Dear Editor,

We have considered the comments from the 2 referees with great attention. We detail below how we intend to address these comments. We have combined our replies to both referees when their major comments were converging / overlapping. We then list all minor comments and respond to them individually.

We hope that you will find our responses convincing and that we can continue with the submission of the 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.*

➔ Both referees highlight a disproportionate effort on model evaluation, biased by a comparison of selected parameters along one specific hydrographic section, while the paper aims at discussing air-sea fluxes, transport and storage of anthropogenic CO2 (Cant) in the North Atlantic Ocean. To respond to these comments, we will expand our model-data comparison by adding:

- a comparison between the simulated air-sea contemporary CO2 fluxes and the data-based reconstruction by Landschützer et al. (2015),
- additional model-data comparisons for mass transport, Cant transport and Cant concentration along the section 25°N (Hernandez-Guerra et al., 2014; Zunino et al., 2015; Guallart et al., 2015), as well as for Cant transport over the Nordic sills (Jeansson et al., 2011).

These additional model-data comparisons will strengthen the assessment of simulated air-sea fluxes, Cant transport and storage rate in the North Atlantic, presented in section *3.4 Budget of Cant in the North Atlantic Ocean (North of 25°N)*.

In addition, we propose :

- to remove section *3.1 Distribution of hydrological and biogeochemical parameters along the OVIDE section*,
- to rearrange part *3 Model evaluation over the OVIDE period* to focus it more clearly on the paper's topic (1. Advective transport of Cant, 2. Air-sea fluxes of anthropogenic CO2, 3. Storage of anthropogenic CO2) and not on the OVIDE section only. It will imply a modification of the current title of this section, from *Model evaluation over the OVIDE period*, to *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.

➔ Referee 2 (i) questions the use of the NEMO-PISCES model for inferring the mechanisms governing the evolution of the Cant inventory in the subpolar gyre and (ii) asks for an improved discussion of 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 in the model simulation by the northward transport of Cant-laden waters coming from South of 25°N, despite the under-estimation of the meridional circulation. Observation-based assessments from Pérez et al. (2013) and Zunino et al. (2014 and 2015) corroborate this driving mechanism. In addition, Figs. 1 and 5, completed in the revised manuscript by the new model-data comparisons described above, show that the Cant is relatively well distributed spatially in the model, despite some under-estimation. We think this justifies the use of our model to document the long-term changes in the Cant inventory of the subpolar gyre and its driving mechanisms. Nevertheless, the limits of the model, and hence of its use to discuss Cant transport / storage, will be stated very clearly.

(ii) Our model analysis reveals an under-estimation of the MOC that is probably in part due to the close to zero contribution of overflow waters entering the North Atlantic Ocean over the Nordic sills (line 302 to 311). This biased representation is clearly a limit of the model and impacts both the export of Cant from the arctic region as well as the intensity of the northward transport from the subtropical region. Our analysis also highlights that air-sea fluxes of Cant in the model are larger than observations (lines 374-375). As suggested in lines 376-382, the over-estimation of air-sea fluxes is a response to and compensates the too low Cant concentration transported towards the subpolar gyre. This compensation contributes to a satisfying reproduction of Cant storage by the model in this region. Nevertheless, as stated above, the primary driving mechanism of Cant storage variability on inter-annual timescales remains the variability of the MOC, and thus the northward transport of Cant-laden waters coming from South of 25°N. This primary driving mechanism is corroborated by observation studies. As far as we can judge from the limited set of observational studies available, and despite the knowledge limits of the model-based approach, the model reproduces at first order the observed balance between Cant air-sea fluxes and transport explaining the variability on interannual timescales of Cant storage, justifying a more in depth analysis. As suggested by the referee, the model limits will be more clearly mentioned in the text and discussed in a new DISCUSSION section.

