Comments Referee 1

General comments: This manuscript deals with temporal variations of anthropogenic CO2 in bottom waters in the Southern Ocean. The Southern Ocean is said to take up 40% of anthropogenic CO2 absorbed by the ocean. Thus, investigations of temporal variability of anthropogenic CO2 are very important to evaluate ocean's capacity of absorbing atmospheric CO2, 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 scare. 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 CO2 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 CO2 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_2 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 µmol.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 µmol.kg^{-1} and +10 µmol.kg^{-1} (Fig. S2). Most of the crossovers that suggest a positive offset for INDIGO3 data (between +6 µmol.kg^{-1} and +10 µmol.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 µmol.kg^{-1}). However, at the OISO-ST11, A_T data from the INDIGO3 cruise are also about 8 µmol.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 µmol.kg^{-1} in the revised manuscript (the impact on C_{ant} is +2 µmol.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 CO2 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. O2 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 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 O_2 and AOU is unnecessary. This is a point also noticed by Reviewer 2. Because we are most discussing the O_2 concentration in the manuscript, we suggest to only present O_2 in Figure 3.

Specific comments:

Line 18 (here around 460): "from about +7 µmol 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 µmol.kg^{-1} calculated for the period 1978-1987 to the averaged concentration of 13 µmol.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): "θ, 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.

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

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

Response: this will be added.

Line 160 (here around 628): “θ and S”, spell out here. ´

Response: this will be added.

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

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

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

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

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

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

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 CO2 (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^{-3}) 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 AT 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 µmol.kg^{-1} and +10 µmol.kg^{-1} (Fig. S2). Most of the crossovers that suggest a positive offset for INDIGO3 data (between +6 µmol.kg^{-1} and +10 µmol.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 µmol.kg^{-1}). However, at the OISO-ST11, A_T data from the INDIGO3 cruise are also about 8 µmol.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 µmol.kg^{-1} in the revised manuscript (the impact on Cant is +2 µmol.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

the increase in Cant between February 1998 and December 1998 remains (from < 6 µmol.kg^{-1} to about 10 µmol.kg^{-1}).

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

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

Specific comments

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

Response: these are not changes, but Cant concentrations. The following rephrasing is suggested: 'from the average concentration of 7 µmol.kg^{-1} calculated for the period 1978-1987 to the average concentration of 13 µmol.kg^{-1} for the period 2010-2018.'

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

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

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

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

L95 (here around 541): I can't find a definition of "AAC" anywhere. Please write out and define the first time.

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

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

Response: The sentence will be rephrased as suggested.

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

Response: The full names will be added explicitly.

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

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

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

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

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

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

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

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

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

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

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

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

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

Response: AT and O2 will be defined here.

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

Response: the reviewer is right about this mistake.

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

Response: Initially, only C/O_2 and N/O_2 ratios were involved in the definition of the parameter 'a' (Touratier and Goyet, 2004b; Lo Monaco et al., 2005b). In the latest definition of the method Touratier et al. (2007) presents an upgraded definition of this parameter by combining the Redfield equation coefficients for CO_2, O_2, HPO_4^{2-} and H^+ and the same rules of construction as Broecker (1974) did for tracers NO or PO. Because we want to keep the explanation simple in the manuscript, we suggest to rephrase L184 as follows : 'where a is defined in Touratier et al. (2007) as combination of

the Redfield equation coefficients for CO_2, O_2, HPO_4^{2-} and H^+. For more details about the definition and the calibration of this parameter, please refer to Touratier et al. (2007).'

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

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

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

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

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

Response: we removed the older data form the Hovmoller plot, but the change in C_{ant} in LCDW remains (from <6 µmol.kg^{-1} in Feb 1998 (similar as for the older data) to about 10 µmol.kg^{-1} for the following cruises).

L240 (here around 719): The maximum in Cant in 2004 is one occasion, and followed by five (almost six) years without any data. I would be cautious to over interpret this. However, it co-incides with a maximum in oxygen, which could indicate a ventilation event.

Response: we agree with the referee about being cautious with the measurements in 2004. Indeed the maximum in C_{ant} is due to the maximum in O_2 (not associated with a maximum in C_T).

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

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

L259-260 (here around 742-743): Is it realistic that the increase in CT is lower than the accumulation of Cant?

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

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

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

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

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

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

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

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

Response: The sentence will be corrected as follows: 'The freshening in S of -0.006 decade^{-1} 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.'

L312-313 (here around 823): ". . .(15 umol kg-1) due to mixing with older CDW."

Response: the sentence will be corrected.

L317 (here around 827): "that contain very high amounts of Cant . . ."

Response: the sentence will be correct as suggested.

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

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

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

Response: "Southern Ocean" will be changed to SO.

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

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

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

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

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

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

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

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

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

Response: the sentence will be corrected.

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

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

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

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

Technical comments

L22: This is, however, modulated. . .

Response: the comas will be added.

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

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

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

Response: the comas will be added.

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

Response: the coma will be added.

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

Response: this will be corrected.

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

Response: the coma will be added.

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

Response: the font will be changed.

L220: Change font for "value for".

Response: the font will be changed.

L306: Add a comma: "2018 (Fig 3a), probably . . ."

Response: the coma will be added.

L340: Add a ".": Pardo et al. (2017)

Response: the dot will be added.

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

Response: this will be corrected.

L449: Remove "." for consistency: (e.g. Frölicher et al., 2014).

Response: the coma will be deleted.

L451: References in chronological order.

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

Variability and stability of anthropogenic CO2 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_2 (C_{ant}) but 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_{ant} in the AABW. This investigation is based on regular observations collected at the same location (63° E/56.5° S) in the frame 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 Cape Darnley ~~bottom water~~Bottom Water (CDBW) and the Weddell Sea ~~deep water~~Deep Water (WSDW). Our calculations reveal that C_{ant} concentrations increased significantly in the AABW, from ~~about ±~~the average concentration of 7 µmol.kg^{-1} ~~in~~calculated for the period 1978-1987 to ~~±~~the average concentration of 13 µmol.kg^{-1} ~~in~~for the period 2010-2018. This is comparable to previous estimates in other SO basins, with the exception of bottom waters close to their formation sites where C_{ant} concentrations are about twice as large. Our analysis shows that ~~the~~ total carbon (C_T) and C_{ant} increasing rates in the AABW are about the same over the period 1978-2018, and we conclude that the long-term change in C_T is mainly due to the uptake of ~~anthropogenic CO_2~~C_{ant} in the different formation regions. This is, however, modulated by significant interannual to ~~pluriannual~~multi-annual variability associated with variations in hydrological (~~Θ,~~ potential temperature (Θ), salinity (S~~)~~)) and biogeochemical (C_T, total alkalinity (A_T~~,~~), dissolved oxygen (O_2~~)~~))) properties. A surprising result is the apparent stability of C_{ant} concentrations in recent years despite the increase in C_T and the gradual acceleration of atmospheric CO_2.

