Response to the editor on the manuscript "Carbonate system distribution, anthropogenic carbon and acidification in the Western Tropical South Pacific (OUTPACE 2015 transect)". bg-2018-163

Dear Editor,

We would like to thank again the referees for their valuable comments that have improved our manuscript. We are pleased to submit a revised version of our manuscript.
In the following document, we provide our detailled answers to both referees and a version of the manuscript that includes track changes in order to easily identify changes from the previous version.

We thank you in advance for the attention you will pay to this new version of our manuscript.

Yours sincerely,

Thibaut Wagener
on behalf of all coauthors

Response to the Interactive comment by « Anonymous Referee #1 » on "Carbonate system distribution, anthropogenic carbon and acidification in the Western Tropical South Pacific (OUTPACE 2015 transect)".

Please note that the referee comments are typesetted with normal characters **and our responses to referee's comments are in bold characters. Text from the manuscript is indicated with** *italic characters* **and changes are highlighted** *in red*. **For minor changes mentioned by the referee, we sometimes just mention that we agree with the referee and we will of course make the corrections in the revised manuscript.**

General comments:
The main goal of the manuscript is to report a new dataset of measurements of the carbon system for the western tropical South Pacific (OUTPACE cruise). The authors describe the distribution of the different variables along the OUTPACE transect highlighting the differences between the western (Melanesian Archipelago, MA) and the eastern (western South Pacific gyre, WGY) part of the transect.
The authors also present results for derived properties (pH, Ω cal and Ω ara) of the carbon system and for anthropogenic carbon (CANT) that has been estimated by the TrOCA method. Making use of ancillary data, the authors present temporal changes in the properties (measured and estimated) observing: 1) a decrease in total alkalinity restricted to the MA area that disappears when using normalized alkalinity; 2) an "over accumulation" of total inorganic carbon and an increase in CANT (close to the thermodynamic value) in the upper thermocline waters; 3) a decrease in pH and shoaling of the aragonite saturation depth.
The dataset reported in the study is of high quality and without any doubt complements the decadal P21 hydrographic line. The manuscript is well written and ordered and the results are well presented.
Nevertheless, I have some specific comments that need to be addressed before being considered for publication.
First, we would like to thank Referee #1 for his/her careful evaluation of our manuscript. We believe that his/her comments will help to improve the manuscript. Please, find hereafter our responses to the concerns raised by Referee #1

Specific comments:

Abstract:
Page 1, line 22. Eliminate "of" after C ANT increases.
We agree with this correction.
Page 1, line 23. "in C ANT " instead of "of C ANT ".
We agree with this correction.
Page 1, line 24. "pH T " instead of "pH".
We agree with this correction.

1 Introduction:
Page 2, line 42. Delete "hereafter named".
We agree with this correction.

2.4 Derived parameters:
Page 5, line 138. "calcite (Ω cal)". There is no need to mention this variable because it is not displayed in the distributions (fig. 3) and its temporal change is not estimated. See comment on section 4.
The reference to (Ω cal) estimates was deleted but a sentence has been added to justify why this parameter was not considered.

Seawater pH on the total scale (pH_T) and the $CaCO_3$ saturation state with respect to aragonite (Ω_{ara}) were derived from A_T and C_T with the "Seacarb" R package (Gattuso and Lavigne, 2009). $CaCO_3$ saturation state with respect to calcite was not considered because seawater up to 2000 dbar was supersaturated with respect to calcite (Ω_{cal}>1).

4 Carbonate chemistry along the OUTPACE transect:
Page 7, line 200. Why is C T slightly lower in bottom waters?
A possible explanation is that in the South Pacific, the deep waters are among the oldest waters in the world ocean with high carbon content whereas the northward moving bottom waters have not had the time to accumulate as much carbon (see for example Murata et al. 2007). The sentence has been modified as follow:
The C_T gradient in the upper water column has been described in Moutin et al. (2008). Below 2000 dbar, C_T is relatively invariant with slightly lower values in the bottom waters (below 4000 dbar) due to the presence of very old deep waters originating from the north Pacific relative to the northward moving bottom waters that have not accumulated as much carbon (Murata et al. 2007).

Page 7, line 211. "pH T " instead of "pH".
We agree with this correction.
Page 8, lines 217-220. Not need to add these sentences or maybe use them in section 2.4 to explain why you're not considering this variable for the temporal changes.
These sentences have been deleted and a sentence has been added earlier in section 2.4 to justify that Ω_{cal} will not be considered.

5 Anthropogenic carbon estimation along the OUTPACE transect:
Page 9, lines 272-279. The authors make the reader notice that denitrification could be affecting their estimates but nothing is concluded. The authors don't explain how they deal with this issue. In section 6 the authors give a reference for the low effect of denitrification over C ANT estimates that could be added in this section as a conclusion of why they don't consider N*.
We agree with this comment of the referee. Based on the suggestions of the referee, we have rephrased this section in order to be clearer.
Finally, it should also be mentioned that, due to the presence of one of the main OMZ area, denitrification occurs in the eastern South Pacific and can be traced by the N parameter (Gruber and Sarminento, 2007). Denitrification, by transforming organic carbon to inorganic carbon without consumption of oxygen, could induce an overestimation of C_{ANT} by the TrOCA method (and other back calculation methods) due to a biological release of C_T that is not taken into account in the formulation of the quasi conservative TrOCA tracer. Horizontal advection by the south equatorial current of the strong negative N* signal originating from the Eastern Pacific towards the western Pacific was previously described (Yoshikawa et al., 2015). Fumenia et al. (2018) have estimated N* along the OUTPACE transect and show slightly negative N* values in the upper thermocline waters at the eastern side of the OUTPACE transect where the highest CANT values are estimated. However, Murata et al. (2007) showed that, based on a direct relation between C_T and N*, the influence of denitrification should be negligible on C_{ANT} estimations in this area. Therefore, the N* correction has not been introduced in the C_{ANT} estimates and the effect of denitrication was not quantified here.*

Page 10, line 285. The year of publication of the reference is 2017.
We agree with this correction. However, following a suggestion of referee #2, this section will be deleted in the revised manuscript.

6 Temporal changes of inorganic carbon in the OUTPACE area:
Section's title: The authors talk about other variables than just inorganic carbon. I suggest to change "inorganic carbon" to "carbonate chemistry".

We agree with this correction.

Page 10, lines 303-305. Add the errors in the trends for A T . What is/are the oceanic process/es behind the change/not change in alkalinity.

Errors have been added for A_T trends. Concerning the main drivers of A_T changes in the ocean (Wolf-Gladrow et al. 2007): The major change in A_T can be attributed to changes in major conservative cations and anions (i.e. salinity). The other important changes in A_T are due to the biological precipitation of calcium carbonate and/or the dissolution of biogenic calcium carbonate. Finally, minor changes in A_T can be attributed to biological assimilation and remineralization of nitrate. A_T in the ocean is not affected by changes in the CO_2 content of the ocean. In our study, when A_T is normalized to salinity, no significant trends in A_T n35 are observed, suggesting that the observed trends in A_T can be attributed to salinity changes. The manuscript has been changed as follow:

However, when A_T is normalized to salinity, no significant trends are observed in $A_{T\ n35}$ suggesting that the observed trends in A_T can be attributed to changes in salinity rather than in calcification.

Page 10, lines 306-307. Add the errors in the trends for C T and C ANT .

We agree with this correction.

Page 10, line 310. Do the authors have an explanation for this “over accumulation”? What is the error in the increase of C T associated to the increase in atmospheric CO2? (line 308).

We agree with the referee that the discussion on this “over accumulation” was not precise enough. In the revised manuscript we will rephrase this section as mentioned in the next comment.

Page 11, lines 315-318. Considering the information given by the authors (page 10, lines 303-305), can the changes in AT still be due to remineralization processes? Can the authors give a possible scenario/explanation for the difference of C ANT between MA and WGY?

As mentioned in the above comment, this section has been rewritten as follow:

At $\sigma_{\theta\ 25}$, a significant decrease of A_T of -0.20 ± 0.07 µmol $kg^{-1}.a^{-1}$ is observed over the entire OUTPACE area. A decrease of -0.30 ± 0.09 µmol.$kg^{-1}.a^{-1}$ is also observed in the MA area, whereas no significant trend is observed for the WGY area. However, when A_T is normalized to salinity, no significant trends are observed in $A_{T\ n35}$ suggesting that the observed trend in A_T can be attributed to salinity changes rather than changes in calcification. Significant negative trends are observed for $[O_2]$ over the entire area (- 0.31 ± 0.10 µmol kg^{-1} a^{-1}) with - 0.35 ± 0.16 µmol kg^{-1} a^{-1} in the MA and - 0.38 ± 0.11 µmol kg^{-1} a^{-1} in the WGY. The decrease in $[O_2]$ which corresponds to a positive trend in AOU suggested an increase in the remineralization of organic matter at $\sigma_{\theta\ 25}$. Significant increasing trends were observed for C_T over the entire area (+ 1.32 ± 0.13 µmol kg^{-1} a^{-1}), in the MA (+ 1.38 ± 0.21 µmol kg^{-1} a^{-1}) and in the WGY (+ 1.57 ± 0.13 µmol kg^{-1} a^{-1}). For C_{ANT}, the trends were slightly slower (+ 1.12 ± 0.07 to 1.2± 0.13 ± 0.09 µmol kg^{-1} a^{-1}) and not significantly different between the MA and the WGY. Taking into account the OUTPACE dataset does not change the overall significance of the observed trends and only minor changes (mostly within the error of the estimates) are observed. If we assume a C_T increase of 0.5 to 1 µmol kg^{-1} a^{-1} (depending on the buffer factors considered) associated to the recent rise in atmospheric CO_2 (see for example Murata et al., 2007), the C_T increase in the OUTPACE area is faster than thermodynamics would govern whereas the C_{ANT} is closer to this thermodynamic value. The higher increase of C_T could be related to an increase in remineralization as deduced from $[O_2]$ trends, with an overall consistency between the rate of C_T increase and the rate of $[O_2]$ decrease. However, the important increase of C_{ANT} observed between 2005 and 2015 between 10°S and 30°S on the P16 line (at the eastern side of the OUTPACE transect) by Carter et al. (2017) is not supported by significant differences in the trends of C_{ANT} observed between MA and WGY in this study.

Page 11, line 319. Add the error for the change in C ANT.

We agree with this correction.

7 Towards an enhanced “Ocean Acidification” in the WTSP?:
Pages 11 and 12, lines 329, 331, 336, 337, 344, 345, 346, 347, 352, 357. “pH T ” instead of “pH”.
We agree with this correction.

Page 11, lines 341-342. Add the errors in the trends for fCO2, C T and pH T .
We agree with this correction.

Page 12, line 362. Add errors in the trends. They are given in the text of reference.
We agree with this correction.

Page 12, lines 363-364. Add the values of the change in Ω ara (with the uncertainty) that you obtained with your data. Give some explanation for the difference between your values and the ones obtained by Murata et al. (2015).
This section was probably unclear. The aim of this section was to discuss our estimates of “anthropogenic Ω_{ara} change” since the preindustrial period. Indeed, we do not discuss decadal Ω_{ara} changes which were not estimated here. The reason why we compared with the Murata et al. study was to point out the interesting longitudinal differences in the Ω_{ara} decrease observed in the recent years (1994 to 2009) in the OUTPACE area which are attributed, at least partially, to changes in sea surface temperature, that we do not observe on our long term estimates. However, we believe this section was confusing for the reader and we will removed this comparison with Murata et al. (2015).

Page 12, line 368. Add the migration rate observed by Feely et al. (2004) and the period of study.
In the study by Feely et al (2004), upward migration of Ω_{ara} horizons between the preindustrial period and present (late 90s) are evaluated by a method comparable to ours and values between 30 and 100 m are given for the Pacific Ocean. These values will be added to the manuscript.

8 Conclusion:
Page 12 line 375. “pH T ” instead of “pH”.
We agree with this correction

Response to the Interactive comment by « Anonymous Referee #2 » on "Carbonate system distribution, anthropogenic carbon and acidification in the Western Tropical South Pacific (OUTPACE 2015 transect)".

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This is a broadly well-written paper that cleanly presents the information data users would need to use the dataset being presented. In these respects, the paper is worthy of publication. However, the paper runs into trouble in the extension of its analysis to Cant. Lacking a direct earlier occupation to compare to or transient tracer information to provide ventilation age information, the dataset is ill equipped to be used for these estimates (as the authors point out). The authors therefore use the TrOCA approach for estimating Cant. TrOCA is convenient and easy to apply, but untrustworthy: The authors discuss this limitation in section 5 (it should also be briefly mentioned in section 2.4), but then go on to discuss comparisons between various regions and literature estimates without further mentioning or propagating the uncertainties from the methods. This leaves the reader to believe that the uncertainties in the fits are appropriate estimates of the uncertainties in the estimates, which seems incorrect. Fortunately, the proximity of the data set to P21 allows the authors' TrOCA estimates to be compared to earlier TrOCA estimates from 1994 and 2009. This analysis should allow much of the TrOCA methodological error to cancel when computing changes in Cant over time (see: https://doi.org/10.5194/bg-7-1789-2010, who briefly provide a bit of evidence to suggest this approach might work okay for estimating rates). However, the change in the sampling grid between the P21 cruises and this cruise could pose a separate problem for this approach (www.biogeosciences.net/10/4801/2013/), particularly when comparing regions to one another (see below).

In terms of improving the Cant discussion, I'd argue the paper should:
1. Remove the discussion of column inventories of Cant, and downplay or remove the discussion of the overall Cant distributions. Presenting these values suggests a belief in the accuracy of the values to within the stated precision of ±6 μmol kg-1 that isn't warranted given Yool et al.'s findings. Instead focus on Cant changes.
2. {Delta}nCT0 or eMLR could be used to compare the P21 section datasets to the new measurements (and to one another), useful to show that rates of change found are not byproducts of the TrOCA methods used.
3. Attempt to estimate Cant uncertainty from Yool et al., and then propagate these estimates through their calculations to estimate uncertainty in each of the values they present.
4. For dealing with the change in the sampling grid, it might be interesting to simply compare the rates found with and without the new dataset. This would allow the rates from "P21 only" to be directly compared to Kouketsu et al. 2013, who use a different method entirely. The differences between those estimates and these could then be discussed in the context of both changes in rates and changes in sampling grids.
Alternately, the paper could likely stand as a simple presentation of the data to ESSD after removing most of the Cant discussion.

First of all, we would like to thank referee #2 for his/her careful evaluation of our manuscript. We believe that his/her comments will help to improve the manuscript. We would also like to acknowledge his/her frank but courteous criticism on our estimates of C_{ANT} by the TrOCA method. We understand that the reviewer does not agree with our estimates of C_{ANT} because (s)he considers the TrOCA method untrustworthy. We are aware of the scientific debate that

exists on the different methods to estimate C_{ANT} in the water column and in particular the TrOCA method.

We believe that this manuscript should be part of the "Interactions between planktonic organisms and biogeochemical cycles across trophic and N2 fixation gradients in the western tropical South Pacific Ocean: a multidisciplinary approach (OUTPACE experiment)" special issue in Biogeosciences. As mentioned in the manuscript, even if the dataset presented has been partially used in Moutin et al. 2018 (this issue), we consider that this manuscript gives a complete information on the carbonate data acquired during the OUTPACE cruise. Comparing the "OUTPACE" data to the GLODAPv2 dataset confirms recent trends observed in the South Pacific in terms of increase of the carbon content and ocean acidification. However, as mentioned by the referee, our dataset "is ill equipped" to be used for C_{ANT} estimates because the OUTPACE transect does not correspond to a earlier occupation in the South Pacific and no transient tracer information to provide ventilation age information is available. Moreover, the horizontal and vertical resolution of our dataset is relatively low and for most of the stations no data have been collected below 2000 dBar. This makes our dataset less extensive compared to the earlier "WOCE lines" cruises that took place in this area (e.g. Carter et al. 2017, Kouketsu et al. 2013).

Despite the limitations of the TrOCA method (which are explicitly exposed in the manuscript, see response below), we decided to apply it because of its simplicity in the case of our dataset. Even if the TrOCA method could produce some wrong estimates of C_{ANT}, we believe that it can be used as a tool to investigate changes in C_T content and that it can be valuable for estimating C_{ANT} accumulation rates.

