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Biogeosciences Discussions

Interactive comment on "Ocean carbonate system variability in the North Atlantic Subpolar surface water (1993–2017)" *by* Coraline Leseurre et al.

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Review 2 (Are Olsen):

"This contribution presents the trends in a very impressive set of data from the western subpolar North Atlantic, collected within the framework of the SURATLANT program. While the paper presents the data and trends more or less adequately, it fails in properly attributing them to (climatic + oceanographic + biological) drivers. These aspects are

unclear, speculative, and somewhat confused. As such, major revisions are required. My major comments are:"

We thank Are Olsen for his fast review, comments and questions that will be taken into account when revising the manuscript. Below we list our responses before preparing a revised manuscript.

1. Samples are collected two to four times a year. My sense is that with such rare sampling one might be very prone to getting false trends because of differences in the timing of the data collection, also in combination with timing of the spring bloom. For example, the strong increase in region C+D+E summer DIC in 2001 -2008 might be generated by earlier sampling/later spring bloom.

"While some effort seems to have been taken to deal with this; reconstructed data; it is not quite clear how this is done, and how well it works. Some improvement is needed in this description (page 5 lines 1-19). Is it possible to test this scheme by using data from a year with sampling in all of the months used (e.g. Jan-March)? At least for SST and SSS this can be done as there should be continuous TSG data vailable, and for fCO2 there are data available from the VOSs Nuka Arctica and Atlantic Companion, which crosses the study area. These can be retrieved from SOCAT and used to test the method".

Response: We fully agree with the reviewer. For the analysis of trends, one should take care of the timing of sampling especially in summer (as well as different tracks for some transects). This is why previous studies focused on winter data (e.g. Metzl et al., 2010; Fröb et al., 2019). Here, in addition to winter 2008-2017 not previously analyzed, we also tried for the first time to evaluate the trends in summer (June-July-August), i.e. after the spring bloom that generally occurs in April-May-June (with a date variable between regions and dates). Do we detect (or not) different view on the trends when comparing winter and summer? On average for 1993-2017, we observed that DIC and fCO2 increase (and pH decrease) in both winter and summer (but with different rates).

Results are much more different when trends are limited to short periods.

Concerning the “strong increase in region C+D+E summer DIC in 2001 -2008”. This is apparently driven by low DIC observed in summer 2001 and high DIC in summer 2007 (medium concentrations observed in 2004-2005, but no data in 2006). Note that for this particular summer 2007 transect samples were taken in early August 2007 and thus correction to standardize to mid-July should not introduce suspicious bias. As suggested by the reviewer, we had a look at the Chl-a time-series (from MODIS) but for now we did not observe a significant difference for the summer 2007 (e.g. low productivity). Also, in summer 2008, DIC and nutrients were significantly lower than in 2007. At that stage we have no firm conclusion to explain the rapid increase of DIC (and fCO2) for summer 2001-2007. Note that in winter 2001-2007 we also found rapid fCO2 increase.

In years when observations were not collected in July or February (selected months for summer and winter trend and driver’s analysis), we use data collected in previous or next month by correcting for the climatological trend between successive months. As suggested by the reviewer we will confirm this scheme with the more regularly-observed SST and SSS data, based on the binned monthly products constructed by Reverdin et al. (2018b).

We also checked the SOCAT data in the region for specific years (e.g. the summer 2010 anomaly). A comparison with SOCAT fCO2 data for the full period would be very interesting but this is beyond the scope of our analysis mainly based on DIC/TA observations. This would be a topic for another publication shared with SOCAT data providers in the North Atlantic.

2. More broadly, it would be interesting to know how representative the data are for large-scale interannual phenomena. This can be tested. For example, the trends in SST can be compared with objectively analysed SST from NOAA. The trends in SSS can be compared with some reanalysis model output. And again, for the time repre-

sentativeness, the continuous data from the TSG, which collects data on all crossings, should be used. It might also be worthwhile to look at remotely sensed Chl a, to evaluate if there are concomitant trends in surface ocean primary production, e.g. a loss in production from 2001 -2008 such as the DIC data seem to indicate and was suggested in the manuscript.

