# **Rebuttal to "Interannual drivers of the seasonal cycle of CO<sub>2</sub> fluxes in the Southern Ocean"**

by Gregor, Kok and Monteiro

## Overview of changes

The authors would like to thank the reviewers for their recommendations to improve the manuscript. The changes are relatively substantial with the manuscript now being more succinct. The major scientific changes are: 1) removal of the two high resolution methods; 2) inclusion of uncertainty estimates. We also apologise for the multitude of typos and figure reference errors throughout the document. We hope that the reviewers now find the manuscript in a more presentable state.

In the document below we show the initial response of the reviewers in blue and the responses to each point in black. Track changes for the manuscript are shown below the response to the reviewer.

# Reviewer 1 Overview

Gregor and co-authors investigate the variability of the Southern Ocean CO2 uptake strength from 1998-2014 analysed for 9 regions (SO divided in basins and Fay and McKinley biomes). The authors combine 5 realizations to form a multi-model mean which is used to investigate seasonality and year-to-year variability of the delta pCO2 and the CO2 flux. 3 of the realizations are independent whereas 2 are simply higher resolution versions of 2 other methods. The authors find that the seasonal variability is the strongest mode of variability in the SO. Additionally, the authors confirm results from past studies that the SO was losing some of its uptake capacity in the early years of their analysis period, then the uptake increased in the subsequent period, whereas in the final years of their analysis period the reinvigoration of the sink stopped again. The authors investigate the cause of this variability in the sink strength by analysing anomaly periods for winter and summer season separately. From this analysis the authors conclude that the drivers are seasonally decoupled with wind being the dominant driver for winter variability and biology being the dominant driver for summer variability.

I found this study to be interesting, comprehensive and in general suitable for the journal. The authors do not only present results from new methods to confirm previous results, they also deepen the analysis by looking at anomalies rather than trends (as previously done) and investigate different seasons.

#### Major Issues

- The study confuses trends and variability. When do the authors talk about trends and when about variability? At the moment, these two terms are mixed up. E.g. take figure 5 all panels. There is clearly some year-to-year variability causing in some years more and in some less uptake but overarching in all panels of figure 5 and 6 one can clearly see an increasing CO2 sink from 1998 onwards, i.e. an increasing trend throughout the entire time period. Furthermore, wording used like "decadal trends" and "interannual trends" contribute to the confusion. What is a decadal trend? Is it the slope of a regression line when considering at least 10 years of data? What are interannual trends? The same only considering 3 years? In the latter case one cannot speak of a trend at all. We have removed most of the references to trends. We have also removed the reference to the decadal mode of variability. We do, however, make the connection between the longer modes of variability in winter and that this is likely linked to the decadal variability mentioned by Landschützer et al. (2016) linked to the SAM.
- Despite being able to do so, the authors do not add uncertainties. There are many sources of uncertainty ignored by the authors, e.g. the measurement uncertainty (which is however neglectably small), the extrapolation error of the method, the building of the

multi-model mean creates an error and finally the calculation of the air-sea flux adds another source of uncertainty (through wind and transfer velocity choice). At the moment, the results are presented overconfidently. It is not clear how much of the explored variability is significant and how much is simply statistical gibberish. I am aware that there is no "perfect" way to represent all uncertainties, but in a data sparse region like the Southern Ocean a study like this needs to add the best possible uncertainty estimate, otherwise, many of the conclusions drawn cannot simply be accepted. Regarding the propagation of errors, we used the same approach used by Landschützer et al. (2014). However, we do note that this error is Gaussian and mechanistically consistent, thus we can make deductions about the changes in the trends we observe. We also incorporate the between-method error from transCom (as in Gurney et al., 2004) at the recommendation of Christian Rödenbeck (pers comm.). We treat this, and a variation thereof as the primary threshold of significance as in Figures 5 and 6 (new numbering).

- On page 11 line 312 I found that the authors claim statistical significance between the mean uptakes, reporting a p-value. It is not clear to the reader what test was used and how significance has been determined. Also, when adding uncertainty, the authors will notice that an uptake of -0.17 PgC/yr is unlikely to be identified as statistically significantly different from -0.19 PgC/yr in the data sparse Southern Ocean.
   We have removed the trends from the FCO2 time-series, so this is no longer an issue. Note that this figure has also moved to the supplementary materials.
- Despite the uncertainty of the CO2 itself there are other sources of concern related to
  uncertainty. Chlorophyll is e.g. also presented without discussing uncertainty. How is
  cloud coverage and missing chlorophyll data effecting the results? Also, wind products
  have been shown to have different trends locally in the Southern Ocean. This has not
  been mentioned. I am also wondering to what extend the use of different products
  hampers the conclusions of the manuscript. SST from Reynolds is based to the extent
  of my knowledge from satellite and in-situ data, whereas ECCO MLD is from an
  assimilation model. I would expect some disagreement between these products that have
  nothing to do with "real world" disagreement. This is not a massive concern, but certainly
  needs to be mentioned as well.

This is a valid point and we agree with the reviewers concern. However, it may be beyond the scope of this study to address the uncertainties driven by the input proxies. This may be a good topic for a collaborative effort of the SOCOM intercomparison. I have to some extent addressed the reviewers points:

**Chlorophyll:** we include the methodology used by Gregor et al. (2017) for the missing chlorophyll, where cloud patches are filled with the climatology for that point, and missing winter values are filled with a low level Gaussian noise.

ECCO2 MLD: This is indeed assimilation model output. Our motivation here is that

using observations (most likely the de Boyer Montégut (2004) MLD climatology) would mean that MLD could not treated as a potential driver of pCO2. However, there may be regions (such as the MIZ) where the MLD estimates are spurious for ECCO2. Choosing between these two products is thus a choice of trade-offs.

**Wind:** We include our justification for using CCMP v2 only. Much of this is based on the study by Swart et al. (2015a) and personal communication with Neil Swart.

• Another source of concern is the length choice of periods P1-P4. Periods P1-P3 are all of the same length, whereas P4 is substantially shorter. The authors claim that substantial year-to-year variability occurs on various timescales (4-6 year and 10 year modes respectively), hence the variation of periods aliases this analysis. We have changed the lengths of the periods to be more or less the same length (5, 4, 4, 4

years respectively). We hope that this approach addresses the reviewers concerns.

All the above points raised are of major concern and must be addressed before the manuscript can be considered for publication.

Minor comments:

• Title: the title only mentions CO2 fluxes whereas in the manuscript largely discusses delta pCO2

We have made the title more generic: CO2

• Line 29: "paucity of observations (Landschützer et al 2015)" – This is not a good reference. Bakker et al 2016 (the SOCATv2 reference) would be a better reference for such a paucity.

We have changed this reference to Bakker et al. (2016)

- Line 62: remove 2nd that removed
- Line 95: wrong reference (Bakker et al 2016) This reference is in fact correct for SOCAT v3. SOCAT v2 is Bakker et al. (2014)
- Table 1: How has the RMSE for the 0.25x0.25 degree product been calculated? SO- CAT offers a gridded product but on 1x1 degrees. Did the authors grid the 0.25 product themselves? If yes, has this been done the "SOCAT" way, i.e per cruise weighted or differently? Same with the 16-day time step. More information is needed here. We have removed the high resolution implementations of RFR and SVR at the request of Reviewer 2 (who felt they resulted in an ensemble biased towards these methods).
- Line 130: How different would the results be if a different transfer velocity was chosen? This may have significant effects on the uncertainty. The fluxes are now only included in the supplementary materials.
- Line 134: where are [ice] data from? I am missing a reference here.

"...sea surface temperature (SST) and sea-ice fraction by Reynolds et al. (2007), ... "

• Line 175-176: "attributed this difference to the clustering step used by the SOM-FFN that created large discrepancies in the Atlantic sector." I have not found any convincing evidence for this in any of the cited papers.

The published version of Gregor et al. (2017) now contains a figure (A3) that the reviewer may find convincing.

• Line 236: Either the authors used the wrong wording or there is a misunderstanding, but I do not see where the authors find that the delta pCO2 is zonally asymmetric within each biome. It looks like the other way around: Figure 4 looks like there is a strong zonal symmetry (besides indeed in panel a).

Included an additional clause that the gradient is during summer: "Apparent also from Figure 3 is that, over and above the latitudinal gradient,  $\Delta pCO_2$  is zonally asymmetric within each biome during summer (Figure 3a), when biological uptake of  $CO_2$  increases."

• Lines 249-254: This paragraph is not clear. Please rephrase

We have tried to make this more clear: "The projected summer minima (dashed lines) are calculated by subtracting the mean seasonal amplitude from the winter maxima (Figure 4, with air-sea  $CO_2$  fluxes shown in Figure S3). The projected summer minima is the expected summer  $\Delta pCO_2$  under the assumption that summer  $\Delta pCO_2$  is dependent on, but not restricted to, the baseline set by winter. Differences between the summer minima and projected minima are highlighted with green and blue patches, highlighting periods of decoupling between summer and winter interannual variability. The green areas indicate periods of strong uptake (relative to winter) that enhance the mean uptake of  $CO_2$  and amplify the seasonal cycle. Conversely, blue areas show periods where weak summer uptake (relative to winter) offsets winter outgassing, thus reducing the mean  $\Delta pCO_2$  as well as supressing the amplitude of the seasonal cycle (Figure 4)."

• Line 273: interannual trend – what is that? Interannual variability. A 3-year trend? A trend that changes sign every year?

We have rephrase this to: "A key feature of Figure 4 is that the mean interannual variability is the net effect of decoupled seasonal modes of variability for summer and winter."

- Lines 275: decadal mode: How are you able to say this. You have 1998-2014 data, i.e. 17 years of data. How can you detect a decadal mode from such a short time series? We have removed this statement and other references to the decadal mode, unless it is associated with previous studies (Lovenduski et al., 2008; Landschützer et al., 2016).
- Line 305: "decadal trend" what is this? A trend of at least 1 decade? Removed decadal trend and see above comment
- Lines 311-312: "-0.19 and -0.17 PgC yr-1 for the Atlantic and Pacific sectors respectively

(where the latter are significantly different with p = 0.01)." how was this calculated, and given the many, many uncertainties that go into such a flux number one cannot possibly believe that this these numbers are indeed statistically significant.

We have moved the fluxes to supplementary materials. We have also removed the trends reported on the figure as this may detract and confuse given that the rest of the study deals with anomalies rather than trends.

- Line 340: What about acidification induced changes linked to changes in the buffer capacity (see e.g. Hauck et al 2015)? A contribution within 17 years is plausible. This is an interesting point, but would lengthen the study too much, and is thus better left for another study.
- Line 439: I suppose ENSE is ENSO but please either way spell out abbreviations when first using them

ENS(E)O is only used once and thus written out in full

Line 505: "The fact that Chl-a is the dominant driver of interannual ΔpCO2 variability should not be surprising" – the authors have not proven that chlorophyll is the dominant driver. From a pool of selected variables, it showed the largest correlation – this can barely be called a "fact" in science.

Rephrased: "Our finding that Chl-a is the dominant driver of interannual  $\Delta pCO_2$  variability should not be surprising given that models and observations support this notion (Hoppema et al., 1999; Bakker et al., 2008; Mahadevan et al., 2011; Wang et al., 2012; Hauck et al., 2013; 2015; Shetye et al., 2015)"

• Figures 5 and 6: Uncertainties need to be added. Without uncertainty I do not trust that the observed variability is significant.

We have added uncertainty estimates of uncertainty as previously mentioned. We keep these relatively simple in Figure 4 (old 5) by using time averages of the between-method errors. We do however include uncertainty thresholds in the final anomaly analysis. We feel that this is perhaps a more pertinent place to show the uncertainties.

• Figures 7 and 8: Wind stress anomalies are interesting, but direction would be equally interesting and provide more evidence.

We include the wind direction for each of the anomaly transitions in the supplementary materials. We find that the wind anomalies do not increase the understanding of the changes in  $\Delta pCO2$  significantly to justify the addition in Figures 5, 6 (old 7 and 8).

# Reviewer 2

#### Overview

The authors present here a lengthy paper in which they aim to provide evidence that the included machine learning products for pCO2 are an appropriate proxy for investigating interannual variability in pCO2 and carbon fluxes in the Southern Ocean. They use these products to analyse transitions between regimes and also investigate the drivers of the changes. However, the paper needs to be proofread prior to submitting as it has errors throughout which distracts from the science. The job of a review is not to be a technical reviewer but to analyse the science. Unfortunately it is difficult to follow the science with the errors included throughout the paper.

#### Major revision

The authors utilize an ensemble of five products for this study, however 2 of the products are just a repetition of two other products produced at a higher resolution. There is no evidence given that this results in 5 independent products for this ensemble. At the very least, more discussion is required for it to be accepted that the high and low resolution versions of the same product can be seen as individual ensemble members.

We acknowledge that these two high resolution implementations may not have contributed results that were mechanistically different to the low resolution implementations. We have thus removed the high resolution data from the study. This means that the now reduced ensemble includes only three machine learning methods: SOM-FFN, SVR, RFR.

Additionally, the analysis should be revised to include the SOM-FFN created with SOCATv3 so that all members are produced using the same dataset. The SOM-FFN product is available based on SOCATv4 now and the authors should at least update it to the version based on SOCATv3 (see <a href="https://www.nodc.noaa.gov/ocads/oceans/SPCO2\_1982\_2015\_ETH\_SOM\_FFN.html">https://www.nodc.noaa.gov/ocads/oceans/SPCO2\_1982\_2015\_ETH\_SOM\_FFN.html</a> ). We now use v2.2 of the SOM-FFN implementation which used SOCAT v4. The results have been updated accordingly.

This paper is wrought with inconsistencies, incomplete definitions, and missed words which distracts the reader throughout. Some examples include the use of MIZ in text but AZ in figures, along with a consistent use of acronyms without first defining them (PFZ, AZ, MIZ, etc). Additionally, using the Fay & McKinley biome boundaries but then referring to the regions as SAZ, PFZ, AZ/MIZ is confusing and inaccurate. The biomes and frontal regions are not interchangeable and the authors need to be consistent throughout the study as to which they are using.

These abbreviations based on the Fay and McKinley (2014) paper where the authors state: "...Southern Ocean regions that we define as STSS, SPSS and ICE biomes. Respectively, these three Southern Ocean biomes are comparable to the Sub-Antarctic Zone (SAZ), the Polar Frontal Zone (PFZ) and Antarctic Zone (AZ)". We chose to use MIZ instead of AZ. These biomes are now defined before they are used. If the reviewer still feels that these should be changed, we would be happy to make these changes.

In Figure 2, the different extents into the ice covered regions of each product will affect the comparison shown in 2c. Ensure that equal regions are being compared. Also, showing how the products compare to the available data in Figure 2 would be helpful.

We have changed the comparison area to be consistent for all models. This means that a large

portion of the MIZ/ICE is omitted. We have thus decided to exclude the MIZ from the rest of the analysis. We do show the MIZ data for both  $\Delta pCO_2$  and  $FCO_2$  in the Supplementary materials. The available data is unfortunately just too sparse to plot alongside the empirical estimates on a monthly scale as shown in the figure below.



Figure 3b is not discussed in the text at all. The definition of the "signal" as the largest difference in trend for a particular grid cell should be referenced as typically it is the mean trend/value that is the "signal" and the noise is the spread around that signal (either standard deviation, standard error, etc).

Figure 3 has been removed from the manuscript. We now take a different approach in addressing the uncertainties at the request of Reviewer 1.

Figures 5 and 6 need to be improved dramatically. The difference between the dark and light curves in Figure 5 is not defined. Additionally, there is no indication of which product (or average of the ensemble) is being plotted here. The captions are wrong (referenced subplots j-r which do not exist). The background grey shading to designate the difference regimes/timescales is difficult to distinguish. Perhaps another method to highlight those would be helpful. Additionally, trends included on these figures should have confidence intervals or uncertainty values included.

