Hypersensitivity of Southern Ocean air-sea carbon fluxes to background turbulent diapycnal mixing

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Abstract

The Southern Ocean (SO) is the worlds largest high nutrient low chlorophyll region and has a plentiful supply of underutilised macronutrients due to light and iron limitation. These macronutrients supply the rest of the neighboring ocean basins, and are hugely important for global productivity and ocean carbon sequestration. Vertical mixing rates in the SO are known to vary by an order of magnitude temporally and spatially, however there is great uncertainty in the parameterization of this mixing, including in the specification of a background mixing value in coarse resolutation Earth System Models. Using a biogeochemical-ocean model we show that SO biomass is highly sensitive to altering the background diapycnal mixing over short timescales. Increasing mixing enhances biomass by altering key biogeochemical and physical parameters. An increased surface supply of iron is responsible for biomass increases in most areas, demonstrating the importance of year round diapycnal fluxes of iron to SO surface waters. These changes to SO biomass could potentially alter atmospheric CO2 concentration over longer timescales, demonstrating the importance of accurate representation of diapycnal mixing in climate models.

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Key Points:

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8	•	Total air-sea carbon fluxes in the Southern Ocean are altered by up to 66% annually
9		by modest background mixing variations.
10	•	Resolving or skillfully parameterising the spatiotemporal variability of small-scale
11		turbulent mixing in the Southern Ocean is essential to model air-sea carbon fluxes.
12	•	The spatiotemporal coverage of available pCO_2 observations is insufficient for con-
13		straining the role of diapycnal mixing rates below the mixed layer on air-sea fluxes

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14 Abstract

The Southern Ocean (SO) connects major ocean basins and hosts large air-sea carbon fluxes 15 due to the resurfacing of deep nutrient and carbon-rich waters. While surface-intensified 16 wind-induced turbulent mixing in the SO surface mixed layer is significant for air-sea fluxes, 17 the orders-of-magnitude weaker background mixing below the mixed layer has not been 18 considered consequential. Topographically induced upward propagating lee waves in the 19 SO, wind-induced downward propagating waves generated at the base of the mixed layer, 20 shoaling of southward propagating internal tides generated in the basins north of the SO, and 21 turbulence under sea ice are among the processes known to induce upper ocean background 22 turbulence but typically are not represented in models. Here, we show that altering the 23 background mixing in the SO over a modest range can lead to a $\sim 40\%$ - 60% annual change in 24 SO air-sea CO_2 fluxes, with bigger changes on a seasonal timescale. This is primarily through 25 altering the temperature and the dissolved inorganic carbon and alkalinity distribution in 26 the surface water. Given the high spatiotemporal variability of processes that induce small-27 scale background mixing, this work demonstrates the importance of their representation in 28 climate models for accurate simulation of global biogeochemical cycles. 29

30 Introduction

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The Southern Ocean (SO), defined here as any region south of 30° S, is a key region 31 for the global carbon cycle due to the upwelling of deep, old, carbon and nutrient-enriched 32 waters, connecting the vast reservoir of nutrients and carbon from below the mixed layer 33 with the surface (Talley et al., 2016). The deep ocean interacts with the atmosphere through 34 less than 4% of the ocean's surface area (Watson & Naveira Garabato, 2006; Klocker, 2018), 35 with 65% of interior waters making first contact with the atmosphere in the SO (DeVries 36 & Primeau, 2011). As the deep ocean contains up to 60 times more carbon than the atmo-37 sphere (Arias et al., 2021), small perturbations to air-sea carbon fluxes can be important 38 for atmospheric carbon content (Adkins, 2013). Therefore, the SO, and especially the up-39 welling branch of circumpolar deep water (Marshall & Speer, 2012), is key in controlling 40 global biogeochemical cycles, the exchange of CO_2 between the atmosphere and the deep 41 ocean, atmospheric CO_2 levels, and the response of the ocean and atmosphere to climate 42 change (Sarmiento et al., 2004; Gruber et al., 2019). 43

Several expeditions have revealed strong cross-density (diapycnal) mixing due to smallscale ocean turbulence in the SO (Garabato et al., 2004; Ledwell et al., 2011; Watson et al., 2013; Garabato et al., 2019), though measurements remain sparse and difficult to scale
up (Tamsitt et al., 2018; Mashayek et al., 2017; Cael & Mashayek, 2021; Mashayek et al., 2022). Given the small scales of diapycnal mixing, it is not resolved in operational models, and so it is parameterised (Gaspar, Grégoris, & Lefevre, 1990; W. G. Large et al., 1994) in two forms:

- 1. Surface mixed layer mixing from storms and other surface winds, as well as convective instabilities.
- Background turbulence induced by bottom generated internal waves due to interaction
 of jets, eddies, and tides with rough topography or due to shoaling and breaking of
 remotely generated internal tides (see de Lavergne et al. (2020); Baker and Mashayek
 (2021, 2022) for reviews of such dynamics).

The 'background' mixing in the ocean interior is typically several orders of magnitude smaller than that in the surface mixed layers. Since the seminal work of Munk (1966), bulk measurements of ocean mixing have found a diapycnal turbulent diffusivity of $K_v \sim \mathcal{O}(10^{-4})$ $m^2 \,\mathrm{s}^{-1}$ required to resurface the abyssal waters and facilitate the closure of the meridional overturning circulation (MOC) (Ganachaud & Wunsch, 2000; Talley et al., 2003; Lumpkin & Speer, 2007; Talley, 2013), while estimates from profiling instruments often find $K_v \sim \mathcal{O}(10^{-5})m^2 \,\mathrm{s}^{-1}$ in the interior of the ocean and much larger values only very close to the seafloor (Waterhouse et al., 2014; Ferrari, 2014). In the Diapycnal and Isopycnal Mixing Experiment in the Southern Ocean (DIMES), estimates of mixing based on microstructure profiles reported $K_v \sim \mathcal{O}(10^{-5})m^2 \text{ s}^{-1}$ at the mean depth of an anthropogenic tracer released upstream of the Drake Passage. Meanwhile, the tracer itself appeared to experience $K_v \sim \mathcal{O}(10^{-4})m^2 \text{ s}^{-1}$ (Watson et al., 2013; Mashayek et al., 2017). The background values used in models typically lie within this range.

Though diapycnal mixing is highly temporally and spatially variable due to its gener-70 ating mechanisms (e.g. strong surface westerly winds and interaction of the currents and 71 72 eddies with rough topography), it is frequently parameterised as temporally invariable and, at times, even spatially constant. Current best estimates of SO diapycnal mixing are based 73 on 'static' maps, produced with numerous limiting assumptions, approximating the contri-74 butions from topographically generated lee waves (Nikurashin & Ferrari, 2011; Shakespeare, 75 2020), wind-induced near-inertial waves (Alford, 2020), and internal tides (de Lavergne et 76 al., 2020). These maps have formed the basis of our representation of such processes in 77 earth system models (A. Melet et al., 2014; A. V. Melet et al., 2022). 78

Diapycnal mixing in the global ocean interior is known to be an important factor in 79 variations in atmospheric carbon levels on centennial to millennial timescales via alterations 80 in ocean circulation (Sigman et al., 2010; Marinov & Gnanadesikan, 2011). Enhanced 81 diapycnal mixing increases deep ocean ventilation via the SO and reduces ocean carbon 82 storage through biological and solubility carbon pumps (Marinov et al., 2008; Marinov & 83 Gnanadesikan, 2011). Climate models are sensitive to the intensity and distribution of global 84 diapycnal mixing, accounting for about 25% of the uncertainty in the estimated range of 85 atmospheric CO_2 concentrations by 2100 (Schmittner et al., 2009). In this work, we are 86 concerned with the response of the air-sea carbon fluxes to mixing below the surface mixed 87 layer on time scales much shorter than those explored in the abovementioned works. We also 88 solely focus on mixing in the SO which has been assumed to be of secondary importance for 89 the global ocean circulation (Nikurashin & Ferrari, 2013) but significant for setting global 90 tracer distributions (Ellison et al., 2021). 91

Given the strong wind-driven isopycnal upwelling in the Southern Ocean, and the in-92 tense diapycnal mixing within the mixed layer induced by strong winds, one may imagine 93 that the modest background mixing below the mixed layer would be inconsequential for 94 setting the air-sea fluxes of CO_2 on short timescales. In this work, we show the contrary. 95 The air-sea flux of CO_2 primarily depends on the difference in the partial pressures of CO_2 96 (pCO_2) between the atmosphere and the ocean. Physical and biological processes, including 97 advective and diffusive transport of tracers, organic matter formations and sinking, and 98 the dilution of tracers due to precipitation, runoff and sea ice melt, all alter the pCO_2 of 99 surface waters (Mahadevan et al., 2011), resulting in high variability in time and space 100 of the CO_2 fluxes in the SO (Rosso et al., 2017). The influence of altering background 101 diapycnal mixing on the surface ocean pCO_2 is complex to predict due to spatiotemporal 102 variability in biological and physical responses to variations in mixing (Dutreuil et al., 2009), 103 and the coupled multivariate dependency of ocean pCO_2 to temperature, salinity, alkalinity 104 and dissolved inorganic carbon. In this work, we will show that modest perturbation of the 105 background mixing strongly alters the SO-integrated air-sea CO₂ fluxes on seasonal, annual, 106 and interannual timescales. 107

108 Experiment Setup

We use the Biogeochemical Southern Ocean state estimate (B-SOSE; Verdy and Mazloff (2017)). The interaction between diapycnal mixing below the mixed layer, the mixed layer dynamics, and the mesoscale processes that advect tracers prove essential for the problem under consideration here. Thus, employing an eddy-resolving model such as B-SOSE is essential and distinguishes this work from studies mentioned earlier that use low-resolution ¹¹⁴ models in global settings to study the role of mixing on carbon fluxes. In return, the ¹¹⁵ timescales of relevance in our study are much shorter than theirs.

The model's optimization procedure solves for adjustments to the prescribed atmo-116 spheric state and initial conditions to bring the solution into better consistency with obser-117 vations. The resulting product is a closed-budget model simulation representing the present 118 SO state. Because it represents the constantly evolving SO state it is not in a steady state, 119 in contrast to what one may expect from analysis of an earth system model undergoing a 120 long spin-up process. The B-SOSE simulation provides an estimate of SO biogeochemistry, 121 122 including air-sea fluxes of heat and carbon and the cycling of nutrients from a biogeochemical and carbon model. 123

The model domain extends from the equator to 78° S with 52 vertical layers of varying 124 thickness. The zonal grid spacing is always $1/6^{\circ}$, but the meridional grid spacing changes 125 with the cosine of the latitude. Ocean physics is represented using the MITgcm. The ocean 126 is forced through an atmospheric boundary layer scheme where bulk formulae determine 127 heat fluxes, freshwater (salt), and momentum (W. Large & Yeager, 2009). The hourly 128 atmospheric conditions of ERA5 (Dee et al., 2011) are applied, with adjustments added 129 that were determined by optimization (Verdy & Mazloff, 2017) using the adjoint method. 130 Sea ice is modelled using 0-layer thermodynamics (Fenty & Heimbach, 2013) and an implicit 131 Line Successive Over Relaxation (LSOR) dynamical solver (Losch et al., 2010). A horizontal 132 biharmonic diffusivity is used with a value of 10^{-8} m⁴s⁻¹. Implicit vertical diffusivity for 133 convection is set to 10 $m^2 s^{-1}$, and no mesoscale eddy parameterisation was implemented. 134 For further details, see Verdy and Mazloff (2017) and Swierczek et al. (2021). 135

The parameterisation of diapycnal mixing is composed of two parts. The GGL90 mixed 136 layer parameterisation of Gaspar, Grégoris, and Lefevre (1990) is used to represent param-137 eterised turbulence and is highly surface-enhanced (Fig.1 D), inducing strong turbulence 138 under the seasonal atmospheric storm tracks, mixing the DIC gradients in the upper few 139 hundreds of meters. In other places, such as under the ice or when there is no strong 140 wind-induced turbulence, the models rely on a prescribed background value for turbulent 141 diffusivity. The background value is behind the sensitivity of fluxes discussed in this work. 142 As discussed in subsequent paragraphs, the background vertical diffusivity is altered in each 143 experimental run. The background mixing value is added to the GGL90 mixing to achieve 144 a total vertical diapycnal mixing value. 145

The Biogeochemistry with LIght, Nutrients and Gases (NBLING) model, described 146 fully in Verdy and Mazloff (2017), forms the biochemical framework within B-SOSE. The 147 original BLING model described in Galbraith et al. (2010) was modified (for B-SOSE) with 148 the addition of nitrogen cycling and improvements in the representation of phytoplankton 149 population dynamics and particle export (Galbraith et al., 2015; Bianchi et al., 2013). Bi-150 ological activity influences the concentrations of carbon and oxygen. At the core of the 151 BLING model is primary production with limitations by light, nitrate, phosphate, iron, and 152 temperature, and subsequent remineralization of organic matter back to inorganic nutri-153 ents. Nine prognostic tracers are simulated in NBLING: dissolved inorganic carbon (DIC), 154 alkalinity (ALK), oxygen (O₂), nitrate (NO₃), phosphate (PO₄), iron (Fe), dissolved or-155 ganic nitrogen (DON), dissolved organic phosphate (DOP) and phytoplankton biomass. The 156 biomass partitioning into species is determined using stored ratios updated every timestep 157 based on growth and decay rates. We include three types of phytoplankton: large, small, 158 and diazotrophs. Small cells represent calcifying organisms; they use calcium carbonate 159 to form shells. Diazotrophs can fix nitrogen, so nitrate availability does not limit them. 160 Phytoplankton loss is expressed as a power law with a size-dependent exponent based on 161 162 (Dunne et al., 2005).