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 explained 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 only difference between both runs is the atmospheric CO2 concentration used to constrain air-sea CO2 fluxes: one with pre-industrial CO2 concentration (NATURAL) and second one with historical trajectory (contemporary). With this model configuration, potential alterations of the natural carbon cycle in response to climate variability are inherent to DFS4 and will thus be detectable on both simulations. Taking the difference of carbon concentrations between both runs removes the alteration in the natural carbon cycle and represents the anthropogenic carbon (see also Orr et al., 2017 where authors explain such protocol) To clarify this essential point in the text, we propose to (i) change "control simulation" by "natural simulation" and (ii) complete the description of both simulations with: "In our model configuration, potential alterations of natural carbon cycle in response to climate variability are inherent to DFS4".

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 the three NAO periods we have selected. Referee 1 suggests to use the approach taken by Iudicone et al (2016) as a template for our study. We agree with these comments and have decided to revise Section 4 accordingly.

We propose:

(i) To revise the analysis according to NAO-phases. The evaluation of the role of this major atmospheric forcing is improved by using the DJFM-NAO index over the period 1959-2011. Figure 9 will be replaced in the revised manuscript by a new figure based on the full time series of this index. In particular, we demonstrate the well-known correlation between the variability of deep water formation, as well as Cant uptake and the NAO index.

(ii) To replace consequently Fig.10 representing boxes correlated to the three major NAO periods as well as Fig. 8 by a new figure showing times series of Cant storage rate, air-sea fluxes and transport divergence over the period 1959-2011 to illustrate the simulated temporal evolution of the anthropogenic carbon budget in the North Atlantic. To be consistent with the model-data comparison, we work between 25°N and the Nordic sills but we consider three boxes instead of the two used for the 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 mid-latitude region (36°N to Ovide) as mentioned in line 409.

This new figure highlights the contribution of variability of both Cant transport and air-sea fluxes on Cant inventory variability.

These changes emphasize two results that had not been discussed in depth in the previous version of the manuscript:

(1) The interannual to decadal variability of the Cant inventory is strongly influenced by the northward advective transport variability. It is sometimes but not always 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) To add 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.
The additional analysis highlights:
(1) the contribution of advected water masses to the anthropogenic CO2 storage: the NACW moving northwards from the subtropical region contributes largely (i) to the Cant storage between 25°N and 36°N and (ii) to the formation of NADW in the subpolar gyre, which is strongly correlated with the DFJM-NAO index and contributes to the storage of Cant between 36°N and OVIDE.
(2) the export of Cant from the subpolar gyre by southward flowing NADW: only 1/3 of the Cant sequestered by NADW formed in the subpolar gyre is exported southward.
(3) the increasing contribution of Cant transport divergence to its storage between 36°N and the OVIDE section from the mid-1990's onward.

These results will be presented in the revised "3. RESULTS" section. They will be discussed in the context of model-data comparison and results from previous studies in the newly added "4. DISCUSSION" section.

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 to the reviewer, we will introduce a section 3.RESULTS and a section 4. DISCUSSION. The discussion will be expanded by considering (i) the contribution of NACW to the storage of Cant in the subpolar North Atlantic gyre, (ii) the role of the atmospheric forcing in NADW formation, (iii) the consequences of the underestimation by the model of overflow waters and (iv) the respective contribution of Cant transport divergence and air-sea fluxes to the Cant storage compare to observation. We will address the influence of these mechanisms on the future evolution of the 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.

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

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.

➔ As referee mentioned, discuss link between air-sea CO2 fluxes, transport and storage of inorganic carbon will be very interested but it is another study.

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] and Guallart et al. [2015] evaluated the consistency of different methods for deriving the concentration of Cant in the North Atlantic Ocean. They obtained a good agreement between methods, each of which has its own uncertainty. The choice of one method over the other would result in a change of the intensity of the under-estimation of Cant transport or the over-estimation of air-sea anthropogenic CO2 fluxes, but it would have no effect on computed trends. We will, however, follow the referee and mention these data inter-comparisons in the revised section 2.2 now labeled *Observation data set (*instead of 2.2. OVIDE data set). As previously mentioned, model limits (e.g. biases and uncertainties) will be discussed in section 4.

