

## **Interactive comment on “Trends of anthropogenic CO<sub>2</sub> storage in North Atlantic water masses” by F. F. Pérez et al.**

**Anonymous Referee #2**

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Review of “Trends of anthropogenic CO<sub>2</sub> storage in North Atlantic water masses” by F. F. Pérez et al. This contribution discusses the development of anthropogenic CO<sub>2</sub> storage in the North Atlantic over the last three decades by considering both the trends in anthropogenic CO<sub>2</sub> concentrations as well as the changes in water mass distribution that have taken place over the years. It is an exiting approach, which provides important information, however, there are many issues that must be dealt with before it can be published. These are given in the following.

1. Unfortunately I found the paper extremely frustrating to read, because of three issues:

(a) The language is far from adequate. There are numerous examples in the text of wrong use of words, odd formulations, strange sentences structures, wrong tense etc. The language must be significantly revised.

REPLY:

We apologize for this language inconvenience. We have been very meticulous about written expression in the revised version of the manuscript and tried our best to correct it. Thank you for the specific recommendations you provided in your review letter as well. They have all been included. It is our hope that the new version has come out clearer, more concise and in proper English.

(b) Section 3 was tedious to read. It contains 10 equations, many of which are essentially repeats of each other, but with minor modifications (e.g. (1), (6) and (10)), they do not appear in order (Eq. (10) is mentioned before Eq. (7)) and there is simply an excessive amount of explanatory text. This section should be shortened, it should be simplified, any adjective-rich subjective evaluation should be removed (for example page 171, line 19, “to produce high performance parameterizations”). Please, briefly outline your line of thought, and then briefly provide the computational framework with the key equations.

REPLY:

The revised version of the manuscript includes a significantly reduced section 3 with only 2 equations instead of 10, following your comments and recommendations. The new section 3 is divided in two sub-sections: 3.1 outlines the  $\varphi C_T^\circ$  method used to estimate  $C_{\text{ant}}$  concentrations; 3.2 describes how  $C_{\text{ant}}$  inventories were calculated in each of the three basins (Irminger, Iceland and ENA). The minutiae for the thickness calculations of the Irminger and Iceland, and  $C_{\text{ant}}$  corrections for the ENA basin have

been moved to the newly added Appendix I. Thus, the new methodology section only includes the most relevant equations to support the line of thought that we followed (how inventories are calculated), as you suggested. We do not include the aforementioned modifications in this reply letter to avoid making it excessively long. Please, refer to Section 3 and Appendix I in the revised manuscript.

(c), the results are also confusing, and I think the paper would benefit if figure 3 was introduced at the very start, the main results on Cant storage rates and their variability was summarized, and then discussed in terms of causes and effects. Table 2 can be replaced with a figure that shows the trends of layer thickness, T, S, AOU and silicate. At least a figure with layer thickness should be included.

REPLY:

We have reorganized and reduced significantly the results section after this comment, so that now more focus is put in results dealing directly with temporal trends of  $C_{ant}$  concentration and storage rates. Also, by having greatly reduced the first descriptive part of section 4 now Fig. 3 is introduced much earlier, which is the most important one as you point out. We have kept the order of figures up to Fig. 3, though (Fig. 4 has moved to Fig. 5 and viceversa), because we needed to show, even if briefly, how the fields of measured  $\theta$ , S and AOU and the estimated  $C_{ant}$  fields looked like in order to be able to come back to them when necessary to explain the  $C_{ant}$  storage trends observed in Fig. 3.

The suggested graph from the data in Table 2 is somewhat already shown in the work from Pérez et al. (2008) dealing with the Irminger basin. However, we were a bit confused about your last suggestion, since old Fig. 4 (Fig. 5 in the revised manuscript) already showed the layer thickness evolution of the different water masses in the Irminger and Iceland basins, as you suggest.

2. Abstract and elsewhere. The term “storage capacity” is used. I think of capacity as unused potential, what can be absorbed, not has been absorbed. The authors seem to use the latter. Another word should be used, in many cases “capacity” can just be deleted.

REPLY:

We have revised the whole text for consistency so that, for instance, when  $\mu\text{mol kg}^{-1} \text{ yr}^{-1}$  is used, we talk about “rate of change of  $C_{ant}$  concentration”,  $\text{Gt C yr}^{-1}$  refers to “storage rates” and  $\text{mol C m}^{-2} \text{ yr}^{-1}$  stands for “ $C_{ant}$  specific inventory rates”.

The abstract has also been reviewed according to the definitions given above. The “capacity” term was misleading, as you noted, and has been deleted in all occurrences:

*“A high-quality inorganic carbon system database spanning over three decades (1981-2006) and comprising 13 cruises has allowed applying the  $\varphi C_T^\circ$  method and coming up with estimates of the anthropogenic  $\text{CO}_2$  ( $C_{ant}$ ) stored in the main water masses of the North Atlantic. In the studied region, strong convective processes convey surface properties, like  $C_{ant}$ , into deeper ocean layers and confer this region an added oceanographic interest from the point of view of air-sea  $\text{CO}_2$  exchanges. Commonly, a tendency for decreasing  $C_{ant}$  storage rates towards the deep layers has been observed. In the Iberian Basin, the North*

*Atlantic Deep Water has low  $C_{ant}$  concentrations and negligible storage rates, while the North Atlantic Central Water in the upper layers shows the largest  $C_{ant}$  values and largest annual increase of its average concentration ( $1.13 \pm 0.14 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ). This unmatched rate of change in the  $C_{ant}$  concentration of the warm upper limb of the Meridional Overturning Circulation decreases towards the Irminger basin ( $0.68 \pm 0.06 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ) due to the lowering of the buffering capacity. The mid and deep waters in the Irminger Sea show rather similar  $C_{ant}$  concentration rates of increase (between  $0.33$  and  $0.45 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ ), whereas in the Iceland basin these layers seem to have been less affected by  $C_{ant}$ . Overall, the  $C_{ant}$  storage rates in the North Atlantic subpolar gyre during the first half of the 1990s, when a high North Atlantic Oscillation (NAO) phase was dominant, are  $\sim 48\%$  higher than during the 1997-2006 low NAO phase that followed. This result suggests that a net decrease in the strength of the North Atlantic sink of atmospheric  $\text{CO}_2$  has taken place during the present decade. The changes in deep-water ventilation are the main driving processes causing this weakening of the North Atlantic  $\text{CO}_2$  sink.”*

3. Abstract, line 22 “Detrimental” is not the right word, use “reduced”.

REPLY:

Done. The last sentence in the abstract now reads as follows: *“The changes in deep-water ventilation are likely the main driving processes causing this weakening of the North Atlantic  $\text{CO}_2$  sink.”*

4. Page 166, line 25. When referring to Sabine, it is “anthropogenic  $\text{CO}_2$  sink” not just “ $\text{CO}_2$  sink”

REPLY:

Corrected. Thank you for noticing.

