

Global Biogeochemical Cycles

Supporting Information for

Drivers of air-sea CO₂ flux in the subantarctic zone revealed by time series observations

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Additional Supporting Information (Files uploaded separately)

Captions for Movies S1 to S6

Movie S1: 8-day SSH anomalies (left panel) and SST (right panel) maps of the study region for 2009. The time stamp is the median of the 8 day period. The magenta triangles denote the location of Argo floats during the same time window. The general location of the SOTS station is indicated by the red pentagram, and the white polygon represents the study area.

Movie S2: Same as Movie S1, but for 2010.

Movie S3: Same as Movie S1, but for 2015.

Movie S4: Same as Movie S1, but for 2016.

Movie S5: Same as Movie S1, but for 2019.

Movie S6: Same as Movie S1, but for 2020.

Section 1

This section includes 5 figures, 1 table and .mp4 movies (uploaded separately). The table lists the SOTS annual voyages on which the HPLC samples used in our study were collected. The .mp4 movies illustrate the movement of water masses during the years of the case studies.

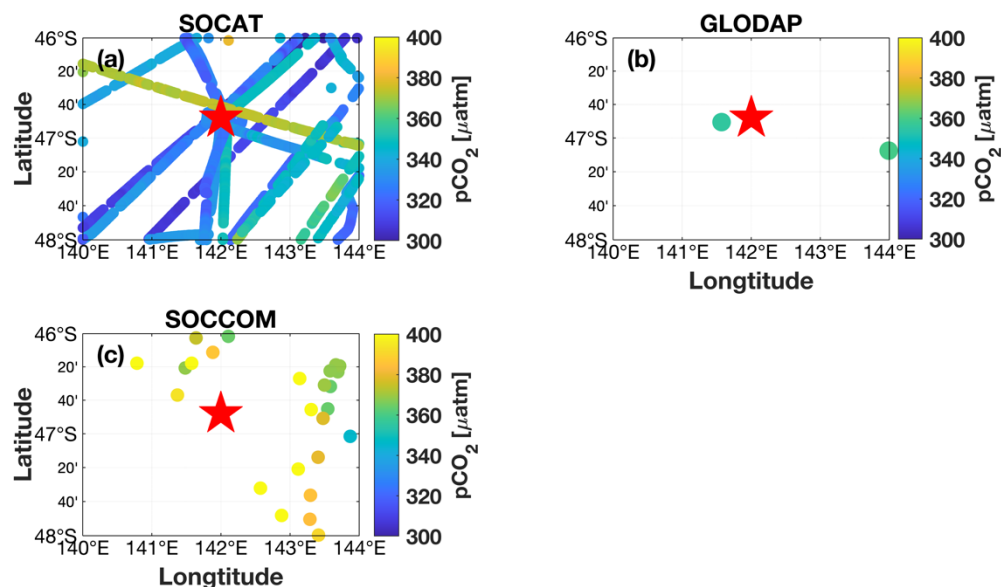


Figure S1. In-situ measurement locations of three different products: (a) SOCAT; (b) GLODAP; (c) SOCCOM, from January 2004 to December 2021, in the white polygon in Figure 1. The red pentagram denotes the general SOTS location.

