1 Review 2 (RC2)

2 We thank the reviewer for the detailed review of our manuscript. The review was indeed

- 3 helpful in strengthening the manuscript. We hope we have addressed all comments
- 4 satisfactorily.

All grammar corrections where done as suggested with minor changes where necessary. Allare detailed in an annotated version of the document.

7 General comments

8 Reviewer: I think the methodology could be better explained. Perhaps the logic of equations 9 1-3 is explained more thoroughly in the previous paper by Mongwe et al 2016, but this 10 critical reference is missing from the reference list. Assuming they mean the paper in Ocean 11 Modelling 106: 90, I will agree that equations 1 and 2 can be derived from equations 3.2-3.4 12 in that paper. But the LHS of equation 2, which does not appear in the previous work, is 13 physically speaking, a fairly nebulous quantity. The Takahashi et al 1993 estimate of 0.0423 14 K⁻¹ mainly expresses the change in partial pressure due to changing temperature for a 15 given concentration of CO_2 ([CO_2^*]), with a small contribution from the partitioning of DIC among $CO_2/HCO^{3-}/CO^{3-}$ due to the temperature dependence of the equilibrium constants. 16 17 DIC does not change as a result of changes in temperature, except indirectly through gas 18 exchange.

19 So what we have here is an observed change of pCO_2 with changing temperature,

20 convoluted into a change of DIC by application of several highly empirical conversion fac-

21 tors (more about this below), as an estimate of the changes in DIC not attributable to

22 biological uptake/remineralization (and therefore primarily attributable to gas exchange).

23 This in itself might be inoffensive, but I would prefer if its relationship to actual physical

24 processes were better explained. The equations that are taken as a starting point are highly

25 empirical, and we should not invest rearrangements of these with an outsized significance.

0.0423 K⁻¹ is intended to be an average value for a broad range of ocean conditions, but it is
stated to be valid for salinities from 34-36 and temperatures from 2-28°C (Takahashi et al
1993). In the Southern Ocean one will encounter conditions outside, or on the far edges of,
these ranges. What are the implications of this for the analysis shown here? This seems like
something that could be evaluated. Similarly, the calculations assume a constant Revelle
factor, but it should be quite straightforward to calculate Revelle factors from the model

outputs, giving a range for the range of environmental conditions characteristic of the study
 area. The conclusions are probably robust to these assumption, I see no reason why not
 tested.

35

36 **Response:** The reviewer points out an overarching request to clarify our methodology, 37 which indicates that it was not clearly presented. This is partly because the link to our 38 previous paper (Mongwe et al., 2016), where we explained this methodology, was poorly 39 made. A more detailed synthesis from Mongwe et al., (2016) was necessary as suggested. 40 This was compounded by some inadvertently missing references, including the Mongwe et 41 al., 2016. Having used referencing software, I neglected a thorough check of the 42 bibliography, which was a mistake and I apologize for this mistake; all references in the 43 revised manuscript have been corrected.

44 This reviewer's comment on the methodology is partly connected to the next comment 45 asking how we ascribed the influence of temperature to DIC and the partitioning of the 46 other DIC terms. We have deferred this part of the response to the next comment, in which 47 this question is raised in a more specific way. In this first part we focus on the question 48 about the uncertainties linked to the constants of the Takahashi et al., (1993) empirical 49 expression as well as the Revelle factor in polar waters. We thank the reviewer for raising 50 the question on limits to the validity of the empirical relationship used to calculate 51 temperature-driven changes to pCO₂ observations and model data.

