

## Comments Referee 1

General comments: This manuscript deals with temporal variations of anthropogenic CO<sub>2</sub> in bottom waters in the Southern Ocean. The Southern Ocean is said to take up 40% of anthropogenic CO<sub>2</sub> absorbed by the ocean. Thus, investigations of temporal variability of anthropogenic CO<sub>2</sub> are very important to evaluate ocean's capacity of absorbing atmospheric CO<sub>2</sub>, information of which is indispensable for the projection of global warming. In terms of oceanic observation, the Southern Ocean is one of the regions, where the number of measurements, especially for chemical and biological properties, is scarce. In this point also, it is worth of being published in the journal. The manuscript is well organized, and is easy to read. The approaches used in the study are not new, but traditional ones. It is not a problem. It would be necessary to adopt an approach, which has been demonstrated to be useful for the detection of small signals of anthropogenic CO<sub>2</sub> variations. The authors attempt also to relate the variations to those of AABW formation, although not clearly found. As a whole, it seems that the manuscript is worthy of publication in the journal, but after a moderate revision. A few major comments are stated in the followings, and the minor ones are stated in the specific comments.

**Response: we are thankful for the quick answer provided by the reviewer. The concerns of the reviewer have been answered here after and have been valuable help to upgrade the manuscript.**

In this paper, temporal variability of anthropogenic CO<sub>2</sub> is examined using historical data collected at OISO. The data have been quality controlled by some data synthesis activities such as GLODAP. Nevertheless, I have a question on this point; the data syntheses have been done with a purpose of obtaining data consistency of a basin-scale. By contrast, the authors examine temporal variability of a local scale. In addition, data consistency is usually confirmed by data in deep layers of > 2000 m. This paper deals with data in deep layers. From these points, it is necessary to show that results obtained in the present study is not influenced by the data synthesis. Furthermore, for the recent data, quality control is made independently. Is there any possibility that the Cant stability is caused by the quality control? I recommend the authors to conduct quality-control on OISO data independently.

**Response: The reviewer is correct. For most of the ocean basins, data consistency is generally based on data in deep layers (> 1500 or 2000 m). However, because in the Southern Ocean anthropogenic CO<sub>2</sub> is also found at depth (> 3000 m), comparison is investigated in "old" deep waters, say around 2000-3000m (LCDW) where Cant (and DIC) should be relatively stable from one year to the next (within error of measurements, 1-3  $\mu\text{mol.kg}^{-1}$ ). Following the reviewer's recommendation, we propose to add a figure in Supplement Material (Fig. S1) showing the consistency of our dataset at the two OISO stations where samples were collected down to the bottom, the OISO-ST11 presented in the manuscript and the OISO-ST17 sampled in the Subtropical Zone (30° S-66° E). This figure shows a limited number of measurements that are out of the range of tolerance, but one has to keep in mind that interannual (or multiannual) variations may occur and this calls for great care before applying an adjustment.**

Since 1987 (when the cruise INDIGO3 was performed), a shift in  $A_T$  is suggested at high latitudes by the comparisons of INDIGO3 data (unadjusted, following the GLODAPv1 and CARINA recommendations) with other cruises data (adjusted, following the GLODAPv2 recommendations). This comparison shows differences that range between -4  $\mu\text{mol.kg}^{-1}$  and +10  $\mu\text{mol.kg}^{-1}$  (Fig. S2). Most of the crossovers that suggest a positive offset for INDIGO3 data (between +6  $\mu\text{mol.kg}^{-1}$  and +10  $\mu\text{mol.kg}^{-1}$ ) are found south of 60°S, suggesting that  $A_T$  may have decreased in deep waters at high latitudes since 1987. This is why we first decided for no adjustment in the submitted manuscript (as in the GLODAPv1 and CARINA data products, whereas the INDIGO3 data in GLODAPv2 were

corrected by  $-8 \mu\text{mol.kg}^{-1}$ ). However, at the OISO-ST11,  $A_T$  data from the INDIGO3 cruise are also about  $8 \mu\text{mol.kg}^{-1}$  higher than the mean value in deep waters (2000-3000m), in good agreement with the other crossovers at high latitudes. In order to reduce the potential bias that could result from either over-adjusting the data (GLODAPv2 recommendation) or not adjusting the data (GLODAPv1 and CARINA recommendations), and because most of the crossovers at mid-latitudes suggest a small positive offset, we propose to apply an intermediate adjustment of  $-4 \mu\text{mol.kg}^{-1}$  in the revised manuscript (the impact on  $C_{\text{ant}}$  is  $+2 \mu\text{mol.kg}^{-1}$ ). The uncertainty regarding this adjustment will be discussed in Supplement Material. Fig. 3 (before Fig. 4) presenting the interannual variability of LAABW properties and Table 2 presenting the calculated trends will be adjusted correspondingly.

Figure S1 also shows that the low  $A_T$  values between late 1998 and 2004 are found both in the Antarctic zone and the Subtropical zone. This is surprising, but there are no reason to believe that the data are biased since CMRs were used for all OISO cruises, and the instrument and data processing were the same during the first OISO cruise in January/February 1998 (showing  $A_T$  values close to the mean in Fig. S1) and the following cruises.

In discussion, the authors attempt to relate variations of anthropogenic  $\text{CO}_2$  in AABW to changes in AABW formation region. It is well discussed, but information of water mass age of AABW is lacking. It is necessary to show that linkages between variations of AABW formation region and observed AABW signals at OISO are appropriate in terms of water mass age.  $\text{O}_2$  and AOU are used simultaneously. I think, it is enough for one of which, probably AOU.

**Response:** we are sorry that there is no measurement related to water mass age in the available data (i.e. no CFCs measured during OISO cruises), other than  $\text{O}_2$  which is too sensitive to biological activity to be used as a water mass age tracer. We agree that the mention of both  $\text{O}_2$  and AOU is unnecessary. This is a point also noticed by Reviewer 2. Because we are most discussing the  $\text{O}_2$  concentration in the manuscript, we suggest to only present  $\text{O}_2$  in Figure 3.

Specific comments:

Line 18 (here around 460): “from about  $+7 \mu\text{mol kg}^{-1}$ ”, increase from what?

**Response:** We guess that what confused the two referees is the positive sign. We will delete the positive sign and rephrase as follows: ‘from the average concentration of  $7 \mu\text{mol.kg}^{-1}$  calculated for the period 1978-1987 to the averaged concentration of  $13 \mu\text{mol.kg}^{-1}$  in the period 2010-2018.’

Line 20 (here around 463): “CT”, this is the first appearance in the abstract. Write it in full.

**Response:** this will be added.

Line 23 (here around 467): “ $\theta$ , S”, they are the first appearance in the abstract. Write them in full.

**Response:** this will be added.

Lines 90-91 (here around 535-536): “station 430”, depth?

**Response:** the depth (4710 m) will be added.

Line 91 (here around 535): “405 km and 465 km”, away from where?

**Response:** These are the distance away from the OISO-ST11 sampling site. This will be rephrased as “located near the OISO-ST11 sampling site (405 km and 465 km away from it, respectively)”

Line 109 (here around 556): “the PET sector”, is it usually used? I do not understand where it is.

88 **Response:** A short sentence will be added to the text, as well as the references mentioned here after  
89 to clarify the use of this name. The PET, Princess Elizabeth Though, is also referred as the Balleny  
90 Though in Orsi et al. (1999), even if more currently mentioned as PET. It corresponds to the ocean  
91 section separating the Kerguelen Plateau from the Antarctic continent. Its deepest point is 3750 m,  
92 deep enough to allow AABWs to flow between the Australian Antarctic Basin and the Enderby Basin  
93 (Heywood et al., 1999). The work of Heywood et al. (1999; Fig. 1) revealed that in the northern part  
94 of the PET the AABW flow from west to east, while in the southern part the flow is from east to west.  
95

96 Line 150 (here around 612): “AT”, Probably this is the first appearance. Spell out here.

97 **Response:** this will be added.

98

99 Line 160 (here around 628): “ $\theta$  and S”, spell out here. ‘

100 **Response:** this will be added.

101

102 Lines 163-165 (here around 633-635): according the description, it seems that the figures are not  
103 accuracy but repeatability.

104 **Response:** The referee is correct. The accuracy is given by the analysis of CRMs. This will be corrected.

105

106 Line 236 (here around 714): “January”, which year. In this paper, all the data are analyzed assuming  
107 that seasonal variations in deep waters are negligible (lines 154-156). It is not appropriate to refer to  
108 months.

109 **Response:** the authors agree with the reviewer. This will be adjusted by mentioning the early and  
110 late 1998 sampling.

111 Line 276 (here around 762): “underlying”, do you mean a water mass below AABW?

112 **Response:** this is a mistake, we meant overlying the AABW (referring to LCDW). This will be corrected  
113 by using ‘LCDW’ instead.

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## Comments Referee 2

### General comments

The study presents results from a time series in the Indian sector of the Southern Ocean, which together with historical relevant data span a 40-year period. Using this time series, the authors evaluate the evolution of anthropogenic CO<sub>2</sub> (Cant) in the Antarctic Bottom Waters (AABW). It is an interesting and generally well written work, and generally good figures and tables. There are some need for clarity in some parts and there is some concern of the treatment of data gaps, but most of this should be rather easily dealt with, and I recommend publication after minor revision. A detailed list of comments follows below.

**The authors are thankful for the fast answer and the positive interest given to the manuscript, as well as for the numerous valuable comments.**

My main comments are related to the definition and subsequent presentation of AABW, and, the data gap between 1987 and 1998 and how this is handled and presented. To start with the definition of AABW, this is not an issue in itself, since the denser definition has been used before, and also, since almost any definition can be accepted as long as it is clearly presented. The latter is the problem here, at least for someone not as familiar with the area and these water masses (I usually work in the high-northern latitudes). The definition and choice is clearly described in 2.3, but, then the reader is referred to Fig. 3, where AABW is noted in the layer above the focus of this study, while the data evaluated is in the layer annotated "Considered data". When then the results of the property evolution of AABW are further presented in Fig. 4, at least I got somewhat confused. Whether this is only me or not, this may call for some added clarity. I would suggest to annotate your AABW layer (hence at neutral density >28.35) as AABW (or AABW\* or similar), to make this clear, and then make a distinction with the more common AABW.

**Response: the authors understand the concern of the reviewer. To solve this potential confusion, we suggest labelling the AABW as define in our manuscript (neutral density >28.35 kg.m<sup>-3</sup>) Lower Antarctic Bottom Water (LAABW).**

Nevertheless, this mostly refers to Fig. 3, and I have several concerns with this figure, as detailed below. Hovmöller plot is a wonderful thing, and can be very illustrative. However, it can also be deceiving, especially when there are gaps in the data, and the gridding is allowed to interpolate over these gaps, which often can create features that give a false picture of actual evolution. Fig. 3 suffers from this when plotting the older data (1978–1987) together with the OISO time-series data starting from 1998. There are several peculiar features in Fig. 3, especially for Cant and AT. The fact that most of the other plotted parameters show overall stable layer properties, over the full period, may seem to reduce this concern, but I am not convinced. In addition, I'm not fully convinced about the benefit of showing depths from 1500 m, when almost all results and discussion are concerned with the layer below 4000 m. Even more so when the upper layers seems to show most of the strange features, for example the minimum in Cant in the older data (which may in part show the issue with the TrOCA method, with even negative concentrations, which are not realistic, in the most upper part of the deep waters).

**Response: The authors agree that the figure needs to be upgraded, clarified and simplified. The suggestions of the referee have been taken into account by redrawing the Fig. 3 (now Fig. 2) using only the OISO data (from 1998 to 2018). The extrapolations were very misleading indeed, so the figure is now drawn with weighted-average gridding (and limited extrapolation around the data point). The aim of this figure is to show the differences in AABW and LCDW characteristics before**

focusing on the variability and trends observed in the bottom layer (it also shows that the neutral density 28.35 is a better definition for a more homogeneous bottom layer that we now define as LAABW). In addition, the control quality of the data is performed in the old deep waters (well characterized in the figure by the maximum in  $C_T$ ). Following the recommendation from the other Referee, we propose to add a figure in Supplement Material (Fig. S1) showing the consistency of our dataset at the two OISO stations where samples were collected down to the bottom, the OISO-ST11 presented in the manuscript and the OISO-ST17 sampled in the Subtropical Zone (30° S-66° E). This figure shows a limited number of measurements that are out of the range of tolerance, but one has to keep in mind that interannual (or multiannual) variations may occur and this calls for great care before applying an adjustment.

Since 1987 (when the cruise INDIGO3 was performed), a shift in  $A_T$  is suggested at high latitudes by the comparisons of INDIGO3 data (unadjusted, following the GLODAPv1 and CARINA recommendations) with other cruises data (adjusted, following the GLODAPv2 recommendations). This comparison shows differences that range between  $-4 \mu\text{mol.kg}^{-1}$  and  $+10 \mu\text{mol.kg}^{-1}$  (Fig. S2). Most of the crossovers that suggest a positive offset for INDIGO3 data (between  $+6 \mu\text{mol.kg}^{-1}$  and  $+10 \mu\text{mol.kg}^{-1}$ ) are found south of 60°S, suggesting that  $A_T$  may have decreased in deep waters at high latitudes since 1987. This is why we first decided for no adjustment in the submitted manuscript (as in the GLODAPv1 and CARINA data products, whereas the INDIGO3 data in GLODAPv2 were corrected by  $-8 \mu\text{mol.kg}^{-1}$ ). However, at the OISO-ST11,  $A_T$  data from the INDIGO3 cruise are also about  $8 \mu\text{mol.kg}^{-1}$  higher than the mean value in deep waters (2000-3000m), in good agreement with the other crossovers at high latitudes. In order to reduce the potential bias that could result from either over-adjusting the data (GLODAPv2 recommendation) or not adjusting the data (GLODAPv1 and CARINA recommendation), and because most of the crossovers at mid-latitudes suggest a small positive offset, we propose to apply an intermediate adjustment of  $-4 \mu\text{mol.kg}^{-1}$  in the revised manuscript (the impact on Cant is  $+2 \mu\text{mol.kg}^{-1}$ ). The Fig. 3 (before Fig. 4) presenting the interannual variability of the LAABW properties and the Table 2 presenting the calculated trends will be adjusted correspondingly. Fig. S2 will be completed by the list of the cruises presented.