The C_{ant} 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 ~~AABW~~AABWs needs to be carefully considered on the extrapolated estimation of C_{ant} sequestration based on sparse observations over several years.

1 Introduction

Carbon dioxide (CO_2) 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_{ant} emissions (Le Quéré et al., 2018; Gruber et al., 2019a). It is widely acknowledged that the Southern Ocean (SO) is responsible for 40

% of the C_{ant} ocean sequestration (Matear, 2001; Orr et al., 2001; McNeil et al., 2003; Gruber et al., 2009; Khatiwala et al., 2009~~; Matear, 2001; McNeil et al., 2003; Orr et al., 2001~~). Ocean C_{ant} uptake and sequestration have the benefit to limit the atmospheric CO_2 increase but also result in a gradual decrease of the ocean pH (Gattuso and Hansson, 2011; Jiang et al., 2019). Understanding the oceanic C_{ant} sequestration and its variability is of major importance to predict future atmospheric CO_2 concentrations, impact on the climate and impact of the pH change on marine ecosystems (de Baar, 1992; Orr et al., 2005; Ridgwell and Zeebe, 2005).

C_{ant} in seawater cannot be measured directly and the evaluation of the relatively small C_{ant} signal from the total inorganic dissolved carbon (C_T; around 3 %; Pardo et al, 2014) is still a challenge to overcome. Different approaches have been developed in the last 40 years to quantify C_{ant} concentrations in the oceans. The 'historical' back calculation method based on C_T measurement and preformed inorganic carbon estimate (C^0) was independently published by Brewer (1978) and Chen and Millero (1979). This method has been often applied at regional and basin scale (Chen, 1982~~, 1993~~; Poisson and Chen, 1987; Chen, 1992; Goyet et al., 1998; Körtzinger et al., 1998, 1999~~; Poisson and Chen, 1987~~; Lo Monaco et al., 2005a). More recently the TrOCA method (Tracer combining Oxygen, dissolved Carbon and total Alkalinity) has been developed (Touratier and Goyet, 2004a, ~~2004b~~b; Touratier et al., 2007) and applied in various regions including the SO (e.g. Lo Monaco et al., 2005b; Sandrini et al., 2007; Van Heuven et al., 2011; Pardo et al., 2014; ~~Roden et al., 2016;~~ Shadwick et al., 2014; ~~Van Heuven~~Roden et al., ~~2011~~2016; Kerr et al., 2018). Comparisons with other data-based methods show significant differences in C_{ant} concentrations, especially at high latitudes and more particularly in deep and bottom waters (Lo Monaco et al., 2005b; Vázquez-Rodríguez et al., 2009; Pardo et al., 2014). Thus, there is a need to better explore the C_T and C_{ant} temporal variability in the deep ocean, especially in the SO where observations are relatively sparse. Antarctic bottom waters (AABWs) are of specific interest for the atmospheric CO_2 and heat regulation as they play a major role in the meridional overturning circulation (Johnson et al., 2008; Marshall and Speer, 2012~~) .~~). AABWs represent a large volume of water by covering the majority of the bottom world ocean (Mantyla and Reid, 1995), and their spreading in the interior ocean through circulation and water mixing ~~(Siegenthaler and Sarmiento, 1993)~~ is a key mechanism for the long-term sequestration of C_{ant} and climate regulation~~.~~ (Siegenthaler and Sarmiento, 1993). The AABW formation is a specific process occurring in few locations around the Antarctic continent (Orsi et al., 1999). In short, the AABW formation occurs when the ~~Dense Shelf Water (DSW)~~Antarctic surface waters flows down along the continental shelf. The ~~DSW~~Antarctic surface waters density required for this process to happen is reached by the increase in salinity (S) due to brine release from the ice formation and by a decrease in temperature due to heat loss to either the ice-shelf or the atmosphere. Importantly, AABW formation process is enhanced by katabatic winds that open areas free of ice called polynyas (Williams et al., 2007). Indeed, katabatic winds are responsible for an intense cooling that enhance the formation of ice constantly pushed away by the wind, leading to cold and salty surface waters in contact with the atmosphere. The variable conditions of wind, ice production, surface water cooling and continental slope shape encountered around the Antarctic continent lead to different types of AABW, hence the AABW characteristics can be used to identify their formation sites.

The ability of AABW to accumulate C_{ant} has been controversial since one can believe that the ice coverage limits the invasion of C_{ant} in Antarctic surface waters (e.g. Poisson and Chen, 1987). This is, however, not the case in polynyas, and several studies have reported significant C_{ant} signals in AABW formation regions, likely due to the uptake of CO_2 induced by high primary production (~~Roden et al., 2016;~~ Sandrini et al., 2007; ~~Shadwick et al., 2014;~~ van Heuven et al., 2011, 2014~~).~~; Shadwick et al., 2014; Roden et al., 2016). However, little is known about the variability and evolution of the CO_2 fluxes in AABW formation regions, and since biological and physical processes are strongly impacted by seasonal and interannual climatic variations (Fukamachi et al., 2000; Gordon et al., 2010, McKee et al., 2011; Gordon et al., 2015; Gruber et al., 2019b~~; McKee et al., 2011~~), 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 Studied area

2.1 AABW ~~samples~~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-~~III~~3 cruise (1987). In our analysis, we also included data from the station 14 (deepest sample taken at 5109 m) of the INDIGO-~~I~~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).