52 We examined the applicability of the Takahashi et al., (1993) linear approximation $\left(\frac{1}{pCO_2}\frac{\partial pCO_2}{\partial SST} \approx 0.0423^o C^{-1}\right)$ in our region of study. In the Sub-Antarctic zone, surface water 53 has a temperature range of ${}^{\sim}4{}^{\circ}C - 12{}^{\circ}C$, which is within the limits (2 – 28 ${}^{\circ}C$) provided by 54 55 Takahashi et al (1993). Since surface temperatures go below 2°C in the Antarctic zone, we 56 have tested whether this relationship can be extrapolated down to -2° C. We do so by 57 comparing the dependence of pCO_2 on temperature for a range of temperature values (4°C 58 to -2° : 0.1°C intervals) using a carbonate equilibrium model and the Takahashi et al., (1993) 59 linearization. Ancillary variables DIC, TAlk, phosphate, silicate and salinity are fixed and the 60 carbonate equilibrium model CO2SYS (Pierrot et al., 2006) was used with K1, K2 from 61 Mehrbach et al., (1973) refitted by Dickson and Millero, (1987), which are the same 62 parameterizations used in the majority of CMIP5 models. We used the mean climatological 63 pCO₂ (Landschützer et al., 2014) and the seasonal mean for nutrients (silicate and

phosphate), salinity, TAlk and DIC from the same Antarctic region in GLODAP2 as was used inthe models.

- As shown in Fig. 1, the response of pCO_2 to temperature below 2°C can still be described
- 67 with the Takahashi et al., (1993) linear relationship. Thus we can extrapolate the Takahashi
- 68 et al., (1993) linear dependence of pCO₂ to temperature for the estimation of temperature
- 69 solubility changes to equivalent DIC changes in the Antarctic zone as explained above.
- 70





78

In a comparable way, we examined the uncertainties arising from a fixed polar Revelle factor in our calculations. We recomputed the Revelle factor in the Sub-Antarctic and Antarctic zones using annual mean climatologies of TAlk, salinity, surface temperature nutrients. Firstly we examined DIC changes for the nominal range of pCO₂ change (340 – 399 µatm, 1 µatm intervals) and then used this dataset to derive the Revelle factor $\left(\gamma_{DIC} = \frac{DIC}{pCO_2} \frac{\partial pCO_2}{\partial DIC}\right)$. The calculated Revelle factors in the Southern Ocean range between $\gamma_{DIC} \sim 12 - 15.5$ with an average of $\gamma_{DIC} = 13.9\pm 1.3$. This justifies our use of $\gamma_{DIC} = 14$ for the conversion of the 86 equivalent solubility driven pCO₂ change to DIC throughout the analysis. As an addition, we 87 now provide the uncertainty in this conversion as it translates into the temperature 88 constraint, by using the upper and lower limits of the Revelle factor ($\gamma_{DIC} = 12 - 15.5$) in the 89 model framework. Below we show an example for observations in the Sub-Antarctic and 90 Antarctic zone, this shows that extremes of the Revelle factor values ($\gamma_{DIC} = 12 - 15.5$) do not 91 alter the phasing or magnitude of the relative controls of temperature or DIC on the 92 seasonal cycle of pCO₂ (Fig 2).

93



Figure. 2 Seasonal cycle of the rate change of surface total DIC for the Landschützer et al (2014) data product (black line) for the Landschützer et al (2014) data product and the estimated temperature driven DIC rate of change $\left(\frac{\partial DIC}{\partial t}\right)_{SST}$ shaded area, for monthly data given in µmol kg⁻¹ month⁻¹ at the Sub-Antarctic zone i.e. Pacific Ocean (first column), Atlantic Ocean (second column) and Indian Ocean (third column). The dotted line shows the uncertainty boundaries for the Revelle factor extremes accounting to range in the Southern Ocean ($\gamma_{DIC} =$ 12 – 15.5).

102

103 Reviewer Finally, isn't the total DIC variability by definition the sum of the various 104 components? So I'm not clear why the temperature driven component should ever be larger 105 than the total. I find equation 3 and the discussion on 304-311 to be the most confusing 106 part. We have an observed rate of change of DIC (which is never actually defined), which 107 one would think would be the sum of the contributions from gas exchange, biological uptake 108 /remineralization and entrainment. But in this case, the index that is considered is that the 109 total is either greater or less than one of these three components (whose physical meaning is nebulous). To confuse matters worse, we have a reference to "the total DIC seasonal cycle (dDIC/dt)" (306-307). Doesn't dX/dt imply an instantaneous rate-of-change that will itself vary over the annual cycle? I really do not understand what is being asserted here. (Also, the text should say something about exactly what sort of discretization was used in calculation of trends, e.g., does delta-X/delta-t for November represent a value for Nov. 1 based on a difference of October and November means, or is it something else? If this is the case, figure axes should indicate that calculated values are for the first of the month and not the mid-