The B-SOSE carbon system is adapted from the MITgcm simple biogeochemical model of Dutkiewicz et al. (2006). DIC and ALK are prognostic variables, and pH and pCO_2 are diagnosed based on Follows et al. (2006), making oceanic pCO_2 a function of DIC, ALK, temperature (T), salinity (S) and silica, where silica is prescribed from the 2013 World Ocean Atlas climatology (Garcia et al., 2013). CO_2 and oxygen air-sea fluxes are calculated following Wanninkhof (1992). Atmospheric pCO_2 is prescribed using values from the CarbonTracker product (Peters et al., 2007).

The state estimate is produced by solving for the model initial and boundary con-170 ditions, the so-called model 'controls', which minimize a weighted least squares sum of 171 model-observation misfits. This is achieved using an adjoint model that provides the gradi-172 ents of the misfit function with respect to the model controls, allowing those controls to be 173 174 efficiently and systematically determined. The model is run for seven years (2012 - 2018)using the adjoint method-based assimilation product. This includes physical and biogeo-175 chemical constraints obtained from Argo floats, including biogeochemical parameters from 176 the SOCCOM float array, satellite altimetry, satellite SST, and ship transect data. The full 177 set of model parameters used in this $1/6^{\circ}$ setup is given in Swierczek et al. (2021); see Verdy 178 and Mazloff (2017) for initial conditions. 179

The B-SOSE's original conditions and atmospheric adjustments were obtained using 180 $10^{-4} m^2 s^{-1}$ as the value for background diapychal diffusivity (added to the GGL90 mixing). 181 We refer to that base simulation as Ex1e-4 hereafter. Using the same initial conditions and 182 atmospheric state adjustments, two additional model simulations were carried out for 2013-183 2018. The first perturbation experiment, Ex1e-5, uses a constant background diffusivity 184 value of $10^{-5}m^2s^{-1}$. The range $10^{-5}m^2s^{-1}$ to $10^{-4}m^2s^{-1}$ is conservative, sandwiched 185 between the two canonical paradigms of mixing often considered in Physical Oceanography. 186 The third experiment, ExVar, uses a spatially variable (but temporally constant) vertical 187 diapycnal mixing map (Fig 1A-C). The map is constructed as the sum of contributions from 188 tides (de Lavergne et al., 2020) and topographically-generated lee waves (Nikurashin & 189 Ferrari, 2013). ExVar features horizontal and vertical variations over a range much broader 190 than 10^{-5} $m^2 s^{-1}$ to 10^{-4} $m^2 s^{-1}$. Although background mixing values dominate over the 191 GGL90 parameterisation in mid-depths, GGL90 is orders of magnitude larger than the 192 background mixing value in the upper ocean (Fig.1D). The three cases together allow for 193 evaluating the impact of the magnitude of mixing and its spatial variations on the carbon 194 flux independently. 195

196 **Results**

¹⁹⁷ Carbon fluxes

The SO is a net sink of atmospheric CO_2 , with most of the uptake occurring between 198 50° S and 30° S, with a peak at 40° S, where around 7 Tg C/yr is taken up (Fig.2A). 40° S 199 is the average latitude of the subtropical front, separating the subtropical waters from the 200 subantarctic mode waters, thus hosting rich mesoscale and submesoscale frontal dynamics 201 and enhanced air-sea exchange of tracers. To the south of the polar front (panel E), on the 202 other hand, the upwelling of deep carbon-rich waters causes carbon outgassing (shown in 203 red panel E). Additional uptake occurs further south around Antarctica due to downwelling 204 (induced by a change in the wind direction from westerly to easterly) and deep water forma-205 tion. Thus, SO fronts, which mark sharp gradients in temperature and carbon chemistry, 206 separate regions of net uptake from regions of outgassing. Higher latitudes show very low 207 mean annual carbon fluxes, partly due to seasonal ice cover. 208

Carbon uptake varies year-on-year during the six years of the state estimate run by almost 2 Tg C/yr at some latitudes, with especially high inter-annual variability at 60°S and 40°S (Fig.2A). The inter-annual range of carbon fluxes for Ex1e-5 are highly nonmonotonic. The inter-annual variations are due to varying oceanic conditions each year, some of which are associated with the Southern Annular Mode (SAM).

Alterations to background diapycnal mixing alter SO carbon fluxes, with ExVar showing smaller differences from Ex1e-5 than Ex1e-4 (Fig.2B dashed vs solid lines). The sensitivity

to altered diapycnal mixing is variable throughout the six years (Fig.2 B). This inter-annual 216 range in sensitivity of around 0.1 Tg C/yr is well within the range of the inter-annual 217 variability of zonally integrated carbon fluxes in Ex1e-5 (Fig.2A,B). A higher difference 218 between Ex1e-5 and Ex1e-4, and to a lesser extent between Ex1e-5 and ExVar occurs in the 219 first three years (2013 to 2015) than in the last three years (2016 to 2018) (Fig.2B). As upper 220 ocean mixing is never in an equilibrium state due to constantly changing winds, eddies and 221 buoyancy fluxes, the response to the mixing perturbation over the first few months of this 222 experiment do not seem unrealistically exaggerated. 223

224 Increasing the background mixing from Ex1e-5 to Ex1e-4 leads to a reduction in annual mean zonally integrated carbon uptake at all latitudes (Fig.2C,F). The most significant 225 reduction is around 55°S (Fig.2C,F). Minor changes occur south of 65°S due to the ice cover. 226 In ExVar, the most significant changes from Ex1e-5 occur further north, at around 45°S. 227 South of 60°S, the difference between ExVar and Ex1e-5 is insignificant (Fig.2C,G). Large 228 areas of ExVar have carbon fluxes unchanged from those in Ex1e-5 (Fig.2G), suggesting 229 that $10^{-4}m^2$ is likely too large of a background mixing value. In Ex1e-5, the mean annual 230 cumulative net flux of carbon into the ocean, integrated from $75^{\circ}S$ northward to $30^{\circ}S$, is 231 1 Pg C/yr (Fig.2D). In Ex1e-4 only 0.6 Pg C/yr is taken up, a reduction of 0.4 Pg C 232 yr^{-1} . The annual uptake of ExVar falls between the other two experiments at around 0.8 233 Pg C/yr. These numbers are for the six-year mean, and as panel B shows, the reductions 234 from Ex1e-5 are much higher over the first three years (almost double). 235

The cumulative carbon fluxes are compared to other estimates of the integrated SO 236 carbon flux from 75° S northward to up to 45° S and 35° S for the period 2015-2017 (Fig.2D; 237 Bushinsky et al. (2019); Landschützer et al. (2016); Rödenbeck et al. (2013)). At 45°S, 238 the Ex1e-5 cumulative flux lies between the three observationally inferred estimates, while 239 the ExVar flux is slightly lower, with Ex1e-4 being the lowest of all three, well below the 240 estimates of the other studies. At 35° S, there is a larger range in carbon uptake between 241 the three model runs. Ex1e-5 is the only experiment that lies within the bounds of the three 242 observational estimates, though it appears towards the lower end, whilst ExVar and Ex1e-4 243 are below. This further suggests that the lower mixing in Ex1e-5 could be a more suitable 244 background mixing value. 245

²⁴⁶ Changes to surface ocean pCO_2

Given that ExVar estimates of carbon fluxes fall between those of Ex1e-4 and Ex1e-5, 247 hereafter, we only focus on the differences between Ex1e-4 and Ex1e-5. Air-sea carbon fluxes 248 exist due to the difference in pCO_2 between the atmosphere and the surface ocean. The 249 high (low) surface ocean pCO_2 values result in regions of low (high) oceanic uptake or even 250 outgassing of CO_2 from the atmosphere (Fig.3A). A region of exception is under sea ice, 251 where the diffusive flux of gases is prevented. The changes in carbon fluxes due to altered 252 mixing, as in figure 2, are due to changes in surface ocean pCO_2 , as atmospheric conditions 253 are constant across experiments. 254

The annual mean pCO_2 of the surface ocean is higher in Ex1e-4 than in Ex1e-5 in almost all regions, reducing the pCO_2 gradient and carbon uptake (Fig.3B). The areas of greatest increase in pCO_2 include south of South Africa and the waters east of the West Antarctic Peninsula. Small regions where the annual mean pCO_2 is reduced in Ex1e-4 include latitudes of around 30°S, especially to the east of Australia, the Argentine basin, and a few small bands just off the coast of Antarctica in the south.

Using the methodology set out by Takahashi et al. (2014), we break down the pCO_2 change to contributions from changes in the upper ocean content of salinity, temperature, DIC and alkalinity (equation 1). The change in pCO_2 as a contribution from each of the four tracers is calculated using equations 2-5, where $\bar{p}CO_2$ is the mean pCO_2 , $\bar{A}lk$ is the mean alkalinity, γ_{CO_2} is the Revelle factor for CO_2 (value used = 11), and γ_{ALK} is the Revelle factor for alkalinity (value used = -10).

$$\Delta pCO_2 = \left(\frac{\delta pCO_2}{\delta T}\right)\Delta T + \left(\frac{\delta pCO_2}{\delta DIC}\right)\Delta DIC + \left(\frac{\delta pCO_2}{\delta Alk}\right)\Delta Alk + \left(\frac{\delta pCO_2}{\delta S}\right)\Delta S \quad (1)$$

$$\frac{\delta p CO_2}{\delta T} \Delta T = 2(p CO_2) [Exp(0.0423(\pm 0.0002)\Delta T/2) - 1]$$
(2)

$$\frac{\delta p C O_2}{\delta D I C}) = \gamma_{CO_2} (\bar{p} C O_2 / \bar{T} C O_2) \tag{3}$$

$$\frac{\delta p C O_2}{\delta A l k} = \gamma_{ALK} \left(\frac{\bar{p} C O_2}{\bar{A} l k} \right) \tag{4}$$

$$\left(\frac{\delta p C O_2}{\delta S}\right) = 0.026(\pm 0.002) \cdot \bar{p} C O_2 \tag{5}$$

The four individual contributions, shown in Fig.3D-G, can be summed together to give 267 the annual mean approximated change in pCO_2 (Fig.3C). This calculated change agrees 268 satisfactorily with the changes in pCO_2 between the two experiments (Fig.3B). This verifies 269 the assumptions made in equations 2-5, and confirms that changes to the distribution of 270 these tracers are key in causing changes to carbon fluxes (Fig.3B,C). The only region where 271 the Takahashi et al. method does not capture the changes is in the north of the SO, west 272 of New Zealand and east of South America in the Argentine basin. This is likely due to 273 enhanced water mass mixing occurring in these regions, making changes in this area complex 274 to approximate with simple assumptions. While the calculations shown in Fig. 3 use the 275 upper 2.6 m of the water column, they are not sensitive to depth and similar results are 276 found down to ~ 55 m. 277

On an annual basis, contributions from changes in DIC and alkalinity concentrations are 278 the main drivers of changes in pCO_2 , with the contributions from salinity and temperature 279 being secondary (Fig.3E,F). An increase in the alkalinity content decreases pCO_2 , whilst 280 an increase in salinity or DIC increases pCO_2 . Where the temperature increases, pCO_2 281 increases due to modulation of the equilibrium DIC. The increase in the DIC content of 282 the surface waters of the southern SO in Ex1e-4 increases pCO_2 , whilst in the north the 283 decrease in DIC concentration decreases pCO_2 . On the contrary, the increase in alkalinity 284 concentration in the south decreases pCO_2 , while the decrease in alkalinity in the north 285 increases pCO_2 . Changes in salinity concentrations act to slightly increase the pCO_2 in 286 Ex1e-4. Temperature changes with enhanced mixing cause a slight decrease in pCO_2 in the 287 north and an increase in pCO_2 in the south in Ex1e-4. 288

Changes in the upper ocean temperature, salinity, DIC and alkalinity are due to alter-289 ations to the diapycnal flux of these tracers. The diapycnal flux for a tracer with concentra-tion C may be approximated by $-K_v \times \frac{\partial C}{\partial z}$. Therefore, if vertical diapycnal mixing K_v is increased, more tracer, e.g. DIC, is mixed downgradient (upward into the surface waters). 290 291 292 This increase in upward flux is the strongest where the vertical gradients are the strongest. 293 Therefore, strong correlations develop between locations with sharp vertical gradients and 294 locations with significantly altered tracer content with enhanced mixing (Fig.4A-D). This 295 correlation is especially clear when examining changes to tracer distributions in the first 296 month of the perturbation experiments, as shown in figure 4. 297

Regions that experience high changes in DIC concentration with enhanced mixing are 298 around the coast of Antarctica, south of 60° S and above depths of 40 m. In these areas, 299 surface waters are fed by wind-induced upwelling of deep waters rich in DIC due to the 300 respiration of organic material. Further to the north, the upper 120 m of the water column 301 has weak vertical gradients of DIC concentration (Fig.4E,F). The dipole pattern shown 302 when looking in a zonal average sense implies the erosion of the sharp gradient by enhanced 303 mixing. The DIC concentration increases with increased mixing in the upper surface waters 304 (shown in red), while concentrations decrease between 40m and 20m depth (shown in blue) 305 due to a flux divergence, as more of this carbon has been mixed upwards into the surface 306 waters (Fig.4G). There is a clear divide at around 20 m; this depth corresponds to the 307 depth of the maximum vertical gradient. Changes in alkalinity and salinity roughly follow 308

³⁰⁹ a pattern similar to DIC (hence not shown). The greatest changes in temperature occur in ³¹⁰ different regions, mainly in the northern SO, especially at around 90°E, in the Argentine ³¹¹ basin, and in the waters surrounding New Zealand.

