Plouzane, Sept. 16th 2017.

Dear Editor,

You will find below the marked-up version of our manuscript regarding major comments expressed by the two referees. To facilitate reading, we choice to remove text in this marked-up version and keep only manuscript outline including changes. We use **black color for unchanged sessions, green color and crossed out text for removed sessions or paragraphs** and **red color for new sessions in response to major comments of referees**. Lines indicated here correspond to the old version of our manuscript. Finally, additional analyses led to add a new collaboration for this manuscript. As I indicated in previous e-mail (Sept. 8th), a new co-authors is added.

Transport and storage of anthropogenic C in the Subpolar North Atlantic : Model – Data comparison

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Abstract

The North Atlantic Ocean is a major sink region for anthropogenic carbon (Cant) and a major contributor to its storage. While it is in general agreed that the intensity of the meridional overturning circulation (MOC) modulates uptake, transport and storage of Cant in the North Atlantic Subpolar Ocean, processes controlling their recent variability and 21st century evolution remain uncertain. This study aims to investigate the relationship between the transport of Cant, the air-sea anthropogenic CO2 fluxes and the storage of Cant in the North Atlantic Subpolar Ocean over the past 44 years. Its relies on the combined analysis of an annual to multi-annual in situ data set and output from a global biogeochemical ocean general circulation model (NEMO/PISCES) at 1/2° spatial resolution forced by the atmospheric reanalysis Drakkar Forcing Set 4. Despite an underestimation of Cant transport and an overestimation of anthropogenic air-sea CO₂ fluxes in the model, Cant storage rate, its variability and driving processes are well simulated. At the interannual time scale, this study confirms that the time rate of changes in Cant storage in NEMO/PISCES is controlled by the divergence of the northward transport of Cant between 25°N and the Greenland-Iceland-Scotland sills. Our results highlight the key role played by the divergence of the NACW transport to the storage of Cant in the upper oceanic layer of the subtropical region and to supply IW then NADW. In addition, this study shows that Cant uptake by NADW in the lower limb of the MOC mainly occurs in the OVIDE-sills box and only one quarter is exported to the subtropical region. Finally, at the multi-decadal scale, the long-term changes in the north Atlantic Cant storage rate is rather driven by the increasing air-sea fluxes of anthropogenic CO₂.

1. Introduction

L. 41 to 95

The objective of the present study is to evaluate the relationship between Cant transport, air-sea fluxes and storage rate in the Subpolar North Atlantic, along with their combined evolution over the past 44

years (1958-2012). It relies on the combination of an annual to multi-annual data set gathered from 25°N to the Greenland-Iceland-Scotland sills over the period 2003-2011 and output from the global biogeochemical ocean general circulation model NEMO/PISCES at 1/2° spatial resolution forced by an atmospheric reanalysis (Bourgeois et al., 2016). The paper is organized as follow: NEMO/PISCES and *in situ* data sets are detailed in Sect. 2 and compared in Sect. 3 to evaluate the model performance; main results of the interannual to decadal change of the North Atlantic Cant fluxes and storage rate as well as the evaluation of their main drivers are presented in Sect. 4 and discussed in Sect. 5 regarding model-data comparison.

2. Material and methods

2.1. <u>NEMO-PISCES model</u>

L.106 to 134

Because climate change trends and natural modes of variability are part of the forcing set used to force both simulations, potential alterations of the natural carbon cycle in response to climate change (e.g. rising sea surface temperature) are thus also captured by the control-natural simulation. The concentration of anthropogenic C, as well as anthropogenic CO_2 fluxes is calculated as the difference between the historical (total C = natural + anthropogenic contribution) and the natural simulations following Orr et al. (2017).

L. 134 to 150

2.2. OVIDE Observation data sets

Observations used to evaluate Cant transport computed from ORCA05-PISCES in the North Atlantic Ocean were collected along the Greenland-Portugal OVIDE section and at 24.5°N following the tracks presented on Fig. 2. Model output of air-sea CO₂ fluxes are compared to the observation-based gridded sea surface pCO₂ product of Landschützer et al. (2015a). Programs and/or data sets are briefly summarized below.