We have changed this figure substantially. The MIZ has been dropped as the small remaining area (after masking for model inconsistency) is not representative of the biome. This has thus simplified the image a lot. We have darkened the shading as we too noticed that these are very light when the manuscript is printed. We have also included the dates of these periods on the figure. Note that the dates of the periods have changed at the request of Reviewer 1. The captions have been corrected.

We have also moved the figure showing the fluxes to the supplementary materials. We feel that this distracts slightly from the story that we are trying to tell. Moreover, we have removed the trends from the Flux figure as it is confusing to work with anomalies and then present trends (Reviewer 1).

Figures 7-8 continue with captions that do not correspond to the figure ("wind stress (d-e), SST (f-h)" when wind stress is in (d-f) and SST in (g-i), etc.

We have corrected these captions.

#### **Technical Corrections**

Below are some obvious technical corrections which I have not already addressed above. I reiterate though that the entire paper needs to be revised and improved, as it is difficult to follow the science with incorrect references to figures and missing words in sentences.

• Line 43: "not due to changes in overturning"

I wasn't completely sure what the reviewer found at fault in this sentence other than being clumsy. It has been rephrased: "While previous studies suggested that changes in wind strength have led to changes in meridional overturning and thus CO<sub>2</sub> uptake (Lenton and Matear, 2007; Lovenduski et al., 2007; Lenton et al., 2009; DeVries et al., 2017), Landschützer et al. (2015) suggested that atmospheric circulation has become more zonally asymmetric since the mid 2000's, which has led to an oceanic dipole of cooling and warming."

- Line 46: "The led to an oceanic dipole. . ." "...which has led to an oceanic dipole..."
- Line 62: repeated word Removed
- Line 141: PFZ and MIZ not defined. MIZ and AZ used inter- changeably throughout paper
  - We now define these when first introduced on L165.
- Line 149: missing a word in "the variability of between the ensemble members. . ." This has been rephrased to: "*The first section examines the uncertainties of the ensemble and its members.*"

- Line 157: how are "scores" calculated? We now define the scores explicitly as the root mean squared error (RMSE).
- Line 199: Should reference Figure 3c I believe Figure has been removed
- Line 213: Figures 2a-c and 4a-c are not the correct reference figures for the points being addressed.
  - The references to figures have (hopefully) been corrected
- Line 221: "CO2" We have corrected all occurrences of CO<sub>2</sub> to be subscripted
- Line 255-260: Figure 5 caption is inaccurate to what is shown in Figure 5 Corrected and figure numbers have changed
- Line 289: Should reference Figure 5a,d rather than Figure 5 b,e. Corrected and figure numbers have changed
- Line 291: remain consistent with capitalizing Southern Ocean All occurrences of Southern Ocean have now been capitalised
- Line 307: Do you mean the seasonal cycle amplitude? We define this at the first occurrence of the phrase on L211
- Line 342: "it was advanced that the explanation. . ." is awkward This section has changed a great deal – this paragraph has been removed
- Line 350: "These studies have in linked the wind stress variability. . ." is awkward and needs to be rewritten.

This has been removed

• Lines 373-376: caption for Figure 7 and Figure 8 need to be corrected to accurately reference the figures.

The captions have been corrected

- Line 387-388: ". . . and surrounds (Figure 7d,j)" is awkward. Consider revising sentence Rephrased to: *This regional sustained saturation corresponds to a shift towards stronger winds and/or deeper MLDs in the west Pacific sector of the SAZ (Figure 5d,j).*
- Line 397: "(-ve shift)". Is this supposed to be negative shift? If so, simply spell out to improve clarity.
   All occurrences of +ve and -ve have been written out as positive and negative respectively.
- Line 439: "ENSE" perhaps should be ENSO? Changed to ENSO

Overall, the science presented in the paper is interesting and could provide a interested look at using various machine learning methods to gain an understanding of the Southern Ocean carbon drivers. However, the lack of proofreading prior to submitting is clear and must be improved before a complete review of the manuscript can be undertaken.

# Interannual drivers of the seasonal cycle of CO<sub>2</sub> fluxes in the Southern

## Ocean

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Abstract. Machine learning methods (support vector regression and random forest regression) were usedhas become a useful tool to map gridded estimates interpolate ship measurements of  $\Delta pCO_2$  in the Southern Ocean from (SOCAT-v3) to a gridded map using satellite data. A low (1° × monthly) and high (0.25° × 16-day) resolution implementation of each of these methods as well as In this study we use an ensemble of three machine learning methods: Support Vector Regression (SVR) and Random Forest Regression (RFR) from Gregor et al. (2017); and the SOM-FFN method of from Landschützer et al. (2014) were added to a five member ensemble. (2016). The ensemble mean  $\Delta pCO_2$  was used to calculate  $FCO_2$ 

- 15 (air-sea CO<sub>2</sub> flux). Data was interpolated data were separated into nine domainsregions defined by basin (Indian, Pacific and Atlantic) and biomes (as defined by Fay and McKinley-(2014, 2014a). The regional approach showed large a meridional gradient and zonal asymmetry in the magnitude of  $\Delta p$ CO<sub>2</sub> and FCO<sub>2</sub> estimates. Importantly, there was a seasonal decoupling of the modes for summer and winter interannual variability. Winter trends-interannual variability had a larger 10 yearlonger mode of variability compared
- to summer trends, which hadvaried on a shorter 4–6 year modetime scale. To understand this variability of  $FCO_2\Delta pCO_2$ , we separately assessed investigated changes in summer and winter  $\Delta pCO_2$  and the drivers thereof. The dominant winter changes were are driven by wind stress variability. Summer variability correlated well This is consistent with the temporal and spatial characteristics of the Southern Annular Mode (SAM), which has a decadal mode of variability (Lovenduski et al., 2008; Landschützer et al.,
- 25 2016). Interannual trends in summer variability of  $\Delta pCO_2$  are consistent with chlorophyll-*a* variability where the latter had high <u>mean seasonal</u> concentrations. In regions of low chlorophyll-*a* concentrations, wind stress and sea surface temperature were lower order<u>emerged as stronger</u> drivers of  $\Delta pCO_2$ . In summary we propose that sub-decadal variability is explained by summer drivers, while winter variability contributes to the long term changes associated with the SAM.

#### 30 1 Introduction

The Southern Ocean plays a key role in the uptake of anthropogenic CO<sub>2</sub> (Khatiwala et al<sub> $\frac{1}{1}$ </sub> 2013; DeVries et al<sub> $\frac{1}{1}$ </sub> 2017). Moreover, it has been shown that the Southern Ocean is sensitive to anthropogenically influenced climate variability, such as the intensification of the westerlies (Le Quéré et al<sub> $\frac{1}{1}$ </sub> 2007; Lenton et al<sub> $\frac{1}{1}$ </sub> 2009; Swart and Fyfe<sub>2</sub> 2012; DeVries et al<sub> $\frac{1}{1}$ </sub> 2017). Until recently, the <u>research</u> community has not

- 35 been able to accurately measure the <u>contemporary</u> changes, let alone understand the drivers, of CO<sub>2</sub> in the <u>contemporary</u> Southern Ocean due to a paucity of observations (<u>LandschützerBakker</u> et al. <u>2015</u>., <u>2016</u>). Empirical models provide an interim solution to this challenge until prognostic ocean biogeochemical models are able to represent the Southern Ocean CO<sub>2</sub> seasonal cycle accurately (Lenton et al., <u>2013</u>; <u>Rödenbeck et al., 2015</u>; <u>Mongwe et al., 2016</u>). <u>-2013</u>; <u>Rödenbeck et al., 2015</u>; <u>Mongwe et al., 2016</u>).
- 40 2016). The research community agrees on large changes in CO<sub>2</sub> fluxes in the Southern Ocean from a source in the 1990's to a sink in the 2000's; however, there is disagreement in the drivers of the changes in CO<sub>2</sub> uptake (Lovenduski et al., 2008; Landschützer et al., 2015; DeVries et al., 2017). This study aims to understand the drivers of the changing CO<sub>2</sub> sink in the Southern Ocean based on an ensemble of empirical estimates using a seasonal analysis framework.

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Empirical methods estimate CO<sub>2</sub> by extrapolating the sparse ship based CO<sub>2</sub> measurements using satellite observable proxies. This approach has allowed for a better understanding of the drivers of CO<sub>2</sub> by providing improved spatial and temporal resolution of the variability. Landschützer et al. (2015) usedshowed, using an artificial neural network (ANN) to show), that thethere was significant 50 strengthening of the Southern Ocean CO<sub>2</sub> uptake during the period 2000-2010 is part of a decadal internal variability in the natural CO<sub>2</sub> flux dynamics. The authors found. While previous studies suggested that the strengthening sink was not due to changes in wind stress as suggested by previous studies strength have led to changes in meridional overturning and thus CO<sub>2</sub> uptake (Lenton and Matear, 2007; Lovenduski et al., 2007; Lenton et al., 2009). Rather, they; DeVries et al., 2017), Landschützer et al. (2015) suggested that atmospheric circulation has become more zonally asymmetric since the mid 2000's-55 This, which has led to an oceanic dipole of cooling and warming whose. The net impact of cooling and warming, together with changes in the DIC/TA was to(Dissolved Inorganic Carbon/Total Alkalinity), led to an increase in the uptake of CO<sub>2</sub> (Landschützer et al., 2015). During this period, in the Atlantic basin, southward advection reduced upwelled DIC in surface waters overcoming the effect of the concomitant 60 warming in the region. Conversely, in the Eastern Pacific sector of the Southern Ocean, strongerstrong cooling overwhelmed increased upwelling (Landschützer et al., 2015). Munro et al. (2015) supported this mechanism, with data from the Drake Passage showing that  $\Delta p CO_2$  decreased between 2002 and 2014.

- In a subsequent study Landschützer et al. (2016) proposed that interannual variability of CO<sub>2</sub> in the Southern Ocean is tied to the decadal variability of the Southern Annular Mode (SAM) – the dominant mode of atmospheric variability in the Southern Hemisphere (Marshall, 2003). This concurs with previous studies, which suggested that the increase in the SAM during the 1990's resulted in the weakening of the Southern Ocean sink (Le Quéré et al=, 2007; Lenton and Matear, 2007; Lovenduski et al=, 2007; Lenton et al=, 2009; DeVries et al. 2017). The work by Fogt et al. (2012) bridges the gap between the proposed asymmetric atmospheric circulation of Landschützer et al. (2015) and the observed correlation with the SAM of Landschützer et al. (2016). Their study showsFogt et al. (2012) show that changes in the SAM
- have been zonally asymmetric and that that this variability is highly seasonal, thus amplifying or suppressing the amplitude of the seasonal mode.
- Assessing the changes through a seasonal framework may thus help shed light on the drivers of CO<sub>2</sub> in the Southern Ocean. Southern Ocean seasonal dynamics suggest that the processes driving  $\Delta p$ CO<sub>2</sub> are complex but with two clear contrasting extremes. In winter, the dominant deep mixing and entrainment processes are zonally uniform driving an increase in  $\Delta p$ CO<sub>2</sub> with the region south of the Polar Front (PF) becoming a net source and weakening the net sink north of the PFZPF (Lenton et al-, 2013). In summer,
- 80 the picture is much-more spatially heterogeneous, with NPP<u>net primary production</u> being the primary driver of variability (Mahadevan et al., 2011; Thomalla et al., 2011; Lenton et al., 2013). The competition between light and iron limitation results in heterogeneous distribution of Chl-*a* in both space and time, with similar implications for  $\Delta p$ CO<sub>2</sub> (Thomalla et al., 2011; Carranza and Gille, 2015). The interaction between the large-scale drivers, such as wind stress, surface heating and mesoscale ocean dynamics, areis
- the primary cause of this complex picture (McGillicuddy, 2016; Mahadevan et al., 2012). Some regions of elevated mesoscale and submesoscale dynamics, mainly in the Sub-Antarctic Zone (SAZ) are also characterized by strong intraseasonal modes in summer primary production and pCO<sub>2</sub>. (Thomalla et al., 2011) and pCO<sub>2</sub> (; Monteiro et al., 2015). The magnitudes of these In general, the opposing seasonal processes are large, resulting effects of mixing and primary production result in the seasonal cycle being the dominant mode of variability in the Southern Ocean. (Lenton et al., 2013).

In this study we examine <u>winter and summer</u> interannual variability in the air-sea fluxes of  $CO_2$ between  $\Delta pCO_2$  from 1998 – to 2014 through interannual changes in the characteristics of the seasonal mode of both  $pCO_2$  and  $FCO_2$ -in the Southern Ocean. We use an ensemble of empirical estimates of  $CO_2$ 

95 that combine *in situ* observations with remotely sensed proxies to perform this analysis to understand the drivers of long term changes in CO<sub>2</sub> uptake.

#### 2 Empirical methods and data

#### 2.1 Empirical methods and data

In this study we made use of three empirical methods combined to an ensemble – these machine learning

- 00 methods-are-: Random Forest Regression (RFR), Support Vector Regression (SVR) and Self-Organising-Maps Feed-Forward Neural Network (SOM-FFN). RFR and SVR are introduced in Gregor et al. (2017) and SOM-FFN is presented in Landschützer et al. (2014). In brief, the RFR approach is an ensemble of decision trees that provides non-linear regression by combining many high variance – low bias estimators (Gregor et al., 2017). Gregor et al. (2017). The advantage of an ensemble over <u>SVRs are in principle</u>
- 05 similar to a single hidden layer FFN, with the difference that SVR statistically determines the complexity of the problem, which is analogous to the hidden layer structure that is typically determined heuristically. The SOM-FFN method approach is that a degree of robustness is added to the estimate, assuming that ensembles have unique biases in time and space. The other important assumption we make here is that the majority methods will be correct, while the minority will be biased. The ensemble mean contains five
- 10 different Δ*p*CO<sub>2</sub> estimate approaches, shown in Table 1, with low- and high-resolution (1°, monthly and 0.25°, 16-day respectively). The SOM-FFN method is defined by Landschützer et al. (2014) and is a two-step neural network approach (trained with SOCAT v2) that first clusters data (SOM) and then applies a regression model (FFN) to each cluster (Bakker et al., 2014). The low resolution.
- The SVR and RFR implementations of Support Vector Regression (SVR) and Random Forest Regression (RFR) methods are introduced in Gregor et al. (2017). Note that these areused in this study are trained with the monthly by 1° gridded SOCAT v3 dataset (Bakker et al., 2016). The SOM-FFN (v2.2) used in this study was trained with SOCAT v3 data (Bakker et al., 2016). The high-resolution implementations of SVR and RFR used in this study are implemented in the same way as described in Gregor et al. (v4 (Landschützer et al., 2017). The high-resolution estimates of ΔpCO<sub>2</sub> are resampled to match the low
- resolution data in the ensemble.

