Review 2

Review of the manuscript egusphere-2024-1388 «Ocean Acidification trends and Carbonate System dynamics in the North Atlantic Subpolar Gyre during 2009-2019» by Curbelo-Hernández et al. submitted for discussion in Biogeosciences

General comment :

This manuscript reports a detailed analysis of carbonate chemistry parameters measured along the entire water column of a transect in the North Atlantic South Polar Gyre during 8 oceanographic cruises conducted between 2009 and 2019. The manuscript gives a nice description of this remarkable dataset. The dataset is used to study the trends of ocean acidification in this oceanic area with a particular focus on the spatial variability between three defined oceanic provinces (The Irminger Basin, the Iceland Basin and the Rockall Trough) and the different water layers encountered in the water column. Due to its importance as an oceanic carbon sink, the north Atlantic basin has been very intensively studied in the last decades. However this study is a significant contribution for (at least) three reasons : (1) it presents a new dataset, (2) it focuses on the last decade (2009-2019) and it (3) shows some original results in the eastern part of the basin. The authors have made a detailed literature review in order to put this study in context. The measurements and calculation methods are carefully described. The figures and tables are of good quality. The manuscript is well written and the results are convincing. However the manuscript is very long. This is certainly due to the fact that it is at the same time a "data paper" and a "scientific paper". My point is not to say that it should be split in two (I appreciate having all the information in one manuscript) but I believe that it could be more synthetic in some parts. I would be glad to support the publication of this manuscript after some revisions (which I believe to be minor). My major concerns (detailed in the following sections) are related to the structure of the manuscript and some methodological points.

Thank you very much for your thoughtful and constructive feedback on our manuscript. We greatly appreciate your recognition of the significance of our study, particularly in presenting a new dataset, focusing on the most recent decade, and revealing original results in the eastern North Atlantic basin. We acknowledge your point regarding the manuscript's length and the balance between being both a "data paper" and a "scientific paper." We have carefully considered your suggestions, which have contributed to enhancing the quality and reliability of the manuscript. Below, we provide a point-by-point response to each of your comments.

Specific comments :

Section 2.2.5 and 2.2.6: It's the same method that is used to deconvolute the drivers of the pH and omega trends. This could go into one section called "deconvolution of the trends". The beginning of section 2.2.6 on the calculation of the omega values could be added to section 2.2.2 "computational methods".

This restructuring has substantially improved the organization and readability of the methodology in the updated version of the manuscript.

On section 2.2 « Data Processing » : I am surprised about the fact that a huge amount of work has been done to compare the measured variable to variable estimated with CANYON-B but I haven't found a simple "internal consistency test" of the three measured variables of the carbonate system. Following the table S2, a table could give some basic statistics about the difference "pHT measured – pHT calculated from AT/CT

“, “AT measured – AT calculated from pHT/CT” or “CT measured – CT calculated from AT/pHT” in the cases where all three variable were measured.

We conducted an extensive and detailed evaluation of the internal consistency of our observations. The measured variables were compared for each cruise with canyon-estimated and CO2sys-computed data, as you suggested. Initially, we included only comparisons with CANYON-B estimates in the table, but following your recommendation, we recognized the importance of also including comparisons with computed variables to further reinforce the reliability of our observations and results. We have now included these basic statistics on the differences between measured and computed pH, AT, and CT in Table S2. Additionally, we have added the following sentence in section 2.2.2: “An internal consistency test was conducted on the three measured MCS variables. The measured variables were compared with canyon-estimated and CO_{2SYS}-computed variables. The average differences and standard deviations were summarized in Table S2 and ensure the consistency of the observations”.

On section 2.2.4 « Water mass characterization » : This section presents mostly results about the different water masses encountered during the cruises. Most of this section should be added (maybe in a more synthetic way) into section 3.1 « Physicochemical characterization of the water column ».

The titles used for these two subsections may be confused and not fully reflect their content. The title of Section 2.2.4 was updated to “Hydrographic Characterization”. This subsection intend to identify the main basins and water masses across the NASPG. This subsection does not present results directly aligned with the research objectives of the article, but describes the principal water masses occupying the Irminger, Iceland, and Rockall basins. It also explains how were delineated the layers by potential density isopycnals following previous studies in the area. This delineation is crucial for the subsequent calculation of means and trends in each basin and water mass. Therefore, we believe this subsection would be more appropriately placed within the methodology section.