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Temporal and seasonal variability of changes in pCO_2

Carbon fluxes show strong seasonal and spatial variations (Fig.5), as for example, dis-313 cussed in Rosso et al. (2017). In the austral summer (December to February), the SO from 314 60° S to 30° S is a net source of CO₂ outgassing (Fig.5A,B Dashed lines). Some outgassing 315 also occurs at the upwelling zone of the polar front, especially in the Atlantic basin (Fig.5C). 316 South of $\sim 60^{\circ}$ S, the SO acts as a slight carbon sink even in summer. Austral winter (June 317 to August) has much higher carbon uptake than summer, with the net uptake occurring in 318 almost all regions of the SO, except beneath sea ice (Fig.5A,B,D). Small outgassing regions 319 exist around the polar front and at the upwelling region off the west coast of South America 320 in the Argentine basin (Fig.5D). 321

While figures 2 and 3 show the changes to carbon fluxes between model runs in an 322 annual mean sense, there are also significant temporal patterns in how mixing perturbation 323 alters carbon fluxes (as will be discussed in Fig.8). Figures 5 and 6 examine the dominant 324 mechanisms for seasonal differences observed in the changes to pCO_2 between Ex1e-4, ExVar 325 and Ex1e-5. The changes in carbon fluxes between experiments are greater in winter than 326 in summer (Fig.5A,E,F). An exception is in the very south, where ice cover during winter 327 reduces gas exchange in all experiments. In winter, Ex1e-4 has a reduced carbon uptake 328 compared to Ex1e-5, while ExVar has a similar carbon uptake to Ex1e-5 (Fig.5A). Cumu-329 latively integrated winter carbon fluxes are reduced from almost 2 Pg C/yr in Ex1e-5 to 330 1.2 Pg C/yr in Ex1e-4. The greatest decreases in uptake occur around 40°S. The Argentine 331 basin is also a region of pronounced diminished carbon uptake (Fig.5F). Three small areas 332 on the edge of the winter ice extent experience increased carbon uptake in the winter due to 333 reduced ice cover, the reason for which is explained subsequently (Blue areas, Fig.5F). Sum-334 mer changes to carbon fluxes are of a smaller magnitude and show more spatial variability 335 than the winter months (Fig.5A,E). In summer, the cumulative intergrated outgassing of 336 Ex1e-5 is higher than Ex1e-4, and ExVar is higher than both Ex1e-4 and Ex1e-5, though 337 the difference between all three runs is less than 0.2 Pg C/yr. At lower latitudes where the 338 SO is a net source of carbon to the atmosphere, outgassing is decreased in Ex1e-4. Further 339 south, where the SO is a carbon sink, CO_2 uptake is reduced in Ex1e-4 (Fig.5E). Changes 340 in flux occur as far south as the Antarctic continent due to diminished summer sea ice. 341

Using Eqs.1-5 and Fig. 3, we next use the Takahashi et al. methodology to examine 342 seasonal changes to tracer contributions and their implications for the pCO_2 and carbon 343 fluxes. The outcome is shown in Fig. 6. Salinity contributions to changes to pCO_2 are not 344 shown as they are negligible compared to DIC, alkalinity, and temperature contributions. 345 The temperature contribution varies greatly between seasons, being stronger in January 346 than in July (Fig.6B,E). Closer to Antarctica, changes in temperature increase the pCO_2 347 of Ex1e-4 surface waters throughout the year. In July, this positive contribution extends 348 further north. In January, the change in temperature causes very strong reductions in 349 pCO_2 , especially in the subtropical gyres. Because the change in surface temperature and 350 associated change to pCO_2 vary with season, the annual mean change in temperature and 351 its contribution to change in pCO_2 appear much smaller (Fig.3D). They are nevertheless 352 key to driving the seasonal response of changing SO carbon fluxes in response to altered 353 diapycnal mixing. 354

The vertical structure of the thermocline and the associated change in surface temperature with enhanced mixing have seasonal trends (Fig.7B,F). In January, surface waters are warm, and the temperature declines rapidly with depth down to 100 m, especially north of 60°S (Fig.7A). South of 60°S and below 100 m, water temperature increases with depth due to the upwelling of deep warm waters of North-Atlantic origin through Ekman transport.

In Ex1e-4, subsurface cooler waters are mixed more strongly towards the surface, cooling 360 the surface waters and warming the subsurface waters relative to Ex1e-5 (Fig.7B). This 361 surface cooling reduces the pCO_2 (Fig.6A). In July the surface waters are well mixed and 362 there is no temperature gradient in the upper 100 m (Fig.7E). Below the winter mixed 363 layer, temperature rises with depth. Enhanced mixing warms surface waters, increasing the 364 pCO_2 (Fig.6E,F). This increase in surface temperature also increases the rate of sea ice melt, 365 reducing the sea ice cover toward the end of winter/ spring in Ex1e-4. This results in small 366 regions of increased carbon uptake around the sea ice edge in winter (Fig.5F). North of 60° S 367 and below the mixed layer, waters still decrease in temperature with depth, so increased 368 mixing cools the surface waters (Fig.7E,F). The vertical gradients of DIC and alkalinity are 369 relatively constant regardless of season (Fig.7C,G). Changes in DIC and alkalinity concen-370 trations have opposing effects on pCO_2 (Fig.3E,F), but together act to increase pCO_2 at all 371 latitudes in summer and winter in Ex1e-4 (Fig.6C,F). The increase to pCO_2 in Ex1e-4 from 372 combined carbonate chemistry changes is stronger in the winter than in summer, especially 373 north of 40° S (Fig.6C,F). 374

Though all six years of the model run exhibit a similar seasonal cycle of changes to 375 carbon fluxes with enhanced mixing in Ex1e-4, important inter-annual differences exist 376 (Fig.8) which would not be appreciated in annual and seasonal means. Some differences in 377 carbon fluxes between the two experiments become more pronounced over time, while other 378 changes become less. North of 40°S, Ex1e-4 has an increase in carbon uptake (or reduced 379 outgassing) during the summer. This becomes more pronounced and extends further south 380 down to 50° S in subsequent summers as the model run progresses. While the winter time 381 reductions in uptake in Ex1e-4 compared to Ex1e-5 around $45^{\circ}S$ become stronger through 382 the six years, the reductions in carbon uptake south of 60°S become weaker. The SO yearly 383 mean change in C flux (red stars, Fig.8B) show a smaller mean change in the carbon flux 384 between Ex1e-4 and Ex1e-5 in the later years of the run compared to earlier years. This is 385 due to opposing signs of change to carbon fluxes over the seasons becoming more pronounced 386 and therefore causing an antagonistic net effect to changes in an annual mean sense. 387

 40° S approximately corresponds to the mean latitude of the subtropical front (STF) and marks a regime in terms of the leading mechanisms responsible for changes in pCO_2 and carbon fluxes. This marks the boundary between the nutrient deplete sub-tropical waters to the north and the nutrient and DIC rich waters to the south (Chapman et al., 2020). Regions to the north of this divide are responsible for the summer increases in carbon uptake in Ex1e-4 in later years of the run. In contrast, regions to the south are responsible for the strong response of reduced carbon uptake in Ex1e-4.

The contributions to the total change in pCO_2 are drastically different across the STF 395 (Fig.8C-E). To its South, opposite changes in pCO_2 due to alkalinity and DIC nearly bal-396 ance, with the latter being slightly larger (Fig.8D). As before, salinity contributions remain 397 negligible at all times. Over the first two years of the perturbation, the total change in pCO_2 398 is positive, meaning pCO_2 is higher in Ex1e-4 than in Ex1e-5. The magnitude of the reduc-399 tion in pCO_2 due to Alkalinity increases over time. The magnitude of total pCO_2 change 400 decreases over time and becomes negative in the summer months, allowing for increased 401 carbon uptake. 402

North of the STF, DIC and alkalinity do not balance each other out (Fig.8E) and changes in temperature between the two model runs are more dominant. Alkalinity increases pCO_2 north of the STF, while changes in DIC initially also increase the pCO_2 in Ex1e-4. By the summer of the third year of the run, changes in DIC begin to reduce the pCO_2 , causing the net total change in pCO_2 to be negative in summer. This causes an increase in carbon uptake in the summer north of the STF in Ex1e-4.

Almost all changes in DIC between Ex1e-4 and Ex1e-5 are due to altered diapycnal fluxes of DIC. However, north of the STF, DIC contribution to decreased pCO_2 in the summer is due to increased productivity in the nutrient depleted waters of the sub tropical

gyre. This increase in productivity does not occur instantaneously, but instead takes around 412 6 months to begin decreasing the DIC contribution (red line, Fig.8E and Fig.9C). While an 413 increase in productivity occurs with higher mixing across the whole SO region, it is only in 414 the north, roughly north of the sub tropical front where increased phytoplankton production 415 and DIC uptake becomes the dominant mechanism in altering DIC concentrations. Thus, 416 in the context of this paper, we consider the STF as the upper boundary of the SO and 417 postpone further discussion on the biologically-dominated change in pCO_2 north of the SO 418 to future work. 419

$_{420}$ Comparison to pCO_2 observational data

The pCO_2 values for Ex1e-4, Ex1e-5 and ExVar can be compared to 2013-2018 observed levels from the Surface Ocean CO₂ Atlas (SOCAT) (Bakker et al., 2016) (Fig.10). Neither clearly matches SOCAT observations better than the other (Fig.10A). Regional trends are also unclear, although from the limited data available, Ex1e-5 appears to better represent the pCO_2 of the northern Pacific Ocean, as well as off the coast of South Africa and Tasmania. Meanwhile estimates from Ex1e-4 are better matched to observations in the western Atlantic and the northern Indian Oceans.

The probability density function for the difference between SOCAT and B-SOSE for the 428 three experiments is broken down over seasons (Fig.10B). In summer the standard deviation 429 of differences between ExVar and data is much larger than those for Ex1e-4 and Ex1e-5. The 430 mean difference of 15.5 μ atm for Ex1e-5 is lower than 17.46 μ atm for Ex1e-4, whilst ExVar 431 has the lowest mean difference from observations. The high-end tails of the distributions 432 are more skewed than the lower ends, implying a systematic over-estimate of pCO_2 by 433 B-SOSE. B-SOSE overestimates the flux of carbon from the ocean to the atmosphere or 434 underestimates the SO carbon uptake from the atmosphere, particularly in the summer. 435

SOCAT data is heavily biased towards summer data due to limitations on data col-436 lection in the winter. The mean difference between SOCAT and B-SOSE is lower for the 437 winter mean than for the summer in all experiments. In the winter, ExVar has the largest 438 mean difference from observations but also the largest standard deviation. In an annual 439 mean sense, ExVar does a better job in matching SOCAT observations, though with a much 440 higher standard deviation. It is interesting to note that, while in the annual mean sense, 441 442 ExVar better matches SOCAT observations of pCO_2 , Ex1e-5 was better able to replicate previous observational estimates of cumulative SO C fluxes (Fig.2D). 443

444 Conclusion

We have demonstrated that the air-sea carbon fluxes in the SO are highly sensitive to modest background mixing variations well within the range of our best estimates. This is despite background mixing rates being orders of magnitude smaller than mixed layer model generated mixing. We find that the overall changes to carbon fluxes depend on the interactive effects of changes to DIC, temperature, and alkalinity, which can compensate or reinforce each other, and the predominant driver varies regionally, seasonally and temporally as additive and opposing feedbacks kick in at varying time scales.

The relevance of diapycnal mixing in setting global carbon fluxes has previously been considered to be through changes to the underlying stratification and of regional and global overturning circulation and ventilation patterns. Although that may be true on centennial or longer timescales, here we show that on much faster timescales mixing directly acts upon tracers such as DIC, alkalinity, temperature, and salinity leading to a significant change in surface ocean carbon fluxes.

The high correlation found between vertical gradients and strong changes in tracer distributions with altered mixing shows that on a timescale of days to months, direct changes in diapycnal mixing fluxes are the predominant drivers of the pCO_2 response in the SO. On longer timescales, from months to years, further feedbacks involving changes to biological productivity and mixed layer depth will also begin to cause further changes to the surface ocean pCO_2 . A latitudinal divide exists at around 40°S, roughly the location of the subtropical front. High vertical tracer gradients cause the direct impact of altered tracer fluxes to dominate trends to the south, whilst changes in biological productivity play a key role in the observed changes to carbon fluxes to the north.