Table 1: FiveThree empirical methods used in the ensemble. RFR-LR and SVR-LR are described in GregorGregor et al. (2017).SOM-FFN is from Landschützer et al. (Landschützer et al. 2014(2016). SST = sea surface temperature, MLD = mixed layerdepth, SSS = sea surface salinity, ADT = absolute dynamic topography, Chl-a = Chlorophyll-a,  $pCO_{2(atm)}$  = fugacity of atmosphericCO2,  $xCO_{2(atm)}$  = mole fraction of atmospheric CO2,  $\Phi(lat, lon) = \frac{N-vectorN-vector}{N-vector}$  transformations of latitude and longitude, $\lambda(t(day of year) = trigonometric transformation of the day of the year. Note that SOM-FFN uses the de Boyer Montégut et al. (2004)climatology for MLD (dBM2004). The root mean squared errors (RMSE) listed in the last column are for the Southern Ocean from Gregor et al. (2017).$ 

Metho	Resol		RMSE
d	ution	Input variables	(µatm)
			Tim
		Space	e
		2	

	RFR- <del>LR</del>	<del>1.0</del> θ°	$M = SST, MLD, SSS, ADT, Chl-a(clim), pCO2(atm), \Phi(\Phi(lat, lon), t(day of year)) h$	<del>17.21<u>16.45</u></del>
	SVR- <del>LR</del>	<del>1.0</del> θ°	$\begin{array}{l} & \label{eq:starses} \begin{tabular}{l} & \end{tabular} \\ &$	<del>21.73</del> <u>24.04</u>
	SOM- FFN	<del>1.0</del> θ°	M o n SST, <u>MLD_MLD_dBM2004</u> , SSS, Chl- <i>a</i> , <i>x</i> CO <sub>2(atm)</sub> t h	<del>15.45<u>14.84</u></del>
0. 0.	25° <del>16-day</del> <del>25° 16-day</del>	+ <del>SST</del> + <del>SST</del>	, <mark>MLD, SSS, ADT, Chl-a<sub>(clim)</sub>, <del>ρCO<sub>2(atm)</sub></del> , MLD, SSS, ADT, Chl-a<sub>(clim)</sub>, <del>ρCO<sub>2(atm)</sub>, Φ(lat, Ion), t(day of year)</del></mark>	<del>12.58</del> <del>19.18</del>

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RFR-HR SVR-HR

Table 1 also-shows the proxy variables used for each of the methods. The sources for the proxy variables are consistent for all methods ensuring a fair comparison. This is particularly important for the assimilated model variables, mixed layer depth (MLD) and seaSea surface salinity (SSS) and mixed layer depth (MLD) for SVR and RFR are from Estimating the Circulation and Climate of the Ocean, Phase II (ECCO<sub>2</sub>) (Menemenlis et al<del>.</del>, 2008). Choosing to The use different of these assimilative modelled products may in some cases produce results that are unrealistic. This may have influenced the use of the de Boyer Montégut et al. (2004) MLD climatology in the SOM-FFN, where ECCO<sub>2</sub> was used in previous iterations of the product. The trade-off of using the climatology is that no changes in MLD are taken into account. We acknowledge that using different proxy variables could result in data driven differences

- 40 (from the same variable).different  $\Delta pCO_2$  estimates, but comparing the different products is beyond the scope of this study. Other data sources that are consistent between methods are: sea surface temperature (SST) (and sea-ice fraction by Reynolds et al. (2007), Chlorophyll-*a* (Chl-*a*) (by Maritorena and Siegel (2005), absolute dynamic topography (ADT) (by Duacs),  $xCO_2$  (CDIAC, 2016) with  $pCO_{2(atm)}$  calculated from interpolated  $xCO_2$  using NCEP2 sea level pressure (Kanamitsu et al. 2002)., 2002). In the case of
- 45 <u>Chl-a for SVR and RFR, Gregor et al. (2017) filled the cloud gaps with climatological Chl-a.</u> Note that ADT coverage is limited to regions of no to very low concentrations of <u>sea-ice cover</u>, thus estimates for SVR and RFR methods do not extend into the ice covered regions during winter. <u>Our analyses are thus limited to the regions without ice cover</u>.

50 Seasonality of the data is preserved by transforming the day of the year (*j*) and is included in both SVR and RFR analyses:

$$t = \begin{pmatrix} \cos\left(j \cdot \frac{2\pi}{365}\right) \\ \sin\left(j \cdot \frac{2\pi}{365}\right) \end{pmatrix}$$
(1)

Transformed coordinate vectors wereare passed to <u>SVR</u> only <u>SVR</u> using *n*-vector transformations of latitude ( $\lambda$ ) and longitude ( $\mu$ ) (Gade<sub>4</sub> 2010; Sasse et al. 2013), with *n* containing:

$$N = \begin{pmatrix} \sin(\lambda) \\ \sin(\mu) \cdot \cos(\lambda) \\ -\cos(\mu) \cdot \sin(\lambda) \end{pmatrix}$$
(2)

Wind speed, while not used in the empirical methods, is used in the assessment of the drivers of  $CO_2$ . We use CCMP v2, which is an observation-based product that combines remote sensing, ship and weather buoy data (Atlas et al., 2011). Swart et al. (2015a) compared a number of wind reanalysis products with

60 CCMP v1 (where CCMP was the benchmark). The authors found that many of the reanalysis products had spurious trends, particularly in the Southern Hemisphere where data is sparse. Our choice of CCMP, which is based on observations, is thus one that aims to minimise the assumptions that are otherwise made by reanalysis products.

#### **2.2 Uncertainties**

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65 The machine learning approaches used in this study are by no means able to estimate  $\Delta pCO_2$  with absolute certainty. To account for the uncertainty we use the same approach as Landschützer et al. (2014) to calculate total errors for each of the methods:

$$\boldsymbol{e}_{(t)} = \sqrt{e_{meas}^2 + e_{grid}^2 + e_{map}^2} \tag{3}$$

where  $e_{m(t)}$  is the total error associated with a method (m);  $e_{meas}$  is the error associated with SOCAT 70 measurements, which is fixed at 5 µatm (Pfeil et al., 2013);  $e_{grid}$  is the 5 µatm error associated with gridding the data into monthly by 1° bins (Sabine et al., 2013).

$$N = \Phi \begin{pmatrix} \frac{\sin(\lambda)}{\sin(\mu) \cdot \cos(\lambda)} \\ -\frac{\cos(\mu) \cdot \sin(\lambda)}{\cos(\lambda)} \end{pmatrix}$$
(2)

#### Air-sea CO<sub>2</sub> fluxes

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Lastly *e<sub>map</sub>* is the root mean squared error (RMSE) calculated for each method as shown in Table 1 taken from Gregor et al. (2017).

These errors are used to calculate the average "within-method" error as defined by Gurney et al. (2004):

$$E_w = \sqrt{\frac{1}{M} \cdot \sum_{m=1}^{M} (e_{m(t)})^2}$$
(4)

where  $e_{m(t)}$  is the method specific error as defined in Equation 1 and *M* is the number of methods (3 in this case). For a measure of the difference between methods we use the "between-method" approach used in Gurney et al. (2004):

$$E_b = \sqrt{\frac{1}{M} \sum_{m=1}^{M} (S_m - \bar{S})^2}$$
(5)

where  $S_m$  is the method estimate of  $\Delta pCO_2$  and  $\overline{S}$  is the mean of the methods. This is analogous to the standard deviation (for a known population size). We later use an adaptation of this metric as a threshold to determine the confidence around anomalies.

#### 85 <u>2.3 Air sea $CO_2$ fluxes are calculated with:</u>

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$$FCO_2 = k_w \cdot K_0 \cdot (pCO_2^{sea} - pCO_2^{atm}) \cdot (1 - [ice])$$
(3)

The gas transfer velocity  $(k_{*})$  is calculated using a quadratic dependency of wind speed with the

90 coefficients of (Wanninkhof et al. 2009). Wind speed is calculated from the *u* and *v* vectors  $(\sqrt{u^2 + v^2})$  of the Cross-Calibrated Multiplatform Product v2 (Atlas et al. 2011). Coefficients from Weiss (1974) are used to calculate  $K_{\theta}$  and  $\Delta p CO_2$  is estimated by the empirical models. The effect of sea-ice cover on  $CO_2$  flux is treated linearly (Butterworth and Miller 2016): the fraction of sea ice cover ([*ice*]) is converted to fraction of open water by subtracting one as shown in Equation (3).

#### 95 Regional Coherence Framework

Southern Ocean CO<sub>2</sub> is spatially heterogeneous both zonally and meridionally (Jones et al. 2012). In order to understand this heterogeneity we used the three southernmost biomes defined by Fay and McKinley (20172014a) as done in Rödenbeck et al. (2015). From north to south these are: <u>sub-tropicalsubtropical</u> seasonally stratified (STSS), sub-polar seasonally stratified (SPSS), seasonally ice covered region (ICE). These three biomes are comparable to the SAZ<sub>7</sub> (Sub-Antarctic Zone), PFZ (Polar-Frontal Zone) and MIZ (Marginal Ice Zone) respectively and will be used throughout the rest of the study. The Southern Ocean is further split into basins where the boundaries are defined by lines of longitude (70°W : Atlantic : 20°E : Indian : 145°E : Pacific : 70°W). Figure 1 depicts these nine regions.



Figure 1: A map showing the regions used throughout this study. The three biomes, SAZ, PFZ and MIZ used in this study are defined by Fay and McKinley (20142014a). The regions are also split by basin with boundaries shown above the map.

#### **3** Results and discussion

- In this section<u>Here</u> we present and discuss the <u>dataresults</u>. The first section examines the <u>variabilityuncertainties</u> of <u>between</u> the ensemble <u>and its</u> members to <u>understand the potential limitations</u> of the dataset. We then <u>look atconsider</u> the seasonal cycle of the ensemble mean in time and space. This is done to lay the foundation <u>knowledge</u> for the interpretation of the results when assessed with the regional framework, which is the following section. In the regional interpretation the data is decomposed
- into <u>the</u> nine regions as shown in Figure 1. The section that follows sets out to make senseLastly we implement a seasonal decomposition of the estimates to interpret the drivers of the trendschanges observed in the regional decomposition.  $\Delta pCO_{2.}$

#### 3.21 Ensemble member performance and variability

In this section we discuss the performance and variability of the ensemble members. The individual ensemble member Table 2: A regional summary of the errors for the different models. Note that the propagated errors are calculated as shown in equation (3) where the measurement and gridding errors are assumed to be constant at 5 μatm each (Pfeil et al., 2013; Sabine et al., 2013). The within-model and between model errors are calculated using equations (4) and (5) respectively.

	Propagated errors (µatm)			Within	Between
Biome	<u>SVR</u>	<u>RFR</u>	<u>SOM-</u>	<u>model</u> (µatm)	<u>model</u> (µatm)

			FFN		
SAZ	17.48	14.50	12.30	<u>14.91</u>	4.88
PFZ	15.94	12.71	13.09	<u>13.99</u>	4.78
MIZ	36.38	24.53	22.46	28.46	10.81
Southern Ocean	<u>25.06</u>	<u>17.91</u>	<u>16.44</u>	<u>20.16</u>	<u>6.79</u>

We use the RMSE scores are shown in Table 1. The RFR-HR is the best performing member, as presented
in Gregor et al. (2017) with abbreviated results shown in Table 1. The SOM-FFN method has the best score (14.84 µatm). SVR scores the lowest RMSE (12.58 µatm). The SVR members score the lowest (21.73 and 19.18 µatm for LR and HR respectively(24.04 µatm), but werewas still included due to the method's sensitivity to sparse data, which is favourable to the poorly sampled winter period (Gregor et al., 2017). This compliments the RFR methodsmethod, which scorescores well (12.58 and 17.21 µatm for HR and LR respectively16.45 µatm), but areis prone to being insensitive to sparse data (Gregor et al., 2017). These RMSE scores are used to calculate the total errors for each method and region using equation (3) where the measurement and mapping errors are both 5 µatm each (Pfeil et al., 2013; Sabine et al., 2013). These results are shown in Table 2.

- 35 Total errors are used to calculate the within-method error, which is an estimate -, 2017). The SOM FFN member has the best of the low-resolution scores (15.45 µatm). However, this is because the SOM-FFN is tested with SOCAT v2 data rather than SOCAT v3, where the latter has a larger-combined total errors of the three machine learning methods (equation 4). The between-method errors are the mean of the standard deviation (32.85 and 36.27 µatm respectively). When RFR-LR and SVR-LR are tested with the standard deviation (32.85 and 36.27 µatm respectively). When RFR-LR and SVR-LR are tested with the SOCAT v2 dataset, the RMSE scores are 15.15 and 19.82 µatm respectively.between the methods (equation 5). The within-method errors are much larger than the between-method errors (Table 2). However, the within-method errors are normally distributed and are mechanistically consistent (Gregor et al., 2017). This allows us to observe changes that are smaller than the within-method error. The between-method error (shown in Figure 2d) serves as a better measure of whether observed variability is
- .45 more than statistical noise as it incorporates the three methodologically different approaches.





Figure 2: Time series of the five<u>three</u> ensemble members for each biome as defined by Fay and McKinley (20142014a): (a) SAZ, (b) PFZ, (c) AZMIZ. (d) shows the standard deviation between ensemble members for the three biomes<u>which is analogous to the between-model error (equation 5)</u>. The SOM-FFN data ends at the end of 2011. This is indicated in (d) by the dashed line. within-method ( $E_w$ ) and between-method ( $E_b$ ) errors are shown for each biome. For a more detailed breakdown of the errors see Table 2.

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Figure 2 shows <u>the Δ*p*CO<sub>2</sub></u> time series <u>of the for</u> each of the methods for the three Southern Ocean biomes as defined by Fay and McKinley (2014)... The methodological and data driven differences between each of the <u>approachesmethods</u> have been addressed in Gregor et al. (2017). In general, there is good agreement amongst the methods with a few notable exceptions. In the SAZ (Figure 2a) <u>The the</u> SOM-FFN differs from <u>all the</u> other methods for summer and autumn from 1998 to 2008. Gregor et al. (2017) attributed this difference to the clustering step <u>used byin</u> the SOM-FFN that <u>created largecreates</u> discrepancies in the

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Atlantic sector. The SVR-LR method overestimates the seasonal amplitude ΔpCO<sub>2</sub> (where the seasonal amplitude is the difference between the winter maxima and summer minima of ΔpCO<sub>2</sub>) relative to the other methods for winter 2012 to 2014. In the PFZ (Figure 2b), the SVR methods (LR and HR) overestimateoverestimates ΔpCO<sub>2</sub> relative to the other methods during winter from 1998 to 2004, likely due to the sensitivity to sparse winter data. The spread of data in the AZ is much larger than the two other regions, but the impact on the fluxes is reduced due These differences contribute to ice cover during winter
.65 (Ishii et al. 1998; Bakker et al. 2008; Butterworth and Miller the between-method error. 2016).



Figure 3: The mean spatial standard deviation (a) of the ensemble members (SVR-LR, SVR-HR, RFR-LR, RFR-HR and SOM-FFN) is shown to represent the "noise" of the ensemble mean. The "signal" (b) is calculated as the mean difference between the minimum and maximum values of the annually averaged  $\Delta pCO_2$ . This shows the signal that needs to be detected by the ensemble. The signal to noise ratio (c) shows regions where the confidence in the ensemble estimate is large, where darker shows higher confidence.

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The results from Figure 2 are summarised with the standard deviation of the ensemble members over time (Figure 2d) and space (Figure 3). As noted, the AZ (Figure 2c) has the largest disagreement amongst methods shown by map (Figure 3a) and the differences between the solid and dashed lines in Figure 2d, particularly during summer and autumn. This is likely due to the inability of the methods to accurately capture the larger intra-seasonal variability and patchiness in the AZ where the rapid reduction of  $pCO_{25}$  due to melting sea ice leads to patchy  $pCO_2$  distributions (Bakker et al. 2008; Chierici et al. 2012). The ensemble members are more coherent in the SAZ and PFZ.

In order to ascertain a degree of coherence and confidence in the ensemble we show the signal to noise ratio in Figure 3c. We define the signal as the largest difference in the trend for a particular point. This is calculated from the largest difference of annual averages of Δ*p*CO<sub>2</sub>. The noise is the mean standard deviation of ensemble member estimates. A large signal to noise ratio (Figure 4c) is indicative of a large trend signal compared to the variability of the ensemble. While signal to noise ratio is > 1 for the entire domain, there are regions where the ratio is < 2: parts of the Atlantic sector of the SAZ and the Indian sector of the PFZ.</p>

With the established baseline of confidence in the ensemble, the ensemble mean of  $\Delta pCO_2$  can be assessed. The seasonal cycle is the strongest mode of  $\Delta pCO_2$  variability in the Southern Ocean (Lenton et al., 2013). It is therefore important that the ensemble mean is understood in the context of our current understanding of the seasonal cycle.