5. Page 167, line 5, replace “bring forth” with “have”

REPLY:

Done.

6. Page 167, line 7. The extent to which a slowdown of the MOC would reduce ocean  $\text{CO}_2$  uptake is a matter of debate, Swingedouw et al, GRL, 2007. Consider to use more than the Sarmiento and LeQuéré reference.

REPLY:

Thank you very much for suggesting this reference, which we have now included in this context. This paper shows from model simulations that opposing processes of less saline and cooler processes tend to limit the effect of the Greenland Ice Sheet (GIS) melting on  $\text{CO}_2$  uptake, which is responsible for up to 25% of the MOC slowdown.

The new line reads as follows:

*“Although the effects of the MOC slowdown are still a matter of debate (Swingedouw et al., 2007), it is likely that they will cast profound consequences on global climate due to the*

*associated decrease in heat transport (Drijfhout et al., 2006) and oceanic  $C_{ant}$  uptake (Sarmiento & Le Quéré, 1996)”*

7. page 167. During high NAO, when LSW formation is reduced, Nordic Seas convection is intensified, and we may expect ISOW with higher loads of Cant into the North Atlantic. Perhaps the authors may quantify the extent to which this cancels the effect of reduced LSW formation on Cant column inventories?

REPLY:

This suggestion is very interesting, but somewhat confusing... We stated several times in the manuscript that during the high NAO period the formation of LSW was intensified due to the stronger convection (Yashayaev et al., 2007; 2008) and not the other way around as you suggest. Anyhow, we understand this might have been a mistake.

In either case, there is literature (even as suggested by yourself in comment #20) stating that the deep waters in the Nordic Seas (overflows ISOW and DSOW) behave quite steadily in terms of formation rates (Olsen et al., 2008), so they could not be expected to counterbalance the LSW contributions to  $C_{ant}$  uptake in the NASPG.

Olsen, S.M., Hansen, B., Quadfasel, D., Osterhus, S, Observed and modelled stability of overflow across the Greenland-Scotland ridge. Nature, vol. 455, 25, 519-523, doi:10.1038/nature07302, 2008.

8. Page 168, line 22. I'd expect Oaces in full caps (i.e. OACES)

REPLY:

Corrected.

9. Page 169, line 17, replace “exceptionally” with “on some occasions”

REPLY:

Done.

10. Page 169. Corrections to the TTO data were suggested by Tanhua and Wallace, 2005. Use these.

REPLY:

Thank you for noticing. We did apply (but forgot to mention) the correction of  $-3.0 \mu\text{mol}\cdot\text{kg}^{-1}$  to the  $C_T$  data from the TTO suggested by Tanhua and Wallace (2005). In fact, this same correction had been previously applied in a previous paper of the authors (Pérez et al., 2008). The sentence has now been extended:

*“...The exception to the latter is the 1981 TTO cruise, where  $C_T$  was determined potentiometrically (Bradshaw et al., 1981) and no CRMs were used. Tanhua and Wallace (2005) performed a cross-over analysis between this cruise and an overlapping more recent one. Based on a comparison with CRM-referenced data, they suggest a correction for TTO-NAS  $C_T$  measurements of  $-3.0 \mu\text{mol}\cdot\text{kg}^{-1}$ , which has been applied to our dataset.”*

11. Page 170, line 6, I do not understand, some words must be missing.

REPLY:

The sentence has been rewritten. We hope the meaning is now clear:

*“...The geographic position and timely date of these two cruises made them assets to this study. Both cruises had comprehensive amounts of coulometric  $C_T$  measurements yet very few potentiometric  $A_T$  data. Given the shortage of  $A_T$  data they were not discarded from our dataset.”*

12. Page 170, line 13. Replace “3-D grid nodes” with sampling depths

REPLY:

Done.

13. page 171, lines 11-24. I question the validity of using an approach that has not yet been published in a peer-reviewed journal, to calculate the anthropogenic CO<sub>2</sub> concentrations. I also question the clearly highly subjective review that is given here. In order to give credibility to their results I think that the authors must (1) use an additional approach, which has been reviewed positively, and evaluate whether this gives the same trends, and (2) they must tone down their review of their own approach. Please state only accuracy.

REPLY:

In the new version of the manuscript we have removed all subjective adjectives dealing with the  $\varphi C_T^\circ$  method review, following your general recommendations and this one (Please, refer to Section 3.1). A paragraph on the accuracy has been added upon your request:

*“...A random propagation of the errors associated with the input variables necessary to calculate  $C_{ant}$  according to the  $\varphi C_T^\circ$  formulation yielded an overall uncertainty of  $\pm 5.2 \mu\text{mol}\cdot\text{kg}^{-1}$  for the  $C_{ant}$  estimates obtained with this methodology. This practice for calculating uncertainties has been successfully used in the past by many authors (Gruber et al., 1996; Lee et al., 2003; Lo Monaco et al., 2005).”*

Regarding the validity of the  $C_{ant}$  calculation method applied, the handling editors of the  $\varphi C_T^\circ$  method manuscript rejected to publish it in BG because they considered that the method was already described well-enough in Vázquez-Rodríguez et al. (2009) (although no equations were provided there) and successfully applied in Pérez et al. (2008), not because it was inconsistent (discussion threads can be followed in the BGD website: <http://www.biogeosciences-discuss.net/6/4527/2009/bgd-6-4527-2009-discussion.html>).

