

Supplementary material

A1. Mechanisms of CO₂ uptake in Sub-Antarctic region

The seasonal cycle of $(dpCO_2/dt)_{nonT}$ in the contemporary Sub-Antarctic region is broadly characterized by a maximum in mid-winter (JJA) and minima in late spring to early summer (NDJ) (Fig. 4e). The increase of $(dpCO_2/dt)_{nonT}$ from mid-autumn to winter coincides with the increase in $dDIC/dt$ (Fig. 5e), surface cooling (Fig. 6i), and deepening of the MLD (Fig. 6c). As the surface ocean cools, the buoyancy flux weakens and MLD deepens. This stimulates the entrainment of the DIC-rich subsurface waters resulting in the increase of $dpCO_2_{nonT}/dt$ during the cool seasons, peaking in winter (Fig. 4e). From early spring when light becomes available, the primary production removes ocean surface DIC to form organic matter. This decreases surface DIC concentrations ($dDIC/dt < 0$) and leads to the $(dpCO_2/dt)_{nonT}$ minima (Fig. 4e & 6a). NPP peak in ESMs coincides with a minimum in $dDIC/dt$ (Fig. 6a & 4e). Furthermore, the timing of NPP peaking coincides with a minimum in the apparent oxygen utilization rate ($dAOU/dt$) (Fig. 6g, h). AOU is defined as the difference between oxygen at saturation (pre-formed estimate; $O_{2, sat}$) and the in situ dissolved oxygen concentration (O_2); here, it is used to estimate near-surface (within the MLD) respiration. Negative $dAOU/dt$ magnitude when NPP is high reflects oxygen production during photosynthesis, whereas positive $dAOU/dt$ magnitudes can be used to roughly estimate the respiration or oxidation of organic matter back to DIC in the near-surface. Indeed, ESMs show positive $dAOU/dt$ at the tail of NPP maxima, and, AOU and DIC rates are aligned which indicates respiration (Fig. 6). Moreover, we find that particularly large $dAOU/dt$ magnitudes are shown by ESMs with extensive NPP magnitudes, (MPI models, Fig. S4 &); in these models, a significant amount of the organic matter formed during the growing season models is respired back to DIC within the MLD. It is worth noting that even if respiration occurs below the MLD, the MLD is still relatively shallow (~80m) when $dAOU/dt$ is at its peak (Fig. 5). It is, therefore, unlikely that a significant amount of organic matter permanently leaves the winter mixed layer, it likely that a significant organic carbon is respired below the winter MLD.

Tables and figures

Table S1. The list of the nine Earth System models used in this study.

Earth System Model	Reference

CanESM5	Swart et al., 2019 ¹
CMCC-ESM2	Lovato et al., 2022 ²
CESM2-WACCM	Danabasoglu et al., 2020 ³
IPSL-CM6A-LR	Dufresne et al., 2013 ⁴
NorESM2-LM	Bentsen et al., 2013 ⁵
MPI-ESM1-2-LR	Mauritsen et al., 2019 ⁶
MPI-ESM1-2-HR	Müller et al., 2018 ⁷
UKESM1-0-LL	Sellar et al., 2019 ⁸
AWI-CM1	Semmler et al., 2019 ⁹

Table S2. Details of observation-based surface pCO₂ products collated in the SeaFlux ensemble product (Fay and Gregor et al. 2021). Note that the abbreviated names all contain the institution(s) and then an abbreviation for the method. Further, NN = neural network, SOM = self-organising map, MLR = multi-linear regression, GBDT = gradient boosted decision trees, SVR = support vector regression. For proxy variables, SST = sea surface temperature, SSS = sea surface salinity, SSH = sea surface height, MLD = mixed layer depth, Chl-a = Chlorophyll-a, SLP = sea level pressure, WIND = wind speed at 10m, xCO₂ = atmospheric CO₂ concentration, LAT = latitude, LON = longitude, MON = month. Note that some of these variables have been transformed in the original study but have simplified for brevity in this table. Please see the original study for full details.

Abbreviated name	Brief method description	Proxy variables	References
JMA-MLR	A regional regression approach where MLR is applied to 44 defined regions	xCO ₂ , SST, SSS, MLD, Chl-a, SLP, WIND	Iida et al. (2020) ¹⁰
NIES-nn	A NN approach that uses no clustering or regions	MON, LAT, LON, SST, SSS, CHL	Zeng et al. (2014) ¹¹
CMEMS-LSCE-	Two-step NN that estimates seasonal	SST, SSS, SSH, MLD, Chl-	Denvil-Sommer et al.

FFNN	variability and the anomalies there from	a, xCO ₂	(2019) ¹²
MPI-SOMFFN	A two-step approach that first clusters data with SOM, and then performs a regression per cluster using NN.	xCO ₂ , SST, SSS, MLD, Chl-a	Landschützer et al. (2014) ¹³
CSIR-ML6	An ensemble two-step cluster-regression approaches. Clustering is performed with K-means, and regression: NN, GBDT, and SVR.	xCO ₂ , SST, SSS, MLD, Chl-a, WIND, MON	Gregor et al. (2019) ¹⁴
Jena-MLS	Mixed Layer Scheme: A bayesian approach that matches pCO ₂ observations to the mixed layer DIC budget.	SST, SSS, and other variables (see Fig 1 in reference)	Rödenbeck et al. (2013) ¹⁵

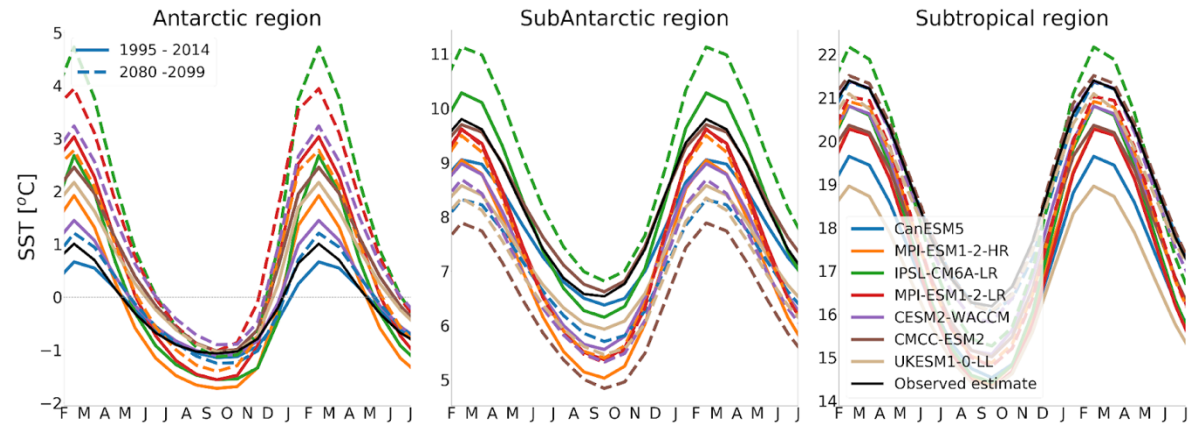


Figure. S1 The seasonal cycle of sea surface temperature. The left panel is the Antarctic region, the middle panel is the Sub-Antarctic region, and the right is the subtropical region.