Response: as suggested by the reviewer, we will compare the SST and SSS trends with monthly fields along AX-2 line constructed by Reverdin et al. (2018b). We found coherent view on the timing and variability of surface properties (more clearly seen in SST and SSS anomalies). This will be added in the revision. We will also use SST reanalysis to confirm the trends for the selected periods. We also started to investigate Chl-a data (from MODIS). Preliminary results suggest that there are some Chl-a trends that might qualitatively explain the variability of DIC (more specifically variations of DIC-natural derived from DIC corrected to anthropogenic signal). To quantify the impact of biological processes on DIC and fCO2, a more specific study should be performed based on primary production (not only biomass). On this issue, Bennington et al (2009) used an ocean model and satellite Chl-a to explore the effect of biological activity on carbon uptake in the North Atlantic and found that biological variability “is not sufficient to be a first-order control on annual subpolar air-sea CO2 flux variability.” This might be explored in more detail with our observations (including nutrients) but for another publication. As a first step, we explored the winter and summer trends of natural N-DIC over 1993-2017 and found no significant trend in summer and winter, suggesting that biological activity variability has a small impact on the DIC and fCO2 long-term changes. This analysis from in-situ data confirms the conclusion from ocean models (Bennington et al, 2009).

The main signal that we can interpret based on satellite Chl-a is for the warm summer 2010 with high Chl-a observed (probably linked to NAO signal) and that could explain part of the DIC changes during that summer. However, this event has little impact on the trends for this specific period (summer 2008-2017). There are no summer 2010

SOCAT data, but the low summer 2010 DIC is confirmed with independent DIC and TA OVIDE-2010 observations.

3. The attribution section is not well done. In particular, I strongly suggest the authors to explicitly account for salinity changes in the pCO2 driver decomposition following the method by Keeling et al. (2004) and recently used in the subpolar North Atlantic by Fröb et al. (2019). The reason is that dilution may completely overwhelm increase of DIC expected from uptake of anthropogenic CO2.

Response: In the revised manuscript, we will consider separately the effect of salinity and N-DIC, N-TA following the reviewer's suggestion.

4. Further, showing trends in salinity normalised DIC and TA is worthwhile, but make sure to use the correct method for salinity normalising as described in Friis et al. (2003).

Response: we will add trend analysis in salinity normalized DIC and TA. In previous work we found about the same trends in 2001-2007 for TA/N-TA (negative) and DIC/N-DIC (positive) but for winter only (Metzl et al 2010). New results suggest that for summer 2001-2007, the trends are also about the same for DIC and N-DIC or TA and N-TA. However, as salinity is decreasing in 2008-2017 (also previously documented by Reverdin et al 2018a,b; Tesdal et al 2018; Fröb et al 2019), trends for N-TA and N-DIC are significantly different compared to TA and DIC for the last decade. This supports the reviewer's suggestion to show both DIC/TA and N-DIC/N-TA trends and discuss processes that drive the observed changes. Of course, we follow the same approach as Friis et al. to estimate the normalized DICA and TA (but with equation in Reverdin et al. (2018a)). Thanks to highlight this issue.

5. Also, there is a lot of mentioning of the change in the air-sea CO2 difference. But this is not illustrated, which leaves a lot to the readers imagination. I therefore recommend to show the actual air-sea fCO2 difference in Fig. 3. You can make room for this by removing one of the panels for Omega (Calcite/Aragonite), as there is no need to show both.

Response: this is a good point and we will add a figure of air-sea fCO2 differences. Replacing Omega is a nice idea, but we think a separate figure should be added to present air-sea CO2 gradients for the full period and seasons.

6. Uncertainties are not properly dealt with. For example, the uncertainty in the SST of 0.1 degrees C results in an fCO2 uncertainty of 2 μatm. There are also uncertainties in DIC and TA. These errors need to be propagated to calculated fCO2, pH etc. This can be done using the most recent CO2SYS from James Orr as this includes error propagation. It is available from GitHub. The errors in fCO2 (and the others) can be propagated to the trends using Monte Carlo.

Response: In Table 2, significant trends are presented with (*). But, as in some cases, there are no trends (this can not be determined by the Student's method), so we chose a maximum error criterion (but maybe not adapted as suggested by Are Olsen). We will correct that with the most recent CO2SYS (from James Orr), thank you for this pertinent comment.

7. I am in particular concerned with the fact that the large summer fCO2 increase in region B 2001-2008 are basically caused by the 'reconstructed' data of 2001 and 2008. The actual observations are pretty steady. How confident are you in these reconstructed values?

Response: the large summer fCO2 increase in 2001-2008 is observed in all boxes. Reviewer is correct that starting (2001) and ending (2007) points were reconstructed for July from data obtained in August (for both boxes B and CDE). If we don't standardize the data to July, the "summer" (June+July+August) trend in Box B would be +6.8 μatm/yr (instead of 8.5 when normalizing to July). The same test for box CDE leads to fCO2 increase of +11.2 μatm/yr (instead of 11.5 when normalizing to July). We should note that for August 2007, TA data were doubtful and we used reconstructed TA from salinity for this specific cruise. Overall, we are confident that in 2001-2007 the fCO2 increased much faster than in the atmosphere.