Anyhow, we did not have too many choices when deciding which  $C_{ant}$  estimation method to apply here, surprising as it sounds: The TTD and  $\Delta C^*$  methods both need CFC data to make their estimates, so that leaves those two candidates out of the list. The eMLR method is based on repeated sections over time, which we did not have in our case except for the meridional cruises TYRO (1990), OACES (1993), CHAOS

(1998) and A16N (2003), which only covered a small area of the Iceland and ENA basins. The  $C_{\text{IPSL}}^{\circ}$  method from Lo Monaco et al. (2005) was proven to yield consistently higher estimates in the Atlantic than the  $\Delta C^*$ , TTD, TrOCA and  $\varphi C_{\text{T}}^{\circ}$  methods (Vázquez-Rodríguez et al., 2009). Therefore, we only really could choose between the  $\varphi C_{\text{T}}^{\circ}$  and TrOCA methods. The TrOCA method tends to yield higher  $C_{\text{ant}}$  inventory estimates than other methods in the NASPG (Fig. 6 in Vázquez-Rodríguez et al., 2009). Also, the recent work from Yool et al. (2010) questions the theoretical foundations of the TrOCA approach and, when compared with model outputs, very large biases (up to 50%) are revealed. These were the reasons why we decided not to include TrOCA (nor any of the above-mentioned methodologies) estimates in our work in the first place, and chose to apply the  $\varphi C_{\text{T}}^{\circ}$  method.

However, since the TrOCA method is very easy to apply, we did the calculations and compared them to the results we obtained, following your advice. The obtained results are summarised in the table included below. We found that the  $C_{\text{ant}}$  concentrations estimated with TrOCA were about 20% higher than those from the  $\varphi C_{\text{T}}^{\circ}$  method, as expected (Vázquez-Rodríguez et al., 2009; Yool et al., 2010). However, when comparing the trends in the  $C_{\text{ant}}$  storage rates, no statistical significant difference was found in most cases between the two sets of slopes (table below).

Specific Inventory Rates ( $\text{mol C m}^{-2} \text{ yr}^{-1}$ )			
Basin	NAO Phase (time period)	$C_{\text{ant}} \varphi C_{\text{T}}^{\circ}$	$C_{\text{ant}} \text{TrOCA}$
<b>IRMINGER</b>	High (1991-1997)	1.74±0.24	1.88±0.51
	Low (1997-2006)	0.40±0.3	0.43±0.31
<b>ICELAND</b>	High (1991-1998)	1.88±0.45	1.91±0.76
	Low (1997-2006)	0.30±0.2	0.55±0.42
<b>ENA</b>	(1981-2006)	0.72±0.03	0.76±0.11

The main points raised in the above discussion and this table have been included in the Appendix II of the revised manuscript, as stated in the first paragraph of section 3.1:

*“...Appendix II discusses further the choice of the  $\varphi C_{\text{T}}^{\circ}$  method with respect to other methodologies, and a comparison of results is made with the TrOCA approach (Touratier et al., 2007).”*

Yool, A., Oschlies, A., Nurser, A. J. G., and Gruber, N.: A model-based assessment of the TrOCA approach for estimating anthropogenic carbon in the ocean, *Biogeosciences*, 7, 723-751, 2010.

14. Page 171, line 22-23. The method does not bring the estimates from the other

methods closer together, it estimates  $C_{ant}$  concentrations which falls between the estimates of the other methods. Please revise section.

REPLY:

This sentence has been now omitted. The paragraph reviewing the  $\varphi C_T^\circ$  method (Section 3.1) stands now as follows:

*“The concentrations of  $C_{ant}$  shown in Fig. 2 (and in the rest of cruises, not plotted) were estimated applying the  $\varphi C_T^\circ$  method (Pérez et al, 2008; Vázquez-Rodríguez et al., 2009a). The  $\varphi C_T^\circ$  method is a process-oriented geochemical approach that attempts to account for the nature and evolution of the phenomena that ultimately have affected oceanic  $C_{ant}$  storage since the 1750s. The method considers processes that control the uptake of  $C_{ant}$  by the ocean: from the biogeochemistry of the marine carbon cycle to the mixing and air-sea exchanges. It also considers the spatiotemporal variability of the  $A_T^\circ$  and  $\Delta C_{dis}$  terms since the pre-industrial era. The subsurface layer reference for water mass formation conditions produced parameterizations of  $A_T^\circ$  and  $\Delta C_{dis}$  that serve to estimate  $C_{ant}$  without the need of any additional zero- $C_{ant}$  references. A random propagation of the errors associated with the input variables necessary to calculate  $C_{ant}$  according to the  $\varphi C_T^\circ$  formulation yielded an overall uncertainty of  $\pm 5.2 \mu\text{mol}\cdot\text{kg}^{-1}$ . This way of calculating uncertainties has been successfully used in the past by many authors (Gruber et al., 1996; Lee et al., 2003; Lo Monaco et al., 2005). The work from Vázquez-Rodríguez et al. (2009b) compared five independent estimation methodologies of  $C_{ant}$  in the Atlantic Ocean. According to this study, the  $\varphi C_T^\circ$  approach consistently yielded the closest values to the average of all five  $C_{ant}$  methodologies over the whole latitudinal range of the Atlantic. Appendix II discusses further the choice of the  $\varphi C_T^\circ$  method with respect to other methodologies, and a comparison of results is made with the TrOCA approach (Touratier et al., 2007).”*

15. The following 7 pages (to 178) must be revised for clarity and brevity, as I have required in part (b) of my first comment.

REPLY:

Done, as explained in the answer to your comment 1b. Please, refer to new section 3 and Appendix I in the revised manuscript version.

16. Eq (1) has density in it, but not eq 6 and 10.

REPLY:

Thank you for noticing that the “density” term was missing. Please, note that old equations 6 and 10 are now equations 1 and 2, respectively.

17. Page 173-174 The approach assumes that the ratio of change in layer thickness (F from Eq. (2)) is constant over the whole basin. How valid is this assumption? This must be evaluated. An initial trial can be carried out by looking at the  $F_{b,l,c}$  term. It should be same at every station on a given cruise. If not, then I question the validity of the approach.

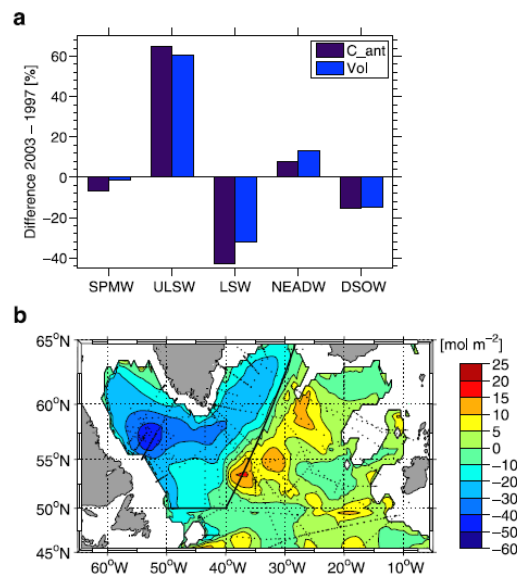
REPLY:

We understand that this is indeed a strong assumption made in our computations, and that you might question it. The introduction of the factor  $F_{b,l,c}$  (old Eq. (2), new Eq.