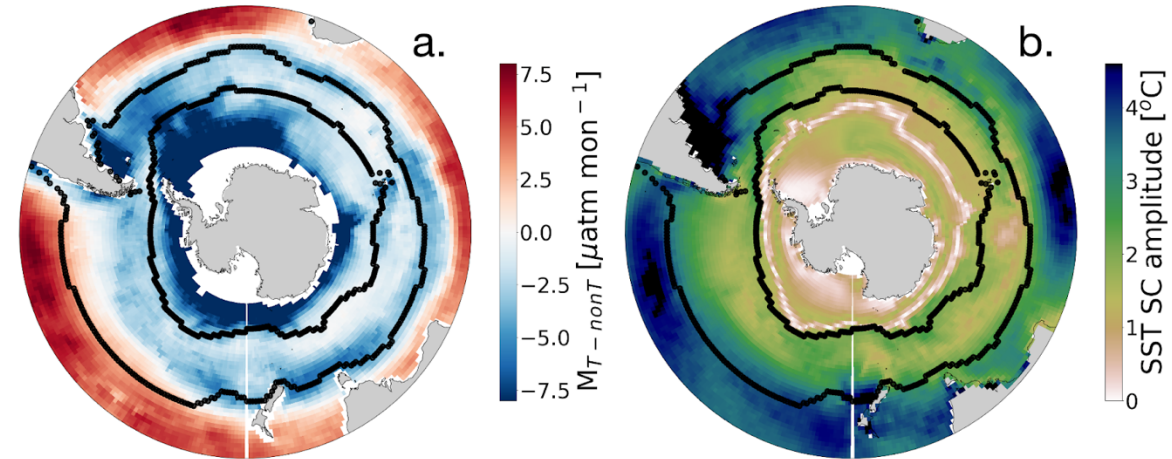


Figure S2. The left panel shows the annual mean of the observation-based estimate of MT-nonT (eq. 3), red (blue) denotes regions where the thermal (nonthermal) component is leading driving monthly pCO₂ changes, and blue denotes nonthermal processes (biological and mixing) are leading the monthly pCO₂ changes. The right panel shows the seasonal amplitude {summer (DJF) vs. winter (JJA)} of sea surface temperature in the Southern Ocean.

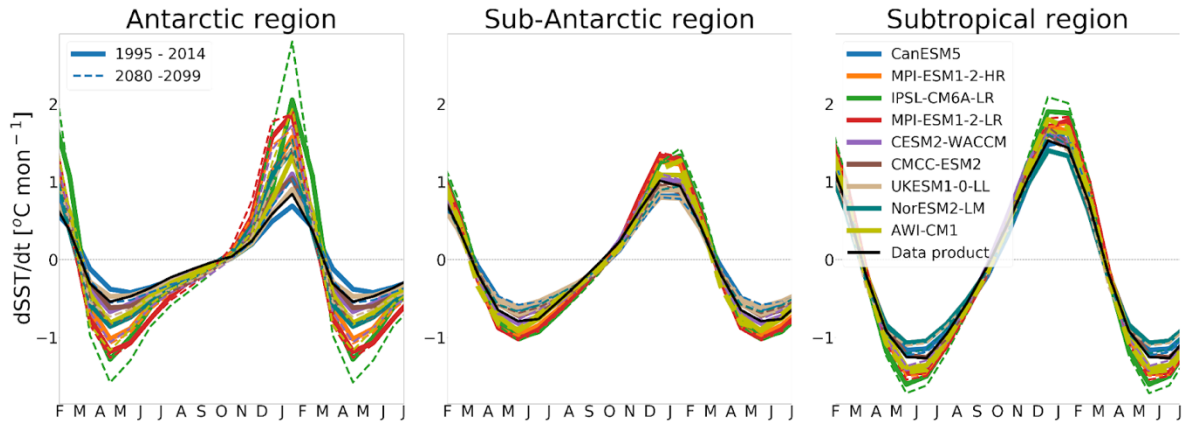


Figure S3. The seasonal cycle of the monthly rate of sea surface temperature. The left panel is the Antarctic region, the middle panel is the Sub-Antarctic region, and the right is the subtropical region.