8. As the other reviewer, I think this contribution confuses anthropogenic and natural CO2. The trends that are observed in pCO2 does not say any on anthropogenic carbon uptake. Further, the North Atlantic is not a big sink of anthropogenic CO2 because the air-sea flux is large. The air-sea flux is a combination of natural and anthropogenic CO2 fluxes. Horizontal advection is likely a big source of anthropogenic CO2 to the North Atlantic. Hence, the data that are presented only informs about the changes in surface pCO2 and air-sea CO2 flux, not on the North Atlantic sink for anthropogenic CO2. This needs to be considered both in the introduction and in the discussion.

Response: Thank you to highlight the anthropogenic versus non-anthropogenic signal issue (same remark by reviewer 1). We therefore address the same response below.

As the DIC and pCO2 variability are large in surface waters, detection of anthropogenic CO2 (C-ant) is difficult (if time-series are limited) and data-based methods such as C0, TrOCA, delta-C*... not suitable to quantify C-ant in surface waters. Indeed, longer-term time-series are required to separate natural and anthropogenic (or climate induced) signals (e.g. at least 30 years in the North Atlantic subpolar gyre, McKinley et al, 2011). We have evaluated the anthropogenic concentrations (C-ant) in this region based on subsurface Glodap-V2 data (Olsen et al., 2016) and TrOCA method, but we did not introduce the results in the present paper as they correspond to a different period (and only for summer). However, in the revision, as recommended by reviewers, we will introduce the trends of C-ant evaluated in subsurface from different methods (our calculations and recently from Gruber et al., 2019). In short, based on Glodap-V2 data in the NASPG we estimate C-ant trend of +0.7 μmol/kg/yr at subsurface (150-200m) for the period 1997-2011. This would explain 75% of the DIC trend of +0.9 μmol/kg/yr observed at subsurface for the same period. On the other hand, using the new e(MLR*) method, Gruber et al (2019) evaluate accumulation of C-ant from 1994 to 2007 in the global ocean. In the North Atlantic and specifically along the SURATLANT line, the accumulated C-ant is +8.5 (+/- 1.7) μmol/kg in the layer 150-200m. This signal is rather homogeneous at depth (150-200m) but with a small gradient between southern

(6 μmol/kg) and northern (10 μmol/kg) NASPG regions. This would correspond to a trend of between +0.6 and +0.7 μmol/kg/yr, very close to our C-ant estimate based on Glodap-V2. If we assume that the subsurface C-ant trend results are correct and also valid for the surface layer, the e(MLR*) method lead to a very homogeneous C-ant accumulation in the layer 0-50m along the SURATLANT line (55N-64N), of +10.1 (+/- 0.8) μmol/kg between 1994 and 2007, i.e. about +0.8 μmol/kg/yr. Interestingly, this is in the range of the long-term (1993-2017) DIC surface trends that we report (table 2 in our paper) between 0.6 and 0.7 μmol/kg/yr in boxes B,C,D,E (depending the season). However, our observations also show that DIC trends could be very different for short periods and north/south regions (see all trends listed in Table 2), and occasionally DIC decreases over time as opposed to C-ant (e.g. 1993-1997 winter, this study; Metzl et al., 2010; Ullman et al 2009). Based on the C-ant estimates, we will also evaluate and discuss the trends of DIC-Cant (C-nat), i.e. the natural part of the signal in the revision. We will discuss the results in relation with biological activity as also suggested by reviewer (using remote sensing Chl-a).

Figure 1 and 2 (attached) show the anthropogenic and natural contributions of DIC to pH change. This overview is obtained with the C-ant set at 0.7 μmol / kg / yr (in each region, period, season). We note that the results presented in boxes C and D-E are substantially the same as when we group them in C-D-E.

9. I think the discussion is pretty disappointing. It is basically a recap of the results + some more exploration of these, combined with some speculation based on published literature. There are no attempts to analyses the relation between the observed trends and likely drivers (for which data exist) – such as NAO or AMV indices, winter/summer mixed layer depths (Argo data), primary production (remotely sensed ocean color), and SGP strength (SPG index). This should be done.

Response: We agree that a further discussion should be attempted to link the observed changes of surface properties with regional or large scale forcing such as NAO, AMV. The same comment was addressed by reviewer 1. We therefore address the same

response below.