The seasonal amplitude of  $\Delta p CO_2$  in the MIZ is much larger than the two other regions. However, this amplitude is likely to be dampened by ice cover (Ishii et al., 1998; Bakker et al., 2008; Butterworth and Miller, 2016). Note that in this study, we do not include regions with sea-ice cover to ensure consistency

95 between methods. Calculated fluxes for this methodologically reduced region will thus under-represent the fluxes of the full extent of the MIZ. We thus exclude the MIZ for the remainder of the study.

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Figure 2d shows the time evolution of between-method errors for each biome. This panel highlights the seasonality of the data, specifically the increased heterogeneity of  $\Delta pCO_2$  in summer and the impact that

00 this has on  $\Delta pCO_2$  estimates. This is due to the more complex competing processes affecting  $\Delta pCO_2$ during summer. To gain a better understanding of the seasonal processes we look at the mean state of each season to characterise the drivers of opposing fluxes.

#### 3.2 Ensemble seasonal cycle



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Figure 4<u>3</u>: The mean seasonal states of  $\Delta pCO_2$  of the empirical ensemble mean  $\frac{Seasonal averages of <math>pCO_2$  for the ensemble of  $\Delta pCO_2$ . These are shown for (a) summer, (b) autumn, (c) winter and (d) spring. The black contour lines show the SAZ, PFZ and  $\frac{AZMIZ}{D}$  from north to south as defined by Fay and McKinley (20142014a).

The seasonal cycle of the  $\Delta p CO_2$  for each biome (Figure 2a-c and Figure 4a3a-c) is coherent with expected 10 seasonal processes based on reported in the literature (Metzl et al., 2006; Thomalla et al., 2011; Lenton et al., 2012; Lenton et al., 2013). In all biomes, uptake of CO<sub>2</sub> is stronger during summer than in winter giving rise to the strong seasonal cycle. This is due to the opposing influences of the dominant winter and summer drivers, partially damped by the seasonal cycle of temperature (Takahashi et al., 2002; Thomalla et al., 2011; Lenton et al., 2013). The dominant processes of mixing and entrainment in winter 15 results result in increased surface  $pCO_2$  and thus outgassing (Takahashi et al., 2009; Lenton et al., 2013; Rodgers et al-, 2014). In summer, stratification also allows for increased biological production and the consequent uptake of  $CO_2$ , thus reducing the entrained winter DIC and associated  $pCO_2$  (Bakker et al-., 2008; Thomalla et al., 2011). However, stratification typically limits entrainment, but does not exclude the occurrence of entrainment during periods of intense mixing driven by storms, which. This has an 20 impact on both primary productivity, DIC and pCO<sub>2</sub> (Lévy et al., 2012; Monteiro et al., 2015; Nicholson et al., 2016; Whitt et al., 2017).

The SAZ (Figure 2a) is a continuous sink where summer uptake (Figure 4a3a) is enhanced by biological production and winter (Figure 4e3c) mixing results in a weaker sink (Metzl et al., 2006; Lenton et al.,

- 25 2012; Lenton et al., 2013). These The same processes produce a similar seasonal amplitude in the PFZ (Figure 2b), but the seasonal fluxes are stronger upwelling and weaker biological uptake result in a positive shift of the mean. This results in an opposing: a sink in net summer (<0  $\mu$ atm)sink and a source in winter (>0  $\mu$ atm)source. However, this is according to the mean state in the PFZ and winter estimates of  $\Delta pCO_2$  do in fact approach 0  $\mu$ atm toward the end of the time series (Figure 2b). The AZMIZ has the strongest
- 30 seasonal cycle due to upwelling of CO<sub>2</sub> during winter and strong biological uptake in summer. However, much of this is dampened by sea ice cover during winter and weaker winds during summer (Ishii et al., 1998; Bakker et al., 2008).

It<u>Apparent also from Figure 3</u> is important to note that, over and above the latitudinal gradient,  $\Delta pCO_2$  is 35 zonally asymmetric within each biome, particularly during summer (Figure 4a)3a), when biological uptake of CO<sub>2</sub> increases. Zonal integration of  $\Delta pCO_2$  and  $FCO_2$ -could thus dampen magnitudes of regional signals $\Delta pCO_2$ . A regional approach is therefore needed to examine the regional characteristics of seasonal and interannual variability of  $\Delta pCO_2$  and  $FCO_2$ -and to understand theirits drivers.

#### 3.3 Regional <u>ApCO<sub>2</sub> and FCO<sub>2</sub> Variability: Zonalvariability: zonal</u> and basin contrasts

40 Here  $\Delta pCO_2$  and  $FCO_2$  are is decomposed into nine domains by biome and basin with the boundaries shown in Figure 1-, but note only six are shown in Figure 4. The data are plotted as time series for  $pCO_2$ (Figure 5) and  $FCO_2$  (Figure 64) showing: the mean annual trends of  $pCO_2$  and  $FCO_2$  (black lines), the maximum winter values (red line) and the projected summer minima (dashed red line) based on adjusting the winter maxima each year by the mean of the difference between the winter maxima and the summer

- minima (Figures 5, 6).). The projected summer minima implies that there is an expectation(dashed lines) 45 are calculated by subtracting the mean seasonal amplitude from the winter maxima (Figure 4, with airsea CO<sub>2</sub> fluxes shown in Figure S3). The projected summer minima are the expected summer  $\Delta p$ CO<sub>2</sub> under the assumption that summer  $\Delta p CO_2$  is dependent on, but not restricted to, the baseline set by the winter maxima. Differences between the ensemble summer minima and projected minima are highlighted 50 with green and blue patches, highlighting periods of decoupling between summer and winter interannual

variability. The green areas indicate periods of strong uptake (relative to winter) that enhance the mean uptake of CO<sub>2</sub>- and amplify the seasonal cycle. Conversely, blue areas show periods where weak summer uptake (relative to winter) offsets winter trendoutgassing, thus reducing the mean trend.  $\Delta p CO_2$  as well as supressing the amplitude of the seasonal cycle (Figure 4).





Figure 54: Figures (a-if) show the ensemble mean of  $\Delta pCO_2$  (dark grey) and (j-r) show  $FCO_2$  (dark grey) plotted by biome (rows) and basin (columns). Biomes are defined by Fay and McKinley (20142014a). The solid red line shows the maximum for each year (winter outgassing) and the dashed line shows the same line less the average difference between the minimum and maximumseasonal amplitude – this is the expected amplitude. The shaded blue (green) area shows when the annual minimum is less (greater) than the expected amplitude.  $\Delta pCO_2E_b$  is the average between-method error and  $\Delta p\overline{CO}_2$  is the average for plots (g-i) was normalised to sea ice cover, but under ice  $\Delta pCO_2$  estimates were still used to find the expected amplitude. the entire time series. Light grey shading in (a-if) shows the proposed periods used in Figure 9 and Figure 10 Light grey shading in (j-r) shows the "saturation" period (1998 to 2001) and the "reinvigoration" period (2002 to 2011).5 and Figure 6.

The data for Δ*p*CO<sub>2</sub> and *F*CO<sub>2</sub> (Figures 5 and 6); (Figure 4) show that the Southern Ocean sink strengthened from 2002 to 2011 in all domains, a period identified as the *reinvigoration* by Landschützer et al. (2015). This was preceded by a period of a net weakening sink in the 1990's referred to as the *saturation* period after Le Quéré et al. (2007). These two periods are highlighted by the grey fill in Figure 6. The saturation of the Southern Ocean CO<sub>2</sub> sink is not as strong in the ensemble, occurring in only five of the nine domains (see the positive trend slopes between 1998 and 2002 in Figure 6). In the last-period (from 2012 to 2014) of the ensemble, three domains (Figure 4a,c,f) go from growing uptake to reducing uptake; however, our confidence in the increasing trends from 2012 to 2014 changing trend is low due to lack of coherence between methods (Figure 2a,b) and only three years of data, with very sparselittle data

in 2014.

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Importantly, these <u>A key feature of Figure 4 is that the mean</u> interannual trends are variability is the integrated net effect of decoupled seasonal modes of variability. for summer and winter. This is particularly evident in the PFZ (Figures 5d4d-f). Here, and in the other biomes, there is a the net strengthening of the CO<sub>2</sub> sink due is mainly linked to a reduction of  $\Delta p$ CO<sub>2</sub> in winter on roughly a decadal mode. for the majority of the time series. This corresponds with the findings of Landschützer et al.,

(2016), who linked the <u>decadal variability</u>reinvigoration to <u>the decadal variability of</u> the Southern Annular Mode (SAM) – the dominant mode of atmospheric variability in the Southern Hemisphere (Marshall,

- 85 2003). In comparison<u>contrast</u>, summer  $\Delta pCO_2$  variability is <u>sub-decadal or shorter (roughly 4 6 years;)</u>, thus impacting the short termproviding inter-annual modulation of longer time-scale winter variability of the annually integrated trend. This is demonstrated well in the Indian sector of the PFZ where a decrease in winter  $\Delta pCO_2$  from 2002 to 2011 is offset by weakening of the summer sink from 2006 to 2010 (Figure 5d, 6d4d). Similarly in the Atlantic and Pacific sectors of the <u>SAZ and PFZ strong</u> decoupling occurs
- 90 from  $\sim 2011$  to the end of 2014 with a rapid increase in the strength of the summer sink.

The mean amplitude of the seasonal cycle of  $\Delta pCO_2$ , the mean difference between the summer minima and the winter maxima, is perhaps a better means of understanding the magnitudestrength of the seasonal drivers for each domain than the mean  $\Delta pCO_2$  (Table A1). For example the Atlantic sectors of the SAZ and PFZ (Figures 5e4c,f) have the strongest seasonal variability (20.5714.11 and 27.6725.83 µatm

- and PFZ (Figures 5e4c, f) have the strongest seasonal variability (20.5714.11 and 27.6725.83 μatm respectively). This contrasts the relatively weak seasonal amplitude in the Indian sector of the Southern Ocean which has mean amplitudes of 8.857.06 and 13.3164 μatm for the SAZ and PFZ respectively (Figures 5b4b,e). This contrast can also be seen by comparing the mean seasonal maps of Δ*p*CO<sub>2</sub> in Figures 4a3a and 4e3c. In summer, strong uptake in the eastern Atlantic sector of the southern ocean is indicative of large biological drawdown of CO<sub>2</sub> by phytoplankton (Thomalla et
- al., 2011). Conversely, relatively low primary production in the Indian sectors of the SAZ and PFZ resultresults in a small seasonal amplitude (Thomalla et al., 2011). This large discrepancy in biological primary production is related to the availability of iron, a micronutrient required for photosynthesis. The lack of large land masses, a source of iron, in the Indian sector of the Southern Ocean could be a contributing factor to the lack of biomass (Boyd and Ellwood, 2010; Thomalla et al., 2011).

The seasonal amplitude in the AZ is much larger due to strong contrast of the upwelling of  $CO_2$  rich deep water contained beneath winter sea ice and the strong biological drawdown in the beginning of summer (Ishii et al., 1998, Bakker et al., 2008). Rapid stratification and iron supply by melting sea ice provide the

•10 environment for phytoplankton to proliferate in the AZ. This results in large seasonal amplitudes of 46.72,
 75.46 and 64.29 μatm for the Indian, Pacific and Atlantic.



+15 Figure 6: Figures (a-i) show  $\Delta pCO_2$  (dark grey) and (j-r) show  $FCO_2$  (dark grey) plotted by biome (rows) and basin (columns). Biomes are defined by Fay and McKinley (2014). The solid red line shows the maximum for each year (winter outgassing) and the dashed line shows the same line less the average difference between the minimum and maximum—this is the expected amplitude. The shaded blue (green) area shows when the annual minimum is less (greater) than the expected amplitude.  $\Delta pCO_2$  for plots (g-i) was normalised to sea ice cover, but under ice  $\Delta pCO_2$  estimates were still used to find the expected amplitude. Light grey shading in (a-i) shows the proposed periods used in Figure 9 and Figure 10 Light grey shading in (j-r) shows the "saturation" period (1998 to 2001) and the "reinvigoration" period (2002 to 2011).

- 20 The air sea fluxes of CO<sub>2</sub>-(FCO<sub>2</sub>) have decadal trends that are coherent with the pCO<sub>2</sub> (Figure 6), but there are notable differences that emerge from the impact of wind on the rate of exchange as well as the surface area of each domain (Figure A1). Most prominent are the changes in the seasonal cycle and the mean seasonal sink of FCO<sub>2</sub> relative to pCO<sub>2</sub> with amplification in the Indian sector (Figures 5a,d and 6a,d) and weakening in the Atlantic Ocean (Figures 5c,f and 6c,f) of the Southern Ocean. The Indian sector of the SAZ (Figure 6a) dominates the uptake of CO<sub>2</sub> with an annual mean flux of -0.25PgC yr-1 compared to -0.19 and -0.17 PgC yr-1 for the Atlantic and Pacific sectors respectively (where the latter are significantly different with p = 0.01). The seasonality of wind stress (see Figure A1) results in a damped seasonal cycle of FCO<sub>2</sub> in the SAZ and increasing intra-seasonal variability (compared to ΔpCO<sub>2</sub>), with stronger winter winds compensating for a weaker ΔpCO<sub>2</sub> gradient (Monteiro et al., 2015).
- -30

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This contrasts the PFZ, where opposition of summer uptake and winter outgassing of  $CO_2$  is amplified by stronger wind stress (Figure 6d-f). Interannual variability is also enhanced, particularly during winter in the Indian sector of the PFZ, where a reduction in outgassing of 0.18 PgC yr<sup>-1</sup> is observed. The decoupling between summer and winter  $FCO_2$  also becomes more pronounced in this region (Figure 6d), resulting in a lag in the decreasing trend. In other words, the trend of  $FCO_2$  for the reinvigoration (2002

through 2011: -10.85 PgC yr-1) would have been stronger if the decoupling had not occurred. Similarly,

the seasonal decoupling in the Pacific sector of the SAZ and PFZ results in a stronger growing sink from 2012 to 2014. In the Atlantic sector of the SAZ and PFZ the earlier onset of the seasonal decoupling (Figure 5c,f) also means that re-coupling occurs sooner, resulting in a positive flux trend (Figure 6c,f).

40 Lastly,  $FCO_2$  in the MIZ is damped during winter due to ice cover and weaker winds during summer when  $\Delta pCO_2$  is low due to the short-lived intense biological uptake of  $CO_2$  (Ishii et al. 1998; Bakker et al. 2008).

#### 3.4 Seasonal deconstruction of interannual variability

Figures 5 and 6 give Figure 4 gives us insight into the magnitude of  $FCO_2$  and  $ApCO_2$ -interannual  $\Delta pCO_2$ variability as well as the character of these changes; *i.e.* decoupling of decadal interannual winter and subdecadal summer interannual modes of variability. This alludes to the factpoint that  $\Delta pCO_2$  and  $FCO_2$ -are is responding to different adjustments of seasonal large scale atmospheric forcing and/or responses of internal ocean dynamics in the Southern Ocean (Landschützer et al., 2015, 2016; DeVries et al., 2017). In the study by Landschützer et al., (2015) it was advanced that the explanation for the reinvigoration of

- 50 Δ*p*CO<sub>2</sub> uptake in the 2000s decade was linked to the net thermal control driven by a response of DIC and temperature to asymmetric atmospheric forcing over the Southern Ocean. More recently, a study by DeVries et al., (2017) proposed that the Southern Ocean uptake was due to the global deceleration of the Meridional Overturning Circulation (MOC). They suggested that the MOC was increasing the oceanic storage and reducing the losses of CO<sub>2</sub> to the atmosphere, particularly in the Southern Ocean. The
- .55 mechanism proposed by DeVries et al. (2017) is the same as that put forward by Le Quéré et al. (2007) and Lovenduski et al. (2008) amongst others, where the changes in outgassing are related to the strength of the westerly winds over the Southern Ocean. These studies have in linked the wind stress variability to SAM.
- In order to capture the decoupled short term variability observed during summer, the data are brokendivided into four interannual periods (P1 to P4)), where P1 is five years and the remaining periods (P2 to P4) are four years as shown by the light grey fills in Figure 5. The first period is the saturation period (P1: 1998 2002) by Le Quéré et al. (2007). The second and third periods are informed by the reinvigoration period (2002 through 2011) split around the start of 2007 an early, weaker reinvigoration
- 65 (P2: 2002 2006) and a late, stronger reinvigoration (P3: 2007 2011). The last period incorporates the three years of new data (P4: 2012 to 2014).4. The small discrepancy in the length of the periods is due to the uneven length of the time series (17 years).