(A1) in Appendix I) was justified for the Irminger and Iceland basins because these are regions where strong convection occurs and large thickness variations are expected over time. As a matter of fact, the  $F_{b,l,c}$  factor was not applied in the ENA basin ( $F_{ENA,l,c}=1$ ) because water mass formation is much less important in this region, and also because of its larger extension and the sparseness of measurements here (see modified paragraph of the manuscript included below, introduced after new Eq. (1)).

“...Equation (1) has been applied in this study to calculate the inventories of  $C_{ant}$  in the Irminger and Iceland basins (“ $b=Irm$ ” and “ $b=Ice$ ”). The procedure to obtain more accurate inventory estimates for the ENA basin is slightly different (Equation (2)). The weaker convection in this region makes layer thickness variability less important compared to the Irminger and Iceland basins. Also, due to its larger extension and the sparseness of measurements, the calculation of  $Th_{b,l,c}^*$  in the ENA basin is not applied, i.e., in this case  $Th_{ENA,l,c} = Th_{ENA,l}^{WOA05}$ .”

In the following we will provide evidence and arguments that support and justify our assumption. On the one hand, the work from Steinfeldt et al. (2009) shows the importance of the LSW contribution to the Atlantic inventory of  $C_{ant}$  and, most outstandingly, how the fluctuations of LSW volume affects the  $C_{ant}$  column inventory (see Fig. 9 from Steinfeldt et al. (2009) included below). In sum, the variation of LSW thickness affects considerably the Atlantic inventory. Thus, accounting for these thickness variations as accurately as possible is vital to make good inventory estimates, and this is a clear aim in our study.



**Figure 9.** (a) Relative changes of volume and  $C_{ant}$  inventory (all values time-corrected toward 2003) between 1997 and 2003 within the northwest Atlantic. In the case of a steady state ocean, both differences should be zero. (b) Changes of  $C_{ant}$  column inventory (2003–1997; the 1997 values are time-corrected toward 2003) for LSW. The northwest Atlantic region is limited by the bold black line.

In addition, this evidence of high LSW thickness variability makes inappropriate the classical transient steady state assumption (TSS, Keeling and Bolin, 1967) assumed in



earlier works that estimate  $C_{ant}$  or CFC inventories (Holfort et al., 1998; Roson et al., 2003; Álvarez et al., 2004; Tanhua et al., 2006).

Alternatively, Kieke et al. (2006) provide solid evidences for the temporal variability of LSW thickness (see their Fig. 12 included below) that is consistent with our results shown in Fig. 4 for the layer thickness variation of the  $u$ LSW and  $c$ LSW in the Irminger and Iceland basins. It must be noticed that our Fig. 4 also gives an idea of the associated uncertainty in layer thickness computations, which is around 10% (equivalent to 100-200 m). These uncertainties were later taken into account when calculating the final uncertainties of  $C_{ant}$  storage rates (see answer to comment 30). The STDs of the “WOA-along-cruise-track” and “observed” thickness values ( $Th_{b,l}^{WOA05}$  and  $Th_{b,l,c}^{obs}$ , respectively) were used to calculate the uncertainties of the  $F_{b,l,c}$  factors in the Irminger and Iceland basins, which are now provided in Tables 2a and 2b.

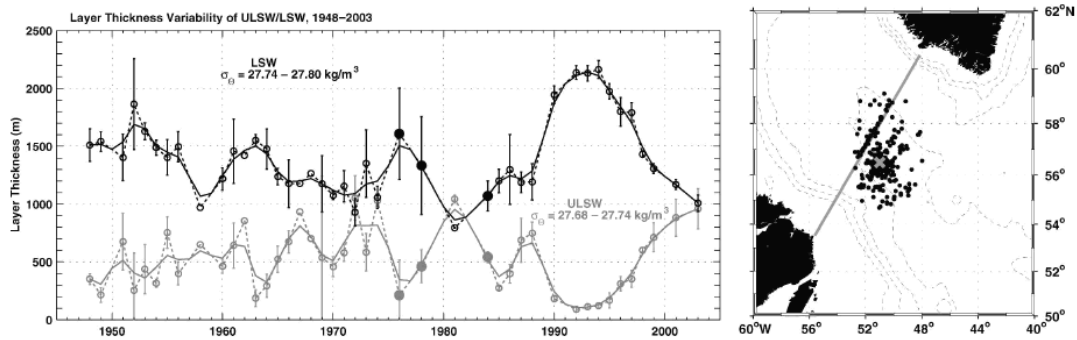
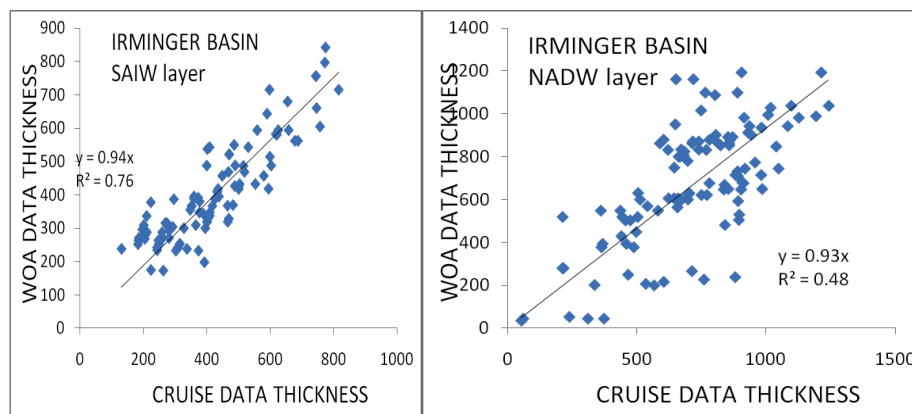
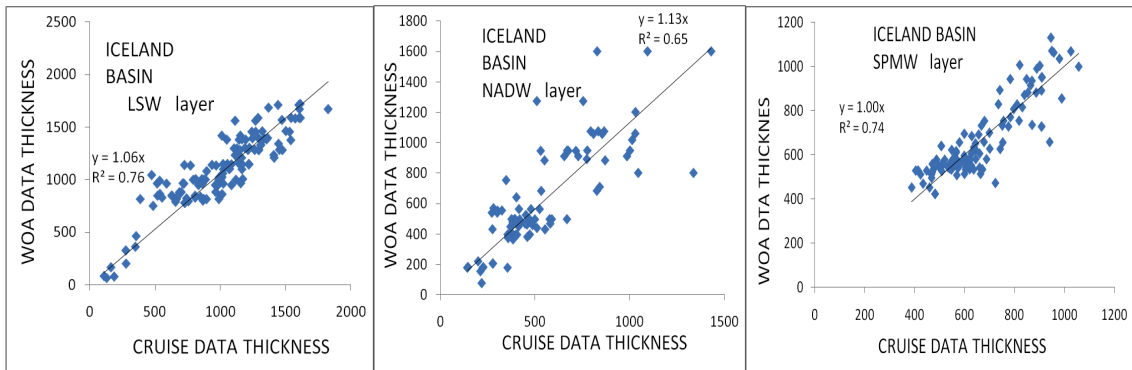