About NAO:

NAO was previously recognized a possible link of the rapid fCO2 increase when NAO shifted from positive to negative phase in 1995-1996 (Corbière et al., 2007). However, this was not confirmed for the period 2001-2007 when NAO did not vary so much around neutral value (Metzl et al., 2010). A possible explanation is that the observed variations of fCO2 in the NASPG, especially rapid trends such as +7 μatm/yr observed in 2001-2007, are driven by superimposed processes linked to climatic signals such as NAO and AMO, and both should be taken into account, as well as other processes involved in the NASPG ocean circulation, ventilation and vertical mixing. AMO, based on SST, is a long-term multidecadal signal (sometimes better called AMV) that experienced a gradual progressive increase from negative values in the 70s to positive values in the early 2000s, and remains positive and relatively stable in 2002-2017. NAO based on sea level atmospheric pressure gradient, shows much shorter variability, with highs and lows occurring in its winter record at interannual to decadal periods. We think that a direct relationship of the variability of pCO2 or CO2 uptake in the North Atlantic with NOA is still ambiguous (e.g. Takahashi et al., 2009; Schuster et al., 2013; Mc Kinley et al. 2017) and the detection of pCO2 changes with climate variability is still challenging from observations (at least for the period we investigated 1993-2017). However, for long-term multi-decadal variability the link between AMO and pCO2 change in the NASPG appears more robust (Breeden and McKinley, 2016; Landschützer et al., 2019).

Summary of NAO and model results cited:

Ocean models can help to understand the link between NAO and biogeochemical cycles, but results from models are still controversial. Keller et al (2012) who investigated several simulations (6 Earth systems models and for winter only, i.e. not the productive season), conclude that on-site entrainment in the subpolar gyre (mixing, upwelling) is

the main driver of carbon sink variability as opposed to advection (as suggested by Thomas et al. 2008). In their model Thomas et al (2008) suggest that negative or neutral NAO conditions result in a substantial decline in CO2 uptake for years 1997-2004 along the NAC and in the eastern subpolar gyre. However, in another modeling study of the North Atlantic, Ullman et al. (2009) conclude that the air-sea CO2 flux increased over the 1992–2006 and this is due to the increasing atmospheric CO2 and not to long-term variability in the physical climate or biogeochemistry. During the transition of NAO (from positive to neutral), their model simulates a decline of the convection and vertical DIC supply to the surface in the subpolar region, counteracting the increase of pCO2 due to warming. This leads to a small net pCO2 increase (compared to atmospheric trend) and increasing CO2 sink. Interestingly in the NASPG, the DIC decrease of -0.75 μmol/kg/yr in winter is more pronounced in the model (Ullman et al., 2009) than in the SURATLANT data (for 1992-2005). That might explain why observations suggest a reduced CO2 sink after the NAO shift in the mid-90s (Corbière et al., 2007), while the model suggest an increasing sink.

Keller et al., (2012) show that simulations with different coupled models lead to different results, and the response to NAO seems modest: typical NAO-driven variations at large-scale are +/- 10 μmol/kg for surface DIC and TA, and +/- 8 μatm for delta-pCO2. Depending on the model, the change of pCO2 in the subpolar region varies between +2/-4 μatm (for NAO+) and +16/-12 μatm (for NAO-), that is even the sign of the response is different between models. Such low variations, if real, are rather difficult to extract from observations and thus the results of the models difficult to validate. Thus, Keller et al (2012) conclude that although the interannual variability in the North Atlantic is largest in the subpolar gyre, the magnitude and responses of the carbon uptake to NAO significantly differ between the models (recall that this conclusion holds only for winter).

These model studies and their controversial results (level of variability and processes at play) show that there is still more work to be performed to understand the link between

NAO and biogeochemistry. As a matter of fact, in a recent analysis on the link between NAO and biology in the North Atlantic, Mc Kinley et al. (2018) conclude that "nowhere is the NAO correlated with biomass variability and that more investigation of the links between North Atlantic climate and biomass variability is clearly warranted". The same is true for biogeochemistry and carbon cycle and we believe that the new data on the carbonate system we analyzed in our submitted paper along the SURATLANT line should offer new information on this issue, even if the main conclusion is that over 1993-2017 there is NO direct link detected between fCO2 and DIC trends with NAO in this region of the NASPG. Of course, at shorter time scale (1-2 years) we might recognize such a link as described below (and see also the large changes in regions with changes in convection, such as in the western Irminger Sea, Fröb et al., 2018).