Two major issues stand in the way of better constraining of the data-assimilating ocean 467 estimates insofar as the role of vertical mixing in the upper ocean is concerned. First, despite the significant investments in observations such as SOCAT, Fig. 10A clearly shows the 469 sparsity of the available observational data. From a statistical perspective, this coverage 470 is insufficient to discern which background mixing value better represents the real ocean 471 despite the strong impact of these choices on pCO_2 . This issue can be resolved only through 472 sustained observations. Knowledge of the seasonal cycle of pCO_2 is worse in the SO than in 473 most other regions of the ocean. The strong seasonality of the sensitivity of carbon fluxes 474 to altered mixing demonstrates the importance of year-round observations. Second, SO 475 diapychal mixing can vary by orders of magnitude over timescales ranging from hourly to 476 seasonally, as well as varying spatially. To achieve a close agreement with observations, a 477 model should have a representation of such variability. ExVar employed our best estimate 478 of a time-mean spatially variable mixing map, resulting in carbon fluxes similar to that 479 obtained with a constant diffusivity of $10^{-5}m^2/s$. Direct observations of diapycnal mixing 480 in the SO have suggested that such maps (a) lack the representation of many key processes 481 that result in higher turbulence in upper surface waters (e.g. bottom-generated lee waves, 482 shoaling of remotely generated internal tides) and (b) do not allow for co-variance of mixing 483 and tracer gradients, key to biological processes. This work highlights the absolute necessity 484 for climate models to resolve the spatio-temporal variability of small-scale turbulent mixing, 485 or to skillfully parameterise the processes responsible for generating them. 486

⁴⁸⁷ 1 Data and material availability

The data sets generated during and/or analysed during the current study are available from the corresponding author on reasonable request and from http://sose.ucsd.edu.

The data used to construct the spatially variable mixing map used for ExVar is available at https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2020MS002065.

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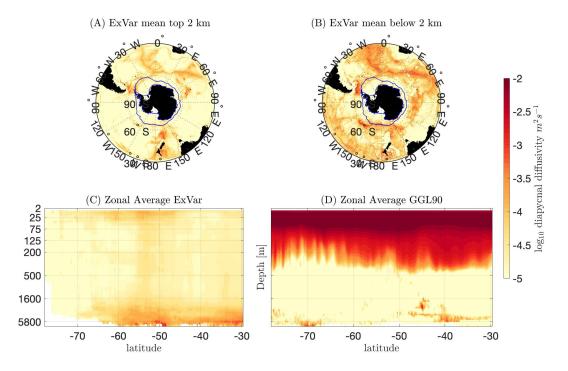


Figure 1: Diapycnal mixing in the Southern Ocean State Estimate (SOSE). The distribution of diapycnal mixing in the Southern ocean, constructed as the sum of contributions from tides and topographically-generated lee waves. This mixing is shown averaged in depth over the top/bottom 2km in panel A/B, and zonally over the Southern Ocean in C. These maps are used in the spatially variable mixing map experiment (ExVar). For reference, a zonally-averaged map of the storm-induced mixing, as parameterised through GGL90 parameterisation in B-SOSE, is also shown in panel D.

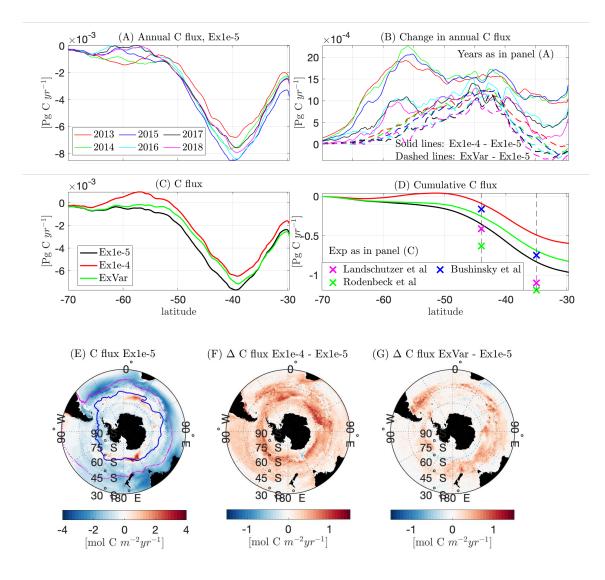


Figure 2: (A) Zonally integrated flux of Carbon for each year of Ex1e-5 (negative = Carbon flux from atmosphere to ocean). (B) Difference in the zonally integrated flux of Carbon between Ex1e-4 and Ex1e-5 (solid lines) and ExVar and Ex1e-5 (dashed lines) for each year of the experiment. (C) zonally integrated annual mean (2013-2018) Carbon flux for Ex1e-4, Ex1e-5 and ExVar. (D) Annual mean (averaged over 2013 to 2018) cumulative integral of carbon fluxes from 70°S northward to 30°S (legend same as the previous panel). Observational markers are included for comparison (Landschützer et al., 2016; Bushinsky et al., 2019; Rödenbeck et al., 2013). (E) Average annual carbon flux for Ex1e-5, the blue line shows the Polar Front, the magenta line shows Sub-tropical Front as defined by Orsi et al. (1995). (F) Annual mean change in Carbon flux (Ex1e-4 – Ex1e-5). (G) Annual mean change in Carbon flux (ExVar – Ex1e-5). Positive values imply reduced carbon uptake or increased outgassing.

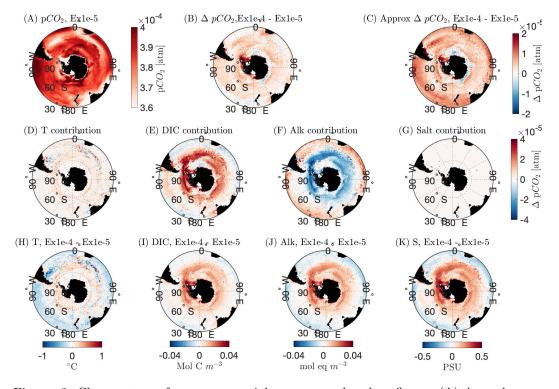


Figure 3: Changes to surface ocean partial pressure and carbon fluxes. (A) Annual mean surface ocean pCO_2 in Ex1e-5. (B) Change in pCO_2 between Ex1e-4 and Ex1e-5. (C) Same as panel B, but this time changes to pCO_2 approximated based on the methodology of Takahashi et al. (2014) that breaks down the change into various contributions as per equations (1-5). The various contributions are shown in panels D-G. (H-K) Changes in annual mean DIC, alkalinity, potential temperature and salinity.

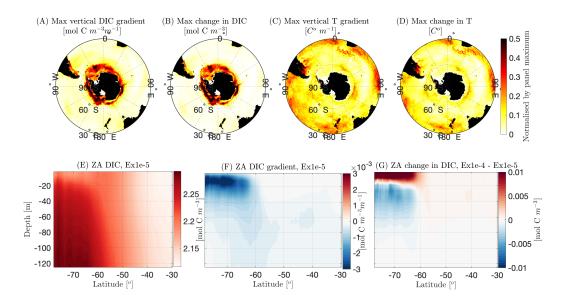


Figure 4: (A) Maximum vertical DIC gradient in the water column for Ex1e-5, normalised by maximum contour value. (B) Maximum change in DIC between Ex1e-4 and Ex1e-5, normalised by the maximum contour value. (C,D) Same as A and B but for temperature. The maximum change in DIC (temperature) is defined as the greatest difference in DIC (temperature) concentration between the two experiments at any depth above 200 m at each latitude and longitude in the domain. (E) Zonal average DIC concentration in Ex1e-5. (F) Zonal average DIC vertical gradient in Ex1e-5; blue indicates a decrease in concentration towards the surface. (G) Zonally averaged change in DIC concentration (Ex1e-4 - Ex1e-5). All shown for December 2012, the first month of all experiments.

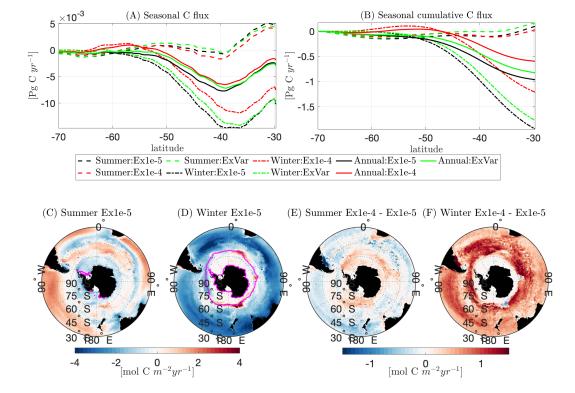


Figure 5: (A) zonally integrated Carbon flux for Ex1e-4, Ex1e-5 and ExVar for summer (dashed), winter (Dotted), and annual mean (solid line). (B) Cumulative sum of carbon fluxes from 70°S northward to 30°S (legend same as the previous panel). (C) Average summer carbon flux for Ex1e-5; magenta lines show the minimum summer ice extent. (D) Average winter carbon flux for Ex1e-5; magenta lines show the maximum winter ice extent. (E) Mean change in summer Carbon flux (Ex1e-4 –Ex1e-5). (F) Mean change in winter Carbon flux (Ex1e-4 –Ex1e-5). Positive values imply reduced carbon uptake or increased outgassing.

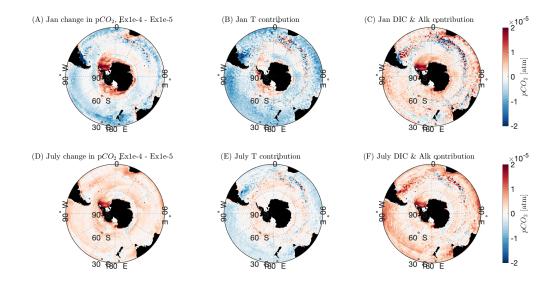


Figure 6: (A) January (summer) 2013-2018 mean change in pCO_2 (Ex1e-4 - Ex1e-5) approximated by the method of Takahashi et al. (2014). (B) Contribution due to changes in temperature. (C) Contribution due to changes in carbon chemistry (DIC and Alkalinity). (D-F) Same as A-C but for July (winter) mean.

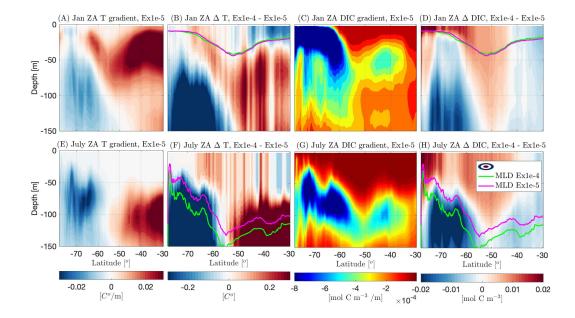


Figure 7: (A) January zonally averaged temperature vertical gradient (red implies increase in temperature towards the surface). (B) January change in temperature (Ex1e-4 - Ex1e-5). Mixed layer depth (MLD) for Ex1e-5 (pink) and Ex1e-4 (green) overlain. (C) January zonally averaged DIC vertical gradient. (D) Change in DIC concentration (Ex1e-4 - Ex1e-5). (E-H) As in A-D but for July mean.

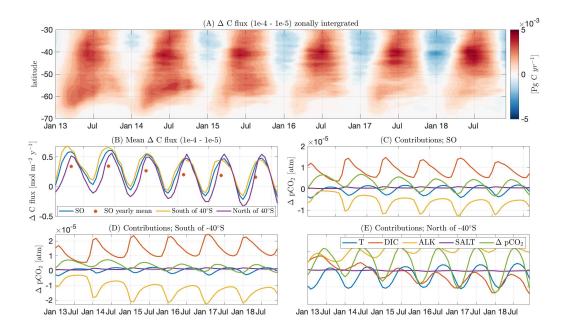


Figure 8: (A) Change in zonally integrated Carbon flux between Ex1e-4 and Ex1e-5 over the six-year time period of Jan 2013 to Dec 2018 (Red shows reduced uptake or increased outgassing in Ex1e-4). (B) Change in the mean carbon flux across the whole SO (blue), the SO North of 40°S (purple) and South of 40°S (yellow) for the same time period. The annual mean change for the whole SO for each year is shown (red star). Using the methodology of Takahashi et al. (2014) as discussed previously, the differences in Carbon flux between the two model runs over time can be attributed to changes in surface ocean pCO_2 (green lines) from alterations to temperature (blue lines), DIC (red liens), alkalinity (yellow lines) and salinity (purple lines). These contributions are shown for the whole SO (C), the SO south of 40°S (D), and the SO north of 40°S (E).

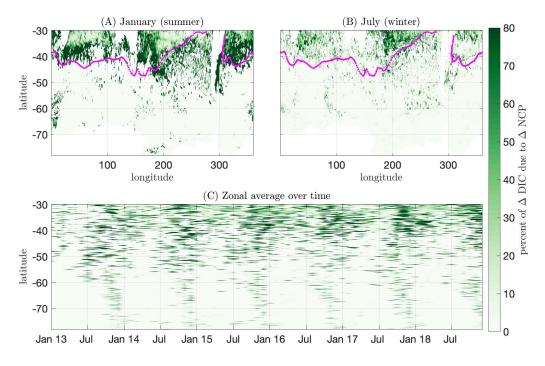


Figure 9: The percentage change of surface water DIC concentration due to changes in biological net community productivity (NCP). Surface water is defined here as waters down to a depth of 55m. Shown as a vertically integrated mean for (A) January (summer) and (B) July (winter). The mean location of the subtropical front, as defined by Orsi et al. (1995), is also shown in pink. (C) The zonal mean of the vertically integrated percentage change due to altered NCP is shown over time.