FIG. 12. (left) Time series of layer thickness (m) for classical (black curve) and ULSW (gray curve), given for the period 1948–2003. Open circles indicate the individual mean estimate derived from summer data with vertical bars as the corresponding standard deviation. Filled circles denote estimates solely based on winter data. A 3-yr filter is applied to smooth the time series (solid lines). (right) Location of included Labrador Sea profiles (black dots). The gray line marks the course of WOCE line AR7W; the gray cross is the position of OWS Bravo.

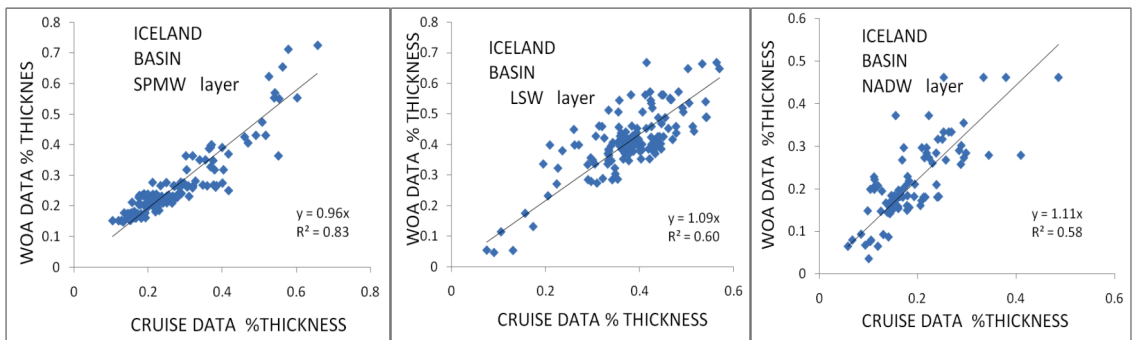
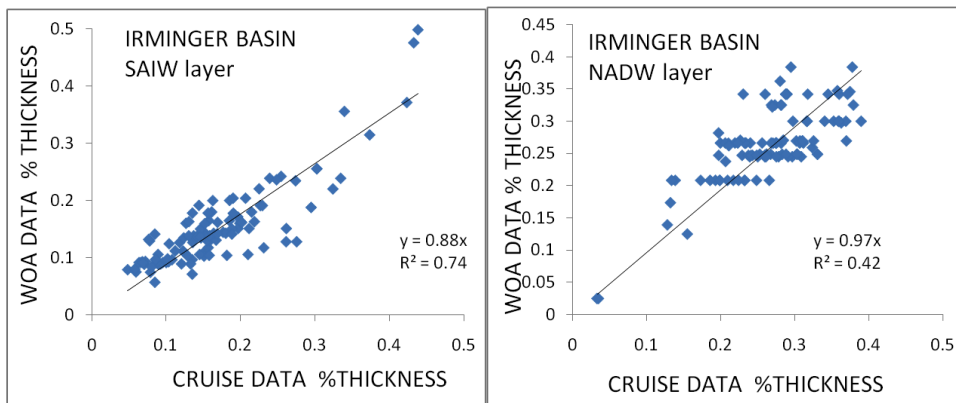
Regarding your request of evaluating how valid was the assumption of a constant  $F_{b,l,c}$  in the Irminger and Iceland basins for a given layer and cruise/year, we provide here the following graphs:

A)  $Th_{b,l,c}^{obs}$  vs  $Th_{b,l,c}^{WOA05}$





B)  $\%Th_{b,l,c}^{obs}$  vs  $\%Th_{b,l,c}^{WOA05}$  (refer to the  $F_{b,l,c}$  equation -A1- in Appendix I)



There are two reasons that explain why  $F_{b,l,c}$  values are not always identical at every station on a given cruise and layer. In first place, there is a high short-scale spatial variability linked with the variability of the mesoscale field (Rodgers et al. 2009), as shown by the first set of five plots “Th WOA05 vs Th cruise”. In addition, the WOA05 gridded fields have been largely smoothed and have less spatial resolution ( $1^\circ \times 1^\circ$ , i.e., each of WOA05’s pixels may include more than one station from the same cruise) than the observations from the cruises. In spite of everything, the above graphs

show that there is a reasonably good correspondence between the thickness estimates obtained from cruise data and WOA05 data (slopes range between 0.93 and 1.13), same as for the case of the set “B” of graphs with the  $\%Th_{b,l,c}^{obs}$  and  $\%Th_{b,l,c}^{WOA05}$  terms.

Even though the procedure introduced in our manuscript to calculate  $C_{ant}$  inventories has certain caveats associated, it is still a more solid and congruent approach than the one previously used that assumed constant mean penetration depths (MPDs) for a whole basin, based on the classical TSS concept from Keeling and Bolin (1967).

Kieke, D., M. Rhein, L. Stramma, W.M. Smethie, D.A. LeBel, W. Zenk, Changes in the CFC inventories and formation rates of Upper Labrador Sea Water, 1997–2001, *J. Phys. Oceanogr.*, 36, 64–86, 2006.

Rodgers, K. B., R. M. Key, A. Gnanadesikan, J. L. Sarmiento, O. Aumont, L. Bopp, S. C. Doney, J. P. Dunne, D. M. Glover, A. Ishida, M. Ishii, A. R. Jacobson, C. Lo Monaco, E. Maier-Reimer, H. Mercier, N. Metzl, F. Perez, A. F. Rios, R. Wanninkhof, P. Wetzel, C. D. Winn, and Y. Yamanaka: Altimetry helps to explain patchy changes in hydrographic carbon measurements. *J. Geophys. Res. Oceans*, 114, C09013, doi:10.1029/2008JC005183, 2009.