These four periods are too short for trend analyses (Fay and McKinley, <u>20142014b</u>), but the intention here is to identify periods that are short enough to resolve interannual changes in theof large-scale drivers of the winter and summer mean values for  $pCO_2$  and  $FCO_2$  that would otherwise be averaged out over longer periods. We perform anthen calculate the relative anomaly analysis between each <u>successive</u> period rather than a trend analysis (for which inflections of  $\Delta pCO_2$  would be more suitable delimiters). The relative anomalies in an anomaly of the drivers are their differences between two

- .75 adjacent periodsmean state (e.g. P2 P1). As a result four periods give rise to three <u>sub-decadal-scale</u> transition anomalies for summer and winter: A (P2 P1), B (P3 P2) and C (P4 P3). We do this separately for each method rather than using the ensemble mean (see S4 for calculations). The mean of the method anomalies for each transition is then taken. These anomalies are considered significant if the absolute estimate of the anomaly is larger than the standard deviation between the methods for each
- •80 period. These calculations along with plots for the standard deviation between methods are shown in the supplementary materials in S4.

Note that, although only summer and winter <u>anomalies</u> are discussed, it is recognised that autumn and spring could be equally mechanistically important. Winter anomalies of  $\Delta p CO_2$ , wind stress, SST and MLD are shown in Figure 75. Summer anomalies of  $\Delta p CO_2$ , wind stress, SST and Chl-*a* are shown in

Figure <u>86</u> where MLD, in winter, is replaced with Chl-*a* for summer as it is potentially a more important driver than the generally shallow <u>MLD in summer MLD</u> (the omitted plots are shown in Figures <u>A2S5</u> and <u>A3S6</u>).

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•90 Figure 7: Transitions of winter  $\Delta pCO_2$ , wind stress, SST and MLD]{Transitions (relative anomalies) of winter  $\Delta pCO_2$  (a-c), wind stress (d-e), sea surface temperature (f-h) and mixed layer depth (i-k) for four periods (as shown above each column). The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and McKinley 2014) and basin boundaries.

#### **3.5 Drivers of the decadal winter trends in** ApCO<sub>2</sub> and FCO<sub>2</sub>

There are two features of interest in the anomalies of winter  $\Delta p CO_2$  and its drivers (Figure 7). First, there .95 is a zonally asymmetric dipole for wind stress between the Pacific and Indian sectors of the Southern Ocean that dominates transitions A and B. Second,  $\Delta p CO_2$ , SST and MLD cohere roughly to the spatial variability of wind stress anomalies, mirroring the wind dipole. These features will be expounded on in the paragraphs that follow.

#### 00 <u>3.5 Drivers of winter △pCO<sub>2</sub> variability</u>

We will limit the interpretation of the changes to the regions where the anomaly is larger than the betweenmethod error of anomalies (see S4 for calculations and maps). This masks out large regions, but three key points still arise from the significant anomalies. Firstly,  $\Delta pCO_2$  is often spatially roughly coherent with wind stress and the inverse of SST. Secondly, there is a dipole in the wind anomalies in the Indian and

O5 Pacific between transitions A and B. This is confirmed by the *u*- and *v*-components of wind shown in the supplementary materials (Figure S5). Lastly, the Indian sector of the Southern Ocean dominates the reinvigoration of the CO<sub>2</sub> sink. These points are now addressed in more detail.

Transition A (the transition from P1, the saturation period, to P2, the start of the reinvigoration – P1)
shows a relative increase of Δ*p*CO<sub>2</sub> in the east Indian and Pacific sectors of the SAZ – suggesting a delay in the onset of the reinvigoration for these basins. This regional sustained saturation corresponds to a shift towards stronger winds and/or deeper MLDs in the west of the Tasman Sea (Pacific sector of the SAZ) and surrounds (Figure 7d5d,j). In contrast, CO<sub>2</sub> uptake in the east Atlantic and west Indian sectors of the SAZ, and the south-eastern Indian sector of the PFZ show a reinvigoration of CO<sub>2</sub> uptake. In the central Pacific, weaker wind stress (green in Figure 7d) start to strengthen, which roughly corresponds with relative warming and shoaling or stagnation of MLD. This is consistent with an invigoration of the Southern Ocean CO<sub>2</sub>-sink (P1 – P2: 1998 – 2007) initiated by a weakening of the mean westerly wind stress in the Pacific and W Indian Oceans, which we are suggesting, reduced winter entrainment and possibly upwelling of CO<sub>2</sub>-rich deep waters (Marshall, 2003; DeVries et al., 2017).the weaker winds.

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Transition B, which corresponds to anomalies between (P3 – P2-to P3 (the two reinvigoration periods),) is characterized by an a further intensification of the invigoration of  $\Delta pCO_2$  (-ve(negative shift) in all basins, but particularly primarily in the Indian basin (Figure 7b). This 5b). Once again the strengthening of the CO<sub>2</sub> uptake corresponds with weaker wind stress, a warming trend in surface waters and shoaling

MLDs in the <u>SEeastern</u> Atlantic and Indian Ocean sectors <u>inof</u> the <u>winterSAZ and PFZ</u> (Figure 7b5b,e,h,k). In the Pacific, the <u>The</u> opposing effects of the dipole are: <u>observed east of New Zealand where</u> stronger wind stress, deeper MLD, and cooler surface waters. <u>These changes are associated correspond</u> with the persistence of a neutral to weak reduction of positive shift in ΔpCO<sub>2</sub>-compared to the Indian sector. All the changes in transitions A and B are coherent to changes in the Pacific Indian wind stress
 dipole.

In transition C (P4 – P3: 2012 – 2015), when we propose), the invigoration trend starts to weaken,  $\Delta pCO_2$ sink strengthens further in the northern and southern extremes of the east Indian and west Pacific basins, albeit in a more spatially heterogeneous way. In \_. This negative shift corresponds well with strong

35 <u>shoaling of the AtlanticMLD (Figure 51). The west Pacific sector of the previous invigoration trend</u>

# reverses completely and the CO<sub>2</sub> sink <u>PFZ</u> shows a positive shift in $\Delta pCO_2$ , which is coherent with an <u>increase</u> in the <u>wind stress and deepening MLD</u>.



40 Figure 5:Transitions (relative anomalies) of winter is shown to weaken (Figure 7b,c). The previously well characterized Pacific Indian dipole is not apparent, suggesting that transition A∆pCO<sub>2</sub> (a-c), wind stress (d-f), sea surface temperature (g-i) and mixed layer depth (j-l) for four periods. The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and B capture well established phasesMcKinley, 2014a) and basin boundaries. Regions with dots in the decadal variability, while transition C may be capturing a transition into the following

45 phase which could take(a-c) are where the system back to anomalies are not significant ie: standard deviation of the same configuration as P1. The notion that this period is a snapshotanomalies between two distinct phases is supported by the relatively heterogeneous spatial structure of wind stress in both the Atlantic and Indian Oceans. models is greater than the absolute mean of method anomalies as described in equations S1 to S3.

#### 3.5.1 Wind dominated interannual variability of pCO<sub>2</sub> in winter

- 50 WeBased on the observations outlined above, we propose that the interannual variability of seasonal the regional (basin-scale) characteristics of winter wind stress in winter may be the dominant driver of the saturation and reinvigoration periods. Moreover, the suggested Pacific Indian wind dipole may be linked to the decadal variability of  $\Delta pCO_2$  observed in the Southern Ocean (Landschützer et al., 2016).
- 55 Increasing These findings suggest that increasing or decreasing interannual winter wind stress variability impacts  $\Delta p CO_2$  (and thus  $FCO_2$ ) by driving strongerchanges in turbulent mixing- that set the magnitudes of winter entrainment. In the transition to and during winter, this mixing is associated with changes in rates of heat loss resulting in athat drive loss of buoyancy or weaker stratification (Abernathey et al-2011). Weaker buoyancy facilitates deepening of the MLD, thus entraining DIC-rich deep waters
- 60 (Abernathey et al., 2011; Lenton et al., 2013). Conversely, decreased wind stress and mixing during winter (on seasonal or interannual time scales) reduces the rate of heat loss (represented as warm anomalies in Figure 75). This results in stronger stratification and shallower winter MLD limits entrainment of DIC, which strengthens the CO<sub>2</sub> winter disequilibrium and leads to a stronger CO<sub>2</sub> sink anomaly (Figure 7). This is5). These are the mechanismmechanisms that results we propose result in
- 65 decreasing or increasing fluxes with <u>interannual and basin-scale</u> changes in wind. However, the direct link between this wind stress mechanism and the reinvigoration was not made by Landschützer et al. (2015). This may be, in part, due to the seasonal decoupling that may lead to biased interpretation of wind stress and SST.
- 70 We propose the link between spatial changes in wind stress and uptake of  $CO_2$  as an alternative hypothesis to temperature being a driver as suggested by Landschützer et al. (2015). Typically an increase in ocean temperature, which reduces  $CO_2$  solubility, results in an increase in  $\Delta pCO_2$  (Takahashi et al., 1993). However, seasonal – regional analysis shows that the observed relationship between  $pCO_2$  and SST is counterintuitive (Figure 5a-c,g-i). On this basis we propose that SST is not a driver of  $pCO_2$  in winter.
- 75 We suggest that this relationship is a product of weaker mixing and Ekman transport that allows warmer waters to shift southward. This also has the impact of strengthening buoyancy that would otherwise bring  $CO_2$  to the surface. In summary, our results suggest that, like  $pCO_2$ , the SST changes are also a response to the wind stress and not in themselves the drivers of  $pCO_2$  changes.

- Given the hypothesis that wind stress is the dominant driver of interannual decadal ΔpCO<sub>2</sub> in winter, it is of interest to understand its potential mechanisms. Past studies have used the SAM as a proxy for wind stress variability over the Southern Ocean, where the multi-decadal increasing trend has been cited as a reason for the saturation in the 1990's (Marshall, 2003; Le Quéré et al., 2007; Lenton and Matear, 2007; Lovenduski et al., 2008). While Landschützer et al. (2016) identified the SAM as being a driver of global CO<sub>2</sub> variability, the index does not explain the reinvigoration of the Southern Ocean-<u>CO<sub>2</sub> sink in the 2000s</u>. The SAM is often represented as a zonally integrating index (Marshall, 2003), but more recent studies have shown that the SAM, as the first empirical mode of atmospheric variability, is zonally asymmetric (Fogt et al-<u>2012-</u>). The zonal asymmetry of the SAM is linked with the El Niño Southern Oscillation and is strongest in winter, particularly over the Pacific sector of the Southern Ocean during a positive phase, thus in accord with the Pacific–Indian winter wind stress dipole observed in Figures 5d,e (Barnes and Hartmann<sub>2</sub> 2010; Fogt et al-<u>2012</u>). Fogt et al. (2012) noted that the SAM has become more
  - zonally symmetric in summer since the 1980's, matching the wind stress anomalies seen in Figure 76d-f.
- DeVries et al. (2017) proposed that slowing down of Meridional Overturning Circulation (MOC) as an alternate mechanism for the reinvigoration of the Southern Ocean CO<sub>2</sub> sink in the 2000's. The authors explain that weaker overturning reduces the natural CO<sub>2</sub> brought from the deep to the surface ocean. Moreover, they suggest that this mechanism may continue to drive intensification of the global CO<sub>2</sub> sink. The longer modes of MOC variability makes it difficult to attribute the change in flux to changes in overturning.

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This poses an interesting question for the Southern Ocean carbon sink when we consider that weakening MOC may counteract the intensification of winds over the Southern Ocean (encapsulated by the increasing SAM). Meredith et al. (2012) found that this question is made more complex by the compensatory effect of increased eddy activity (measured by eddy kinetic energy EKE) to enhanced northward Ekman transport driven by intensified winds (Meredith and Hogg, 2006; Abernathey et al., 2011; Marshall and Speer, 2012). Moreover, the inclusion of these eddies in a high resolution model reduced CO<sub>2</sub> outgassing driven by increased Ekman transport by one third by entraining alkalinity to the surface water (Dufour et al., 2013). As it stands, this is an unresolved question and more work will have to be undertaken to understand the effect of these two counteracting mechanisms of CO<sub>2</sub> transport.

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In summary, we propose that interannual variability of wind stress and its regional expression in winter is the dominant interannual driver of  $FCO_2$ - $\Delta pCO_2$  variability in the Southern Ocean. The interannual variability of wind stress is linked to the SAM, but this relationship is nuanced by the zonally (regional) 15 Indian sectors of the Southern Ocean.



Figure 8: Transitions of summer  $\Delta pCO_2$ , wind stress, SST and Chl-a]{Relative anomalies of summer  $\Delta pCO_2$  (a-c), wind stress (d-e), sea surface temperature (f-h) and mixed layer depth (i-k) for four periods (as shown above each column). The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and McKinley 2014) and basin boundaries.

#### 3.6 Trends in the anomalies Anomalies of $\Delta pCO_2$ and its summer drivers in summer

The most <u>importantmarked</u> difference between the summer and winter anomalies, is that <u>wind</u> <u>stress $\Delta pCO_2$ </u> (Figures <u>8d-f6a-c</u>) does not correlate to  $\Delta pCO_2$  with wind stress (Figures <u>8a-e6d-f</u>), thus ruling out wind as a first order driver of summer CO<sub>2</sub>. Rather,  $\Delta pCO_2$  has the strongest coherence with Chl-a (an inverse relationship), which suggests that primary production may be a first order driver of the

observed  $\Delta p CO_2$  variability. Another difference between summer and winter is that the magnitudes of the transition anomalies are much larger in summer, and thus there are larger regions of significant anomalies (Figure 6a-c).

Looking more specifically at the significant variability of  $\Delta p CO_2$ , Transition transition A (P2 – P1 in

- Figure <u>8a6a</u>) is marked by <u>patchy decreases a decrease of CO<sub>2</sub> in regions of high EKE (Agulhas retroflection and the SAZ (Tasman shelf) in the SAZ and AZ, region), mirrored by an increase in Chl-a. The Atlantic and Indian sectors of the PFZ remain <u>mostly</u> neutral/weak sources marked by a reduction in phytoplankton biomass (Figure <u>8j6j</u>). Transition B (P3 P2 in Figure <u>8b6b</u>), shows invigoration of CO<sub>2</sub> uptake in: the Atlantic sector of the SAZ and PFZ; the Indian sectorand in parts of the AZ; and patchy</u>
- strengthening in the Pacific Ocean. Once again, the reduction of  $\Delta pCO_2$  from P2 to P3 in the aforementioned regions correlate<u>correlates</u> well with Chl-*a*<u>increases</u>. In transition C (P4 – P3 in Figure <u>8e6c</u>) the reduction of the  $\Delta pCO_2$  is widespread in the Indian and Pacific Oceans in all three biomes, though this does not necessarily correspond with as the increase in Chl-*a*. There is a strong decrease<u>similarly widespread</u>. Conversely, there is a reduction in Chl-*a* and concomitant increase in
- $\Delta p \text{CO}_2$  along Polar front in the Atlantic sector, coinciding with position of the ACC<sub>a</sub> which has high EKE (Meredith, 2016). These examples demonstrate that  $\Delta p \text{CO}_2$  is driven primarily by Chl-*a* in summer. However, understanding Chl-*a* variability is perhaps-more complex as there is seemingly no set rule between Chl-*a*, SST and wind stress (Thomalla et al., 2011).