OVIDE data set

L. 154 to 164

L. 169 to 179

24.5°N data set

Description of new data set used for model-data comparison (Sect. 3)

L. 164 to 169

<u>pCO₂ data base</u>

Description of new data set used for model-data comparison (Sect. 3)

2.3. Diagnostic of Cant transport and budget

Transport of Cant across a section

Rewrite to clarify online/offline estimation in response to minor comment of referee#1 <u>Budget of Cant in the North Atlantic Ocean</u>

L. 201 to 208

L. 208 to 210

3. Model evaluation over the OVIDE period 2003-2011

3.1 Distribution of hydrological and biogeochemical parameters along the Greenland-

Portugal OVIDE section

L. 215 to 262 (all Sect. 3.1)

L. 342 to 357 from Sect. 3.4 of old version of manuscript

3.1 Advective transport of Cant

L. 358 to L.360 from Sect. 3.4 of old version of manuscript

 3.2
 Mass transport across the Greenland-Portugal OVIDE section and 25°N

 L.265 to L. 278

L.278 to L. 316 from Sect. 3.4 of old version of manuscript

Completed by the model-data comparison of mass transport at 25°N

- 3.3 Cant distribution along the Greenland-Portugal OVIDE section and 25°N
 L.319 to 339 from Sect. 3.4 of old version of manuscript
 Completed by the model-data of Cant distribution along 25°N
- L. 362 to 370 Sect. 3.4 of old version of manuscript

3.2. Air-sea fluxes of anthropogenic CO2

L. 373 to 376

Model-data comparison of total and anthropogenic air-sea CO₂ fluxes.

3.4-3.3. Storage rate of Cant Budget of Cant in the North Atlantic Ocean (north of 25° N)

L. 379 to 395 from on session 3.4

L. 397 to 401 rewrite as follow:

Despite an underestimation of Cant transport and an overestimation of anthropogenic air-sea CO₂ fluxes, modeled storage rate, its variability and driving processes are coherent with observations allowing the simulation to be used to study drivers of changes in Cant storage rate since 1958.

4. Cant fluxes and storage rate in the Subpolar North Atlantic Ocean (North of 25°N) since

1958 Long-term change in Cant fluxes and storage rate in the Subpolar North Atlantic region

In this section, we present the analysis of the full period covered by our simulations (1958-2012) with the objective of better understanding 1) the relative contribution of the variability of circulation and

the increase in Cant concentration to the variability of Cant transport through the North Atlantic Ocean, and 2) the long-term change of the Cant inventory in this region as well as driving processes the interannual to decadal variability of the Cant inventory in the North Atlantic Ocean as well as its driving processes. The study area, from 25°N to the Greenland-Iceland-Scotland sills, is divided in 3 boxes instead of 2 in section 3: the first box extends from 25°N to 36°N; the second box from 36°N to the OVIDE section and the third box is between the OVIDE section and the Greenland-Iceland-Scotland sills. The section 36°N was added to delimit the northern part of the subtropical region from the Subpolar gyre (Mikaloff-Fletcher et al., 2003).

4.1 <u>Contribution of variability of circulation and Cantaccumulation on Canttransport</u> <u>variability</u>

L. 415 to 438

Completed by a third box, 25°N-36°N.

4.2. <u>Long-term change in Cant storage rate and driving processes</u> L. 439 to 489

4.2. Interannual to decadal variability of the North Atlantic Cant inventory

4.3 Contribution of advection of water masses to the storage rate of Cant

5. Discussion and conclusion

Discussion of old and news results regarding model performance. L. 492 to 501

Table capture

Table 1 (updated with 24.5N-data information) Table 2 (simplified) Table 3 Table 3 Table 4 (old table 3 but simplified) Table 5 and Table 6

Figure Capture

old Figs. 3, 4, 9 and 10 Fig 1 Fig 2 : modified Fig 3: old Fig. 7 Fig. 4: old Fig. 5 Fig. 5 = Old Fig. 6 Fig. 6 to 9 Fig. 10 : old Fig. 8 Fig. 11 to 16 Plouzane, June 4th 2017

Dear Editor,

You will find below our response regarding major and minor comments expressed by the two referees. We read with attention and we hope we have positively replied to all comments to continue with the submission of revised manuscript.

