Ocean, south of \( \sim 60^\circ \text{S} \). A further decomposition also reveals some significant longitudinal patterns and is illustrated in Table 4 and Figure 4.

### 3.3.1. Northern Hemisphere (oceanic uptake)

In the North Pacific and North Atlantic the net \( \text{CO}_2 \) flux is into the ocean, as a result of solubility-driven uptake in the poleward flowing Gulf Stream and Kuroshio western boundary currents, which cool, and due to biological uptake. Disequilibrium-driven outgassing is large, which is a combination of strong convective mixing, particularly around southern Greenland, bringing remineralized carbon (and nutrients) to the surface, as well as subduction of surface waters into the ocean interior before coming to equilibrium with the atmosphere.

There are three different patterns governing zonal variation of \( \text{CO}_2 \) fluxes in the Northern Hemisphere (Figure 4a and Table 4):

1. In the western boundary current dominated Northwest Pacific and Northwest Atlantic as mentioned above, the swift northward boundary current flow produces rapid cooling that is partially compensated by the subduction of mode waters that are incompletely equilibrated with the atmosphere causing negative \( C_{\text{res}} \) component fluxes. As a result,
the absolute values of the disequilibrium fluxes are significantly larger than the opposing biologically-driven fluxes.

2. In the Northeast Atlantic and high latitude Northern Atlantic, the cooling of surface waters and deep water formation mostly drives the CO$_2$ uptake. This deep convective mixing, meanwhile, brings nutrient-rich waters to the surface that drives biological CO$_2$ uptake, but also subducts deep waters from the surface before they have fully equilibrated with the atmosphere, again leading to outgassing disequilibrium fluxes.

3. In the more quiescent Northeast Pacific, CO$_2$ uptake is dominated by biological activity which is partially offset by the residual fluxes, with little influence of heat fluxes.

3.3.2. Equatorial–Subtropical region (oceanic outgassing)

The equatorial-subtropical region generally outgasses CO$_2$ to the atmosphere. Thermally-driven outgassing is countered by net biologically-driven CO$_2$ uptake, since individually, the residual component flux is completely counteracted by biological activity, although only partial equilibrium after heating may reduce the $C_{res}$-driven flux. Furthermore, assuming that all the nutrients required to drive the biological uptake are supplied by the Equatorial upwelling then suggests that these waters had outgassed CO$_2$ when they were last in contact with the atmosphere. If this were not the case, the $C_{res}$-driven outgassing flux would closely balance the biologically-driven uptake flux.

There are also longitudinal gradients in the regional drivers of the air–sea CO$_2$ flux at low latitudes, particularly in the West and East Equatorial Pacific (Figure 4b and Table 4). The strongest upwelling of cool waters occurs in the East Pacific, associated with the sea surface temperature gradient and shoaling of the thermocline, with subsequently greater outgassing through warming due to heat fluxes. This is the region of strongest CO$_2$
outgassing, and its variability in the real ocean is heavily influenced by El Niño–La Niña conditions, associated with weakening (strengthening) of the longitudinal thermocline gradient and reduced (increased) upwelling of cool waters and residual carbon.

### 3.3.3. Southern Ocean midlatitudes (oceanic uptake)

In southern midlatitudes, net uptake of CO$_2$ by the ocean occurs in all three basins (Table 3). Over the entire region, biologically-driven CO$_2$ uptake is completely compensated by the $C_{res}$-driven outgassing, which are mechanistically linked, leaving CO$_2$ uptake driven by cooling to dominate the net flux.

Considering the zonal variation (Figure 4c and Table 4), the cooling flux is dominated by the Indian sector of the Southern Ocean where there is strong heat loss at the boundary between subtropical waters and the Antarctic Circumpolar Current, falling to one-third of the size in the Pacific sector of the Southern Ocean. However, in the Indian Ocean sector the significant excess of $C_{res}$-driven outgassing compared to biologically-driven uptake suggesting that the cooling waters are incompletely equilibrated before subduction. Compare this to the Pacific sector, where biological activity closely compensates the residual flux.