The title of subsection 3.1 was removed as detailed in the response to the following comment. The content of the updated section 3 differs from that of subsection 2.2.4, making their unification challenging. Section 3 presents the spatio-temporal distribution of physicochemical variables once the hydrographic characterization was performed, which will be discussed in detail in Section 4.

On section 3.2. “ Temporal evolution of the physicochemical properties”. There are no real results in this section, most of the information here is more relevant to the method “section”.

It is true. Our initial idea was to divide the Results section into two subsections: one focused on explaining the distribution of variables in the water column and another addressing temporal changes. The second subsection was intended to present the trends that would be discussed in depth in Section 4, but it included extensive information related to trend calculation and statistical analysis. We have considered your suggestions and moved this information to a new subsection in the Methodology titled "Data Adjustment for Trends Computation." Additionally, we have removed the titles of the two subsections in the Results section and unified them in the revised version of the manuscript.

On section 4.2 and Appendix B: I don't really understand the reasons to give the information of the CT and AT trends in appendix. I would suggest putting this information directly in the main text.

Initially, this information from Appendix B was incorporated into Section 4.2. To synthetize this section and reduce the overall text length and considering its lesser relevance to the main results discussed in this subsection and the central ideas being conveyed, these paragraphs were moved to Appendix B and considered supplementary to the discussion performed in section 4.2. Based on your feedback, we have decided to reintegrate the paragraph focused on CT and NCT trends into section 4.2, while trends in AT and NAT, which are complementary results that support the discussion but fall outside the article's primary objectives, will remain in the appendix.

Sections 4.3 "Acidification trends", section 4.4 "Drivers pH" and section 4.5 " Interannual changes in …" are very long. I really believe that these three sections could be synthesized into one single section. This is not an easy task but the drivers of the pH trends and the omega trends are mostly the same (I am of course aware that some differences exist) and could be treated in a more synthetic manner.

We have carefully considered this comment and discussed various possible changes to these three sections of the discussion. Our goal was to ensure that the key results and ideas are clearly conveyed while maintaining readability for the reader. We agree that this was not an easy task, as there was a risk of omitting relevant results and presenting the information in a disorganized manner. Additionally, these sections had already been revised according to suggestions from Reviewer 1.

Due to the various topics addressed in this part of the discussion, we believe that the best approach is to separate the text into subsections. Therefore, we have considered both the titles and content of these sections. In the revised manuscript, we have included the following subsections:

4.3. Acidification trends: this section has been condensed, incorporating some modifications based on the suggestions from Reviewer 1. We addressed here the pH_T trends encountered in the different layers and basins, which were compared to previous studies.

4.4. Interannual changes in Ω_{Ca} and Ω_{Arag}: We considered important to separate this section from the previous one, as it specifically addresses the effects of OA on individual organisms, ecosystems, and biological processes through the analysis of changes in Ω. In this subsection, we removed the analysis of the drivers of Ω_{Ca} and Ω_{Arag} that was included in the corresponding part of the text in the previous version of the manuscript.

4.5. Processes controlling OA and Ω trends: After analysing the interannual trends in pH_T (subsection 4.3) and Ω_{Ca} and Ω_{Arag} (subsection 4.4), we provided in this subsection a detailed analysis of the processes driving their trends quantifying its contributions. The combination of the analysis of drivers for pH_T and Ω, performed by following your suggestion, significantly aids in synthesizing this part of the discussion, making it more concise and easier to understand.

Conclusion: This is a really long conclusion! I would suggest just giving the main messages in conclusion and It could certainly stand in 20 to 30 lines.

We agree that the conclusion was somewhat lengthy, as also noted by Reviewer 1. To enhance readability and ensure that the key messages of this research are effectively

conveyed and better assimilated by readers, we have condensed the conclusion. We have done so by avoiding the inclusion of information already covered in the discussion section, in line with your recommendations and those provided by Reviewer 1.