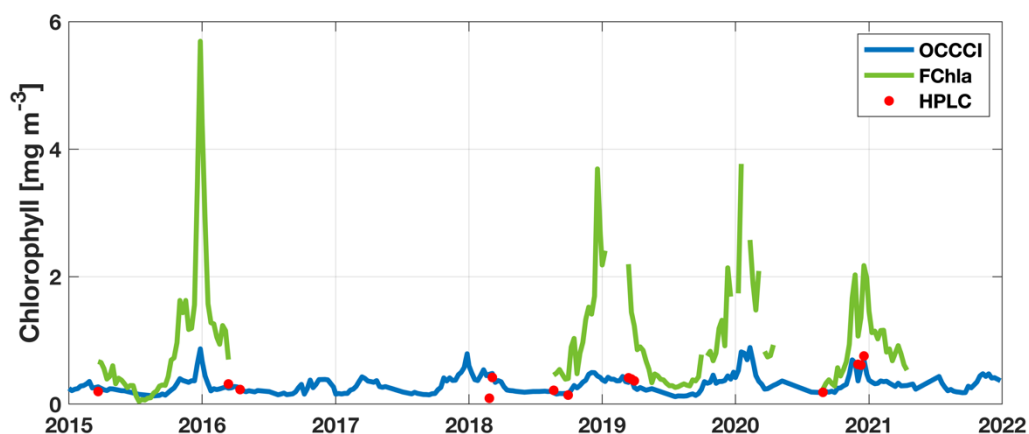


Figure S2. Comparison of different chlorophyll concentration measurements. OCCCI, satellite measurements; FChla, SOTS estimate based on fluorescence measurements at 30m depth; HPLC, shipboard in-situ HPLC data.