In order to respond to the general concerns of referee 2, we propose the following changes in the manuscript:

1. We will remove our estimates of C_{ANT} column inventories. We will delete lines 251 to 254, lines 280 to 288, line 376 and lines 380 to 382 of the manuscript. We will also remove the first panel of Table 2 (which will become Table 3) and we will remove the C_{ANT} inventories on figure 6 (last panel). The new Figure 6 and Table 3 can be seen at the end of this document. We propose to modify lines 18 to 20 of the abstract as follow:

Along the OUTPACE transect, a deeper penetration of C_{ANT} in the intermediate waters was observed in the MA, whereas highest C_{ANT} concentrations were detected in the sub-surface waters of the WGY.

2. We will expose more clearly the motivations and limitations of our dataset and describe more clearly what we want to state with our manuscript. We propose the following changes to the introduction:

The aim of this paper is to report a new dataset of oceanic inorganic carbon (based on measurements of C_T and total alkalinity (A_T)) acquired in the WTSP during the OUTPACE (Oligotrophic to UltTra oligotrophic PACific Experiment) cruise performed in 2015 (Moutin et al., 2017). The main focus of the OUTPACE cruise was to study the complex interactions between planktonic organisms and the cycle of biogenic elements on different scales, motivated by the fact that the WTSP has been identified as a hot spot of N_2 fixation (Bonnet et al., 2017). The data presented here have been partially used in another paper of the special issue (Moutin et al., 2018) in order to study the biological carbon pump in the upper (surface to 200m) water column. In this paper we will explore the carbonate data between the surface and 2000 m depth. The OUTPACE transect (Figure 1) is close to existing WOCE and GO-SHIP lines in the South Pacific : it is parallel to the zonal P21 line (18° S visited in 1994 and 2009) and the P06 line (32° S visited in 1992, 2003 and 2010), it is crossed by the meridional P14 line (180° E visited in 1994 and 2007) and P15 line (170° W visited in 2001, 2009 and 2016) and it is situated at the eastern side of the P16 line (150° W visited in 1992, 2005 and 2014). However, the OUTPACE transect does not correspond to any earlier occupation of the "WOCE lines" in the South Pacific and no tracers of water mass age were measured during the cruise, which limits the possibilities of a robust analysis of C_{ANT} accumulation in the area. Moreover, the horizontal and vertical resolution of the OUTPACE

dataset is low. In consequence, the OUTPACE dataset cannot be used to look at decadal changes in C_{ANT} content in the South Pacific (e.g. Carter et al. 2017, Kouketsu et al. 2013). Here, C_{ANT} estimates based on the TrOCA (Tracer combining Oxygen, inorganic Carbon and total Alkalinity) method will be used as a tool to investigate changes in C_T. Moreover, by comparing our data with the high quality data (internally consistent through a secondary quality control (Olsen et al., 2016)) available in the Global Ocean Data analysis Project version 2 (GLODAPv2 database), will allow to evaluate C_T, A_T, C_{ANT} (for TrOCA) and pH_T (pH on total scale) trends in sub surface waters and at depth.

3. We will mention in the Sect. 2.4 the limitation of the C_{ANT} estimates by the TrOCA method and we will refer to the section 5 for a longer discussion on these limitations. We propose the following changes to the Sect. 2.4 :

This formulation is based on an adjustment of the TrOCA coefficients using $\Delta^{14}C$ and CFC-11 from the GLODAP-V1 database (Key et al., 2014). Touratier et al. (2007) estimated the overall uncertainty of the C_{ANT} with TrOCA method to ca. 6 µmol kg^{-1} based on the random propagation of the uncertainties on the variables (C_T, A_T, $[O_2]$ and θ) and coefficients used in Eq. 1. The limitations and validity of the TrOCA method will be discussed in detail in Sect. 5.

4. We will expose more clearly the limitations of the TrOCA method in section 5 and we will give estimates of the error associated to the TrOCA method based on the study by Yool et al. 2009. We propose following changes to the Sect. 5 :

As no tracers of water mass age were measured during the OUTPACE cruise, the main motivation for using the TrOCA method was to make C_{ANT} estimations based on a simple calculation from parameters acquired within the cruise as done in other cruises conducted in south tropical Pacific waters (e.g. Azouzi et al., 2009; Ganachaud et al., 2017). Even if C_{ANT} estimates from TrOCA could be biased, the application of a simple back-calculation method that accounts for biologically induced relative changes in C_T is used here to identify some spatial features in the distribution of the carbonate system along the OUTPACE transect. Based on Yool et al. (2010), the error on the TrOCA C_{ANT} estimates will be considered here as the normalized standard deviation of 1.67 for the TrOCA variant optimized with world ocean data (See Table 2 in Yool et al. 2010).

6. In section 6, based on the suggestion of the Referee, we will estimate the trends with and without the OUTPACE data in order to illustrate that the OUTPACE data confirms the trends observed with GLODAP_v2. However, we believe that the differences between those two estimates cannot illustrate the influence of changes in sampling grids because the estimation of the trend is not only based on P21 data but on all available data in the OUTPACE area. We will change Table 3 (that will become Table 2) with the estimates of the trends with and without the OUTPACE data. In addition to the new Table 2 (that can be seen at the end of this document), we propose the following changes to the Sect. 6 :

Based on the available GLODAPv2 data, temporal changes in the OUTPACE area have been assessed (Fig. 5 and Table 3). The variation of oceanic parameters with time are estimated on two isopycnal layers : A layer with 25 kg m^{-3} < σ_θ < 25,5 kg m^{-3} (hereafter named $\sigma_{\theta\,25}$) and a layer with 27 kg m^{-3} < σ_θ < 27.2 kg m^{-3} (hereafter named $\sigma_{\theta\,27}$). These two layers correspond to the features in C_{ANT} discussed in the former section. $\sigma_{\theta\ 25}$ can be considered as characteristic of the upper thermocline waters (core of the salinity maximum, Fig 2) whereas $\sigma_{\theta\ 27}$ can be considered as characteristic of intermediate waters of southern origin (core of the salinity minimum). All the values associated to these two layers are spread between 145 and 301 dbar for $\sigma_{\theta\ 25}$ and between 571 and 896 dbar for $\sigma_{\theta\ 27}$. It must be mentioned that the study of temporal changes is based on a large sampling grid which covers the entire OUTPACE transect (see Sect. 2.5. and Fig. 1). This could add a spatial variability that may interfere in the estimation of temporal changes.

Temporal variations of C_T and C_{ANT} between 1970 and 2015 are presented on Fig 5. As mentioned earlier, even if C_{ANT} estimates from TrOCA could be biased, a former study by Perez et al. 2009 suggests that the TrOCA method gives similar values than other methods for estimating C_{ANT} accumulation rates. A linear fit was applied to the observed temporal variations for A_T, $[O_2]$, C_T and C_{ANT} to check for significant trends on data collected between 1980 and 2015 (OUTPACE

cruise). The results of the performed regression analyses are presented on table 2. Trends are evaluated with and without the OUTPACE cruise data in order to estimate the influence of this new dataset on the observed trends. Trends are evaluated for the entire OUTPACE area and for the MA and the WGY areas. Even if presented on Figure 5, data collected before 1980 from the GLODAPv2 database are disregarded in the estimation of the temporal trends. Indeed, for the OUTPACE area, data prior to 1980 originates from one single GEOSEC cruise in 1974, with only one measured point at $\sigma_{\theta 27}$ *for WGY and no points at* $\sigma_{\theta 25}$ *for WGY and MA.*

7. We will change Figure 1. We will only present the GLODAP V2 data that have been used in this study and not all data that exists in the area covered in the map. We believe that this might have been confusing in the manuscript. The new Figure 1 can be seen at the end of this document.

Finally, we think that a complementary analysis of C_{ANT} based on other methods as suggested by the referee would be out of the scope of this study and would completely modify the objectives that we assigned to this manuscript. Moreover such a complementary analysis could be difficult to realize due to the reasons mentioned earlier.

Specific comments:
13. carbonate parameters
We agree with this correction.
27. recommend: 10 PgC or GtC
We changed 10^{13} kg to 10 PgC.

29. Socean/EFF=0.26 (from LeQuere), which is closer to 25% than 30%. 30% is closer to the historical average sequestration.
We agree with this comment and we will change 30% to 25% to more correctly represent the estimate in Le Quéré et al. (2018).

136. These five. . .
We agree with this correction.

180. Consider also the “potential vorticity minimum” definition of SAMW.
We will change this sentence as follow :
Hartin et al. (2011) defines SAMW with σ_θ *values between 26.80 and 27.06 kg* m^{-3} *corresponding to a minimum in potential vorticity, and AAIW with* σ_θ *values between 27.06 and 27.40 kg* m^{-3}.

207. Recommended “The estimated offsets are XXX and XXXX. These offsets are smaller than the estimated repeatability of the measurements.”
- However, if this suggested text is an accurate phrasing of the idea being conveyed then it implies you believe that the repeatability is a good estimate of the potential bias… potential measurement biases on the order of the listed repeatability would completely hide decadal Cant accumulation at 95% confidence. Pehaps rephrase simply as "The estimated offsets are XXXX and XXXX, suggesting measurement biases are likely no larger.
We understand this concern of the reviewer. We rephrased the sentence following his/her last suggestion : *“The estimated offsets are -2.0 ± 4.2 µmol* kg^{-1} *for* A_T *and -2.0 ± 4.4 µmol* kg^{-1} *for* C_T *suggesting measurement biases are likely no larger”*.

236. depths
We agree with this correction.

248. singularities should be another word. . . perhaps “features”

We have changed “singularities” to “features”.

285. the high 0.8 mol C / mˆ2 estimates in Carter et al. were only for the last decade or so. The estimates in this region were smaller for the WOCE-CLIVAR period. If we assume 1994 to 2005 with accumulations of ∼0.3 mol C /mˆ2 per year (approximated from the figure in Carter et al.) with 2005 to 2015 accumulations of 0.8 per year... it suggests a total change of ∼11 mol C /mˆ2 or so, rather than the 20 mol C /mˆ2 since 1994 found here.
As mentioned earlier this section on C_{ANT} inventories has been removed from the manuscript. However we agree that combining the estimates “WOCE-CLIVAR” from Sabine et al. 2008 to the CLIVAR-GOSHIP estimates from Carter et al. 2017 was inconsistent.

298. what does it mean to be adjusted to a linear model? Possible recommended rephrase: “a line was fit to the data. . .”
We agree that the sentence “to be adjusted to a linear model” was unappropriated and was replaced by “a linear fit was applied to...” . See point 6 in the general response.

300. what did they distinguish?
This sentence was confusing and has been rephrased. See point 6 in the general response.

305. trends
We agree with this correction

312. I do not understand this sentence. How could Cant accumulation be related to denitrification? Denitrification does not change Cant.
We agree that denitrification does not change C_{ANT} accumulation , but we believe that it could lead to overestimations of C_{ANT} by the TrOCA method. Based on the suggestions of referee #1, we have rephrased this section to be clearer.
Finally, it should also be mentioned that, due to the presence of one of the main OMZ area, denitrification occurs in the eastern South Pacific and can be traced by the N parameter (Gruber and Sarmiento, 2007). Denitrification, by transforming organic carbon to inorganic carbon without consumption of oxygen, could induce an overestimation of C_{ANT} by the TrOCA method (and other back calculation methods) due to a biological release of C_T that is not corrected in the formulation of the quasi conservative TrOCA tracer. Horizontal advection by the south equatorial current of the strong negative N* signal originating from the Eastern Pacific towards the western Pacific was previously described (Yoshikawa et al., 2015). Fumenia et al. (2018) have estimated N* along the OUTPACE transect and show slightly negative N* values in the upper thermocline waters at the eastern side of the OUTPACE transect where the highest CANT values are estimated. However, Murata et al. (2007) showed that, based on a direct relationship between CT and N*, the influence of denitrification should be negligible on C_{ANT} estimations in this area. Therefore, the N* correction has not been introduced in the C_{ANT} estimates and the effect of denitrication was not quantified here.*

319. The change in the sampling grids means you can’t trust these linear fits in the MA region. Your measurements in MA are south of the P21 section where, being closer to the ventilation regions for AAIW and SAMW, you would expect higher Cant. The fact that your measurements are higher relative to P21 here than elsewhere is potentially attributable to that alone.
As we mentioned earlier, the estimated trends with GLODAP are not only based on the P21 section in the considered area. We agree that by being closer to the ventilation regions for AAIW and SAMW, we would expect higher C_{ANT}. However, a careful observation of Fig. 5(c), does not seem to indicate that the trend is due to the latitudinal position of the observations considered. Moreover, the trend is also observed without considering the OUTPACE dataset (see the new table 2).

325 is -> are
We agree with this correction.

Section 7. Here the authors compare their subsurface pHT changes to some surface pHT changes in literature. These are not valid comparisons because subsurface Cant is frequently lower and because the impact of Cant on pH is increased in the surbsurface where Revelle factors are higher.
We agree that these comparisons are subject to caution. However, the aim of this comparison was to compare the order of magnitude. We propose to add a sentence to point out the differences in buffer factors that exist between surface and subsurface.
These rates of acidification are higher than the values reported by Waters et al. (2011) in the Western South Pacific along the P06 Line (south of OUTPACE area at 32°S) between two visits in 1992 and 2008. They are also higher than the surface rates of pH_T decrease of –0.0016 ± 0.0001 a^{-1} recorded at the HOT time-series station in the tropical North Pacific and of –0.0017 ± 0.0001 a^{-1} and –0.0018 ± 0.0001a^{-1} in the tropical North Atlantic at BATS and ESTOC stations respectively (Bates et al., 2014). Differences in buffer factors between surface and subsurface can partially explain these differences. Nevertheless, our results in subsurface ($\sigma_{\theta 25}$) based on GLODAPv2 and OUTPACE data (C_T and A_T), are similar to pH_T trends derived from fCO2 surface observations (e.g. Lauvset et al, 2015).

328. why 20C?
20°C represents the mean temperature on the sigma level 25 (20.2 +- 0.7 °C). This has been added in the manuscript.

376. observations
We agree with this correction.

Table 3. Commas are used for decimal points at places in this manuscript while periods are used in other places.
We agree with the referee that a mixture of commas and periods were used for decimals in the submitted manuscript. We will use consistently periods for decimal points over the entire manuscript.

Figures. The section plots are tessellated (faint lines going everywhere on them), which is a problem that seems to happen for Matlab 2014b+ when exporting to vector graphics. Consider exporting to high resolution raster files instead. Please ignore if this is just a function of the review-proof.
We are not able to identify this problem on the section plots we have produced (Using the Generic Mapping Tools, GMT, Version 5.2.1, see http://gmt.soest.hawaii.edu/). However, we will of course correct any problems on the figures if the problem persists.

Colormaps: With the exception of Figure 6 (which would be impossible for people who are red-green colorblind to read), there are no changes to the colormaps that need to be made for this paper to be publishable. However, the authors should give this resource a read:
https://matplotlib.org/cmocean/
At the end of the webpage there are links to papers making the case that rainbow colormaps are not ideal for communicating science. The rest of the page is dedicated to providing alternatives
We thank the referee for sharing this interesting information on the use of color palettes. After reading some of the links suggested by reviewer, we have changed the color palettes on Figure 6 from red-green to blue-green which seems to be more adapted to colorblind readers. Concerning the rainbow color palette, we discovered with interest all the disadvantages of this none sequential color palette. However, we believe that, in the case of our figures, the rainbow color palette does not lead to a misinterpretation of the data that would justify a change.

Table 2 : Estimated trends on A_T, $[O_2]$, C_T , C_{ANT} and pH_T changes in two different layers of the water column defined by isopycnal layers between 1980 and 2015 based on GLODAPv2 with (column WITH) and without (column WITHOUT) OUTPACE data added. Estimated trends are obtained from slope values of a linear regression between the studied parameters and time.