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~~eastward, while the Coastal Antarctic Current (CAC) flows ~~Westward~~westward (Fig. 1). The ACC and the CAC influence the circulation of the entire water column, including the AABWs. The main AABW formation sites are the Weddell Sea, where Weddell Sea Deep Water and Weddell Sea Bottom Water are produced ~~deep and bottom waters~~ (WSDW and WSBW, respectively; Gordon, 2001; Gordon et al., 2010), the Ross Sea for the Ross Sea Bottom Water (RSBW; Gordon et al., ~~2015,~~ 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 ~~in the Prydz Bay~~ has also been observed ~~(~~in the Prydz Bay (Yabuki et al., 2006; Rodehacke et al., 2007~~; Yabuki et al., 2006) from~~). There, three polynyas and two ice shelves ~~flowing into the Prydz Channel~~ have been identified as Prydz Bay Bottom Water (PBBW) production hotspots from seal tagging data (Williams et al., 2016~~)~~). This PBBW flows out the Prydz Bay through the Prydz Channel and ~~mixing~~get mixed with the CDBW. The mix of CDBW and ~~Prydz Bay bottom waters~~PBBW (hereafter called CDBW) ~~represent~~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~~) and~~). The WDW is a slightly modified Low

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Circumpolar Deep Water (LCDW) by mixing with High Salinity Surface Water (HSSW) when the ~~WSBW.~~LCDW enters the Weddell basin (see Fig. 2 in van Heuven et al., 2011). The WSDW ~~in the ACC and Weddell Gyre~~ mixes with the ~~Circumpolar Deep Water (CDW).~~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~~, East of~~ (area separating the Kerguelen Plateau~~,~~ from the Antarctic continent), where it mixes with other types of AABWs (Heywood et al., 1999; Orsi et al., ~~1999).~~ 1999). The PET deepest point is 3750 m, deep enough to allow AABWs to flow between the Australian Antarctic Basin and the Enderby Basin (Heywood et al., 1999).

~~In~~At the east of the PET~~ sector~~, the CAC transports a mixture of RSBW and ALBW and accelerates ~~Northward~~northward along the ~~Eastern~~eastern side of the Kerguelen Plateau (Mantyla and Reid, 1995; Fukamachi et al., 2010). Part of the ALBW-RSBW mixture also reaches the ~~Western~~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 ~~mix~~mixes with the CDBW. The mixture of CDBW and ALBW-RSBW either flows ~~Westward~~westward with the CAC and dilutes with the ~~CDW~~LCDW (Meijers et al., 2010) or ~~flow Northward~~flows northward (Ohshima et al., 2013) and ~~mix~~mixes with the ~~older~~ WSDW before reaching the location of our time-series station in the eastern Enderby Basin.

Figure 1~~.~~

2.3 AABW definition

~~Nowadays, the~~The distinction of water masses is usually performed according to neutral density (γ^n) layers. In the SO, ~~CDW~~LCDW and AABW properties are generally well defined in the range 28.15-28.27 kg.m^{-3} 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 ~~AABWs~~AABW core at our location, we prefer to adjust the AABW ~~layer in~~ definition to a narrow ~~band, and select the samples for~~(more homogeneous) layer that we call Lower Antarctic Bottom Water (LAABW), characterised by $\gamma^n > 28.35$ kg.m^{-3} (~~range starting at~~roughly ranging from 4200m to ~~4600m depending on the year~~4800m, see Fig. 3). ~~$\gamma^n > 28.35$ kg.m^{-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).

~~2.4 AABW composition at OISO-ST11~~

~~At each formation site, AABWs experienced 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_{ant} concentrations during AABW formation (Shadwick et al., 2013). The Θ-S diagram constructed from yearly averaged data in bottom waters (Fig. 2) shows that the AABW at OISO-ST11 is a complex mixture of WSDW, CDBW, RSBW and ALBW. The coldest type of AABW was observed at the GEOSECS station at 60° S (-0.56 °C), probably because it experienced less mixing with CDW compared to the warmer type of AABW observed at the INDIGO-1 station at 53° S (-0.44 °C). For the other cruises and years, Θ in AABW ranges from -0.51 to -0.45 °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.s 1 and 2) and~~

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~~the significant C_{ant} concentrations that we estimated at depth (Fig. 3), the main contribution at our location is likely the younger and colder CDBW for which relatively high Cant 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 CDW along the Antarctic shelf and the continental slope between 80°E and 30°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_{ant} concentrations observed in the bottom layer at OISO-ST11 are probably not linked to a single AABW source, but are likely a complex interplay of AABW from different sources with different biogeochemical properties.~~

~~Figure 2.~~

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3 Material and methods

3.1 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 ~~stations~~datasets from GEOSECS, INDIGO-1 and INDIGO-3 were first qualified in ~~GLODAP-v1~~GLODAPv1 (Key et al., 2004) and ~~previously~~ used for the first C_{ant} estimates in the Indian Ocean (Sabine et al., 1999). The ~~data~~adjustments recommended for ~~INDIGO III (1987)~~these historical datasets have been revisited in ~~GLODAP-v2 but the correction~~CARINA and GLODAPv2. In this paper we used the revised adjustments applied ~~on A_T values leads~~ to ~~a suspicious offset and we decided to use~~ the GLODAPv2 data product, with one exception for the total alkalinity (A_T) data from INDIGO-3 for which we applied an intermediate adjustment ~~proposed in GLODAP-v1 and~~ between the recommendation from GLODAPv1 (confirmed in CARINA~~.~~) for no adjustment and the adjustment by -8 µmol.kg^{-1} applied to the GLODAPv2 data product (justification in Supp. Mat.).

For the recent OISO cruises conducted in 2012-2018 not yet ~~qualified~~included in the ~~GLODAP project~~most recent GLODAPv2 product, we have proceeded to a data quality control ~~mainly based on repeated observations~~ in deep waters ~~(CDW)~~ where C_{ant} concentrations are low and subject to very small changes from year to year~~. At this location, the seasonal variations of all properties are only observed in the mixed-layer, about 50 m in austral summer and 150 m in winter (Metzl et al., 2006). Therefore, for deep water analysis, we used the observations available for all seasons (Table 1).~~ (see Supp. Mat.).