Summary of observations with new data related or not with the NAO:

The update made for the last decade 2008-2017 (data not included in our previous work, Corbière et al., 2007; Metzl et al., 2010) adds observations obtained during a strong negative NAO in 2010 and a positive NAO phase in 2015.

The 2010 event was associated with a warming and freshening (and low density) found in both SURATLANT discrete sampling (in August 2010) and monthly reconstructed Binned products (Reverdin et al 2018b). In August 2010 we observed low DIC (and also high δ13CDIC as discussed by Racapé et al 2014), a signal also revealed in high Chl-a concentrations (identified from MODIS data). However, as this was associated with a warming (observed positive SST anomaly up to +1.5 °C), the fCO2 (and pH) values were not very different from previous summers, illustrating the competitive effect of warming and higher production on fCO2 for this event. We also note that in summer 2010, DIC/TA was also sampled during the OVIDE-2010 cruise (in late June). In the NASPG, surface DIC concentrations for OVIDE-2010 were around 2070-2090 μmol/kg, i.e. just between SURATLANT data obtained in early June (2100-2110 μmol/kg) and in mid-August (2050-2070 μmol/kg); for TA, concentrations were the same for all cruises. This confirms the summer 2010 anomaly (low DIC) apparently associated with higher

productivity during the negative NAO phase, but with no significant impact on fCO2 and trends.

We also identified a cold year in 2015 (SST anomaly around -1 °C), when NAO was in a positive phase: for that year, DIC was higher in winter (near the maximum observed on average in winter in our time series) but again, as the temperature also lowers fCO2, the fCO2 value were not so different from other winters, now illustrating the competitive effect of cooling and deep mixing on fCO2.

With the new data introduced in this manuscript for 2008-2017 that corresponds to a period when NAO presents large IAV compared to 1995-2007, we observed more variability in the DIC data (and fCO2, pH) that might result in less clear trends over 5-10 years. We have identified two specific years (2010 and 2015) that experienced very low NAO (-3 in 2010) and high NAO (+2 in 2015), leading respectively to observed warming (cooling), freshening (saltier) and low (high) DIC; these anomalies could be explained by an increase in productivity in 2010 and deeper mixing in 2015. As these anomalies have been clearly recognized, we have tested the sensitivity of the trends analysis with and without these NAO events. Not surprisingly, for the 2015 anomaly and because we have no winter data after 2015, we derived significant different trends when 2015 is or not considered for the period 2008-2015. For example, for the northern boxes CDE the winter trends evaluated for years 2008-2015 were +1.1 μmol/kg/yr for DIC and 0.6 μatm/yr for fCO2. If we restrict to the period 2008-2014, trends become negative, i.e. -0.14 μmol/kg/yr for DIC and -0.8 μatm/yr for fCO2. We thus have to be careful when selecting (and interpreting) the periods. On the other hand, if we test the sensitivity of the trends for summer season in 2008-2017 (with or without the 2010 anomaly), results are basically the same. These specific results recall that fCO2 trends (and here also for DIC) are highly sensitive to the choice of starting and ending years as was illustrated by Mc Kinley et al (2011).

We also tested the impact of these NAO events (high and low) on the long-term trends, 1993-2017 for summer and 1994-2015 for winter. In that case, the results are more

robust. For example, for the northern boxes CDE the winter trends evaluated for years 1994-2015 were +0.6 μmol/kg/yr for DIC and +1.4 μatm/yr for fCO2. If we restrict to the period 1994-2014, trends are lower, i.e. +0.5 μmol/kg/yr for DIC and +1.3 μatm/yr for fCO2. In both cases, the DIC trend appears close to the increase due to anthropogenic uptake (estimated around +0.6 to +0.8 μmol/kg/yr in this region; see our response to other reviewer comments below). For summer, the trend in 1993-2017 with or without the 2010 NAO anomaly leads to the same results: +1 μmol/kg/yr for DIC and +2 μatm/yr for fCO2. In that case, the NAO has no effect on the trends. For the full period, the summer trends appear faster than derived from winter data and this needs to be clarified. This is also why it is important to separate the full period in 3 parts (as presented in the submitted paper) to better investigate the process involved, especially with the cooling and freshening observed after 2008. If the NAO events are relatively well characterized with our observations and could impact the trends when evaluated over 5-10 years, for long-term trends (24 years) this seems a secondary effect (as also suggested from long-term ocean simulations, Breeden and McKinley 2016).