Steinfeldt, R., M. Rhein, J. L. Bullister, and T. Tanhua, Inventory changes in anthropogenic carbon from 1997–2003 in the Atlantic Ocean between 20°S and 65°N, *Global Biogeochem. Cycles*, 23, GB3010, doi:10.1029/2008GB003311, 2009.

18. Page 176, line 10 to page 178, line 10 These two pages describes the approach that is used to correct for any bias that may arise following sparse spatial sampling coverage in the ENA basin, i.e that the cruise data may not be representative of the average conditions in the ENA basin at the time the cruise was carried out. It uses the cruise data to determine the MLR fit between  $C_{ant}$  and (AOU, theta, and S). This MLR fit is then applied WOA and cruise data at the “same locations of the considered cruise track” to calculate the corrections, eq. (8). I do not understand how this can correct for biases in average values that arise from too poor sampling coverage, I mean – the equation is applied to data from the cruise track only, and regardless whether it is WOA or cruise data, spatial biasing may occur. This needs to be fixed. If I misunderstand, then the section must be revised for meaning and clarity.

REPLY:

We apologize for the awkwardness. As mentioned in earlier answers (comment 1b) section 3 has been revised and shortened for clarity. Perhaps we went into excessive detail and ended up complicating a calculation that is meant as a minor correction only. The details are now clearer in Appendix I. You are right in the fact that the equation was applied to cruise data only, but the equation coefficients were obtained using average climatological data as a reference against which “deviations” can be assessed. As it is stated in section B of Appendix I:

“...The “ $\Delta C_{ant}^{ENA,l,c}$ ” terms are computed using an extrapolation method based on covariations with WOA05 properties. These small “ $\Delta C_{ant}^{ENA,l,c}$ ” biases are expectable because, for each layer, a spatial gradient in  $C_{ant}$  exists in the ENA due to the different ventilation stages and rates of each water mass. As a matter of fact, the AOU in the ENA basin displays a positive southward gradient for all layers. Perez et al. (2008) found for the Irminger

*basin a clear relationship between AOU (a proxy for ventilation) and  $C_{ant}$  saturation for different water masses. The " $\Delta C_{ant}^{ENA,I,c}$ " terms were computed from cruise data and expressed as individual correction elements for each cruise and layer in the ENA basin."*

Section 3 now focuses more on presenting the line of thought and the necessary evidence to support it. We hope the ENA correction is now more straightforward to follow in the revised version (please, refer to new Section 3 and Appendix I).

19. Page 180, line 7-20. This section discusses the extent to which different cruise tracks has had an influence on T, S, AOU and silicate. Why is this effect not discussed for anthropogenic CO<sub>2</sub> at all? It might be important. It must be dealt with.

REPLY:

Thank you for this suggestion. The revised and shortened version (after your comment 1c) focuses more on how the described T, S and AOU variability may affect the  $C_{ant}$  distributions obtained and thus the storage rates. Also, since some of the description of the measured properties was already provided in detail in the previous work from Pérez et al. (2008), we have avoided repetitions and left only the very specific and new features in this paragraph.

20. Page 181, line 9, the statement on reduced convection must be referenced. In fact, when I think of it, I do not think that anybody has seen reduced overflow from the Nordic Seas in the recent years (and so this is relevant for my comment 7), see Steffen et al, Nature, 2008.

REPLY:

The statement is now referenced:

*"When the strong convection period relaxed afterwards (Lab Sea Group, 1998), this trend of  $C_{ant}$  increase also weakened and yielded a noisier pattern in  $C_{ant}$  increase tendencies. The effect of weaker convection on LSW propagates deep in the water column and it ca be expected to affect NADW (Yashayaev et al., 2008)."*

Since the mentioned reduction on convection also affected LSW it can be expected that NADW will sense some of this alteration (Yashayaev et al., 2008).

Lab Sea Group. The Labrador Sea Deep Convection Experiment. Bulletin of the American Meteorological Society 79(10) 2033-2058, 1998.

Yashayaev I., Holliday, N. P., Bersch, M., van Aken, H.M., The history of the Labrador Sea Water: Production, Spreading, Transformation and Loss. In "Arctic-Subarctic Ocean Fluxes: defining the role of the Northern Seas in climate", Robert R. Dickson, J. Meincke, P. Rhines. Springer, P.O. Box 17, 3300 AA Dordrecht, The Netherlands, pp. 569-612, 2008.

22. Page 182, line 9-15. Is the concentration of  $C_{ant}$  in the uNADW the same as in the INADW? I would expect otherwise, since uNADW is younger (as reflected in AOU). If it is the same, please explain why.

REPLY:

What is said is that the tendencies/patterns of the average  $C_{\text{ant}}$  concentrations are similar, not the actual average values of  $C_{\text{ant}}$  concentration for 1981-2006, which are  $9.6 \pm 1.1$  and  $5.1 \pm 1.0 \mu\text{mol kg}^{-1}$  (from Table 2c) for the uNADW and INADW, respectively. These data and clarification have been added in the revised manuscript:

*“The temporal trends of the average  $C_{\text{ant}}$  concentrations in the deep waters of the ENA basin (uNADW and INADW) are very similar. As expected from their location in the water column, far from upper layer influences, their  $C_{\text{ant}}$  concentrations are the lowest ones found in the study area. Actually, no significant trends of  $C_{\text{ant}}$  increase are detected. However, the average concentrations of  $C_{\text{ant}}$  in these two layers for the 1981-2006 period (Table 2c) are somewhat different:  $9.6 \pm 1.1$  and  $5.1 \pm 1.0 \mu\text{mol kg}^{-1}$  for the uNADW and INADW, respectively. The warm component of NADW (uNADW) is less influenced by AABW than the cold component (INADW), as reflected by the low  $\text{Si}(\text{OH})_4$  values of the former compared with those of the latter. Also, the higher influence of LSW/ISOW in the uNADW is revealed by its imprint in the AOU and  $\text{Si}(\text{OH})_4$  values, which are lower than the observed in the INADW layer.”*

23. Page 182, lines 15-18. Any method would give lowest concentrations of  $C_{\text{ant}}$  in these watermasses, therefore it is not valid to use this as a support for the Vázquez-Rodríguez method.