	$25 < \sigma_\theta < 25.5$		$27 < \sigma_\theta < 27.2$	
	WITH	WITHOUT	WITH	WITHOUT
	Trend on A_T in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	-0.20 ± 0.07 (n = 167) *	-0.30 ± 0.07 (n = 142) *	-0.12 ± 0.07 (n = 180)	-0.01 ± 0.06 (n = 174)
MA	-0.30 ± 0.09 (n = 85) *	-0.47 ± 0.10 (n = 70) *	-0.16 ± 0.09 (n = 99)	-0.10 ± 0.09 (n = 92)
WGY	-0.20 ± 0.14 (n = 28)	-0.20 ± 0.19 (n = 22)	-0.20 ± 0.14 (n = 35)	-0.01 ± 0.13 (n = 31)
	Trend on $[O_2]$ in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	-0.31 ± 0.10 (n = 167)*	-0.61 ± 0.09 (n = 143)*	0.05 ± 0.11 (n = 183)	0.07 ± 0.10 (n = 178)
MA	-0.35 ± 0.16 (n = 84)*	-0.78 ± 0.17 (n = 70)*	0.06 ± 0.11 (n = 99)	0.04 ± 0.11 (n = 93)
WGY	-0.38 ± 0.11 (n = 27)*	-0.35 ± 0.14 (n = 23)*	-0.11 ± 0.30 (n = 38)	-0.22 ± 0.29 (n = 34)
	Trend on C_T in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	1.32 ± 0.13 (n = 174) *	1.63 ± 0.13 (n = 149) *	0.23 ± 0.13 (n = 189)	0.27 ± 0.11 (n = 183) *
MA	1.38 ± 0.21 (n = 85) *	1.87 ± 0.21 (n = 70) *	0.31 ± 0.16 (n = 100)	0.44 ± 0.17 (n = 93) *
WGY	1.57 ± 0.18 (n = 31) *	1.57 ± 0.23 (n = 25) *	0.23 ± 0.29 (n = 40)	0.23 ± 0.29 (n = 36)
	Trend on C_{ANT} in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	1.12 ± 0.07 (n = 166) *	1.25 ± 0.06 (n = 142) *	0.32 ± 0.05 (n = 179) *	0.25 ± 0.04 (n = 174) *
MA	1.18 ± 0.08 (n = 84) *	1.31 ± 0.08 (n = 70) *	0.40 ± 0.06 (n = 98) *	0.40 ± 0.06 (n = 92) *
WGY	1.20 ± 0.09 (n = 28) *	1.18 ± 0.10 (n = 22) *	0.13 ± 0.09 (n = 35)	0.11 ± 0.08 (n = 31)
	Trend on pH_{TINSI} in a^{-1}			
OUTPACE	-0.0022 ± 0.0003 (n=167) *	-0.0031 ± 0.0002 (n=142) *	-0.0001 ± 0.0003 (n=181)	-0.0002 ± 0.0002 (n=175)
MA	-0.0022 ± 0.0004 (n = 85) *	-0.0033 ± 0.0004 (n = 70) *	-0.0004 ± 0.0003 (n=100)	-0.0007 ± 0.0003 (n=93) *
WGY	-0.0027 ± 0.0004 (n = 28) *	-0.0030 ± 0.0004 (n = 22) *	-0.00008 ± 0.0006 (n=35)	-0.0007 ± 0.0006 (n=31)

* : trend significant (p-level < 0.05)

Table 3 : Estimated depth of the $\Omega_{ara} = 1$ horizon along the OUTPACE cruise (see text for details). No values are available for stations where data up to 2000 dbar were not available (SD2 and SD13). For the depth of the $\Omega_{ara} = 1$ horizon , no values were estimated for stations with $C_{ANT} < -6$ µmol kg^{-1}.

Station	Longitude	Latitude	Depth of the $\Omega_{ara} = 1$ horizon (in m)		
			OUTPACE	Pre-indu.	Difference*
SD1	159,.9425	-17.,9088	1225	NA	NA
SD2	162.,1248	-18,6078	NA	NA	NA
SD3	165,.0082	-19,.4907	928	NA	NA
A	164.,5787	-19.,2233	1032	1185	153
SD4	168,.0157	-19,.98	1029	1193	164
SD5	169.,9965	-21.,9997	1126	1256	130
SD6	172,.1193	-21,.3758	1097	1233	136
SD7	174.,2512	-20.,7677	1015	1235	220
SD8	176,.364	-20,.6945	1010	1171	161
SD9	178.,6087	-20.,9963	1214	NA	NA
SD11	-175,.6475	-20,.0057	1055	1172	117
SD12	-172.,7813	-19.,5368	1013	1112	99
B	-170,.7385	-18,.1745	948	1046	98
SD13	-169.,0728	-18.,2007	NA	NA	NA
C	-165,.7792	-18,.4842	854	941	87
SD14	-162.,9992	-18.,3952	889	1006	117
SD15	-159,.9913	-18,.2618	917	1043	126

* Difference (in m) between the depth of the $\Omega_{ara} = 1$ horizon at the pre-industrial period and the OUTPACE cruise.

Figures

Fig. 1: Map of the OUTPACE cruise transect. The outpace stations are distinguished between Melanesian Archipelago (MA) stations with darkgreen large dots and the Western GYre (WGY) stations with dark blue large dots. Stations outside of these two areas are in grey. The station with a red indication corresponds to the station where the deep cast and intercomparaison cast was made. Station from the GLODAPv2 database are indicated with small cross: small green dots correspond to GLODAPv2 stations considered for comparaison in the MA area, small blue dots correspond to GLODAPv2 stations considered for comparaison in the WGY area and small grey dots are the other GLODAPv2 stations considered for comparaison.

Fig. 6: Longitudinal variations of (a) pH_T changes and (b) Ω_{ara} changes between pre-industrial and present time along the OUTPACE transect between surface and 2000m depth (See text for details). Black contour lines represent the isopycnal horizons based on potential density referenced to a pressure of 0 dBar.

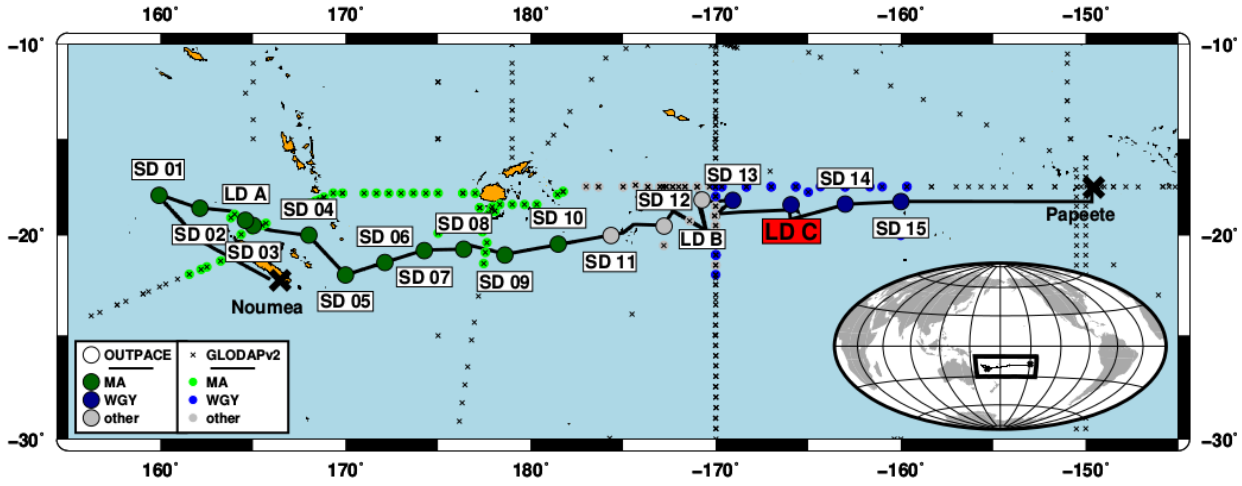
160°
170°
180°
−170°
−160°
−150°
−10°
−20°
−30°
SD 01
LD A
SD 04
SD 02
SD 03
SD 06
SD 08
SD 10
SD 12
SD 13
SD 14
LD B
LD C
SD 15
Papeete
Noumea
SD 05
SD 07
SD 09
SD 11
OUTPACE
MA
WGY
other
GLODAPv2
MA
WGY
other

Figure 1

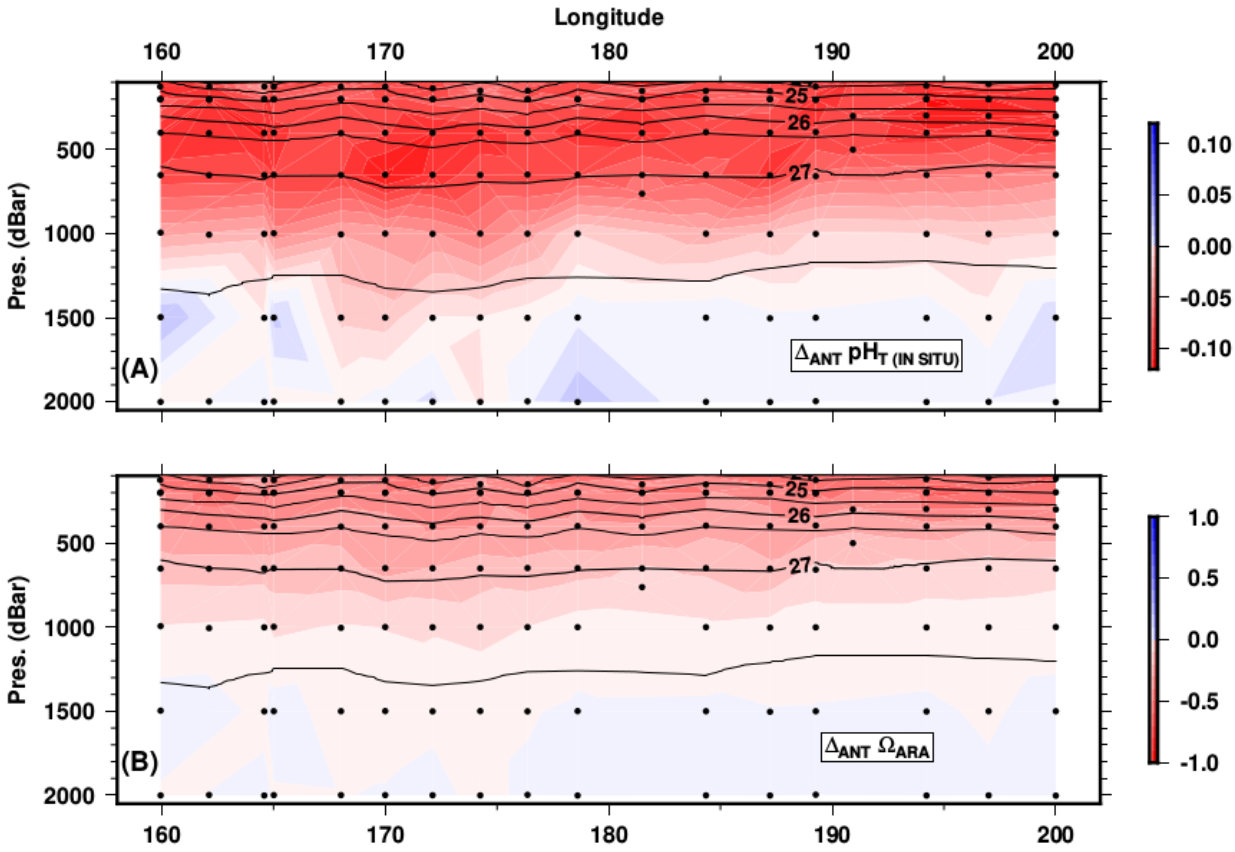
Longitude
160
170
180
190
200
Pres. (dBar)
500
1000
1500
2000
25
26
27
(A)
Δ_{ANT} $pH_{T\ (IN\ SITU)}$
0.10
0.05
0.00
-0.05
-0.10
(B)
Δ_{ANT} Ω_{ARA}
1.0
0.5
0.0
-0.5
-1.0

Figure 6

Carbonate system distribution, anthropogenic carbon and acidification in the Western Tropical South Pacific (OUTPACE 2015 transect)

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Abstract.

The western tropical South Pacific was sampled along a longitudinal 4000 km transect (OUTPACE cruise, 18 Feb., 3 Apr. 2015) for measurement of carbonates parameters (total alkalinity and total inorganic carbon) between the Melanesian Archipelago (MA) and the western part of the South Pacific gyre (WGY). This manuscript reports this new dataset and derived properties: pH on the total scale (pH_T) and the $CaCO_3$ saturation state with respect to calcite (Ω_{cal}) and aragonite (Ω_{ara}). We also estimate anthropogenic carbon (C_{ANT}) distribution in the water column using the TrOCA method (Tracer combining Oxygen, inorganic Carbon and total Alkalinity). Along the OUTPACE transect, C_{ANT} inventories of 37 – 43 mol m^{-2} were estimated with higher C_{ANT} inventories in MA waters (due to a deeper penetration of C_{ANT} in the intermediate waters was observed in the MA than in the WGY waters), although whereas highest C_{ANT} concentrations were detected in the sub-surface waters of the WGY. By combining our OUTPACE dataset with data available in GLODAPv2 (1974-2009), temporal changes in oceanic inorganic carbon were evaluated. An increase of 1.3 to 1.6 µmol kg^{-1} a^{-1} for total inorganic carbon in the upper thermocline waters is estimated whereas C_{ANT} increases of 1.1 to 1.2 µmol kg^{-1} a^{-1}. In the MA intermediate waters (27 kg m^{-3} < σ_θ < 27.2 kg m^{-3}) an increase of 0.4 µmol kg^{-1} a^{-1} of C_{ANT} is detected. Our results suggest a clear progression of ocean acidification in the western tropical South Pacific with a decrease of the oceanic pH_T of up to -0.0027 a^{-1} and a shoaling of the saturation depth for aragonite of up to 200 m since the pre-industrial period.

1 Introduction

Human activities inject about 10^{13}10 PgC kg of carbon per year to the atmosphere which might have major consequences on climate. It is recognized that the ocean plays a key role in the control of atmospheric CO_2 through uptake by the so called "oceanic carbon pump". Through this "pump", the ocean sequesters ca. 3025% of the CO_2 injected annually in the atmosphere by human activities (Le Quéré et al., 2018). A consequence of the ocean carbon uptake is a decrease of the

oceanic pH (Feely et al, 2004) which is described as ocean acidification (the so-called "other" CO_2 problem). Effects of ocean acidification have been observed on marine organisms and could affect the marine ecosystems (Riebesell et al., 2000). Improving our understanding of the oceanic CO_2 uptake relies primarily on observations of the marine carbonate cycle. Studies on the oceanic carbonate cycle have been mostly conducted in the frame of international programs. The World Ocean Circulation Experiment (WOCE) and the Joint Global Flux Study (JGOFS) in the 90's have coordinated oceanographic cruises along large sections in the ocean to collect samples through the water column and to perform accurate measurements of carbonate parameters and ancillary parameters (temperature, salinity, dissolved oxygen, nutrients,...). Since 2000, efforts have been made to "revisit" oceanic sections according to the WOCE strategy in order to assess oceanic changes at the scale of a decade. These programs have generated important databases for oceanic carbonate chemistry (e.g. GLODAP_V2 – Olsen et al., 2016).

In order to better assess the role of the ocean on the global carbon cycle, the concept of oceanic anthropogenic carbon (~~hereafter named~~ C_{ANT}) has been introduced and refers to the fraction of dissolved inorganic carbon (C_T) in the ocean that originates from carbon injected by human activities in the atmosphere since the industrial revolution. As C_{ANT} is not a directly measurable quantity, it can only be estimated through assumptions that are subjected to intense scientific debate (Sabine and Tanhua, 2010). In particular, it has been recently recognized that ocean circulation changes drive significant variability in carbon uptake (De Vries et al., 2017). Detecting, separating and attributing decadal changes of the carbonates system (C_T and A_T), C_{ANT} and pH in the ocean at global or regional scales remains challenging.

Within this context, the Pacific Ocean is a particularly challenging area to study due to its size (ca. one third of the Earth's and one half of the oceanic surface). Even if, due to its remoteness from land, it remains largely under-explored by oceanographic vessels compared to other oceanic areas, the Pacific Ocean has been covered by cruises along long sections (the "P sections" from the WOCE program). Most of these sections have been revisited during the last years (see for example Sabine et al., 2008 or Kouketsu et al., 2013). In a recent study based on repeated sections in the Pacific (P16 at 150W), Carter et al (2017) observed significant increase of Cant in the top 500m around 10°S-30°S and a local carbon storage maximum around 20°S in recent years (between 2005 and 2014). In this context, the OUTPACE data presented in this study, associated to historical observations (since the pioneer 1974 GEOSECS) offer a new view to evaluate variability and decadal changes of C_T, C_{ANT} and pH in the tropical Pacific, here focused in the western tropical south pacific (WTSP).

