

## Reviewer 1

Review of the paper: egusphere-2024-1388: Ocean Acidification trends and Carbonate System dynamics in the North Atlantic Subpolar Gyre during 2009-2019, by David Curbelo-Hernández et al.

### General Comment:

Quantifying and understanding the variations of anthropogenic CO<sub>2</sub> (Cant) in the oceans is important not only to reduce the uncertainty in the present Cant inventories but also to better predict the climate/carbon coupling, i.e. how the ocean will capture atmospheric CO<sub>2</sub> in the future. In addition the increase of Cant leads to ocean acidification and potential damage for marine species. The North Atlantic Ocean is an important CO<sub>2</sub> sink (Takahashi et al. 2009) and this region contains high concentrations of anthropogenic CO<sub>2</sub> (Cant) in the water column (Khatiwala et al 2013). Decadal variations of the Cant inventories were recently identified at basin scale probably linked to the change of the Atlantic Meridional Overturning Circulation (AMOC) (Gruber et al, 2019; Müller et al, 2023; Perez et al, 2024). It is worth noting that biases of AMOC in the GOBMs have been identified (Terhaar et al, 2024) that may explain differences of the Cant inventories between simulations and observations. Therefore, to better evaluate and correct current GOBMs comparisons with observed CT, AT and data-based Cant estimates are highly needed.

In this context David Curbelo-Hernández and co-authors present a detail analysis of the carbonate system (CS) changes based on 8 cruises conducted between 2009 and 2019 in the North Atlantic, here the NASPG region. After a nice introduction authors described in detail the dataset and the methods (measurements and calculations) used to investigate the temporal changes in the water column. I am convinced with all results that offer new views of the CS changes in the north Atlantic. The manuscript is clear, figures and tables adapted. The data presented here will be useful when revisiting the decadal changes of Cant inventories in the ocean (for RECCAP 3 ?), especially in the north Atlantic where decadal change of the Cant inventory could reached -1 PgC per decade (Müller et al, 2023). I support publication of this paper with minor revisions. Few comments and suggestions are listed below.

### Specific comments:

C-00: Title: “Ocean Acidification trends and Carbonate System dynamics in the North Atlantic Subpolar Gyre during 2009-2019”. As you investigate acidification trends in the water column (other studies analyzed this in the surface only, e.g. Lauvset et al, 2015; Leseurre et al, 2020) you may change the title: “Ocean Acidification trends and Carbonate System dynamics in the North Atlantic Subpolar Gyre water masses during 2009-2019”.

[The title will be updated in the new version of the manuscript.](#)

C-01: In the Introduction, maybe recall that acidification rates were also investigated from surface data (Lauvset et al 2015; Chau et al, 2024). For example, in the Irminger Sea, Chau et al (2024) evaluated trend over 1985-2021 for pH ( $-0.016 \pm 0.001$  per decade) and for War ( $-0.039 \pm 0.009$  per decade).

We have added at the beginning of the second-to-last paragraph the surface OA trend for the entire North Atlantic Subpolar biome (RECCAP biome) reported by Lauvset et al., 2015) and for the Irminger and Iceland basins recently reported by Chau et al., 2024.

C-02: Line 149: Authors write: “A detailed overview and metadata of the cruises is given in Table 1.”

I guess the metadata are not listed in Table 1: add a DOI or a reference for each cruise in Table 1?

The cruise reports are not published, thus cannot be referenced. However, we have completed the table with metadata information (number of sampling stations and measured variables during each cruise). A further description of the dataset is also provided in Zenodo (<https://zenodo.org/records/10276222>; DOI: 10.5281/zenodo.10276221).

C-03: Line 162: accuracy of  $\pm 1.5 \mu\text{mol kg}^{-1}$  for AT and  $\pm 1.0 \mu\text{mol kg}^{-1}$  for CT: this is very good. Impressive this is lower than the “climate goal” of  $\pm 2.0 \mu\text{mol kg}^{-1}$  (Newton et al, 2015).

That is what we get by using the full set of CRMs in each cruise.

C-04: Line 181: Are you sure that DO measurement is described in Dickson and Goyet (1994) ? Maybe refer to Dickson (1995).

Done in the updated version.

C-05: Line 382: Authors write: “The AT show a well-correlated direct relationship with salinity throughout the section ( $r^2=0.89$ ),...”. Curiosity: authors discussed significant changes in N-AT (Figure S4), is there a detectable difference of AT/S relationship over time ?

Considering the total amount of data collected from surface-to-bottom along the entire section, there is a linear relationship between  $A_T$  and Salinity with high degree of correlation ( $r^2 = 0.90$  and  $p\text{-value} < 0.01$ ). In the updated version of the manuscript, we provide the linear equation [ $A_T = 54.57 (\pm 0.36) \text{ Salinity} + 396.7 (\pm 12.7)$ ], that can be used to estimate the  $A_T$  content along the NASPG based on salinity data with a standard error of estimate of  $2.9 \mu\text{mol kg}^{-1}$  ( $< 0.1\%$ ).