3.2 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 (Θ ~~(in °C~~) and salinity (S ~~(no unit~~) measurements have an accuracy of 0.~~01 of their respective units. C_T~~002 °C and ~~A_T~~0.005 respectively. A_T and C_T were sampled in 500 mL glass bottles and poisoned with 100 µL of ~~$HgCl_2$~~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 ~~accuracy~~repeatability for C_T and A_T varies from 1 to 3.5 µmol.kg^{-1} (depending on the cruise) and is determined by sample duplicates (in surface, at 1000 m and in bottom waters~~. All~~). The accuracy of C_T and A_T measurements ~~were calibrated with~~was ensured by daily analyses of Certified Reference Materials (CRMs) provided by A.G. Dickson laboratory (Scripps Institute of Oceanography). ~~O_2~~Dissolved oxygen (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). ~~Thiosulphate~~The thiosulphate solution used ~~in~~for the Winkler titration was calibrated using iodate standard solution ~~to provide~~(provided by Ocean Scientific International Limited) to ensure the standard O_2 accuracy of 2 µmol.kg^{-1}. Nitrate (NO_3) and Silicate (Si) concentrations were measured onboard or ~~offshore~~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) since 2009. Based on replicate measurements for deep samples we estimate an error of about 0.3 % for both nutrients. NO_3 ~~concentrations~~data are not available for all the cruises used in this analysis. The mean NO_3 concentrations in the ~~AABW~~LAABW at OISO-ST11 is 32.8 ± 1.2 µmol.kg^{-1} while the average value derived from the GLODAP-v2 database in bottom waters south of 50°S in the South Indian Ocean is 32.4 ± 0.6 µmol.kg^{-1}. The lack of NO_3 data for few cruises has been palliated by considering a ~~standard~~climatological value of 33 µmol.kg^{-1} with a limited impact on C_{ant} determined by the C° method ~~(from 0.1 µmol.kg^{-1} to 1.7~~(<2 µmol.kg^{-1} on ~~the mean annual values~~estimates based on the differences observed between NO_3 measurements and the climatological value).

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3.3 C_{ant} calculation using the TrOCA method

The TrOCA method was first presented by Touratier and Goyet (2004a, ~~2004b~~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 O_2, C_T and A_T, following:

$$TrOCA = O_2 + a\left(C_T - \frac{1}{2}A_T\right), \qquad (1)$$

where a is ~~the Redfield ratio.~~

defined in Touratier et al. (2007) as combination of the Redfield equation coefficients for CO_2, O_2, HPO_4^{2-} and H^+. For more details about the definition and the calibration of this parameter, please refer to Touratier et al. (2007).

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The temporal change in TrOCA is independent of biological processes and can be attributed to anthropogenic carbon (Touratier and Goyet, 2004a). Therefore, C_{ant} can be directly calculated from the difference between TrOCA and its pre-industrial value TrOCA°:

$$C_{ant} = \frac{TrOCA - TrOCA^0}{a}, \qquad (2)$$

where TrOCA° is evaluated as a function of θ and A_T (Eq. 3):

$$TrOCA^0 = e^{\left[b-(c).\theta-\frac{d}{A_T{}^2}\right]}, \qquad (3)$$

In these expressions, coefficients a, b, c and d were adjusted by Touratier et al. (2007) from free anthropogenic CO_2 deep waters using the tracers $\Delta^{14}C$ and CFC-11 from the ~~GLODAP-V1~~GLODAPv1 database (Key et al., 2004). The final expression used to calculate C_{ant} is:

$$C_{ant} = \frac{O_2 + 1{,}279\left(C_T - \frac{1}{2}A_T\right) - e^{\left[7{,}511-(1{,}087.10^{-2}).\theta-\frac{7{,}81.10^5}{A_T{}^2}\right]}}{1{,}279}, \qquad (4)$$

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

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3.4 C_{ant} calculation using the preformed inorganic carbon (C^0) method

To support the C_{ant} trend determined with the TrOCA method, C_{ant} was also estimated using a back-calculation approach noted ~~C°~~C^0 (Brewer, 1978; Chen and Millero, 1979), previously adapted for C_{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_{bio}) and the preindustrial preformed C_T (~~$C_{0,PI}$~~C^0_{PI}):

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

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

$$C_{bio} = 0.5\Delta A_T - (C/O_2 + 0.5N/O_2)\Delta O_2\ , \quad (6)$$

Where C/O_2 = 106/138 and N/O_2 = 16/138. ΔA_T and ΔO_2 are the difference between the measured values (A_T and O_2) and the ~~preindustrial~~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 Θ, 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):

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

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). ~~The~~A correction of ~~$O_{2,sat}$~~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~~. ΔO_2~~ 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 α~~=~~ = 12 % (Anderson et al., 1991) according to Eq. 9:

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

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

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

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

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

Where the reference water term is a constant for a given time of observation, corresponding to the time when C^0_{obs} 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 µmol.kg^{-1}. This number has been computed using an optimum multiparametric (OMP) model ~~and defined as 51 µmol.kg^{-1} from~~to estimate the mixing ratio of the North Atlantic deep water ~~(Lo Monaco et al., 2005a) and~~ in the SO (used as reference water, i.e. old water mass where $C_{ant} = 0$).

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

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

For more details ~~in~~about the ~~C°~~C^0 method, which has a final error of ± 6 µmol.kg^{-1}, ~~especially on the determination of reference water terms and on the errors of this method,~~ 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~~. 3~~ 2. The LCDW layer (γ^n = 28.15-28.27 kg.m^{-3}) is characterized by ~~maximum AOU values (Fig. 3c), maximum C_T~~ minimum O_2 concentrations (Fig. ~~3d~~2c), higher C_T (Fig. 2b) and ~~minimum~~lower C_{ant} concentrations than in the AABW (Fig. ~~3a~~2a). C_{ant} concentrations were not significant in the LCDW until the end of the 1990s (<6 µmol.kg^{-1}), then our data show ~~a sudden~~an increase in C_{ant} between ~~January and December~~the two 1998 reoccupations, followed by relatively constant C_{ant} concentrations (10±3 µmol.kg^{-1}). In the ~~core of AABW~~LAABW (γ^n > 28.35 kg.m^{-3}), well identified by low Θ, low S~~,~~ and high O_2 ~~and low AOU~~, C_{ant} concentrations are higher than in the overlying ~~deep waters~~UAABW and LCDW (Fig. ~~3a) and~~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±4 µmol.kg^{-1} in 1978~~,~~ and 7±4 µmol.kg^{-1} in the mid-1980s to 13±2 µmol.kg^{-1} at the end of the 1990s and up to 19±2 µmol.kg^{-1} in 2004 (Fig. ~~4a~~3a). Figure ~~4a~~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 ~~core of AABW~~LAABW. Our results show a mean C_{ant} trend in ~~AABW~~the LAABW of +1.~~6~~4 µmol.kg^{-1}.decade^{-1} over the full period and a maximum trend of the order of +~~6.~~5.2 µmol.kg^{-1}.decade^{-1} over 1987-2004 (Table 2). 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 µatm.year^{-1} over 1978-2018), the theoretical C_{ant} trend at the AABW formation sites would be of the order of +8 µmol.kg^{-1}.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 ~~AABW~~AABWs with older ~~CDW waters~~LCDW that contain less C_{ant} over their transit (Fig. ~~3~~2a).