The Figure S1 also shows that the low  $A_T$  values between late 1998 and 2004 are found both in the Antarctic zone and the Subtropical zone. This is surprising, but there are no reason to believe that the data are biased since CMRs were used for all OISO cruises, and the instrument and data processing were the same during the first OISO cruise in January/February 1998 (showing  $A_T$  values close to the mean in Fig. S1) and the following cruises.

The interpolation of this minimum patch leads to unfortunate wordings in the results, such as on line 236, with “a sudden increase. . . between January and December 1998” seems to refer to the low values calculated for the 1987 data and the clearly higher concentrations calculated for the OISO data. (I also don’t really understand the “between Jan and Dec 1998” part, since the first OISO data were sampled in Feb 1998, and the next in Dec the same year.) Apparently there are some need for clarifications here, but also to be cautious when interpreting interpolated values over large gaps. One way to solve this is of course to exclude the older data from the Hovmöller plots. These can still be used in the comparison/evaluation, and included in Fig. 4.

**Response:** the reviewer is right about the issue for the 1998 samplings mentioned (same as Reviewer 1). This is because the first OISO cruise started in January 1998, but the station 11 was actually sampled in the beginning of February as mentioned in Table 1. This will be corrected. We also agree that extrapolation can be misleading and we thank the Reviewer for pointing this issue. Having removed the GEOSECS and INDIGO data from the Hovmöller plots (Fig 2., before was Fig. 3), the extrapolation is no more an issue for interpreting the signal observed for the first OISO cruises, but

215 the increase in Cant between February 1998 and December 1998 remains (from < 6  $\mu\text{mol.kg}^{-1}$  to  
216 about 10  $\mu\text{mol.kg}^{-1}$ ).

217  
218 To continue on this figure (Fig. 3), for the bottom layer, the fact that it is stretched below the deepest  
219 samples seems to create at least the distinct maximum in mid-2000s. Perhaps this will be reduced if  
220 the maximum depth/pressure is set to the deepest sample, to exclude extrapolations below that  
221 depth.

222  
223 **Response: Having removed the INDIGO1 data from the Hovmöller plots, this no more an issue**  
224 **because the deepest sample is collected at the same depth for all cruises**

225  
226  
227 Specific comments

228 L18 (here around 460): Do the changes here (+7 and +13, respectively) refer to the whole period?  
229 Please clarify.

230 **Response: these are not changes, but Cant concentrations. The following rephrasing is suggested:**  
231 **‘from the average concentration of 7  $\mu\text{mol.kg}^{-1}$  calculated for the period 1978-1987 to the average**  
232 **concentration of 13  $\mu\text{mol.kg}^{-1}$  for the period 2010-2018.’**

233  
234 L23 (here around 467): A rather tiny remark, but the use of “pluriannual” may be grammatically correct  
235 (I’m not a native English speaker), but consider using “multiannual” (or multi-annual), which are more  
236 common (I believe). The same is used on L360.

237 **Response: We agree that this is maybe not the best word to use. It will be replaced by ‘multi-annual’.**

238 L59 (here around 502): I’m expecting a reference in the end of this sentence. This may be refer to the  
239 reference in the previous line, but you may consider moving this to the end.

240 **Response: We agree that the reference is misplaced. It will be moved to the end of the sentence.**

241 L95 (here around 541): I can’t find a definition of “AAC” anywhere. Please write out and define the first  
242 time.

243 **Response: We agree that the definition of ACC is missing (Antarctic Circumpolar Current). That will**  
244 **be corrected.**

245 L96-97 (here around 542-543): Unclear sentence. Need some rephrasing/re-writing. Suggestion: “. .  
246 .Weddell Sea, where deep and bottom waters are produced. . .”.

247 **Response: The sentence will be rephrased as suggested.**

248 L98-100 (here around 544-546): In the same sentence, there are several instances where the full water  
249 mass name is not spelled out, for example “the Ross Sea (RSBW; . . .”. This may be intuitive, but I don’t  
250 think the full names of some of these are written out at any place in the manuscript so would suggest  
251 to consider doing that at some place.

252 **Response: The full names will be added explicitly.**

253 L100 (here around 546): Rephrase: In the Prytz Bay, AABW formation has also. . . This sentence is  
254 overall quite unclear, especially the last part, so please consider rewriting for clarification.

255 **Response: It is indeed quite unclear. We propose the following rewriting : ‘AABW formation has also**  
256 **been observed in the Prydz Bay (Rodehacke et al., 2007; Yabuki et al., 2006). There, three polynyas**

257 and two ice shelves have been identified as Prydz Bay Bottom Water (PBBW) production hotspots  
258 from seal tagging data (Williams et al., 2016). This PBBW flows out the Prydz Bay through the Prydz  
259 Channel and get mixed with the CDBW.'

260  
261 L105 (here around 552): The "Warm Deep Water" is not described, so not easy to follow without a  
262 previous knowledge of the area and the present water masses. Please clarify.

263 **Response: we agree that it may be difficult to follow. The Warm Deep Water is slightly modified**  
264 **Circumpolar Deep Water (by mixing with surface waters when it enters the Weddell Basin). For**  
265 **simplification, we suggest rewriting as follows: The exported WSDW originates from the**  
266 **Circumpolar Deep Water (CDW) that enters the Weddell basin and mixes with WSBW and High**  
267 **Salinity Surface Water (HSSW) (see Fig.2 in van Heuven et al., 2011).**

268  
269 Section 2.4: Part of this section, and in particular from L133, deals with results of Cant from the  
270 methods not yet described. I would suggest to move this to the Result section, at least the Cant parts,  
271 or maybe part of the Discussion.

272 **Response: the authors agree that this section does not fit in the material and method part of the**  
273 **manuscript, but rather in the discussion section as suggested.**

274  
275 L152 (here around 614): Since the "P" in GLODAP refers to "Project", the "project" after should be  
276 avoided (I think). You could rephrase this into something like: . . .not yet qualified (or included in) the  
277 most recent GLODAPv2 product.

278 **Response: the mention of GLODAP will be rephrased as suggested.**

279  
280 L161 (here around 629): The stated accuracy for temperature and salinity seems too low. The standard  
281 CTD accuracy, for example found at the GO-SHIP home page (Hydro-manual) is 0.002 for both. Please  
282 check.

283 **Response: the authors agree and will correct the accuracy for temperature (0.002°C) and salinity**  
284 **(0.005 for measurements using a salinometer).**

285  
286 L161 (here around 629): As far as I can see, this is the first time "AT" is mentioned, but not defined.  
287 Please add this.

288 L166 (here around 636): Same for "O2" as for AT above. Please define first time.

289 **Response: AT and O2 will be defined here.**

290  
291 L170 (here around 641): You mean "onshore"?

292 **Response: the reviewer is right about this mistake.**

293  
294 L184 (here around 656): Clarify which "Redfield ratio". You mean the C:O ratio? Please add this.

295 **Response: Initially, only C/O<sub>2</sub> and N/O<sub>2</sub> ratios were involved in the definition of the parameter 'a'**  
296 **(Touratier and Goyet, 2004b; Lo Monaco et al., 2005b). In the latest definition of the method**  
297 **Touratier et al. (2007) presents an upgraded definition of this parameter by combining the Redfield**  
298 **equation coefficients for CO<sub>2</sub>, O<sub>2</sub>, HPO<sub>4</sub><sup>2-</sup> and H<sup>+</sup> and the same rules of construction as Broecker (1974)**  
299 **did for tracers NO or PO. Because we want to keep the explanation simple in the manuscript, we**  
300 **suggest to rephrase L184 as follows : 'where a is defined in Touratier et al. (2007) as combination of**

301 the Redfield equation coefficients for  $\text{CO}_2$ ,  $\text{O}_2$ ,  $\text{HPO}_4^{2-}$  and  $\text{H}^+$ . For more details about the definition  
302 and the calibration of this parameter, please refer to Touratier et al. (2007).'

303 L217 (here around 691): Either remove "after", so it reads "... and only impacted by. . .", or if more  
304 correct, add "subduction", so it reads "and after subduction only impacted by. . .".

305 **Response: the word 'subduction' will be added as suggested.**

306  
307 L233: "LCBW" is here mentioned for the first time, without definition or any description anywhere in  
308 the manuscript, as far as I can see. Please add this.

309 **Response: LCDW refers to the Lower Circumpolar Deep Water laying above AABW in the entire**  
310 **Southern Ocean. Details about this water mass will be added in Section 2.2 where it is first**  
311 **mentioned.**

312  
313 L235-236: This is what was commented on in the general comments above, with the "sudden  
314 increase". Please revise and clarify. It is more likely that there was a more gradual evolution, and none  
315 of the other parameters calls for any sudden changes. Also, the data quality and methods between the  
316 older data and the OISO data may differ, so extra caution is taken when comparing them.

317 **Response: we removed the older data from the Hovmöller plot, but the change in  $C_{\text{ant}}$  in LCDW**  
318 **remains (from  $<6 \mu\text{mol.kg}^{-1}$  in Feb 1998 (similar as for the older data) to about  $10 \mu\text{mol.kg}^{-1}$  for the**  
319 **following cruises).**

320  
321 L240 (here around 719): The maximum in  $C_{\text{ant}}$  in 2004 is one occasion, and followed by five (almost  
322 six) years without any data. I would be cautious to over interpret this. However, it co-incides with a  
323 maximum in oxygen, which could indicate a ventilation event.

324 **Response: we agree with the referee about being cautious with the measurements in 2004. Indeed**  
325 **the maximum in  $C_{\text{ant}}$  is due to the maximum in  $\text{O}_2$  (not associated with a maximum in  $C_T$ ).**

326  
327 L256-260 (here around 739-743): The lower concentrations of AT in the years around 2000 at all depths  
328 below (at least) 1500 m (have you checked the whole water column?) seems a bit odd. Especially when  
329 this is not seen in any of the other parameters. Also, when comparing two years in the 1980s with data  
330 more than a decade later, one should be extra cautious in the interpretation, not the least when the  
331 two years/occasions in 1985/87 show the highest concentrations seen over the evaluated period.  
332 Certainly the years after 2000 show much lower concentrations, which may be a phase due to a change  
333 in different forcing, but to suggest reduced calcification from only a few years/occupations of data is  
334 very speculative, and clearly something that change a few years later.

335 **Response: As mentioned in the general comments, the low  $A_T$  values between late 1998 and 2004**  
336 **are found both in the Antarctic zone and the Subtropical zone (Figure S1), but they are not observed**  
337 **in the surface layer (this will be added in the revised manuscript). The hypothesis about reduced**  
338 **calcification could explain this contrast between the surface waters and the deep ocean.**

339 L259-260 (here around 742-743): Is it realistic that the increase in  $C_T$  is lower than the accumulation  
340 of  $C_{\text{ant}}$ ?

341 **Response: The small increase in  $C_T$  over the period 1987-2004 could be caused by a reduction in  $C_{T,\text{nat}}$**   
342 **around the year 2000 (associated with the low  $A_T$  values). This said, we also have to keep in mind**  
343 **the uncertainty on the  $C_{\text{ant}}$  calculations. This will be clarified in the results and in the discussion.**



344 L261 (here around 744): While there is a rather clear trend in oxygen during this period – although I  
345 would be careful in talking about trends over such short periods, especially when comparing to a year  
346 with a maximum (2004) – there is no trend in Cant. Instead the latter shows some clear interannual  
347 variability. Also, the “trend” in temperature is indeed very small, and even if not significant, the change,  
348 or better, variability, in salinity is rather large. Consider these points when revising this part. Your  
349 statement on L267-268 highlights this issue.

350 **Response: we agree with the reviewer that there is no clear trend in Cant over 2004-2018. We will**  
351 **change “decrease in  $C_{ant}$ ” for “no increase in  $C_{ant}$ ”. The same is true for temperature and salinity.**

352 L270-271 (here around 755): There is also a maximum in temperature in 1985, so this could indicate  
353 more mixing with WSDW, which are both fresher and warmer.

354 **Response: we agree with the reviewer that more mixing with WSDW (or CDBW) could also explain**  
355 **the higher  $C_{ant}$  concentrations and lower S in 1985 (the signal in temperature is not well marked due**  
356 **to the large error bars). This will be added in the text.**

357  
358 L275-278 (here around 761-764): This is a very long sentence. I suggest to divide it, with period after  
359 “. . .the underlying deep waters.” Then remove “and”, and start on “Since”, or change the start of the  
360 sentence. For the last part of this sentence (L277-278), the suggestion of increased contribution from  
361 the Ross Sea is not clear to me since the oxygen decrease, while the salinity goes up and down. Or are  
362 you only referring to the one occupation in 2012? (If this is the case, it seems to detailed to explain a  
363 single year taken out of a long time series.)

364 **Response: the suggestion made by the reviewer to shorten the sentence will be used. Our aim is to**  
365 **discuss the variability in Cant concentrations that could reflect variations in the contribution of**  
366 **different types of AABWs. We suggest that the lower Cant concentrations observed in 2011, 2012**  
367 **and 2013 may be due to an increased contribution of older types of AABW. We agree that pointing**  
368 **to RSBW as a possible candidate because salinity was higher in 2012 is too speculative. This will be**  
369 **removed.**

370  
371 L280 (here around 767): The stated freshening of 0.01, for which period is that observed? Please clarify.

372 **Response: The sentence will be corrected as follows: ‘The freshening in S of -0.006 decade<sup>-1</sup> between**  
373 **2004 and 2018 that we observed on the Western side of the Kerguelen Plateau was also observed**  
374 **on the Eastern side of the Plateau by Menezes et al. (2017) over a similar period.’**

375 L312-313 (here around 823): “. . .(15  $\mu\text{mol kg}^{-1}$ ) due to mixing with older CDW.”

376 **Response: the sentence will be corrected.**

377 L317 (here around 827): “that contain very high amounts of Cant . . .”

378 **Response: the sentence will be correct as suggested.**

379 L318-320 (here around 829-830): The last sentence of this paragraph basically repeats what have been  
380 said above. Consider to remove.

381 **Response: the authors agree with the reviewer and will remove this sentence.**

382 L325 (here around 835): Here you write out “Southern Ocean” after having used the abbreviation  
383 throughout the manuscript, even the sentence before. Consider to revise.

384 **Response: “Southern Ocean” will be changed to SO.**

385 L340 (here around 845): “evaluated” should here instead be “estimated”, or “calculated”, or “found”  
386 (I think).

387 **Response: “evaluated” will be replaced by “calculated”.**

388 L386 (here around 898): Consider rewording “. . . vary in a very large range. . .”. Suggestion: “show a  
389 very large variability”, or maybe, “vary over a very large range”.