Analysing fluctuations in the  $A_T/S$  relationship can greatly contribute to understanding the observed interannual changes in  $A_T$ ,  $NA_T$ , and  $S$ . To achieve this, we examined the linear relationship between  $A_T$  and  $S$  for each year of observation and plotted the slope values along with their respective standard deviations (Figure below). The linear regression indicates an increase in the  $A_T/S$  ratio with a statistical confidence level higher than 99%.

The interannual increase in the  $A_T/S$  ratio was related to the freshening (Figure 4) and the progressive increase of  $A_T$ -rich water inflows through upper layers (observed in the positive trends of  $NA_T$  in SPMW and ENACW; Figure S4). This was likely associated with the stagnation of  $A_T$ -rich subtropical waters in the upper layers due to the slowdown of the NASPG since the mid-90s (e.g., Böning et al., 2006; Häkkinen and Rhines, 2004), along with changes in the spreading of waters from higher latitudes influenced by melting.

The  $A_T/S$  ratio for the year 2012 was considered an outlier and excluded from the linear fitting. The high  $A_T/S$  value in 2012 compared to adjacent years was explained by a combination of factors: (1) The enhanced deepening of the MLD during winter in the Irminger Sea, which promoted intense ventilation confined to the surface layer (observed in low AOU, Figure S6) and increased ocean heat loss (observed in a lower temperature signal, Figure 4), as reported by Frob et al., 2016. (2) The eastward transport likely contributed to the depletion of  $A_T$  and  $NA_T$  at intermediate and deeper layers in the Irminger Basin while increasing them in the Iceland and Rockall Basins. (3) Fluctuations in the spreading of ENACW into the Rockall Trough, observed in relatively low temperature and high salinity signals (Figure 4).

We have addressed the interannual variation observed for the  $A_T/S$  relationship in the updated version of the manuscript (Appendix B).

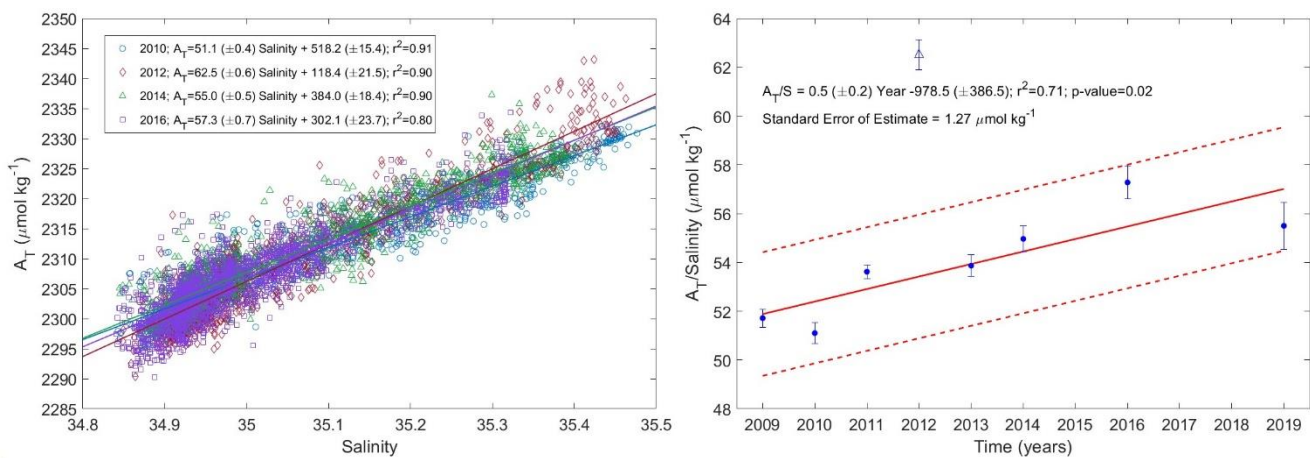


Figure. (a) Observations of  $A_T$  versus Salinity during the cruises in 2010, 2012, 2014 and 2016 plotted along with the regression lines, equations and  $r^2$  (statistically significant at the 99% level of confidence). (b) Temporal variations in the  $A_T/S$  relationship with error bars representing the standard deviations. The continuous red line depicts the linear regression, whose slope represents the interannual change in  $A_T/S$ . The linear equation,  $r^2$ , p-value and standard error of estimate is shown in the panel. The  $A_T/S$  for the cruise of 2012 was considered an outlier and excluded from the fitting. The dashed red lines represent the error margins of the prediction ( $1\sigma$ ).

C-06: Line 392: Figure S2 presents the N-AT distribution for 2009 and 2016. Curiosity: Why in 2016 the N-AT distribution appeared noisier and concentrations higher in the eastern sector compared to 2009? Is the same observed on other recent cruises 2014 or 2019?