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Figure 6: Relative anomalies of summer  $\Delta pCO_2$  (a-c), wind stress (d-f), sea surface temperature (g-i) and mixed layer depth (j-l) for four periods (as shown above each column). The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and McKinley, 2014a) and basin boundaries.

There are regions in the Southern Ocean where summer Chl-*a* variability does not coincide with  $\Delta pCO_2$ variability, particularly in the Indian and Pacific sectors of the SAZ (Figures <u>8a6a</u>-c and <u>8j6j</u>-l). This may be due to <u>the fact that low</u> chlorophyll concentrations, and anomalies thereof, <u>are low</u> in these regions (Thomalla et al., 2011). <u>This mayAs a</u> result <u>in</u> the other variables, SST and wind stress, <u>becomingmay</u> <u>be</u> higher order drivers <u>in low chlorophyll regions</u>, as found by Landschützer et al. (2015) and Munro et al. (2015).

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It is thus important to understand the variability of SST and wind stress in summer. Large SST anomalies between the western Atlantic and eastern Pacific sectors vary as ana zonally asymmetric dipole. As in winter, there is a summer wind stress anomaly dipole, but rather than being zonally asymmetric (*e.g.* Pacific–Indian), the dipole has annular, north-south variability (Figures 7,8d5,6d-f). We suggest that these dipoles in the variability may indicate that the Southern Ocean, as a system, transitions between different

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Lastly, there An important note is seasonal variability in that the magnitudes of  $\Delta p CO_2$  and the its drivers. The have different magnitudes seasonally. For example, the anomalies of  $\Delta p CO_2$  and SST have agree

165 larger magnitudein summer than thein winter anomalies. Conversely, the wind stress anomalies are larger for winter than in summer. This is an important consideration for analyses that aim to understand the driving mechanisms, where annual averaging would make it difficult to decompose the true drivers of change.

#### 3.6.1 Chlorophyll dominated interannual anomalies of pCO<sub>2</sub> in summer

states forced by atmospheric variability (Landschützer et al., 2015).

- The fact<u>Our finding</u> that Chl-*a* is the dominant driver of interannual  $\Delta pCO_2$  variability should not be surprising given that models and observations support this <u>notion</u> (Hoppema et al., 1999; Bakker et al., 2008; <u>Mahadevan et al., 2011;</u> Wang et al., 2012; Hauck et al., 2013; 2015; Shetye et al., 2015). However, our data show that the dominance of interannual Chl-*a* variability over  $\Delta pCO_2$  is largely limited to regions where Chl-*a* is high, such as the Atlantic, the Agulhas retroflection and south of Australia and Naw Zealand (Figure 97).
- New Zealand (Figure 97).

However, the <u>The</u> spatial variability of high Chl-*a* regions in the Southern Ocean is complex due to the dynamics of light and iron limitation (Arrigo et al., 2008; Boyd and Ellwood, 2010; Thomalla et al., 2011; Tagliabue et al., 2014; 2017). This complexity is highlighted in Thomalla et al. (2011), where the

Chl-*a* is characterized into regions of concentration and seasonal cycle reproducibility (Figure 97). The seasonal cycle reproducibility (SCR) is calculated as the correlation between the mean annual seasonal cycle and the observed chlorophyll time series. Here we use the approach of Thomalla et al. (2011), in Figure 97, as a conceptual framework to understand the interannual variability of  $\Delta p$ CO<sub>2</sub>.



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Figure 9: Chl-a seasonal cycle reproducibility and iron supply mechanisms in the Southern Ocean]{(a) Regions of chlorophyll biomass and seasonal cycle reproducibility from Thomalla et al. (2011) (using ScaWIFS data). Seasonality is calculated as the correlation between the mean annual seasonal cycle compared to the observed chlorophyll time series. A correlation threshold of 0.4 was applied to each time series to distinguish between regions of high and low seasonality; similarly, a threshold of 0.25 mg m<sup>-3</sup> was used to distinguish between low or high chlorophyll waters. Black lines showing the fronts are the same as described in figure.

#### 3.6.2 High chlorophyll regions

While regions of high SCR (dark green in Figure 97) do not correspond with the interannual variability of Chl-*a* (Figure 8j6j-l), the framework by Thomalla et al. (2011) does present a hypothesis by which the variability of Chl-*a* and its drivers can be interpreted. This is, that the variability of Chl-*a* in a region is a complex interaction of the response of the underlying physics (mixing vs<sub>2</sub> buoyancy forcing<sub>5</sub>), which modulate light (via the MLD) and iron supply, to the interannual variability in the drivers (SST and wind stress). This complexity is exemplified by strong warming in the Atlantic during transition B, which results in both an increase and decrease in Chl-*a*, with inverse consequences for Δ*p*CO<sub>2</sub>. The effect is an even stronger transition C, where strong cooling in the Atlantic results in both a decrease and increase of Chl-*a* (Figure 8i6i,l). In both transition A and B the respective increase and decrease of Chl-*a* occur roughly over the ACC, while the opposing effects during transitions A and B occur roughly to the north and south of the ACC region. These temperature changes may impact the stratification of the region, but

complex interaction with the underlying physics results in variable changes in Chl-a.



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Figure 9: Chl-a seasonal cycle reproducibility and iron supply mechanisms in the Southern Ocean (a) Regions of chlorophyll biomass and seasonal cycle reproducibility from Thomalla et al. (2011) (using SeaWIFS data). Seasonality is calculated as the correlation between the mean annual seasonal cycle compared to the observed chlorophyll time series. A correlation threshold of 0.4 was applied to each time series to distinguish between regions of high and low seasonality; similarly, a threshold of 0.25 mg m<sup>-3</sup> was used to distinguish between low or high chlorophyll waters. Black lines show the Southern Ocean fronts calculated by sea surface height (Swart et al., 2010).

It is clear that, while there is a relationship between Chl-*a* and  $pCO_2$  as well as a relationship between wind stress and SST in summer, the relationship between wind forcing and, Chl-*a* and  $pCO_2$  is not as strong as in the winter anomalies (Figure 7). The reason for this is likely to 5). It may be that enhanced summer buoyancy forcing resulting from summer warming and mixed layer eddies drives a more complex response to wind stress in the form of vertical velocities (w) and mixing  $(Kz)_{72}$  which influence the iron supply and the depth of mixing depth (McGillicuddy, 2016; Mahadevan et al., 2012).

Mesoscale and sub-mesoscale processes may have a part to play in these dynamic responses of Chl-*a* to changes in SST and wind stress (amongst other drivers). For example, eddy-driven slumping is a submesoscale process that actscould act to rapidly shoal the mixed layer (Mahadevan et al-, 2012; Swart et al., 2015b; du Plessis et al., 2017). This allows phytoplankton to remain within the euphotic zone and thus grow (while iron is not limiting). Similarly, Nicholson et al. (2016) and Whitt et al. (2017) demonstrated that a combination of high and low frequency oscillation of down-front winds are able to enhance nutrient

<sup>25</sup> entrainment (includingsubmesoscale processes could supply iron) into to the mixed layer on the less dense side of a front. This has important implications for Southern Ocean fronts, where Chl-*a* may benefit from this entrainment mechanism combined with eddy-driven slumping that could subsequently rapidly shoal the mixed layer (Du Plessis et al., by submesoscale 2017).

'30 Storm driven, intra-seasonal mixing is another sub-mesoscale process that could alleviate iron limitation through shear-driven mixing along the base of the mixed layer (Nicholson et al., 2016). Importantly, both storm driven entrainment and the oscillatory enhancement of entrainment, these mechanisms rely on a mixing transition layer that has sufficient iron that is able to sustain growth—weak. Weak dissolved iron gradients in the Pacific and east Indian sectors of the Southern Ocean wouldcould explain the lack of phytoplankton in these regions (Tagliabue et al=., 2014; Nicholson et al., 2016). Much of the spatial character of the transition anomalies occurs at mesoscale, which strengthens the view that these mesoscale and sub-mesoscale processes may be key to explain changes in Chl-a (Figure 8j-1). This level of

#### mechanistic detail was not part of this study.6j-l).

#### 3.6.3 Low chlorophyll regions

- Entrainment and stratification can explain much of the variability in the eastern Pacific and Indian sector of the PFZ (with the exception of the wake of the Kerguelen Plateau). For example, in the eastern Pacific in transition A (Figure 8a6a,d,g), strong warming and weaker winds have little impact on Chl-*a*, but a decrease in  $\Delta pCO_2$  is observed. Conversely, cooling in the west Indian sector of the PFZ results in a weak increase in  $\Delta pCO_2$  during the same transition. In both these cases, the effect of cooling or warming on  $\Delta pCO_2$  is negligible relative to the impact of entrainment or stratification respectively. The effect is reversed in the eastern Pacific during transition B where strong cooling results in a weak reduction of  $\Delta pCO_2$  rather than the increase that would be expected from entrainment. This is the mechanism that Landschützer et al. (2015) describe in the Pacific, where enhanced entrainment of DIC and TA is compensated for by cooling. This emphasises that the balance between SST (as a driver of stratification)
- <sup>50</sup> and wind stress is far more important than in winter(2015) ascribed to the reduction of  $\Delta p CO_2$  in the Pacific, but the effect observed in Figure 6b is weak.

In summary, regions with high biomass, Chl-*a* integrates the complex interactions between SST, wind stress, MLD and sub-mesoscale variability resulting in large interannual *p*CO<sub>2</sub> variability compared to
 low biomass regions. In low Chl-*a* regions, wind driven entrainment/stratification are in general dominant over thermally driven changesmore likely drivers of Δ*p*CO<sub>2</sub>.

#### **4** Synthesis

In this study, <u>an</u> ensemble <u>meansmean</u> of empirically estimated  $\Delta p CO_2$  and <u>FCO<sub>2</sub></u> were is used to investigate the trends and the drivers of these trends in the Southern Ocean. The <u>ensemble mean</u>

<sup>60</sup> of <u>estimated</u>  $\Delta p CO_2$  <u>showed shows</u> that the seasonal cycle is the dominant mode of variability imposed upon a weaker interannual trend. The interannual variability was in accordance with past studies with the saturation of the Southern Ocean CO<sub>2</sub>-sink in the 1990's followed by the reinvigoration in the 2000's (Le Quéré et al., 2007; Landschützer et al., 2015).

- <sup>165</sup> Data was. The data are separated into-nine domains defined by functional biomes and oceanic basins to account for the roughly basin scale zonal asymmetry observed in preliminary analyses of  $\Delta p CO_2$  (Fay and McKinley, 2014). 2014a). A seasonal decomposition wasis applied to the nine-domains, revealing that winter and summer interannual trends are variability is decoupled for each region. It was shown that winter interannual variability has a decadal period The increase and summer interannual variability has a
- <sup>70</sup> shorter four to six year period.subsequent decrease of  $pCO_2$  (and air-sea  $CO_2$  fluxes) is in accordance with recent studies showing a saturation of the Southern Ocean  $CO_2$  sink in the 1990's followed by the reinvigoration in the 2000's (Le Quéré et al., 2007; Landschützer et al., 2015).

The AWe suggest that changes in the characteristics of the seasonal cycle of the drivers of pCO<sub>2</sub> define
 the interannual variability pCO<sub>2</sub>. In other words, the mechanisms that drive interannual modes of variability are embedded in the seasonal cycle.

We propose that winter trends, agreed with  $\Delta p CO_2$  variability is driven primarily by changes in winter wind stress variability, where the latter corresponds with the decadal variability associated with the .

- which influences the resulting convective entrainment of deep DIC-rich water masses (Lenton et al., 2009; 2013). This winter variability has a longer mode than summer inter-annual variability. We attribute this longer winter mode of variability to the Southern Annular Mode, which has a decadal mode (Lovenduski et al., 2008; Fogt et al., 2012; Landschützer et al<del>.</del>, 2016). We propose that  $\Delta pCO_2$  variability is driven by increased (decreased) wind stress which drives deeper (shallower) mixing, thus bringing  $CO_2$ -rich waters
- '85 to the surface (Lenton et al. 2009; 2013). This mechanism is strongest<u>likely dominant</u> in winter due to weaker buoyancy gradients. its role in large seasonal net heat losses that drive convective overturning of the water column.
- Our findings show<u>We suggest</u> that interannual summer variability of  $\Delta pCO_2$  occurs from a baseline set by an interannual winter trend. The<u>Moreover, the</u> shorter time-scale summer <u>interannual</u> variability of  $\Delta pCO_2$  (roughly 4 – 6 years) wasis driven primarily by Chl-*a*. Wind stress and sea surface temperature still influencedinfluence  $\Delta pCO_2$  in summer, but wereare lower order drivers. We propose that changes in atmospheric circulation influence primary production through the interannual variability of the summer seasonal peak is linked to the complex interactionsinteraction of mid-latitude storms with surface temperature the strong mesoscale and mixing at small scales (sub-mesoscale), gradients in the Southern
- <u>Ocean.</u>

#### **A Additional Materials**

#### A1 Mean ApCO<sub>2</sub> for Southern Ocean domains

The SAZ (Figures 5a-c) accounts for the majority of uptake in the Southern Ocean with a mean  $\Delta pCO_2$ of -25.31 µatm. However, there is a relatively large difference between the three sectors of the SAZ, with

- 00 mean ApCO<sub>2</sub>-values of -24.39, -22.24 and -30.48 µatm for the Indian, Pacific and Atlantic respectively. In the PFZ (Figures 5d-f) the sink is far weaker due to the opposing summer uptake and winter outgassing. with ApCO<sub>2</sub>-values of 0.23, -2.06 and -6.57 again in the respective order Indian, Pacific and Atlantic. Similarly, in the AZ (Figures 5g-i) mean estimates of  $\Delta pCO_2$  are muted by opposing seasonal signals with mean estimates of 4.83, -3.01 and -0.76 uatm.
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Table 2: Mean ApCO2 for each of the Southern Ocean domains shown in Figure 5, where the domains are defined according to Figure 1.

BIOME	Indian	<b>Pacific</b>	<b>Atlantic</b>
SAZ	<del>-24.39</del>	<del>-22.25</del>	<del>-30.48</del>
PFZ	<del>0.23</del>	<del>-2.06</del>	<del>-6.57</del>
AZ	<del>4.83</del>	<del>-3.01</del>	<del>-0.76</del>

 $A^2$ 

- 10 Overall, we propose that although the winter wind stress linked mechanisms explain the decadal trends in the strengthening and weakening of CO<sub>2</sub> uptake by the Southern Ocean, summer drivers may explain the inter-annual variability in the decadal trends (Lovenduski et al., 2008; Landschützer et al., 2015).
- Lastly the ensemble of machine learning methods shows that there is still considerable disagreement between the different approaches. This is likely driven by the lack of  $pCO_2$  measurements in the Southern 15 Ocean as found by Rödenbeck et al. (2015). Autonomous sampling platforms will likely play a role in closing this "observation gap", but strategic deployment and sampling strategies will be critical to constrain and improve our understanding of CO<sub>2</sub> in the non-stationary context (McNeil and Matear, 2013; Monteiro et al., 2015).