390 **Response: the rewording ‘vary over a very large range’ will be used.**

391 L387-388 (here around 901): “(–221 mmol C m<sup>-2</sup> d<sup>-1</sup>; Roden et al., 2016).

392 **Response: we will correct this according to the reviewer suggestion.**

393 L416 (here around 928): Both these water masses (RSBW and ALBW) have higher salinity, and while  
394 oxygen show a reduced trend the salinity goes up and down, so this explanation does not hold for all  
395 years during this period.

396 **Response: we understand the concern of the reviewer. The mention of the WSDW will be added, as**  
397 **for the response of the comment L275-278.**

398 L424 (here around 937): “explains most, but not all, of the observed. . .”

399 **Response: the sentence will be corrected.**

400 L463 (here around 977): GLODAPv2 version are written as “GLODAPv2.2021 (.2020 is soon to be  
401 released). You do mean 2021 and not 2020?

402 **Response: the data will not be included in GLODAP in the 2020 version, but in the following one.**

403  
404 L851-853 (here around 1353-1354): Table 2 (and in general): You may want to consider if you want to  
405 keep AOU as parameter, when you mostly refer to oxygen. The trends are almost exactly the same (but  
406 opposite of course), and gives the same message.

407 **Response: we agree with the reviewer. AOU will be removed from Table 2 and from Figure 2 (and**  
408 **from the corresponding parts in the text).**

409  
410 Technical comments

411 L22: This is, however, modulated. . .

412 **Response: the comas will be added.**

413 L35: The references should, typically, be chronologically ordered. Please check throughout the  
414 manuscript. (There are more examples of this, but I won’t comment on this more.)

415 **Response: we agree with the referee. We will check for other occurrences.**

416 L71: This is, however, not the. . .

417 **Response: the comas will be added.**

418 L91: “. . . (405 and 465 km, respectively).”

419 **Response: the coma will be added.**

420 L107-113: Exemplified with “. . .East of the Kerguelen. . .”, this section has many of these  
421 “directions/locations” (east/west/. . .) spelled with a large letter, even not part of a name. I think this  
422 is not correct, and if so, please change.

423 **Response: this will be corrected.**

424 L118-119: . . .28.27-bottom, respectively. . .

425 **Response: the coma will be added.**

426 L172-173: Change font; the part of the sentence from “for deep samples. . .” are in a different font  
427 (maybe “Cambria”).

428 **Response: the font will be changed.**

429 L220: Change font for “value for”.

430 **Response: the font will be changed.**

431 L306: Add a comma: “2018 (Fig 3a), probably . . .”

432 **Response: the coma will be added.**

433 L340: Add a “.”: Pardo et al. (2017)

434 **Response: the dot will be added.**

435 L347: For consistency, change “South-Western” to “South-western” (similar as on L325).

436 **Response: this will be corrected.**

437 L449: Remove “. ” for consistency: (e.g. Frölicher et al., 2014).

438 **Response: the coma will be deleted.**

439 L451: References in chronological order.

440 **Response: we agree with the referee. This will be corrected.**

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448 **Answer to the Topic Editor comments:**

449 Comments to the Author:

450 Dear Dr. Mahieu and co-authors,

451 Thank you for the revised submission. I am generally satisfied with the changes you made following  
452 the comments by the referees. Going through the manuscript myself, I have listed my comments  
453 below. Please prepare the final version of your manuscript taking into account these comments.

454 **Response: Dear Dr. Hoppema, we are thankful for your comments. Please find hereafter our**  
455 **responses.**

456 In the title: Antarctic Bottom Water (without –s) as this study measured only one type at one  
457 location.

458 **Response: we wanted to insist on the mix of AABW from different sources by writing it this way.**  
459 **This will be corrected as suggested.**

460 Section 2.1 is clearly part of the methods and should thus be moved to Section 3.

461 **Response: this section will be moved as suggested.**

462 I suggest to call the water mass defined here as Lower AABW, not as Low AABW. Just like other well-  
463 known water masses like Lower CDW, etc.

464 **Response: this will be corrected as suggested.**

465 Please place all 2 in CO<sub>2</sub> in subscript.

466 **Response: this will be corrected in the title and the references.**

467 Please check the references because many are incomplete.

468 **Response: this will be done. DOIs will be updated and page numbers checked. The last references**  
469 **without page numbers do not mention any online.**

470 As to the data used in this study, there are several more OISO cruises (as also in the GLODAP tables).  
471 Please provide the arguments for including the cruises that the authors did, while excluding others.

472 **Response: the missing OISO cruises in this study correspond to the cruises when this station was**  
473 **not re-occupied. To clarify this, the following sentence will be added to section AABW sampling: ‘In**  
474 **our analysis, we included all the data available for the OISO-ST11 location (which has not been**  
475 **sampled during each cruise for logistical reasons).’**

476 As to the supplement Table S1 (and discussion in the main text) with the adjustments from the  
477 different quality control efforts, it is shown that AT at the OISO cruises did not receive any  
478 adjustments. However, this is not the complete story. The GLODAP table says that there is not  
479 sufficient data for comparison in this region, upon which the OISO data did not get an adjustment  
480 because this could not be argued safely. This is actually the same as getting no quality control. This  
481 should be made clear in the manuscript.

482 **Response: we agree and will clarify this point as follows:**

483 '... this calls for great care before applying an adjustment. This is the case for  $A_T$  data that did not  
484 get an adjustment in GLODAP because this could not be argued safely due to the limited number of  
485 data in this region.'

486 L5 Shouldn't the University of Liverpool be mentioned?

487 **Response: this is missing indeed and will be added.**

488 L9 Antarctic bottom water (AABW) is known ...

489 **Response: this will be corrected.**

490 L9 ... but the sink is hardly quantified ...

491 **Response: this will be added.**

492 L13 in the framework of ...

493 **Response: this will be corrected.**

494 L16 At this location, the main sources of AABW are the low-saline ... (fresh is not the word here,  
495 because this is a saline water mass; I suggest to skip "younger" because: younger against what?)

496 **Response: we understand your concern and will correct this sentence as suggested.**

497 L20 SO has not been defined before

498 **Response: this will be added.**

499 L24 hydrographic (not: hydrological)

500 **Response: this will be corrected.**

501 L27 AABW

502 **Response: this will be corrected.**

503 L27-28 This sentence is trivial, and if not followed by which of these processes are important or how  
504 they function, not necessary/useful.

505 **Response: we agree and will remove this sentence.**

506 L43 3% is more like the maximum. Mostly Cant is much less. I suggest to write here: less than 3%

507 **Response: we agree and will correct this.**

508 L53-55 "Thus, there is a need to better explore the CT and Cant temporal variability in the deep  
509 ocean, especially in the SO where observations are relatively sparse." I cannot understand the  
510 connection of this concluding sentence with the previous text in this paragraph. Please modify.

511 **Response: we agree that the sentence has no clear link with the previous statements. We suggest  
512 to remove it.**

513 L56 AABW (without -s) Please change this throughout the manuscript.

514 **Response: this will be corrected.**

515 L58 ... by covering a major part of the world ocean floor ...

516 **Response: this will be corrected.**

517 L84 Study area

518 **Response: this will be corrected.**

519 L86 framework

520 **Response: this will be corrected.**

521 L98 is dominated by (instead of: is mainly governed)

522 **Response: this will be corrected.**

523 L111 ... Lower Circumpolar ...

524 **Response: this will be corrected.**

525 L112 I think HSSW is generally the abbreviation for High Salinity Shelf Water

526 **Response: this is correct, the abbreviation will be removed.**

527 L116-117 The PE deepest point of the PET is 3750 m, ...

528 **Response: this will be corrected as 'The deepest point of the PET is 3750 m...'**

529 L158-160 "The accuracy of CT and AT measurements was ensured by daily analyses of Certified  
530 Reference Materials (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of  
531 Oceanography)." This is indeed important to warrant the accuracy. For the interpretation it is also  
532 important to know the accuracy. Please give the accuracy here.

533 **Response: A single accuracy value for all cruises is difficult to specify. Although we used the same  
534 technic (and data processing) accuracy range between around 1.5 and 3  $\mu\text{mol}/\text{kg}$  for both AT and  
535 CT depending on the cruise. A complete list of CRMS batch number used during OISO cruise is  
536 available at NCEI/OCADS with information on duplicates for each cruise  
537 ([https://www.nodc.noaa.gov/ocads/oceans/VOS\\_Program/OISO.html](https://www.nodc.noaa.gov/ocads/oceans/VOS_Program/OISO.html)). As this information is  
538 available at NCEI/OCADS (and the link recall in the section "Data Availability"), we think it was not  
539 appropriate to list all CRM batch values for each cruise in the manuscript. We suggest to correct as  
540 follows: 'The accuracy of  $C_T$  and  $A_T$  measurements (always better than  $\pm 3 \mu\text{mol.kg}^{-1}$  for all cruises  
541 since 1998) was ensured...'**

542 L164 silicate (no capital)

543 **Response: this will be corrected.**

544 L171 using (instead of: considering)

545 **Response: this will be corrected.**

546 L171 I do not understand why the value of 33  $\mu\text{mol}/\text{kg}$  was used, as the mean value from GLODAPv2  
547 is 32.4  $\mu\text{mol}/\text{kg}$ . Even if the error because of this is small, it does increase it for no good reason.

548 **Response: this is correct. The value has been changed to 32.4  $\mu\text{mol.kg}^{-1}$ . The change on the  $C_{\text{ant}}$   
549 values calculated with  $C^\circ$  is -0.3  $\mu\text{mol.kg}^{-1}$ .**

550 L187-188 from deep waters free of anthropogenic  $\text{CO}_2$  ...

551 **Response: this will be corrected.**

552 L245-246 “the theoretical  $C_{\text{ant}}$  trend at the AABW formation sites would be of the order of +8  
553  $\mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ .” How was this calculated? Only part of the AABW, when it is formed, contains  
554 water that has been at the surface. Only that part could follow the atmospheric increase on  $\text{CO}_2$ .  
555 What percentage of surface water was assumed as contributing to AABW?

556 **Response:** This value was listed to give a taste of the theoretical  $C_{\text{T}}$  increase in Antarctic surface  
557 waters assuming that ocean  $f\text{CO}_2$  follows the atmospheric  $\text{CO}_2$  increase. In the Prydz Bay region  
558 Roden et al (2016) concluded that “surface waters in the seasonal ice zone track the atmospheric  
559 increase in  $f\text{CO}_2$ ”. For our calculation we used the mean properties in Antarctic surface waters  
560 observed in the Prydz Bay by Roden et al. (2016):  $\text{SST}=-1^\circ\text{C}$ ,  $\text{SSS}=34.2$ ,  $A_{\text{T}}=2291 \mu\text{mol/kg}$  and  $f\text{CO}_2=$   
561  $376 \mu\text{atm}$  in 2006. Assuming that oceanic  $f\text{CO}_2$  increased at a rate of  $1.8 \mu\text{atm}$  we calculated  $C_{\text{T}}$  and  
562 we derived a trend in  $C_{\text{T}}$  of  $+8 \mu\text{mol/kg/decade}$  in the Antarctic surface water (assuming no change  
563 in temperature, salinity and alkalinity). Note that this value is close to the theoretical trend in  $C_{\text{T}}$   
564 calculated by Van Heuven et al. (2014) in the Weddell Sea (about  $+0.8 \mu\text{mol/kg/yr}$ , the red circle in  
565 Figure 4a in Van Heuven et al., 2014). We suggest to revise following: “Due to the mixing of AABW  
566 with old CDW ( $C_{\text{ant}}$  free), these trends are lower than the theoretical trend expected from the  
567 increase in atmospheric  $\text{CO}_2$ . Indeed, assuming that the surface ocean  $f\text{CO}_2$  follows the  
568 atmospheric growth rate ( $+1.8 \mu\text{atm.year}^{-1}$  over 1978-2018) in the seasonal ice zone (Roden et al.,  
569 2016), the theoretical  $C_{\text{ant}}$  trend at the AABW formation sites would be of the order of  $+8 \mu\text{mol.kg}^{-1}.$   
570  $\text{decade}^{-1}$  in the Antarctic surface water. This is close to the theoretical  $C_{\text{T}}$  trend estimated for  
571 freezing shelf water in the Weddell Sea (Van Heuven et al 2014).”

572 L288 experiences

573 **Response:** this will be corrected.

574 L310 and ends ... (instead of: and lasts)

575 **Response:** this will be corrected.

576 L315 ... in the 1980s in the Indian sector of the Southern Ocean ...

577 **Response:** this will be corrected.

578 L316 quality control (instead of: qualification)

579 **Response:** this will be corrected.

580 L427 “recognized freshening of AABWs over the last decades (Rintoul, 2007).” With a reference from  
581 2007, this is not about the last decades. Please change wording or give a different reference.

582 **Response:** we agree that there is a lack of consistency between the sentence and reference. We  
583 suggest to change the sentence as follow: ‘recognized freshening of the AABW (Rintoul, 2007;  
584 Anilkumar et al., 2015).’

585 L484 change to: GLODAPv2.2021

586 **Response:** this will be corrected.

587 L504 Please add info on what kind of this reference is and possibly where it can be found online.

588 **Response:** we suggest to replace the current reference by the following:  
589 Coverly, S. C., Aminot, A., and R. K  rouel, 2009. Nutrients in Seawater Using Segmented Flow  
590 Analysis, In Practical Guidelines for the Analysis of Seawater, Ed. Oliver Wurl, CRC Press, June 2009,  
591 doi: 10.1201/9781420073072.ch8.