Figure ~~3.~~2

~~To investigate changes in the accumulation of C_{ant} in AABW, Fig. 4 shows the evolution of C_T, A_T, O_2, Θ and S (properties used to estimate C_{ant}), as well as the "natural" component of C_T (C_{Tnat} calculated as the difference between C_T and C_{ant}).~~ Over the full period, C_T increased by 2.0±0.5 µmol.kg^{-1}.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±0.4 µmol.kg^{-1}.decade^{-1} over the 40-years period (Fig. ~~4c~~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 µmol.kg^{-1}.decade^{-1} over 50 years (1960-2010), which is consistent with our data. We did not detect any significant trend in A_T, Θ and S over the full period, but on shorter periods our data show a significant decrease in A_T ~~from the mid-1980s to 2004 (Fig. 4d, Table 2) that is also observed in the overlying deep waters (Fig. 3f). This~~. 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 ~~hence~~ a decrease in ~~C_{Tnat} (i.e. for~~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 ~~C_{Tnat}~~the ‘natural’ component of C_T (C_{nat}, calculated as the difference between C_T and C_{ant}) since 2004 associated to a ~~rapid~~ decrease in O_2 ~~(~~and no increase in ~~AOU) and a decrease in~~ C_{ant} (Table 2). These ~~recent~~ trends were not associated with a ~~small increase in θ (Fig. 4e, Table 2), but no~~ significant trend in θ or S (Fig. ~~4f~~3e,f, Table 2). The increase in ~~C_{Tnat}~~C_{nat} is thus unlikely originating ~~solely~~ 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 ~~nitrate~~NO_3 did not show any significant trend (Table 2).

Table 2~~.~~

Figure ~~4.~~3

Importantly, our data show substantial interannual variations in ~~AABW~~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 µmol.kg^{-1}) compared to 1978 ~~and 1987~~ (5 µmol.kg^{-1}~~).~~) and 1987 (7 µmol.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 ~~AABW~~waters such as the WSDW or ~~reduced mixing with saltier LCDW (Fig. 3h).~~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 θ) 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 ~~nitrate~~NO_3 concentrations. ~~These~~Since these anomalies ~~point to a change in AABW characteristics rather than a change in mixing with the underlying deep waters, and since they~~ 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 ~~(possibly from the Ross Sea due to higher S, Fig. 2).~~. A few years later our data show a S minimum (correlated to lower θ), 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.~~01~~006 decade^{-1} in S between 2004 and 2018 that we observed on the ~~Western~~western side of the Kerguelen Plateau was also observed on the ~~Eastern~~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 ~~salinity~~S by about -0.008~~.~~ decade^{-1} from 2007 to 2016 (against -0.002~~.~~ 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 °C.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 experienced 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 Cant concentrations during AABW formation (Shadwick et al., 2013). The Θ-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° S (-0.56

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°C), while the warmer type of LAABW observed at the INDIGO-1 station at 53° S (-0.44 °C). These extreme Θ values could be a natural feature or may be related to specific sampling. For the other cruises, Θ in LAABW ranges from -0.51 to -0.45 °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_{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 Cant 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° E and 30° 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 Cant 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_{ant} concentrations

In order to compare our C_{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_{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 when C_{ant} concentrations in ~~AABW~~the LAABW are maximum (Fig. ~~4a~~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_{ant}. The mean C_{ant} concentrations for each period are 7, 14 and 13 µmol.kg^{-1}, respectively, which is consistent with the results from other studies (Table 3). The C_{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 (Sabine et al~~.~~., 1999) and because of potential biases for historical data despite their careful qualification 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_{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_{ant} concentrations were low in the 1970s and 1980s, and reached values of the order of 10 µmol.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_{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_{ant} from 1994 to 2007 is not uniform and ranges between 0 and 8 µmol.kg^{-1} (Gruber et al., 2019a). At our station, in the ~~CDW~~LCDW (2000-3000m) the C_{ant} concentrations were not significant in 1978-1987 (-2 to 5 µmol.kg^{-1}) but increase to an average of ~~8.7~~9±3~~.0~~ µmol.kg^{-1} in 1998-2018 (Fig. ~~3a)~~2a), probably due to mixing with ~~AABW~~AABWs that contain more C_{ant}. Interestingly, this value is close but in the high range of the C_{ant} accumulation estimated from 1994 to 2007 in deep waters of the south Indian Ocean (Gruber et al., 2019a).

Not surprisingly, high C_{ant} concentrations are detected in the AABW formation regions (Table 3). The highest C_{ant} concentrations in bottom waters (up to 30 µmol.kg^{-1}) were observed in the ventilated shelf waters in the Ross Sea

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(Sandrini et al., 2007). In the Adélie and Mertz Polynya regions, Shadwick et al. (2014) observed high C_{ant} concentrations in the subsurface shelf waters (40-44 µmol.kg^{-1}) but lower values in the ALBW (15 µmol.kg^{-1}) ~~when it mixed~~due to mixing with older ~~CDW~~LCDW. In WSBW, all C_{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 µmol.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_{ant} concentrations in bottom waters (25 µmol.kg^{-1}) resulting from the shelf waters that contain very high amounts of C_{ant} (50 µmol.kg^{-1}). The comparison with other studies confirms that far from the AABW formation sites, contemporary C_{ant} concentrations are not exceeding 16 µmol.kg^{-1} on average. ~~However, higher C_{ant} concentrations are not unrealistic (Sandrini et al., 2007; Roden et al., 2016; this study in 2004) and likely related to ventilation and water masses variability.~~

Table 3.