As a final test, we investigated the SOCAT data (version V6, Bakker et al, 2016) in this region corresponding to the NAO events. Unfortunately, there is no fCO2 data for July-August 2010 that could be used to support the low DIC concentrations we observed in August 2010, but also in late June 2010 during the OVIDE-2010 cruise. However, for 2015 (high NAO), SOCAT data suggest relatively high fCO2 around 390-415 μatm in Jan-Feb 2015 in the NASPG, not far from our average value of around 400 μatm. A comparison with SOCAT fCO2 data for the full period would be interesting but this is beyond the scope of our analysis mainly based on DIC/TA observations. This would be a topic for another publication shared with SOCAT data providers in the north Atlantic.

Based on our results and sensitivity analysis described above, we will introduce the NAO events in the revision and discuss how NAO may be linked (or not) to the CO2 trends. We thank again Are Olsen to highlight this issue in his comment.

10. Finally, Metzl et al (2010) suggested deep mixing as the cause for the sharp win-

ter fCO2 trend in 2001-2008. Fröb et al (2019) demonstrated unequivocally that deep mixing leads to strong increases in winter fCO2 in NAO positive years because of intensified deep mixing (bringing remineralised DIC to the surface). In light of this, it is interesting that the smallest winter fCO2 trends from SURATLANT are observed in the period 2008-2017, as it is well known that this has been a period of rather large deep mixing compared to 2001-2008. I am not sure how this can be reconciled, i.e. the large fCO2 trend in the period of little deep mixing 2001-2008 vs the small fCO2 trend in the period of frequent deep mixing in 2008-2017. It seems worthwhile to delve into this. While doing so, keep in mind that deep-mixing events seemed (in Fröb et al., 2019) to cause year-to-year anomalies and not so much an anomalous trend. Therefore, I recommend to reconsider the use of three periods - the trend in each of these can be strongly affected by the fCO2 in the start and/or end year, which might just be an anomalous year. Therefore, the authors might be doing themselves a disfavor by sticking to the three periods, which are largely defined because of 'historical'/'traditional' reasons related to sampling and previous SURATLANT papers.

Response: Reviewer is partially correct. Not surprisingly, the trends for each period strongly depend on starting and ending points. This somehow recalls the sensitivity analysis presented by McKinley et al. for decadal trends (2011, their figure 2a). The 3 periods we selected were based on (i) available DIC and TA data, (ii) variability of SST and salinity (more specifically trends of SST and SSS anomalies as derived from monthly binned products, Reverdin et al 2018) and (iii) fCO2 (and pH) observed changes.

We also agree that deep-mixing events (for winter) or high/low productivity (in summer) would drive large anomalies for specific years, but might not impact much the multi-year trends. As a matter of fact, we identified some years with significant anomalies (e.g. summer 2010 or winter 2015 probably linked to NAO) that might change the trend if these events are or not taken into account (i.e. if these events were just not observed and missing in the time-series). We will discuss more clearly in the revised manuscript

that in winter 2015 (positive NAO), and because we have no winter observations after 2015, this specific year leads to a relatively large trend. If high DIC and fCO2 in winter 2015 are not taken into account the trends for winter 2008-2014 are significantly different. Interestingly, in a recent analysis based on SOCAT data, Denvil-Sommer et al (2019) suggests large variability in the NASPG (more specifically the NASPSS biome, Fig S5) for years 2001-2015, especially after 2013 (but with different responses between data-based methods). The observations that will be conducted during the next 5 years or so would support (or not) what we have learned from data presently available. On the opposite, if we filtered the summer 2010 anomaly, the trends in summer for 2008-2017 are almost the same. Finally, we note that if the trends are evaluated for the full period (1993-2017), the winter 2015 anomaly has less impact. In the revision, as suggested by reviewer, we propose to show first the long-term trend and then separate the period to better explain the drivers and contrasting response between region and periods. We will also investigate mixed-layer variability based on either in-situ data (ARGO) or models outputs.

Figure 3 (attached) show a link the same variability of temperature observed for two different data-set, in like to AMO. Regarding the depth of the mixing layer, the index presented in Figure 4 (very low frequency) does not seem conclusive, in contrast to the temperature. Thus, these results seem to support again that the NAO does not have too much impact on trends, which reinforces our previous conclusions.

Other comments:

“Page 2, line 4. Takahashi’s estimates of air-sea CO2 flux cannot be equated to anthropogenic CO2 flux.