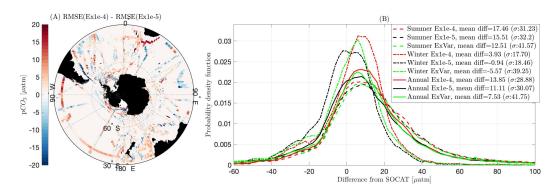


Figure 10: Comparison of modelled pCO_2 to observations from Surface Ocean CO₂ Atlas (SOCAT) between 2012 and 2018 (Bakker et al., 2016). (A) Comparison of the root mean squared error between Ex1e-4 and Ex1e-5. Red/blue shows regions where Ex1e-5/Ex1e-4 is closer to the observations. (B) Probability density function showing the misfit between observed carbon fluxes from SOCAT and the model outputs for pCO_2 in Ex1e-5 (black), Ex1e-4 (red), and ExVar (green).

Hypersensitivity of Southern Ocean air-sea carbon fluxes to background turbulent diapycnal mixing

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Key Points:

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8	•	Total air-sea carbon fluxes in the Southern Ocean are altered by up to 66% annually
9		by modest background mixing variations.
10	•	Resolving or skillfully parameterising the spatiotemporal variability of small-scale
11		turbulent mixing in the Southern Ocean is essential to model air-sea carbon fluxes.
12	•	The spatiotemporal coverage of available pCO_2 observations is insufficient for con-
13		straining the role of diapycnal mixing rates below the mixed layer on air-sea fluxes

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14 Abstract

The Southern Ocean (SO) connects major ocean basins and hosts large air-sea carbon fluxes 15 due to the resurfacing of deep nutrient and carbon-rich waters. While surface-intensified 16 wind-induced turbulent mixing in the SO surface mixed layer is significant for air-sea fluxes, 17 the orders-of-magnitude weaker background mixing below the mixed layer has not been 18 considered consequential. Topographically induced upward propagating lee waves in the 19 SO, wind-induced downward propagating waves generated at the base of the mixed layer, 20 shoaling of southward propagating internal tides generated in the basins north of the SO, and 21 turbulence under sea ice are among the processes known to induce upper ocean background 22 turbulence but typically are not represented in models. Here, we show that altering the 23 background mixing in the SO over a modest range can lead to a $\sim 40\%$ - 60% annual change in 24 SO air-sea CO_2 fluxes, with bigger changes on a seasonal timescale. This is primarily through 25 altering the temperature and the dissolved inorganic carbon and alkalinity distribution in 26 the surface water. Given the high spatiotemporal variability of processes that induce small-27 scale background mixing, this work demonstrates the importance of their representation in 28 climate models for accurate simulation of global biogeochemical cycles. 29

30 Introduction

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The Southern Ocean (SO), defined here as any region south of 30° S, is a key region 31 for the global carbon cycle due to the upwelling of deep, old, carbon and nutrient-enriched 32 waters, connecting the vast reservoir of nutrients and carbon from below the mixed layer 33 with the surface (Talley et al., 2016). The deep ocean interacts with the atmosphere through 34 less than 4% of the ocean's surface area (Watson & Naveira Garabato, 2006; Klocker, 2018), 35 with 65% of interior waters making first contact with the atmosphere in the SO (DeVries 36 & Primeau, 2011). As the deep ocean contains up to 60 times more carbon than the atmo-37 sphere (Arias et al., 2021), small perturbations to air-sea carbon fluxes can be important 38 for atmospheric carbon content (Adkins, 2013). Therefore, the SO, and especially the up-39 welling branch of circumpolar deep water (Marshall & Speer, 2012), is key in controlling 40 global biogeochemical cycles, the exchange of CO_2 between the atmosphere and the deep 41 ocean, atmospheric CO_2 levels, and the response of the ocean and atmosphere to climate 42 change (Sarmiento et al., 2004; Gruber et al., 2019). 43

Several expeditions have revealed strong cross-density (diapycnal) mixing due to smallscale ocean turbulence in the SO (Garabato et al., 2004; Ledwell et al., 2011; Watson et al., 2013; Garabato et al., 2019), though measurements remain sparse and difficult to scale
up (Tamsitt et al., 2018; Mashayek et al., 2017; Cael & Mashayek, 2021; Mashayek et al., 2022). Given the small scales of diapycnal mixing, it is not resolved in operational models, and so it is parameterised (Gaspar, Grégoris, & Lefevre, 1990; W. G. Large et al., 1994) in two forms:

- 1. Surface mixed layer mixing from storms and other surface winds, as well as convective instabilities.
- Background turbulence induced by bottom generated internal waves due to interaction
 of jets, eddies, and tides with rough topography or due to shoaling and breaking of
 remotely generated internal tides (see de Lavergne et al. (2020); Baker and Mashayek
 (2021, 2022) for reviews of such dynamics).

The 'background' mixing in the ocean interior is typically several orders of magnitude smaller than that in the surface mixed layers. Since the seminal work of Munk (1966), bulk measurements of ocean mixing have found a diapycnal turbulent diffusivity of $K_v \sim \mathcal{O}(10^{-4})$ $m^2 \,\mathrm{s}^{-1}$ required to resurface the abyssal waters and facilitate the closure of the meridional overturning circulation (MOC) (Ganachaud & Wunsch, 2000; Talley et al., 2003; Lumpkin & Speer, 2007; Talley, 2013), while estimates from profiling instruments often find $K_v \sim \mathcal{O}(10^{-5})m^2 \,\mathrm{s}^{-1}$ in the interior of the ocean and much larger values only very close to the seafloor (Waterhouse et al., 2014; Ferrari, 2014). In the Diapycnal and Isopycnal Mixing Experiment in the Southern Ocean (DIMES), estimates of mixing based on microstructure profiles reported $K_v \sim \mathcal{O}(10^{-5})m^2 \text{ s}^{-1}$ at the mean depth of an anthropogenic tracer released upstream of the Drake Passage. Meanwhile, the tracer itself appeared to experience $K_v \sim \mathcal{O}(10^{-4})m^2 \text{ s}^{-1}$ (Watson et al., 2013; Mashayek et al., 2017). The background values used in models typically lie within this range.

Though diapycnal mixing is highly temporally and spatially variable due to its gener-70 ating mechanisms (e.g. strong surface westerly winds and interaction of the currents and 71 72 eddies with rough topography), it is frequently parameterised as temporally invariable and, at times, even spatially constant. Current best estimates of SO diapycnal mixing are based 73 on 'static' maps, produced with numerous limiting assumptions, approximating the contri-74 butions from topographically generated lee waves (Nikurashin & Ferrari, 2011; Shakespeare, 75 2020), wind-induced near-inertial waves (Alford, 2020), and internal tides (de Lavergne et 76 al., 2020). These maps have formed the basis of our representation of such processes in 77 earth system models (A. Melet et al., 2014; A. V. Melet et al., 2022). 78

Diapycnal mixing in the global ocean interior is known to be an important factor in 79 variations in atmospheric carbon levels on centennial to millennial timescales via alterations 80 in ocean circulation (Sigman et al., 2010; Marinov & Gnanadesikan, 2011). Enhanced 81 diapycnal mixing increases deep ocean ventilation via the SO and reduces ocean carbon 82 storage through biological and solubility carbon pumps (Marinov et al., 2008; Marinov & 83 Gnanadesikan, 2011). Climate models are sensitive to the intensity and distribution of global 84 diapycnal mixing, accounting for about 25% of the uncertainty in the estimated range of 85 atmospheric CO_2 concentrations by 2100 (Schmittner et al., 2009). In this work, we are 86 concerned with the response of the air-sea carbon fluxes to mixing below the surface mixed 87 layer on time scales much shorter than those explored in the abovementioned works. We also 88 solely focus on mixing in the SO which has been assumed to be of secondary importance for 89 the global ocean circulation (Nikurashin & Ferrari, 2013) but significant for setting global 90 tracer distributions (Ellison et al., 2021). 91

Given the strong wind-driven isopycnal upwelling in the Southern Ocean, and the in-92 tense diapycnal mixing within the mixed layer induced by strong winds, one may imagine 93 that the modest background mixing below the mixed layer would be inconsequential for 94 setting the air-sea fluxes of CO_2 on short timescales. In this work, we show the contrary. 95 The air-sea flux of CO_2 primarily depends on the difference in the partial pressures of CO_2 96 (pCO_2) between the atmosphere and the ocean. Physical and biological processes, including 97 advective and diffusive transport of tracers, organic matter formations and sinking, and 98 the dilution of tracers due to precipitation, runoff and sea ice melt, all alter the pCO_2 of 99 surface waters (Mahadevan et al., 2011), resulting in high variability in time and space 100 of the CO_2 fluxes in the SO (Rosso et al., 2017). The influence of altering background 101 diapycnal mixing on the surface ocean pCO_2 is complex to predict due to spatiotemporal 102 variability in biological and physical responses to variations in mixing (Dutreuil et al., 2009), 103 and the coupled multivariate dependency of ocean pCO_2 to temperature, salinity, alkalinity 104 and dissolved inorganic carbon. In this work, we will show that modest perturbation of the 105 background mixing strongly alters the SO-integrated air-sea CO₂ fluxes on seasonal, annual, 106 and interannual timescales. 107

108 Experiment Setup

We use the Biogeochemical Southern Ocean state estimate (B-SOSE; Verdy and Mazloff (2017)). The interaction between diapycnal mixing below the mixed layer, the mixed layer dynamics, and the mesoscale processes that advect tracers prove essential for the problem under consideration here. Thus, employing an eddy-resolving model such as B-SOSE is essential and distinguishes this work from studies mentioned earlier that use low-resolution ¹¹⁴ models in global settings to study the role of mixing on carbon fluxes. In return, the ¹¹⁵ timescales of relevance in our study are much shorter than theirs.

The model's optimization procedure solves for adjustments to the prescribed atmo-116 spheric state and initial conditions to bring the solution into better consistency with obser-117 vations. The resulting product is a closed-budget model simulation representing the present 118 SO state. Because it represents the constantly evolving SO state it is not in a steady state, 119 in contrast to what one may expect from analysis of an earth system model undergoing a 120 long spin-up process. The B-SOSE simulation provides an estimate of SO biogeochemistry, 121 122 including air-sea fluxes of heat and carbon and the cycling of nutrients from a biogeochemical and carbon model. 123

The model domain extends from the equator to 78° S with 52 vertical layers of varying 124 thickness. The zonal grid spacing is always $1/6^{\circ}$, but the meridional grid spacing changes 125 with the cosine of the latitude. Ocean physics is represented using the MITgcm. The ocean 126 is forced through an atmospheric boundary layer scheme where bulk formulae determine 127 heat fluxes, freshwater (salt), and momentum (W. Large & Yeager, 2009). The hourly 128 atmospheric conditions of ERA5 (Dee et al., 2011) are applied, with adjustments added 129 that were determined by optimization (Verdy & Mazloff, 2017) using the adjoint method. 130 Sea ice is modelled using 0-layer thermodynamics (Fenty & Heimbach, 2013) and an implicit 131 Line Successive Over Relaxation (LSOR) dynamical solver (Losch et al., 2010). A horizontal 132 biharmonic diffusivity is used with a value of 10^{-8} m⁴s⁻¹. Implicit vertical diffusivity for 133 convection is set to 10 $m^2 s^{-1}$, and no mesoscale eddy parameterisation was implemented. 134 For further details, see Verdy and Mazloff (2017) and Swierczek et al. (2021). 135

The parameterisation of diapycnal mixing is composed of two parts. The GGL90 mixed 136 layer parameterisation of Gaspar, Grégoris, and Lefevre (1990) is used to represent param-137 eterised turbulence and is highly surface-enhanced (Fig.1 D), inducing strong turbulence 138 under the seasonal atmospheric storm tracks, mixing the DIC gradients in the upper few 139 hundreds of meters. In other places, such as under the ice or when there is no strong 140 wind-induced turbulence, the models rely on a prescribed background value for turbulent 141 diffusivity. The background value is behind the sensitivity of fluxes discussed in this work. 142 As discussed in subsequent paragraphs, the background vertical diffusivity is altered in each 143 experimental run. The background mixing value is added to the GGL90 mixing to achieve 144 a total vertical diapycnal mixing value. 145

The Biogeochemistry with LIght, Nutrients and Gases (NBLING) model, described 146 fully in Verdy and Mazloff (2017), forms the biochemical framework within B-SOSE. The 147 original BLING model described in Galbraith et al. (2010) was modified (for B-SOSE) with 148 the addition of nitrogen cycling and improvements in the representation of phytoplankton 149 population dynamics and particle export (Galbraith et al., 2015; Bianchi et al., 2013). Bi-150 ological activity influences the concentrations of carbon and oxygen. At the core of the 151 BLING model is primary production with limitations by light, nitrate, phosphate, iron, and 152 temperature, and subsequent remineralization of organic matter back to inorganic nutri-153 ents. Nine prognostic tracers are simulated in NBLING: dissolved inorganic carbon (DIC), 154 alkalinity (ALK), oxygen (O₂), nitrate (NO₃), phosphate (PO₄), iron (Fe), dissolved or-155 ganic nitrogen (DON), dissolved organic phosphate (DOP) and phytoplankton biomass. The 156 biomass partitioning into species is determined using stored ratios updated every timestep 157 based on growth and decay rates. We include three types of phytoplankton: large, small, 158 and diazotrophs. Small cells represent calcifying organisms; they use calcium carbonate 159 to form shells. Diazotrophs can fix nitrogen, so nitrate availability does not limit them. 160 Phytoplankton loss is expressed as a power law with a size-dependent exponent based on 161 162 (Dunne et al., 2005).