- 117 month.)
- 118 **Response**: We thank the reviewer for this comment, it was important to clarify this point.
- 119 We apologize for giving this impression. We, regretfully, neglected to provide an adequate
- $120\,$ $\,$ description of how we separated the terms contributing to the total DIC surface layer $\,$
- 121 changes. We have clarified this part in the revised manuscript. The total rate of change of
- 122 DIC $\left(\frac{\partial DIC}{\partial t}\right)_{Tot}$ in the surface layer consists of the contribution of air-sea exchanges,
- 123 biological, vertical and horizontal transport-driven changes (eq. 1).

124
$$\left(\frac{\partial DIC}{\partial t}\right)_{Tot} = \left(\frac{\partial DIC}{\partial t}\right)_{air-sea} + \left(\frac{\partial DIC}{\partial t}\right)_{Bio} + \left(\frac{\partial DIC}{\partial t}\right)_{Vert} + \left(\frac{\partial DIC}{\partial t}\right)_{Hor}$$
 (eq.1)

125 In our method, we assumed that the horizontal term can be neglected because we used126 zonal means from medium resolution models

127 The discretized form of the total rate of change is written as:

128
$$\left(\frac{\partial DIC}{\partial t}\right)_{Tot} = \left(\frac{\Delta DIC}{\Delta t}\right)_{n,l} = \frac{DIC_{n+1,l} - DIC_{n,l}}{1 \text{ month}}$$
 (eq. 2)

129 where n is time in month, I is vertical level (in this case the surface, I=1). We here take the

130 forward derivative such that November rate the is difference between December the 15th

131 and November the 15^{th} , thus being centered at the interval between the months.

- 132 In order to compare the role of temperature in scalable terms with DIC in pCO_2 and FCO_2 , we
- 133 convert the instantaneous pCO₂ changes driven by solubility $\left(\text{using } \left(\frac{\partial pCO_2}{\partial DIC}\right)\frac{1}{pCO_2}\right) =$
- 134 $0.0423^{\circ}C^{-1}$ from Takahashi et al (1993) to an equivalent DIC change using the Revelle

135 factor
$$\left(\frac{\ln (pCO_2)}{\ln (DIC)} \approx \gamma_{DIC}\right)$$
.

136
$$\left(\frac{\partial DIC}{\partial t}\right)_{SST} = \frac{DIC}{\gamma_{DIC} \times pCO_2} \left(\frac{\partial pCO_2}{\partial t}\right)_{SST}$$
 (eq. 3)

This equivalent DIC rate of change (eq. 2) driven by temperature (solubility) allow us to convert the influence of solubility into DIC units, which can then be directly compared with the other terms in eq. 1. We hope that this now clarifies the construction of our indicator metrics,

141
$$M_{T-DIC} = \left| \left(\frac{\partial DIC}{\partial t} \right)_{SST} \right| - \left| \left(\frac{\partial DIC}{\partial} \right)_{Tot} \right|$$
 (eq. 4)

142 which is used to compare the estimated change of (equivalent) DIC as driven by

143 temperature-controlled solubility with the actual DIC change simulated by the models and

144 obtained from the observational data product. When $M_{T-DIC} > 0$ indicates that the pCO₂

145 variability is dominated by the rate of change of temperature and when $M_{T-DIC} < 0$ indicates

146 that the pCO_2 variability is controlled mainly by DIC changes.

147 We have now revised manuscript to better reflect the underlying methodology.

148 **Reviewer**: The discussion of entrainment is also confusing and poorly connected to actual

physical processes. Equation 4 does not have the units of a flux, but rather of a rate of

150 change within the surface layer. The proper quantity here is not DIC concentration at MLD

151 (T+1) but rather the difference between DIC at MLD(T+1) and at MLD(T).