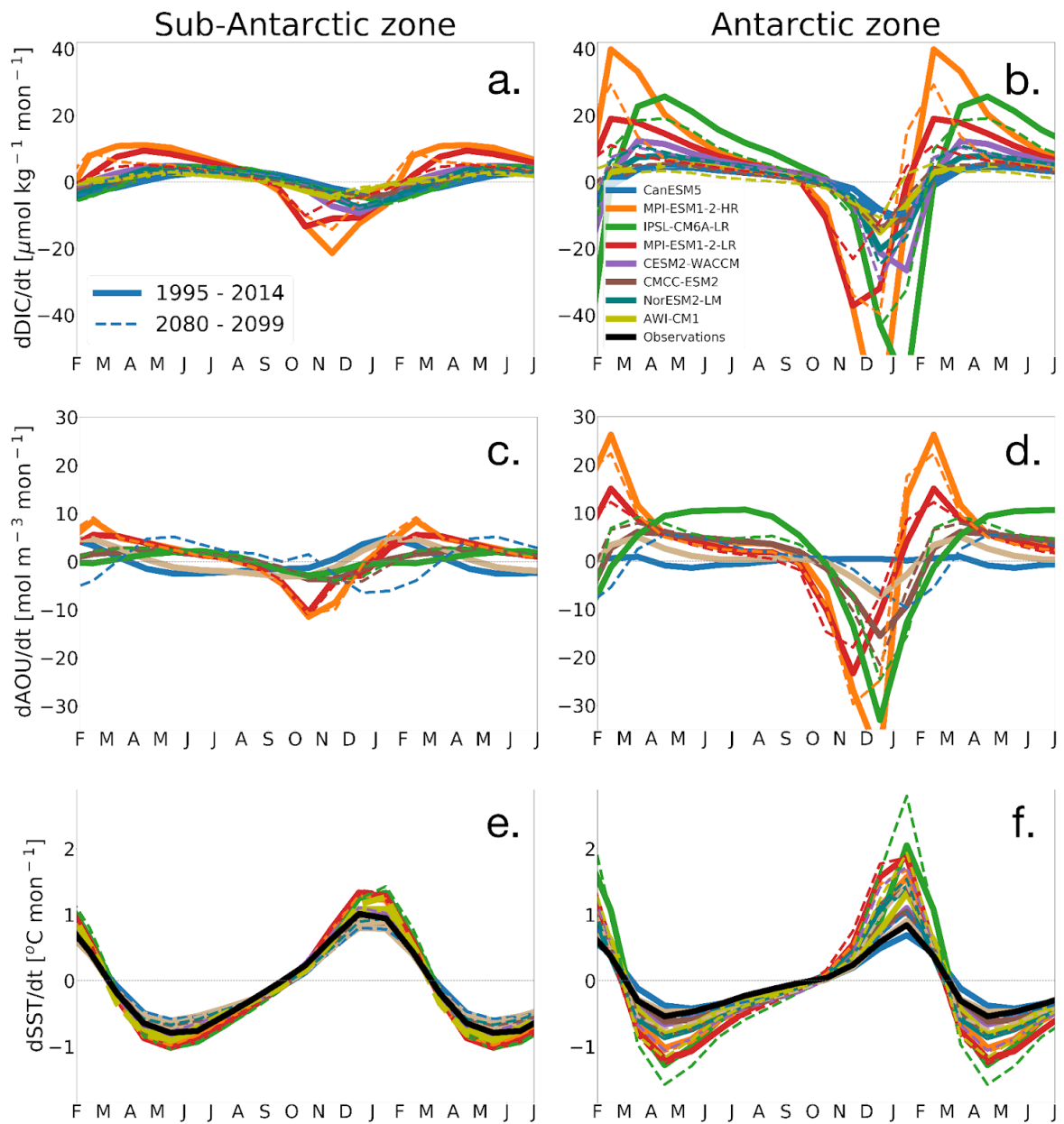


Figure S4. The monthly rate of change of surface dissolved inorganic carbon ($dDIC/dt$, top row panel), the monthly rate of change of the oxygen utilization rate ($dAOU/dt$, second row panel) and the monthly rate of surface temperature change ($dSST/dt$, bottom panel) in the Sub-Antarctic region (left panel) and Antarctic region (right panel). The solid lines are the present climate (1995 – 2014) and dotted lines are the end of the 21st century (2080 – 2099).

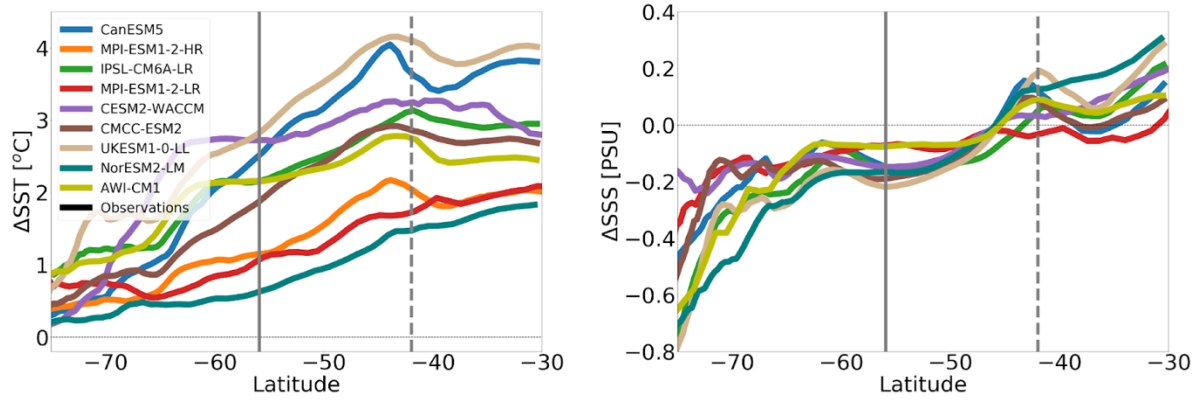


Figure S5 Zonal averages of sea surface temperature and salinity change (2080-2099 vs 1995-2014).

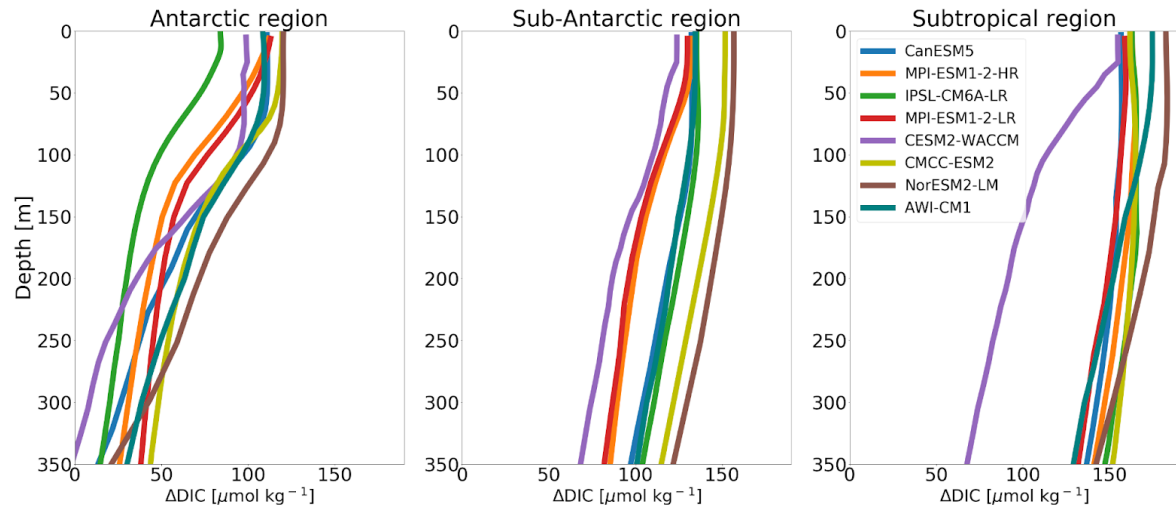


Figure S6: Vertical profiles of net DIC change (2080 - 2099 vs 1995 - 2014 vs. 2080 - 2099). The left panel is the Antarctic region, the middle panel is the Sub-Antarctic region, and the right is the subtropical region.