It is true that the section for NAT during 2016 looks noisier than for 2009. It is due to the enhanced spatial resolution of the most recent cruises in comparison with the first ones. The number of sampling stations increased since 2009. The black dots, which represent data points, are closer one to each other in the panel of 2016 and thus interpolations were performed (using ODV) with higher degree of certainty than for 2009.

C-07: Line 438: Authors write: “The interannual ratios are presented along with their respective standard error of estimate and correlation factors ( $r^2$  and p-value) in Table 3 and S4.”. Correct: Table 2 (not 3).

Done in the updated version.

C-08: Line 451: “SEANOE [<https://www.seanoe.org/>], Pascale et al., 2022”. Correct name: Lherminier et al 2022.

Done in the updated version.

C-09: Line 517: Authors write: “while the  $C_{nat}$  experienced a slightly decrease throughout the region (Figure 5 and Table 2).” Any chance to specify the origin of this decrease ? (e.g. less export over time?)

See C-11

C-10: Line 519: Authors write: “The increase in the ventilation rates during this decade...”. Which decade ? Maybe specify the period: “The increase in the ventilation rates over 2009-2019...”

Done in the updated version.

C-11: Line 521: Authors write: “explained the higher growth in  $C_{ant}$  than expected due to the atmospheric  $CO_2$  increase.” Could you recall the expected  $C_{ant}$  trend due to the atmospheric  $CO_2$  increase? +0.5, +1, +1.5  $\mu\text{mol/kg/yr}$ ?

The entire second paragraph in section 4.2 was modified in the updated version of the manuscript to address the comments from C-09 to C-11. We have discussed the comment C-11 and we think that the statement “The increase in the ventilation rates over 2009-2019, shown by the negative AOU trends (Figure S6 and Table S4), explained the higher growth in  $C_{ant}$  than expected due to the atmospheric  $CO_2$  increase” is controversial and does not reflect what our results show. Therefore, we have decided to reformulate this part of the discussion.

What we wanted to emphasize is the relevant role of deep-water formation on modulating the  $C_{ant}$  and  $C_{nat}$  content. The 2009-2019 was a period of increasing ventilation in which the surface-to-bottom transport of water has enhanced. It favoured the surface waters with high  $C_{ant}$  (due to the increasing uptake of atmospheric  $CO_2$ ) and low  $C_{nat}$  (due to the high biological production) to be injected into the interior ocean, where  $C_{ant}$  is lower and  $C_{nat}$  is higher (mainly due to the enhanced remineralization). As a result, the  $C_{ant}$  has increased following quasi-linear trends throughout the layers and basins, while  $C_{nat}$  has slightly decreased (mainly in the Irminger Sea due to the higher ventilation of the western NASPG). It addressed the comment C-09. The differences between years in the biological carbon pump behave as a source of variation for  $C_{nat}$  and explained its non-significant trends at several layers.

Here we presented the updated paragraph:

“The entrance of  $C_{ant}$  through the air-sea interface and its accumulation dominated the observed increase in  $C_T$  (Figure 5 and Table 2). The increase in ventilation over 2009-2019, shown by the negative AOU trends (Figure S6 and Table S4), favoured the vertical

mixing. The upper waters, due to be in contact with the atmosphere and have high biological production rates during the warm months, show high  $C_{\text{ant}}$  and low  $C_{\text{nat}}$  contents. The enhanced transport of upper waters toward the interior ocean explained the rapid growth in  $C_{\text{ant}}$  at intermediate and deep layers. The  $C_{\text{ant}}$  trends ranged between 0.85 and 1.77  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  (statistically significant at the 99% level). They were higher than the observed on a decadal to multidecadal scale since the late 20<sup>th</sup> century in the Irminger and Iceland basins (0.21-0.89  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  during 1991-2015, García-Ibáñez et al., 2016; and 0.38-1.15  $\mu\text{mol kg}^{-1} \text{yr}^{-1}$  during 1983-2013, Pérez et al., 2021), which show the enhancement in the  $C_{\text{ant}}$  accumulation on interannual scales during periods of high ventilation, as previously reported by Perez et al., (2008). The  $C_{\text{nat}}$  show an inverse relationship with  $C_{\text{ant}}$  at intermediate and deep layers ( $r^2 > 0.5$ ; statistically significant at the 95% level of confidence) and weakly decreased across the western deep-convection NASPG (Figure 5 and Table 2). The increasing ventilation depleted  $C_{\text{nat}}$  in the upper waters by transporting it toward the interior ocean. The  $C_{\text{nat}}$  showed a weaker decrease at intermediate and deep layers due to the dominance of remineralization, which was not intense enough at this time of the year to neutralize the downward transport of low- $C_{\text{nat}}$  water from the surface but accounted to partially compensate for its effect. The observed variations in  $C_{\text{nat}}$  between years were strongly linked with fluctuations in the biological processes explained its non-significant trends at several layers. The changes in the circulation pattern of the NASGP and thus in the horizontal advection related with the climatological forcing (Balmaseda et al., 2007; Desbruyères et al., 2013; Mercier et al., 2015; Thomas et al., 2008; Xu et al., 2013) could behave as a source of variability for both  $C_{\text{ant}}$  and  $C_{\text{nat}}$  and also infers differences between consecutive years”.