5.~~2.~~3 C_{ant} trends and variability

Comparison of long-term C_{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_{ant} trends in the ~~Southern Ocean~~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_{ant} have also been investigated in other SO regions, but limited to 2 to 4 reoccupations (~~Murata et al., 2019;~~ Williams et al., 2015; Pardo et al., 2017~~).~~; Murata et al., 2019). Given the C_{ant} variability depicted at our location (Fig. ~~4a~~3a), different trends can be deduced from limited reoccupations. As an example, Murata et al., (2019) evaluated the change in C_{ant} from data collected 17 years apart (1994–1996 and 2012–2013) along a transect around 62° S and found a small increase at our location (< 5 µmol.kg^{-1} around 60° E). This result appears very sensitive to the time of the observation given that we found a minimum in C_{ant} concentrations between 2011 and 2014 (Fig. ~~4a~~3a) associated with a marked C_T minimum (Fig. ~~4b~~3b). In addition, our results show that the detection of C_{ant} trends appears very sensitive to the time period considered (Table 2). As an extreme case, the C_{ant} trend ~~estimated~~calculated for the period 1987-2004 is +~~6.~~5.2 µmol.kg^{-1}.decade^{-1} (relatively close to the theoretical C_{ant} trend of +8 µmol.kg^{-1}.decade^{-1}), but it reverses to -3.5 µmol.kg^{-1}.decade^{-1} for the period 2004-2018.

The long-term C_T trend that we estimated in ~~AABW~~the LAABW in the eastern Enderby Basin (2.0±0.5 µmol.kg^{-1}.decade^{-1}) is slightly faster than the C_T trends estimated in the WSBW in the Weddell Gyre: +1.2±0.5 µmol.kg^{-1}.decade^{-1} over the period 1973-2011 and +1.6±1.4 µmol.kg^{-1}.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) ~~evaluated~~calculated a C_T trend of +2.4±0.2 µmol.kg^{-1}.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_{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 ~~silicate~~Si concentrations during 1995-2011 (Pardo et al., 2017). This is a clear example of decoupling between C_T and C_{ant} trends in deep and bottom waters as observed at our location in the last decade (Table 2). For C_{ant}, our 40-years trend estimate (1.~~6~~4±0.~~6~~5 µmol.kg^{-1}.decade^{-1}) appears close to the trend reported by Rios et al. (2012) in the ~~South-Western~~south-western Atlantic AABW from 6 reoccupations between 1972 and 2003 (+1.5 µmol.kg^{-1}.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 µmol.kg^{-1}.decade^{-1}).

At our location, the C_{ant} trend over 40 years (+1.~~6~~4±0.~~6~~5 µmol.kg^{-1}.decade^{-1}) explains most of the observed C_T increase (+2.0±0.5 µmol.kg^{-1}.decade^{-1}). The residual of +0.4 µmol.kg^{-1}.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_{Tnat}~~C_{nat}) mirrors the observed decrease in O_2 by -0.~~80~~8±0.4 µmol.kg^{-1}.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 (Schmidtko et al. 2017), suggesting that the changes observed at 56.5°S~~-~~/63°E are related to large-scale processes, possibly due to a decrease in AABW formation (Purkey and Johnson, 2012).

5.~~3.~~4 Recent C_{ant} stability

Although most studies suggest a gradual accumulation of C_{ant} in the AABW, our time-series highlights significant ~~pluriannual~~multi-annual changes, in particular over the last decade when C_{ant} concentrations were as low as around the year 2000 (Fig. ~~4a~~3a) and decoupled from the increase in C_T (Fig. ~~4b~~3b). This result is difficult to interpret because at our location, away from AABW sources (Fig. 1), the temporal variability observed in the ~~AABW~~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~~), but~~). Additionally, internal processes during the transport of AABWs (such as organic matter remineralization, carbonate dissolution and mixing with surrounding waters) must also be taken into account. The apparent steady C_{ant} feature suggests that AABWs found at our location has stored less C_{ant} in recent years. This might be linked to reduced 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 ~~carbon~~C_T-rich deep waters and reduced CO_2 uptake (Lenton et al., 2009). The reconstructed pCO_2 fields by Landschützer et al. (2015) suggest that the reduced 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 fCO_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 fCO_2 data are sparse, especially before 1990, and cruises were mainly conducted in austral summer when the spatio-temporal fCO_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 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 fCO_2 is low and where katabatic winds prevail, very strong instantaneous CO_2 sink can occur at the local scale (up to -250 mmol C.m^{-2}.d^{-1} in Terra Nova Bay in the Ross Sea according to ~~DeJong~~De Jong and Dunbar, 2017). In the Prydz Bay region where CDBW is formed, recent studies show that surface fCO_2 in austral summer vary ~~in~~over a very large range (150-450 µatm), with the lowest fCO_2 observed in the shelf region generating very strong local CO_2 sink (-221 mmol $C.m^{-2}.d^{-1}$~~ according to~~; Roden et al. 2016). The carbon uptake was particularly enhanced near Cape Darnley and coincided with the highest C_{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 fCO_2 values in 1993-1995 were as low as 100 µatm (Gibson and Trull, 1999) leading to a strong local CO_2 uptake of -30 mmol $C.m^{-2}.d^{-1}$ in summer. In addition, Roden et al. (2013) found a large C_T increase over 16 years (+34 $µmol.kg^{-1}$) in the Prydz Bay, which is much higher than the anthropogenic signal alone (+12 $µmol.kg^{-1}$) 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_{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 CO_2 uptake in this region using the qualified fCO_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_{ant} stability that we observed in the ~~AABW~~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_{ant} in AABW could also be directly related to changes in physical processes occurring in AABW formation ~~region~~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 AABWs over the last decades (Rintoul, 2007). 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_{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_{Tnat}~~C_{nat}) not associated with an increase in C_{ant}, and a decrease in O_2 (as observed in recent years in Fig. ~~4a~~3a,b,c). Finally, it is also possible that the ~~AABW~~LAABW observed in recent years at our location is the result of a larger contribution of older RSBW~~ and/or~~, 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 AABWs 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 Low Antarctic Bottom Water (LAABW) as described in the Section 2.3. Our results suggest that the accumulation of C_{ant} explains most~~ of (~~, 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~~AABWs 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 µmol.kg^{-1}.decade^{-1} between 1987 and 2004 and an apparent stability in the last 20 years (despite an increase in C_T). This suggests that AABWs 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 ~~AABW~~the LAABW since 1998 could also be directly linked to a decrease in ~~AABW~~AABWs formation in the 1990s (Purkey and Johnson, 2012) or a change in the contributions of AABWs 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 ~~CDW~~LCDW along AABWs transit would also explain the decoupling between C_{ant} and C_T (increase in ~~C_{Tnat}~~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 ±2 µmol.kg^{-1}, Bockmon and Dickson, 2015) and uncertainties of data-based methods (±6 µmol.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_2 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~~; Heuzé et al., 2014~~; 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.