592 L506 Cycles (also in other cases where this journal is concerned)  
593 **Response: this will be corrected.**  
594 L538 pages: 205-206  
595 **Response: this will be corrected.**  
596 L551 pCO<sub>2</sub>  
597 **Response: this will be corrected.**  
598 L602 should be cited as: 18, GB1042, doi:10.1029/2002GB002017  
599 L606 should be cited differently, similar as above  
600 L624 pages: 346-349  
601 L636 pages: 1221-1224  
602 In many cases the references are incomplete, for example missing page numbers. Please go through  
603 the references and correct them.  
604 **Response: all the references will be checked and updated. The DOIs will be updated, and the page**  
605 **numbers checked. The last references without page numbers do not mention any online.**  
606 Figure 1 Please add that these are very rough transport paths. The dashed line for the ACC gives the  
607 position, says the caption. What position? The ACC is wide; please explain. The path of the AABW in  
608 the Weddell Sea is not correct. Neither is the path of the AABW from Prydz Bay and Cape Darnley,  
609 which flows along the coast to the west and enters the Weddell circulation.  
610 **Response: the mention will be added and the figure updated.**  
611 Figure 2 The term is Hovmöller diagram.  
612 **Response: this will be corrected.**  
613 Thank you and best wishes  
614 Mario Hoppema

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# Variability and stability of anthropogenic CO<sub>2</sub> in Antarctic Bottom Waters observed in the Indian sector of the Southern Ocean, 1978-2018

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## Abstract

Antarctic bottom waters (AABWs) are known as a long term sink for anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) but the sink is hardly quantified because of the scarcity of the observations, specifically at an interannual scale. We present in this manuscript an original dataset combining 40 years of carbonate system observations in the Indian sector of the Southern Ocean (Enderby Basin) to evaluate and interpret the interannual variability of C<sub>ant</sub> in the AABW. This investigation is based on regular observations collected at the same location (63° E/56.5° S) in the framework of the French observatory OISO from 1998 to 2018 extended by GEOSECS and INDIGO observations (1978, 1985 and 1987).

At this location the main sources of AABW sampled is the ~~fresh and younger low-saline~~ Cape Darnley Bottom Water (CDBW) and the Weddell Sea Deep Water (WSDW). Our calculations reveal that C<sub>ant</sub> concentrations increased significantly in the AABW, from the average concentration of 7 μmol.kg<sup>-1</sup> calculated for the period 1978-1987 to the average concentration of 13 μmol.kg<sup>-1</sup> for the period 2010-2018. This is comparable to previous estimates in other ~~Southern Ocean (SO)~~ basins, with the exception of bottom waters close to their formation sites where C<sub>ant</sub> concentrations are about twice as large. Our analysis shows that total carbon (C<sub>T</sub>) and C<sub>ant</sub> increasing rates in the AABW are about the same over the period 1978-2018, and we conclude that the long-term change in C<sub>T</sub> is mainly due to the uptake of C<sub>ant</sub> in the different formation regions. This is, however, modulated by significant interannual to multi-annual variability associated with variations in hydrographic (potential temperature (Θ), salinity (S)) and biogeochemical (C<sub>T</sub>, total alkalinity (A<sub>T</sub>), dissolved oxygen (O<sub>2</sub>)) properties. A surprising result is the apparent stability of C<sub>ant</sub> concentrations in recent years despite the increase in C<sub>T</sub> and the gradual acceleration of atmospheric CO<sub>2</sub>. ~~The C<sub>ant</sub> sequestration by AABWs is more variable than expected and depends on a complex combination of physical, chemical and biological processes at the formation sites and during the transit of the different AABWs.~~ The interannual variability at play in AABWs needs to be carefully considered on the extrapolated estimation of C<sub>ant</sub> sequestration based on sparse observations over several years.

## 1 Introduction

Carbon dioxide (CO<sub>2</sub>) atmospheric concentration has been increasing since the start of the industrialization (Keeling and Whorf, 2000). This increase leads to an ocean uptake of about a quarter of C<sub>ant</sub> emissions (Le Quéré et al., 2018; Gruber et al., 2019a). It is widely acknowledged that the Southern Ocean (SO) is responsible for 40

659 % of the  $C_{\text{ant}}$  ocean sequestration (Matear, 2001; Orr et al., 2001; McNeil et al., 2003; Gruber et al., 2009;  
 660 Khatiwala et al., 2009). Ocean  $C_{\text{ant}}$  uptake and sequestration have the benefit to limit the atmospheric  $\text{CO}_2$  increase  
 661 but also result in a gradual decrease of the ocean pH (Gattuso and Hansson, 2011; Jiang et al., 2019). Understanding  
 662 the oceanic  $C_{\text{ant}}$  sequestration and its variability is of major importance to predict future atmospheric  $\text{CO}_2$   
 663 concentrations, impact on the climate and impact of the pH change on marine ecosystems (de Baar, 1992; Orr et  
 664 al., 2005; Ridgwell and Zeebe, 2005).  
 665  $C_{\text{ant}}$  in seawater cannot be measured directly and the evaluation of the relatively small  $C_{\text{ant}}$  signal from the total  
 666 inorganic dissolved carbon ( $C_T$ ; ~~around less than 3 %~~; Pardo et al., 2014) is still a challenge to overcome. Different  
 667 approaches have been developed in the last 40 years to quantify  $C_{\text{ant}}$  concentrations in the oceans. The ‘historical’  
 668 back calculation method based on  $C_T$  measurement and preformed inorganic carbon estimate ( $C^0$ ) was  
 669 independently published by Brewer (1978) and Chen and Millero (1979). This method has been often applied at  
 670 regional and basin scale (Chen, 1982; Poisson and Chen, 1987; Chen, 1992; Goyet et al., 1998; Körtzinger et al.,  
 671 1998, 1999; Lo Monaco et al., 2005a). More recently the TrOCA method (Tracer combining Oxygen, dissolved  
 672 Carbon and total Alkalinity) has been developed (Touratier and Goyet, 2004a, b; Touratier et al., 2007) and applied  
 673 in various regions including the SO (e.g. Lo Monaco et al., 2005b; Sandrini et al., 2007; Van Heuven et al., 2011;  
 674 Pardo et al., 2014; Shadwick et al., 2014; Roden et al., 2016; Kerr et al., 2018). Comparisons with other data-based  
 675 methods show significant differences in  $C_{\text{ant}}$  concentrations, especially at high latitudes and more particularly in  
 676 deep and bottom waters (Lo Monaco et al., 2005b; Vázquez-Rodríguez et al., 2009; Pardo et al., 2014). ~~Thus, there~~  
 677 ~~is a need to better explore the  $C_T$  and  $C_{\text{ant}}$  temporal variability in the deep ocean, especially in the SO where~~  
 678 ~~observations are relatively sparse.~~  
 679 Antarctic bottom waters (AABW\*) are of specific interest for the atmospheric  $\text{CO}_2$  and heat regulation as they  
 680 play a major role in the meridional overturning circulation (Johnson et al., 2008; Marshall and Speer, 2012).  
 681 AABWs represent a large volume of water by covering ~~the majority of the bottom~~ a major part of the world ocean  
 682 floor (Mantyla and Reid, 1995), and their spreading in the interior ocean through circulation and water mixing is  
 683 a key mechanism for the long-term sequestration of  $C_{\text{ant}}$  and climate regulation (Siegenthaler and Sarmiento, 1993).  
 684 The AABW formation is a specific process occurring in few locations around the Antarctic continent (Orsi et al.,  
 685 1999). In short, the AABW formation occurs when the Antarctic surface waters flows down along the continental  
 686 shelf. The Antarctic surface waters density required for this process to happen is reached by the increase in salinity  
 687 (S) due to brine release from the ice formation and by a decrease in temperature due to heat loss to either the ice-  
 688 shelf or the atmosphere. Importantly, AABW formation process is enhanced by katabatic winds that open areas  
 689 free of ice called polynyas (Williams et al., 2007). Indeed, katabatic winds are responsible for an intense cooling  
 690 that enhance the formation of ice constantly pushed away by the wind, leading to cold and salty surface waters in  
 691 contact with the atmosphere. The variable conditions of wind, ice production, surface water cooling and continental  
 692 slope shape encountered around the Antarctic continent lead to different types of AABW, hence the AABW  
 693 characteristics can be used to identify their formation sites.  
 694 The ability of AABW to accumulate  $C_{\text{ant}}$  has been controversial since one can believe that the ice coverage limits  
 695 the invasion of  $C_{\text{ant}}$  in Antarctic surface waters (e.g. Poisson and Chen, 1987). This is, however, not the case in  
 696 polynyas, and several studies have reported significant  $C_{\text{ant}}$  signals in AABW formation regions, likely due to the  
 697 uptake of  $\text{CO}_2$  induced by high primary production (Sandrini et al., 2007; van Heuven et al., 2011, 2014; Shadwick  
 698 et al., 2014; Roden et al., 2016). However, little is known about the variability and evolution of the  $\text{CO}_2$  fluxes in

AABW formation regions, and since biological and physical processes are strongly impacted by seasonal and interannual climatic variations (Fukumachi et al., 2000; Gordon et al., 2010; McKee et al., 2011; Gordon et al., 2015; Gruber et al., 2019b), the amount of  $C_{ant}$  stored in the AABWs may be very variable, which could bias the estimates of  $C_{ant}$  trends derived from data sets collected several years apart (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019). In this context of potentially high variability in  $C_{ant}$  uptake at AABW formation sites, as well as in AABW export, circulation and mixing, we used repeated observations collected in the Indian sector of the Southern Ocean to explore the variability in  $C_{ant}$  and  $C_T$  in the AABW and evaluate their evolution over the last 40 years.

## 2 Study area

### 2.1 AABW sampling during the last 40 years

Most of the data used in this study were obtained in the frame of the long-term observational project OISO (Ocean Indien Service d'Observations) conducted since 1998 onboard the R.S.V. Marion-Dufresne (IPEV/TAAF). During these cruises, several stations are visited, but only one station is sampled down to the bottom (4800 m) south of the Polar Front at 63.0° E and 56.5° S (hereafter noted OISO-ST11). This station is located in the Enderby Basin on the Western side of the Kerguelen Plateau (Fig. 1) and coincides with the station 75 of the INDIGO-3 cruise (1987). In our analysis, we also included data from the station 14 (deepest sample taken at 5109 m) of the INDIGO-4 cruise (1985) and the station 430 (deepest sample taken at 4710 m) of the GEOSECS cruise (1978) located near OISO-ST11 sampling site (405 km and 465 km away from it, respectively; Fig. 1). All the re-occupations used in this analysis are listed in Table 1. Since seasonal variations are only observed in the surface mixed layer (Metzl et al., 2006), we used the observations available for all seasons (Table 1).

Table 1

### 2.2 AABWs circulation in the Atlantic and Indian sectors of the Southern Ocean

The circulation in the SO is mainly governed by the Antarctic Circumpolar Current (ACC) that flows eastward, while the Coastal Antarctic Current (CAC) flows westward (Fig. 1) (Carter et al., 2008). The ACC and the CAC influence the circulation of the entire water column, including the AABWs and generate gyres, crucial drivers of SO circulation (Carter et al., 2008). The most important gyres encountered around the Antarctic continent correspond to major AABW formation sites (Fig. 1). The main AABW formation sites are the Weddell Sea, where Weddell Sea Deep Water and Weddell Sea Bottom Water are produced (WSDW and WSBW, respectively; Gordon, 2001; Gordon et al., 2010), the Ross Sea for the Ross Sea Bottom Water (RSBW; Gordon et al., 2009, 2015), the Adelie Land coast for the Adelie Land Bottom Water (ALBW; Williams et al., 2008, 2010) and the Cape Darnley Polynya for the Cape Darnley Bottom Water (CDBW; Ohshima et al., 2013). AABW formation has also been observed in the Prydz Bay (Yabuki et al., 2006; Rodehacke et al., 2007). There, three polynyas and two ice shelves have been identified as Prydz Bay Bottom Water (PBBW) production hotspots from seal tagging and mooring data (Williams et al., 2016). This PBBW flows out the Prydz Bay through the Prydz Channel and get mixed with the CDBW. The mix of CDBW and PBBW (hereafter called CDBW) represents a significant AABW export (13 % of all AABWs exports; Ohshima et al., 2013).

The largest bottom water source of the global ocean is the Weddell Sea (Gordon et al., 2001). The exported WSDW is a mixture of the WSBW and Warm Deep Water (WDW). The WDW is a slightly modified Low ~~er~~ Circumpolar Deep Water (LCDW) by mixing with ~~h~~High ~~s~~Salinity ~~s~~Surface wWater (~~HSSW~~) when the LCDW enters the Weddell basin (see Fig. 2 in van Heuven et al., 2011). The WSDW mixes with the LCDW during its transit ~~from the Weddell basin~~. A part of the WSDW deflecting southward with the ACC in the Enderby Basin reaches the north-western part of the Princess Elizabeth Trough (PET) region (area separating the Kerguelen Plateau from the Antarctic continent), where it mixes with other types of AABW~~s~~ (Heywood et al., 1999; Orsi et al., 1999). The ~~PET~~deepest point ~~of the PET~~ is 3750 m, deep enough to allow AABW~~s~~ to flow between the Australian Antarctic Basin and the Enderby Basin (Heywood et al., 1999).

At the east of the PET, the CAC transports a mixture of RSBW and ALBW and accelerates northward along the eastern side of the Kerguelen Plateau (Mantyla and Reid, 1995; Fukamachi et al., 2010) ~~following the Australian-Antarctic gyre, also called Kerguelen gyre (Vernet et al. 2019)~~. Part of the ALBW-RSBW mixture ~~also~~ reaches the western side of the Kerguelen Plateau by the southern part of the PET (Heywood et al., 1999; Orsi et al., 1999; Van Wijk and Rintoul, 2014) and mixes with the CDBW. The mixture of CDBW and ALBW-RSBW ~~either~~ flows westward with the CAC and dilutes with the LCDW (Meijers et al., 2010) ~~or flows northward (Ohshima et al., 2013) and mixes with the WSDW before reaching the location of our time-series station in the eastern Enderby Basin until it reaches the Weddell gyre (Carter et al., 2008)~~.

Figure 1

## 2.3.2 AABW definition

The distinction of water masses is usually performed according to neutral density ( $\gamma^n$ ) layers. In the SO, LCDW and AABW properties are generally well defined in the range 28.15-28.27 kg.m<sup>-3</sup> and 28.27-bottom, respectively (Orsi et al., 1999; Murata et al 2019). However, to interpret the long-term variability of the properties in the AABW core at our location, we prefer to adjust the AABW definition to a narrow (more homogeneous) layer that we call Lower Antarctic Bottom Water (LAABW), characterised by  $\gamma^n > 28.35$  kg.m<sup>-3</sup> (roughly ranging from 4200m to 4800m, see Fig. 3). This definition corresponds to the AABW characteristics observed at higher latitudes in the Indian SO sector (Roden et al., 2016). The layer above the LAABW is hereafter called Upper Antarctic Bottom Water (UAABW).