The South Atlantic region as a whole displays a different pattern, with small heat flux-driven CO$_2$ fluxes (that actually suggest outgassing), leaving biological drivers to dominate the overall CO$_2$ uptake, incompletely compensated by $C_{res}$ driven outgassing. A patch of outgassing in the South Atlantic at 40°S (Figures 1b and 3a), which does not appear in the climatology (Figures 1a), may be a model artifact associated with the incorrect positioning of the subtropical front where the ACC detaches from the coast of Argentina. Anomalously cold waters of the ACC would induce strong surface temperature relaxation in the model,
which is seen in the heat flux-driven CO$_2$ flux component (Figure 3b). However, there is also a strong uptake flux associated with the $C_{res}$ component (Figure 2e), suggesting that the swift current advects the surface waters into a neighboring cooling region before the warming effect is fully achieved. When integrated over the entire south Atlantic region, where cooling is also occurring in common with the rest of the Southern Ocean midlatitudes, the final result is that these opposing heat fluxes closely compensate in this region. Furthermore, the pattern of outgassing (due to upwelling of regenerated carbon) and uptake (due to incomplete equilibration to the saturated concentration determined by the heat forcing) in the disequilibrium component results in a comparatively small $C_{res}$-driven flux.

### 3.3.4. Southern Ocean south of 60°S (neutral)

Finally, in the highest southern latitudes, south of roughly 60°S, the average flux is around zero (Table 3) although many of the individual components are significantly larger, indicating considerable compensation and interplay. Cooling of waters adjacent to Antarctica induces a positive CO$_2$ flux associated with the saturated carbon pool. The largest individual flux in this region comes from the residual, disequilibrium component driven by upwelling of deep waters rich in regenerated carbon and nutrients. While biological activity partly compensates, the net effect is negative (leading to outgassing) because the upwelled nutrients are incompletely utilized and biological activity is restricted from its full potential, limited by light and/or micronutrient concentrations. Additionally, cooling with incomplete equilibration may result in $C_{res}$-driven outgassing with the atmosphere before subduction into the deep ocean. In the Weddell Sea of this model in particular, there is a relatively strong CO$_2$ uptake associated with cooling due to heat fluxes, twice
that of the East Antarctica and the Ross Sea regions, which is partly compensated by outgassing associated with the residual flux, suggesting the bottom waters formed here are incompletely equilibrated.

4. Summary and Outlook

We have developed diagnostic framework for interpreting the regional drivers of the air–sea flux of CO$_2$ which can be expressed succinctly and generally as:

$$F_{CO_2} = \gamma \frac{F_{heat}}{\rho C_p}$$

$$+ \frac{F_W}{\rho_{fw}} (\gamma S S + \gamma A T - \overline{C_T})$$

$$- R_{C_T:P}(-\nabla \cdot (\vec{u} P) + \nabla \cdot (\kappa \nabla P)) h$$

$$- \frac{1}{2} R_{CaCO_3} R_{C_T:P}(-\nabla \cdot (\vec{u} P) + \nabla \cdot (\kappa \nabla P)) h$$

$$+ (\nabla \cdot (\vec{u} C_{res}) + \nabla \cdot (\kappa \nabla C_{res})) h$$

Drivers of the air–sea CO$_2$ flux are:

1. The effect of heat and freshwater fluxes on the saturated ocean carbon concentration, which depends on surface temperature, salinity and alkalinity at steady state atmospheric pCO$_2$.

2. The direct effect of freshwater dilution on surface $C_T$ concentrations.

3. Carbon uptake and export by biological activity (as both soft tissue and carbonate) through nutrient utilization, and

4. Atmosphere-ocean disequilibrium, which is influenced by upwelling and entrainment of remineralized carbon- and nutrient-rich waters from the ocean interior as well as rapid subduction of surface waters before reaching full equilibrium.
Here, in order to cleanly illustrate the framework, we have diagnosed the relative importance of air–sea flux drivers from a numerical model simulation. We demonstrated that the sum of the individually determined components is close to the known total flux of the simulation. In breaking down the contributions in the simulation, we find a dominant role for the air–sea heat flux forcing, as has been suggested by sensitivity experiments [Follows et al., 1996; Murnane et al., 1999; Sarmiento et al., 2000; Takahashi et al., 2002, 2009]. However, the analysis reveals that this is not universal, with a dominant role for biological- and air–sea disequilibrium driven fluxes. In the upwelling regions of the Southern Ocean, the combined effect of biological activity and disequilibrium opposes the heat flux-driven CO₂ uptake. Here, biological activity is limited from its maximum efficiency and so the upwelling of regenerated carbon is not completely compensated. Subduction of cooling waters before equilibrium also reinforces the C_{res} outgassing. In the Equatorial upwelling regions, biological CO₂ uptake is generally larger than the disequilibrium-driven fluxes, offsetting the warming-driven outgassing.