C-12: Line 561: Authors write: “keeping approximately constant the CT (Table 3)”. Correct to Table 2. Maybe recall in the text the value of the CT trend = +0.05  $\mu\text{mol/kg/yr}$  in ENACW.

Done in the updated version.

C-13: Line 583: Authors write: “...where strong slowdowns in ventilation were observed from 2009 to 2010 and from 2013 to 2014, resulted in a relatively increase in  $C_{\text{nat}}$  and decrease in  $C_{\text{ant}}$  observed in SPMW”. Is this signal could be associated with the low NAO index in 2010?

We updated this paragraph as follow:

“The extreme negative NAO index of 2009-2010 (Jung et al., 2011) weakened the wind forcing, which infers variability in the circulation patterns and physical properties of the surface waters, consequently reducing deep convection. This was observed in the slowdown in ventilation from 2009 to 2010 (Figure S6) in the Irminger and Iceland basins which caused a relatively increase in  $C_{\text{nat}}$  and decrease in  $C_{\text{ant}}$  (Figure 5)”.

C-14: Line 635: Authors write: “The year-to-year variability in the biogeochemical patterns after 2012 may be attributed to the fluctuations in the spreading into the Rockall Trough of several water masses occupying different depths coming from the south and east”. As there is also variability in AT and N-AT, would that be also associated to local

or regional biological processes (e.g. Cocco Blooms) and linked to the NAC transports (as discussed lines 556-559).

Our results show that the ENACW in the Rockall Trough was well-oxygenated (low AOU values; Figure S4) but the ventilation was highly variable over time (several differences were found for AOU between consecutive years). This likely introduced differences in the biological carbon pump and in the horizontal and vertical advection which collectively introduced heterogeneities in the temporal distribution of  $A_T$ ,  $NA_T$ ,  $C_T$  and  $NC_T$  (Figure 5, S4 and S5). We have added the following statement to the updated version of the manuscript: “The heterogeneities in the ventilation of the ENACW between consecutive years likely influenced the biological and advective patterns, introducing differences in  $A_T$ ,  $NA_T$ ,  $C_T$  and  $NC_T$  (Figure 5, S4 and S5)”.

C-15: Line 672: Authors write: “The substantial variability introduced by these processes made it difficult to discern the pattern of acidification and its drivers on an interannual scale in the shallow Rockall Trough. Therefore, long-term monitoring and the development of multidecadal-scale studies are required in this area to derive significant conclusions.” I agree that maintaining long-term monitoring is important and it is often difficult to detect and explain the pH variations: however it seems that, and opposed to other regions, the Cant trend in the ENACW is relatively well estimated (quasi-linear) in the Rockall Trough (Figure 5) with value of  $0.85 \pm 0.11 \mu\text{mol/kg/yr}$  and that  $C_{\text{nat}}$  decrease is also well evaluated ( $-0.84 \pm 0.50 \mu\text{mol/kg/yr}$ ). For this water mass would it be possible to suggest some process that explain the  $C_{\text{nat}}$  decreasing trend since 2009 (physics and/or biology ?).

We have modified this paragraph in the updated version of the manuscript. We decided to remove that statement as is not appropriate in this part of the Manuscript. The long-term monitoring is important in regions which present high variability at different timescales such as the Rockall Trough. It is required to evaluate with higher certainty (with and statistical level of confidence higher than the 95%) the variations in the physical and biogeochemical seawater properties. As the differences between consecutive years encountered for MCS properties over 2009-2019 have behaves as a source of uncertainty at some layers as for example the ENACW, we would like to highlight the need of continue monitor and develop observation-based studies in such crucial areas, as we detailed in the conclusions.

In the updated paragraph (section 4.3), we suggest that the observed variability in the MCS through the ENACW is linked with changes in the advection, which modified the ventilation and spreading of water masses with different physical and biogeochemical patterns, and in the biological processes. Holliday et al., (2020) reported the freshening of the eastern NASPG led the weakening in the poleward advection of saline waters and strengthening in the eastward recirculation of freshwater during 2012-2016. The observed decrease in salinity (Figure 4) and its impact on  $A_T$  compared to  $NA_T$  (Figure S4) reflect this behaviour. The decrease in  $C_{\text{nat}}$  offsetting the increase in  $C_{\text{ant}}$  was also discussed: the depletion in  $C_{\text{nat}}$  could be related to the increasing biological uptake (Ostle et al., 2022) and physical and biogeochemical changes driven by fluctuations in the lateral advection (Holliday et al., 2020).