## 3 Material and methods

### 3.1 AABW sampling during the last 40 years

Most of the data used in this study were obtained in the framework of the long-term observational project OISO (Ocean Indien Service d'Observations) conducted since 1998 onboard the R.S.V. Marion-Dufresne (IPEV/TAAF). During these cruises, several stations are visited, but only one station is sampled down to the bottom (4800 m) south of the Polar Front, at 63.0° E and 56.5° S (hereafter noted OISO-ST11). This station is located in the Enderby Basin on the Western side of the Kerguelen Plateau (Fig. 1) and coincides with the station 75 of the INDIGO-3 cruise (1987). In our analysis, we included all the data available for the OISO-ST11 location (which has not been sampled during each cruise for logistic reasons). We also included data from the station 14 (deepest sample taken at 5109 m) of the INDIGO-1 cruise (1985) and the station 430 (deepest sample taken at 4710 m) of the GEOSECS

cruise (1978) located near OISO-ST11 sampling site (405 km and 465 km away from it, respectively; Fig. 1). All the re-occupations used in this analysis are listed in Table 1. Since seasonal variations are only observed in the surface mixed layer (Metzl et al., 2006), we used the observations available for all seasons (Table 1).

### 3.2.1 Validation of the data

For 1998-2004, the OISO data were quality controlled in CARINA (Lo Monaco et al., 2010) and for 2005 and 2009-2011 in GLODAPv2 (Key et al., 2015; Olsen et al., 2016, 2019). The 3 additional datasets from GEOSECS, INDIGO-1 and INDIGO-3 were first qualified in GLODAPv1 (Key et al., 2004) and used for the first  $C_{\text{ant}}$  estimates in the Indian Ocean (Sabine et al., 1999). The adjustments recommended for these historical datasets have been revisited in CARINA and GLODAPv2. In this paper we used the revised adjustments applied to the GLODAPv2 data product, with one exception for the total alkalinity ( $A_T$ ) data from INDIGO-3 for which we applied an intermediate adjustment between the recommendation from GLODAPv1 (confirmed in CARINA) for no adjustment (in reason of the lack of available observations in this region for robust comparison) and the adjustment by  $-8 \mu\text{mol.kg}^{-1}$  applied to the GLODAPv2 data product (justification in Supp. Mat.). For the recent OISO cruises conducted in 2012-2018 not yet included in the most recent GLODAPv2 product, we have proceeded to a data quality control in deep waters where  $C_{\text{ant}}$  concentrations are low and subject to very small changes from year to year (see Supp. Mat.).

### 3.2.3 Biogeochemical measurements

Measurement methods during OISO cruises were previously described (Jabaud-Jan et al., 2004; Metzl et al., 2006). In short, measurements were obtained using Conductivity-Temperature-Depth (CTD) casts fixed on a 24 bottles rosette equipped with 12 L General Oceanics Niskin bottles. Potential temperature ( $\Theta$ ) and salinity ( $S$ ) measurements have an accuracy of  $0.002^\circ\text{C}$  and  $0.005$  respectively.  $A_T$  and  $C_T$  were sampled in 500 mL glass bottles and poisoned with 100  $\mu\text{L}$  of mercuric chloride saturated solution to halt biological activity. Discrete  $C_T$  and  $A_T$  samples were analyzed onboard by potentiometric titration derived from the method developed by Edmond (1970) using a closed cell. The repeatability for  $C_T$  and  $A_T$  varies from 1 to  $3.5 \mu\text{mol.kg}^{-1}$  (depending on the cruise) and is determined by sample duplicates (in surface, at 1000 m and in bottom waters). The accuracy of  $C_T$  and  $A_T$  measurements (always better than  $\pm 3 \mu\text{mol.kg}^{-1}$  for all cruises since 1998) was ensured by daily analyses of Certified Reference Materials (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of Oceanography). Dissolved oxygen ( $\text{O}_2$ ) concentration was determined by an oxygen sensor fixed on the rosette. These values were adjusted using measurements obtained by Winkler titrations using a potentiometric titration system (at least 12 measurements for each profile). The thiosulphate solution used for the Winkler titration was calibrated using iodate standard solution (provided by Ocean Scientific International Limited) to ensure the standard  $\text{O}_2$  accuracy of  $2 \mu\text{mol.kg}^{-1}$ . Nitrate ( $\text{NO}_3$ ) and silicate ( $\text{Si}$ ) concentrations were measured onboard or onshore with an automatic colorimetric Technicon analyser following the methods described by Tréguer and Le Corre (1975) until 2008, and the revised protocol described by Aminot and Kérouel (2007) Coverly et al. (2009) since 2009. Based on replicate measurements for deep samples we estimate an error of about 0.3 % for both nutrients.  $\text{NO}_3$  data are not available for all the cruises used in this analysis. The mean  $\text{NO}_3$  concentrations in the LAABW at OISO-ST11 is  $32.8 \pm 1.2 \mu\text{mol.kg}^{-1}$  while the average value derived from the GLODAP-v2 database in bottom waters south of  $50^\circ\text{S}$  in the

South Indian Ocean is  $32.4 \pm 0.6 \mu\text{mol.kg}^{-1}$ . The lack of  $\text{NO}_3$  data for few cruises has been palliated by considering using a climatological value of  $32.43 \mu\text{mol.kg}^{-1}$  with a limited impact on  $C_{\text{ant}}$  determined by the  $C^\circ$  method ( $<2 \mu\text{mol.kg}^{-1}$  on estimates based on the differences observed between  $\text{NO}_3$  measurements and the climatological value).

### 3.43 $C_{\text{ant}}$ calculation using the TrOCA method

The TrOCA method was first presented by Touratier and Goyet (2004a, b) and revised by Touratier et al. (2007). Following the concept of the quasi-conservative tracer NO (Broecker, 1974), TrOCA is a tracer defined as a combination of  $\text{O}_2$ ,  $C_T$  and  $A_T$ , following:

$$\text{TrOCA} = \text{O}_2 + a \left( C_T - \frac{1}{2} A_T \right), \quad (1)$$

where  $a$  is defined in Touratier et al. (2007) as combination of the Redfield equation coefficients for  $\text{CO}_2$ ,  $\text{O}_2$ ,  $\text{HPO}_4^{2-}$  and  $\text{H}^+$ . For more details about the definition and the calibration of this parameter, please refer to Touratier et al. (2007). The temporal change in TrOCA is independent of biological processes and can be attributed to anthropogenic carbon (Touratier and Goyet, 2004a). Therefore,  $C_{\text{ant}}$  can be directly calculated from the difference between TrOCA and its pre-industrial value  $\text{TrOCA}^\circ$ :

$$C_{\text{ant}} = \frac{\text{TrOCA} - \text{TrOCA}^\circ}{a}, \quad (2)$$

where  $\text{TrOCA}^\circ$  is evaluated as a function of  $\theta$  and  $A_T$  (Eq. 3):

$$\text{TrOCA}^\circ = e^{\left[ b - (c) \cdot \theta - \frac{d}{A_T^2} \right]}, \quad (3)$$

In these expressions, coefficients  $a$ ,  $b$ ,  $c$  and  $d$  were adjusted by Touratier et al. (2007) from deep waters free of anthropogenic  $\text{CO}_2$  deep-waters using the tracers  $\Delta^{14}\text{C}$  and CFC-11 from the GLODAPv1 database (Key et al., 2004). The final expression used to calculate  $C_{\text{ant}}$  is:

$$C_{\text{ant}} = \frac{\text{O}_2 + 1.279 \left( C_T - \frac{1}{2} A_T \right) - e^{\left[ 7.511 - (1.087 \cdot 10^{-2}) \cdot \theta - \frac{7.81 \cdot 10^5}{A_T^2} \right]}}{1.279}, \quad (4)$$

The consideration of the errors on the different parameters involved in the TrOCA method results in an uncertainty of  $\pm 6.25 \mu\text{mol.kg}^{-1}$  (mostly due to the parameter  $a$ , leading to  $\pm 3.31 \mu\text{mol.kg}^{-1}$ ). As this error is relatively large compared to the expected  $C_{\text{ant}}$  concentrations in deep and bottom SO waters (Pardo et al., 2014) we will compare the TrOCA results using another indirect method to interpret  $C_{\text{ant}}$  changes over 40 years.

### 3.54 $C_{\text{ant}}$ calculation using the preformed inorganic carbon ( $C^0$ ) method

To support the  $C_{\text{ant}}$  trend determined with the TrOCA method,  $C_{\text{ant}}$  was also estimated using a back-calculation approach noted  $C^0$  (Brewer, 1978; Chen and Millero, 1979), previously adapted for  $C_{\text{ant}}$  estimates along the WOCE-I6 section between South Africa and Antarctica (Lo Monaco et al., 2005a). This method consists in the correction of the measured  $C_T$  for the biological contribution ( $C_{\text{bio}}$ ) and the preindustrial preformed  $C_T$  ( $C_{\text{PI}}^0$ ):

$$C_{\text{ant}} = C_T - C_{\text{bio}} - C_{\text{PI}}^0, \quad (5)$$

$C_{\text{bio}}$  (Eq. 6) depends on carbonate dissolution and organic matter remineralization, taking account of the corrected  $\text{C}/\text{O}_2$  ratio from Kortzinger et al. (2001):

$$C_{\text{bio}} = 0.5 \Delta A_T - (C/\text{O}_2 + 0.5 N/\text{O}_2) \Delta \text{O}_2, \quad (6)$$

Where  $C/O_2 = 106/138$  and  $N/O_2 = 16/138$ .  $\Delta A_T$  and  $\Delta O_2$  are the difference between the measured values ( $A_T$  and  $O_2$ ) and the preformed values ( $A_T^0$  and  $O_2^0$ ).  $A_T^0$  (Eq. 7) has been computed by Lo Monaco et al. (2005a) as a function of  $\Theta$ ,  $S$  and the conservative tracer  $PO$ :

$$A_T^0 = 0.0685PO + 59.79S - 1.45\Theta + 217.1, \quad (7)$$

$PO$  (Eq. 8) has been defined by Broecker (1974) and depends on the equilibrium of  $O_2$  with phosphate ( $PO_4$ ). When  $PO_4$  data are not available, nitrate ( $NO_3$ ) can be used instead as follows (the N/P ratio of 16 is from Anderson and Sarmiento, 1994):

$$PO = O_2 + 170PO_4 = O_2 + (170/16)NO_3, \quad (8)$$

To determine  $O_2^0$ , it is assumed that the surface water is in full equilibrium with the atmosphere ( $O_2^0 = O_{2,sat}$ ; Benson and Krause, 1980) and that after subduction  $O_2$  in a given water mass is only impacted by the biological activity (Weiss, 1970). A correction of  $O_2^0$  has been proposed by Lo Monaco et al. (2005a) to take account of the undersaturation of  $O_2$  due to sea-ice cover at high latitudes.  $O_2^0$  is, therefore, corrected by assuming a mean mixing ratio of the ice-covered surface waters  $k=50\%$  (Lo Monaco et al., 2005a), and a mean value for  $O_2$  undersaturation in ice-covered surface waters  $\alpha = 12\%$  (Anderson et al., 1991) according to Eq. 9:

$$\Delta O_2 = (1 - \alpha k)O_{2,sat} - O_2 = AOU, \quad (9)$$

$C_{PI}^0$  in equation 5 is a function of the current preformed  $C_T$  ( $C_{obs}^0$ ) and a reference water term (Eq. 10):

$$C_{PI}^0 = C_{obs}^0 + [C_T - C_{bio} - C_{obs}^0]_{REF}, \quad (10)$$

$C_{0,obs}$  has been computed similarly as  $A_T^0$  (Eq. 11):

$$C_{obs}^0 = -0.0439PO + 42.79S - 12.02\Theta + 739.8, \quad (11)$$

Where the reference water term is a constant for a given time of observation, corresponding to the time when  $C_{obs}^0$  is parameterized. In this paper, we used the parameterization given by Lo Monaco et al., (2005a) and their estimated value for the reference term of  $51 \mu\text{mol.kg}^{-1}$ . This number has been computed using an optimum multiparametric (OMP) model to estimate the mixing ratio of the North Atlantic deep water in the SO (used as reference water, i.e. old water mass where  $C_{ant} = 0$ ). For more details about the  $C^0$  method, which has a final error of  $\pm 6 \mu\text{mol.kg}^{-1}$ , please see Lo Monaco et al. (2005a).

## 4 Results

The vertical distribution of hydrological and biogeochemical properties observed in deep and bottom waters and their evolution over the last 40 years are displayed in Fig 2. The LCDW layer ( $\gamma^n = 28.15\text{-}28.27 \text{ kg.m}^{-3}$ ) is characterized by minimum  $O_2$  concentrations (Fig. 2c), higher  $C_T$  (Fig. 2b) and lower  $C_{ant}$  concentrations than in the AABW (Fig. 2a).  $C_{ant}$  concentrations were not significant in the LCDW until the end of the 1990s ( $<6 \mu\text{mol.kg}^{-1}$ ), then our data show an increase in  $C_{ant}$  between the two 1998 reoccupations, followed by relatively constant  $C_{ant}$  concentrations ( $10 \pm 3 \mu\text{mol.kg}^{-1}$ ). In the LAABW ( $\gamma^n > 28.35 \text{ kg.m}^{-3}$ ), well identified by low  $\Theta$ , low  $S$  and high  $O_2$ ,  $C_{ant}$  concentrations are higher than in the overlying UAABW and LCDW (Fig. 2a). The evolutions of the mean properties in the LAABW over 40 years are shown in Fig. 3. In this layer,  $C_{ant}$  concentrations increased from  $5 \pm 4 \mu\text{mol.kg}^{-1}$  in 1978 and  $7 \pm 4 \mu\text{mol.kg}^{-1}$  in the mid-1980s to  $13 \pm 2 \mu\text{mol.kg}^{-1}$  at the end of the 1990s and up to  $19 \pm 2 \mu\text{mol.kg}^{-1}$  in 2004 (Fig. 3a). Figure 3a also shows a very good agreement between the TrOCA method and the  $C^0$  method for both the magnitude and variability of  $C_{ant}$  in the LAABW. Our results show a mean  $C_{ant}$  trend in the LAABW of  $+1.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over the full period and a maximum trend of the order of  $+5.2 \mu\text{mol.kg}^{-1}.$

<sup>1</sup>.decade<sup>-1</sup> over 1987-2004 (Table 2). Due to the mixing of AABW with old CDW ( $C_{ant}$  free), these trends are lower than the theoretical trend expected from the increase in atmospheric  $CO_2$ . Indeed, assuming that the surface ocean  $fCO_2$  follows the atmospheric growth rate (+1.8  $\mu atm \cdot year^{-1}$  over 1978-2018) in the seasonal ice zone (Roden et al., 2016), the theoretical  $C_{ant}$  trend at the AABW formation sites would be of the order of +8  $\mu mol \cdot kg^{-1} \cdot decade^{-1}$  in the Antarctic surface water. This is close to the theoretical  $C_T$  trend estimated for freezing shelf water in the Weddell Sea (van Heuven et al 2014). These trends are lower than the theoretical trend expected from the increase in atmospheric  $CO_2$ . Indeed, assuming that the surface ocean  $fCO_2$  follows the atmospheric growth rate (+1.8  $\mu atm \cdot year^{-1}$  over 1978-2018), the theoretical  $C_{ant}$  trend at the AABW formation sites would be of the order of +8  $\mu mol \cdot kg^{-1} \cdot decade^{-1}$ . The observed slow  $C_{ant}$  trends can be partly explained by the transit time for AABW to reach our study site and the mixing of AABWs with older LCDW that contain less  $C_{ant}$  over their transit (Fig. 2a).