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.

➔ Model output for the online analysis is only available for the period 2003-2011 so we can't analyze long term change. Nevertheless, the comparison between the online and offline approaches demonstrates that the net transport of Cant is dominated by the advection term and that the contribution of eddy and diffusive transport can be neglected. We will reformulate lines 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 at the beginning of this letter.

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

➔ Vertical accumulated arrangement (Fig 5a) refers to the cumulative distribution of integrated mass transport in density space (sigma 1 with 0.01 kg m-3 resolution); horizontal accumulated arrangement (fig. 5b) corresponds to the vertically integrated mass transport cumulated from Greenland to Portugal. All of these refer to the stream function. We will reformulate lines 280-282 as "the stream function simulated by ORCA05-PISCES is quite similar to those estimated from the OVIDE data set".

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

➔ We agree with the referee that this bias is important. As mentioned above, it will be discussed in the revised manuscript 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 the 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.

➔ Please refer to the reply to 'major comments' for our suggestion on how to address these issues.

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.

➔ Please refer to the reply to 'major comments' for our suggestion on how to address these issues.

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.

➔Please refer to the reply to 'major comments' for our suggestion on how to improve the structure and balance of the manuscript.

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.

➔Please refer to the reply to 'major comments' for our suggestion on how to address these issues.

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.

➔Please refer to the reply to 'major comments' for our suggestion on how to address these issues.

p15, line 492. Wouldn't it make sense to add here a Conclusion section? Otherwise, this last paragraph makes little sense.

➔We agree with the referee and we will add a 'CONCLUSION' section to the revised manuscript.

p15, line 497 "preconditioning". As far as I was able to discern, this preconditioning has not been shown.

➔Here "preconditioning" refers to the Cant uptake through the air-sea interface mainly in the subtropical North Atlantic, preconditioning the waters that flow northward.

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 address all comments during revisions

Anonymous Referee #2 – Technical comments
➔ All technical comments will be corrected in the revised manuscript and references added.

<u>References</u>

Guallart et al., (2015) : Trends in anthropogenic CO2 in water masses of the Subtropical North Atlantic Ocean. Progress in Oceanography, doi: http://dx.doi.org/101016/j.pocean.2014.11 .006

Hernandez-Guerra et al., (2014) : Meridional overturning transoprt at 7.5N and 24 ?5N in the Atlantic Ocean during 1992-93 and 2010-11. Progress in Oceanography 128, 98-114. http://dx.doi.org/10.1016/j.pocean.2014.08.016

Jeansson et al. (2011) : The Nordic Seas carbon budget : sources, sinks and uncertainties. Global Biogeochemical Cycles, 25(4).

Landschützer et al. (2015): A 30 years observation-based global monthly gridded sea surface pCO2 product from 1982 through 2011. http://cdiac.ornl.gov/ftp/oceans/SPCO2_1982_2011_ETH _SOM_FFN. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, US Department of Energy, Oak Ridge, Tennessee. Doi:10.3334/CDIAC/OTG.SPCO2_1982_2011_ETH_ SOM-FFN

Orr et al. (2017): OMIP biogeochemical protocols and diagnostics for CMIP6. Geoscientific Model Development. In press

Pérez et al. (2013) : Atlantic Ocean CO2 uptake reduced by weakening of the meridional overturning circulation, Nature Geoscience, 6(2), 146-152, doi: 10.1038/NGEO1680, 2013

Vázquez-Rodríguez et al. (2009): An upgraded carbon-based method to estimate the anthropogenic fraction of dissolved CO2 in the Atlantic Ocean, 685 Biogeosciences Discuss., 6, 4527–4571, doi:10.5194/bgd-6-4527-2009

Zunino et al., (2015): Transports and budgets of anthropogenic CO2 in the tropical North Atlantic in 1992–1993 and 2010–2011, Global Biogeochem Cy, 29(7), 1075-1091.