REPLY:

OK. We have deleted this sentence. However, what we meant is that some methods like the  $\Delta C^*$  or TrOCA tend to give negative  $C_{\text{ant}}$  concentrations in very old waters like these ones (Vázquez-Rodríguez et al., 2009), and the fact that a method does not give negative values but very low values is a positive feature. Anyhow, this discussion was kind of off-topic so we are OK to remove it.

24. Page 183, line 1-2 Neither Corbiere nor Schuster attributed the reduced air-sea flux to increased stratification.

REPLY:

In the abstract from Schuster and Watson (2007) work clearly state that “...Declining rates of wintertime mixing and ventilation between surface and subsurface waters **due to increasing stratification**, linked to variation in the North Atlantic Oscillation, are suggested as the main cause of the change”

Schuster, U., and A. J. Watson, A variable and decreasing sink for atmospheric CO<sub>2</sub> in the North Atlantic, *J. Geophys. Res.*, 112, C11006, doi:10.1029/2006JC003941, 2007.

25, page 183, line 6. Le Quéré does not quantify how much the changes in the NA has contributed to the increase of atmospheric CO<sub>2</sub>, we have no idea whether it is significant or not.

REPLY:

OK. We have removed that one and the rest of references. The sentence now remains as:

*“The consequent convection weakening accompanied by a strong stratification are the main reasons for the overall decline of the northern North Atlantic CO<sub>2</sub> sink.”*

26. page 186, line 21-24. Corbiere, as well as Schuster et al, evaluated changed in air-sea CO<sub>2</sub> fluxes. This paper evaluates anthropogenic CO<sub>2</sub> storage. Since air-sea fluxes have a natural component as well, and since water moves around, transporting CO<sub>2</sub> as well, these are not the same things. It is therefore not correct to state that these results support each other or are in good agreement, since they are not comparable.

REPLY:

The concentration of  $C_{ant}$  in the Irminger Sea changes over time due to the atmospheric xCO<sub>2</sub> increase that, once it enters into the water column, is later transported into the ocean interior thanks to the deep-convection processes. Thus, the strength of such convection events in the Irminger is also a determining factor for the  $C_{ant}$  that ultimately goes into these particular waters. Pérez et al. (2008) showed that in the Irminger basin the % of the  $C_{ant}$  saturation concentration varies (it is actually inversely correlated) with AOU, which is a proxy for ventilation (NB: using % $C_{ant}$  sat. “removes” the contribution of the temporal atm. xCO<sub>2</sub> increase from this relationship, since the % saturation concentration is always relative to the corresponding atmospheric xCO<sub>2</sub>, i.e., the relationship % $C_{ant}$  sat-AOU establishes the direct dependence between  $C_{ant}$  content and convection). Since AOU is controlled by the natural cycles of ventilation plus the remineralization of organic matter, and it is not affected by the anthropogenic effect (it assumes 100% saturation of oxygen at the air-sea interface), this means that in the subpolar gyre the natural cycles and the entrainment of the anthropogenic signal are directly linked. This relationship is driven by the interannual variability of winter convection. The authors of the present manuscript therefore maintain that the changes observed in the uptake of natural and anthropogenic CO<sub>2</sub> in this region are indeed linked and that, consequently, the observed decrease in air-sea CO<sub>2</sub> exchange over the last decade (Omar and Olsen, 2006; Corbiere et al., 2007; Schuster et al., 2007) runs parallel to the weakening of  $C_{ant}$  storage in the NASPG. As discussed in previous answers, this weakening stems from the NAO-driven changes in stratification and convection.

The sentence has now been modified and reads as follows:

*“The changes in  $C_{ant}$  storage rates here obtained are consistent with the results in Omar and Olsen (2006), Corbière et al. (2007) and Schuster and Watson (2007), who found analogous decreasing rates in the air-sea CO<sub>2</sub> exchanges from surface fCO<sub>2</sub> measurements in the North Atlantic that, overall, contribute to the decrease of  $C_{ant}$  storage rates in the NASPG. Such air-sea CO<sub>2</sub> exchange results can be legitimately compared to the ones here obtained for  $C_{ant}$  storage rates since, according to Pérez et al. (2008), the cycles and uptake of natural and anthropogenic CO<sub>2</sub> in the NASPG are linked. Consequently, the observed decrease in air-sea CO<sub>2</sub> exchange over the last decade (Omar and Olsen, 2006; Corbière et al., 2007; Schuster et al., 2007) must have occurred simultaneously (and most probably linked) to the weakening of  $C_{ant}$  storage in the NASPG that, as shown here, stems from NAO-driven changes of stratification and convection intensity.”*

27. Table 2. Include basin name in each table header.

REPLY:

Done. Please, note that Tables 2 have been revised and extended with new columns after your comment 17.

28. Include a figure with at least the layer thicknesses over the years.

REPLY:

The requested figure already existed in the previous version of the ms (Fig. 4). After revising Section 4 and reorganising the numbering, the figure moved to Fig. 5 in the revised ms.

29. Quantify the relative contribution of layer thickness and  $C_{ant}$  changes on the inventory trends.

REPLY:

If the thickness corrections we have applied and developed in section 3 are considered when calculating  $C_{ant}$  inventories (and storage rates) then we obtain the results given in Table 3 (re-numbered in revised manuscript), whereas if this correction is not done (i.e., only  $C_{ant}$  changes contribute to the differences in the storage rates), then we obtain the results in Table 3.2 (only included below).

When the thickness variability is not taken into account in calculations the differences between high-NAO and low-NAO  $C_{ant}$  storage rates in the OVIDE box reduce by about 40%, i.e., from 0.028 Gt C yr<sup>-1</sup> in Table 3 to 0.017 Gt C yr<sup>-1</sup> in Table 3.2. This result is something similar to what was found by Steinfeldt et al. (2009). This evaluation has been included in the revised manuscript, towards the end of section 4:

*“...Additionally, an assessment was performed of the impact or relative contribution of considering the temporal variability of layer thickness (section 3 and Appendix I) together with  $C_{ant}$  changes on the obtained inventory trends from Table 3. When the thickness variability is not taken into account in calculations then the differences between high-NAO and low-NAO  $C_{ant}$  storage rates in the NASPG reduce by about 40%: from 0.028 Gt C yr<sup>-1</sup> (Table 3) to 0.017 Gt C yr<sup>-1</sup>. This result is consistent with what was found by Steinfeldt et al. (2009). In their Fig. 9 they showed how the fluctuations of, particularly, LSW volume affects the  $C_{ant}$  column inventory.”*