#### Figure 2

Over the full period,  $C_T$  increased by  $2.0 \pm 0.5 \mu mol \cdot kg^{-1} \cdot decade^{-1}$ , mostly due to the accumulation of  $C_{ant}$  (Table 2). Our data also show a significant decrease in  $O_2$  concentrations by  $0.8 \pm 0.4 \mu mol \cdot kg^{-1} \cdot decade^{-1}$  over the 40-years period (Fig. 3c, Table 2) that could be caused by reduced ventilation, as suggested by Schmidtko et al. (2017) who observed significant  $O_2$  loss in the global ocean. In the deep Indian SO sector, these authors found a trend approaching -1  $\mu mol \cdot kg^{-1} \cdot decade^{-1}$  over 50 years (1960-2010), which is consistent with our data. We did not detect any significant trend in  $A_T$ ,  $\theta$  and  $S$  over the full period, but on shorter periods our data show a significant decrease in  $A_T$ . The low  $A_T$  values observed over 2000-2004 (Fig. 3d) could suggest reduced calcification in the upper ocean leading to less sinking of calcium carbonate tests and a decrease in  $A_T$  in deep and bottom waters over this period (Fig. 2d). For this period the increase in  $C_T$  was lower than the accumulation of  $C_{ant}$ , but such feature is disputable in view of the uncertainty on the  $C_{ant}$  calculation. This event is followed by an increase in the 'natural' component of  $C_T$  ( $C_{nat}$ , calculated as the difference between  $C_T$  and  $C_{ant}$ ) since 2004 associated to a decrease in  $O_2$  and no increase in  $C_{ant}$  (Table 2). These trends were not associated with a significant trend in  $\theta$  or  $S$  (Fig. 3e,f, Table 2). The increase in  $C_{nat}$  is thus unlikely originating from increased mixing with LCDW during bottom waters transport, confirming that our LAABW definition exclude mixing with the LCDW. Enhanced organic matter remineralization is also unlikely since  $NO_3$  did not show any significant trend (Table 2).

#### Table 2

#### Figure 3

Importantly, our data show substantial interannual variations in LAABW properties, which could significantly impact the trends estimated from limited reoccupations (e.g. Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019). For example, we found relatively higher  $C_{ant}$  concentrations in 1985 (10  $\mu mol \cdot kg^{-1}$ ) compared to 1978 (5  $\mu mol \cdot kg^{-1}$ ) and 1987 (7  $\mu mol \cdot kg^{-1}$ ). This is linked to a signal of low  $S$  in 1985 (Fig. 3f) that could be due to a larger contribution of fresher waters such as the WSDW or CDBW. This could also be related to the different sampling locations. Over the last decade (2009-2018), our data show large and rapid changes in  $S$  that are partly reflected on  $C_T$  and  $O_2$ , and that could explain the relatively low  $C_{ant}$  concentrations observed over this period. Indeed, the  $S$  maximum observed in 2012 (correlated to higher  $\theta$ ) is associated with a marked  $C_T$  minimum (surprisingly almost as low as in 1987), as well as low  $A_T$  (hence low  $C_{Tnat}$ ), and low  $NO_3$  concentrations. Since these anomalies were associated with a decrease in  $C_{ant}$  concentrations, one may argue for an increased contribution of bottom waters ventilated far away from our study site. A few years later our data show a  $S$  minimum (correlated to lower  $\theta$ ), associated with a rapid increase in  $C_T$  and a rapid decrease in  $O_2$  between 2013 and 2016, suggesting



the contribution of a closer AABW type such as the CDBW. The freshening of  $-0.006 \text{ decade}^{-1}$  in S between 2004 and 2018 that we observed on the western side of the Kerguelen Plateau was also observed on the eastern side of the Plateau by Menezes et al. (2017) over a similar period. In this region, Menezes et al. (2017) evaluated a change in S by about  $-0.008 \text{ decade}^{-1}$  from 2007 to 2016 (against  $-0.002 \text{ decade}^{-1}$  between 1994 and 2007), suggesting an acceleration of the AABW freshening in recent years. However, they also reported a warming by  $+0.06 \text{ }^{\circ}\text{C}.\text{decade}^{-1}$ , while we observed cooler temperature in 2016-2018. This suggests that we sampled a different mixture of AABWs.

Figure 4

## 5 Discussion

### 5.1 LAABW composition at OISO-ST11

At each formation site, AABWs experience significant temporal property changes, mostly recognized at decadal scale (e.g. freshening in the South Indian Ocean, Menezes et al., 2017) with potential impact on carbon uptake and  $C_{\text{ant}}$  concentrations during AABW formation (Shadwick et al., 2013). The  $\Theta$ -S diagram constructed from yearly averaged data in bottom waters (Fig. 4) shows that the LAABW at OISO-ST11 is a complex mixture of WSDW, CDBW, RSBW and ALBW. The coldest type of LAABW was observed at the GEOSECS station at  $60^{\circ}$  S ( $-0.56^{\circ}\text{C}$ ), while the warmer type of LAABW observed at the INDIGO-1 station at  $53^{\circ}$  S ( $-0.44^{\circ}\text{C}$ ). These extreme  $\Theta$  values could be a natural feature or may be related to specific sampling. For the other cruises,  $\Theta$  in LAABW ranges from  $-0.51$  to  $-0.45^{\circ}\text{C}$  with no clear indication on the specific AABW origin. The S range observed in the bottom waters at OISO-ST11 (34.65-34.67) illustrates either changes in mixing with various AABW sources or temporal variations at the formation site. Given the knowledge of deep and bottom waters circulation and characteristics (Fig. 1 and 4) and the significant  $C_{\text{ant}}$  concentrations that we calculated in the LAABW (Fig. 3a), the main contribution at our location is likely the younger and colder CDBW for which relatively high  $C_{\text{ant}}$  concentrations have been recently documented (Roden et al., 2016). From its formation region, the CDBW can either flow westward with the CAC or flow northward in the Enderby Basin (Ohshima et al., 2013, Fig. 1). In the CAC branch, the CDBW mixes with the LCDW along the Antarctic shelf and the continental slope between  $80^{\circ}$  E and  $30^{\circ}$  E (Meijers et al., 2010; Roden et al., 2016). On the western side of the Kerguelen Plateau, CDBW also mixes with RSBW and ALBW (Orsi et al., 1999; Van Wijk and Rintoul, 2014). In this context, the  $C_{\text{ant}}$  concentrations observed in the bottom layer at OISO-ST11 are probably not linked to one single AABW source, but are likely a complex interplay of AABWs from different sources with different biogeochemical properties.

### 5.2 $C_{\text{ant}}$ concentrations

In order to compare our  $C_{\text{ant}}$  estimates with other studies, we separated the 40-years time-series into 3 periods: the first period (1978-1987) corresponds to historical data when  $C_{\text{ant}}$  is expected to be low; the second period (1998-2004) starts when the first OISO cruise was conducted (and using CRMs for  $A_T$  and  $C_T$  measurements) and **lasts ends** when  $C_{\text{ant}}$  concentrations in the LAABW are maximum (Fig. 3a); the third period consists in the observations performed in late 2009 to 2018 when the observed variations are relatively large for S and small for  $C_{\text{ant}}$ . The mean  $C_{\text{ant}}$  concentrations for each period are 7, 14 and  $13 \mu\text{mol.kg}^{-1}$ , respectively, which is consistent with the results from other studies (Table 3). The  $C_{\text{ant}}$  values for 1978-1987 can hardly be compared to other studies because very

few observations were conducted in the 1980s in the ~~SO~~ Indian sector ~~of the SO~~ (Sabine et al., 1999) and because of potential biases for historical data despite their careful ~~qualification-quality control~~ in GLODAP and CARINA (Key et al., 2004; Lo Monaco et al., 2010; Olsen et al., 2016). In addition, the different methods used to estimate  $C_{\text{ant}}$  can lead to different results, especially in deep and bottom waters of the SO (Vázquez-Rodríguez et al., 2009). Overall, Table 3 confirms that  $C_{\text{ant}}$  concentrations were low in the 1970s and 1980s, and reached values of the order of  $10 \mu\text{mol.kg}^{-1}$  in the 1990s, a signal not clearly captured in global data-based estimates (Gruber, 1998; Sabine et al., 2004; Waugh et al., 2006; Khatiwala et al., 2013). The observations presented in this analysis, although regional, offer a complement to recent estimates of  $C_{\text{ant}}$  changes evaluated between 1994 and 2007 in the top 3000 m for the global ocean (Gruber et al., 2019a). In the Enderby Basin at the horizon 2000-3000 m, the accumulation of  $C_{\text{ant}}$  from 1994 to 2007 is not uniform and ranges between 0 and  $8 \mu\text{mol.kg}^{-1}$  (Gruber et al., 2019a). At our station, in the LCDW (2000-3000\_m) the  $C_{\text{ant}}$  concentrations were not significant in 1978-1987 ( $-2$  to  $5 \mu\text{mol.kg}^{-1}$ ) but increase to an average of  $9 \pm 3 \mu\text{mol.kg}^{-1}$  in 1998-2018 (Fig. 2a), probably due to mixing with AABWs that contain more  $C_{\text{ant}}$ . Interestingly, this value is close but in the high range of the  $C_{\text{ant}}$  accumulation estimated from 1994 to 2007 in deep waters of the south Indian Ocean (Gruber et al., 2019a). Not surprisingly, high  $C_{\text{ant}}$  concentrations are detected in the AABW formation regions (Table 3). The highest  $C_{\text{ant}}$  concentrations in bottom waters (up to  $30 \mu\text{mol.kg}^{-1}$ ) were observed in the ventilated shelf waters in the Ross Sea (Sandrini et al., 2007). In the Adélie and Mertz Polynya regions, Shadwick et al. (2014) observed high  $C_{\text{ant}}$  concentrations in the subsurface shelf waters ( $40-44 \mu\text{mol.kg}^{-1}$ ) but lower values in the ALBW ( $15 \mu\text{mol.kg}^{-1}$ ) due to mixing with older LCDW. In WSBW, all  $C_{\text{ant}}$  concentrations estimated from observations between 1996 and 2005 and with the TrOCA method (Table 3) lead to about the same values ranging between 13 and  $16 \mu\text{mol.kg}^{-1}$  (Lo Monaco et al., 2005b; van Heuven et al., 2011). In bottom waters formed near the Cape Darnley (CDBW), Roden et al. (2016) estimated high  $C_{\text{ant}}$  concentrations in bottom waters ( $25 \mu\text{mol.kg}^{-1}$ ) resulting from the shelf waters that contain very high amounts of  $C_{\text{ant}}$  ( $50 \mu\text{mol.kg}^{-1}$ ). The comparison with other studies confirms that far from the AABW formation sites, contemporary  $C_{\text{ant}}$  concentrations are not exceeding  $16 \mu\text{mol.kg}^{-1}$  on average.

Table 3.

### 5.3 $C_{\text{ant}}$ trends and variability

Comparison of long-term  $C_{\text{ant}}$  trends in deep and bottom waters of the SO is limited to very few regions where repeated observations are available. To our knowledge, only 3 other studies evaluated the long-term  $C_{\text{ant}}$  trends in the SO based on more than 5 reoccupations: in the South-western Atlantic (Rios et al., 2012) and in the Weddell Gyre along the Prime meridian section (van Heuven et al., 2011, 2014). Temporal changes of  $C_T$  and  $C_{\text{ant}}$  have also been investigated in other SO regions, but limited to 2 to 4 reoccupations (Williams et al., 2015; Pardo et al., 2017; Murata et al., 2019). Given the  $C_{\text{ant}}$  variability depicted at our location (Fig. 3a), different trends can be deduced from limited reoccupations. As an example, Murata et al., (2019) evaluated the change in  $C_{\text{ant}}$  from data collected 17 years apart (1994–1996 and 2012–2013) along a transect around  $62^\circ \text{S}$  and found a small increase at our location ( $< 5 \mu\text{mol.kg}^{-1}$  around  $60^\circ \text{E}$ ). This result appears very sensitive to the time of the observation given that we found a minimum in  $C_{\text{ant}}$  concentrations between 2011 and 2014 (Fig. 3a) associated with a marked  $C_T$  minimum (Fig. 3b). In addition, our results show that the detection of  $C_{\text{ant}}$  trends appears very sensitive to the time period considered (Table 2). As an extreme case, the  $C_{\text{ant}}$  trend calculated for the period 1987-2004 is  $+5.2 \mu\text{mol.kg}^{-1}$

<sup>1</sup>.decade<sup>-1</sup> (relatively close to the theoretical  $C_{\text{ant}}$  trend of  $+8 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ), but it reverses to  $-3.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  for the period 2004-2018.