Going beyond this proof-of-concept analysis of a single simulation, we believe that this diagnostic approach has potential for interesting applications. The approach could be applied offline with simulation data from climate models to reveal differences in the balance of drivers acting across an ensemble of simulations. Potentially more interesting still is the application to observed climatologies and the real-world air–sea flux of CO₂. Using monthly-averaged fluxes calculated “offline”, the framework is still able to successfully reconstruct air–sea fluxes of CO₂ compared to the model simulation, albeit with a degree of error introduced into the biological activity and disequilibrium terms. Finally, there is
also the prospect of extending the approach to include a time-varying atmospheric pCO$_2$.

This is clearly challenging but should be explored.

In summary, we have developed and presented a new framework for analyzing the relative contributions of different physical and biogeochemical drivers to the air–sea flux of CO$_2$. Using numerical model simulations, where the correct solution is known, we have demonstrated that the method can work given appropriate input data. We suggest that applications to ensembles of model simulations and, in particular, climatological observations may be fruitful.
Appendix A: The upper ocean carbon budget

Taking the model surface layer of uniformly 50 m depth over which (2) is integrated, the
carbon budget can be partitioned into six components, combining the effects of soft-tissue
and order of magnitude smaller carbonate biogenic activity (Figure 5). Divergence of the
horizontal and vertical advective fluxes (including parameterized eddy bolus advection)
taken separately are an order of magnitude larger than the sum (Figure 5a) and therefore
show a large degree of compensation. For example, upwelling of carbon in the equatorial
regions and the Southern Ocean is largely, but incompletely redistributed by the horizontal
gyre circulation leading to small, but significant, accumulation of carbon. Similarly, in
the subtropical gyres there is convergence of $C_T$ due to horizontal advection but this is
counteracted by downwelling and deepened pycnoclines leading to net decrease in $C_T$.

Divergence of horizontal and vertical diffusive fluxes of $C_T$ (Figure 5b) are almost en-
tirely due to vertical diffusion, increasing $C_T$ concentrations everywhere, but particularly
elevated in the eastern equatorial Pacific, parts of the Southern Ocean and western bound-
dary current regions where vertical gradients are large and convective mixing is important.

Upwelling of carbon to the surface is also accompanied by dissolved nutrients that drive
widespread decrease in surface $C_T$ concentrations due to biological fixation of carbon
at the surface and vertical export to depth as sinking particles (Figure 5c). The rate
of carbon concentration change here is prognostically calculated by the biogeochemical
model as the net effect of biological productivity depending on the availability of light
and nutrients, export fluxes and remineralization of dissolved and particulate organic and
inorganic matter. Biological activity particularly occurs in the equatorial and high latitude
oceans where nutrients are abundant in the model, although in some places limited by
availability of light and micronutrients [e.g. Dutkiewicz et al., 2006], with low biological drawdown in the oligotrophic subtropical gyres.

Net surface freshwater fluxes can induce changes in surface carbon concentration (Figure 5d). Regions where precipitation dominates (including relaxation to surface salinity climatology) such as low latitudes under the Intertropical Convergence zone, the $C_T$ concentration is decreased through dilution, whereas in the subtropical regions where evaporation is the dominant process, $C_T$ concentration increases by roughly the same amounts.

Accumulating the effects of advection, diffusion, biological activity and freshwater fluxes leads to regions of net $C_T$ accumulation or reduction at the surface, which drive oceanic pCO$_2$ variations and ultimately air–sea fluxes of CO$_2$. The CO$_2$ fluxes in Figure 5e (redrawn from Figure 1b) are computed by the biogeochemistry model using (1) and show outgassing in the equatorial-subtropical regions and uptake at the midlatitudes.

Appendix B: Development of the carbon flux diagnostic framework

This section lays out the specifics of the new, mechanistic carbon flux framework (8). Terms are defined and described in Table 1.