Anonymous Referee #1• (i) Evaluation:

On the one hand, the authors spend a lot of text and effort on the evaluation of the model results. But on the other hand, this evaluation is biased in that they nearly exclusively rely on the data from the Ovide section. In order to assess the relationship between air-sea fluxes, transport and storage of anthropogenic CO2 in the model, it is necessary, in my opinion, to assess all of these elements and not just the data along the southern boundary. A particularly glaring gap is the lack of assessment of the air-sea CO2 fluxes. Of course, the data provide constraints on the total air-sea CO2 flux and not only on that of anthropogenic CO2, but a demonstration that the model is capturing the observed variability in the total flux would substantially strengthen the analysis. Further elements to assess include also the transport across the northern boundary, which is as large as that through the well assessed southern boundary.

Anonymous Referee #2 (1) Model-data comparison: This part of the paper is way too long and descriptive. Furthermore, it seems problematic to compare the model mainly along one section. What if the model's circulation-field is slightly displaced and shows an acceptable overall performance for the subpolar North Atlantic, but fails to do it exactly for the OVIDE section? A comparison with more data-points would be preferential, or at least to use a larger surrounding of the OVIDE-section.

→ The two Referees highlight a disproportionate effort on the model evaluation biased by a comparison of only few parameters along one section while the paper treats about air-sea fluxes, transport and storage of anthropogenic CO2 in the North Atlantic Ocean. To respond to these comments, we suggest to join a new comparison of air-sea contemporary CO2 fluxes with Landschützer et al. (2015) as well as additional comparisons of mass transport and Cant concentration along the section 25°N (Hernandez et al., 2014; Zunino et al., 2015) like it is provided for OVIDE section (section 3.2 and 3.3) and of Cant transport along the Nordic sills (Jeansson et al., 2011). These new investigations will increase the existing but very short comparative analysis of air-sea fluxes, Cant transport and storage rate in the North Atlantic described in section 3.4 Budget of Cant in the North Atlantic Ocean (North of 25°N). In addition, we propose removing section 3.1 Distribution of hydrological and biogeochemical parameters along the OVIDE section, and rearranging part 3. Model evaluation over the OVIDE period to focus it on the paper subject (1. Advective transport of Cant, 2. Air-sea fluxes of anthropogenic CO2, 3. Storage of anthropogenic CO2) and not on the OVIDE section. This last comment includes to change the current title of this section, *Model evaluation over the OVIDE period*, by Model evaluation over the period 2003-2011.

Anonymous Referee #2 (2) Limits of the findings: In comparison with Perez et al. (2013), the considered model clearly seems to under-estimate transport into and out of the subpolar gyre box. Is it the justified to use this model to infer about mechanisms governing the subpolar gyre? If so, then the limits should at least be clearly mentioned. It is possible that the weak MOC of the model does not allow for general conclusions.

→ The referee 2 questions initially (i) on the use of the model NEMO-PISCES to infer about mechanisms governing evolution of Cant inventory in the subpolar gyre and then (ii) on the model limits.

(i) As we mentioned in section 3.4 lines 389 to 395, the time rate of inter-annual change in Subpolar Cant inventory over the period 2003-2011 is controlled by the northward transport of Cant-laden waters coming from South of 25°N, despite the under-estimation of the meridional circulation. This driving mechanism is corroborated by observation-based assessments from Perez et al. (2013) and Zunino et al. (2014 and 2015). In addition, Figs. 1 and 5, completed in the revised manuscript by new comparisons described above, show that the anthropogenic carbon is relatively well distributed in

the model, despite its under-estimation. These results made possible the use of our model to document the long term change in the Cant inventory of the subpolar gyre and its driving mechanisms.

(ii) Model analysis reveals in fact an under-estimation of the mass transport that is probably and partially due to the close to zero contribution of overflows water in the North Atlantic Ocean (line 302 to 311). The bad representation of this water masses is clearly a limit of this model that impacts both export of Cant concentration from arctic region and intensity of northward transport from subtropical region. Our analysis also reveals large air-sea anthropogenic CO2 fluxes compared to regional observations (lines 374-375). As suggested inherently lines 376-382, this over-estimation of air-sea fluxes is a response to the low Cant concentration transported inside the subpolar gyre and leads model to well store the anthropogenic Carbon in this region. As suggested by Referee, these limits will be clearly mentioned in the text and discussed in new section DISCUSSION.