152 **Response:** The reviewer is correct in this regard and we made this correction in the revised 153 text. The definition of entrainment at the base of the MLD has been improved using a more 154 appropriate notation (please refer to the discretization in the answer above that is now 155 included in the revised manuscript). Entrainment is physically considered as advection of 156 preformed DIC at the base of the mixed layer. It is therefore based on the advection term.

157

158
$$RE = U_e \left(\frac{\partial DIC}{\partial z}\right)_{MLD}$$
 (eq. 5)

159
$$RE_n = \left(\frac{\Delta MLD_n}{\Delta t}\right) \left(\frac{\Delta DIC}{\Delta z}\right)_{n,MLD}$$
 (eq. 6)

$$160 \qquad \Delta MLD = \frac{MLD_{n+1} - MLD_n}{1 Month}$$
(eq. 7)

161
$$\left(\frac{\Delta DIC}{\Delta z}\right)_{n,MLD} = \frac{DIC_{n,MLD_{n+1}} - DIC_{n,MLD_n}}{\Delta z}$$
 (eq. 8)

In which Ue is an equivalent velocity based on the rate of change of the mixed layer depth. This approximation of vertical entrainment is necessary as it is not possible to compute this term from the CMIP5 data because the vertical DIC distribution is only available as annual means. We clarified that we use the estimated rate of change of DIC at the base of the mixed to examine surface DIC changes driven by subsurface/bottom DIC changes. We also updated F_{DIC} estimate with GLODAP version 2 dataset.

169 **Reviewer:** Why use only ten years of model output (124)? The results could be biased by 170 internal variability; the more usual averaging period would be 20 or even 30 years. With a 171 reference year of 2000, this would require using emissions scenarios, which is perhaps a 172 reason not to do it, but the differences among scenarios are very small in 2005-2015 173 (because the scenarios are constructed precisely around the assumption that there is some 174 inertia in human societies and abrupt changes are unlikely). I think the authors should (a) 175 pick one model, recalculate the results for 1990-2010 and 1985-2015, and estimate the 176 potential error associated with aliasing of internal variability. And (b) if this error turns out 177 to be large, repeat the calculation for the full suite of models.

178 Response: The choice of the period was to match a period closest to the available 179 observational data product (Landschützer et al (2014), 1998 – 2011). However, the reviewer 180 is correct to highlight that we assumed that the seasonal cycle of CO₂ does not vary 181 significantly on decadal timescales. We have now investigated this assumption for a few 182 models (here we present HadGEM2-ES and CanESM2), comparing the mean seasonal cycle 183 climatology of FCO₂ at the Sub-Antarctic zone for 30 years (1975 – 2005) and 10 years (1995 184 – 2005) of the historical scenario (Fig. 3). It shows that the seasonal cycle of FCO₂ remain 185 the small (R = 0.99) in both HadGEM2-ES and CanESM2 over 30 year.

186 We have now added this sentence in the method section: "The choice of the 10 year period 187 was done to match the simulated pCO_2 values to the period of the observations. We tested 188 the interannual variability of the seasonal cycle over a period longer than the reference 10 189 years for a few models and found no significant variation in the monthly standard

190 deviation."



191

Figure 3. Compares the seasonal cycle of Sea-Air CO_2 fluxes over 30 year (1975 – 2005 and 10 years (1995 – 2015). The shaded area shows the standard deviation.

194

Reviewer: The use of chlorophyll as a proxy is not really explained, when primary production
and export production are generally available as model output fields. One might justify this
by saying that observations are available only for chlorophyll, but this should be stated
explicitly. There are also observation-based estimates of primary production available (see
below Terminology).

200 **Response:** Our initial choice for chlorophyll was because of the availability of an

201 observational data product for comparison and that it was available for most models (9 out

202 of 10) in the CMIP5 portal. In the revised manuscript, we added net primary production

203 (NPP), carbon export and oxygen for the models with available data. This addition of NPP,

204 carbon export and oxygen was indeed useful in examining impact of biological driven DIC

- 205 changes in particular it helped isolate/constrain the role of primary production on the
- 206 seasonal cycle of pCO₂.