Considering (3), the most difficult terms to infer are the effects of biological activity, $S_{bio}$ and $S_{CaCO3}$, on the surface flux of CO$_2$. A similar equilibrium relationship to (2) can be written considering the steady state phosphate concentration (B1), from which the $S_{bio}$ component can be obtained from the advective and diffusive fluxes of nutrients (B2).
Nitrate could also be used here instead.

\[ \frac{\partial P}{\partial t} = -\nabla \cdot (\vec{u}P) + \nabla \cdot (\kappa \nabla P) - S_{bio} \]  
\[ S_{bio} = -\nabla \cdot (\vec{u}P) + \nabla \cdot (\kappa \nabla P) \]  

Rearranging the steady state budget of alkalinity \((A_T, B3)\) could yield a similar expression for the \(S_{CaCO_3}\) component from the advective and diffusive fluxes of alkalinity, the consumption of charged nutrients and dilution or concentration of \(A_T\) by surface freshwater fluxes \((B4)\).

\[ \frac{\partial A_T}{\partial t} = -\nabla \cdot (\vec{u}A_T) + \nabla \cdot (\kappa \nabla A_T) - 2S_{CaCO_3} - R_{N:P}S_{bio} - \frac{F_w A_T}{\rho_{fw} h} \]  
\[ S_{CaCO_3} = \frac{1}{2} \left[ -\nabla \cdot (\vec{u}A_T) + \nabla \cdot (\kappa \nabla A_T) - R_{N:P}S_{bio} - \frac{F_w A_T}{\rho_{fw} h} \right] \]  

For simplicity, however, \(S_{CaCO_3}\) can also be estimated by assuming an organic carbon to inorganic carbonate production/remineralization ratio \((R_{CaCO_3})\), in which case:

\[ S_{CaCO_3} = \frac{1}{2} R_{CaCO_3} R_{CT:P} S_{bio} \]  

**B2. Saturated and disequilibrium carbon reservoirs.**

The advective and diffusive fluxes of \(C_T\) in the carbon budget \((3)\) can be further broken down into several different constituents, linking surface carbon fluxes to interior carbon reservoirs \([Brewer, 1978; Gruber et al., 1996; Ito and Follows, 2005; Williams and Follows, 2011]\). In turn, this allows atmosphere-ocean carbon fluxes to be linked to physical forcing of the ocean.

At the sea surface, a parcel of water in chemical equilibrium with the atmosphere will have a dissolved carbon concentration equal to the saturated concentration, \(C_{sat}\) \((B6)\). However, since CO\(_2\) gas exchange is not instantaneous, having a timescale of the order
of 1 year [Ito et al., 2004], there will be a residual disequilibrium concentration, $C_{res}$, which results from other processes that drive the surface $C_T$ concentration away from the “potential” chemical equilibrium concentration. This measure of incomplete air–sea gas exchange is largely influenced by the upwelling of regenerated carbon of biogenic origin from the deep ocean [Ito and Follows, 2013] and rapid subduction of waters from the surface before equilibration with the atmosphere. In some regions, outside the surface layer, there may also be a contribution to $C_T$ from the remineralization of sinking soft tissue ($C_{soft}$) and carbonate ($C_{carb}$) particles of biogenic origin, however these reservoirs are zero by definition in the surface ocean.

\[ C_T = C_{sat} + C_{res} \]  

(B6)

The concentration of $C_{sat}$ in the surface ocean depends on atmospheric pCO$_2$ and the surface temperature, salinity and alkalinity. Again, away from the surface ocean there is a contribution to the alkalinity from remineralization of calcium carbonate particles and addition of charged nutrients ($A_T = A_{pre} + A_{reg}$), however these reservoirs are zero by definition in the surface ocean.

It is possible to separate the individual effects of the latter three terms by linearizing the buffering capacity of seawater at constant pCO$_2$ such that perturbations of $C_{sat}$ can be evaluated from surface changes in $\theta$, $S$ and $A_T$ (B7).

\[ \delta C_{sat} \simeq \gamma_\theta \delta \theta + \gamma_S \delta S + \gamma_{A_T} \delta A_T + \delta pCO_2 \frac{C_{sat}}{BpCO_2} \]  

(B7)

The solubility coefficients ($\gamma_\theta$, $\gamma_S$ and $\gamma_{A_T}$) that represent the tight linear relationships between equilibrium carbon concentrations and forcing can be determined by calculating $C_{sat}$ over a range of values for temperature, salinity or alkalinity while holding the others.
(including atmospheric CO₂) at surface mean values [Lewis and Wallace, 1998; Goodwin and Lenton, 2009; Ito and Follows, 2013] (Table 2).