Anonymous Referee #2 (3) Anthropogenic Carbon: The carbon difference between the historical and control run is NOT anthropogenic carbon. Instead, this kind of difference includes also climate change induced alterations in the natural carbon cycle, even though these alterations are probably small (see, for example, Frölicher, 2015).

As we explain in section 2.1, the historical and the control runs are forced with the same atmospheric reanalysis products, DFS4.2 over the period 1870 to 2001 and DFS4.4 between 2002 and 2012. The unique difference between both runs is the atmospheric CO2 concentration used to constrain air-sea CO2 fluxes. With our model configuration, potential alterations of natural carbon cycle in response to climate variability, which is inherent to DFS4, will be detectable on both simulations. The carbon difference between the historical and the control runs removes this potential bias and represents well the anthropogenic carbon. To clarify this essential point in the text, we propose to complete the sentence "Cant concentrations and anthropogenic CO2 fluxes were calculated as the difference between historical (total C) minus control (natural C component) simulations", lines 134-136 with, "Because potential alterations of natural carbon cycle in response to climate variability is inherent to DFS4, [...] in this model configuration".

Anonymous Referee #1 • (ii) Depth of analysis: This is my most important concern. As it stands, the paper is imbalanced in that too much effort is spent on evaluating the model results (also for aspects that are not so relevant for the question at hand), while the main objective of the paper is not covered in sufficient depth. As it stands, the manuscript remains essentially descriptive in its discussion of the variability in the different terms making up the budget, but does not really identify and discuss the underlying mechanisms. For example, it would be important to know and understand what drives the variability in transport, fluxes and storage. A correlation with an index is not really insightful enough here. What is needed is a disentanglement of the relative contribution of mass transport, changes in concentrations, and residence times within different water masses (density classes). The approach taken by Daniele Iudicone could serve as an excellent template here, i.e., Iudicone et al. (2016).

Anonymous Referee #2 (4) NAO-phases: I am not convinced by the MOC weak and strong phases as presented in Figure 9. I would much rather see this presented together with the NAO-index of the model over this time-period. With this approach, it should also be possible to find more than 3 different NAO-phases and back the results up for all positive and negative NAO phases. After all, that is the advantage of a model over data.

Anonymous Referee #2 (5) Mechanisms: The paper should focus a bit more on the detailed mechanisms that the model simulates that are leading to these different NAO-responses, i.e. what mechanism is behind a different carbon uptake. Are those mechanisms seen in reality or by other models?

→ The two referees find that section 4 is essentially descriptive and doesn't cover the objective of this paper in sufficient depth. They also express some reservations on the evaluation of driving mechanisms according to three NAO periods. Referee 1 suggests to use the approach taken by ludicone et al (2016) as a template for our study. We have considered these most important and justified reviews. To enhance our paper without change it in depth, we have revised this section and we propose :

- (i) Removing analysis according to NAO-phases. The role of this major atmospheric forcing is nevertheless evaluated using DJFM- NAO index over the period 1959-2011. Figure 9 will be thus changed in the revised manuscript by new figure with a time series of DJFM-NAO index.
- (ii) Changing Figs. 8 and 10 by a new fig. showing times series of Cant storage rate, air-sea Cant fluxes and transport divergence of Cant over the period 1959-2011 to document temporal evolution of the anthropogenic carbon budget in the North Atlantic. To be consistent with model-data comparison, we work between 25°N and the Nordic sills but we considered in this section three boxes instead of two (like in section model data comparison): 25°N-36°N; 36°N-OVIDE; OVIDE-Sills. Boundary 36°N is added to exclude the northern part of the subtropical region (25°N to 36°N) from the subpolar region (36°N to Sills) as mentioned line 409.

This new figure, completed by a new table of correlations, highlights clearly the contribution of variability of (i) both Cant transport and air-sea fluxes on Cant inventory variability and (ii) both circulation and Cant accumulation on Cant transport variability. We obtain two majors results not clearly explained or interpreted in old manuscript:

(1) The interannual to decadal variability of the Cant inventory is strongly influenced by the northward advective transport whatever atmospheric conditions, which is sometimes, reinforced by air-sea fluxes.

(2) At the multi-decadal time scale, air-sea Cant fluxes and Cant transport contribute to the long term change in Cant storage rate with a ratio of 50/50 between 25°N and 36°N, from ~90/10 to 75/25 between 36°N and OVIDE and 100/0 between OVIDE and the Nordic sills.