**Table 3 (computed considering the effect of thickness variation on  $C_{ant}$  inventory)**

Basin	NAO Phase (time period)	$C_{\text{ant}}$ Specific Inventory Rates (mol C m <sup>-2</sup> yr <sup>-1</sup> )	Storage Rate (Gt yr <sup>-1</sup> )
<b>IRMINGER</b>	High (1991-1997)	1.74±0.18	0.013±0.002
	Low (1997-2006)	0.4±0.3	0.006±0.002
<b>ICELAND</b>	High (1991-1998)	1.88±0.45	0.022±0.005
	Low (1997-2006)	0.3±0.2	0.0035±0.003
<b>ENA</b>	(1981-2006)	0.72±0.07	0.019±0.002
<b>NASPG</b> (OVIDE box)	High NAO	1.18±0.12	0.054±0.006
	Low NAO	0.56±0.08	0.026±0.004

Table 3.2 (computed without considering the effect of thickness variation on  $C_{\text{ant}}$  inventory)

Basin	NAO Phase (time period)	$C_{\text{ant}}$ Specific Inventory Rates (mol C m <sup>-2</sup> yr <sup>-1</sup> )	Storage Rate (Gt yr <sup>-1</sup> )
<b>IRMINGER</b>	High (1991-1997)	1.69±0.20	0.012±0.0015
	Low (1997-2006)	0.72±0.2	0.005±0.0015
<b>ICELAND</b>	High (1991-1998)	1.91±0.2	0.022±0.002
	Low (1997-2006)	1.13±0.2	0.013±0.002
<b>ENA</b>	(1981-2006)	0.72±0.07	0.019±0.002
<b>NASPG</b> (OVIDE box)	High NAO	1.18±0.12	0.054±0.003
	Low NAO	0.56±0.08	0.037±0.003

Steinfeldt, R., M. Rhein, J. L. Bullister, and T. Tanhua, Inventory changes in anthropogenic carbon from 1997–2003 in the Atlantic Ocean between 20°S and 65°N, *Global Biogeochem. Cycles*, 23, GB3010, doi:10.1029/2008GB003311, 2009.

30. My final comment is on the uncertainties, which are not dealt with at all. There are many sources, for example (1) uncertainty of anthropogenic CO<sub>2</sub> estimates (2) from spatial biasing of average values (see comment 19), and (3) the assumption of a constant  $F_{b,l,C}$  (comment 17), and measurement errors. How significant are the trends after the effect of these uncertainties have been taken into account? Please identify, quantify and propagate all errors you can think of, and evaluate the significance of the trends in light of these.

REPLY:

There are two major sources of uncertainties in the calculation of  $C_{\text{ant}}$  inventories (and thus, storage rates) in our work: a) The uncertainties associated with the  $C_{\text{ant}}$  estimation



method; b) The uncertainties associated with the calculation of layer thickness.

The table given below includes the specific inventory rates of  $C_{\text{ant}}$  ( $\text{mol C m}^{-2} \text{ yr}^{-1}$ )  $\pm$  the standard errors of the estimate calculated in three additional ways to how it was done to calculate the std. errs. given in Table 3 (single linear regression):

(1) Column “Perturbation  $2\sigma C_{\text{ant}}$  method”: The standard errors of the  $C_{\text{ant}}$  estimates obtained for each cruise and layer (given in Tables 2a,b,c) were randomly propagated 100 times. Afterwards, the  $\text{AVG}\pm\text{STD}$  of all 100 slopes was calculated. This is the value shown in column 4 of the table below. (NB: The uncertainty in  $C_{\text{ant}}$  estimation when applying the  $\varphi C_T^0$  method is  $\pm 5.2 \mu\text{mol kg}^{-1}$  –see reply to comment 14).

(2) Column “Perturbation  $2\sigma \text{Th}$  factor”: The standard errors of the  $F_{\text{b,l,c}}$  estimates obtained for each cruise and layer (given in Tables 2a and 2b) in the Irminger and Iceland basins were randomly propagated 100 times. In the case of the ENA basin, the Th calculated from WOA05 data is applied ( $F_{\text{ENA,l,c}} = 1$ ), so a constant error of 5% is assumed. Afterwards, the  $\text{AVG}\pm\text{STD}$  of all 100 slopes was calculated. This is the value shown in column 5 of the table below.

(3) Column “Perturbation both  $C_{\text{ant}}$  & Th”: Both sources of error in the previous two columns are combined and then randomly propagated 100 times to get, again, the  $\text{AVG}\pm\text{STD}$  of all 100 slopes. This is the value shown in column 6 of the table below.

**Specific Inventory Rates ( $\text{mol C m}^{-2} \text{ yr}^{-1}$ ): Slopes  $\pm$  Std. errs.**

Basin	NAO Phase	Linear regression (as in Table 3)	Perturbation ( $2\sigma C_{\text{ant}}$ method)	Perturbation ( $2\sigma \text{Th}$ factor)	Perturbation ( $C_{\text{ant}}$ & Th)
<b>IRMINGER</b>	High (1991-1997)	1.74 $\pm$ 0.24	1.74 $\pm$ 0.15	1.73 $\pm$ 0.10	1.72 $\pm$ 0.19
	Low (1997-2006)	0.4 $\pm$ 0.3	0.43 $\pm$ 0.11	0.43 $\pm$ 0.05	0.43 $\pm$ 0.12
<b>ICELAND</b>	High (1991-1998)	1.88 $\pm$ 0.45	1.85 $\pm$ 0.19	1.85 $\pm$ 0.16	1.85 $\pm$ 0.26
	Low (1997-2006)	0.3 $\pm$ 0.2	0.34 $\pm$ 0.12	0.34 $\pm$ 0.11	0.34 $\pm$ 0.17
<b>ENA</b>	(1981-2006)	0.72 $\pm$ 0.03	0.76 $\pm$ 0.05	0.76 $\pm$ 0.06	0.76 $\pm$ 0.07

In the case of the ENA basin, the std. err. in column 6 ( $\pm 0.07$ ) is larger than the one previously shown in table 4 (column 3, i.e.,  $\pm 0.03$ ). Thus, Table 3 has been updated with the largest std.err. value for the ENA: 0.72 $\pm$ 0.07  $\text{mol C m}^{-2} \text{ yr}^{-1}$ . The opposite occurred in the cases of the Irminger and Iceland basins (std.err. in column 3 is larger than that in column 6), so the same std.err. values have remained in the updated Table 3. This way, we are always providing the upper limits of our uncertainty sources.

This evaluation of uncertainties has been included in the newly added Appendix II.