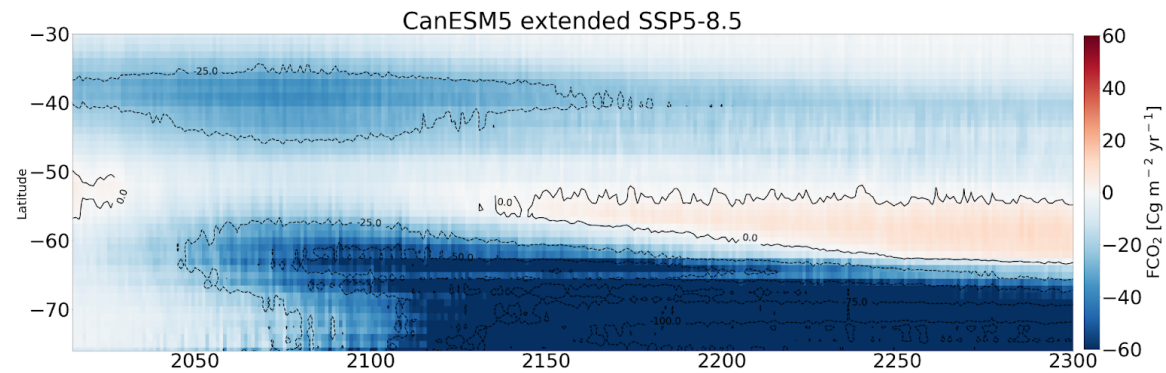


Figure S7. Annual and zonal mean air-sea CO_2 fluxes (FCO_2) south of 30° in CanESM5 for SSP5-8.5 climate scenario.

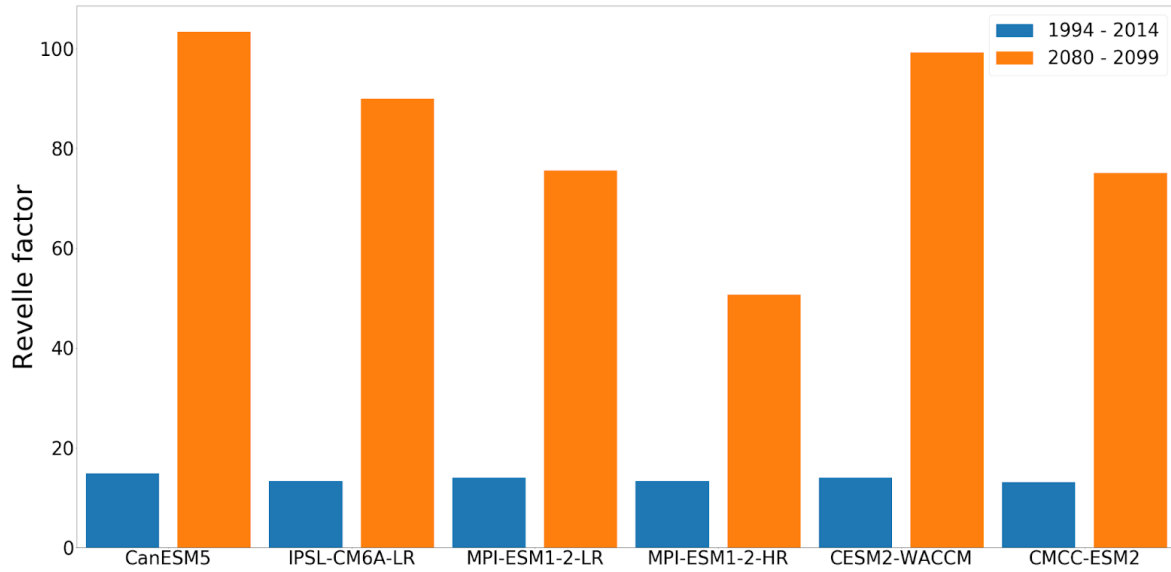


Figure S8 Revelle factor estimate South of 30°S for the present climate (1995 - 2014) and the end of the 21st century (2080 - 2099), $R = \text{DIC}/$

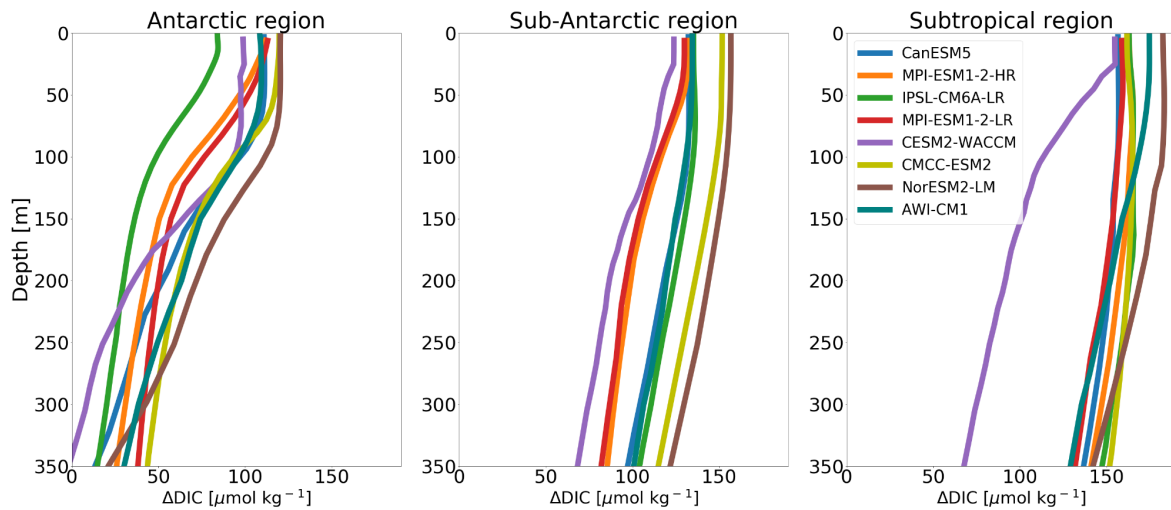


Figure S9: Vertical profiles of net DIC change (2080 - 2099 vs 1995 - 2014). The left panel is the Antarctic region, the middle panel is the Sub-Antarctic region, and the right is the subtropical region.