(iii) Adding a new analysis in density classes as suggested by Referee 1. Based on the water column distribution of mass transport integrated into density (sigma 1), we identify 3 classes: Classe 1 = North Atlantic Central Water (NACW) transported northward; Classe 2: Intermediate water; Classe 3: North Atlantic Deep Water (NADW). Mass transport, Cant transport and change in concentration were estimated for each density class over the period 1959-2011.

We obtain three major results from this additional analysis concerning:

(1) the contribution of advected water masses on the anthropogenic CO2 storage: Indeed, NACW coming from subtropical region contributes largely (i) to the Cant storage between 25°N and 36°N and (ii) to the formation of the NADW in the subpolar gyre, whereas NADW, whose production is strongly correlated with the DFJM-NAO index, contributes to the storage of Cant between 36°N and OVIDE.

(2) the exportation of Cant stored in the deep ocean in the subpolar gyre: Only 1/3 of the NADW formed in the subpolar gyre is exported toward south region.

(3) the reason of the increase of the contribution of the Cant transport on the storage of the anthropogenic CO2 between 36°N and OVIDE from mid-1990's to the detriment of air-sea fluxes regarding.

These new results will be clearly mentioned in new part 3. RESULTS and discussed in new part 4. DISCUSSION regarding first results from model-data comparison and into the context of other people's work.

Anonymous Referee #1 • (iii) Discussion: The article does not really contain a discussion, i.e., a place where the results from this study are put back into the context of other people's work. I also miss a thorough assessment of the robustness of the conclusions given the uncertainties and biases in the model and the data. Finally, there is also no discussion right now about what this all means and what we should conclude from this regarding the future uptake of CO2.

→ To clarify our message and respond reviewer, we introduced a section 3.RESULTS and a section 4. DISCUSSION where we will discussed amongst other think of (i) the important role of NACW in the storage of Cant in the subpolar North Atlantic gyre, (ii) the role of the atmospheric forcing in the NADW production, (iii) The consequences of the bad representation in the model of overflows waters and (iv) the compensation of the transport of Cant by the air-sea fluxes regarding the future uptake of CO2.

Anonymous Referee #1 Minor comments

Abstract: The abstract is a good example to illustrate my most important concern. In essence not much more than 4 lines (end of line 30 to the beginning of line 35) is devoted to the results and the underlying drivers, while the remaining 13 lines are devoted to motivation, method, and outlook. This is not a good balance, in my opinion. Concretely, I suggest to shorten the introduction part (lines 20 through 26) and the method and evaluation parts (lines 26 to 30), in order to generate the necessary space for a more in depth discussion of the results and the key governing processes. Introduction, line 44: Add uncertainty to uptake fraction. Introduction: The introduction provides a nice summary and concludes with a clear objective. However, I am wondering about a missed opportunity here. By focusing exclusively on the role of anthropogenic CO2, the authors forgo the opportunity to truly link air-sea CO2 fluxes, transport and storage of inorganic carbon. This is merely a thought, and by no means a request to substantially alter the orientation and scope of this paper. But it may serve as a motivation for taking the next step.

→ Abstract will be rewritten regarding new results described above and referee comments.

Methods: p5, line 166ff: 'CT method: Each of the different methods to separate Cant from the background comes with its uncertainties and biases. It would be actually quite insightful to investigate the robustness of the conclusions with regard to the choice of separation technique. Would it be possible to use estimates from the TTD or the _C* methods?

→ Vázquez Rodríguez et al. [2009] or Guallart et al. [2015] evaluate the consistence between different methods estimating Cant concentration in the North Atlantic Ocean and at the 24.5°N respectively. They obtain a good agreement between methods. Estimations are in fact associated to an uncertainty but, regarding these inter-comparisons, choice one or two method to evaluate Cant concentration from observations data set will change the intensity of the under-estimation of Cant transport or the over-estimation of air-sea anthropogenic CO2 fluxes but not the direction/trend. However, to improve our paper and regarding referee comments, we will mention these inter-comparisons in the new section 2.2 *Observation data set* (instead of OVIDE data set regarding first part of our response), and, as previously explained, model limits will be discussed in section 4 regarding bias and uncertainties.