Data availability

GEOSECS, INDIGO and OISO 1998-2011 data are publicly available at the Ocean Carbon Data System (OCADS; https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2_2019). OISO original data ~~stations~~ are available at: www.nodc.noaa.gov/ocads/oceans/RepeatSections/clivar_oiso.html. OISO 2012-2018 will be available in GLODAPv2_2021.

Author contributions

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

~~**Author contributions**~~

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

We thank the captains and crew of the R.S.V. Marion Dufresne and the staff at the French Polar Institute (IPEV) for their important contribution to the success of the cruises since 1998. We are also very grateful to all colleagues, students and technicians who helped to obtain the data. We extend our gratitude to P. C. Pardo, S. R. Rintoul and B. Legresy for the discussions during the preparation of the manuscript and to M. K. Shipton for the valuable comments. We thank two anonymous reviewers for their comments and constructive suggestions that helped improve the manuscript. The OISO program was and is supported by the French institutes INSU ~~and~~, IPEV and OSU Ecce-Terra and the French program SOERE/Great-Gases. Support from the European Integrated Projects CARBOOCEAN (511176) and CARBOCHANGE (264879) is also acknowledged.

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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

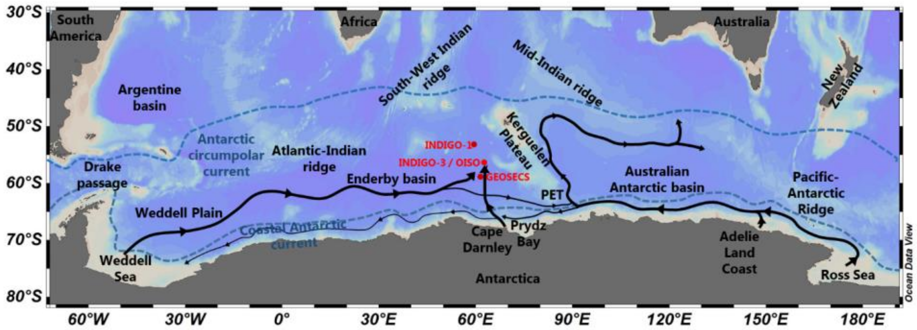

Figure 1. The AABWs circulation from the literature (Fukamachi et al., 2010; Orsi et al., 1999) and this study, with geographic indications (black text), SO currents (blue text and dash lines for the approximative positions) and stations considered in this study (red text and dots). PET: Princess Elizabeth Trough. Figure produced with ODV (Schlitzer et al., 2019).

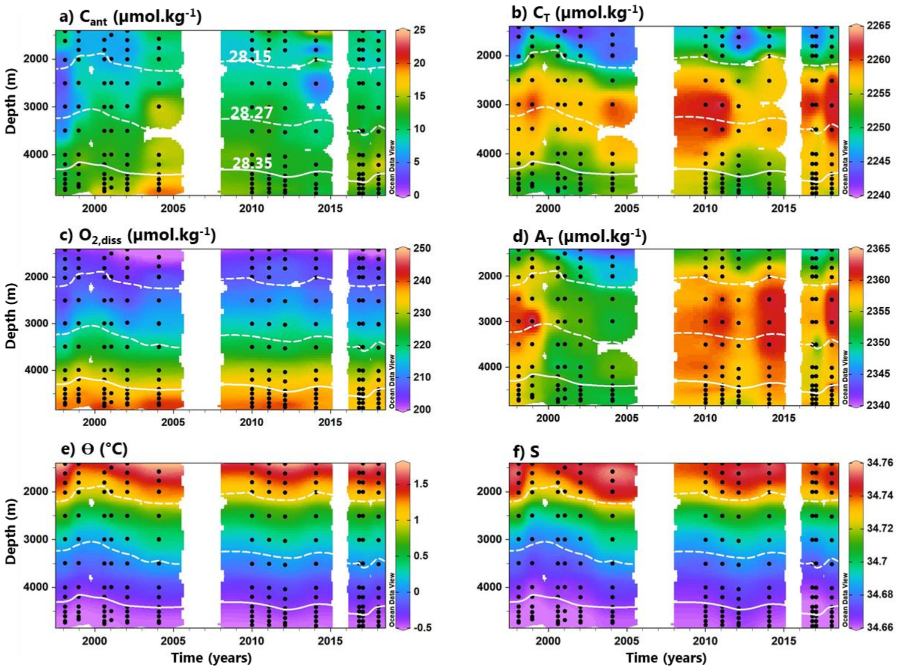

Figure 2. Hovmöller section of (a) C_{ant} via TrOCA, (b) C_T, (c) O_2, (d) A_T, (e) θ and (f) S based on the OISO data presented in Table 1. Data points are represented by black dots. The white isolines represent the water masses separation by γ^n (from the bottom: LAABW, UAABW and LCDW). Figure produced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