The long-term  $C_T$  trend that we estimated in the LAABW in the eastern Enderby Basin ( $2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) is slightly faster than the  $C_T$  trends estimated in the WSBW in the Weddell Gyre:  $+1.2 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  over the period 1973-2011 and  $+1.6 \pm 1.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  when restricted to 1996-2011 (van Heuven et al., 2014). Along the SR03 line (south of Tasmania) reoccupied in 1995, 2001, 2008 and 2011, Pardo et al. (2017) calculated a  $C_T$  trend of  $+2.4 \pm 0.2 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  in the AABW, composed of ALBW and RSBW in this sector. This is higher than the  $C_T$  trends found at our location and in the Weddell Gyre, but surprisingly, this was not associated with a significant increase in  $C_{\text{ant}}$ . The  $C_T$  trend in AABW along the SR03 section was likely due to the intrusion of old and  $C_T$ -rich waters also revealed by an increase in Si concentrations during 1995-2011 (Pardo et al., 2017). This is a clear example of decoupling between  $C_T$  and  $C_{\text{ant}}$  trends in deep and bottom waters as observed at our location in the last decade (Table 2). For  $C_{\text{ant}}$ , our 40-years trend estimate ( $1.4 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) appears close to the trend reported by Rios et al. (2012) in the south-western Atlantic AABW from 6 reoccupations between 1972 and 2003 ( $+1.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). However, if we limit our result to the period 1978-2002 or 1978-2004 (about the same period as in Rios et al., 2012), our trend is much larger ( $+3-4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). At our location, the  $C_{\text{ant}}$  trend over 40 years ( $+1.4 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ) explains most of the observed  $C_T$  increase ( $+2.0 \pm 0.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ ). The residual of  $+0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  reflects changes in natural processes affecting the carbon content (different AABW sources, ventilation, mixing with deep waters, remineralization or carbonates dissolution). Although this is a weak signal, the natural  $C_T$  change ( $C_{\text{nat}}$ ) mirrors the observed decrease in  $O_2$  by  $-0.8 \pm 0.4 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$ . This  $O_2$  decrease detected in the Enderby Basin appears to be a real feature that was documented at large scale for 1960-2010 in deep SO basins (Schmidtke et al. 2017), suggesting that the changes observed at ~~56.5°S/63°E~~~~56.5°S~~ are related to large-scale processes, possibly due to a decrease in AABW formation (Purkey and Johnson, 2012).

#### 5.4 Recent $C_{\text{ant}}$ stability

Although most studies suggest a gradual accumulation of  $C_{\text{ant}}$  in the AABW, our time-series highlights significant multi-annual changes, in particular over the last decade when  $C_{\text{ant}}$  concentrations were as low as around the year 2000 (Fig. 3a) and decoupled from the increase in  $C_T$  (Fig. 3b). This result is difficult to interpret because at our location, away from AABW sources (Fig. 1), the temporal variability observed in the LAABW layer can result from many remote processes occurring at the AABW formation sites (such as wind forcing, ventilation, sea-ice melting, thermodynamic, biological activity and air-sea exchanges). Additionally, internal processes during the transport of AABW<sub>s</sub> (such as organic matter remineralization, carbonate dissolution and mixing with surrounding waters) must also be taken into account. The apparent steady  $C_{\text{ant}}$  feature suggests that AABW<sub>s</sub> found at our location has stored less  $C_{\text{ant}}$  in recent years. This might be linked to reduced  $\text{CO}_2$  uptake in the AABW formation regions, as recognized at large-scale in the SO from the late 1980s to 2001 (Le Quéré et al., 2007; Metzl, 2009; Lenton et al., 2012; Landschützer et al., 2015). This large-scale response in the SO during a positive trend in the Southern Annular Mode (SAM) is mainly associated to stronger winds driven by accelerating greenhouse gas emissions and stratospheric ozone depletion, leading to warming and freshening in the SO (Swart et al., 2018), change in the ventilation of the  $C_T$ -rich deep waters and reduced  $\text{CO}_2$  uptake (Lenton et al., 2009). The reconstructed  $\text{pCO}_2$  fields by Landschützer et al. (2015) suggest that the reduced  $\text{CO}_2$  sink in the 1990s is identified

at high latitudes in the SO (see Fig. 2a and S9 in Landschützer et al., 2015). However, as opposed to the circumpolar open ocean zone (e.g. Metzl, 2009; Takahashi et al., 2009, 2012; Munro et al., 2015; Fay et al., 2018), the long-term trend of surface  $f\text{CO}_2$  and carbon uptake deduced from direct observations are not clearly identified in the seasonal ice zone (SIZ) and shelves around Antarctica, and thus in the AABW formation regions of interest to interpret our results (Laruelle et al., 2018). There, surface  $f\text{CO}_2$  data are sparse, especially before 1990, and cruises were mainly conducted in austral summer when the spatio-temporal  $f\text{CO}_2$  variability is very large and driven by multiple processes at regional or small scales, such as primary production, sea-ice formation and retreat, and water circulation and mixing. This leads to various estimates of the air-sea  $\text{CO}_2$  fluxes around Antarctica depending on the region and period and large uncertainty when attempting to detect long-term trends (Gregor et al., 2018).

In particular, in polynyas and AABW formation regions where  $f\text{CO}_2$  is low and where katabatic winds prevail, very strong instantaneous  $\text{CO}_2$  sink can occur at the local scale (up to  $-250 \text{ mmol C.m}^{-2}.\text{d}^{-1}$  in Terra Nova Bay in the Ross Sea according to De Jong and Dunbar, 2017). In the Prydz Bay region where CDBW is formed, recent studies show that surface  $f\text{CO}_2$  in austral summer vary over a very large range ( $150\text{--}450 \mu\text{atm}$ ), with the lowest  $f\text{CO}_2$  observed in the shelf region generating very strong local  $\text{CO}_2$  sink ( $-221 \text{ mmol C.m}^{-2}.\text{d}^{-1}$ ; Roden et al. 2016). The carbon uptake was particularly enhanced near Cape Darnley and coincided with the highest  $C_{\text{ant}}$  concentrations that Roden et al. (2016) estimated in the dense shelf waters that subduct to form AABW. In the Prydz Bay coastal region, surface  $f\text{CO}_2$  values in 1993-1995 were as low as  $100 \mu\text{atm}$  (Gibson and Trull, 1999) leading to a strong local  $\text{CO}_2$  uptake of  $-30 \text{ mmol C.m}^{-2}.\text{d}^{-1}$  in summer. In addition, Roden et al. (2013) found a large  $C_T$  increase over 16 years ( $+34 \mu\text{mol.kg}^{-1}$ ) in the Prydz Bay, which is much higher than the anthropogenic signal alone ( $+12 \mu\text{mol.kg}^{-1}$ ) and likely explained by changes in primary production that would have been stronger in 1994. To our knowledge, this is the only direct observation of decadal  $C_T$  change in surface waters in a region of AABW formation (here the Prydz Bay) and it highlights the difficulty not only to evaluate the  $C_T$  and  $C_{\text{ant}}$  long-term trends in these regions but also to separate natural and anthropogenic signals when this water reaches the deep ocean. We attempted to detect long-term changes in  $\text{CO}_2$  uptake in this region using the qualified  $f\text{CO}_2$  data available in the SOCAT database (Bakker et al., 2016), but our estimates (not shown) were highly uncertain due to very large spatial and temporal variability. To conclude, all previous studies conducted near or in AABW formation sites clearly reveal that these regions are potentially strong carbon sinks, but how the sink changed over the last decades is not yet evaluated, and thus we are not able to certify that the recent  $C_{\text{ant}}$  stability that we observed in the LAABW at our location is directly linked to the weakening of the carbon sink that was recognized at large-scale in the SO from the 1980s to mid-2000s (Le Quéré et al., 2007; Landschützer et al., 2015).

Changes in the accumulation of  $C_{\text{ant}}$  in AABW could also be directly related to changes in physical processes occurring in AABW formation regions. Decadal decreasing of sea-ice production and melting of sea-ice have been documented in several regions including Cape Darnley polynyas (Tamura et al., 2016; Williams et al., 2016). The consequent changes in Antarctic surface waters properties are transmitted into the deep ocean, notably the well-recognized freshening of the AABW ~~over the last decades~~ (Rintoul, 2007; Rintoul, 2007; Anilkumar et al., 2015). The warming of bottom waters was also documented in the Enderby basin (Couldrey et al., 2013) as well as at a larger scale in all deep SO basins (Purkey and Johnson, 2010; Desbruyères et al., 2016). Associated to a decrease in AABW formation in the 1990s (Purkey and Johnson, 2012), these physical changes could explain the recent stability of  $C_{\text{ant}}$  concentrations in AABW observed at our location. As AABWs from different sources spread and mix with  $C_T$ -rich deep waters before reaching our location (Fig. 1), less AABW formation and export would result

in an increase in  $C_T$  (increase in  $C_{nat}$ ) not associated with an increase in  $C_{ant}$ , and a decrease in  $O_2$  (as observed in recent years in Fig. 3a,b,c). Finally, it is also possible that the LAABW observed in recent years at our location is the result of a larger contribution of older RSBW, ALBW or even WSBW that have lower  $C_{ant}$  and  $O_2$  concentrations compared to CDBW formed at Cape Darnley and Prydz Bay.

## 6 Conclusion

The distribution and evolution of  $C_{ant}$  in the bottom layer of the SO are related to complex interactions between climatic forcing, air-sea  $CO_2$  exchange at formation sites, as well as biological and physical processes during AABW<sub>s</sub> circulation. The dataset that we collected regularly in the Enderby basin over the last 20 years (1998-2018) in the frame of the OISO project, together with historical observations obtained in 1978, 1985 and 1987 (GEOSECS and INDIGO cruises), allows the investigation of  $C_{ant}$  changes in AABW over 40 years in this region. The focus on the AABW variability is made by defining a Lower Antarctic Bottom Water (LAABW) as described in the Section 2.3. Our results suggest that the accumulation of  $C_{ant}$  explains most, but not all, of the observed increase in  $C_T$ . We also detected a decrease in  $O_2$  that is consistent with the large-scale signal reported by Schmidtko et al. (2017), possibly due to a decrease in AABW<sub>s</sub> formation (Purkey and Johnson, 2012). Our data further indicate rapid anomalies in some periods suggesting that for decadal to long-term estimates care have to be taken when analyzing the change in  $C_{ant}$  from data sets collected 10 or 20 years apart (e.g. Williams et al., 2015; Murata et al., 2019). Our results also show different  $C_{ant}$  trends on short periods, with a maximum increase of  $6.5 \mu\text{mol.kg}^{-1}.\text{decade}^{-1}$  between 1987 and 2004 and an apparent stability in the last 20 years (despite an increase in  $C_T$ ). This suggests that AABW<sub>s</sub> have stored less  $C_{ant}$  in the last decade, but our understanding of the processes that explain this signal is not clear. This might be the result of the reduced  $CO_2$  uptake in the SO in the 1990s (Le Quéré et al., 2007; Landschützer et al., 2015), but this is not yet verified from direct  $C_T$  or  $fCO_2$  observations in AABW formation regions due to the lack of winter data and very large variability during summer. This calls for more data collection and investigations in these regions. The apparent stability of  $C_{ant}$  in the LAABW since 1998 could also be directly linked to a decrease in AABW<sub>s</sub> formation in the 1990s (Purkey and Johnson, 2012) or a change in the contributions of AABW<sub>s</sub> from different sources, especially in the Prydz Bay region (Williams et al., 2016). In these scenarios, an increased contribution of  $C_T$ -rich and  $O_2$ -poor older LCDW along AABW<sub>s</sub> transit would also explain the decoupling between  $C_{ant}$  and  $C_T$  (increase in  $C_{nat}$ ) and decrease in  $O_2$  concentrations observed in recent years, even if we tried to isolate this specific feature in our data selection. The decoupling between  $C_{ant}$  and  $C_T$  is not a unique feature, as it was also reported along the SR03 section between Tasmania and Antarctica, most probably due to advection of  $C_T$ -rich waters (Pardo et al., 2017). This highlights the importance of the ocean circulation in influencing the temporal  $C_T$  and  $C_{ant}$  inventories changes (De Vries et al., 2017) and the need to better separate anthropogenic and natural variability based on time-series observations. The evaluation and understanding of decadal  $C_{ant}$  changes in deep and bottom ocean waters are still challenging, as the  $C_{ant}$  concentrations remain low compared to  $C_T$  measurements accuracy (at best  $\pm 2 \mu\text{mol.kg}^{-1}$ , Bockmon and Dickson, 2015) and uncertainties of data-based methods ( $\pm 6 \mu\text{mol.kg}^{-1}$ ). Long-term repeated and qualified observations (at least 30 years) are needed to accurately detect and separate the anthropogenic signal from the internal ocean variability; we thus only start to document these trends that should now help to identify shortcomings in models regarding the carbon storage in the deep SO (e.g. Frölicher et al., 2014). As changes in

the SO (including warming, freshening, oxygenation/deoxygenation, CO<sub>2</sub> and acidification) are expected to accelerate in the future in response to anthropogenic forcing and climate change (e.g. Heuzé et al., 2014; Hauck et al., 2015; Ito et al., 2015, Yamamoto et al., 2015), it is important to maintain time-series observations to complement the GO-SHIP strategy, and to occupy more regularly other sectors of the SO (Rintoul et al., 2012). In this context, we hope to maintain our observations in the Southern Indian Ocean in the next decade, and with ongoing synthetic products activities such as GLODAPv2 (Olsen et al., 2016, 2019), SOCAT (Bakker et al., 2016) and more recently the SOCCOM project (Williams et al., 2018), to offer a solid database to validate ocean biogeochemical models and coupled climate/carbon models (Russell et al. 2018), and ultimately reduce uncertainties in future climate projections.

#### 1125 Data availability

1126 GEOSECS, INDIGO and OISO 1998-2011 data are publicly available at the Ocean Carbon Data System (OCADS;  
1127 [https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2\\_2019](https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2_2019)). OISO original data are available at:  
1128 [www.nodc.noaa.gov/ocads/oceans/RepeatSections/clivar\\_oiso.html](https://www.nodc.noaa.gov/ocads/oceans/RepeatSections/clivar_oiso.html). OISO 2012-2018 will be available in  
1129 GLODAPv2\_2021.

#### 1130 Author contributions

1131 LM, CLM, NM, JF and CM performed the sampling and carried out the measurements of the OISO data. LM  
1132 prepared the manuscript with contributions from CLM and NM.

#### 1133 Competing interests

1134 The authors declare that they have no conflict of interest.

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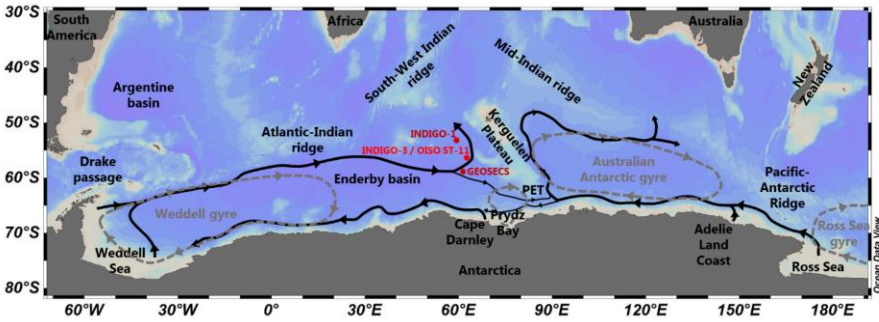
Table 1. List of the cruises used in this study.