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#### **S Supplementary Materials**

#### **<u>S1</u>** Wind speed and regional surface area

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The regional magnitude of integrated air-sea CO<sub>2</sub> fluxes areis in part determined by the wind speed and surface area of the specific region. Figure 11aS1a shows the average wind speeds for summer and winter for each of the regions as defined in Figure 1. The wind product used is CCMP v2 (Atlas et al. 2011). Figure 11bS1b shows the surface area of each of the regions. Note that the Indian sector of the PFZ has both the highest average wind speed and has the largest surface area. This explains the dominance of the region in the determination of interannual trendsvariability of  $FCO_{25}$  (Figure S2), even though  $\Delta pCO_2$  trends are(Figure 4) variability is relatively weak.



Figure A1<u>S1</u>: (a) Average wind speeds for each of the <u>bimoesbiomes</u> for summer (dark) and winter (light). The ocean basins are shown by the <u>colorscolours</u> as shown in the key for (b). (b) shows the size of each region <u>seperated separated</u> by biome and basin.

#### A3 Additional transition anomaly figures

Figure A2 and A3 augment Figures 7 and 8 respectively. These were omitted from the main text figures as we found that these variables (Chl-*a* in winter and MLD in summer) do little to aid our understanding of changes in  $\Delta p$ CO<sub>2</sub>. These are included for the sake of completeness.



Figure A2: Winter Chl-a transitions for each of the three anomaly periods (relating to Figure 7).



Figure A3: Summer MLD transitions

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Figure S2: The regional breakdown of the seasonal averages for  $\Delta pCO_2$ . The seasonal mean for summer (solid) and winter (dashed) for each method is represented by the different coloured lines as shown in the key, where MLS is the Mixed Layer Scheme. The other methods are as in the main text. The grey fill is the ensemble mean  $\Delta pCO_2 \pm E_b$  where  $E_b$  is the between-method error calculated as in Eq (5).

Figure S2 shows the seasonal time series for each of the three anomaly periods (relating to Figure 8).region maintaining separate seasonal averages for each method. We also include the Marginal Ice Zone plots with all plots showing the average between-method error.

The Mixed Layer Scheme (MLS) method by Rödenbeck et al. (2013) is also included. Note that the scale of MLDMLS is not a machine learning method as it incorporates prior knowledge of the system. The method results in divergent estimates of ΔpCO<sub>2</sub>, particularly in the SAZ. The MLS fails to produce a seasonal cycle with winter and summer ΔpCO<sub>2</sub> having the same magnitude. Further work will have to be done to understand the cause for this difference. We do not include MLS in the main ensemble as we cannot explain this difference. The methods are in much better agreement in the PFZ and MIZ.

#### S3 Air-Sea CO<sub>2</sub> Fluxes

Air sea CO<sub>2</sub> fluxes are calculated with:

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$$FCO_2 = k_w \cdot K_0 \cdot (pCO_2^{\text{sea}} - pCO_2^{\text{atm}})$$
(S1)

The gas transfer velocity  $(k_w)$  is calculated using a quadratic dependency of wind speed with the coefficients of (Wanninkhof et al., 2009). Coefficients from Weiss (1974) are used to calculate  $K_{\ell}$  and  $\Delta pCO_2$  is estimated by the empirical models. figureWind speed is calculated from the *u* and *v* vectors  $(\sqrt{u^2 + v^2})$  of the Cross-Calibrated Multiplatform Product (CCMP) v2 (Atlas et al., 2011; Wentz et al., 2015). Wind speed is one of the largest contributors to the uncertainty in flux estimates, thus the choice of the wind product could have a large impact on flux estimates as well as interpretation of the drivers of  $CO_2$  (Takahashi et al., 2009). We use the ensemble mean  $\Delta pCO_2$  from Figure 4 to calculate fluxes - note that this does not matchinclude the scale of MLDMLS shown in Figure S2.



Figure S3: FCO<sub>2</sub> (dark grey) plotted by biome (rows) and basin (columns). Biomes are defined by Fay and McKinley (2014a). The solid red line shows the maximum for each year (winter outgassing) and the dashed line shows the same line less the average difference between the minimum and maximum – this is the expected amplitude. Lighter grey shading in (a-i) shows periods used in Figure 7.-5 and 6. Note that fluxes in the MIZ are calculated from a reduced surface area to maintain consistency between methods.

- Mean FCO<sub>2</sub> is shown in Figure S3. Note that the apparent weak fluxes in the MIZ are due to the reduction of the surface area and thus flux to maintain equal weighting between machine learning methods. The SAZ clearly dominates the annual uptake of CO<sub>2</sub> in the Southern Ocean, but the interannual variability is dominated by the PFZ. An interesting point of the SAZ is that the seasonal cycle of wind speed (strong in winter, weak in summer) opposes that of ΔpCO<sub>2</sub> sink (weak in winter, strong in summer). The net result is that, compared to ΔpCO<sub>2</sub>, the seasonal amplitude of FCO<sub>2</sub> is reduced. The same effect shifts the
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mean flux in the PFZ, but does not affect the amplitude, where outgassing is amplified in winter and the sink is weaker than if wind speed was constant. Lastly, Figures S3a,d show that the Indian sector of the Southern Ocean dominate both uptake (SAZ) and the interannual variability (PFZ).

#### **S4 Uncertainty of the transition anomalies**

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90 The transition anomalies are not calculated from the mean of the three methods. Rather we calculate the anomalies for each individual method with:

$$a_{n(p')} = \overline{s_{n(p)}} - \overline{s_{n(p-1)}} \tag{S2}$$

where *s* are the estimates for a particular model, *n* represents an individual model and *p* represents P1 to P4. The result,  $a_{n(p')}$  thus represents the anomaly for two periods for a particular model. We then calculate the average of the anomalies with:

$$a_{p'} = \frac{1}{N} \cdot \sum_{n=1}^{N} a_{n(p')}$$
(S3)

where *N* is 3, the number of models. We then calculate the standard deviation of the three anomalies  $(e_{p'})$ , which is analogous to the between-model error, with:

$$e_{p'} = \sqrt{\frac{1}{N} \cdot \sum_{n=1}^{N} (a_{n(p')} - \overline{a_{n(p')}})^2}$$
(S4)

<sup>100</sup> where the terms are consistent with those above. We use  $e_{p'}$  as an uncertainty threshold where anomalies are only considered significant if  $|a_{p'}| > e_{p'}$ . These regions are masked in Figures 5a-c and <u>6a-c</u>. Figure S4 shows the winter (a-c) and summer (d-f)  $e_{p'}$  for each transition anomaly.



<sup>105</sup> Figure S4: Maps of the standard deviation between empirical methods for the anomalies. These are used as thresholds for  $\Delta pCO_2$  in Figures 5(a-c) and 6(a-c) for winter and summer respectively. When the standard deviation exceeds the absolute value average anomaly, the values are masked as shown in Figures 5 and 6.

#### **S5 Additional driver variables**

Here we show additional variables that accompany Figures 5 and 6. Figure S5 shows winter Chl-*a*, uand v-components of wind and Figure S6 shows summer MLD, u- and v-components of wind. These variables were not included in the main analyses as they did not contribute significant information to the

'15 proxy variables already present (wind stress, SST and MLD/Chl-a). It is interesting to note that the uand v- components of wind speed highlight the zonally asymmetric dipole during winter (Figures S5d,e,g,h) and the annular dipole during summer (Figures S6d,e).



<sup>1</sup>20 Figure S5: Relative anomalies of winter chlorophyll-a (a-c), u- (d-f), and v-components (g-i) of wind for four periods (as shown above each column). The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and McKinley, 2014a) and basin boundaries.



Figure S6: Relative anomalies of summer mixed layer depth (a-c), u- (d-f), and v-components (g-i) of wind for four periods (as shown above each column).. The thin black lines show the boundaries for each of the nine regions described by the biomes (Fay and McKinley, 2014a) and basin boundaries.

#### References

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- Abernathey, R., Marshall, J. C., & Ferreira, D. (2011). The Dependence of Southern Ocean Meridional Overturning on Wind Stress. *Journal of Physical Oceanography*, 41(12), 2261–2278. https://doi.org/10.1175/JPO-D-11-023.1
- Arrigo, K. R., van Dijken, G. L., & Bushinsky, S. (2008). Primary production in the Southern Ocean, 1997-2006. Journal of Geophysical Research, 113(8), 1997–2006. https://doi.org/10.1029/2007JC004551
- Atlas, R., Hoffman, R. N., Ardizzone, J., Leidner, S. M., Jusem, J. C., Smith, D. K., & Gombos, D. (2011). A Crosscalibrated, Multiplatform Ocean Surface Wind Velocity Product for Meteorological and Oceanographic Applications. *Bulletin of the American Meteorological Society*, 92(2), 157–174. https://doi.org/10.1175/2010BAMS2946.1
- Bakker, D. C. E., Hoppema, M., Schr, M., Geibert, W., & Baar, H. J. W. De. (2008). A rapid transition from ice covered CO2-rich waters to a biologically mediated CO2 sink in the eastern Weddell Gyre. *Biogeosciences*, *5*, 1373–1386.
- Bakker, D. C. E., Pfeil, B., Landa, C. S., Metzl, N., O'Brien, K. M., Olsen, A., ... Xu, S. (2016). A multi-decade record of high-quality fCO2 data in version 3 of the Surface Ocean CO2 Atlas (SOCAT). *Earth System Science Data*, 8(2), 383– 413. https://doi.org/10.5194/essd-8-383-2016
- Bakker, D. C. E., Pfeil, B., Smith, K., Hankin, S., Olsen, A., Alin, S. R., ... Watson, A. J. (2014). An update to the surface ocean CO2 atlas (SOCAT version 2). *Earth System Science Data*, 6(1), 69–90. https://doi.org/10.5194/essd-6-69-2014
  - Barnes, E. A., & Hartmann, D. L. (2010). Dynamical Feedbacks of the Southern Annular Mode in Winter and Summer. *Journal of the Atmospheric Sciences*, 67(2007), 2320–2330. https://doi.org/10.1175/2010JAS3385.1
- Boyd, P. W., & Ellwood, M. J. (2010). The biogeochemical cycle of iron in the ocean. *Nature Geoscience*, *3*(10), 675–682. https://doi.org/10.1038/ngeo964
  - Butterworth, B. J., & Miller, S. D. (2016). Air-sea exchange of carbon dioxide in the Southern Ocean and Antarctic marginal ice zone. *Geophysical Research Letters*, 43(13), 7223–7230. https://doi.org/10.1002/2016GL069581
  - Carranza, M. M., & Gille, S. T. (2015). Southern Ocean wind-driven entrainment enhances satellite chlorophyll-a through the summer. *Journal of Geophysical Research: Oceans*, *120*(1), 304–323. https://doi.org/10.1002/2014JC010203
- <sup>50</sup> CDIAC. (2017). *Multi-laboratory compilation of atmospheric carbon dioxide data for the period 1957-2016*. NOAA Earth System Research Laboratory, Global Monitoring Division. https://doi.org/10.15138/G3CW4Q
  - Chierici, M., Signorini, S. R., Mattsdotter-Björk, M., Fransson, A., & Olsen, A. (2012). Surface water fCO 2 algorithms for the high-latitude Pacific sector of the Southern Ocean. *Remote Sensing of Environment*, 119, 184–196. https://doi.org/10.1016/j.rse.2011.12.020
- '55 <u>de Boyer Montégut, C., Madec, G., Fischer, A. S., Lazar, A., & Iudicone, D. (2004). Mixed layer depth over the global ocean: An examination of profile data and a profile-based climatology. *Journal of Geophysical Research*, 109(12), 1–20. https://doi.org/10.1029/2004JC002378</u>
  - DeVries, T., Holzer, M., & Primeau, F. (2017). Recent increase in oceanic carbon uptake driven by weaker upper-ocean overturning. *Nature*, *542*(7640), 215–218. https://doi.org/10.1038/nature21068
- '60 Dufour, C. O., Sommer, J. Le, Gehlen, M., Orr, J. C., Molines, J.-M., Simeon, J., & Barnier, B. (2013). Eddy compensation and controls of the enhanced sea-to-air CO2 flux during positive phases of the Southern Annular Mode. *Global Biogeochemical Cycles*, 27(3), 950–961. https://doi.org/10.1002/gbc.20090
  - Fay, A. R., & McKinley, G. A. (2014). Global open-ocean biomes: Mean and temporal variability. *Earth System Science Data*, 6(2), 273–284. https://doi.org/10.5194/essd-6-273-2014
- '65 Fay, A. R., & McKinley, G. A. (2017). Correlations of surface ocean pCO2 to satellite chlorophyll on monthly to interannual timescales. *Global Biogeochemical Cycles*, 31(3), 1–20. https://doi.org/10.1002/2016GB005563
  - Fay, A. R., McKinley, G. A., & Lovenduski, N. S. (2014). Southern Ocean carbon trends: Sensitivity to methods. Geophysical Research Letters, 41(19), 6833–6840. https://doi.org/10.1002/2014GL061324
- Fogt, R. L., Jones, J. M., & Renwick, J. (2012). Seasonal zonal asymmetries in the southern annular mode and their impact on regional temperature anomalies. *Journal of Climate*, 25(18), 6253–6270. https://doi.org/10.1175/JCLI-D-11-00474.1
  - Gade, K. (2010). A Non-singular Horizontal Position Representation. *Journal of Navigation*, 63(3), 395–417. https://doi.org/10.1017/S0373463309990415
- Gregor, L., Kok, S., & Monteiro, P. M. S. (2017). Empirical methods for the estimation of Southern Ocean CO2: support vector and random forest regression. *Biogeosciences*, 14(23), 5551–5569. https://doi.org/10.5194/bg-14-5551-2017
  - Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Pak, B. C., Baker, D., ... Taguchi, S. (2004). Transcom 3 inversion intercomparison: Model mean results for the estimation of seasonal carbon sources and sinks. *Global Biogeochemical Cycles*, 18(1), n/a-n/a. https://doi.org/10.1029/2003GB002111
- Hauck, J., Völker, C., Wang, T., Hoppema, M., Losch, M., & Wolf-Gladrow, D. A. (2013). Seasonally different carbon flux changes in the Southern Ocean in response to the southern annular mode. *Global Biogeochemical Cycles*, 27(4), 1236–1245. https://doi.org/10.1002/2013GB004600
  - Hoppema, M., Fahrbach, E., Stoll, M. H. C., & de Baar, H. J. W. (1999). Annual uptake of atmospheric CO2 by the Weddell sea derived from a surface layer balance, including estimations of entrainment and new production. *Journal of Marine Systems*, *19*(4), 219–233. https://doi.org/10.1016/S0924-7963(98)00091-8

- '85 Ishii, M., Inoue, H. Y., Matsueda, H., & Tanoue, E. (1998). Close coupling between seasonal biological production and dynamics of dissolved inorganic carbon in the Indian Ocean sector and the western Pacific Ocean sector of the Antarctic Ocean. *Deep-Sea Research Part I: Oceanographic Research Papers*, 45(7), 1187–1209. https://doi.org/10.1016/S0967-0637(98)00010-7
- Jones, S. D., Le Quéré, C., & Rdenbeck, C. (2012). Autocorrelation characteristics of surface ocean pCO2 and air-sea CO2 fluxes. *Global Biogeochemical Cycles*, *26*(2), 1–12. https://doi.org/10.1029/2010GB004017
- Kanamitsu, M., Ebisuzaki, W., Woollen, J., Yang, S. K., Hnilo, J. J., Fiorino, M., & Potter, G. L. (2002). NCEP-DOE AMIP-II reanalysis (R-2). Bulletin of the American Meteorological Society, 83(11), 1631–1643+1559. https://doi.org/10.1175/BAMS-83-11-1631
- Khatiwala, S., Tanhua, T., Mikaloff Fletcher, S. E., Gerber, M., Doney, S. C., Graven, H. D., ... Sabine, C. L. (2013). Global ocean storage of anthropogenic carbon. *Biogeosciences*, 10(4), 2169–2191. https://doi.org/10.5194/bg-10-2169-2013
- Landschützer, P., Gruber, N., & Bakker, D. C. E. (2016). Decadal variations and trends of the global ocean carbon sink. *Global Biogeochemical Cycles*, 30(10), 1396–1417. https://doi.org/10.1002/2015GB005359