208 Reviewer: I think the authors should acknowledge that the FCO₂ data product is not really 209 'observed' in the sense that pCO_2 is. I think they should compare modelled and observed 210 pCO_2 , and then discuss what this means for modelled estimates of CO_2 flux, without 211 referring to the Landschuter FCO₂ estimates as observations. CO₂ fluxes in models and data 212 products like this are actually quite different conceptually. When you estimate CO2 flux from 213 observed pCO2, the errors in the flux are a linear function of errors in wind speed (or u², 214 assuming a quadratic parameterization) and the piston velocity. In numerical models, pCO₂ 215 and DIC self-regulate to dampen these errors when monthly averaged fluxes are considered, 216 e.g., if both the wind speed and the DIC are too large, the enhanced outgassing flux will 217 reduce the pCO₂ and DIC error. Higher wind speed or piston velocity will tend to drive pCO_2 218 towards atmospheric, and not necessarily towards the 'correct' value if over- or 219 undersaturation exists, so there is no straightforward way to correct for this difference. But I 220 think that the authors should acknowledge that it exists, and that in comparing modelled 221 and 'observed' fluxes they are to some degree comparing apples and oranges. (With regard 222 to point (4) above, if there is no observed primary production, there is no observed CO_2 flux 223 either: both of these are extrapolated from the primary observed field using models of 224 unknown accuracy.)

225 **Response:** It is indeed true that FCO_2 observations we use are estimates, data products 226 derived from empirical methods of gap filling. We now refer to them in this answer and in 227 the revised manuscript as "data products". We tried to make the point much clearer in the 228 revised manuscript and also provided the uncertainty as given by Landschützer et al (2014) 229 and used the interannual standard deviation over 14 years when comparing the seasonal 230 cycle. However, it also important to note that the majority of our analysis is based on pCO_2 231 estimates rather than FCO_2 in particular. Thus, we do acknowledge the role of wind in 232 providing the kinematic forcing for sea-air fluxes and although the wind influences the 233 magnitude of the fluxes, the direction is determined by delta pCO₂, which is here considered 234 the primary driver of the seasonal cycle of FCO_2 . We are mindful that differences in the 235 parameterization of the Sea-Air interactions and wind products chosen are likely to affect 236 the resulting FCO₂ (Feely et al., 2004; Swart et al., 2014).

The uncertainty in the data products used for the assessment is an important point that we
neglected to highlight in our first version of the manuscript. We now aknolwedge the
limitations of the data products and have further addressed the issue of uncertainties as

requested by other reviewers as well.

241 We have examined the uncertainty of our reference data set by comparing Landschützer et 242 al., 2014 data product with the more recent Gregor et al (2017a) data product, which uses 243 Support Vector Regression (SVR) and Random Forest Regression (RFR), as well as Takahashi 244 et al (2009) to derive a seasonal climatology pCO_2 in the SAZ and AZ of the Southern Ocean 245 in the supplementary material (Fig. 5). Part of the reason we focus on pCO_2 in these data 246 products instead of FCO₂ is firstly, because Gregor et al., (2017a) only focuses on fugacity 247 and pCO₂. Thus we mindful that the choice of wind product and tranfer velocity constant in 248 computing FCO₂ is likely to increase the level of uncertainty for the compared data products 249 (Swart et al., 2014). Secondly, while we evaluate FCO_2 biases as the final aim of the paper, 250 the major part of our diagnostic analysis is based on pCO_2 rather FCO₂. Fig. 4 below shows 251 the climatology of the seasonal cycle of pCO₂ in the Sub-Antarctic zone and Antarctic zone 252 with interannual standard deviation between 1998 – 2011. All three datasets mostly agree in 253 the phasing of the seasonal cycle of pCO_2 in the Sub-Antarctic, but show significant 254 differences in the magnitude. Takahashi et al. (2009) shows an amplified impact of primary 255 production on pCO₂ in summer. We see this as a bias in the Takahashi et al., (2009) dataset 256 arising from a period when the space – time coverage of pCO_2 observations was still limited 257 and strongly biased towards summer. In the Antarctic zone all three observationally-based 258 data products agree in both phasing and amplitude. At this stage it is not clear whether this 259 agreement is due to all the methods being equally exposed to the same limited observations 260 or if it is due to a more marked CO₂ seasonal cycle in the Antarctic zone (relative to Sub-261 Antarctic) that can be captured with less observations