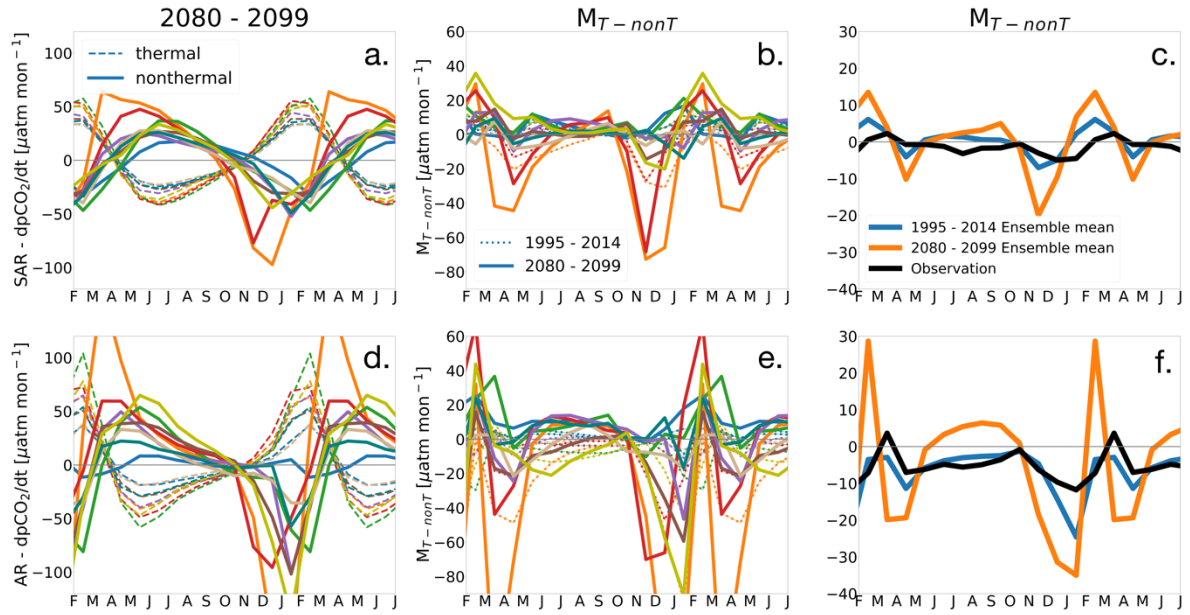


Fig. S10. The seasonal cycle of the rate of change of the thermal ($\text{dpCO}_2^{\text{thermal}}/\text{dt}$) and nonthermal ($\text{dpCO}_2^{\text{nonthermal}}/\text{dt}$) ocean pCO_2 components for the present climate (left panel), the absolute difference between thermal and non-thermal components (middle panel; $M_{T-\text{non}T}$) in the Sub-Antarctic region (SAZ, top panel) and Antarctic region (AZ, bottom panel). The right panel show the ensemble mean of the $M_{T-\text{non}T}$ for present climate (blue), end of the 21st century (orange) and observations (black). The colors legend for each ESM in the first two columns follow the previous figure.

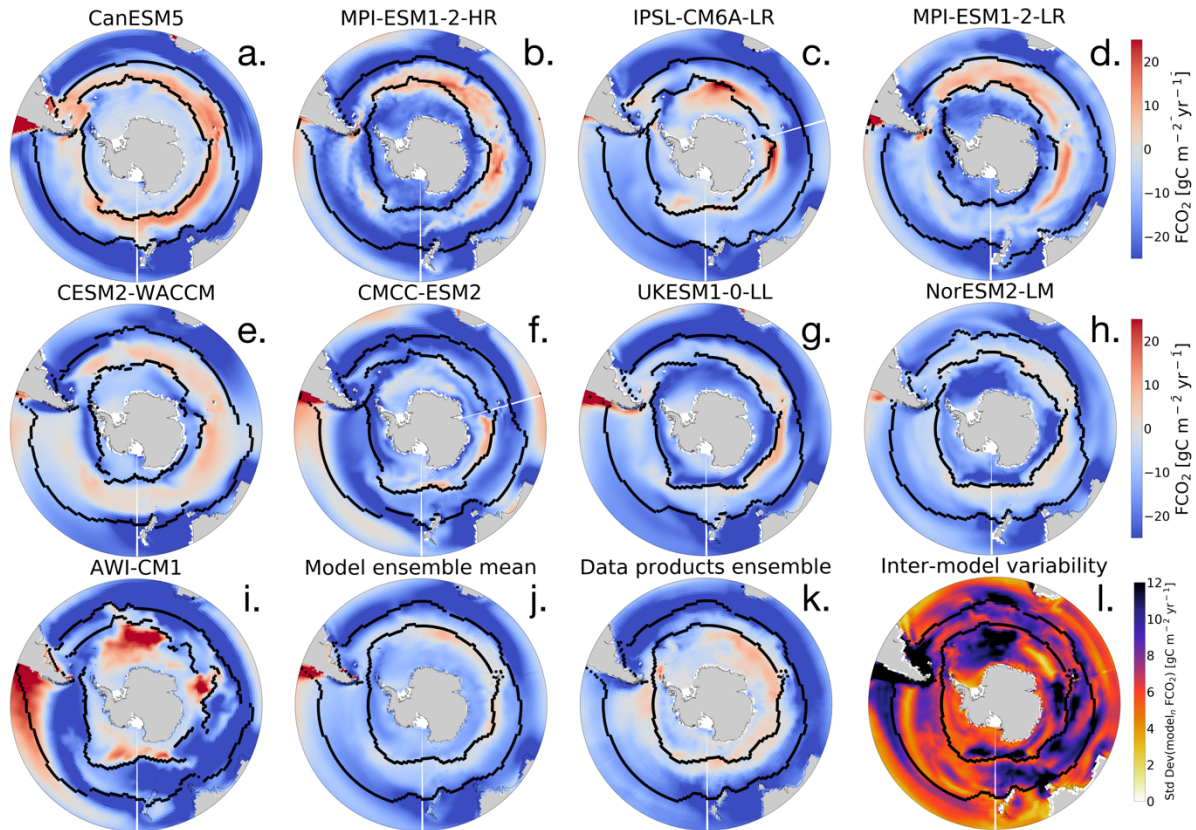


Figure S11. The climatological mean of air-sea CO_2 flux (FCO_2) in the Southern Ocean (south of 30°S) for nine CMIP6 ESMs for historical period (1995 - 2014), negative indicates flux into the ocean and positive outgassing for (a- i), the multi model mean (j), the ensemble mean of six pCO_2 products (k) and the ESM inter model variability (l), all given in $\text{gC m}^{-2} \text{yr}^{-1}$. The fronts are defined according to Orsi et al. (1995) as black lines with

the Subtropical Front to the North and the Polar Front to the South. The zones are defined as the subtropical region north of the Subtropical Front (outer line), the Sub-Antarctic region between the two fronts and the Antarctic region to the south of the Polar Front (inner line).