Response: The reviewer is correct (same comment as reviewer 1). The sentence will be revised as: “The North Atlantic is one of the strongest ocean sinks for natural (Takahashi et al., 2009) and anthropogenic atmospheric CO2 (Sabine et al. 2004; Khatiwala et al., 2013).” At that stage in the introduction, we don’t really need to specify

number such as 0.27 PgC/yr (in our analysis we do not evaluate integrated flux over the domain).

“Page 2, line 6. I don’t think any has ascribed the variations in the North Atlantic CO2 sink to climate change. Careful with such statements.”

Response: Mc Kinley et al 2011 found that in the North Atlantic subtropical gyre, the increase of pCO2 by a warming trend is partially due to anthropogenic forcing (i.e. interpreted as climate change). For clarity, we will revise the following: However, repeated observations have shown important variations in this natural sink for anthropogenic CO2 in response to climate variability (Corbière et al., 2007; Metzl et al., 2010; McKinley et al., 2011; Landschützer et al., 2013).

“Page 2, line 21. “based on Total Alkalinity (TA) and Dissolved Inorganic Carbon (DIC) observations [. . .]”.

Response: will be corrected

“Page 3, line 7. ’which’, not ’witch’.”

Response: will be corrected

“Page 3, line 14-15. ’to this end’, not ’to this aim’.”

Response: will be corrected

“Page 3, line 15. ’Iceland’, not ’Island’.”

Response: will be corrected

“Page 3, line 17. These acronyms have already been defined.”

Response: will be corrected

“Page 4, equation 1. I think it would be useful to recap the accuracy of this relationship, and how well it works for the data that are presented here.”

Response: The TA/S relation was established by Reverdin et al (2018) and used here when TA data were not available in 1993-1997 and occasionally when TA data were flagged “dubious or bad”. For a regional view (as described in Reverdin et al), the TA/S relation was obtained for salinity above 34. These authors also note: “For the lower salinities found on the Newfoundland shelf, different sources of freshwater (from the Arctic or resulting from continental or sea ice melt inputs) contribute to deviations from the relation.” For the southern region (box A and box B for some periods when salinity is low), one should be careful when using this relation. This is also why in this paper, we did not describe the long-term trend in the southern region (box A) which would require a dedicated analysis. In addition, one cannot exclude the impact of blooms of coccolithophorids observed in the NASPG, as variability of such blooms may also impact on TA concentrations and thus deviate from the TA/S regional relation. Recently, Loveday and Smyth (2018) show significant differences of such blooms over decades in the North Atlantic. An interesting topic would be to re-explore TA distribution and TA/S relations for different periods. This is why we recall that a specific study of TA spatiotemporal distribution and drivers of TA variability should be performed, but beyond the scope of the present analysis. Details of the TA/S relation, including comparisons with other relations were presented in Reverdin et al., 2018, Appendix figure B1, B2, B3).

“Page 4. Atmospheric fCO2 calculation. What atmospheric pressure was used?”

Response: We used standard pressure (1 atm).

“Page 4, decomposition equation. As mentioned above, please use the method that explicitly accounts for the effects of dilution/concentration. Note also, that the equation as written is wrong. The dX/dt term should be dX/dz, where z is the driver in question. You further need to explain what values you used for these sensitivities, and how they were derived.”

Response: Correct. Equation will be rewritten.

“Page 5, line 5. How many times were data excluded from box B because of SSS being outside the 34-35 range?”

Response: Over 470 samples in Box-B (50-54N) for the full period with DIC data, 399 data were in the salinity range 34-35 (38 data excluded for S<34 and 33 data excluded for S>35).

“Page 5. Also, please consider the grouping of these regions for the trend analysis from Figure 2 SSS and TA, E and D appears quite similar while C and B both appear different. My sense this that combining E and D, while keeping C and B separate might be the best approach.”

Response: Trends and contributions have been tested accordingly (see table 1 and figure 5). Figure 5 show the decomposition of the trends in surface pH only (the same observation between boxes C, D-E and C-D-E is observed). Trends averaged one boxes C and D-E are overall similar to within the error bars. In all cases, they illustrate the large differences in the trends between the three different periods that match what we commented in the previous draft of the paper.

“Page 5, lines 10-19. Please collect this information in one section. Please also write that these are the reconstructed values in Fig. 3 (If I understand correctly the e.g Jan/March values adjusted to February are the ’reconstructed’ values in Fig 3).

Response: The reviewer is correct. For the trend analysis we standardized winter to February and summer to July. Reconstructed values were adjusted based on the mean seasonal cycle. Of course, when no observation is available for the previous or the next month, there is no reconstructed value (i.e. gaps in the time series for some years).