262

Fig. 4 pCO_2 (µatm) spatial (climatology) and seasonal cycle differences in Landschützer et al (2014) (L14), Gregor et al (2017) (G17), Takahashi et al (2009) (T09) datasets in the Southern Ocean. The seasonal cycle climatology of pCO_2 in the Sub-Antarctic and Antarctic zone is based on the period 1998 – 2011. The shaded areas show the standard deviation of the interannual variability of the seasonal cycle for this period. The uncertainty in the correlation coeffecient is based on the correlation coefficient of the mean plus stardard deviations seasonal cycle(s).

- Reviewer: I would like to see some discussion of the possibility that the apparently greater
 temperature control in the Pacific sector (259-263) is a real effect that arises from iron
 limitation. Because terrestrial sources of iron are much greater in Atlantic sector and the
 western half of the Indian sector (see e.g., Graham et al 2015 DSR I 104: 9; Tagliabue et al.,
 2012 Biogeosciences 9: 2333), it seems logical that the effect of seasonal biological
 drawdown on pCO₂ would be greater than in the Pacific and in the eastern half of the Indian
 sector. These regions also overlap the regions where the wind speed and the amplitude of
- its annual cycle are greatest (e.g., Trenberth et al., 1990, JPO 20: 1742), which will also tend
- 277 to reduce the influence of biological uptake
- 278 **Response:** Thank you for this important suggestion, it is indeed likely that differences in
- 279 wind, iron supply, primary productivity and MLD across the basins are most likely
- 280 responsible for contrasting variability in the three basins. We have added this discussion in

the revised manuscript;

282	"The observed differences in the seasonal cycle of FCO_2 across the three basins is likely due
283	to differences in the basin properties of the Southern Ocean. Recent studies have
284	highlighted significant basin scale differences in pCO_2 and FCO_2 ascribed to large-scale
285	differences in temperature (Landschutzer et al., 2015), winds (Gregor et al., 2017b and
286	primary production as reflected in surface ocean phytoplankton biomass (Thomalla et al.,
287	2011). The relatively higher chlorophyll biomass (Graham et al., 2015; Thomalla et al., 2011)
288	in the Atlantic Ocean, is likely linked to lower wind speeds (Trenberth et al., 1990) and
289	higher supplies of iron from continental shelves (Thomalla et al., 2011; Boyd and Ellwood,
290	2010; (Tagliabue et al., 2014)2; 2014). This is likely associated with longer periods of
291	shallower MLD (Dec – Mar, Fig. 7), which favor sustained primary production leading to a
292	stronger CO ₂ sink in the Atlantic Ocean with respect to the Indian and Pacific Ocean (Figs.
293	3a-c; 6a-c). In contrast, shorter periods of shallow MLD and lower iron concentrations in the
294	Pacific Ocean, as pointed out by Tagliabue et al. (2012), likely account for lower chlorophyll
295	biomass and stronger thermal control of the seasonal cycle of pCO_2 and FCO_2 (Fig. 6b, Fig. 3
296	here). In the Indian Ocean stronger wind speeds are likely responsible for the early
297	deepening of the MLD (Fig. 6c), and thus chlorophyll biomass are lower (Fig. 9). In the Indian
298	Ocean, stronger wind speeds (Trenberth et al., 1990) are likely responsible for the early
299	deepening of the MLD (Fig. 7c), limiting primary production and lower rates of change of
300	temperature (Fig. 5c), ultimately resulting in a relatively constant FCO_2 for about half the
301	year (Dec – Jun). Our plots indicate that CMIP5 models mostly don't show these basin-
302	specific features highlighted in observational products (Landschutzer et al., 2015; Gregor et
303	al., 2017a and Thomalla et al., 2011) with the exception of three group B models (i.e.
304	CESM1-BGC, CanESM2 and CMCC-CESM) in the Indian Ocean (Fig. 2, 3 a-c). This poses a
305	challenge to the new generation of Earth Systems Models in CMIP6"
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