The Conclusion was updated as follow:

“This research has evaluated the interannual changes in the basin-wide MCS dynamics along the NASPG during 2009-2019. Despite the observational period is relatively short to quantify long-term trends and to formulate significant future projections, the finding has allowed to evaluate the ocean response, in terms of MCS dynamics and on an interannual scale, to changes in deep-water convection and to isolate events affecting the physical patterns. The assessment of OA within the Irminger and Iceland basins was enhanced by supplying novel data and trends spanning a decade in which the physical patterns reversed. Additionally, the study provides an unprecedent analysis of the physico-chemical variations in the Rockall Trough, which is crucial for the assessment of the entire longitudinal span of the NASPG. It facilitates a more accurate understanding of the mechanisms dictating basin-scale acidification processes and advances our understanding of OA in the North Atlantic and Global Ocean.

Overall, the entrance and accumulation of Cant and interannual acidification trends were strongly affected by the cooling, freshening and enhancement in the oxygenation during this decade. The longitudinal span of the NASPG and the differences in circulation patterns, water masses and bathymetry behaved as a source of spatio-temporal variability. The interannual acidification trends of the main water masses across the NASPG ranged between 0.0006-0.0032 units yr-1 and caused a decline in the ΩCa and ΩArag of 0.004-0.021 and 0.003-0.013 units yr-1, respectively. The convective processes increased the accumulation rates of Cant in the interior ocean by 50-86% and accelerated the acidification rates by around 10% compared to previous decades in the Irminger and Iceland basins. The shallower hydrography of the Rockall Trough and the poleward circulation patterns accounted for differences in the acidification rates respect to surrounding waters.

The Cant-driven increase in NCT was found to govern the acidification of the NASPG with contributions exceeding 60%. The combined effect of the decreasing temperature, salinity and NAT neutralized close to one-half of the acidification along the entire longitudinal span of the SPMW. The enhanced deep-water ventilation in the western NASPG slowdown the cooling and freshening toward the interior ocean, weakening the physical counterbalance of acidification.

The present investigation emphasizes the progressively increase in the uptake and accumulation of Cant and subsequent acceleration of OA along the NASPG. Novel data and results provided could be compared with other repeated hydrographic section data at mid and high latitudes in the North Atlantic, such as the A02, A25, AR07E and AR28 framed in the GO-SHIP program, as well as used in conjunction to develop future investigations. Additionally, they contribute to the improvement of the projections pertaining to the future state of the oceans run by models and forecast. Considering the important variability in the mechanism controlling the distribution of the physico-biogeochemical properties and particularly the OA in the North Atlantic, this research aims to highlight the necessity of continue monitoring and sampling the whole water column through repeated hydrographic sections, especially through the highly variable but less assess easternmost part.”

Technical corrections

L54 : What is meant by « conservative scenario » ?

In this context, we refer to the IPCC's Representative Concentration Pathways (RCPs) scenarios (Van Vuuren et al., 2011; Moss et al., 2010), which project various future trajectories of greenhouse gas concentrations. By "conservative scenario," we intended to refer to the RCP2.6 scenario, which is based on lower future CO_2 emissions and predicts more moderate impacts compared to more aggressive emission pathways.

We have revised this part of the introduction and we now discuss the RCP2.6 scenario as the more optimistic projection and the RCP8.5 scenario as the more pessimistic one. We also cited IPCC AR5 and AR6 reports: "According to the IPCC's Representative Concentration Pathways (RCPs) scenarios (Van Vuuren et al., 2011; Moss et al., 2010), which project various future trajectories of greenhouse gas concentrations, the model projections estimate a potential pH decrease of 0.3–0.4 units by the end of the century under the RCP8.5 scenario, which assumes continued high CO_2 emissions. In contrast, the most conservative RCP2.6 scenario, which includes significant emission reductions, anticipates a pH drop of 0.2–0.3 units (IPCC 2013 and 2021)".

L55 : Please define what is meant by « Anthropogenic carbon » before using it for the first time.

We added the definition given by Sarmiento et al., 1992: "fraction of inorganic carbon resulted from human emissions".

Sarmiento, J. L., Orr, J. C., and Siegenthaler, U. (1992). A perturbation simulation of CO2 uptake in an ocean general circulation model. Journal of Geophysical Research: Oceans, 97(C3), 3621-3645.

L84 : Missing reference "Gonzalez-Davilla and Santana-Casiano 2023"

It was included in the reference list.

L133 : The concept of « hydrographic CLIVAR 59.5°N Section » is not clear. Is this an historical WOCE section ?