00

45

- Landschützer, P., Gruber, N., & Bakker, D. C. E. (2017). An updated observation-based global monthly gridded sea surface pCO2 and air-sea CO2 flux product from 1982 through 2015 and its monthly climatology (NCEI Accession 0160558) Version 2.2.
- Landschützer, P., Gruber, N., Bakker, D. C. E., & Schuster, U. (2014). Recent variability of the global ocean carbon sink. *Global and Planetary Change*, 927–949. https://doi.org/10.1002/2014GB004853.Received
- Landschützer, P., Gruber, N., Haumann, F. A., Rödenbeck, C., Bakker, D. C. E., Van Heuven, S. M. A. C., ... Wanninkhof, R. H. (2015). The reinvigoration of the Southern Ocean carbon sink. *Science*, *349*(6253), 1221–1224.
   https://doi.org/10.1126/science.aab2620
- Le Quéré, C., Rödenbeck, C., Buitenhuis, E. T., Conway, T. J., Langenfelds, R., Gomez, A., ... Heimann, M. (2007). Saturation of the Southern Ocean CO2 Sink Due to Recent Climate Change. *Science*, *316*(5832), 1735–1738. https://doi.org/10.1126/science.1137004
- Lenton, A., Codron, F., Bopp, L., Metzl, N., Cadule, P., Tagliabue, A., & Le Sommer, J. (2009). Stratospheric ozone depletion reduces ocean carbon uptake and enhances ocean acidification. *Geophysical Research Letters*, 36(12), 1–5. https://doi.org/10.1029/2009GL038227
  - Lenton, A., & Matear, R. J. (2007). Role of the Southern Annular Mode (SAM) in Southern Ocean CO2 uptake. *Global Biogeochemical Cycles*, *21*(2), 1–17. https://doi.org/10.1029/2006GB002714
- Lenton, A., Metzl, N., Takahashi, T. T., Kuchinke, M., Matear, R. J., Roy, T., ... Tilbrook, B. (2012). The observed evolution of oceanic pCO2 and its drivers over the last two decades. *Global Biogeochemical Cycles*, 26(2), 1–14. https://doi.org/10.1029/2011GB004095
  - Lenton, A., Tilbrook, B., Law, R. M., Bakker, D. C. E., Doney, S. C., Gruber, N., ... Takahashi, T. T. (2013). Sea-air CO2 fluxes in the Southern Ocean for the period 1990-2009. *Biogeosciences*, 10(6), 4037–4054. https://doi.org/10.5194/bg-10-4037-2013
- 120 Lévy, M., Iovino, D., Resplandy, L., Klein, P., Madec, G., Tréguier, A. M., ... Takahashi, K. (2012). Large-scale impacts of submesoscale dynamics on phytoplankton: Local and remote effects. *Ocean Modelling*, 43–44, 77–93. https://doi.org/10.1016/j.ocemod.2011.12.003
  - Lovenduski, N. S., Gruber, N., & Doney, S. C. (2008). Toward a mechanistic understanding of the decadal trends in the Southern Ocean carbon sink. *Global Biogeochemical Cycles*, *22*(3), 9. https://doi.org/10.1029/2007GB003139
- 125 Lovenduski, N. S., Gruber, N., Doney, S. C., & Lima, I. D. (2007). Enhanced CO2 outgassing in the Southern Ocean from a positive phase of the Southern Annular Mode. *Global Biogeochemical Cycles*, 21(2), 1–14. https://doi.org/10.1029/2006GB002900
  - Mahadevan, aA., D'Asaro, E., Lee, C., & Perry, M. J. (2012). Eddy-Driven Stratification Initiates North Atlantic Spring Phytoplankton Blooms. *Science*, *337*(6090), 54–58. https://doi.org/10.1126/science.1218740
- 130 Mahadevan, A., Tagliabue, A., Bopp, L., Lenton, A., Mémery, L., & Lévy, M. (2011). Impact of episodic vertical fluxes on sea surface pCO2. *Philosophical Transactions of the Royal Society*, 369(1943), 2009–2025. https://doi.org/10.1098/rsta.2010.0340
  - Maritorena, S., & Siegel, D. A. (2005). Consistent merging of satellite ocean color data sets using a bio-optical model. *Remote Sensing of Environment*, 94(4), 429–440. https://doi.org/10.1016/j.rse.2004.08.014
- Marshall, G. J. (2003). Trends in the Southern Annular Mode from observations and reanalyses. *Journal of Climate*, 16(24), 4134–4143. https://doi.org/10.1175/1520-0442(2003)016<4134:TITSAM>2.0.CO;2
  - Marshall, J. C., & Speer, K. (2012). Closure of the meridional overturning circulation through Southern Ocean upwelling. *Nature Geoscience*, 5(3), 171–180. https://doi.org/10.1038/ngeo1391
- McGillicuddy, D. J. (2016). Mechanisms of Physical-Biological-Biogeochemical Interaction at the Oceanic Mesoscale.
   Annual Review of Marine Science (Vol. 8). https://doi.org/10.1146/annurev-marine-010814-015606
- McNeil, B. I., & Matear, R. J. (2013). The non-steady state oceanic CO2 signal: Its importance, magnitude and a novel way to detect it. *Biogeosciences*, 10(4), 2219–2228. https://doi.org/10.5194/bg-10-2219-2013
  - Menemenlis, D., Campin, J., Heimbach, P., Hill, C. N., Lee, T., Nguyen, A., ... J-M. Campin. (2008). ECCO2 : High Resolution Global Ocean and Sea Ice Data Synthesis. *Mercator Ocean Quarterly Newsletter*, 31(October), 13–21.
  - Retrieved from http://www.mercator-ocean.fr/content/download/691/5904/version/1/file/lettre\_31\_en.pdf#page=13

- Meredith, M. P. (2016). Understanding the structure of changes in the Southern Ocean eddy field. *Geophysical Research Letters*, 43(11), 5829–5832. https://doi.org/10.1002/2014JC010066.Received
- Meredith, M. P., & Hogg, A. M. (2006). Circumpolar response of Southern Ocean eddy activity to a change in the Southern Annular Mode. *Geophysical Research Letters*, 33(16), 2–5. https://doi.org/10.1029/2006GL026499
- Meredith, M. P., Naveira Garabato, A. C., Hogg, A. M., & Farneti, R. (2012). Sensitivity of the overturning circulation in the Southern Ocean to decadal changes in wind foreing. *Journal of Climate*, 25(1), 99–110. https://doi.org/10.1175/2011JCLI4204.1
- Metzl, N., Brunet, C., Jabaud-Jan, A., Poisson, A., & Schauer, B. (2006). Summer and winter air-sea CO2 fluxes in the
   Southern Ocean. *Deep-Sea Research Part I: Oceanographic Research Papers*, 53(9), 1548–1563. https://doi.org/10.1016/j.dsr.2006.07.006
  - Mongwe, N. P., Chang, N., & Monteiro, P. M. S. (2016). The seasonal cycle as a mode to diagnose biases in modelled CO 2 fluxes in the Southern Ocean. *Ocean Modelling*, *106*, 90–103. https://doi.org/10.1016/j.ocemod.2016.09.006
- Monteiro, P. M. S., Gregor, L., Lévy, M., Maenner, S., Sabine, C. L., & Swart, S. (2015). Intraseasonal variability linked to sampling alias in air-sea CO2 fluxes in the Southern Ocean. *Geophysical Research Letters*, 42(20), 8507–8514. https://doi.org/10.1002/2015GL066009
  - Munro, D. R., Lovenduski, N. S., Takahashi, T. T., Stephens, B. B., Newberger, T., & Sweeney, C. (2015). Recent evidence for a strengthening CO2 sink in the Southern Ocean from carbonate system measurements in the Drake Passage (2002-2015). *Geophysical Research Letters*, 42(18), 7623–7630. https://doi.org/10.1002/2015GL065194
- 165 Nicholson, S. A., Lévy, M., Llort, J., Swart, S., & Monteiro, P. M. S. (2016). Investigation into the impact of storms on sustaining summer primary productivity in the Sub-Antarctic Ocean. *Geophysical Research Letters*, 43(17), 9192– 9199. https://doi.org/10.1002/2016GL069973
  - <u>Pfeil, B., Olsen, A., Bakker, D. C. E., Hankin, S., Koyuk, H., Kozyr, A., ... Yoshikawa-Inoue, H. (2013). A uniform, quality controlled Surface Ocean CO2 Atlas (SOCAT). *Earth System Science Data*, 5(1), 125–143. https://doi.org/10.5194/essd-5-125-2013</u>
- Reynolds, R. W., Smith, T. M., Liu, C., Chelton, D. B., Casey, K. S., & Schlax, M. G. (2007). Daily high-resolution-blended analyses for sea surface temperature. *Journal of Climate*, 20(22), 5473–5496. https://doi.org/10.1175/2007JCLI1824.1
- Rödenbeck, C., Bakker, D. C. E., Gruber, N., Iida, Y., Jacobson, A. R., Jones, S. D., ... Zeng, J. (2015). Data-based estimates of the ocean carbon sink variability First results of the Surface Ocean pCO2 Mapping intercomparison (SOCOM). *Biogeosciences*, 12(23), 7251–7278. https://doi.org/10.5194/bg-12-7251-2015
- <u>Rödenbeck, C., Keeling, R. F., Bakker, D. C. E., Metzl, N., Olsen, A., Sabine, C. L., & Heimann, M. (2013). Global surfaceocean pCO2 and sea-Air CO2 flux variability from an observation-driven ocean mixed-layer scheme. Ocean Science, 9(2), 193–216. https://doi.org/10.5194/os-9-193-2013</u>
- Rodgers, K. B., Aumont, O., Mikaloff Fletcher, S. E., Plancherel, Y., Bopp, L., de Boyer Montégut, C., ... Wanninkhof, R. H. (2014). Strong sensitivity of Southern Ocean carbon uptake and nutrient cycling to wind stirring. *Biogeosciences*, *11*(15), 4077–4098. https://doi.org/10.5194/bg-11-4077-2014
  - Sabine, C. L., Hankin, S., Koyuk, H., Bakker, D. C. E., Pfeil, B., Olsen, A., ... Yoshikawa-Inoue, H. (2013). Surface Ocean CO2 Atlas (SOCAT) gridded data products. *Earth System Science Data*, 5(1), 145–153. https://doi.org/10.5194/essd-5-145-2013
- 185 Sasse, T. P., McNeil, B. I., & Abramowitz, G. (2013). A novel method for diagnosing seasonal to inter-annual surface ocean carbon dynamics from bottle data using neural networks. *Biogeosciences*, 10(6), 4319–4340. https://doi.org/10.5194/bg-10-4319-2013
  - Shetye, S. S., Mohan, R., Patil, S., Jena, B., Chacko, R., George, J. V., ... Sudhakar, M. (2015). Oceanic pCO2 in the Indian sector of the Southern Ocean during the austral summer–winter transition phase. *Deep Sea Research Part II: Topical Studies in Oceanography*, *118*, 250–260. https://doi.org/10.1016/j.dsr2.2015.05.017
- Studies in Oceanography, 118, 250–260. https://doi.org/10.1016/j.dsr2.2015.05.017
   Swart, N. C., & Fyfe, J. C. (2012). Observed and simulated changes in the Southern Hemisphere surface westerly windstress. Geophysical Research Letters, 39(16), 6–11. https://doi.org/10.1029/2012GL052810
  - Swart, N. C., Fyfe, J. C., Gillett, N., & Marshall, G. J. (2015). Comparing trends in the southern annular mode and surface westerly jet. *Journal of Climate*, 28(22), 8840–8859. https://doi.org/10.1175/JCLI-D-15-0334.1
- 195 Swart, S., Speich, S., Ansorge, I. J., & Lutjeharms, J. R. E. (2010). An altimetry-based gravest empirical mode south of Africa: 1. Development and validation. *Journal of Geophysical Research*, 115(C3), 1–19. https://doi.org/10.1029/2009JC005299
- Swart, S., Thomalla, S. J., & Monteiro, P. M. S. (2015). The seasonal cycle of mixed layer dynamics and phytoplankton<br/>biomass in the Sub-Antarctic Zone: A high-resolution glider experiment. Journal of Marine Systems, 147, 103–115.00https://doi.org/10.1016/j.jmarsys.2014.06.002
- Tagliabue, A., Bowie, A. R., Philip, W., Buck, K. N., Johnson, K. S., & Saito, M. A. (2017). The integral role of iron in ocean biogeochemistry. *Nature*, 543(7643), In Press. https://doi.org/10.1038/nature21058
  - Tagliabue, A., Sallée, J.-B., Bowie, A. R., Lévy, M., Swart, S., & Boyd, P. W. (2014). Surface-water iron supplies in the Southern Ocean sustained by deep winter mixing. *Nature Geoscience*, 7(March), 314–320.
- 05 https://doi.org/10.1038/NGEO2101

50

 $\overline{70}$ 

- Takahashi, T. T., Olafsson, J., Goddard, J. G., Chipman, D. W., & Sutherland, S. C. (1993). Seasonal variation of CO 2 and nutrients in the high-latitude surface oceans: A comparative study. *Global Biogeochemical Cycles*, 7(4), 843–878. https://doi.org/10.1029/93GB02263
- Takahashi, T. T., Sutherland, S. C., Sweeney, C., Poisson, A., Metzl, N., Tilbrook, B., ... Nojiri, Y. (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
  - Takahashi, T. T., Sutherland, S. C., Wanninkhof, R. H., Sweeney, C., Feely, R. A., Chipman, D. W., ... de Baar, H. J. W. (2009). Climatological mean and decadal change in surface ocean pCO2, and net sea–air CO2 flux over the global oceans. *Deep-Sea Research Part II: Topical Studies in Oceanography*, 56(8–10), 554–577.

https://doi.org/10.1016/j.dsr2.2008.12.009

15

- Thomalla, S. J., Fauchereau, N., Swart, S., & Monteiro, P. M. S. (2011). Regional scale characteristics of the seasonal cycle of chlorophyll in the Southern Ocean. *Biogeosciences*, 8(10), 2849–2866. https://doi.org/10.5194/bg-8-2849-2011
  Wang, S., & Moore, J. K. (2012). Variability of primary production and air-sea CO2 flux in the Southern Ocean. *Global*
- 20 Wang, S., & Moore, J. K. (2012). Variability of primary production and air-sea CO2 flux in the Southern Ocean. *Gl* Biogeochemical Cycles, 26(1), 1–12. https://doi.org/10.1029/2010GB003981
  - Wanninkhof, R. H., Asher, W. E., Ho, D. T., Sweeney, C., & McGillis, W. R. (2009). Advances in Quantifying Air-Sea Gas Exchange and Environmental Forcing\*. *Annual Review of Marine Science*, 1(1), 213–244. https://doi.org/10.1146/annurev.marine.010908.163742
- Weiss, R. (1974). Carbon dioxide in water and seawater: the solubility of a non-ideal gas. *Marine Chemistry*, 2(3), 203–215.
   https://doi.org/10.1016/0304-4203(74)90015-2
  - Wentz, F. J., Scott, J., Hoffman, R., Leidner, M., Atlas, R., & Ardizzone, J. (2015). Remote Sensing Systems Cross-Calibrated Multi-Platform (CCMP) 6-hourly ocean vector wind analysis product on 0.25 deg grid, Version 2.0. Retrieved March 5, 2017, from www.remss.com/measurements/ccmp
- Whitt, D. B., Lévy, M., & Taylor, J. R. (2017). Low-frequency and high-frequency oscillatory winds synergistically enhance nutrient entrainment and phytoplankton at fronts. *Journal of Geophysical Research*, *122*(2), 1016–1041. https://doi.org/10.1002/2016JC012400

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