Cruise	Station	Location	Year	Month
GEOSECS	430	61.0°E / 60.0°S	1978	February
INDIGO-1	14	58.9°E / 53.0°S	1985	March
INDIGO-3	75	63.2°E / 56.5°S	1987	January
OISO-01	11	63.0°E / 56.5°S	1998	February
OISO-03	11	63.0°E / 56.5°S	1998	December
OISO-05	11	63.0°E / 56.5°S	2000	August
OISO-06	11	63.0°E / 56.5°S	2001	January
OISO-08	11	63.0°E / 56.5°S	2002	January
OISO-11	11	63.0°E / 56.5°S	2004	January
OISO-18	11	63.0°E / 56.5°S	2009	December
OISO-19	11	63.0°E / 56.5°S	2011	January
OISO-21	11	63.0°E / 56.5°S	2012	February
OISO-23	11	63.0°E / 56.5°S	2014	January
OISO-26	11	63.0°E / 56.5°S	2016	October
OISO-27	11	63.0°E / 56.5°S	2017	January
OISO-28	11	63.0°E / 56.5°S	2018	January

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Figure 1. The AABWs circulation rough transport paths from the literature (Fukamachi et al., 2010; Orsi et al., 1999; Carter et al., 2008; Fukamachi et al., 2010; Williams et al., 2010; Vernet et al., 2019) and this study, with geographic indications (black text), main SO currents-gyres (blue-dark yellow text and dash lines for the approximative boundaries/locations) and stations considered in this study (red text and dots). PET: Princess Elizabeth Trough. Figure produced with ODV (Schlitzer et al., 2019).

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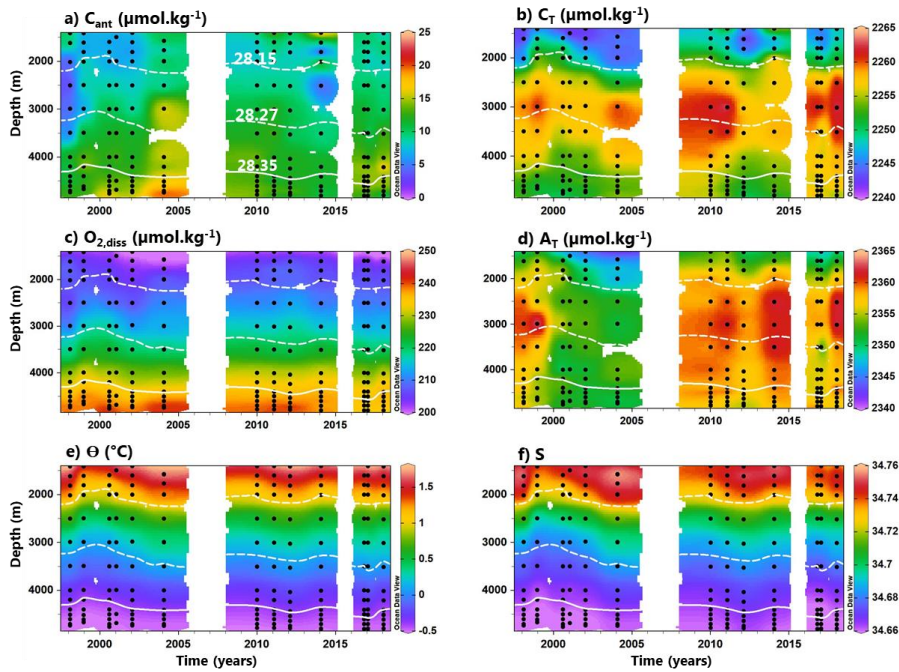


Figure 2. Hovmöller section diagram of (a)  $C_{ant}$  via TrOCA, (b)  $C_T$ , (c)  $O_2$ , (d)  $A_T$ , (e)  $\theta$  and (f)  $S$  based on the OISO data presented in Table 1. Data points are represented by black dots. The white isolines represent the water masses separation by  $\gamma^n$  (from the bottom: LAABW, UAABW and LCDW). Figure produced with ODV (Schlitzer et al., 2019).

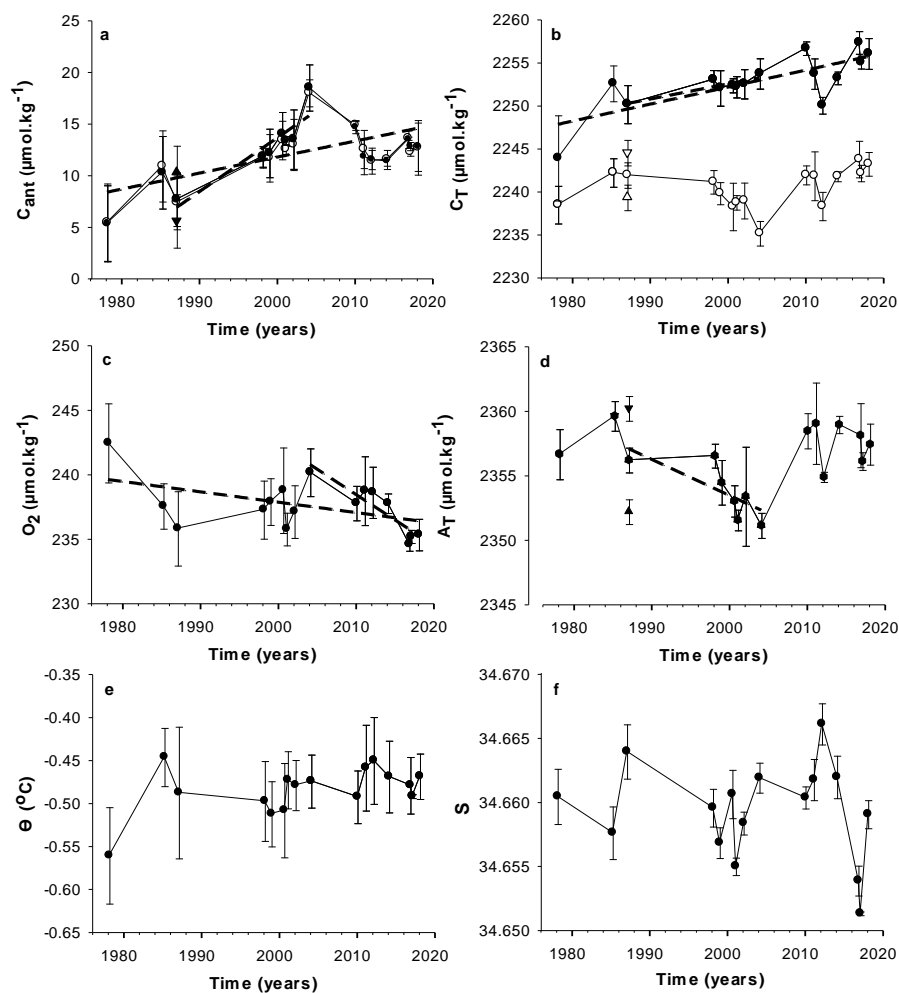


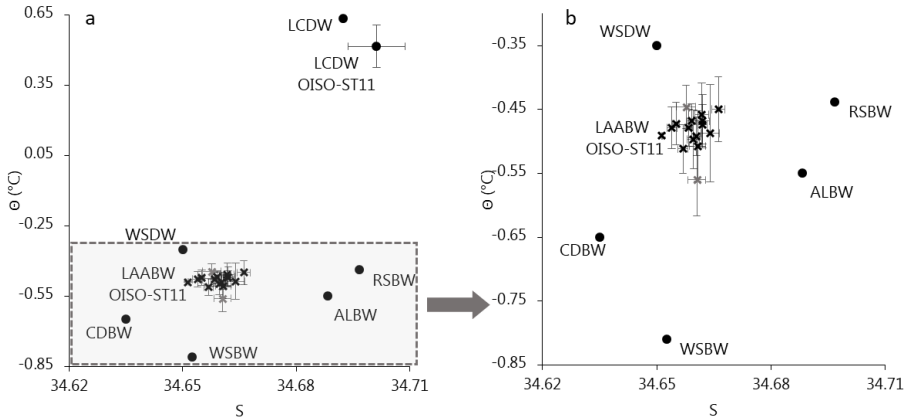
Figure 3. Interannual variability (dash lines lines) and significant trends (at 95 %, see Table 2; dotted lines) for the 40 years of observation of the OISO-ST11 LAABW properties, including (a)  $C_{ant}$  by the TrOCA (black circles and triangles) and the  $C^0$  (open circles) method, (b)  $C_T$  (black circles) and  $C_{nat}$  (open circles), (c)  $O_2$ , (d)  $A_T$ , (e)  $\Theta$  and (f)  $S$ . For (a)  $C_{ant}$ , (b)  $C_{nat}$  and (d)  $A_T$ , the triangles pointing down and up correspond to INDIGO-3 value without and with  $-8 \mu\text{mol.kg}^{-1}$  of correction on the  $A_T$ , respectively (see Supp. Mat. for more details).

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**Table 2: Trends (per decade) of observed and calculated properties in the LAABW estimated over different periods (in bold: significant trends at 95 % confidence level).**

Period	S	$\Theta$ °C	Si $\mu\text{mol.kg}^{-1}$	$\text{NO}_3$ $\mu\text{mol.kg}^{-1}$	$\text{O}_2$ $\mu\text{mol.kg}^{-1}$	$\text{Ar}$ $\mu\text{mol.kg}^{-1}$	$\text{C}_\text{T}$ $\mu\text{mol.kg}^{-1}$	$\text{C}_\text{ant TrOCA}$ $\mu\text{mol.kg}^{-1}$
1978-2018	-0.001 $\pm$ 0.001	0.01 $\pm$ 0.01	-1.2 $\pm$ 0.9	0.2 $\pm$ 0.2	<b>-0.8 <math>\pm</math> 0.4</b>	-0.1 $\pm$ 0.1	<b>2.0 <math>\pm</math> 0.5</b>	<b>1.4 <math>\pm</math> 0.5</b>
1987-2018	-0.001 $\pm$ 0.001	0.01 $\pm$ 0.01	-1.9 $\pm$ 1.4	0.3 $\pm$ 0.4	-0.3 $\pm$ 0.5	0.6 $\pm$ 0.1	<b>1.6 <math>\pm</math> 0.5</b>	1.1 $\pm$ 0.8
1987-2004	-0.003 $\pm$ 0.002	0.01 $\pm$ 0.01	<b>-6.5 <math>\pm</math> 1.8</b>	0.9 $\pm$ 0.9	1.7 $\pm$ 1.0	<b>-1.9 <math>\pm</math> 1.1</b>	<b>1.8 <math>\pm</math> 0.4</b>	<b>5.2 <math>\pm</math> 1.1</b>
2004-2018	-0.006 $\pm$ 0.003	0.01 $\pm$ 0.01	-1.8 $\pm$ 4.5	-0.5 $\pm$ 1.0	<b>-3.9 <math>\pm</math> 0.7</b>	3.4 $\pm$ 0.2	1.7 $\pm$ 1.9	-3.5 $\pm$ 1.5

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**Figure 4. (a) Full  $\Theta$ -S diagram of studied water masses and (b) zoomed on bottom waters. Values are from literature for the WSBW (Fukamachi et al., 2010; van Heuven, 2013; Pardo et al., 2014; Robertson et al., 2002), the WSDW (Carmack and Foster, 1975; Fahrbach et al., 1994; van Heuven, 2013; Robertson et al., 2002), the RSBW (Fukamachi et al., 2010; Gordon et al., 2015; Johnson, 2008; Pardo et al., 2014), the ALBW (Fukamachi et al., 2010; Johnson, 2008; Pardo et al., 2014), the CDBW (Ohshima et al., 2013) and the LCDW (Lo Monaco et al., 2005a; Pardo et al., 2014; Smith and Treguer, 1994), and from the OISO-ST11 dataset for the OISO-ST11 LAABW and OISO-ST11 LCDW. Error bars are calculated from the individual annual averaged values for the OISO-ST11 LAABW and from all data for the OISO-ST11 LCDW. For the OISO-ST11 LAABW, the grey cross are the GEOSECS (lowest  $\Theta$ ) and INDIGO-1 (highest  $\Theta$ ) values.**

Table 3. Compilation of  $C_{ant}$  sequestration investigations in the AABW<sub>s</sub> ( $\gamma^n \geq 28.25 \text{ kg.m}^{-3}$ ) using the TrOCA method. The  $C_{ant}$  estimation of Pardo et al. (2014) is calculated using theoretical AABW mean composition (with 3% of ALBW) and the carbon data from the GLODAPv1 and CARINA databases. Sandrini et al. (2007) values has been measured at the bottom in the Ross Sea and correspond to recently sink high salinity shelf surface water (HSSW). The mean values published by Roden et al. (2016) for the AABW<sub>s</sub> present WSDW characteristics but can be a mix of CDBW and LCDW.

Source	Location	Water masses considered	Year	$C_{ant}$ $\mu\text{mol.kg}^{-1}$
Pardo et al. (2014) Fig. 5	Averaged AABW composition	WSBW-RSBW-ALBW	1994	12
Lo Monaco et al. (2005b) Fig. 4b	WOCE line I6 (30° E; 50°-70° S)	WSBW CDBW	1996	15 20
Sandrini et al. (2007) Fig. 4a	Ross Sea	HSSW (previous RSBW)	2002/2003	Max. of 30
Shadwick et al. (2014) Table 2	Mertz polynya and Adelie depression	ALBW	2007/2008	15
Roden et al. (2016) Table 2	South Indian ocean (30°-80° E; 60°-69° S)	WSDW-LCDW- CDBW	2006	25
van Heuven et al. (2011) Fig.13	Weddell gyre (0° E; 55°-71°S)	WSBW	2005	16
This study	Enderby basin (56.5° S/63° E)	LAABW (mix of WSDW- CDBW-RSBW- ALBW)	1978-1987	8 ± 3
			1987-1998	10 ± 4
			1987-2004	13 ± 4
			1998-2004	14 ± 2
			2010-2018	13 ± 1
			<b>1978-2018</b>	<b>12 ± 3</b>

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