The relationship between \( C_{sat} \) and pCO₂ is non-linear due to the buffering capacity of seawater, and an expression involving the Revelle buffer factor, \( B \), is required [Ito and Follows, 2005; Goodwin et al., 2007] to correctly diagnose changes in the saturated carbon associated with changes in atmospheric CO₂ (B8), however we do not develop this part of the framework, keeping atmospheric CO₂ at a constant value in (5) neglecting anthropogenic carbon emissions.

\[
B = \left( \frac{\delta p\text{CO}_2}{\delta C_T} \right) \left( \frac{C_T}{p\text{CO}_2} \right)
\]  

(B8)

Thus the concentration of \( C_{sat} \) can be diagnosed for other data sources as will be described in the next section, however knowledge of \( C_T \) is still needed to determine \( C_{res} \) using (B6).

**B3. Evaluating individual components of the saturated carbon flux.**

Individual contributions from \( \delta \theta \), \( \delta S \) and \( \delta A_T \) (B7) to the “potential” equilibrium air–sea CO₂ flux can be calculated from the steady state budgets for temperature (B9), salinity (B10) and alkalinity (B11). Note that the biological formation of calcium carbonate is not included in the alkalinity equation because the dominant balance at steady state is between the fluxes of alkalinity due to ocean circulation and air-sea freshwater fluxes. Biological fluxes are an order-of-magnitude smaller. The dominance of freshwater at the surface produces a close linear relationship between surface concentrations of alkalinity and salinity. This does not suggest that we totally disregard the small contribution of calcium carbonate cycling in the surface layer, but, by the definition of (B6) we have instead partitioned it into the \( C_{res} \) pool. This is consistent with the treatment of the
biological fixation of soft-tissue carbon, which is also accounted for in the disequilibrium pool since increased productivity drives undersaturation of $C_T$, making the concentration of $C_{\text{res}}$ more negative, which leads to CO$_2$ uptake.

\[
\frac{\partial \theta}{\partial t} = -\nabla \cdot (\bar{u} \theta) + \nabla \cdot (\kappa \nabla \theta) - \frac{F_{\text{heat}}}{\rho C_p h} \tag{B9}
\]

\[
\frac{\partial S}{\partial t} = -\nabla \cdot (\bar{u} S) + \nabla \cdot (\kappa \nabla S) - \frac{F_w S}{\rho_{fw} h} \tag{B10}
\]

\[
\frac{\partial A_T}{\partial t} = -\nabla \cdot (\bar{u} A_T) + \nabla \cdot (\kappa \nabla A_T) - \frac{F_w A_T}{\rho_{fw} h} \tag{B11}
\]

At steady state, setting $\partial \theta/\partial t$, $\partial S/\partial t$ and $\partial A_T/\partial t$ to zero, the changes in saturated carbon ($\delta \theta$, $\delta S$ and $\delta A_T$) and the residual carbon concentrations can be inferred directly from surface heat and freshwater fluxes and substituted in (2) and (3) for the advective and diffusive fluxes of $C_T$ (B12).

\[
\nabla \cdot (\kappa \nabla C_T) - \nabla \cdot (\bar{u} C_T) = \frac{\gamma_{\theta} F_{\text{heat}}}{\rho C_p} - \frac{F_w}{\rho_{fw} h} \left( \gamma_S S + \gamma_{A_T} A_T \right) - \nabla \cdot (\bar{u} C_{\text{res}}) + \nabla \cdot (\kappa \nabla C_{\text{res}}) \tag{B12}
\]

Again, note that the contributions to the saturated carbon exchange due to temperature, salinity and alkalinity are potential fluxes that are only fully realized at chemical equilibrium. In reality, the individual temperature, salinity and alkalinity fluxes will only be partially fulfilled, with the remainder collected in the residual, disequilibrium flux ($C_{\text{res}}$).