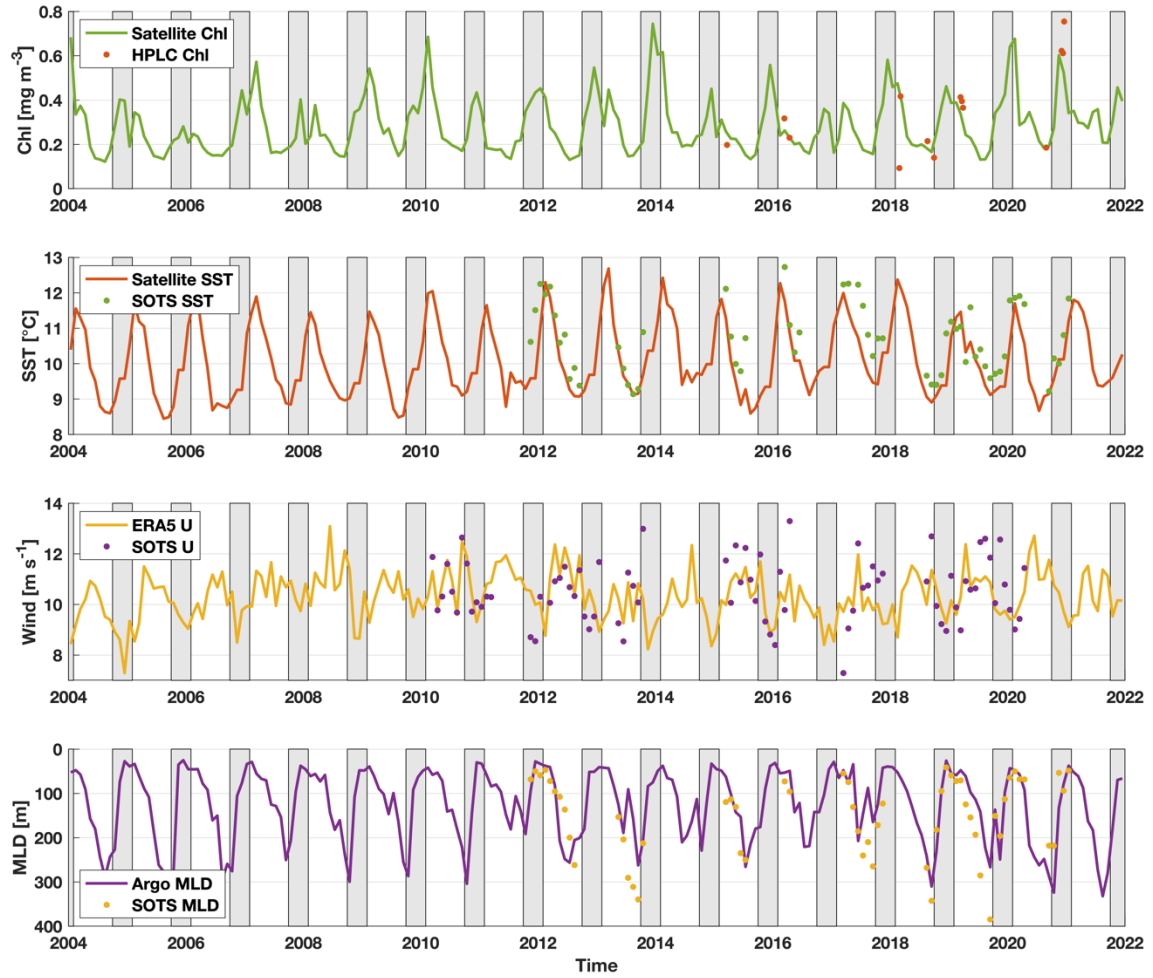


Figure S3. Time series of input parameters (auxiliary data, see section 2.2) to the MLR model at monthly temporal resolution, (a) chlorophyll concentration, (b) surface temperature, (c) 10 m wind speed, (d) mixed layer depth spanning January 2004 to December 2021. Gray shading denotes autotrophic season, October to February.

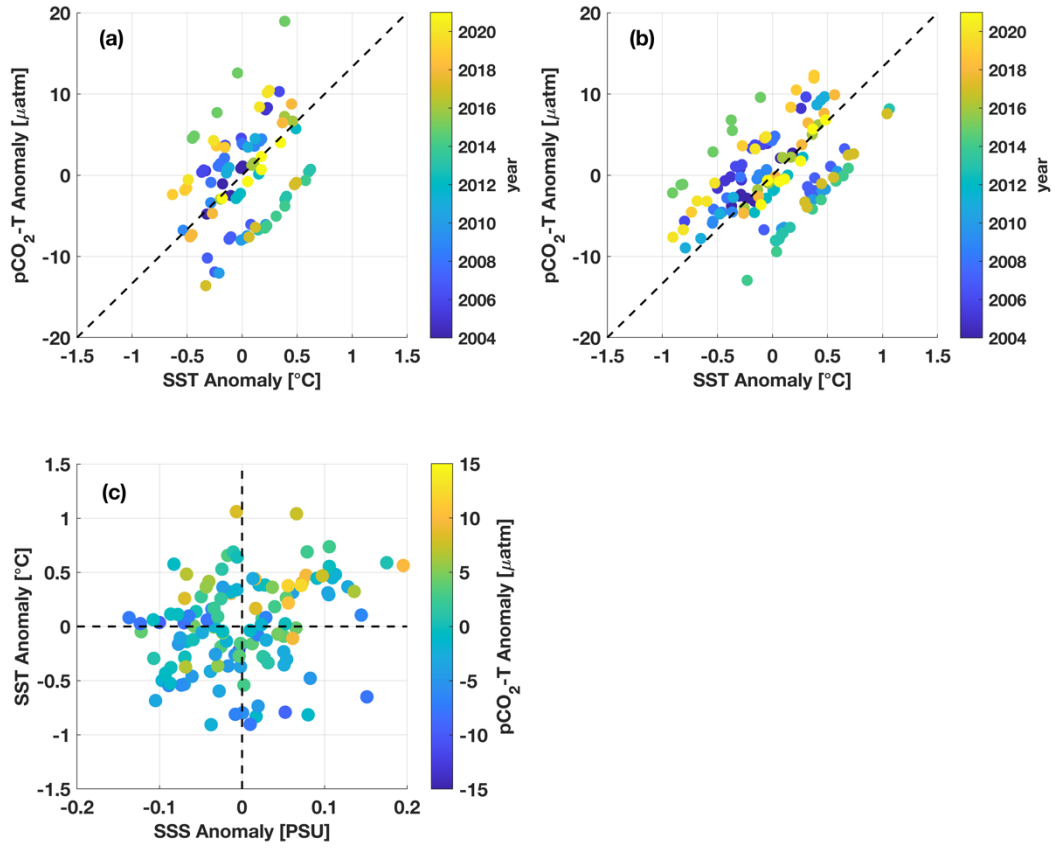


Figure S4. The relationship between anomalies of pCO₂-T and SST in (a) autotrophic seasons and (b) heterotrophic seasons. Relationship of (c) pCO₂-T anomalies with respect to combination of SST and SSS, during the heterotrophic seasons. During 2013 to 2015, the pCO₂-T anomalies are scattered on the edge of the cloud, this is because pCO₂-T was very low in 2013 but increased dramatically in 2015.