The aim of this paper is to report a new dataset of oceanic inorganic carbon (based on measurements of C_T and total alkalinity (A_T)) acquired in the WTSP during the OUTPACE (Oligotrophic to UltTra oligotrophic PACific Experiment) cruise performed in 2015 (Moutin et al., 2017). The main focus of the OUTPACE cruise was to study the complex interactions between planktonic organisms and the cycle of biogenic elements on different scales, motivated by the fact that the WTSP has been identified as a hot spot of N_2 fixation (Bonnet et al., 2017). The data presented here have been partially used in another paper of the special issue (Moutin et al., 2018) in order to study the biological carbon pump in the upper (surface to 200m) water column. In this paper we will explore the carbonate data between the surface and 2000 m depth. The OUTPACE transect (Figure 1) is close to existing WOCE and GO-SHIP lines in the South Pacific : it is parallel to the zonal

P21 line (18° S visited in 1994 and 2009) and the P06 line (32° S visited in 1992, 2003 and 2010), it is crossed by the meridional P14 line (180° E visited in 1994 and 2007) and P15 line (170° W visited in 2001, 2009 and 2016) and it is situated at the eastern side of the P16 line (150° W visited in 1992, 2005 and 2014). However, the OUTPACE transect is not corresponding to any earlier occupation of the "WOCE lines" in the South Pacific and no tracers of water mass age were measured during the cruise, which limits the possibilities of a robust analysis of C_{ANT} accumulation in the area. Moreover, the horizontal and vertical resolution of the OUTPACE dataset is low. In consequence, the OUTPACE dataset can not be used to look at decadal changes in C_{ANT} content in the South Pacific (e.g. Carter et al. 2017, Kouketsu et al. 2013). Here, C_{ANT} estimates based on the TrOCA (Tracer combining Oxygen, inorganic Carbon and total Alkalinity) method will be used as as a tool to investigate changes in C_T. Moreover, ~~This will allow us to~~by compar~~e~~ing our data with ~~these~~the high quality data (internally consistent through a secondary quality control (Olsen et al., 2016)) available in the Global Ocean Data analysis Project version 2 (GLODAPv2 database), will allow ~~and~~to evaluate ~~both~~ C_T, A_T, C_{ANT} (for TrOCA) and pH_T (pH on total scale) trends in sub surface waters and at depth.

The paper is organized as follows: After describing the methods used to acquire the dataset and the way the auxiliary data have been used in Sect. 2, we briefly present the hydrographic context of the cruise in Sect. 3. We then present in Sect. 4, the carbonate dataset acquired during the cruise. In Sect. 5, estimated C_{ANT} values in the water column are presented, the validity of these estimates based on the TrOCA ~~(Tracer combining Oxygen, inorganic Carbon and total Alkalinity)~~ method is discussed and geographical patterns are evoked. In Sect. 6, the temporal changes in oceanic inorganic carbon in the WTSP combining data available in GLODAPv2 and our OUTPACE dataset are presented and discussed. Finally, in Sect. 7, some features in relation to ocean acidification are inferred from our dataset.

2 Material and Methods

2.1 Cruise and sampling strategy

The OUTPACE cruise took place between 18 February and 3 April 2015 from Noumea (New Caledonia) to Papeete (French Polynesia), in the WTSP on board the French research vessel "L'Atalante" (Fig. 1). A total of 18 stations were sampled mostly in the top 2000 m of the water column along a ~4000 km transect from the Melanesian archipelago to the South Pacific gyre (Moutin et al., 2017). A CTD-Rosette was deployed ~~(1)~~ to acquire data with CTD and associated sensors along vertical profiles and ~~(2)~~ to collect discrete seawater samples from 24 12-L Niskin bottles for chemical analysis. Due to technical failures on the main CTD-Rosette, for two of the casts considered in this study, a trace metal clean CTD rosette (TM-R) equipped with 24 teflon-lined GO-FLO bottles devoted to trace metal analyses was used. The configurations of both CTD Rosettes are detailed elsewhere (Moutin et al., 2017).

For carbonate parameters, seawater was sampled from 31 casts over the 18 stations. At each station, on a regular basis, samples were collected at 12 depths between the surface and 2000 m on two distinct casts: 6 samples on a 0-200 m cast and 6 samples on a 0-2000 m cast. At station SD-13, only one cast was sampled down to 500 m depth. In addition, at station LD-

C, samples were collected at 24 depths on a deep cast (down to 5000 m) and 12 samples were collected at the same depth (25 m) on a "repeatability" cast. Details on the casts performed for this study are summarized in Table 1.

2.2 Chemical measurements on discrete samples

All samples were collected within less than 1 hour after arrival of the CTD rosette on deck.

2.2.1 Total alkalinity and dissolved inorganic carbon

Samples for A_T and C_T were collected in one 500 mL borosilicate glass flask (Schott Duran®) and poisoned immediately after collection with $HgCl_2$ (final concentration 20 $mg.L^{-1}$). Samples were stored at 4°C during transport and were analyzed (within 10 days) 5 months after the end of the cruise at the SNAPO-CO_2 (Service National d'Analyse des paramètres Océaniques du CO_2- LOCEAN – Paris). A_T and C_T were measured on the same sample based on a potentiometric titration in a closed-cell (Edmond, 1970). A non-linear curve fitting approach was used to estimate A_T and C_T from the recorded titration data (Dickson 1981, DOE 1994). Measurements were calibrated with Certified Reference Material (CRM) provided by Dr. A Dickson, Univ. Southern California (Batch 139 - C_T : 2023.23 ± 0.70 µmol kg^{-1} and A_T: 2250.82 ± 0.60 µmol kg^{-1}, see Dickson, 2010). The reproducibility, expressed as the standard deviation of the CRM analysis (n=15), was 4.6 µmol kg^{-1} for A_T and 4.7 µmol kg^{-1} for C_T . Based on replicate measurements at station LD-C (cast out_c_194, see Table 1) the reproducibility, expressed as the standard deviation of the analysis of the replicates collected at the same depth (ca. 25 m, n=12) from different Niskin bottles was 3.6 µmol kg^{-1} for A_T (average value = 2324.7 µmol kg^{-1}) and 3.7 µmol kg^{-1} for C_T (average value = 1969.7 µmol kg^{-1}).

2.2.2 Oxygen concentration

Dissolved oxygen concentration [O_2] was measured following the Winkler method (Winkler, 1888) with potentiometric endpoint detection (Oudot et al., 1988). For sampling, reagents preparation and analysis, the recommendations from Langdon (2010) were carefully followed. The thiosulfate solution was calibrated by titrating it against a potassium iodate certified standard solution of 0.0100N (CSK standard solution – *WAKO*). The reproducibility, expressed as the standard deviation of replicates samples was 0.8 µmol kg^{-1} (n=15, average value= 195.4 µmol kg^{-1}).

2.3 Vertical profiles of hydrological and biogeochemical parameters

2.3.1 CTD measurements

CTD measurements were ensured by a Seabird™ 911+ underwater unit which interfaced an internal pressure sensor, two redundant external temperature probe (SBE3plus) and two redundant external conductivity cells (SBE4C). The sensors were calibrated pre- and post-cruise by the manufacturer. No significant drift between the redundant sensors was observed. For vertical profiles, full resolution data (24 Hz) were reduced to 1 dbar binned vertical profiles on the downcast with a suite of

processing modules using the Seabird™ dedicated software (*SbeDataProcessing*). For values at the closure of the Niskin bottles, values collected at 24 Hz were averaged 3 s before and 5 s after closure of the bottle. In this study, for temperature and conductivity the signal of the first sensors has been systemically used. For the two TM-R casts, no significant difference with the main CTD-Rosette on temperature and conductivity was observed.

2.3.2 Oxygen measurements

$[O_2]$ was also measured with a SBE43 electrochemical sensor interfaced with the CTD unit. The raw voltage was converted to oxygen concentration with 13 calibration coefficients based on the Seabird™ methodology derived from Owens and Millard (1985). Three of these coefficients (the oxygen signal slope, the voltage at zero oxygen signal, the pressure correction factor) were adjusted with the concentrations estimated with the Winkler method on samples collected at the closure of the bottles. One unique set of calibration coefficients has been applied to all oxygen profiles from the cruise because no significant drift of the sensor was observed during the time of the cruise. For the two TM-R casts, values have been corrected with a drift and offset based on the comparison of 15 pairs of casts (main CTD-rosette / TM-R) collected close in time (less than 2 h) and space (less than 1 nautical mile) over the entire OUTPACE transect.

2.4 Derived parameters

Practical salinity (S_P) was derived from conductivity, temperature and pressure with the EPS-78 algorithm. Absolute salinity (S_A), potential temperature (θ), conservative temperature (Θ) and potential density (σ_θ) were derived from Sp, temperature, pressure and the geographic position with the TEOS-10 algorithms (Valdarez et al., 2011). Thesesis five derived parameters were calculated within the processing with *Seabird*™ dedicated software.

Seawater pH on the total scale (pH_T) and the $CaCO_3$ saturation state with respect to ~~calcite (Ω_{cal}) and~~ aragonite (Ω_{ara}) were derived from A_T and C_T with the "Seacarb" R package (Gattuso and Lavigne, 2009). $CaCO_3$ saturation state with respect to calcite was not considered because seawater up to 2000 dbar was supersaturated with respect to calcite ($\Omega_{cal}>1$). Following the recommendations from Dickson et al. (2007), the constants for carbonic acid K_1 and K_2 from Lueker et al. (2000), the constant for hydrogen fluoride K_F from Perez and Fraga (1987) and the constant for hydrogen sulfate K_S from Dickson (1990) were used. Orthophosphate and silicate concentration were considered in the calculation. Methods for nutrients measurement are presented in details in Fumenia et al. (2018). When nutrient data were not available (Station SD-8), silicate and orthophosphate were estimated from the nutrient profile measured on cast out_c_163 (interpolated values). Apparent Oxygen Utilization (AOU) was computed from the difference between oxygen solubility (at p=0 dbar, θ and S_P) estimated with the "Benson and Krause coefficients" in Garcia and Gordon (1992) and in situ $[O_2]$.

For estimation of C_{ANT}, the TrOCA method was used. The TrOCA approach was first proposed in Touratier and Goyet (2004a, b) with improvements in Touratier et al. (2007). In brief, the TrOCA parameter is defined as a combination of A_T, C_T and $[O_2]$ that accounts for biologically induced relative changes among these parameters (with constant stoichiometric ratios). TrOCA is thus a quasi-conservative tracer derived from C_T in the ocean. Within a defined water mass, changes in

TrOCA over time are independent from biology and can be attributed to the penetration of C_{ANT}. In consequence C_{ANT} can be calculated in a parcel of water from the difference between current and pre-industrial TrOCA (TrOCA°) divided by a stoichiometric coefficient. The simplicity of the TrOCA method relies on the fact that a simple formulation for TROCA° has been proposed based on potential temperature and alkalinity and thus an estimation of C_{ANT} can be done by a simple calculation using C_T, A_T, $[O_2]$ and θ. In this study, the formulation proposed in eq. 11 in Touratier et al. (2007) is used to calculate C_{ANT} and is reminded here in Eq. (1).

$$C_{ANT} = \frac{[0_2] + 1.279\left(C_T - \frac{A_T}{2}\right) - \exp\left(7.511 - 1.087.10^{-2}\theta - \frac{7.81.10^{-5}}{A_T^2}\right)}{1.279} \quad (1)$$

This formulation is based on an adjustment of the TrOCA coefficients using $\Delta^{14}C$ and CFC-11 from the GLODAP-V1 database (Key et al., 2014). Touratier et al. (2007) estimated the overall uncertainty of the C_{ANT} with TrOCA method to c.a. 6 µmol kg^{-1} based on the random propagation of the uncertainties on the variables (C_T, A_T, $[O_2]$ and θ) and coefficients used in Eq. 1. The limitations and validity of the TrOCA method will be discussed in details in Sect. 5.

2.5 Data from available databases

For comparison with existing values of carbonate chemistry in the area of the OUTPACE cruise, relevant data were extracted from GLODAPv2 database (NDP-93 - Olsen et al. 2016). The specific data file for the Pacific Ocean was used (downloaded from https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2/ on December 14, 2017). For comparison with OUTPACE data, GLODAPv2 data were selected between 22°S and 17°S and between 159°E and 159° W (going westwards). For specific comparisons in the Melanesian archipelago (MA) and the South Pacific Western Gyre waters (WGY) a zonal subset of the extracted data was used: 159°E and 178° W for MA and 170°W to 159°W for WGY (see Fig. 1).

3 Hydrological context along the OUTPACE transect

The hydrological context encountered during the OUTPACE transect is presented with a $\Theta - S_A$ diagram between 0 and 2000 dbar on Fig. 2. A detailed description of the water masses encountered during the OUTPACE cruise can be found in Fumenia et al. (2018). Briefly, from the surface to 2000 dbar, the following features are distinguished: the surface waters ($\sigma_\theta < 23.5$) were characterized by temperatures over 25 °C with increasing temperature and salinity towards the east and AOU close to zero. Under the the surface water, the upper thermocline waters (UTW) presented a maximum in salinity reaching values higher than 36 g kg^{-1} in the eastern part of the cruise. In the lower thermocline waters, S_A decreased with depth with a more pronounced decrease in the eastern part than in the western part whereas AOU is higher in the eastern part than in the western part of the studied area. These differences in lower thermocline waters have been described for South Pacific Central Waters (SPCW) with more saline western (WSPCW) and less saline eastern (ESPCW) waters (Tomczack and Godfrey, 2001). Below the thermocline, intermediate waters are constituted of Sub-Antarctic Mode Waters (SAMW) and Antarctic

Intermediate Waters (AAIW). AAIW have a salinity minimum close to the σ_θ = 27 kg m^{-3} isopycnal . Hartin et al. (2011) defines SAMW with σ_θ values between 26.80 and 27.06 kg m^{-3} corresponding to a minimum in potential vorticity, and AAIW with σ_θ values between 27.06 and 27.40 kg m^{-3}. The separation of both waters is not trivial in the subtropical area. SAMW is generally associated to lower AOU than AAIW. Finally deep waters constituted of Upper Circumpolar Deep Waters (UCDW) correspond to an increase in salinity and AOU for depth corresponding to $\sigma_\theta > 27.4$ kg m^{-3}.

In this study, discussion will sometimes make distinction between two sub-regions along the OUTPACE transect: MA and WGY (See Sect. 2.5 for definition). This distinction is mainly based on geographic and oceanographic arguments. Indeed, these two sub-regions are geographically separated by the Tonga volcanic arc. WGY is characterized by higher surface temperature and a higher salinity in the upper thermocline waters than MA. The difference between these sub-regions is evidenced by the difference in oligotrophy (Moutin et al., 2018). Due to specific conditions in the transition area between the MA and WGY (de Verneil et al., 2017), SD11, SD12 and LDB were discarded from both groups in this study following the arguments in Moutin et al. (2018).

4 Carbonate chemistry along the OUTPACE transect

A_T and C_T measured along the OUTPACE transect are presented on Fig 3a and 3b. All vertical profiles for A_T, A_T normalized to S_A = 35 g kg^{-1} ($A_{T\,n35}$) and C_T are presented on Fig. 3e, 3f and 3G. A_T ranged between 2300 and 2400 µmol.kg^{-1}. Below the surface, a pronounced maximum in A_T was observed associated to the saltier upper thermocline waters. When normalized to S_A = 35 g kg^{-1}, A_T values are remarkably constant in the upper 500 dbar with values between 2270 and 2310 µmol kg^{-1}. Below 500 dbar, A_T increases with depth up to ca. 2400 µmol kg^{-1} indicating that alkalinity changes are mostly due to salinity changes in the upper water column whereas the increase in the deep waters is mainly due to carbonate biominerals remineralization. C_T values are close to 1950 µmol kg^{-1} in the surface and increase with depth up to 2300 µmol kg^{-1} at 2000 dbar. The C_T gradient in the upper water column has been described in Moutin et al. (2008). Below 2000 dbar, C_T is relatively invariant with slightly lower values in the bottom waters (below 4000 dbar) due to the presence of very old deep waters originating from the north Pacific relative to the northward moving bottom waters that have not accumulated as much carbon (Murata et al. 2007). A_T and C_T values in deep waters measured during OUTPACE are in good agreement with the data of the GLODAPv2 database (Fig. 3E, 3f et 3g). No systematic adjustment of the OUTPACE dataset with the GLODAPv2 dataset was performed because only very few data are available in the deep ocean where crossover comparison can be performed for cruises carried out in different decades. Nevertheless, for the only "deep" cast performed during OUTPACE (out_c_163 at station C), we performed a simple crossover analysis with the station 189 (located at 107 km kilometers from OUTPACE station C) of the Japanese "P21 revisited" cruise in 2009. We compared interpolated profiles on density surfaces values ($27.75 < \sigma_\theta < 27.83$ corresponding to pressure levels of ca. 3000 to 5500 dbar) . The estimated offsets are -2.0 ± 4.2 µmol kg^{-1} for A_T and -2.0 ± 4.4 µmol kg^{-1} for C_T ~~lies within the repeatability of method (see Sect. 2).~~ suggesting

measurement biases are likely no larger. This simple quality control procedure seems to indicate that no systematic adjustment is needed.