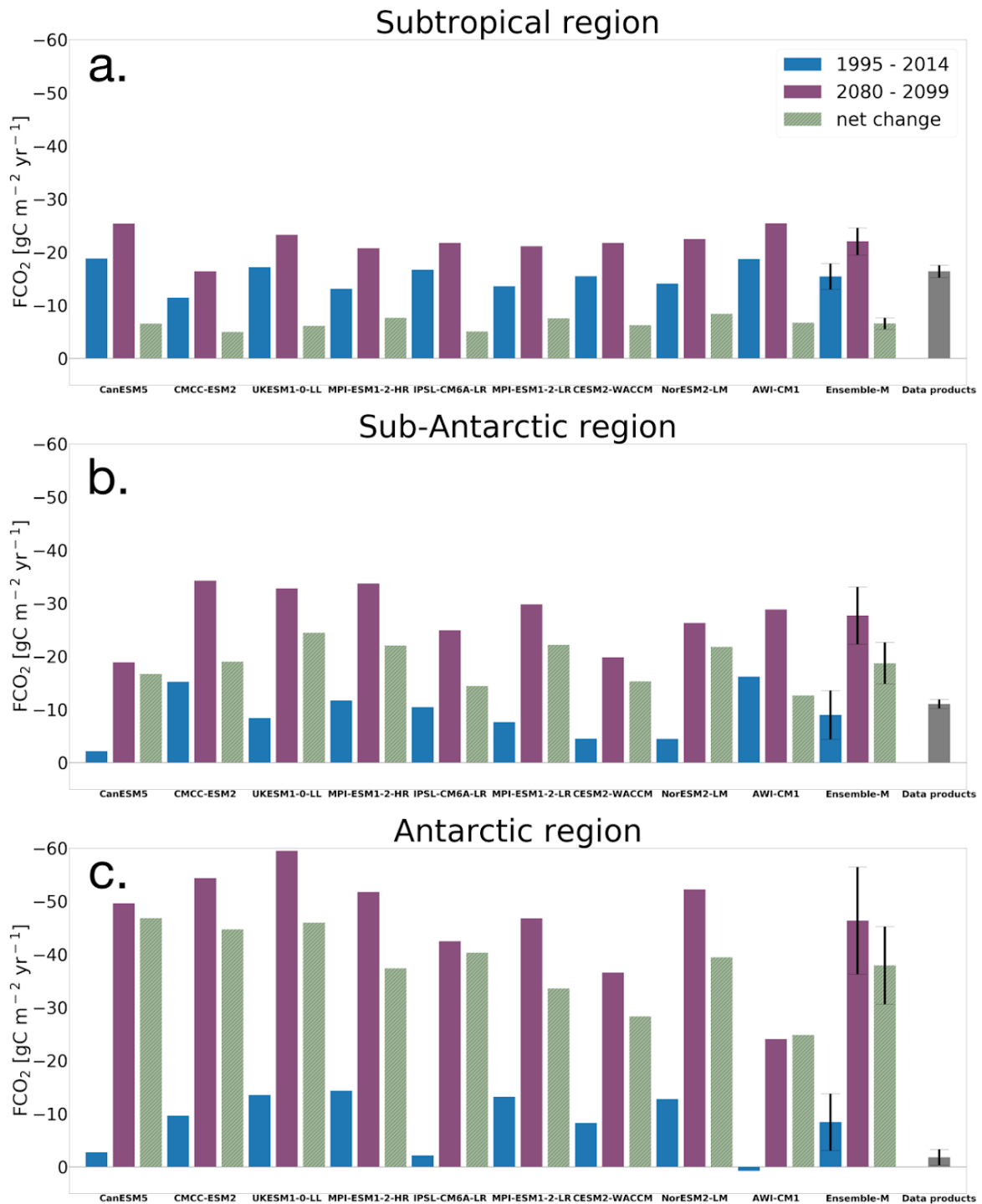


Figure S12 Annual mean air-sea CO₂ fluxes (FCO₂) in the Southern Ocean (south of 30°S) for nine CMIP6 ESMs, the multi model mean, and ensemble mean of six pCO₂ products, given in gC m⁻² yr⁻¹. The error bars reflect the standard deviation.