The updated paragraph is presented here:

“The upper waters of the Rockall Trough presented the maximum  $pH_T$  throughout the transect (8.02-8.08 units). The observed strong  $pH_T$  fluctuations between years related with interannual changes in the NAC do not allow to discern trends with a statistically interval of confidence equal or higher than the 90%. The interannual decrease in  $pH_T$  in the ENACW ( $\sim 0.001$  units  $yr^{-1}$ ) was half than the observed along southernmost transects in the Rockall Trough between 1991 and 2010 ( $\sim 0.002$  units  $yr^{-1}$ , McGrath et al., 2012a). The temporal distribution of the average  $pH_T$  (Figure 6) highly influenced by the high-ventilation (seen in minimum AOU values highly variables between years and which tend to decrease with 99% statistical confidence; Figure S6 and Table S4) allow to discern two periods: the approximately constant ventilation rates keep a steady state in terms of  $pH_T$  during 2009-2011, while the progressively renewal and oxygenation of subsurface waters after 2012 (and peaking in this year) increase the  $pH_T$ . The renewal of waters in the shallow Rockall Trough, in contrast with the westernmost NASPG, was not primarily driven by vertical but by lateral advection. The modifications of the ENACW through air-sea exchange and mixing with adjacent waters modulated its properties at different time scales (Holliday et al., 2000) and caused the observed variations in the MCS. The variations in  $pH_T$  between consecutive years after 2012 may be attributed to the fluctuations in the spreading into the Rockall Trough of several water masses occupying different depths coming from the south and east (Ellett et al., 1986; Pollard et al., 1996). Holliday et al., 2020 reported the reduction in the spreading of saline subsurface waters from subtropical latitudes and diversion of Arctic freshwater from the western boundary into the eastern NASPG during 2012-2016. The subsequent freshening of the ENACW compensated for the increase in  $A_T$  expected without the effect of salinity (see in the decreasing  $A_T$  against the increasing  $NA_T$ ; Figure S4 and Table S4) and weakened the increase in  $C_T$  expected due to poleward advection (see in the slowdown in the rise of  $C_T$  in comparison with those of NCT; Figure 5 and S5 and Table 2 and S4). The  $C_T$  remains approximately constant (Figure 5 and Table 2) due to the increase in  $C_{ant}$  ( $0.85 \pm 0.11 \mu mol kg^{-1} yr^{-1}$ ; p-value  $< 0.01$ ) was neutralized by the decrease in  $C_{nat}$  ( $-0.84 \pm 0.50 \mu mol kg^{-1} yr^{-1}$ ; p-value  $< 0.1$ ). These findings suggest that the atmospheric  $CO_2$  invasion was offset by the growing phytoplankton biomass favouring its biological uptake (Ostle et al., 2022) and the weakening transport of remineralized and saline water from the south (Holliday et al., 2020), thus compensating the acidification of the ENACW”.

C-16: Line 697: Authors write: “the positive NAT trends encountered in the upper layers lead a rise in pHT, while the diminished NAT contributed to decrease the pHT toward the interior ocean.” Is the N-AT trend could be related to change in Cocco blooms distributions or the signal is too low to interpret the link with the various biological processes in the north atlantic (Ostle et al, 2022) ?

The many processes involved in the distribution of  $NA_T$  difficult to assess its trend. The evaluation of the  $NA_T$  drivers required to consider the biogeochemical processes influencing the bicarbonate-carbonate equilibrium in seawater. The variations in  $NA_T$  could be related to changes in biological production, but its direct influence is not significant. However, it has an indirect effect through the production/decomposition of organic matter as well as through its influence on biogeochemical cycles (i.e. nutrients). The variations in pH have an important role by changing the fundamental chemical equilibrium in seawater: the acidification favour the dissolution of carbonates, which increases  $NA_T$ . Lastly, the freshwater inflows with high levels of bicarbonate could increase  $NA_T$ .

The positive  $NA_T$  trends encountered in subsurface layers (SPMW and ENACW) could be related to the diminishing in  $pH_T$ , which favour the dissolution of carbonates, combined with increasing biological production reported for upper layers across the NASPG (Ostle et al, 2022). It contrasts with the constant to weakly decreasing  $NA_T$  trends at intermediate and deep layers, in which the accelerated acidification was compensated by the dominance of remineralization processes over lower biological uptake. The  $NA_T$  trends were not-statistically significant in most of the layers and basins due to the year-to-year differences in the processes involved in its variability. This statement was including in this part of the discussion.

The increasing phytoplankton biomass and subsequent enhancing in primary production reported for the NASPG by Ostle et al, (2022) was evidenced in the decrease in  $C_{nat}$ , although the effect of advection also introduced variations. It has been discussed at some points of the new version of the manuscript.

C-17: Line 742: Figure 7, Table 2: The trend of  $W$  in the Irminger Sea of around  $-0.07$  per decade seems high compared to that deduced from reconstructed products (e.g.  $-0.039 \pm 0.009$  per decade, Chau et al, 2024). Could that be discussed ? Would that be explained by seasonal difference of the trends (e.g. Leseurre et al, 2020) ?