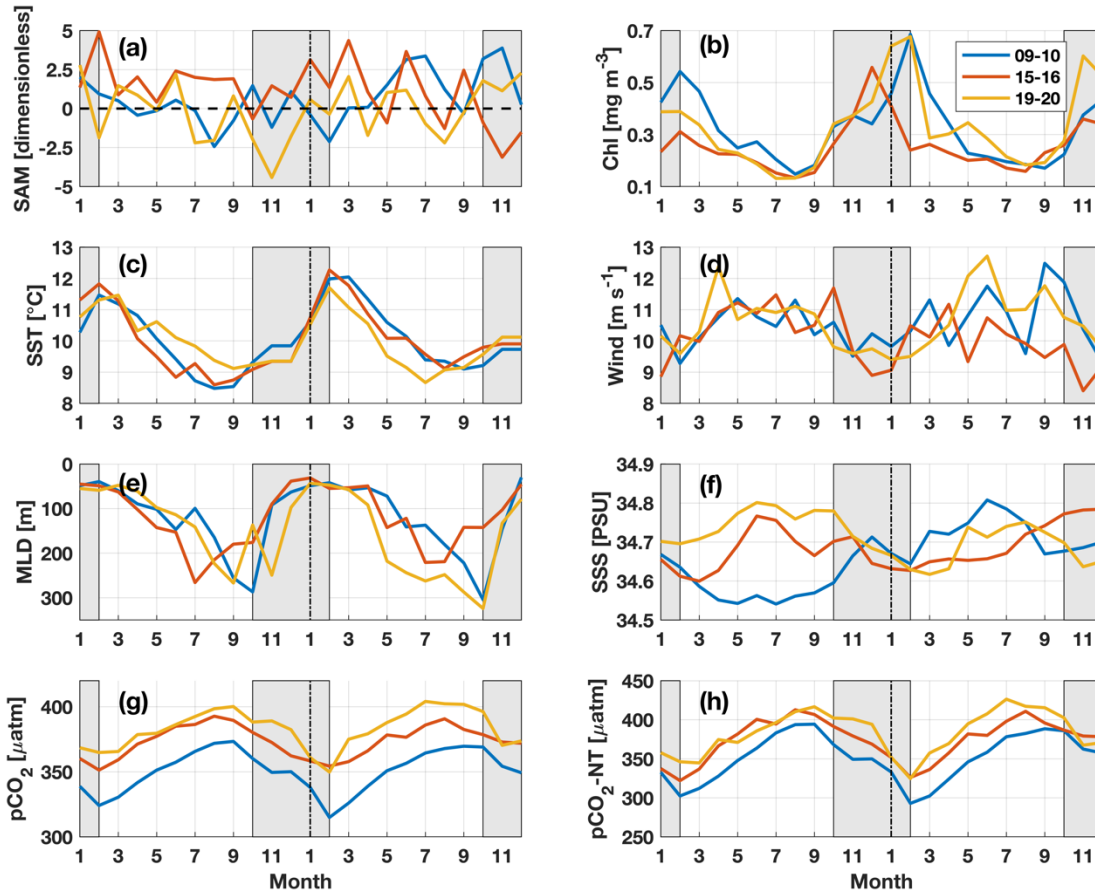


Figure S5. Time series of (a) SAM index, (b) chlorophyll concentration, (c) surface temperature, (d) 10 m wind speed, (e) mixed layer depth, (f) surface salinity, (g) sea water pCO₂-MLR, and (h) pCO₂-NT during the period of case studies. Grey shading indicates the autotrophic season (October to February).

Table S1. Citation details for the SOTS annual voyage reports on which HPLC samples were collected.