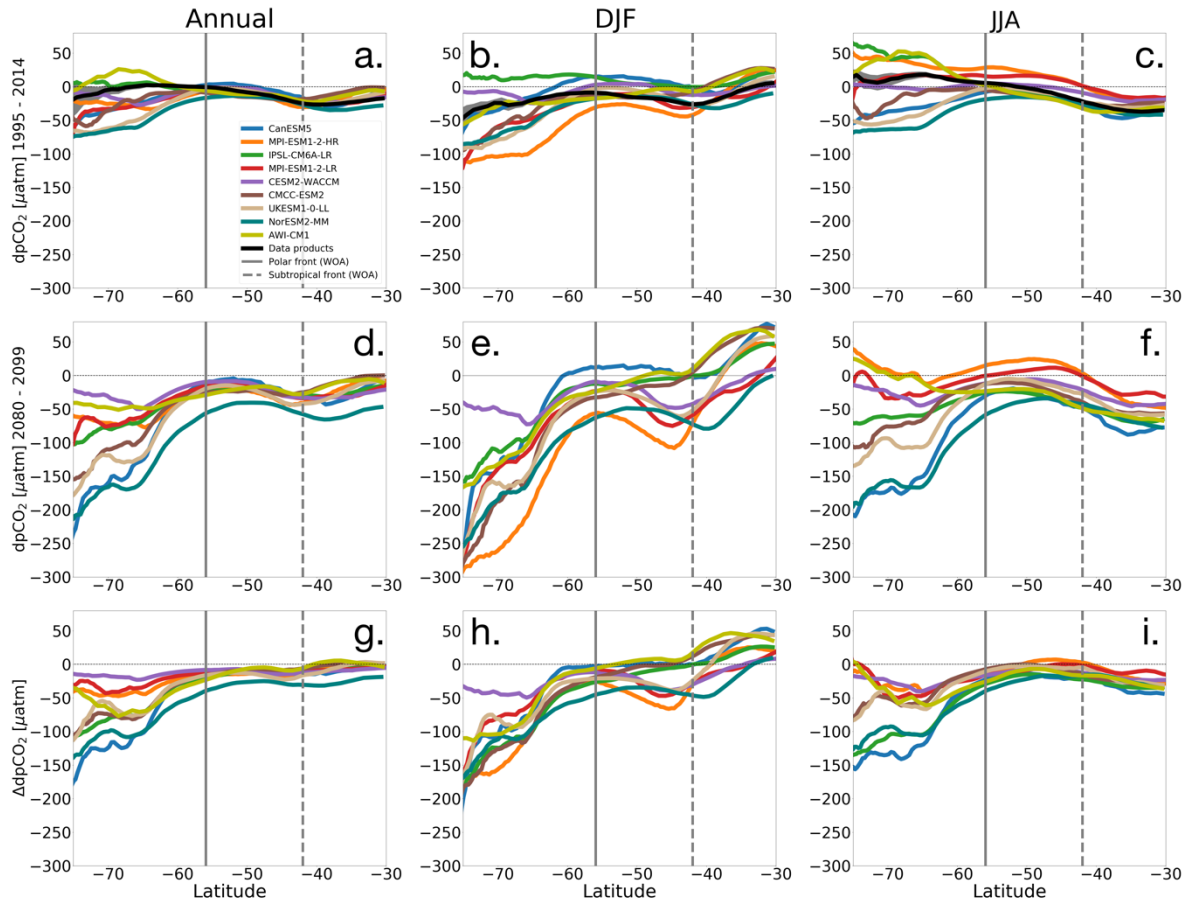


Figure S13. Zonally averaged $\Delta p\text{CO}_2$ in the Southern Ocean (south of 30°S) for nine CMIP6 ESMs, and ensemble mean of six $p\text{CO}_2$ products, given in μatm units. The top panels show the present climate averages, middle panels show the end of the century (2080 – 2099) averages and net changes are shown in the bottom panel. The first, second, and third column depict the annual mean, the austral summer (DJF), and austral winter (JJA) averages, respectively. The vertical grey lines denote frontal positions, solid line the Polar Front and dotted the Subtropical Front.

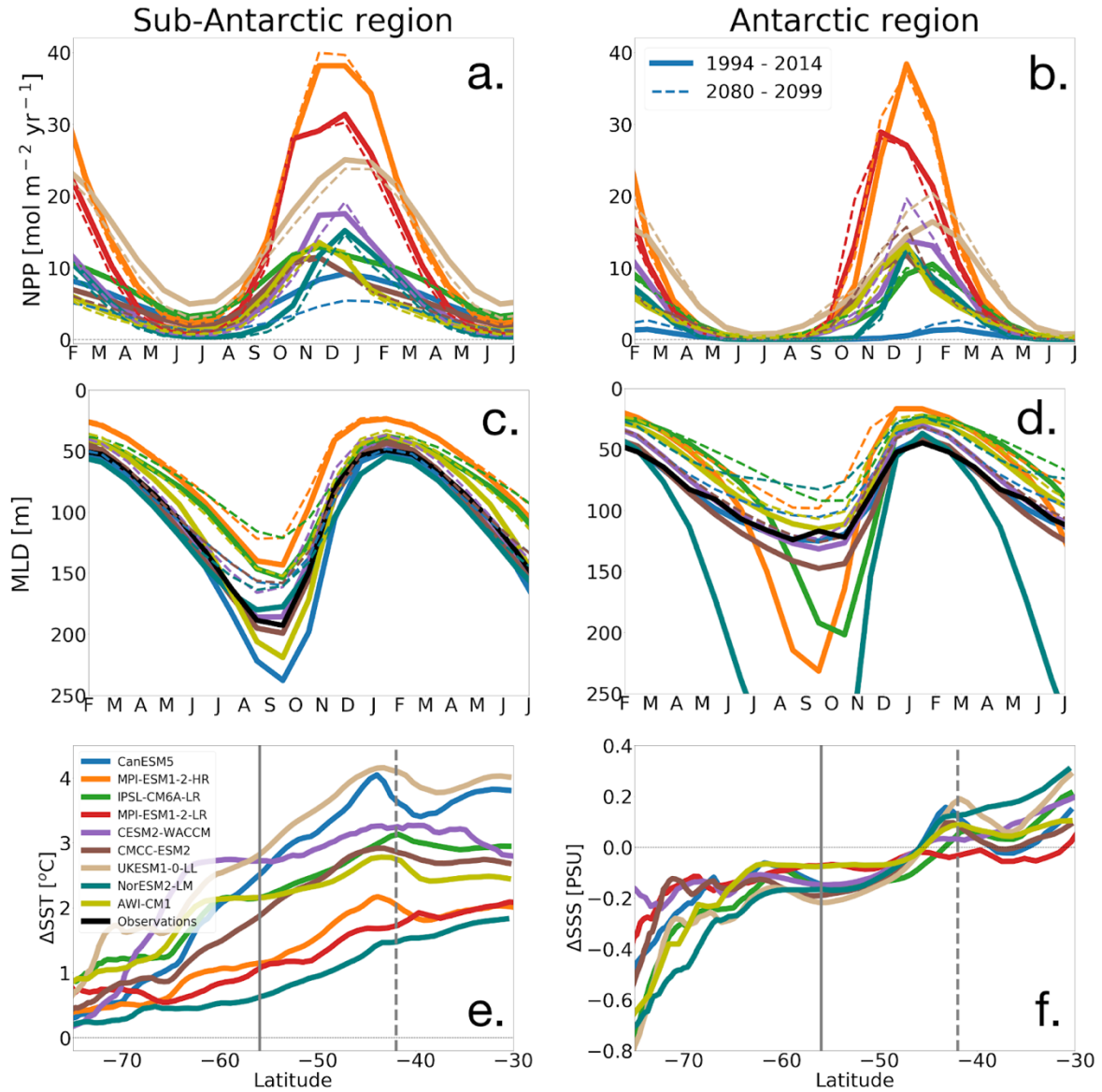


Figure S14 The seasonal cycle of net primary production (NPP, top panel), the seasonal cycle of the ocean mixed layer depth (MLD, second row panel). The solid lines are the present climate (1995 – 2014) and dotted lines are the end of the 21st century (2080 – 2099). The bottom panel shows zonal averages of sea surface temperature and surface salinity net change (2080-2099 vs 1995-2014).