Derived parameters from the A_T and C_T measurements are presented on Fig. 3c for pH_T values (estimated at in situ temperature and pressure). pH_T decreases from values close to 8.06 in surface to values close to 7.84 at 2000 m. Surface values of pH_T are typical of subtropical warm waters and are in a similar range as the austral summer values estimated by Takahashi et al. (2014) in this area (8.06 - 8.08). Figure 3d represents the vertical distribution of computed values of Ω_{ara} along the OUTPACE transect. Seawater is supersaturated with respect to aragonite ($\Omega_{ara}>1$) at surface with Ω_{ara} values of ca. 4.0 again in good agreement with the austral summer values of 4 – 4.4 estimated by Takahashi et al. (2014) in this area. Values of Ω_{ara} decrease with depth and seawater becomes undersaturated with respect to aragonite ($\Omega_{ara}<1$) at an horizon situated below 1000 dbar in the west and above 1000 dbar in the eastern part of the cruise, with a general shoaling of the Ω_{ara} values from west to east, in good agreement with a previous study by Murata et al. (2015) in this area. ~~As expected, values for Ω_{cal} show the overall same pattern as Ω_{ara} with the important difference that seawater up to 2000 dbar was supersaturated with respect to calcite ($\Omega_{cal}>1$) (data not shown). At the deep cast, seawater becomes undersaturated with respect to calcite ($\Omega_{cal}<1$) at an horizon situated below 3000 dbar.~~

5 Anthropogenic carbon estimation along the OUTPACE transect

The TrOCA method is a way to quantify C_{ANT} in the ocean based on C_T, A_T, $[O_2]$ and θ. This method has been used and compared to other methods in different oceanic areas (e.g. Lo Monaco et al., 2005; Alvarez et al., 2009; Vazquez-Rodriguez et al.,2009) : based on specific C_{ANT} inventories in the water column, the TrOCA method reasonably agreed with the other methods (including transient tracer based method). However, Yool et al. (20~~09~~10) "tested" the TrOCA method within an ocean general circulation model and argued that the use of globally uniform parametrization for the estimation of the preindustrial TrOCA is a source of significant ~~errors~~overestimation but also that even with regionally "tunned" parameters a global positive bias in the method exists. As no tracers of water mass age were measured during the OUTPACE cruise, the main motivation for using the TrOCA method was to make C_{ANT} estimations based on a simple calculation from parameters acquired within the cruise as done in other cruises conducted in south tropical Pacific waters (e.g. Azouzi et al., 2009; Ganachaud et al., 2017). Even if C_{ANT} estimates from TrOCA could be biased, the application of a simple back-calculation method that accounts for biologically induced relative changes in C_T is used here to identify some spatial features in the distribution of the carbonate system along the OUTPACE transect. Based on Yool et al. (2010), the error on the TrOCA C_{ANT} estimates will be considered here as to the normalized standard deviation of 1,67 for the TrOCA variant optimized with world ocean data (See Table 2 in Yool et al. 2010).

As mentioned by Touratier et al. (2007), C_{ANT} estimates cannot be considered within the mixed layer because the underlying hypotheses used in the formulation of TrOCA may not be verified due to biological activity and gas transfers across the air–sea interface. To avoid this issue, C_{ANT} estimates are generally used below the "permanent" mixed layer depth (e.g. Alvarez

et al., 2009, Carter et al., 2017). For the OUTPACE area, Moutin et al. (2018) shows that the mixed layer depth do not exceed 70 m in the area. Even if the depths of the deep chlorophyll maximum was encountered below 100 dbar along the transect, we will consider C_{ANT} values up to 100 dbar. It can be mentioned that the C_{ANT} values of 50-60 µmol kg^{-1} in the top of the water column (100 dbar), are in reasonable agreement with a rough estimate of thermodynamic consistent C_T changes: by assuming that CO_2 in surface seawater is in equilibrium with the atmosphere, we estimated that with a partial pressure of CO_2 (pCO_2) of 280 µatm at the pre-industrial period, a pCO_2 of 380 µatm during OUTPACE (Moutin et al., 2018) and a constant A_T over time of 2300 µmol kg^{-1}, C_T change in surface waters between pre-industrial and 2015 is of ca. 65 µmol kg-1 for a temperature of surface waters between 25 and 28 °C. For OUTPACE, C_{ANT} estimates below 1000 dbar, were not significantly different from 0 µmol kg-1 with a standard deviation of 6.3 µmol kg^{-1}~~, in the range of the error estimate~~.

C_{ANT} distribution along the OUTPACE transect is presented on Fig. 4a and all vertical profiles for C_{ANT} are presented on Fig. 4b with a more detailed view of the first 1500 dbar of the water column on Fig. 4c. Figures 4b and 4c distinguish values from the MA and the WGY area. The C_{ANT} vertical profiles suggest a penetration of anthropogenic carbon up to 1000 dbar. As mentioned before, estimated values of C_{ANT} reach values of 60 ± 40 µmol kg^{-1} at depth of 100 dbar, then regularly decreases to values close to 10 - 20 ± 13 µmol kg^{-1} at a depth of 1000 dbar and reaches values close to 0 µmol kg^{-1} below 1500 dbar. The zonal C_{ANT} section along the OUTPACE transect (Fig. 4a) presents two ~~singularities~~features: (1) a deeper penetration of C_{ANT} in the western part of the transect with values of C_{ANT} reaching 40 µmol kg^{-1} around the isopycnal layer of 27 kg m^{-3} (ca. 700 dbar) with a coherent behavior with the distribution of AOU and (2) a larger accumulation of C_{ANT} in the eastern part of the transect centered around the isopycnal layer of 25 kg m^{-3} (ca. 200 dbar). ~~This singularity in the distribution of C_{ANT} is reflected in the estimated inventories in the 0 - 1000 dbar water ($C_{ANT}INV_{1000}$) and 0- 2000 dbar ($C_{ANT}INV_{2000}$) water column presented on Table 2 : for stations in the MA area $C_{ANT}INV_{1000}$ represents between 82% and 90% of the $C_{ANT}INV_{2000}$, whereas it represents more than 90% in the WGY area indicating a lower penetration depth of C_{ANT} in the eastern part of the cruise.~~

Several studies have identified deeper C_{ANT} penetration in the Western South Pacific than in the Eastern South Pacific at tropical and subtropical latitudes. The primary reason for this longitudinal difference might be associated to deeper convection in the western part and upwelling in the eastern part. AAIW has been described as the lower limit of the penetration of C_{ANT} in the ocean interior of the South Pacific (Sabine et al., 2004). Moreover, a recent study by deVries et al., (2017) shows that ocean circulation variability is the primary driver for changes in oceanic CO_2 uptake at decadal scales. Based on C_T changes between the two repeated visits of the longitudinal P21 line (18° S close to the OUTPACE transect) in 1994 and 2009, Kouketsu et al. (2013) shows faster increase of C_{ANT} in the western part than in the eastern part of the section. They also postulate that C_{ANT} may have been transported by deep circulation associated to the AAIW. In the subtropical Pacific along the P06 line (longitudinal section at ca. 32°S), Murata et al. (2007), also identified an increase of C_{ANT} in the SAMW and AAIW. Waters et al. (2011), based on the extended multiple linear regression (eMLR) method along the P06 line (and taking into account a third visit) attributes the deeper penetration of C_{ANT} in the western part of the section to the local formation of subtropical mode water in the area.

In the eastern part of the OUTPACE cruise, the detected accumulation of C_{ANT} in the upper thermocline waters may be related to recent observations of a significant accumulation of C_{ANT} at latitudes around 20°S on the P16 meridional transect along 150°W by Carter et al. (2017). This change in C_{ANT} accumulation is attributed to changes in the degree of the water mass ventilation due to variability in southern Pacific subtropical cell. Along the P16 line, Carter et al. (2017) observed high values of C_{ANT} (up to 60 µmol.kg^{-1}) for the upper water column at the latitude of OUTPACE area in good agreement with our estimates in WGY in the upper water column. Finally, ~~I~~it should also be mentioned that, due to the presence of one of the main OMZ area, denitrification occurs in the eastern South Pacific and can be traced by the N* parameter (Gruber and Sarminento, 2007). Denitrification, by transforming organic carbon to inorganic carbon without consumption of oxygen, could induce an overestimation of C_{ANT} by the TrOCA method (and other back calculation methods) due to a biological release of C_T that is not taken into account in the formulation of the quasi conservative TrOCA tracer. Horizontal advection by the south equatorial current of the strong negative N* signal originating from the Eastern Pacific towards the western Pacific was previously described (Yoshikawa et al., 2015). Fumenia et al. (201~~0~~8) have estimated N* along the OUTPACE transect and show slightly negative N* values in the upper thermocline waters at the eastern side of the OUTPACE transect where the highest C_{ANT} values are estimated. However, Murata et al. (2007) showed that, based on a direct relation between C_T and N*, the influence of denitrification should be negligible on C_{ANT} estimations in this area. Therefore, the N* correction has not been introduced in the C_{ANT} estimates and the effect of denitrification was not quantified here.

~~7) estimated a recent year column inventory change of up to 0.8 mol m^{-2} a^{-1} at 20°S along the P16 line (170°W). When adding the C_{ANT} inventories of the early 90s to the changes in inventories of the recent years, our inventories $C_{ANT}INV_{2000}$ are slightly higher but in reasonable agreement. Our results are also in the range of C_{ANT} inventory evaluated around 40 - 50 mol m^{-2} in this region for a reference year 2010 (Khatiwala et al., 2013). 0Finally, if we consider that all C_{ANT} is distributed in the upper 2000 dbar of the water column, our $C_{ANT}INV_{2000}$ of 43 ± 4 mol m^{-2} for MA and 37.3 ± 0.4 mol m^{-2} for WGY (Table 2) can be considered as water column C_{ANT} inventories. The C_{ANT} is signigicantly higher in MA than in WGY. The C_{ANT} concentrations during OUTPACE are about twice the C_{ANT} estimated in 1994 (P21 line, Sabine et al., 2002; their Fig. 5) leading to a doubling of C_{ANT} inventory, from about 20 mol m^{-2} in 1994 to 40 mol m^{-2} in 2015. Sabine et al., (2008) estimated a column inventory change of 0.5 mol m^{-2} a^{-1} for the years 2000 and Carter et al. (20~~

6 Temporal changes of ~~inorganic carbon~~carbonate chemistry in the OUTPACE area

Based on the available GLODAPv2 data, temporal changes in the OUTPACE area have been assessed (Fig. 5 and Table 3). The variation of oceanic parameters with time are estimated on two isopycnal layers : A layer with 25 kg m^{-3} < σ_θ < 25,5 kg m^{-3} (hereafter named $\sigma_{\theta 25}$) and a layer with 27 kg m^{-3} < σ_θ < 27.2 kg m^{-3} (hereafter named $\sigma_{\theta 27}$). These two layers correspond to the features~~singularities~~ in C_{ANT} discussed in the former section. $\sigma_{\theta 25}$ can be considered as characteristic of the upper thermocline waters (core of the salinity maximum, Fig 2) whereas $\sigma_{\theta 27}$ can be considered as characteristic of intermediate waters of southern origin (core of the salinity minimum). All the values associated to these two layers are spread between

145 and 301 dbar for $\sigma_{\theta 25}$ and between 571 and 896 dbar for $\sigma_{\theta 27}$. It must be mentioned that the study of temporal changes is based on a large sampling grid which covers the entire OUTPACE transect (see Sect. 2.5. and Fig. 1). This could add a spatial variability that may interfere in the estimation of temporal changes.

Temporal variations of C_T and C_{ANT} between 1970 and 2015 are presented on Fig 5. As mentioned earlier, even if C_{ANT} estimates from TrOCA could be biased, a former study by Perez et al. 2009 suggests that the TrOCA method gives similar values than other methods for estimating C_{ANT} accumulation rates. A linear fit was applied to ~~T~~the observed temporal variations for A_T, $[O_2]$, C_T and C_{ANT} ~~were adjusted to a linear model~~ to check for significant trends on data collected between 1980 and 2015 ~~(OUTPACE cruise)~~. The results of the performed regression analyses are presented on table 2. Trends are evaluated with and without the data of the OUTPACE cruise in order to estimate the influence of this new dataset on the observed trends. Trends are evaluated for the entire OUTPACE area and ~~Values from~~for ~~-~~the MA and the WGY area ~~distinguished~~. Even if presented on Figure 5, data collected before 1980 from the GLODAPv2 database are disregarded in the estimation of the temporal trends. Indeed, for the OUTPACE area, data prior to 1980 originate from one single GEOSEC cruise in 1974, with only one measured point for $\sigma_{\theta 27}$ at WGY and no points at $\sigma_{\theta 25}$ for WGY and WMA.

At $\sigma_{\theta 25}$, a significant decrease of A_T of ~~-0.2~~-0.20 ± 0.07 µmol $kg^{-1}.a^{-1}$ is observed over the entire OUTPACE area. A decrease of -0.30 ± 0.09~~- 0.2~~ µmol.$kg^{-1}.a^{-1}$ is also observed in MA area, whereas no significant trend is observed for the WGY area. However, when A_T is normalized to salinity, no significant trends are observed in $A_{T\,n35}$ suggesting that the observed trend in A_T can be attributed to salinity changes rather than changes in calcification. ~~No s~~Significant negative trends are observed for $[O_2]$ over the entire area (- 0.31 ± 0.10 µmol kg^{-1} a^{-1}), in MA (- 0.35 ± 0.16 µmol kg^{-1} a^{-1}) and in WGY (- 0.38 ± 0.11 µmol kg^{-1} a^{-1}) ~~are observed for $[O_2]$~~. The decrease in $[O_2]$ wich corresponds to a positive trend in AOU suggested an increase in the remineralization of organic matter at $\sigma_{\theta 25}$. Significant increasing trends were observed for C_T over the entire area (+ 1.32 ± 0.13 µmol kg^{-1} a^{-1}), in MA (+ 1.~~4~~38 ± 0.21 µmol kg^{-1} a^{-1}) and in WGY (+ 1.~~6~~57 ± 0.13 µmol kg^{-1} a^{-1}) ~~are observed for C_T~~. For C_{ANT}, the trends ~~are~~were slightly slower (+ 1.12 ± 0.07 to 1.2± 0.13 ± 0.09 µmol kg^{-1} a^{-1}) and not significantly different between MA and WGY. Taking into account the OUTPACE dataset does not change the overall significance of the observed trends and only minor changes (mostly within the error of the estimates) are observed. If we assume a C_T increase of 0.5 to 1 µmol kg^{-1} a^{-1} (depending on the buffer factors considered) associated to the recent rise in atmospheric CO_2 (see for example Murata et al., 2007), the C_T increase in the OUTPACE area is faster than thermodynamics would govern whereas the C_{ANT} is closer to this thermodynamic value. The higher increase of C_T could be related to an increase in remineralization processes as deduced from $[O_2]$ trends, with an overall consistency between the rate of C_T increase and the rate of decrease in $[O_2]$. However, the important increase of C_{ANT} observed between 2005 and 2015 between 10°S and 30°S on the P16 line (at the eastern side of the OUTPACE transect) by Carter et al. (2017) is not supported by significant differences in the trends of C_{ANT} observed between MA and WGY in this study. ~~This suggests that even if the higher levels of C_{ANT} observed at $\sigma_{\theta 25}$ in WGY (see former section) than in MA would be related to denitrification, they are not associated to a faster accumulation rates of C_{ANT} induced by changes in denitritification. Indeed, Murata et al. (2007) showed that based on a direct relation between C_T and N*, the influence of denitrification should be negligible on C_{ANT} accumulation rates in this area. The small~~

~~estimated change in A_T (less than - 3 µmol kg^{-1} per decade) which could indicate some changes in remineralization processes has a very low impact on TrOCA° used in the estimation of C_{ANT} and cannot explain the difference of C_{ANT} between MA and WGY observed in the upper thermocline waters. trend of C_{ANT} between MA and WGY. in the observed area significant differences N .observed by Carter et al. (2017) at 20°S on the P16 line on C_{ANT} changes estimated between 2005 and 2015has been "over accumulation" Such an~~

At $\sigma_{\theta 27}$, the only significant trend observed is an increase in C_{ANT} of ca. 0.40 ± 0.06 µmol.kg^{-1}.a^{-1} in the MA area. When the OUTPACE dataset is not considered, a similar trend is observed for C_T in the MA area. This trend is compatible with the observed increase of C_{ANT} by Kouketsu et al. (2013) along the P21 line close to the isopycnal layer 27 kg m^{-3}. As this increase is not observed in WGY and if we assume that the $\sigma_{\theta 27}$ is filed with AAIW waters, this suggest that the accumulation of C_{ANT} in AAIW is faster west of 170°W line than to the east, but no clear explanation for this trend can be given.