The B-SOSE carbon system is adapted from the MITgcm simple biogeochemical model of Dutkiewicz et al. (2006). DIC and ALK are prognostic variables, and pH and pCO_2 are diagnosed based on Follows et al. (2006), making oceanic pCO_2 a function of DIC, ALK, temperature (T), salinity (S) and silica, where silica is prescribed from the 2013 World Ocean Atlas climatology (Garcia et al., 2013). CO_2 and oxygen air-sea fluxes are calculated following Wanninkhof (1992). Atmospheric pCO_2 is prescribed using values from the CarbonTracker product (Peters et al., 2007).

The state estimate is produced by solving for the model initial and boundary con-170 ditions, the so-called model 'controls', which minimize a weighted least squares sum of 171 model-observation misfits. This is achieved using an adjoint model that provides the gradi-172 ents of the misfit function with respect to the model controls, allowing those controls to be 173 174 efficiently and systematically determined. The model is run for seven years (2012 - 2018)using the adjoint method-based assimilation product. This includes physical and biogeo-175 chemical constraints obtained from Argo floats, including biogeochemical parameters from 176 the SOCCOM float array, satellite altimetry, satellite SST, and ship transect data. The full 177 set of model parameters used in this $1/6^{\circ}$ setup is given in Swierczek et al. (2021); see Verdy 178 and Mazloff (2017) for initial conditions. 179

The B-SOSE's original conditions and atmospheric adjustments were obtained using 180 $10^{-4} m^2 s^{-1}$ as the value for background diapychal diffusivity (added to the GGL90 mixing). 181 We refer to that base simulation as Ex1e-4 hereafter. Using the same initial conditions and 182 atmospheric state adjustments, two additional model simulations were carried out for 2013-183 2018. The first perturbation experiment, Ex1e-5, uses a constant background diffusivity 184 value of $10^{-5}m^2s^{-1}$. The range $10^{-5}m^2s^{-1}$ to $10^{-4}m^2s^{-1}$ is conservative, sandwiched 185 between the two canonical paradigms of mixing often considered in Physical Oceanography. 186 The third experiment, ExVar, uses a spatially variable (but temporally constant) vertical 187 diapycnal mixing map (Fig 1A-C). The map is constructed as the sum of contributions from 188 tides (de Lavergne et al., 2020) and topographically-generated lee waves (Nikurashin & 189 Ferrari, 2013). ExVar features horizontal and vertical variations over a range much broader 190 than $10^{-5} m^2 s^{-1}$ to $10^{-4} m^2 s^{-1}$. Although background mixing values dominate over the 191 GGL90 parameterisation in mid-depths, GGL90 is orders of magnitude larger than the 192 background mixing value in the upper ocean (Fig.1D). The three cases together allow for 193 evaluating the impact of the magnitude of mixing and its spatial variations on the carbon 194 flux independently. 195

196 **Results**

¹⁹⁷ Carbon fluxes

The SO is a net sink of atmospheric CO_2 , with most of the uptake occurring between 198 50° S and 30° S, with a peak at 40° S, where around 7 Tg C/yr is taken up (Fig.2A). 40° S 199 is the average latitude of the subtropical front, separating the subtropical waters from the 200 subantarctic mode waters, thus hosting rich mesoscale and submesoscale frontal dynamics 201 and enhanced air-sea exchange of tracers. To the south of the polar front (panel E), on the 202 other hand, the upwelling of deep carbon-rich waters causes carbon outgassing (shown in 203 red panel E). Additional uptake occurs further south around Antarctica due to downwelling 204 (induced by a change in the wind direction from westerly to easterly) and deep water forma-205 tion. Thus, SO fronts, which mark sharp gradients in temperature and carbon chemistry, 206 separate regions of net uptake from regions of outgassing. Higher latitudes show very low 207 mean annual carbon fluxes, partly due to seasonal ice cover. 208

Carbon uptake varies year-on-year during the six years of the state estimate run by almost 2 Tg C/yr at some latitudes, with especially high inter-annual variability at 60°S and 40°S (Fig.2A). The inter-annual range of carbon fluxes for Ex1e-5 are highly nonmonotonic. The inter-annual variations are due to varying oceanic conditions each year, some of which are associated with the Southern Annular Mode (SAM).

Alterations to background diapycnal mixing alter SO carbon fluxes, with ExVar showing smaller differences from Ex1e-5 than Ex1e-4 (Fig.2B dashed vs solid lines). The sensitivity

to altered diapycnal mixing is variable throughout the six years (Fig.2 B). This inter-annual 216 range in sensitivity of around 0.1 Tg C/yr is well within the range of the inter-annual 217 variability of zonally integrated carbon fluxes in Ex1e-5 (Fig.2A,B). A higher difference 218 between Ex1e-5 and Ex1e-4, and to a lesser extent between Ex1e-5 and ExVar occurs in the 219 first three years (2013 to 2015) than in the last three years (2016 to 2018) (Fig.2B). As upper 220 ocean mixing is never in an equilibrium state due to constantly changing winds, eddies and 221 buoyancy fluxes, the response to the mixing perturbation over the first few months of this 222 experiment do not seem unrealistically exaggerated. 223

224 Increasing the background mixing from Ex1e-5 to Ex1e-4 leads to a reduction in annual mean zonally integrated carbon uptake at all latitudes (Fig.2C,F). The most significant 225 reduction is around 55°S (Fig.2C,F). Minor changes occur south of 65°S due to the ice cover. 226 In ExVar, the most significant changes from Ex1e-5 occur further north, at around 45°S. 227 South of 60°S, the difference between ExVar and Ex1e-5 is insignificant (Fig.2C,G). Large 228 areas of ExVar have carbon fluxes unchanged from those in Ex1e-5 (Fig.2G), suggesting 229 that $10^{-4}m^2$ is likely too large of a background mixing value. In Ex1e-5, the mean annual 230 cumulative net flux of carbon into the ocean, integrated from $75^{\circ}S$ northward to $30^{\circ}S$, is 231 1 Pg C/yr (Fig.2D). In Ex1e-4 only 0.6 Pg C/yr is taken up, a reduction of 0.4 Pg C 232 yr^{-1} . The annual uptake of ExVar falls between the other two experiments at around 0.8 233 Pg C/yr. These numbers are for the six-year mean, and as panel B shows, the reductions 234 from Ex1e-5 are much higher over the first three years (almost double). 235

The cumulative carbon fluxes are compared to other estimates of the integrated SO 236 carbon flux from 75° S northward to up to 45° S and 35° S for the period 2015-2017 (Fig.2D; 237 Bushinsky et al. (2019); Landschützer et al. (2016); Rödenbeck et al. (2013)). At 45°S, 238 the Ex1e-5 cumulative flux lies between the three observationally inferred estimates, while 239 the ExVar flux is slightly lower, with Ex1e-4 being the lowest of all three, well below the 240 estimates of the other studies. At 35° S, there is a larger range in carbon uptake between 241 the three model runs. Ex1e-5 is the only experiment that lies within the bounds of the three 242 observational estimates, though it appears towards the lower end, whilst ExVar and Ex1e-4 243 are below. This further suggests that the lower mixing in Ex1e-5 could be a more suitable 244 background mixing value. 245

²⁴⁶ Changes to surface ocean pCO_2

Given that ExVar estimates of carbon fluxes fall between those of Ex1e-4 and Ex1e-5, 247 hereafter, we only focus on the differences between Ex1e-4 and Ex1e-5. Air-sea carbon fluxes 248 exist due to the difference in pCO_2 between the atmosphere and the surface ocean. The 249 high (low) surface ocean pCO_2 values result in regions of low (high) oceanic uptake or even 250 outgassing of CO_2 from the atmosphere (Fig.3A). A region of exception is under sea ice, 251 where the diffusive flux of gases is prevented. The changes in carbon fluxes due to altered 252 mixing, as in figure 2, are due to changes in surface ocean pCO_2 , as atmospheric conditions 253 are constant across experiments. 254

The annual mean pCO_2 of the surface ocean is higher in Ex1e-4 than in Ex1e-5 in almost all regions, reducing the pCO_2 gradient and carbon uptake (Fig.3B). The areas of greatest increase in pCO_2 include south of South Africa and the waters east of the West Antarctic Peninsula. Small regions where the annual mean pCO_2 is reduced in Ex1e-4 include latitudes of around 30°S, especially to the east of Australia, the Argentine basin, and a few small bands just off the coast of Antarctica in the south.

Using the methodology set out by Takahashi et al. (2014), we break down the pCO_2 change to contributions from changes in the upper ocean content of salinity, temperature, DIC and alkalinity (equation 1). The change in pCO_2 as a contribution from each of the four tracers is calculated using equations 2-5, where $\bar{p}CO_2$ is the mean pCO_2 , $\bar{A}lk$ is the mean alkalinity, γ_{CO_2} is the Revelle factor for CO_2 (value used = 11), and γ_{ALK} is the Revelle factor for alkalinity (value used = -10).

$$\Delta pCO_2 = \left(\frac{\delta pCO_2}{\delta T}\right)\Delta T + \left(\frac{\delta pCO_2}{\delta DIC}\right)\Delta DIC + \left(\frac{\delta pCO_2}{\delta Alk}\right)\Delta Alk + \left(\frac{\delta pCO_2}{\delta S}\right)\Delta S \quad (1)$$

$$\frac{\delta p CO_2}{\delta T} \Delta T = 2(p CO_2) [Exp(0.0423(\pm 0.0002)\Delta T/2) - 1]$$
(2)

$$\frac{\delta p C O_2}{\delta D I C}) = \gamma_{CO_2} (\bar{p} C O_2 / \bar{T} C O_2) \tag{3}$$

$$\frac{\delta p C O_2}{\delta A l k} = \gamma_{ALK} \left(\frac{\bar{p} C O_2}{\bar{A} l k} \right) \tag{4}$$

$$\left(\frac{\delta p C O_2}{\delta S}\right) = 0.026(\pm 0.002) \cdot \bar{p} C O_2 \tag{5}$$

The four individual contributions, shown in Fig.3D-G, can be summed together to give 267 the annual mean approximated change in pCO_2 (Fig.3C). This calculated change agrees 268 satisfactorily with the changes in pCO_2 between the two experiments (Fig.3B). This verifies 269 the assumptions made in equations 2-5, and confirms that changes to the distribution of 270 these tracers are key in causing changes to carbon fluxes (Fig.3B,C). The only region where 271 the Takahashi et al. method does not capture the changes is in the north of the SO, west 272 of New Zealand and east of South America in the Argentine basin. This is likely due to 273 enhanced water mass mixing occurring in these regions, making changes in this area complex 274 to approximate with simple assumptions. While the calculations shown in Fig. 3 use the 275 upper 2.6 m of the water column, they are not sensitive to depth and similar results are 276 found down to ~ 55 m. 277

On an annual basis, contributions from changes in DIC and alkalinity concentrations are 278 the main drivers of changes in pCO_2 , with the contributions from salinity and temperature 279 being secondary (Fig.3E,F). An increase in the alkalinity content decreases pCO_2 , whilst 280 an increase in salinity or DIC increases pCO_2 . Where the temperature increases, pCO_2 281 increases due to modulation of the equilibrium DIC. The increase in the DIC content of 282 the surface waters of the southern SO in Ex1e-4 increases pCO_2 , whilst in the north the 283 decrease in DIC concentration decreases pCO_2 . On the contrary, the increase in alkalinity 284 concentration in the south decreases pCO_2 , while the decrease in alkalinity in the north 285 increases pCO_2 . Changes in salinity concentrations act to slightly increase the pCO_2 in 286 Ex1e-4. Temperature changes with enhanced mixing cause a slight decrease in pCO_2 in the 287 north and an increase in pCO_2 in the south in Ex1e-4. 288

Changes in the upper ocean temperature, salinity, DIC and alkalinity are due to alter-289 ations to the diapycnal flux of these tracers. The diapycnal flux for a tracer with concentra-tion C may be approximated by $-K_v \times \frac{\partial C}{\partial z}$. Therefore, if vertical diapycnal mixing K_v is increased, more tracer, e.g. DIC, is mixed downgradient (upward into the surface waters). 290 291 292 This increase in upward flux is the strongest where the vertical gradients are the strongest. 293 Therefore, strong correlations develop between locations with sharp vertical gradients and 294 locations with significantly altered tracer content with enhanced mixing (Fig.4A-D). This 295 correlation is especially clear when examining changes to tracer distributions in the first 296 month of the perturbation experiments, as shown in figure 4. 297

Regions that experience high changes in DIC concentration with enhanced mixing are 298 around the coast of Antarctica, south of 60° S and above depths of 40 m. In these areas, 299 surface waters are fed by wind-induced upwelling of deep waters rich in DIC due to the 300 respiration of organic material. Further to the north, the upper 120 m of the water column 301 has weak vertical gradients of DIC concentration (Fig.4E,F). The dipole pattern shown 302 when looking in a zonal average sense implies the erosion of the sharp gradient by enhanced 303 mixing. The DIC concentration increases with increased mixing in the upper surface waters 304 (shown in red), while concentrations decrease between 40m and 20m depth (shown in blue) 305 due to a flux divergence, as more of this carbon has been mixed upwards into the surface 306 waters (Fig.4G). There is a clear divide at around 20 m; this depth corresponds to the 307 depth of the maximum vertical gradient. Changes in alkalinity and salinity roughly follow 308

³⁰⁹ a pattern similar to DIC (hence not shown). The greatest changes in temperature occur in ³¹⁰ different regions, mainly in the northern SO, especially at around 90°E, in the Argentine ³¹¹ basin, and in the waters surrounding New Zealand.