Drawing together these different components into (3) with the additional parameters from (B2), (B5), (B6) and (B12) substituted in for the soft-tissue and carbonate components of biological activity, saturated carbon advection and diffusion from temperature, salinity and alkalinity surface forcing and finally $C_{\text{res}}$, which results from other processes
such as upwelling into the surface layer that drive the surface $C_T$ concentration away from equilibrium gives a new quantitative, mechanistic framework to diagnose regional drivers of the global air–sea CO$_2$ fluxes (8).
Figure 1: Air–sea CO$_2$ fluxes (mol C m$^{-2}$ s$^{-1}$) for (a) the contemporary Takahashi et al. [2009] climatology (including anthropogenic carbon uptake) interpolated onto the same grid used by the model (approximately $3^\circ \times 3^\circ$), (b) simulated MITgcm preindustrial CO$_2$ fluxes using (1), (c) zonally-averaged fluxes for the Takahashi et al. [2009] climatology (blue) and the model simulated preindustrial CO$_2$ fluxes (red) with ±1 standard deviation bounds to illustrate zonal variations and (d) zonally-averaged fluxes for the simulated MITgcm preindustrial CO$_2$ fluxes (red) and two reconstructions using the framework detailed in (8) with “online” advective and diffusive fluxes averaged into monthly fields (blue) and “offline” advective and diffusive fluxes calculated with monthly average velocity, diffusivity and scalar fields (orange). Again, ±1 standard deviation bounds are shown to illustrate zonal variations. Positive values indicate oceanic uptake while negative values indicate outgassing.

Figure 2: Component drivers of the air–sea flux of CO$_2$ (mol C m$^{-2}$ s$^{-1}$) for the flux due to (a) surface heat fluxes $\left(\gamma_\theta \frac{F_{heat}}{\rho C_p}\right)$, (b) changes in salinity driven by freshwater fluxes $\left(\gamma_S \frac{F_{W}}{\rho_{fw}}\right)$, (c) changes in alkalinity driven by freshwater fluxes $\left(\gamma_{AT} \frac{A_T F_{W}}{\rho_{fw}}\right)$, (d) changes in carbon driven by freshwater fluxes $\left(\gamma_{CT} \frac{C_T F_{W}}{\rho_{fw}}\right)$, (e) divergence of total $C_{res}$, and (f) biological activity (soft-tissue and carbonate terms, from divergence of nutrient concentration). Positive values indicate oceanic uptake from the atmosphere while negative values indicate outgassing from the ocean.

Figure 3: (a) reconstruction of the air–sea flux of CO$_2$ (mol C m$^{-2}$ s$^{-1}$) as the sum of the component drivers, the majority of which is accounted for by (b) the CO$_2$ flux due to surface heat fluxes, and (c) the balance of the divergence of total $C_{res}$ and the opposing flux due to biological activity. There is a negligible contribution from (d) the sum of CO$_2$.
fluxes due to changes in salinity, alkalinity and carbon concentrations that are all driven by net freshwater fluxes. Positive values indicate oceanic uptake from the atmosphere while negative values indicate outgassing from the ocean.

**Figure 4:** Area-integrated air–sea CO$_2$ fluxes (red) broken down into the CO$_2$ flux due to heat fluxes (yellow), biological activity (grey), the residual, C$_{res}$, component (light blue) and the total effect of freshwater fluxes (dark blue, the sum of salinity, alkalinity and $C_T$ changes) all in PgC yr$^{-1}$. Positive values indicate ocean uptake, while negative values indicate outgassing. The regions are primarily divided by whether they are a net source or a net sink of CO$_2$ as in Table 3, with ocean uptake of CO$_2$ occurring in the crosshatched regions, and further by the grey shaded areas of (a) the Northwest and Northeast regions of the Atlantic and Pacific, (b) the equatorial and subtropical Indian ocean, and the west and east regions of the Atlantic and Pacific, (c) the Indian, Pacific and Atlantic midlatitude sectors of the Southern Ocean (north of $\sim$60$^\circ$S), and (d) the East Antarctic, Ross Sea and Weddell Sea regions (south of $\sim$60$^\circ$S).