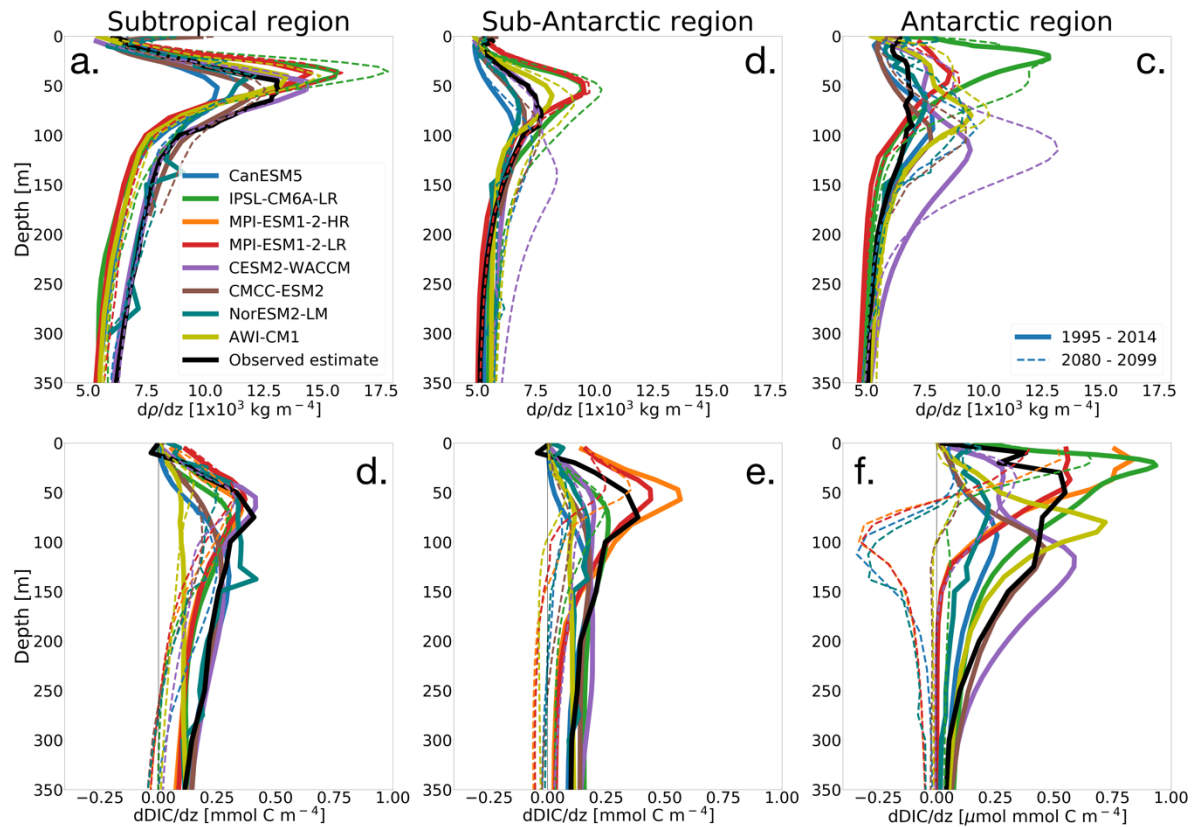


Figure S15. Vertical density gradient (dp/dz , top panel) and dissolved inorganic carbon vertical gradient ($dDIC/dz$, bottom panel) for Subtropical (left panel), Sub-Antarctic (middle panel) and Antarctic (right panel) regions. Solid lines depict the present climate (1995 – 2014) and dotted lines the end of the 21st century (2080 – 2099).

References

- Swart, N. C. *et al.* The Canadian Earth System Model version 5 (CanESM5.0.3). *Geosci Model Dev* **12**, 4823–4873 (2019).
- Lovato, T. *et al.* CMIP6 Simulations With the CMCC Earth System Model (CMCC-ESM2). *J Adv Model Earth Syst* **14**, (2022).
- Danabasoglu, G. *et al.* The Community Earth System Model Version 2 (CESM2). *J Adv Model Earth Syst* **12**, (2020).
- Vial, J., Dufresne, J. L. & Bony, S. On the interpretation of inter-model spread in CMIP5 climate sensitivity estimates. *Clim Dyn* **41**, 3339–3362 (2013).
- Bentsen, M. *et al.* The Norwegian Earth System Model, NorESM1-M – Part 1: Description and basic evaluation of the physical climate. *Geosci Model Dev* **6**, 687–720 (2013).
- Mauritsen, T. *et al.* Developments in the MPI-M Earth System Model version 1.2 (MPI-ESM1.2) and Its Response to Increasing CO₂. *J Adv Model Earth Syst* **11**, 998–1038 (2019).
- Müller, W. A. *et al.* A Higher-resolution Version of the Max Planck Institute Earth System Model (MPI-ESM1.2-HR). *J Adv Model Earth Syst* **10**, 1383–1413 (2018).

8. Sellar, A. A. *et al.* UKESM1: Description and Evaluation of the U.K. Earth System Model. *J Adv Model Earth Syst* **11**, 4513–4558 (2019).
9. Semmler, T. *et al.* Simulations for CMIP6 With the AWI Climate Model AWI-CM-1-1. *J Adv Model Earth Syst* **12**, (2020).
10. Iida, Y., Takatani, Y., Kojima, A. & Ishii, M. Global trends of ocean CO₂ sink and ocean acidification: an observation-based reconstruction of surface ocean inorganic carbon variables. *J Oceanogr* **77**, 323–358 (2021).
11. Zeng, J., Nojiri, Y., Landschützer, P., Telszewski, M. & Nakaoka, S. A global surface ocean fCO₂ climatology based on a feed-forward neural network. *J Atmos Ocean Technol* **31**, 1838–1849 (2014).
12. Denvil-Sommer, A., Gehlen, M. & Vrac, M. Observation system simulation experiments in the Atlantic Ocean for enhanced surface ocean pCO₂ reconstructions. *Ocean Science* **17**, 1011–1030 (2021).
13. Landschützer, P., Gruber, N., Bakker, D. C. E. & Schuster, U. Recent variability of the global ocean carbon sink. *Global Biogeochem Cycles* **28**, 927–949 (2014).
14. Gregor, L., Lebehot, A. D., Kok, S. & Scheel Monteiro, P. M. A comparative assessment of the uncertainties of global surface ocean CO₂ estimates using a machine-learning ensemble (CSIR-ML6 version 2019a)-Have we hit the wall? *Geosci Model Dev* **12**, 5113–5136 (2019).
15. Rödenbeck, C. *et al.* Global surface-ocean pCO₂ and sea-Air CO₂ flux variability from an observation-driven ocean mixed-layer scheme. *Ocean Science* **9**, 193–216 (2013).