7 Towards an enhanced "Ocean Acidification" in the WTSP?

Temporal variations of pH_T between 1970 and 2015 are~~is~~ presented on Fig. 5c and 5f with rates of pH_T decrease of -0.0022 ± 0.0004 a^{-1} for MA and -0.0027 ± 0.0004 a^{-1} for WGY at $\sigma_{\theta 25}$ (Table 3) between 1980 and 2015. Based on the C_{ANT} rates estimated in the previous section (1.1 to 1.2 µmol kg^{-1} a^{-1}), and based on a constant value of A_T of 2285 µmol kg^{-1} (mean value of A_{Tn35} on $\sigma_{\theta 25}$) and a constant temperature of 20°C (mean value of temperature on $\sigma_{\theta 25}$), we can estimate a pH_T decrease rate of -0.0023 to -0.0025 a^{-1}. This indicates that rates of oceanic pH_T decrease (ocean acidification) can mostly be explained by the increase of C_{ANT}. These rates of acidification are higher than the values reported by Waters et al. (2011) in the Western South Pacific along the P06 Line (south of OUTPACE area at 32°S) between two visits in 1992 and 2008. They are also higher than the surface rates of pH_T decrease of –0.0016 ± 0.0001 a^{-1} recorded at the HOT time-series station in the tropical North Pacific and of –0.0017 ± 0.0001 a^{-1} and –0.0018 ± 0.0001a^{-1} in the tropical North Atlantic at BATS and ESTOC stations respectively (Bates et al., 2014). However, differences in buffer factors between surface and subsurface can partially explain these differences. Nevertheless, our results in subsurface ($\sigma_{\theta 25}$) based on GLODAPv2 and OUTPACE data (C_T and A_T), are similar ~~can be compared with~~to pH_T trends derived from fCO2 surface observations (e.g. Lauvset et al, 2015). In the southern subtropical and equatorial Pacific regions, using SOCAT version 2, Lauvset et al. (2015) evaluate contrasting fCO2 and pH_T trends, ranging between +1.1 µatm a^{-1} and +3.5 µatm a^{-1} for fCO2 and between -0.001 a^{-1} and -0.0023 a^{-1} for pH_T. If we revisit these estimates, using surface fCO2 observations available in the OUTPACE region (18-22°S/170-200°E) in SOCAT version 6~~5~~ (Bakker et al., 2016; www.socat.info) and assuming a constant alkalinity (2300 µmol/kg, average of surface data), we can calculate pH_T and C_T from fCO2 and temperature data. The resulting long-term trends for the period 1980-2016 for fCO2, C_T and pH_T are respectively +1.27 ± 0.01~~3~~ µatm a^{-1}, +1.03 + ± 0.01 µmol kg^{-1} a^{-1} and -0.0013 ± 0.0001 a^{-1}. Interestingly for the period 2000-2016 the trends are +2.53 ± 0.02~~6~~ µatm a^{-1}, +2.02 ± 0.02 µmol kg^{-1} a^{-1} and -0.0025 ± 0.0003 a^{-1}, suggesting an acceleration of the signals in recent years. These results based on fCO2 observations in surface waters, confirm the trends we detected for C_T and pH_T in subsurface layers ($\sigma_{\theta 25}$).

On Fig. 6, estimates of the so-called "Anthropogenic pH_T change" ($\Delta^{ANT}pH_T$) and "Anthropogenic Ω_{ara} change" ($\Delta^{ANT}\Omega_{ara}$) which corresponds to the difference of pH_T and Ω_{ara} between the time of the OUTPACE cruise (modern time) and the pre-industrial period are presented. The pH_T and Ω_{ara} correspond to the values presented on Fig. 3, whereas the pre-industrial values corresponds to pH_T and Ω_{ara} estimated with C_T minus C_{ANT}. All other parameters (temperature, salinity, alkalinity and nutrients) are assumed to remain constant over time. The main features for the distribution of $\Delta^{ANT}pH_T$ and $\Delta^{ANT}\Omega_{ara}$ logically reflect the distribution of the estimated C_{ANT} in this study because C_{ANT} is the only driving force in these estimations. The estimated pH_T decrease reaches values slightly higher than 0.1 and the estimated Ω_{ara} decrease reaches values of 0.75 since the pre-industrial period for areas with the highest C_{ANT} accumulation. When considering an error on C_{ANT} of 6 µmol kg^{-1}, we can assume that we are able to distinguish changes of 0.0012 for pH_T and 0.06 for Ω_{ara}. Decreases of pH_T and Ω_{ara} are thus detectable below 1000 dbar in the MA waters and above 1000 dbar in WGY waters.

A decrease of pH_T of ~0.1 units since the pre-industrial period is a generally accepted value for oceanic waters affected by C_{ANT} penetration (see e.g. Raven et al., 2005). Several studies have assessed the rate of ocean acidification based on successive visits to different oceanic areas. For the South Pacific Ocean, Carter et al. (2017) reports decreases of oceanic pH_T since the pre-industrial period of -0.09 and -0.11 pH_T units for the latitude band from 10 to 20°S and from 20 to 30°S, respectively, along the P16 line (150°W) situated on the eastern side of the OUTPACE area. These are in good agreement with our estimates in this area.

~~Decreases of Ω_{ara} in the WTSP have been discussed in detail along the P21 line by Murata et al. (2015). Based on mean values in the upper water column (< 400 dbar) in regions defined by specific features in the dataset, a decrease of Ω_{ara} at a rate of 0.017 a^{-1} is observed for the western part of the OUTPACE area whereas a much smaller decrease of 0.005 a^{-1} is estimated for the eastern part of the OUTPACE area. This important difference between the western and the eastern part of the OUTPACE area are not observed in our analysis.~~ Based on an interpolation of the estimated Ω_{ara} during OUTPACE and the pre-industrial $\Omega_{ara,}$, we calculated the depth of the horizon where $\Omega_{ara} = 1$ for the different stations of the OUTPACE transect (Table 2) in 2015 and the pre-industrial period base on the $\Delta^{ANT}\Omega_{ara}$ estimates. We observed an upward migration of aragonite saturation horizon of up to 220 m in the MA area along the OUTPACE transect (Table 2 and Fig 6c). These upward migration of the $\Omega_{ara} = 1$ horizon is higher than the migration of 30 to 100m observed between the 90th and the pre-industrial period in early studies (Feely et al. 2004) in the ~~South Equatorial~~ Pacific based on the WOCE dataset illustrating the continuous acidification of the WTSP.

8 Conclusion

Based on $A_{T,}$ and C_T data and related properties collected during the OUTPACE cruise, we estimated different parameters of the carbonate system along a longitudinal section of nearly 4000 km and up to 2000 dbar in WTSP. Even if the vertical and horizontal resolution is low compared to the WOCE lines and precludes a rigorous comparison with this high quality dataset, we estimated that the measured carbonate chemistry parameters are in good agreement with previous data collected in this

area. Based on estimation of C_{ANT} from the TrOCA method, we find C_{ANT} penetration in the WTSP and impacts on pH_T and saturation state of calcium carbonate since the pre-industrial period that are in good agreement with previous observations in this area. ~~Our estimates of C_{ANT} inventories are confirming a recent increase of C_{ANT} accumulation in the WTSP.~~ As mentioned above, C_{ANT} from TrOCA estimates are not reliable in surface layer. However, based on GLODAPv2 and SOCAT database, our estimation of C_{ANT} in sub surface seems to be in good agreement with expected changes in surface waters. ~~We therefore extended to the surface, our estimates of C_{ANT} at 100 dbar in order compute C_{ANT} Inventories in the water column. Even if other parameters needs to be considered, Fig. 6e illustrates the link between accumulation of C_{ANT} in the water column and ocean acidification along the OUTPACE transect.~~ The enhanced impact of ocean acidification in the Subtropical South Pacific suggested by our study highlight the necessity of sustained research efforts in this largely under-explored part of the World Ocean. The presented dataset collected along the OUTPACE transect could complement existing section visited nearly every decade in the South Pacific ocean and in particular the P21 line which was last visited in 2009.

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Table 1 : General description of the cast sampled for carbonate chemistry parameters during the OUTPACE cruise.

Cast	Station	Longitude (°E)	Latitude (°N)	Time (UTC)	Max Pres.	Type*	Rosette**
out_c_006	SD 1	159,,9255	-17,,9418	2015/02/22 03:08:00	202	SHAW	CLA
out_t_002		159,,9425	-17,,9088	2015/02/22 07:43:00	2000	INT	TMC
out_c_010	SD 2	162,,1248	-18,,6078	2015/02/23 00:11:00	199	SHAW	CLA
out_c_016		162,,1112	-18,,5845	2015/02/23 08:16:00	1998	INT	CLA
out_c_019	SD 3	165,,0093	-19,,4955	2015/02/24 05:58:00	200	SHAW	CLA
out_c_020		165,,0082	-19,,4907	2015/02/24 08:14:00	1999	INT	CLA
out_c_066	LD A	164,,5877	-19,,2242	2015/03/02 14:39:00	200	SHAW	CLA
out_c_067		164,,5787	-19,,2233	2015/03/02 16:10:00	2002	INT	CLA
out_c_070	SD 4	168,,0118	-19,,9832	2015/03/04 10:55:00	201	SHAW	CLA
out_c_071		168,,0157	-19,,98	2015/03/04 12:43:00	1999	INT	CLA
out_c_074	SD5	169,,9943	-22,,0002	2015/03/05 08:48:00	201	SHAW	CLA
out_c_075		169,,9965	-21,,9997	2015/03/05 10:27:00	1999	INT	CLA
out_c_078	SD6	172,,1198	-21,,3732	2015/03/06 07:27:00	200	SHAW	CLA
out_c_079		172,,1193	-21,,3758	2015/03/06 09:08:00	1999	INT	CLA
out_c_082	SD7	174,,25	-20,,7697	2015/03/07 05:09:00	201	SHAW	CLA
out_c_083		174,,2512	-20,,7677	2015/03/07 06:37:00	2000	INT	CLA
out_c_086	SD8	176,,3778	-20,,7027	2015/03/08 02:31:00	201	SHAW	CLA
out_c_087		176,,364	-20,,6945	2015/03/08 04:19:00	1997	INT	CLA
out_c_091	SD9	178,,6087	-20,,9963	2015/03/09 04:57:00	2002	INT	CLA
out_t_012		178,,6062	-20,,9892	2015/03/09 06:46:00	201	SHAW	TMC
out_c_094	SD10	-178,,5105	-20,,4417	2015/03/10 04:10:00	200	SHAW	CLA
out_c_095		-178,,5105	-20,,44	2015/03/10 05:48:00	762	INT	CLA
out_c_098	SD11	-175,,6542	-20,,0028	2015/03/11 00:53:00	207	SHAW	CLA
out_c_099		-175,,6475	-20,,0057	2015/03/11 02:46:00	2000	INT	CLA
out_c_102	SD12	-172,,7885	-19,,5237	2015/03/12 00:38:00	200	SHAW	CLA
out_c_103		-172,,7813	-19,,5368	2015/03/12 02:26:00	2001	INT	CLA
out_c_150	B	-170,,7433	-18,,179	2015/03/20 12:38:00	204	SHAW	CLA
out_c_151		-170,,7385	-18,,1745	2015/03/20 14:16:00	1997	INT	CLA
out_c_152	SD13	-169,,0728	-18,,2007	2015/03/21 10:27:00	501	INT	CLA
out_c_163	C	-165,,9315	-18,,4282	2015/03/24 12:23:00	5027	DEEP	CLA
out_c_194		-165,,8647	-18,,4952	2015/03/28 02:01:00	25	REPRO	CLA
out_c_198		-165,,7915	-18,,4912	2015/03/28 12:42:00	298	SHAW	CLA
out_c_199		-165,,7792	-18,,4842	2015/03/28 14:32:00	2001	INT	CLA
out_c_209	SD14	-163,,001	-18,,395	2015/03/30 05:19:00	300	SHAW	CLA
out_c_210		-162,,9992	-18,,3952	2015/03/30 07:03:00	2000	INT	CLA
out_c_212	SD15	-159,,9913	-18,,265	2015/03/31 04:01:00	300	SHAW	CLA
out_c_213		-159,,9913	-18,,2618	2015/03/31 05:41:00	2002	INT	CLA

*: SHAW stands for casts up to 200 dbar, INT stands for casts up to 2000m, DEEP stands for the deep cast and REPRO stands for the cast with reproducibility measurements

**: CTD rosette used for the cast. CLA is the normal CTD rosette and TMC is the trace metal clean rosette (See Sect. 2.1)

Table 2 : Estimated trends on A_T, $[O_2]$, C_T , C_{ANT} and pH_T changes in two different layers of the water column defined by isopycnal layers between 1980 and 2015 based on GLODAPv2 with (column WITH) and without (column WITHOUT) OUTPACE data added. Estimated trends are obtained from slope values of a linear regression between the studied parameters and time.

	$25 < \sigma_\theta < 25.5$		$27 < \sigma_\theta < 27.2$	
	WITH	WITHOUT	WITH	WITHOUT
	Trend on A_T in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	-0.20 ± 0.07 (n = 167) *	-0.30 ± 0.07 (n = 142) *	-0.12 ± 0.07 (n = 180)	-0.01 ± 0.06 (n = 174)
MA	-0.30 ± 0.09 (n = 85) *	-0.47 ± 0.10 (n = 70) *	-0.16 ± 0.09 (n = 99)	-0.10 ± 0.09 (n = 92)
WGY	-0.20 ± 0.14 (n = 28)	-0.20 ± 0.19 (n = 22)	-0.20 ± 0.14 (n = 35)	-0.01 ± 0.13 (n = 31)
	Trend on $[O_2]$ in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	-0.31 ± 0.10 (n = 167)*	-0.61 ± 0.09 (n = 143)*	0.05 ± 0.11 (n = 183)	0.07 ± 0.10 (n = 178)
MA	-0.35 ± 0.16 (n = 84)*	-0.78 ± 0.17 (n = 70)*	0.06 ± 0.11 (n = 99)	0.04 ± 0.11 (n = 93)
WGY	-0.38 ± 0.11 (n = 27)*	-0.35 ± 0.14 (n = 23)*	-0.11 ± 0.30 (n = 38)	-0.22 ± 0.29 (n = 34)
	Trend on C_T in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	1.32 ± 0.13 (n = 174) *	1.63 ± 0.13 (n = 149) *	0.23 ± 0.13 (n = 189)	0.27 ± 0.11 (n = 183) *
MA	1.38 ± 0.21 (n = 85) *	1.87 ± 0.21 (n = 70) *	0.31 ± 0.16 (n = 100)	0.44 ± 0.17 (n = 93) *
WGY	1.57 ± 0.18 (n = 31) *	1.57 ± 0.23 (n = 25) *	0.23 ± 0.29 (n = 40)	0.23 ± 0.29 (n = 36)
	Trend on C_{ANT} in $\mu mol.kg^{-1}.a^{-1}$			
OUTPACE	1.12 ± 0.07 (n = 166) *	1.25 ± 0.06 (n = 142) *	0.32 ± 0.05 (n = 179) *	0.25 ± 0.04 (n = 174) *
MA	1.18 ± 0.08 (n = 84) *	1.31 ± 0.08 (n = 70) *	0.40 ± 0.06 (n = 98) *	0.40 ± 0.06 (n = 92) *
WGY	1.20 ± 0.09 (n = 28) *	1.18 ± 0.10 (n = 22) *	0.13 ± 0.09 (n = 35)	0.11 ± 0.08 (n = 31)
	Trend on pH_{TINSI} in a^{-1}			
OUTPACE	-0.0022 ± 0.0003 (n=167)*	-0.0031 ± 0.0002 (n=142*	-0.0001 ± 0.0003 (n=181)	-0.0002 ± 0.0002 (n=175)
MA	-0.0022 ± 0.0004 (n=85) *	-0.0033 ± 0.0004 (n=70) *	-0.0004 ± 0.0003 (n=100)	-0.0007 ± 0.0003 (n=93)*
WGY	-0.0027 ± 0.0004 (n=28) *	-0.0030 ± 0.0004 (n=22) *	-0.00008 ± 0.0006 (n=35)	-0.0007 ± 0.0006 (n=31)

* : trend significant (p-level < 0.05)

595 **Table 23 :** ~~Estimated inventories of C_{ANT} up to 1000 and 2000 meters (see text for details) and e~~Estimated depth of the $\Omega_{ara} = 1$ horizon along the OUTPACE cruise (see text for details). No values are available for stations where data up to 2000 dbar were not available (SD2 and SD13). For the depth of the $\Omega_{ara} = 1$ horizon, no values were estimated for stations with $C_{ANT} < -6$ µmol kg^{-1}.