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Temporal and seasonal variability of changes in pCO_2

Carbon fluxes show strong seasonal and spatial variations (Fig.5), as for example, dis-313 cussed in Rosso et al. (2017). In the austral summer (December to February), the SO from 314 60° S to 30° S is a net source of CO₂ outgassing (Fig.5A,B Dashed lines). Some outgassing 315 also occurs at the upwelling zone of the polar front, especially in the Atlantic basin (Fig.5C). 316 South of $\sim 60^{\circ}$ S, the SO acts as a slight carbon sink even in summer. Austral winter (June 317 to August) has much higher carbon uptake than summer, with the net uptake occurring in 318 almost all regions of the SO, except beneath sea ice (Fig.5A,B,D). Small outgassing regions 319 exist around the polar front and at the upwelling region off the west coast of South America 320 in the Argentine basin (Fig.5D). 321

While figures 2 and 3 show the changes to carbon fluxes between model runs in an 322 annual mean sense, there are also significant temporal patterns in how mixing perturbation 323 alters carbon fluxes (as will be discussed in Fig.8). Figures 5 and 6 examine the dominant 324 mechanisms for seasonal differences observed in the changes to pCO_2 between Ex1e-4, ExVar 325 and Ex1e-5. The changes in carbon fluxes between experiments are greater in winter than 326 in summer (Fig.5A,E,F). An exception is in the very south, where ice cover during winter 327 reduces gas exchange in all experiments. In winter, Ex1e-4 has a reduced carbon uptake 328 compared to Ex1e-5, while ExVar has a similar carbon uptake to Ex1e-5 (Fig.5A). Cumu-329 latively integrated winter carbon fluxes are reduced from almost 2 Pg C/yr in Ex1e-5 to 330 1.2 Pg C/yr in Ex1e-4. The greatest decreases in uptake occur around 40°S. The Argentine 331 basin is also a region of pronounced diminished carbon uptake (Fig.5F). Three small areas 332 on the edge of the winter ice extent experience increased carbon uptake in the winter due to 333 reduced ice cover, the reason for which is explained subsequently (Blue areas, Fig.5F). Sum-334 mer changes to carbon fluxes are of a smaller magnitude and show more spatial variability 335 than the winter months (Fig.5A,E). In summer, the cumulative intergrated outgassing of 336 Ex1e-5 is higher than Ex1e-4, and ExVar is higher than both Ex1e-4 and Ex1e-5, though 337 the difference between all three runs is less than 0.2 Pg C/yr. At lower latitudes where the 338 SO is a net source of carbon to the atmosphere, outgassing is decreased in Ex1e-4. Further 339 south, where the SO is a carbon sink, CO_2 uptake is reduced in Ex1e-4 (Fig.5E). Changes 340 in flux occur as far south as the Antarctic continent due to diminished summer sea ice. 341

Using Eqs.1-5 and Fig. 3, we next use the Takahashi et al. methodology to examine 342 seasonal changes to tracer contributions and their implications for the pCO_2 and carbon 343 fluxes. The outcome is shown in Fig. 6. Salinity contributions to changes to pCO_2 are not 344 shown as they are negligible compared to DIC, alkalinity, and temperature contributions. 345 The temperature contribution varies greatly between seasons, being stronger in January 346 than in July (Fig.6B,E). Closer to Antarctica, changes in temperature increase the pCO_2 347 of Ex1e-4 surface waters throughout the year. In July, this positive contribution extends 348 further north. In January, the change in temperature causes very strong reductions in 349 pCO_2 , especially in the subtropical gyres. Because the change in surface temperature and 350 associated change to pCO_2 vary with season, the annual mean change in temperature and 351 its contribution to change in pCO_2 appear much smaller (Fig.3D). They are nevertheless 352 key to driving the seasonal response of changing SO carbon fluxes in response to altered 353 diapycnal mixing. 354

The vertical structure of the thermocline and the associated change in surface temperature with enhanced mixing have seasonal trends (Fig.7B,F). In January, surface waters are warm, and the temperature declines rapidly with depth down to 100 m, especially north of 60°S (Fig.7A). South of 60°S and below 100 m, water temperature increases with depth due to the upwelling of deep warm waters of North-Atlantic origin through Ekman transport.

In Ex1e-4, subsurface cooler waters are mixed more strongly towards the surface, cooling 360 the surface waters and warming the subsurface waters relative to Ex1e-5 (Fig.7B). This 361 surface cooling reduces the pCO_2 (Fig.6A). In July the surface waters are well mixed and 362 there is no temperature gradient in the upper 100 m (Fig.7E). Below the winter mixed 363 layer, temperature rises with depth. Enhanced mixing warms surface waters, increasing the 364 pCO_2 (Fig.6E,F). This increase in surface temperature also increases the rate of sea ice melt, 365 reducing the sea ice cover toward the end of winter/ spring in Ex1e-4. This results in small 366 regions of increased carbon uptake around the sea ice edge in winter (Fig.5F). North of 60° S 367 and below the mixed layer, waters still decrease in temperature with depth, so increased 368 mixing cools the surface waters (Fig.7E,F). The vertical gradients of DIC and alkalinity are 369 relatively constant regardless of season (Fig.7C,G). Changes in DIC and alkalinity concen-370 trations have opposing effects on pCO_2 (Fig.3E,F), but together act to increase pCO_2 at all 371 latitudes in summer and winter in Ex1e-4 (Fig.6C,F). The increase to pCO_2 in Ex1e-4 from 372 combined carbonate chemistry changes is stronger in the winter than in summer, especially 373 north of 40° S (Fig.6C,F). 374

Though all six years of the model run exhibit a similar seasonal cycle of changes to 375 carbon fluxes with enhanced mixing in Ex1e-4, important inter-annual differences exist 376 (Fig.8) which would not be appreciated in annual and seasonal means. Some differences in 377 carbon fluxes between the two experiments become more pronounced over time, while other 378 changes become less. North of 40°S, Ex1e-4 has an increase in carbon uptake (or reduced 379 outgassing) during the summer. This becomes more pronounced and extends further south 380 down to 50° S in subsequent summers as the model run progresses. While the winter time 381 reductions in uptake in Ex1e-4 compared to Ex1e-5 around $45^{\circ}S$ become stronger through 382 the six years, the reductions in carbon uptake south of 60°S become weaker. The SO yearly 383 mean change in C flux (red stars, Fig.8B) show a smaller mean change in the carbon flux 384 between Ex1e-4 and Ex1e-5 in the later years of the run compared to earlier years. This is 385 due to opposing signs of change to carbon fluxes over the seasons becoming more pronounced 386 and therefore causing an antagonistic net effect to changes in an annual mean sense. 387

40°S approximately corresponds to the mean latitude of the subtropical front (STF) and marks a regime in terms of the leading mechanisms responsible for changes in pCO₂ and carbon fluxes. This marks the boundary between the nutrient deplete sub-tropical waters to the north and the nutrient and DIC rich waters to the south (Chapman et al., 2020). Regions to the north of this divide are responsible for the summer increases in carbon uptake in Ex1e-4 in later years of the run. In contrast, regions to the south are responsible for the strong response of reduced carbon uptake in Ex1e-4.

The contributions to the total change in pCO_2 are drastically different across the STF 395 (Fig.8C-E). To its South, opposite changes in pCO_2 due to alkalinity and DIC nearly bal-396 ance, with the latter being slightly larger (Fig.8D). As before, salinity contributions remain 397 negligible at all times. Over the first two years of the perturbation, the total change in pCO_2 398 is positive, meaning pCO_2 is higher in Ex1e-4 than in Ex1e-5. The magnitude of the reduc-399 tion in pCO_2 due to Alkalinity increases over time. The magnitude of total pCO_2 change 400 decreases over time and becomes negative in the summer months, allowing for increased 401 carbon uptake. 402

North of the STF, DIC and alkalinity do not balance each other out (Fig.8E) and changes in temperature between the two model runs are more dominant. Alkalinity increases pCO_2 north of the STF, while changes in DIC initially also increase the pCO_2 in Ex1e-4. By the summer of the third year of the run, changes in DIC begin to reduce the pCO_2 , causing the net total change in pCO_2 to be negative in summer. This causes an increase in carbon uptake in the summer north of the STF in Ex1e-4.

Almost all changes in DIC between Ex1e-4 and Ex1e-5 are due to altered diapycnal fluxes of DIC. However, north of the STF, DIC contribution to decreased pCO_2 in the summer is due to increased productivity in the nutrient depleted waters of the sub tropical

gyre. This increase in productivity does not occur instantaneously, but instead takes around 412 6 months to begin decreasing the DIC contribution (red line, Fig.8E and Fig.9C). While an 413 increase in productivity occurs with higher mixing across the whole SO region, it is only in 414 the north, roughly north of the sub tropical front where increased phytoplankton production 415 and DIC uptake becomes the dominant mechanism in altering DIC concentrations. Thus, 416 in the context of this paper, we consider the STF as the upper boundary of the SO and 417 postpone further discussion on the biologically-dominated change in pCO_2 north of the SO 418 to future work. 419

$_{420}$ Comparison to pCO_2 observational data

The pCO_2 values for Ex1e-4, Ex1e-5 and ExVar can be compared to 2013-2018 observed levels from the Surface Ocean CO₂ Atlas (SOCAT) (Bakker et al., 2016) (Fig.10). Neither clearly matches SOCAT observations better than the other (Fig.10A). Regional trends are also unclear, although from the limited data available, Ex1e-5 appears to better represent the pCO_2 of the northern Pacific Ocean, as well as off the coast of South Africa and Tasmania. Meanwhile estimates from Ex1e-4 are better matched to observations in the western Atlantic and the northern Indian Oceans.

The probability density function for the difference between SOCAT and B-SOSE for the 428 three experiments is broken down over seasons (Fig.10B). In summer the standard deviation 429 of differences between ExVar and data is much larger than those for Ex1e-4 and Ex1e-5. The 430 mean difference of 15.5 μ atm for Ex1e-5 is lower than 17.46 μ atm for Ex1e-4, whilst ExVar 431 has the lowest mean difference from observations. The high-end tails of the distributions 432 are more skewed than the lower ends, implying a systematic over-estimate of pCO_2 by 433 B-SOSE. B-SOSE overestimates the flux of carbon from the ocean to the atmosphere or 434 underestimates the SO carbon uptake from the atmosphere, particularly in the summer. 435

SOCAT data is heavily biased towards summer data due to limitations on data col-436 lection in the winter. The mean difference between SOCAT and B-SOSE is lower for the 437 winter mean than for the summer in all experiments. In the winter, ExVar has the largest 438 mean difference from observations but also the largest standard deviation. In an annual 439 mean sense, ExVar does a better job in matching SOCAT observations, though with a much 440 higher standard deviation. It is interesting to note that, while in the annual mean sense, 441 442 ExVar better matches SOCAT observations of pCO_2 , Ex1e-5 was better able to replicate previous observational estimates of cumulative SO C fluxes (Fig.2D). 443

444 Conclusion

We have demonstrated that the air-sea carbon fluxes in the SO are highly sensitive to modest background mixing variations well within the range of our best estimates. This is despite background mixing rates being orders of magnitude smaller than mixed layer model generated mixing. We find that the overall changes to carbon fluxes depend on the interactive effects of changes to DIC, temperature, and alkalinity, which can compensate or reinforce each other, and the predominant driver varies regionally, seasonally and temporally as additive and opposing feedbacks kick in at varying time scales.

The relevance of diapycnal mixing in setting global carbon fluxes has previously been considered to be through changes to the underlying stratification and of regional and global overturning circulation and ventilation patterns. Although that may be true on centennial or longer timescales, here we show that on much faster timescales mixing directly acts upon tracers such as DIC, alkalinity, temperature, and salinity leading to a significant change in surface ocean carbon fluxes.

The high correlation found between vertical gradients and strong changes in tracer distributions with altered mixing shows that on a timescale of days to months, direct changes in diapycnal mixing fluxes are the predominant drivers of the pCO_2 response in the SO. On longer timescales, from months to years, further feedbacks involving changes to biological productivity and mixed layer depth will also begin to cause further changes to the surface ocean pCO_2 . A latitudinal divide exists at around 40°S, roughly the location of the subtropical front. High vertical tracer gradients cause the direct impact of altered tracer fluxes to dominate trends to the south, whilst changes in biological productivity play a key role in the observed changes to carbon fluxes to the north.

Two major issues stand in the way of better constraining of the data-assimilating ocean 467 estimates insofar as the role of vertical mixing in the upper ocean is concerned. First, despite the significant investments in observations such as SOCAT, Fig. 10A clearly shows the 469 sparsity of the available observational data. From a statistical perspective, this coverage 470 is insufficient to discern which background mixing value better represents the real ocean 471 despite the strong impact of these choices on pCO_2 . This issue can be resolved only through 472 sustained observations. Knowledge of the seasonal cycle of pCO_2 is worse in the SO than in 473 most other regions of the ocean. The strong seasonality of the sensitivity of carbon fluxes 474 to altered mixing demonstrates the importance of year-round observations. Second, SO 475 diapychal mixing can vary by orders of magnitude over timescales ranging from hourly to 476 seasonally, as well as varying spatially. To achieve a close agreement with observations, a 477 model should have a representation of such variability. ExVar employed our best estimate 478 of a time-mean spatially variable mixing map, resulting in carbon fluxes similar to that 479 obtained with a constant diffusivity of $10^{-5}m^2/s$. Direct observations of diapycnal mixing 480 in the SO have suggested that such maps (a) lack the representation of many key processes 481 that result in higher turbulence in upper surface waters (e.g. bottom-generated lee waves, 482 shoaling of remotely generated internal tides) and (b) do not allow for co-variance of mixing 483 and tracer gradients, key to biological processes. This work highlights the absolute necessity 484 for climate models to resolve the spatio-temporal variability of small-scale turbulent mixing, 485 or to skillfully parameterise the processes responsible for generating them. 486

⁴⁸⁷ 1 Data and material availability

The data sets generated during and/or analysed during the current study are available from the corresponding author on reasonable request and from http://sose.ucsd.edu.