Methods: p6, line 190ff: Offline approach: It is unclear why some of the calculations were done offline. Wouldn't it have been easier to do all analyses online? This would have avoided the need to neglect the contributions from diffusion and eddy transport.

 \rightarrow Online version is only available for the period 2003-2011, but as show on figures S2 and S4 in supplementary:

(Advective Tcant) $_{online} \approx (Advective Tcant + eddy Tcant + diffusive Tcant)_{online}$

(Advective Tcant)_{offline} ≈ (Advective Tcant + eddy Tcant + diffusive Tcant)_{online}

We could have worked only with offline version and estimated advective transport of Cant, but using online version show that the anthropogenic carbon in the model is well advected and not transported with eddies or by diffusion. We will reformulate line 190-192 to avoid confusion.

Model evaluation, p7ff: This section is overly long, and in many respects also not that relevant. Further, as mentioned in my first major comment, it is a bit biased in the sense that the focus is almost exclusively on the data from the Ovide section, thereby omitting important other constraints. For example, there is no need to evaluate the modeled nutrient and oxygen distributions as they are not relevant for this paper. I thus recommend to substantially shorten this section, so that more space is available for the really novel aspect of the paper, i.e. section 4

→ This section has been rearranged as mentioned above.

Model evaluation, p9, line 281: "accumulated arrangement". This is unclear. What is meant here?

→ Vertical accumulated arrangement correspond to Fig 5a, i.e. water column distribution of integrated mass transport in density (sigma 1 with 0.01 kg m-3 resolution); horizontal accumulated arrangement correspond to fig. 5b, i.e. integrated mass transport from Greenland to Portugal. We will reformulate this line.

Model evaluation, p9, line 307ff: This bias is clearly important, but its implications are only partially discussed later on. This should be improved.

→ As mentioned above, this bias will be discussed in revised version in the new section 4. Discussion.

p10, line 336: change "processes" to "process".

p11, line 367: mTadv Cant: This is very cryptic. Is it really necessary to use a symbol that is not intuitive and that one needs to look up 2 pages above instead of simply writing the advective transport of Cant?

→ These will be corrected in revised manuscript.

p11, line 376ff: "overestimation": This is a reasonable interpretation, but key here is really the surface ocean concentration of Cant, and not really the concentrations at depth. Thus, I recommend to discuss this more specifically.

Model evaluation, p12, lines 390ff: This statement is hard to understand and follow without a more thorough discussion of the underlying mechanisms. This is a key finding, and thus needs to be given the emphasis it needs.

Long-term change, p12ff: This is where the paper starts to become really interesting. Unfortunately, only a little bit more than 2 pages are devoted to this most novel aspect of this study. This is clearly unbalanced when considering that section 3 was given more than 5 pages.

Long-term change, p13, section 4.1: This section needs to be improved. As it stands it is very difficult to understand. For starters, I would re-evaluate whether the symbols are really a good strategy to provide clarity (in my opinion, they don't). Also, the authors are providing too many details (also too many numbers), so that the important message gets drowned. Further, the writing is complex and lacks a good storyline.

Long-term change, p13-14, section 4.2: A good fraction of the analysis here builds on correlations. While this provides a good starting point, it does not provide a fruitful avenue to develop a good understanding of the mechanisms and processes driving the responses. As suggested in my major comment above, I think a more process oriented framework would be very helpful here. On top of this, also section 4.2 is not that well written, and like 4.1 could be much improved to increase its readability.

p15, line 492. Wouldn't it make sense to add here a Conclusion section? Otherwise, this last paragraph makes little sense. p15, line 497 "preconditioning". As far as I was able to discern, this preconditioning has not been shown.

→ All of these minor comments echo major comments to which we answer above

Figure 4: I strongly recommend to add an estimate of the uncertainty to the observation-based estimates of transport. Figure 5: Caption. Replace "Shadows" with "shaded band".

Figure 5: Caption. Please specify location of the two estimates more explicitly.

Figure 6: Unclear what the standard deviations is based upon and what its meaning is.

Figure 9: Grey bands are not visible in my printed version.

→ We will consider figure comments in the revised manuscript

Anonymous Referee #2 – Technical comments

→ All technical comments will be corrected in the revised manuscript and references added.