SOTS annual report	Voyages	Citation
Southern Ocean Time Series SOTS Annual Reports: 2013/2015	IN2015_V01	(Wynn-Edwards et al., 2021)
Southern Ocean Time Series SOTS Annual Reports: 2016/2017	IN2016_V02	(Wynn-Edwards et al., 2020a)
Southern Ocean Time Series SOTS Annual Reports: 2018/2019	IN2018_V02 IN2018_V04 IN2018_V07 IN2019_V02	(Wynn-Edwards et al., 2020b)

Southern Ocean Time Series SOTS Annual Reports: 2020/2021	IN2020_V09 IN2021_V02	(Wynn-Edwards et al., 2022)
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Section 2

This section includes one figure and one table to compare and explain in more detail the non-thermal changes in $p\text{CO}_2$ derived via two different methods. We refer to $p\text{CO}_2\text{-NT}$ calculated using eq. 4 from Takahashi et al. (2002) as $p\text{CO}_2\text{-NT}[\text{Takahashi}]$. We refer to $p\text{CO}_2\text{-NT}$ calculated as a residual (i.e. $p\text{CO}_2\text{-NT} = p\text{CO}_2\text{-MLR} - p\text{CO}_2\text{-T}$) as $p\text{CO}_2\text{-NT}[\text{Residual}]$. The magnitudes of the seasonal cycle of $p\text{CO}_2\text{-NT}[\text{Takahashi}]$ (mean: $86.80 \mu\text{atm}$, standard deviation: $11.49 \mu\text{atm}$) and $p\text{CO}_2\text{-NT}[\text{Residual}]$ (mean: $88.77 \mu\text{atm}$, standard deviation: $12.21 \mu\text{atm}$) are very similar (Figure S6), as is the correlation with the SAM index (Table S2). By adding the annual mean $p\text{CO}_2$ to the $p\text{CO}_2\text{-NT}[\text{Residual}]$, we obtain a result very close to $p\text{CO}_2\text{-NT}[\text{Takahashi}]$ (Figure S6c). This suggests that the difference between $p\text{CO}_2\text{-NT}[\text{Residual}]$ and $p\text{CO}_2\text{-NT}[\text{Takahashi}]$ is mainly caused by the annual mean $p\text{CO}_2$ in eq. 2. In this study, we followed the Takahashi et al. (2002) method because it already contained the decadal growth trend information, but we believe that these two approaches would yield consistent results. So we simply follow the empirical method of Takahashi et al. (2002), and we refer to $p\text{CO}_2\text{-NT} [\text{Takahashi}]$ as $p\text{CO}_2\text{-NT}$.