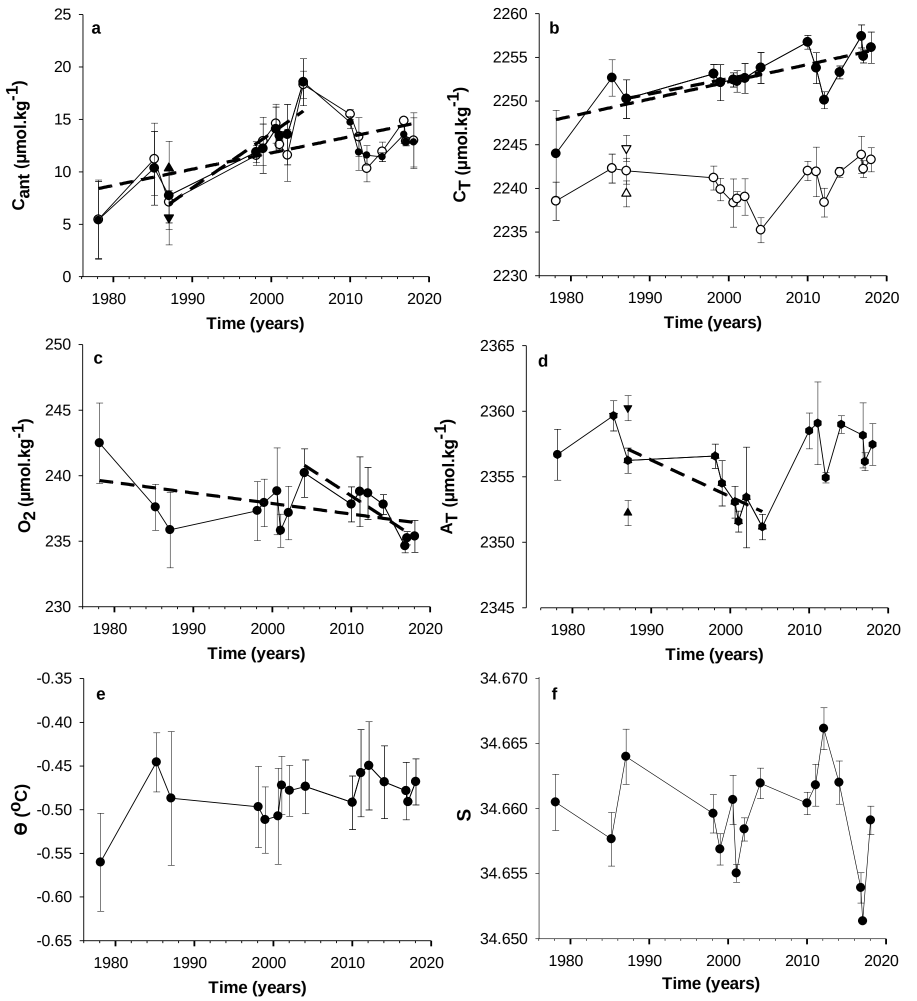

Figure 3. Interannual variability (dash lines lines) and significant trends (at 95 %, see Table 2; dotted lines) for the 40 years of observation of the OISO-ST11 LAABW properties, including (a) C_{ant} by the TrOCA (black circles and triangles) and the C^0 (open circles) method, (b) C_T (black circles) and C_{nat} (open circles), (c) O_2, (d) A_T, (e) Θ 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 µmol.kg^{-1} of correction on the A_T, respectively (see Supp. Mat. for more details).

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	Θ °C	Si $\mu mol.kg^{-1}$	NO_3 $\mu mol.kg^{-1}$	O_2 $\mu mol.kg^{-1}$	A_T $\mu mol.kg^{-1}$	C_T $\mu mol.kg^{-1}$	C_{ant} TrOCA $\mu mol.kg^{-1}$
1978-2018	-0.001 ± 0.001	0.01 ± 0.01	-1.2 ± 0.9	0.2 ± 0.2	**-0.8 ± 0.4**	-0.1 ± 0.1	**2.0 ± 0.5**	**1.4 ± 0.5**
1987-2018	-0.001 ± 0.001	0.01 ± 0.01	-1.9 ± 1.4	0.3 ± 0.4	-0.3 ± 0.5	0.6 ± 0.1	**1.6 ± 0.5**	1.1 ± 0.8
1987-2004	-0.003 ± 0.002	0.01 ± 0.01	**-6.5 ± 1.8**	0.9 ± 0.9	1.7 ± 1.0	**-1.9 ± 1.1**	**1.8 ± 0.4**	**5.2 ± 1.1**
2004-2018	-0.006 ± 0.003	0.01 ± 0.01	-1.8 ± 4.5	-0.5 ± 1.0	**-3.9 ± 0.7**	3.4 ± 0.2	1.7 ± 1.9	-3.5 ± 1.5

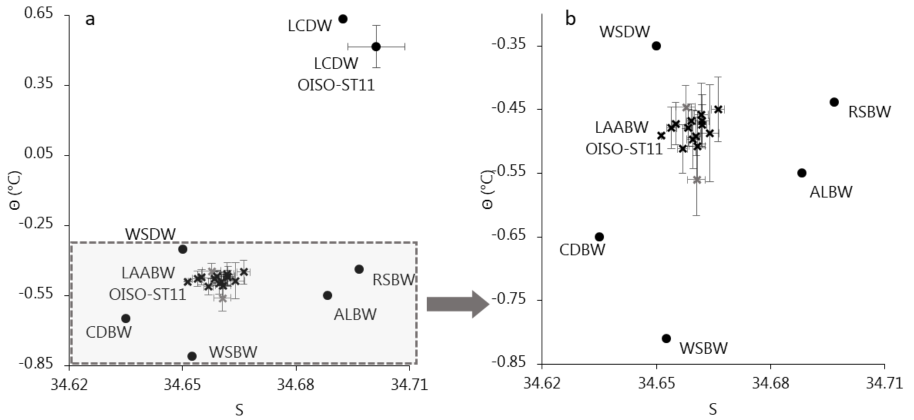

Figure 4. (a) Full Θ-S diagram of studied water masses and (b) zoomed on bottom waters. Values are from literature for the WSBW (Fukamachi et al., 2010; van Heuven, 2013; Pardo et al., 2014; Robertson et al., 2002), the WSDW (Carmack and Foster, 1975; Fahrbach et al., 1994; van Heuven, 2013; Robertson 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 Θ) and INDIGO-1 (highest Θ) values.

Table 3. Compilation of C_{ant} sequestration investigations in the AABWs (γ^n > 28.25 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 surface water. The mean values published by Roden et al. (2016) for the AABWs present WSDW characteristics but can be a mix of CDBW and LCDW.

Source	Location	Water masses considered	Year	C_{ant} µmol.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)	WSDW-CDBW-RSBW-ALBW	1978-1987	8 ± 3
			1987-1998	10 ± 4
			1987-2004	13 ± 4
			1998-2004	14 ± 2
			2010-2018	13 ± 1
			1978-2018	**12 ± 3**