Chau et al, 2024 highlighted the uncertainty in their own  $pH$  and  $\Omega_{Arag}$  estimations in the Irminger and Iceland basins (also for the estimations performed by Bates et al., 2014) due to low data-sampling frequency at their monitoring sites. We have updated this part of the discussion by including these statements: "...The  $\Omega_{Arag}$  trend estimated for SPMW in the Irminger basin ( $-0.007 \pm 0.003$  units  $yr^{-1}$ ) is consistent with that reported for surface waters by Bates et al., (2014) over 1983-2014 ( $-0.008 \pm 0.004$  units  $yr^{-1}$ ) and fall within the range of those estimated during summer by Leseurre et al., 2020 over 2008-2017 ( $-0.005 \pm 0.001$  units  $yr^{-1}$ ). Chau et al., 2014 recently deduced from reconstructed products a slower decrease ( $-0.004 \pm 0.001$  units  $yr^{-1}$ ), highlighting the large uncertainty in the estimations of interannual trends for  $pH$  and  $\Omega_{Arag}$  across the NASPG due to the low-data sampling frequency at their monitoring sites..."

C-18: Line 811: Authors write: "The driver analysis exhibited the strongest interannual decrease in  $\Omega$  in the upper layers governed by the uptake of  $C_{nat}$  weakly compensated by the increase in  $NA_T$  and favoured by the cooling and freshening." Here, again, you identify that the  $NA_T$  increase explains part of the  $W$  changes: what is the process associated to this variations (Cocco bloom ?) ?

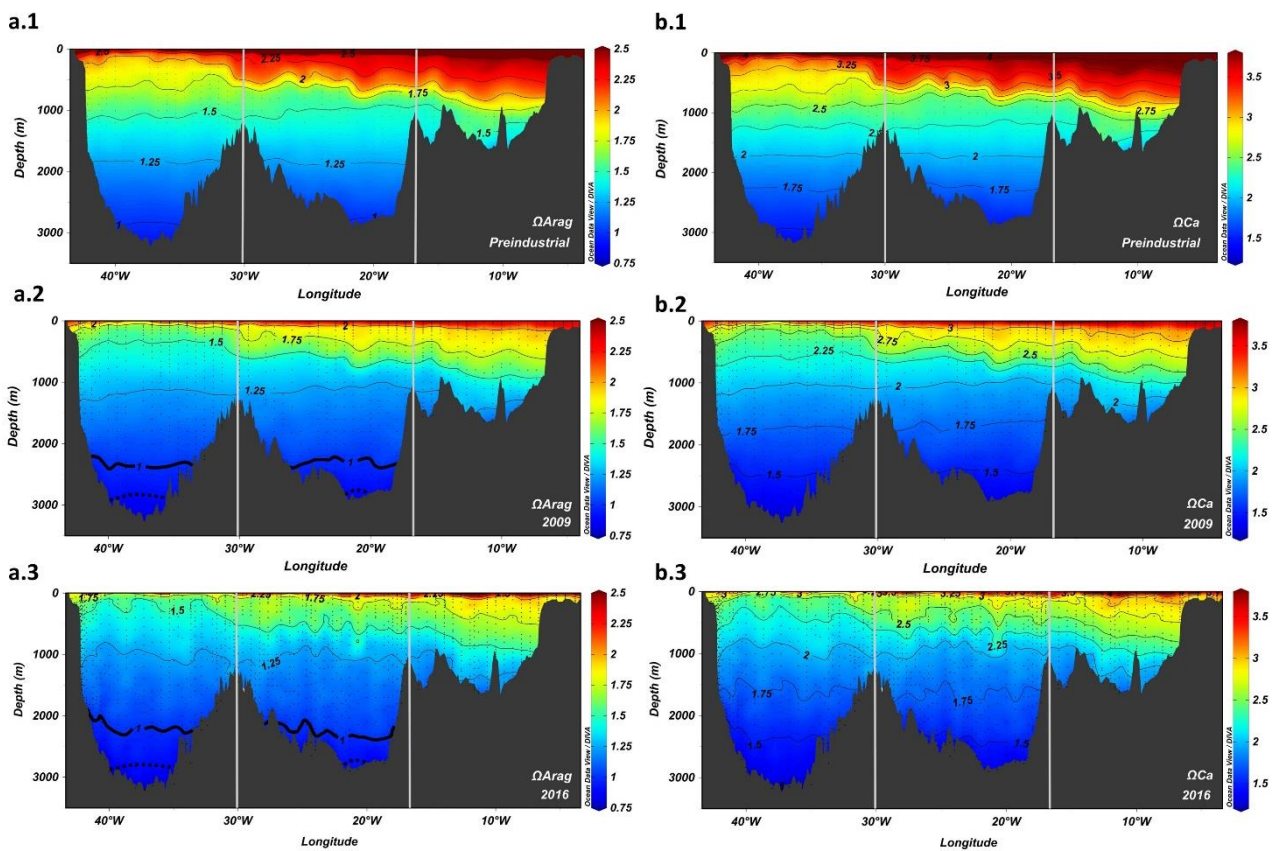
We explained it in the updated version of the manuscript and provided a detailed description of the processes involved in the variation of  $NA_T$  to response comment C-16. We have added the following statement in section 4.4: "...The interannual increase in  $NA_T$  in upper layers could be related to the diminishing in  $pH_T$ , which favour the dissolution of carbonates, combined with increasing biological production reported for upper layers across the NASPG (Ostle et al, 2022). It contrasts with the constant to weakly decrease in  $NA_T$  at intermediate and deep layers, in which the accelerated acidification was compensated by the dominance of remineralization processes over lower biological uptake. Consequently, the positive  $NA_T$  trends encountered in the upper layers lead a rise in  $pH_T$ , while the diminished  $NA_T$  contributed to decrease the  $pH_T$  toward the interior ocean ...".