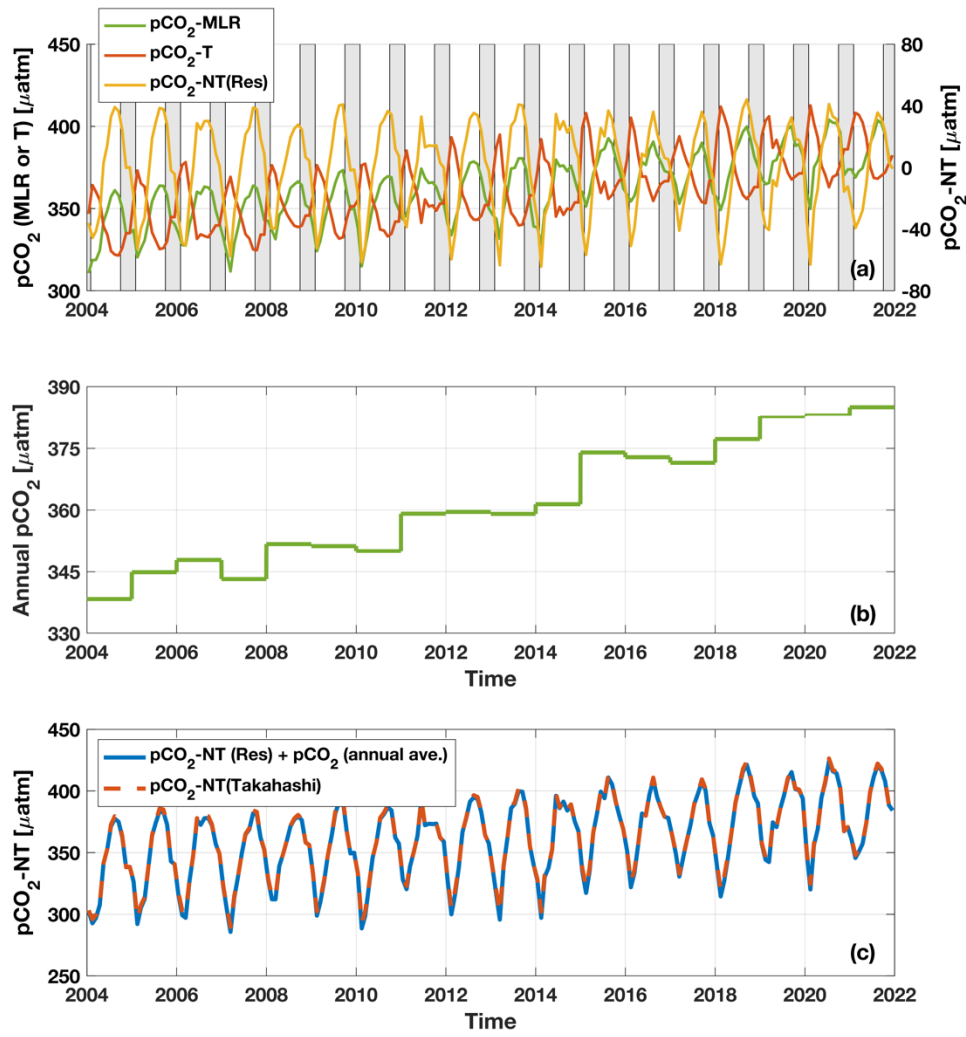


Figure S6. (a) Time series of pCO₂-MLR decomposed into thermal components using eq.2 and pCO₂-NT[Residual]. Gray shading denotes the autotrophic season, October to February. (b) Time series of annual average pCO₂, which was used in equations. 3 and 4. (c) Comparison of pCO₂-NT[Takahashi] and pCO₂-NT[Residual] plus the annual average pCO₂.

Table S2. The correlation of the two pCO₂-NT parameters with the SAM index for 2004 to 2021.

	r	p	Time lag
pCO ₂ -NT [Takahashi]	0.17	0.01	4 months
pCO ₂ -NT [Residual]	0.14	0.04	4 months

Reference list

- Takahashi, T., Sutherland, S. C., Sweeney, C., Poisson, A., Metzl, N., Tilbrook, B., et al. (2002). Global sea-air CO₂ flux based on climatological surface ocean pCO₂, and seasonal biological and temperature effects. *Deep-Sea Research Part II: Topical Studies in Oceanography*, 49(9–10), 1601–1622. [https://doi.org/10.1016/S0967-0645\(02\)00003-6](https://doi.org/10.1016/S0967-0645(02)00003-6)
- Wynn-Edwards, C. A., Davies, D. M., Eriksen, R. S., Jansen, P., Trull, T. W., & Shadwick, E. H. (2020a). *Southern Ocean Time Series SOTS Annual Reports: 2016/2017 Report 2. Samples Version 1.0*. <https://doi.org/10.26198/mbf0-ry85>
- Wynn-Edwards, C. A., Davies, D. M., Jansen, P., Shadwick, E. H., & Trull, T. W. (2020b). *Southern Ocean Time Series SOTS Annual Reports: 2018/2019 Report 2. Samples Version 1.0*. <https://doi.org/10.26198/r9ny-r549>
- Wynn-Edwards, C. A., Davies, D. M., Eriksen, R., Jansen, P., Bray, S. G., Shadwick, E. H., & Trull, T. W. (2021). *Southern Ocean Time Series SOTS Annual Reports: 2013/2015 Report 2. Samples Version 1.0*. <https://doi.org/10.26198/6504-se58>
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