**Figure 5:** Upper ocean $C_T$ budget: $0 = \nabla \cdot (\bar{u} C_T) h + \nabla \cdot (\kappa \nabla C_T) h + R_{C_T,P} S_{bio} h + \frac{1}{2} S_{CaCO_3} h + \frac{E_{CO_2}}{\rho_{fw}} + F_{CO_2}$ (mol C m$^{-2}$ s$^{-1}$) comprising (a) the divergence of three dimensional advection of carbon, (b) divergence of horizontal and vertical diffusion of carbon, (c) biological activity (soft tissue and carbonate combined) directly calculated by the model, including biological community productivity, export fluxes and remineralization of dissolved and particulate organic and inorganic matter, (d) the effect of freshwater fluxes (precipitation or evaporation) on the concentration or dilution of surface $C_T$ concentrations, (e) the air–sea exchange of carbon dioxide using (1), and (f) the budget residual which is the sum of components (a) through (e). Positive values indicate in-
creasing carbon concentration tendency, while negative values indicate decreasing carbon concentration tendency. The location of the zero contour is plotted in black.

**Table 1:** Definition and description of terms in equations.

**Table 2:** Values of the linear solubility coefficients used in the attribution of saturated carbon changes. Coefficients were empirically diagnosed by calculating $C_{\text{sat}}$ over a range of values for temperature, salinity or alkalinity while holding the others (including atmospheric $\text{CO}_2$) at surface mean values [Lewis and Wallace, 1998; Goodwin and Lenton, 2009; Ito and Follows, 2013] and finding the gradient by linear regression.

**Table 3:** Area-integrated carbon flux drivers (PgC yr$^{-1}$). The regions are divided by whether they are a net source or a net sink of $\text{CO}_2$ or neutral with respect to the zero contour of the simulated net air–sea $\text{CO}_2$ flux (see figure 1b). Positive values indicate oceanic uptake of $\text{CO}_2$ from the atmosphere, while negative values indicate oceanic outgassing of $\text{CO}_2$ to the atmosphere.

**Table 4:** Area-integrated carbon flux drivers (PgC yr$^{-1}$). The regions are divided by whether they are a net source or a net sink of $\text{CO}_2$ or neutral with respect to the zero contour of the simulated net air–sea $\text{CO}_2$ flux and then by longitude (see shaded regions in Figure 4). Positive values indicate oceanic uptake of $\text{CO}_2$ from the atmosphere, while negative values indicate oceanic outgassing of $\text{CO}_2$ to the atmosphere.

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References


Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.

(a) advection, (b) diffusion, (c) biological activity, (d) freshwater flux, (e) CO₂ flux, (f) budget residual.
Table 1.

<table>
<thead>
<tr>
<th>Term</th>
<th>Units</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_{CO_2}$</td>
<td>mol C m$^{-2}$ s$^{-1}$</td>
<td>Air–sea flux of CO$_2$</td>
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<td>$K$</td>
<td>mol C m$^{-2}$ s$^{-1}$ µatm$^{-1}$</td>
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### Table 2.

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### Table 3.

<p>|                  | Global &amp; North Atlantic &amp; North Pacific &amp; Subtropical &amp; Southern Ocean | Southern Ocean |
|------------------|---------------------------------------------------------------|----------------|-----------------|----------------|----------------|
| Heat Fluxes      | 0.00                                                          | 1.02           | -2.27           | 1.00           | 0.26           |
| Salt Fluxes      | -0.00                                                         | 0.03           | -0.08           | 0.02           | 0.03           |
| Alkalinity Fluxes| -0.00                                                         | -0.24          | 0.73            | -0.19          | -0.29          |
| Freshwater Fluxes| 0.00                                                          | 0.26           | -0.77           | 0.21           | 0.31           |
| Net Freshwater Fluxes | 0.00                                                          | 0.04           | -0.13           | 0.03           | 0.05           |
| Residual Flux    | -6.63                                                         | -1.52          | -1.84           | -2.22          | -1.06          |
| Biological Activity | 6.79                                                        | 1.24           | 2.68            | 2.11           | 0.77           |
| Residual + Biological | 0.16                                                          | -0.29          | 0.84            | -0.11          | -0.29          |
| Sum of Components| 0.16                                                          | 0.78           | -1.56           | 0.92           | 0.02           |
| Net CO$_2$ Flux  | 0.00                                                          | 0.75           | -1.60           | 0.87           | -0.01          |</p>
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<tr>
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