C-19: Line 816: Authors write: “The progressive reduction in  $\Omega_{\text{Arag}}$  is driving a long-term decrease in the depth of the aragonite saturation horizon ( $\Omega_{\text{Arag}}=1$ ) by 80-400 m since the preindustrial era”. Curiosity: could you show a section (or profiles) of preindustrial War calculated with Cnat to compare with modern values (to add in Supp Mat ?).

We agree that this can greatly enrich this section of the discussion. We have computed the preindustrial  $\Omega_{\text{Arag}}$  and  $\Omega_{\text{Ca}}$  with the  $\text{CO}_{2\text{sys}}$  programme run in the MATLAB software and based on  $C_{\text{nat}}$ . The vertical sections of preindustrial  $\Omega_{\text{Arag}}$  and  $\Omega_{\text{Ca}}$  are depicted along with those for the cruises of 2009 and 2016 in the updated version of the Supplementary Material (Figure S3). Additionally, the isolines for the aragonite saturation horizon for preindustrial and present times are remarked in the new vertical sections.

The updated figure and capture are shown here:



“Figure S3. Water-column distribution along the longitudinal transect of (a)  $\Omega_{\text{Arag}}$  and (b)  $\Omega_{\text{Ca}}$  for preindustrial times and for the cruises of 2009 and 2016. The preindustrial  $\Omega_{\text{Arag}}$  and  $\Omega_{\text{Ca}}$  values were computed with the  $\text{CO}_{2\text{sys}}$  programme (Lewis and Wallace, 1998) run with the MATLAB software (van Heuven et al., 2011; Orr et al., 2018; Sharp et al., 2023) using as input  $\text{CO}_2$  system variables the  $A_{\text{T}}$  and the  $C_{\text{nat}}$ . In panels a.2 and a.3, the highlighted continuous black isolines represent the aragonite saturation state horizon during the cruises of 2009 and 2016, respectively, while dash isolines show the aragonite saturation state horizon in preindustrial times. The vertical white lines show the limits between basins”.

We also added the following sentence to the discussion (section 4.5):

“The vertical section of  $\Omega_{\text{Arag}}$  in Figure S3 shows the shallower aragonite saturation horizon during 2009 and 2016 compared to preindustrial times”.

C-20: Line 834: Authors write “In fact, several studies reported that CWC ecosystems are anticipated to be among the first deep-sea ecosystems to experience acidification threats”. Maybe refer to Gehlen et al (2014) (and also in the introduction).

Done in the updated version.

C-21: Line 856: Authors write “The observational period is relatively short to quantify long-term trends and to formulate significant future projections.” Maybe delete here, as this was also written on line 840: “Despite the observational period is relatively short to quantify long-term trends and to formulate significant future projections,...”.

Done in the updated version.

C-22: 900: “to favour the entrance of Cant in intermediate and deep-layers and this its acidification,”

It was a typo and was removed

C-23: Conclusion: Maybe reduce the numbers of values listed in the conclusion (already listed in the MS).

Done in the updated version.

Figures:

C-24: Figures: check the color code for blind (see accepted colors for the journal Biogeoscience)...

We checked the readability of our figures and maps by readers with color vision deficiencies using the Color Blindness Simulator (Coblis) provided by the journal Biogeosciences. We have observed that the color scheme used for the vertical sections in Figures 1, 2, 3, S2, and S3 effectively highlights the contrasts between maximum and minimum values, allowing for a clear identification of the vertical gradient observed for each variable. Additionally, we included contour lines or isolines to facilitate the interpretation of the results and enhance the understanding of the distribution of intermediate values, where the color scheme introduces some brightness.

In contrast, the color scheme used for Figures 4, 5, 6, 7, S4, S5, and S6 may be confusing for those with dichromatic vision (protanopia and deuteranopia). We have adjusted the hues of the cool colors (violet and green) to enhance the contrast with the warm colors (orange and red) for individuals with protanopia and deuteranopia. Additionally, we have replaced the orange color with red in the uLSW layer to increase contrast with the other colors used in the Irminger and Iceland basins. We hope these changes will improve the accessibility of the figures and enhance the interpretation and impact of the results.