The data used to construct the spatially variable mixing map used for ExVar is available at https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2020MS002065.

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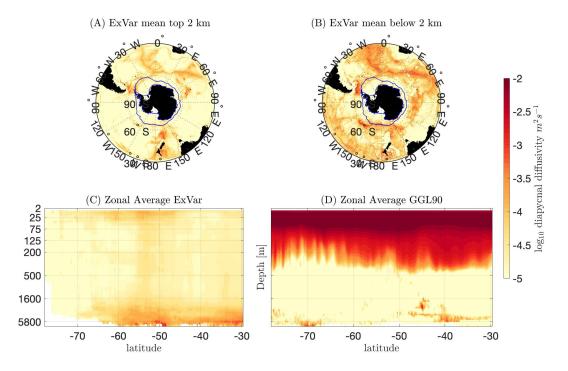


Figure 1: Diapycnal mixing in the Southern Ocean State Estimate (SOSE). The distribution of diapycnal mixing in the Southern ocean, constructed as the sum of contributions from tides and topographically-generated lee waves. This mixing is shown averaged in depth over the top/bottom 2km in panel A/B, and zonally over the Southern Ocean in C. These maps are used in the spatially variable mixing map experiment (ExVar). For reference, a zonally-averaged map of the storm-induced mixing, as parameterised through GGL90 parameterisation in B-SOSE, is also shown in panel D.

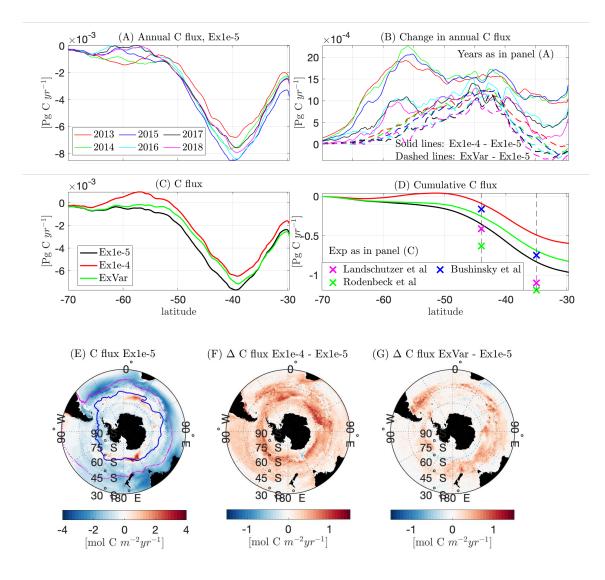


Figure 2: (A) Zonally integrated flux of Carbon for each year of Ex1e-5 (negative = Carbon flux from atmosphere to ocean). (B) Difference in the zonally integrated flux of Carbon between Ex1e-4 and Ex1e-5 (solid lines) and ExVar and Ex1e-5 (dashed lines) for each year of the experiment. (C) zonally integrated annual mean (2013-2018) Carbon flux for Ex1e-4, Ex1e-5 and ExVar. (D) Annual mean (averaged over 2013 to 2018) cumulative integral of carbon fluxes from 70°S northward to 30°S (legend same as the previous panel). Observational markers are included for comparison (Landschützer et al., 2016; Bushinsky et al., 2019; Rödenbeck et al., 2013). (E) Average annual carbon flux for Ex1e-5, the blue line shows the Polar Front, the magenta line shows Sub-tropical Front as defined by Orsi et al. (1995). (F) Annual mean change in Carbon flux (Ex1e-4 – Ex1e-5). (G) Annual mean change in Carbon flux (ExVar – Ex1e-5). Positive values imply reduced carbon uptake or increased outgassing.

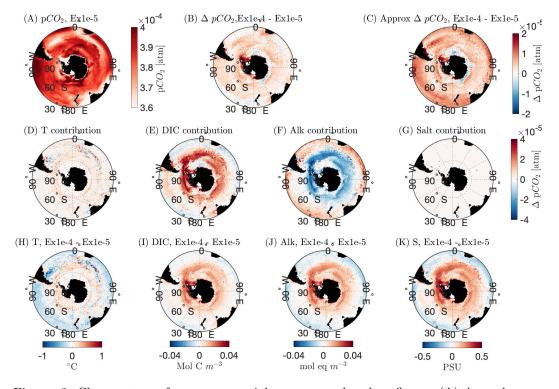


Figure 3: Changes to surface ocean partial pressure and carbon fluxes. (A) Annual mean surface ocean pCO_2 in Ex1e-5. (B) Change in pCO_2 between Ex1e-4 and Ex1e-5. (C) Same as panel B, but this time changes to pCO_2 approximated based on the methodology of Takahashi et al. (2014) that breaks down the change into various contributions as per equations (1-5). The various contributions are shown in panels D-G. (H-K) Changes in annual mean DIC, alkalinity, potential temperature and salinity.

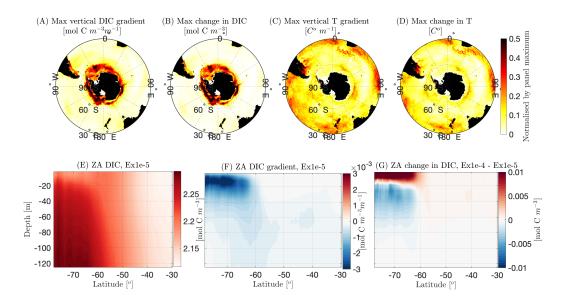


Figure 4: (A) Maximum vertical DIC gradient in the water column for Ex1e-5, normalised by maximum contour value. (B) Maximum change in DIC between Ex1e-4 and Ex1e-5, normalised by the maximum contour value. (C,D) Same as A and B but for temperature. The maximum change in DIC (temperature) is defined as the greatest difference in DIC (temperature) concentration between the two experiments at any depth above 200 m at each latitude and longitude in the domain. (E) Zonal average DIC concentration in Ex1e-5. (F) Zonal average DIC vertical gradient in Ex1e-5; blue indicates a decrease in concentration towards the surface. (G) Zonally averaged change in DIC concentration (Ex1e-4 - Ex1e-5). All shown for December 2012, the first month of all experiments.

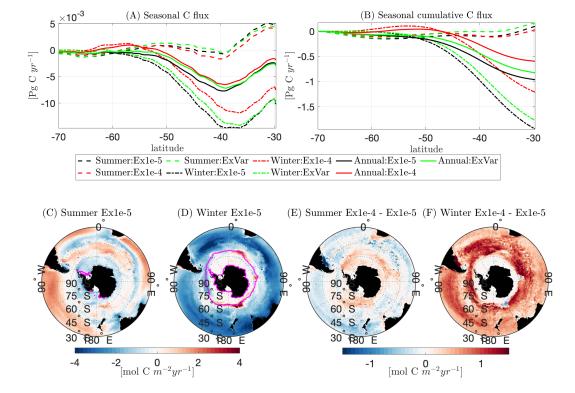


Figure 5: (A) zonally integrated Carbon flux for Ex1e-4, Ex1e-5 and ExVar for summer (dashed), winter (Dotted), and annual mean (solid line). (B) Cumulative sum of carbon fluxes from 70°S northward to 30°S (legend same as the previous panel). (C) Average summer carbon flux for Ex1e-5; magenta lines show the minimum summer ice extent. (D) Average winter carbon flux for Ex1e-5; magenta lines show the maximum winter ice extent. (E) Mean change in summer Carbon flux (Ex1e-4 –Ex1e-5). (F) Mean change in winter Carbon flux (Ex1e-4 –Ex1e-5). Positive values imply reduced carbon uptake or increased outgassing.

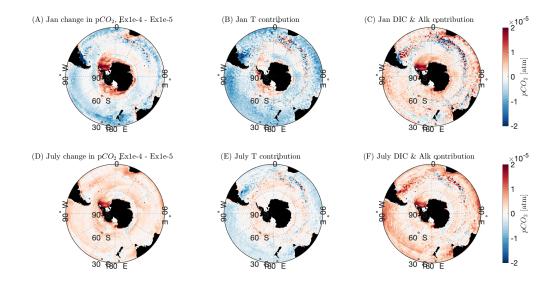


Figure 6: (A) January (summer) 2013-2018 mean change in pCO_2 (Ex1e-4 - Ex1e-5) approximated by the method of Takahashi et al. (2014). (B) Contribution due to changes in temperature. (C) Contribution due to changes in carbon chemistry (DIC and Alkalinity). (D-F) Same as A-C but for July (winter) mean.

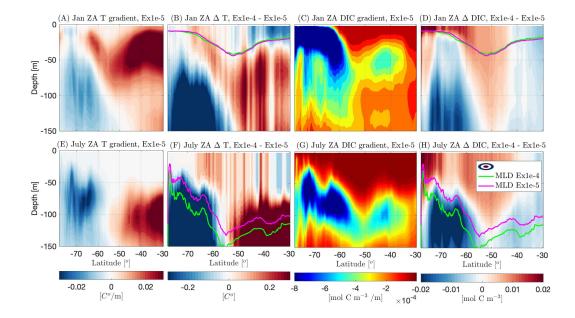


Figure 7: (A) January zonally averaged temperature vertical gradient (red implies increase in temperature towards the surface). (B) January change in temperature (Ex1e-4 - Ex1e-5). Mixed layer depth (MLD) for Ex1e-5 (pink) and Ex1e-4 (green) overlain. (C) January zonally averaged DIC vertical gradient. (D) Change in DIC concentration (Ex1e-4 - Ex1e-5). (E-H) As in A-D but for July mean.

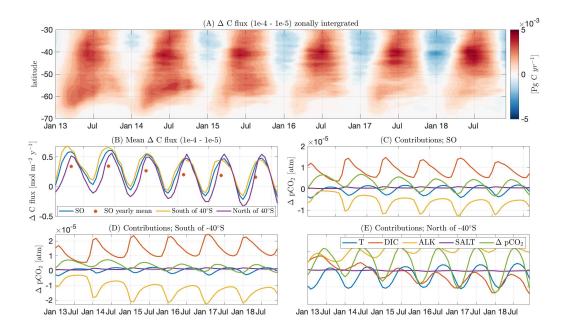


Figure 8: (A) Change in zonally integrated Carbon flux between Ex1e-4 and Ex1e-5 over the six-year time period of Jan 2013 to Dec 2018 (Red shows reduced uptake or increased outgassing in Ex1e-4). (B) Change in the mean carbon flux across the whole SO (blue), the SO North of 40°S (purple) and South of 40°S (yellow) for the same time period. The annual mean change for the whole SO for each year is shown (red star). Using the methodology of Takahashi et al. (2014) as discussed previously, the differences in Carbon flux between the two model runs over time can be attributed to changes in surface ocean pCO_2 (green lines) from alterations to temperature (blue lines), DIC (red liens), alkalinity (yellow lines) and salinity (purple lines). These contributions are shown for the whole SO (C), the SO south of 40°S (D), and the SO north of 40°S (E).

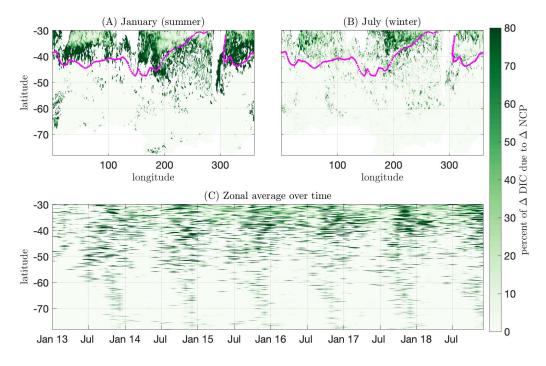


Figure 9: The percentage change of surface water DIC concentration due to changes in biological net community productivity (NCP). Surface water is defined here as waters down to a depth of 55m. Shown as a vertically integrated mean for (A) January (summer) and (B) July (winter). The mean location of the subtropical front, as defined by Orsi et al. (1995), is also shown in pink. (C) The zonal mean of the vertically integrated percentage change due to altered NCP is shown over time.

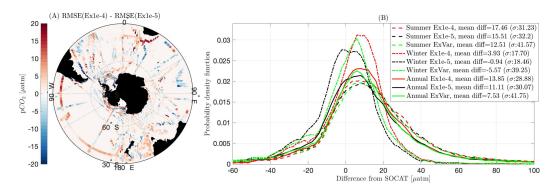


Figure 10: Comparison of modelled pCO_2 to observations from Surface Ocean CO₂ Atlas (SOCAT) between 2012 and 2018 (Bakker et al., 2016). (A) Comparison of the root mean squared error between Ex1e-4 and Ex1e-5. Red/blue shows regions where Ex1e-5/Ex1e-4 is closer to the observations. (B) Probability density function showing the misfit between observed carbon fluxes from SOCAT and the model outputs for pCO_2 in Ex1e-5 (black), Ex1e-4 (red), and ExVar (green).