The hydrographic section is part of the World Climate Research Programme (WCRP) within the framework of the CLIVAR (Climate and Ocean: Variability, Predictability and Change) project. We updated the manuscript with the following statements: "Data were collected from eight summer cruises conducted along the transverse hydrographic section at 59.5ºN between 2009 and 2019 (Daniault et al., 2016; Gladyshev et al., 2016b, 2017, 2018; Sarafanov et al., 2018). This repeat section is part of the World Climate Research Programme (WCRP) within the framework of the CLIVAR (Climate and Ocean: Variability, Predictability and Change) project and covers the length of the Subpolar North Atlantic between Scotland and Greenland (4.5-43.0ºW), crossing the Irminger and Iceland basins and the Rockall Trough (Figure 1)"

L160 : What is meant by « in-situ calibrated » ?

We have removed "in situ" from this sentence for a better understanding. We wanted to say that the VINDTA was systematically on-board calibrated using CRMs.

L170-173 : This point is confusing. An uncertainty of 0.0047 pH Units does not necessarily correspond to a systematic bias. Please clarify this point?

We have reformulated this point. The uncertainty of 0.0047 pH units reported by DelValls and Dickson (1998) was identified as an average difference rather than a systematic bias.

This value was derived from comparing pH values calculated using the pK* values of m-cresol purple with those obtained from more accurate pH determinations performed by DelValls and Dickson (1998).

Following their recommendations, we applied the correction of +0.0047 units to our pH measurements to address this identified discrepancy and align the pH values with those determined from the more accurate pK* values. While the uncertainty itself does not directly indicate systematic bias, applying this correction helps ensure that our pH data are consistent with the updated pK* values and thus improves the accuracy of our measurements. We have included this adjustment to reflect the more reliable pK* values and maintain consistency with the literature recommendations. In addition, after applying this correction, we got pH values with a negligible difference (0.0002 units) compared to those estimated by CANYON-B (this point was added to subsection 2.2.1), which infers confidence to the correction applied.

L199 : CANYON-B is trained and validated using GLODAPv2 data. Argo profiles are just used for comparison but not for formal validation.

The statement was updated in the new version of the manuscript: "This neural network is trained on and validated against bottle data from GLODAPv2 and recent GO-SHIP profiles and compared with sensor data from Argo floats".

L372-373 : This sentence is confusing. This section is not devoted to evaluating the temporal changes between 2009 and 2016.

In the updated version of the manuscript, we revised the beginning of this paragraph to clarify our intent. Our goal was to highlight that Figures 2, 3, S2, and S3 allow for a comparison of the surface-to-bottom distribution of variables between the 2009 cruise (left panels) and the 2016 cruise (right panels). After revision, we simplified this idea to: "The vertical distribution of the physical and biogeochemical variables is depicted for the cruises of 2009 and 2016 in Figures 2, 3, S2 and S3".

L550 – 553 : This sentence is not completely clear to me. Why is the limitation of the ventilation to the subsurface water related to an enhanced entrance of CANT through the air-sea interface ?

We noticed there was controversy in this part of the paragraph as the enhanced ventilation not necessarily means an increase in the invasion of CO_2 from the atmosphere which rise the C_{ant} in the subsurface waters. Considering the path of the ENACW from the south and subsequent warming of this layer at subpolar latitude, the high C_{ant} encountered in subsurface waters may be more closely related to the northward transport of C_{ant}-rich water than to the invasion of atmospheric CO_2 (as surface warming reduced its solubility). We modified this part of the paragraph as follow:

"The enhanced oxygenation of the ENACW (AOU <20 µmol kg-1 and reaching the oxygen saturation after 2014) was related with its high rates of renovation due to its path from the south (Pollard et al., 1996) and its mixing with waters moving eastward (Ellett et al., 1986). This favoured the transport subsurface waters with relatively high C_{ant} content from lower latitudes into the Rockall Trough and introduced wide differences respect to adjacent deeper layers moved from the western NASPG which strength the stratification."

L651 : The name of this section « Drivers pH » should be rephrased

The previous section titled “Drivers pH” was modified by including the results from the decomposition of both pHT and Ω trends as explained above. The new title of this section (4.5) is “Processes controlling pHT and Ω trends”.

L674-676 : This sentence is repeated in the conclusion. It could be removed here.

The entire conclusion was modified