Station	Longitude	Latitude	~~Inventory for C_{ANT} (mol.m^{-2})~~			Depth of the $\Omega_{ara} = 1$ horizon (in m)		
			~~$C_{ANT}INV_{2000}$~~	~~$CANTINV_{1000}$~~	~~% in 1000m*~~	OUTPACE	Pre-indu.	Difference**
SD1	159,9425	-17,9088	~~43~~	~~38,6~~	~~89,7~~	1225	NA	NA
SD2	162,1248	-18,6078	~~NA~~	~~NA~~	~~NA~~	NA	NA	NA
SD3	165,0082	-19,4907	~~44,8~~	~~39,6~~	~~88,3~~	928	NA	NA
A	164,5787	-19,2233	~~39,7~~	~~35,2~~	~~88,6~~	1032	1185	153
SD4	168,0157	-19,98	~~43,7~~	~~36,2~~	~~82,8~~	1029	1193	164
SD5	169,9965	-21,9997	~~49,4~~	~~41,2~~	~~83,4~~	1126	1256	130
SD6	172,1193	-21,3758	~~47~~	~~41,2~~	~~87,6~~	1097	1233	136
SD7	174,2512	-20,7677	~~44,6~~	~~36,7~~	~~82,2~~	1015	1235	220
SD8	176,364	-20,6945	~~42,7~~	~~38,3~~	~~89,6~~	1010	1171	161
SD9	178,6087	-20,9963	~~36,6~~	~~34,8~~	~~95,1~~	1214	NA	NA
SD11	-175,6475	-20,0057	~~37,2~~	~~34,1~~	~~91,6~~	1055	1172	117
SD12	-172,7813	-19,5368	~~39,7~~	~~37,4~~	~~94,2~~	1013	1112	99
B	-170,7385	-18,1745	~~36~~	~~34,5~~	~~95,8~~	948	1046	98
SD13	-169,0728	-18,2007	~~NA~~	~~NA~~	~~NA~~	NA	NA	NA
C	-165,7792	-18,4842	~~37,7~~	~~36,1~~	~~95,7~~	854	941	87
SD14	-162,9992	-18,3952	~~37,3~~	~~34,4~~	~~92,2~~	889	1006	117
SD15	-159,9913	-18,2618	~~36,9~~	~~35,3~~	~~95,6~~	917	1043	126

~~* Percentage of $CANTINV_{2000}$ estimated in $C_{ANT}INV_{1000}$~~

** Difference (in m) between the depth of the $\Omega_{ara} = 1$ horizon at the pre-industrial period and the OUTPACE cruise.

: Estimated trends on A_t, $[O_2]$, C_t , C_{ANT} and pH_t changes in two different layers of the water column defined by isopycnal layers between 1980 and 2015 based on GLODAPv2 and OUTPACE. Estimated trends are obtained from slope values of a linear regression between the studied parameters and time.3Table

	$25 < \sigma_\theta < 25.5$	$27 < \sigma_\theta < 27.2$
	Trend on AT	
OUTPACE area	µmol.kg-1.a-1,-0.20 ± 0,.07 µmol.kg-1.a-1 (n = 167) *	,-0.12 ± 0,.07 µmol.kg^{-1}.a^{-1} (n = 180)
MA area	µmol.kg-1.a-1-0.30 ± 0.09 µmol.kg-1.a-1 (n = 85) *	-0.16 ± 0.09 µmol.kg^{-1}.a^{-1} (n = 99)
WGY area	,-0.,20 ± 0.14 µmol.kg^{-1}.a^{-1} (n = 28)	,-0,.20 ± 0.14 µmol.kg^{-1}.a^{-1} (n = 35)
	Trend on $[O_2]$ in µmol.kg^{-1}.a^{-1}	
OUTPACE area	-0.31 ± 0.10 µmol.kg^{-1}.a^{-1} (n = 167)	0.05 ± 0.11 µmol.kg^{-1}.a^{-1} (n = 183)
MA area	-0.35 ± 0.16 µmol.kg^{-1}.a^{-1} (n = 84)	0.06 ± 0.11 µmol.kg^{-1}.a^{-1} (n = 99)
WGY area	-0.38 ± 0.11 µmol.kg^{-1}.a^{-1} (n = 27)	,-0,.11 ± 0.30 µmol.kg^{-1}.a^{-1} (n = 38)
	Trend on C_T in µmol.kg^{-1}.a^{-1}	
OUTPACE area	1.32 ± 0.13 µmol.kg^{-1}.a^{-1} (n = 174) *	0.23 ± 0.13 µmol.kg^{-1}.a^{-1} (n = 189)
MA area	1.38 ± 0.21 µmol.kg^{-1}.a^{-1} (n = 85) *	0.31 ± 0.16 µmol.kg^{-1}.a^{-1} (n = 100)
WGY area	1.57 ± 0.18 µmol.kg^{-1}.a^{-1} (n = 31) *	0.23 ± 0.29 µmol.kg^{-1}.a^{-1} (n = 40)
	Trend on C_{ANT} in µmol.kg^{-1}.a^{-1}	
OUTPACE area	1.12 ± 0.07 µmol.kg^{-1}.a^{-1} (n = 166) *	0.32 ± 0.05 µmol.kg^{-1}.a^{-1} (n = 179) *
MA area	1.18 ± 0.08 µmol.kg^{-1}.a^{-1} (n = 84) *	0.40 ± 0.06 µmol.kg^{-1}.a^{-1} (n = 98) *
WGY area	1.20 ± 0.09 µmol.kg^{-1}.a^{-1} (n = 28) *	0.13 ± 0.09 µmol.kg^{-1}.a^{-1} (n = 35)
	Trend on pH_{TINSI} in a^{-1}	
OUTPACE area	-0.0022 ± 0.0003 a^{-1} (n=167) *	-0.0001 ± 0.0003 a^{-1} (n=181)
MA area	-0.0022 ± 0.0004 a^{-1} (n = 85) *	-0.0004 ± 0.0003 a^{-1} (n=100)
WGY area	-0.0027 ± 0.0004 a^{-1} (n = 28) *	-0.00008 ± 0.0006 a^{-1} (n=35)

* : trend significant (p-level < 0.05)

Figures

Fig. 1: Map of the OUTPACE cruise transect. The outpace stations are distinguished between Melanesian Archipelago (MA) stations with darkgreen large dots and the Western GYre (WGY) stations with dark blue large dots. Stations outside of these two areas are in grey. The station with a red indication corresponds to the station where the deep cast and intercomparaison cast was made. Station from the GLODAPv2 database are indicated with small crossesdots: small green dots correspond to GLODAPv2 stations considered for comparaison in the MA area, small blue dots correspond to GLODAPv2 stations considered for comparaison in the WGY area and small grey dots are the other GLODAPv2 stations considered for comparison.

Fig. 2: $\Theta - S_A$ diagram with colors indicating the AOU. Black contour lines represent the isopycnal horizons based on potential density referenced to a pressure of 0 dBar (σ_θ).

Fig. 3: Longitudinal variations of (a) A_T, (b) C_T, (c) pH_T and (d) Ω_{ara} along the OUTPACE transect between surface and 2000m depth. Black contour lines represent the isopycnal horizons based on potential density referenced to a pressure of 0 dBar. Vertical profiles of (e) A_T, (f) A_T normalized to SA =35 and (g) C_T of the entire OUTPACE dataset (red dots) superimposed on the GLODAPv2 data corresponding to the OUTPACE area (grey dots).

Fig. 4: Longitudinal variations C_{ANT} (Estimated with the TROCA method) along the OUTPACE transect between surface and 2000m depth (a). Black contour lines represents the isopycnal horizons based on potential density referenced to a pressure of 0 dBar. Vertical profiles of C_{ANT} for the entire OUTPACE dataset superimposed on the values estimated from the GLODAPv2 data (b) and vertical profiles of of C_{ANT} between surface and 1500m superimposed on the values estimated from the recent (after 2005) GLODAPv2 data (c). Color code for the dots is the same as for Figure 1

Fig. 5: Temporal evolution in the OUTPACE area of C_T (a and c), C_{ANT} (b and d) and pH_{Tinsi} (c and e) based on GLODAPv2 and OUTPACE data along two isopycnal layers: 25 – 25.5 $kg.m^{-3}$ (left side panels) and 27 - 27.2 $kg.m^{-3}$ (right side panels). Color code for the dots is the same as for Figure 1.

Fig. 6: Longitudinal variations of (a) pH_T changes and (b) Ω_{ara} changes between pre-industrial and present time along the OUTPACE transect between surface and 2000m depth (See text for details). Black contour lines represent the isopycnal horizons based on potential density referenced to a pressure of 0 dBar. ~~Panel (c) represents longitudinal variations of inventories of C_{ANT} ($C_{ANT}INV_{2000}$) plotted in grey (see table 2 for details) and the depth of the Ω_{ara} = 1 horizon (in m) during OUTPACE (red dots) and at the pre-industrial period (Black dots). The red area illsutrates the shoaling of the Ω_{ara} = 1 horizon since the pre-industrial period.~~

Fig. 1

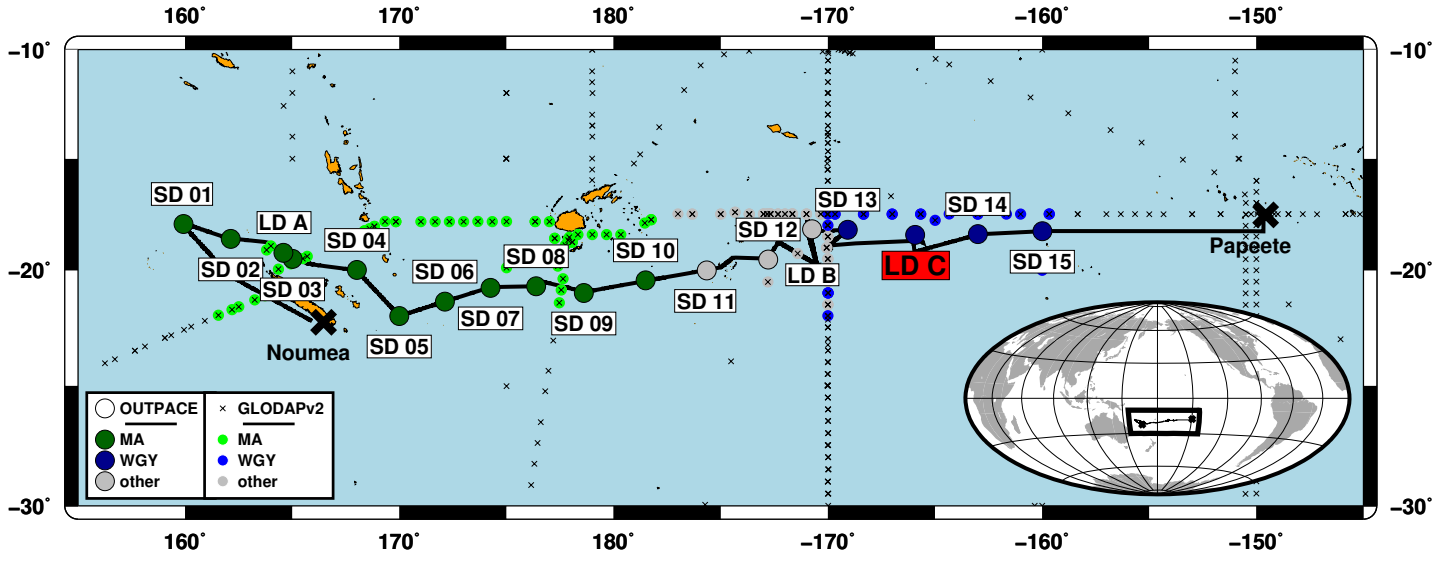
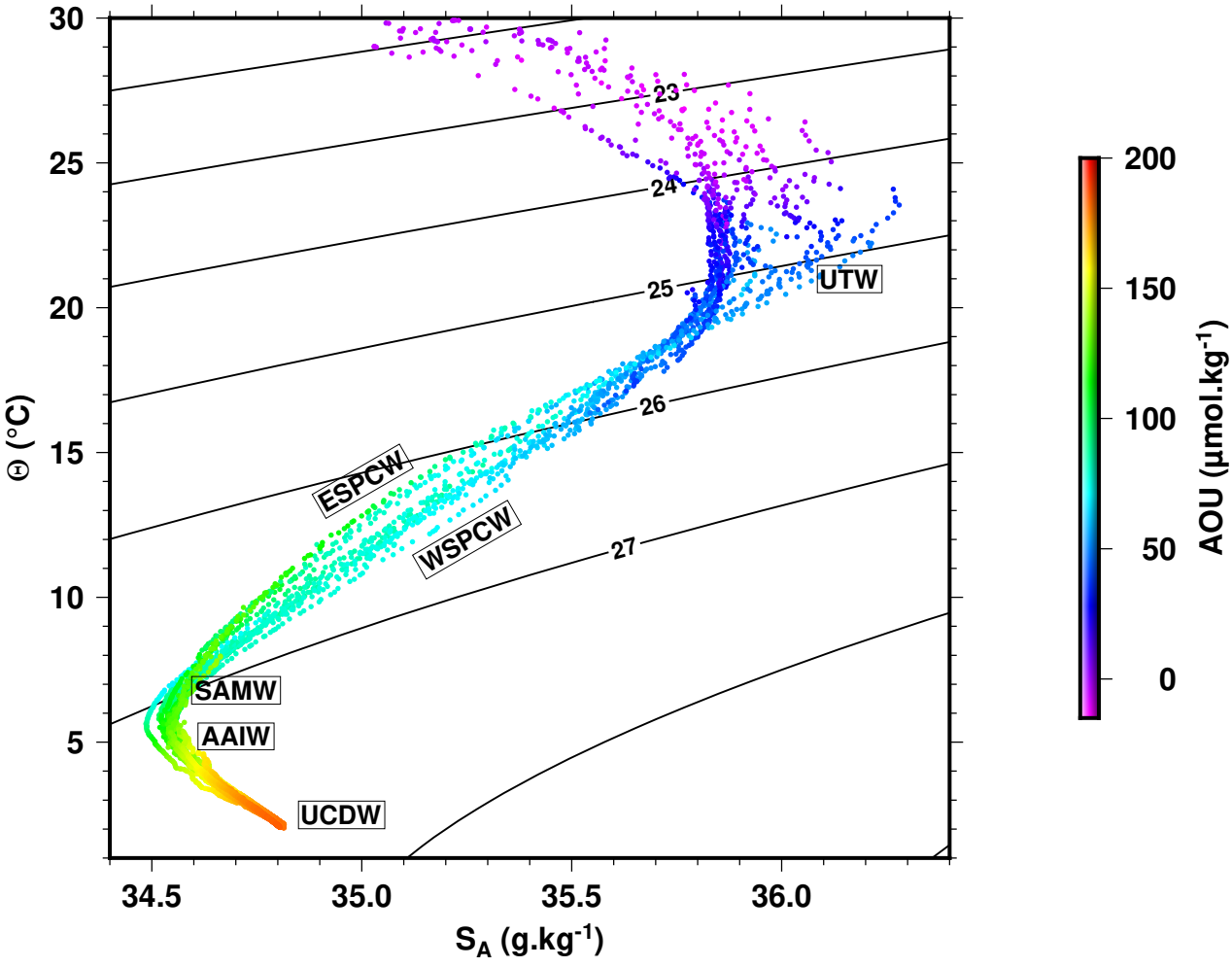

160˚
170˚
180˚
−170˚
−160˚
−150˚
−10˚
−20˚
−30˚
SD 01
LD A
SD 02
SD 03
SD 04
SD 05
SD 06
SD 07
SD 08
SD 09
SD 10
SD 11
SD 12
SD 13
SD 14
SD 15
LD B
LD C
Noumea
Papeete
OUTPACE
MA
WGY
other
GLODAPv2
MA
WGY
other

Fig. 2


30
25
20
15
10
5
Θ (°C)
34.5
35.0
35.5
36.0
S_A (g.kg^-1)
23
24
25
26
27
UTW
ESPCW
WSPCW
SAMW
AAIW
UCDW
200
150
100
50
0
AOU (µmol.kg^-1)