C-25: For Figures 4, 5, 6, 7, a suggestion: change the color code for ENACW (i.e. different than for uLSW)

Done in the updated version.

C-26: In figure 5 the CT and Cant concentrations in ENACW were low in 2014. I think this is not discussed. Any idea to explain this anomaly ? A strong bloom in 2014 in the eastern NASPG or a shift in the data for this cruise ?

During 2014, the ENACW presented low  $C_{nat}$  which relatively decrease the  $C_T$  (Figure 5) and high  $A_T$  and  $NA_T$  (Figure S4). These changes occurred in phase with the warming observed from 2012 to 2014 (Figure 4) and likely indicated that the increase in carbonate and bicarbonate concentrations rising  $A_T$  and  $NA_T$  was compensated by the depletion in dissolved  $CO_2$ . The relatively high temperature and  $NA_T$  in 2014 indicates an improved spreading of subsurface waters from subtropical latitudes into the Rockall Trough. The enhanced biological production in these waters, together with the reduction in solubility due to warming which favour the  $CO_2$  evasion to the atmosphere, account for decreasing  $C_{nat}$  and thus  $C_T$ .

This explanation was added to the discussion in the new version of the manuscript.

References:

C-27: Friedlingstein et al, 2022: add full reference, journal, doi

C-28: Pascale, L., 2022: change to

Lherminier P., Perez, F. F., Branellec, P., Mercier, H., Velo, A., Messias, M. J., Castrillejo, M., Reverdin, G., Fontela, M., Baurand, F. (2022). GO-SHIP A25 - OVIDE 2018 Cruise data. SEANOE. <https://doi.org/10.17882/87394>

References were corrected in the updated version of the manuscript.

;;;;;;;;;;;;; References added in this review not cited in the MS

Chau, T.-T.-T., Gehlen, M., Metzl, N., and Chevallier, F.: CMEMS-LSCE: a global, 0.25°, monthly reconstruction of the surface ocean carbonate system, Earth Syst. Sci. Data, 16, 121–160, <https://doi.org/10.5194/essd-16-121-2024>, 2024.

Dickson, A. D. 1995. Determination of dissolved oxygen in sea water by Winkler titration. WOCE Operations Manual, Part 3.1.3 Operations & Methods, WHP Office Report WHPO 91-1.

Gehlen, M., Séférian, R., Jones, D. O. B., Roy, T., Roth, R., Barry, J., Bopp, L., Doney, S. C., Dunne, J. P., Heinze, C., Joos, F., Orr, J. C., Resplandy, L., Segschneider, J., and Tjiputra, J.: Projected pH reductions by 2100 might put deep North Atlantic biodiversity at risk, Biogeosciences, 11, 6955-6967, [10.5194/bg-11-6955-2014](https://doi.org/10.5194/bg-11-6955-2014), 2014.

Lauvset, S. K., Gruber, N., Landschützer, P., Olsen, A., and Tjiputra, J.: Trends and drivers in global surface ocean pH over the past 3 decades. *Biogeosciences*, 12, 1285-1298, doi:10.5194/bg-12-1285-2015, 2015

Lherminier P., Perez, F. F., Branellec, P., Mercier, H., Velo, A., Messias, M. J., Castrillejo, M., Reverdin, G., Fontela, M., Baurand, F. (2022). GO-SHIP A25 - OVIDE 2018 Cruise data. SEANOE. <https://doi.org/10.17882/87394>

Müller, J. D., Gruber, N., Carter, B., Feely, R., Ishii, M., Lange, N., et al.: Decadal trends in the oceanic storage of anthropogenic carbon from 1994 to 2014. *AGU Advances*, 4, e2023AV000875. <https://doi.org/10.1029/2023AV000875>, 2023

Newton, J.A., Feely, R. A., Jewett, E. B., Williamson, P. and Mathis, J.: Global Ocean Acidification Observing Network: Requirements and Governance Plan. Second Edition, GOA-ON, <https://www.iaea.org/sites/default/files/18/06/goa-on-second-edition-2015.pdf>, 2015.

Ostle C., P. Landschützer, M. Edwards, M. Johnson, S. Schmidtko, U. Schuster, A. J. Watson and C. Robinson, 2022. Multidecadal changes in biology influence the variability of the North Atlantic carbon sink. *Environ. Res. Lett.* 17, 114056, DOI : 10.1088/1748-9326/ac9ecf

Terhaar, J., Goris, N., Müller, J. D., DeVries, T., Gruber, N., Hauck, J., et al. (2024). Assessment of global ocean biogeochemistry models for ocean carbon sink estimates in RECCAP2 and recommendations for future studies. *Journal of Advances in Modeling Earth Systems*, 16, e2023MS003840. <https://doi.org/10.1029/2023MS003840>