635 Fig. 3

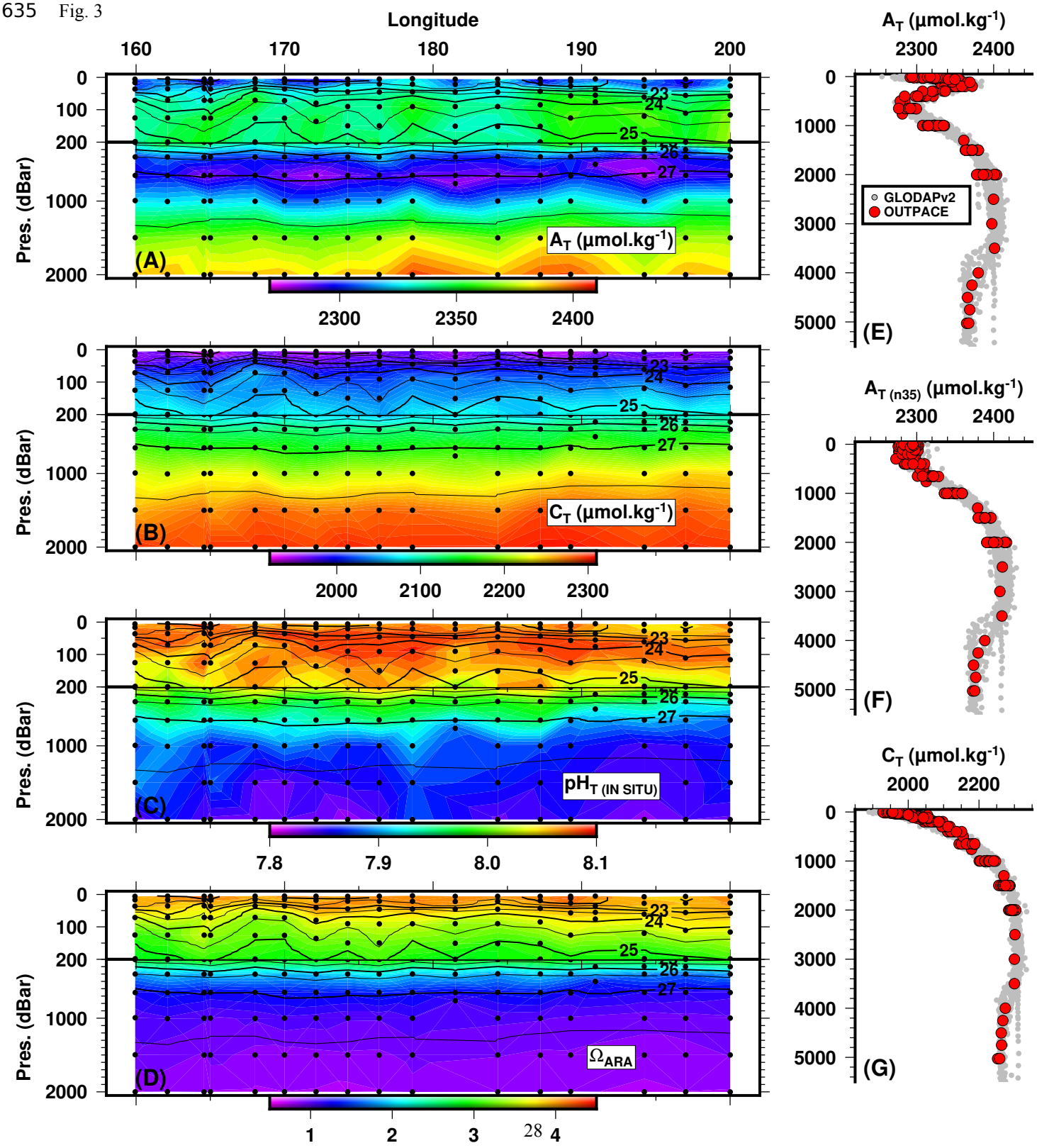

Longitude
160
170
180
190
200
Pres. (dBar)
0
100
200
1000
2000
23
24
25
26
27
(A)
A_T (µmol.kg^{-1})
2300
2350
2400
(B)
C_T (µmol.kg^{-1})
2000
2100
2200
2300
(C)
$pH_{T\ (IN\ SITU)}$
7.8
7.9
8.0
8.1
(D)
Ω_{ARA}
1
2
3
4
A_T (µmol.kg^{-1})
2300
2400
3000
4000
5000
GLODAPv2
OUTPACE
(E)
$A_{T\ (n35)}$ (µmol.kg^{-1})
(F)
C_T (µmol.kg^{-1})
(G)

Fig. 4

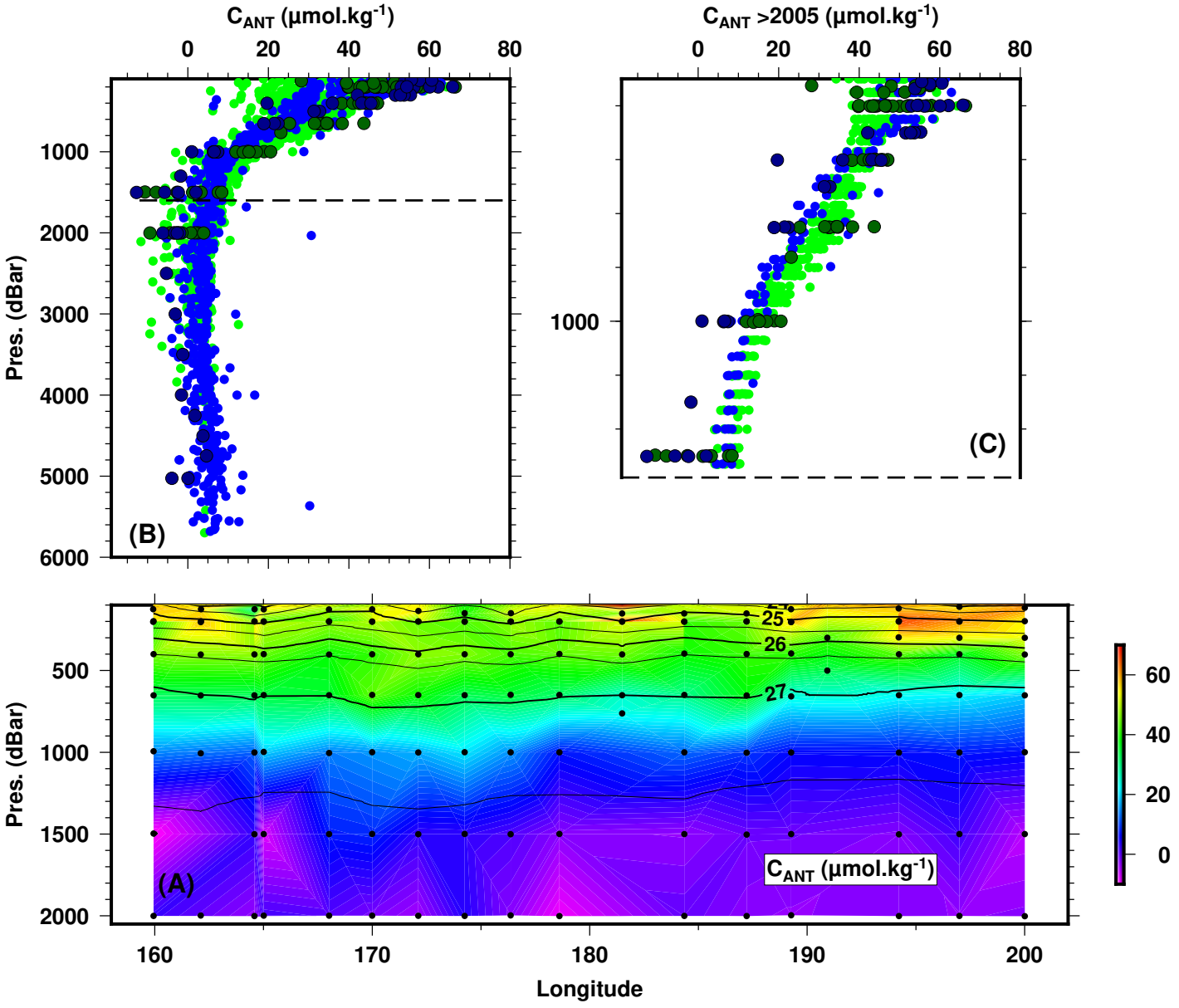

C_ANT (µmol.kg-1)
0
20
40
60
80
1000
2000
3000
4000
5000
6000
Pres. (dBar)
(B)
C_ANT >2005 (µmol.kg-1)
0
20
40
60
80
1000
(C)
500
1000
1500
2000
Pres. (dBar)
25
26
27
(A)
C_ANT (µmol.kg-1)
60
40
20
0
160
170
180
190
200
Longitude

Fig. 5

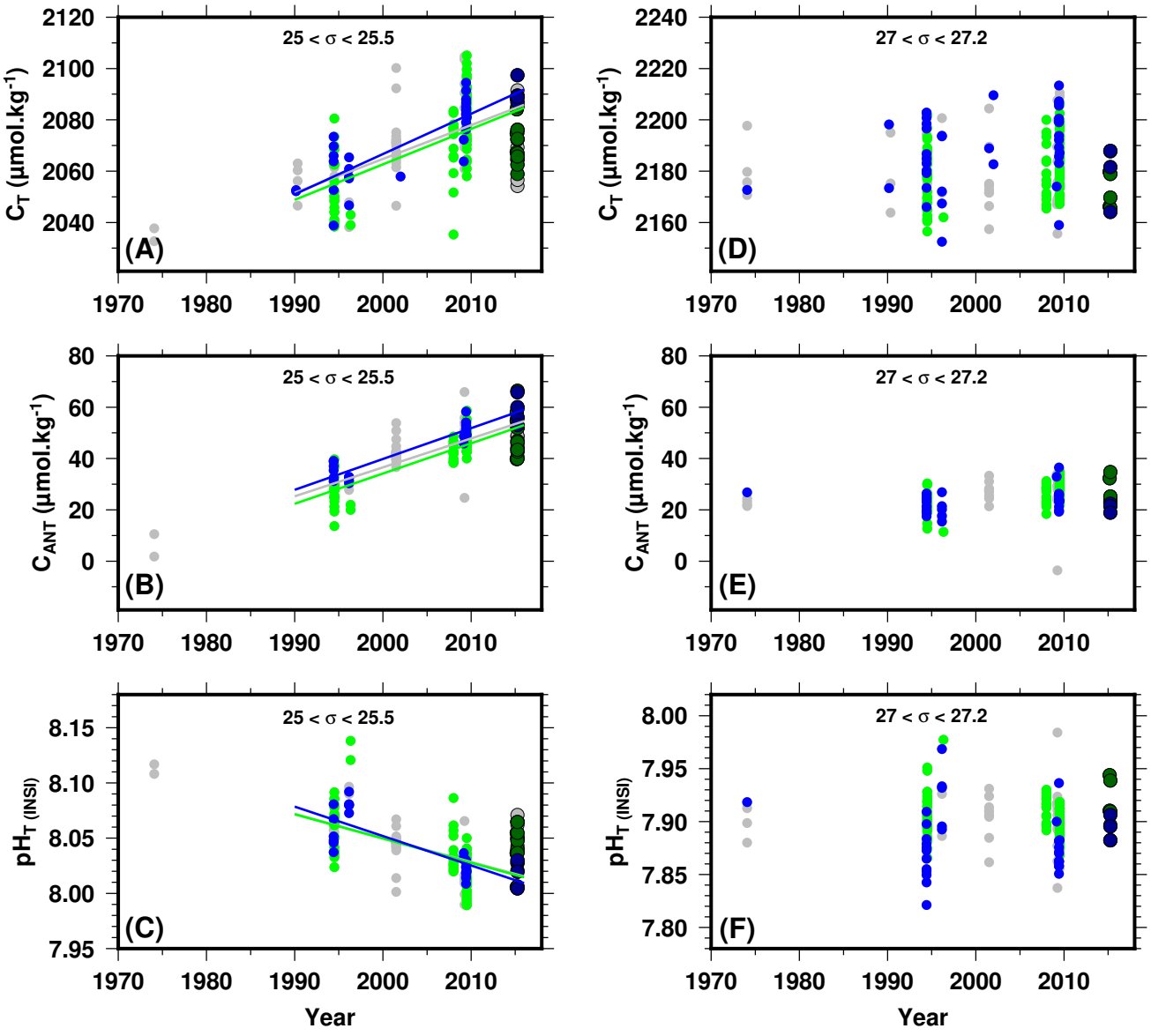
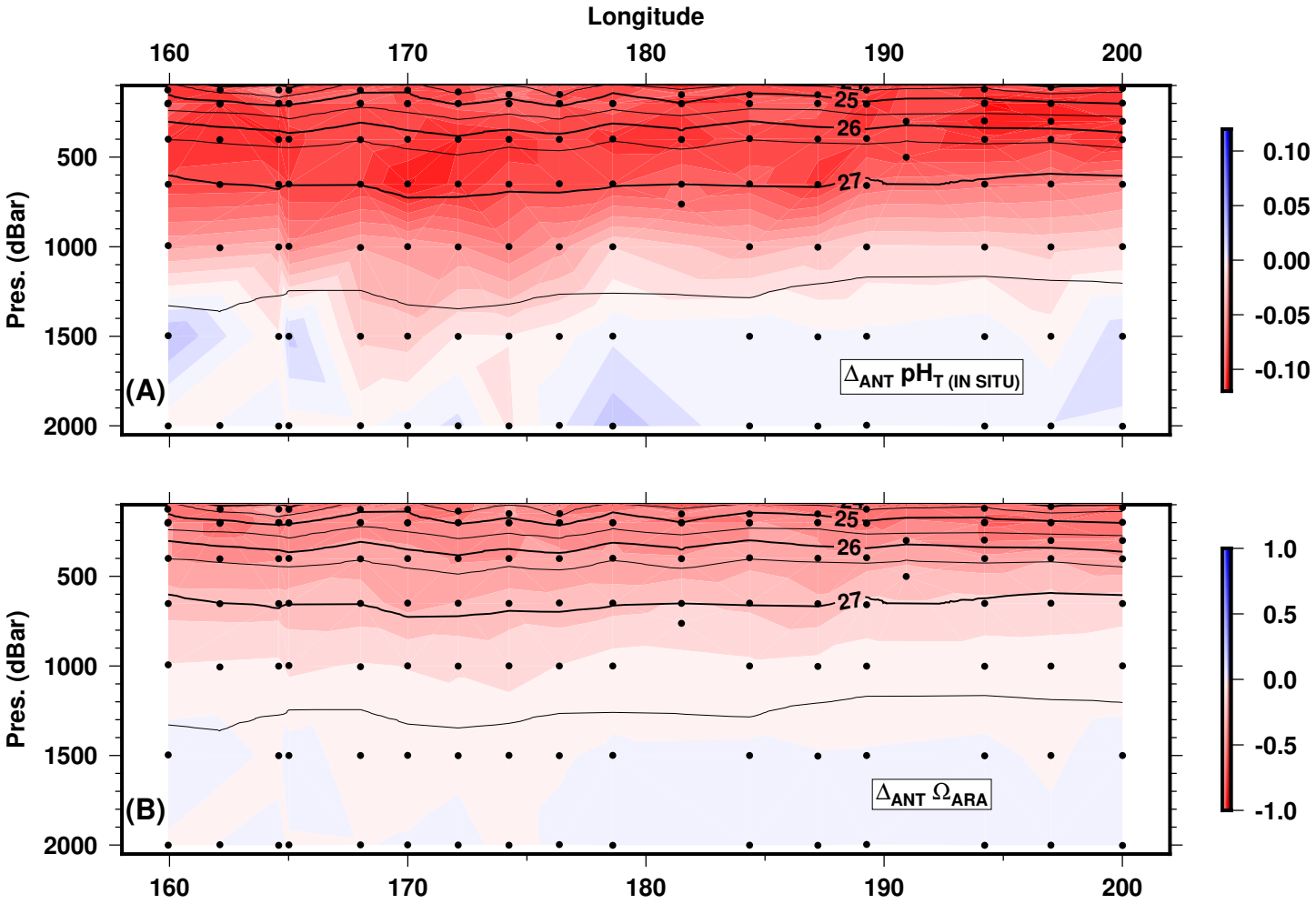

25 < σ < 25.5
27 < σ < 27.2
(A)
(B)
(C)
(D)
(E)
(F)
C_T (µmol.kg^{-1})
C_{ANT} (µmol.kg^{-1})
$pH_{T\ (INSI)}$
Year
1970
1980
1990
2000
2010

Fig. 6


Longitude
160
170
180
190
200
Pres. (dBar)
500
1000
1500
2000
25
26
27
(A)
Δ_{ANT} $pH_{T\ (IN\ SITU)}$
0.10
0.05
0.00
-0.05
-0.10
(B)
Δ_{ANT} Ω_{ARA}
1.0
0.5
0.0
-0.5
-1.0