“Page 6, lines 2-3. As mentioned above I am not convinced that these are ’pluriannual’ trends. You might be doing yourself a disfavor by splitting the data into three time periods. It might make more sense to look at the timeseries as a whole and instead look at anomalies from the long-term trend. In particular this should be done from

2001 onwards. Consider to relate anomalies to mixed layer depths similar to Fröb et al. (2019)."

Response: In the revision, we propose to show first the long-term trend and then separate the period to better explain the drivers and contrasting response between region and periods. We will also explore mixed-layer depth as suggested.

"Page 6, lines 7 onwards. Here a panel showing air-sea fCO2 difference, as suggested above would help, as it will make the discussion more quantitative."

Response: Good point. We will add a separate figure to present air-sea dCO2 difference for the full period and seasons.

"Page 6, line 20. I think 'near-stagnation' is the wrong word here. In winter the increase appears significant, at 1 μatm/yr. Generally, please make the summary of the results quantative."

Response: This will be reformulated (depending on the new results based on revised calculations for the new Box definition).

"Page 7, line 11. Remove 'Thereafter'."

Response: Will be corrected.

"Page 7, line 15-16. Enclose 'much faster than the atmospheric signal', with commas and: 'suggest larger productivity in the beginning of the period than at the end'. BTW this can and should be checked with Chl a data."

Response: There is no Chl-a measurements in SURATLANT and Chl-a from remote sensing started in 1998. Unfortunately, we cannot explore a link between DIC and Chl-a trends for the period 1993-1997. From our results we can only suggest "an increase in productivity at the beginning of the period compared to the end" to explain why DIC was lower in 1993.

"Page 7, line 19, replace 'than' with 'to'."

Response: will be corrected

"Page 7, line 21-22. It is interesting that the slower increase in fCO2 is associated with strengthening of the winds and enhanced deep mixing. As noted above, Metzl et al., (2010) suggested deep mixing as the cause for the larger increase in the 2001-2008 period. Recently Fröb et al. (2019) found anomously high fCO2 during years of deep mixing. What is suggested here, is thus at odds with these papers. This needs to be explored or revised." Response: This is an interesting difference between the region further west investigated by Fröb et al. (2019) and the central gyre/Reykjanes Ridge region that is investigated here. Contrary to the western Irminger Sea, the area surveyed by SURATLANT does not encounter deep convection. Winter convection in the northern part of the SURATLANT area is limited to mode waters, such as the Reykjanes Ridge mode water (Thierry et al., 2007). These waters vary interannually, but not necessarily in phase with the deep convection of the western Irminger Sea. Furthermore, they only reach as deep as 400 to 650m depending on the year, and with a potential density close to 27.51. This is much shallower and less dense that the deep winter mixing in the western Irminger Sea) that was observed to reach the middle ocean layer (same density as Labrador Sea water). This depth-range difference can induce strong differences in the properties mixed to the surface. We would also like to comment that there is a difference between the effect of local winter mixing that will be strongly modulated with NAO (at least in central and northern part of SURATLANT) and the longer-term trends that are mostly explored here.

"Page 7, line 25. Set the '2' as subscript."

Response: will be corrected

Page 8, line 9 -10. The link between the trends in the carbon system and NAO+AMV that are described here are not backed up with any statistics. It comes across as very speculative. The statements need to be backed up with for example correlation analyses.

Response: we will study that.

“Page 8, line 11. Fröb et al (2018) shows in particular a large increase of anthropogenic DIC inventory in deep mixing years, and tendencies for a loss of natural carbon. Fröb et al. (2019) shows an outgassing of CO2 during deep mixing years. The coupling between the inventory changes and the variability in fluxes has yet to be made. Some discussion around this would be interesting.”

Response: we will try to add a discussion on this interesting issue, but this will also depend on revised calculations and if possible, link with MLD. To be done.

“Page 9, line 5. ’Makes it difficult to predict the evolution of CO2 uptake...’ I suggest to read the paper by Li et at 2016, on decadal predictions of North Atlantic CO2 uptake, this might provide some relevant information.”

Response: Regarding the prediction of carbon uptake, Li et al 2016 is a very good suggestion. The comparison with Li et al 2016 is however not clear as these authors indicate that (from simulations) when the mixing is enhanced in the western SPG this leads to more CO2 uptake, whereas weaker mixing in the eastern SPG leads to less CO2 uptake (their figure 1b and Supp Fig 1b). On this topic, we would also refer to Couldrey et al 2019.

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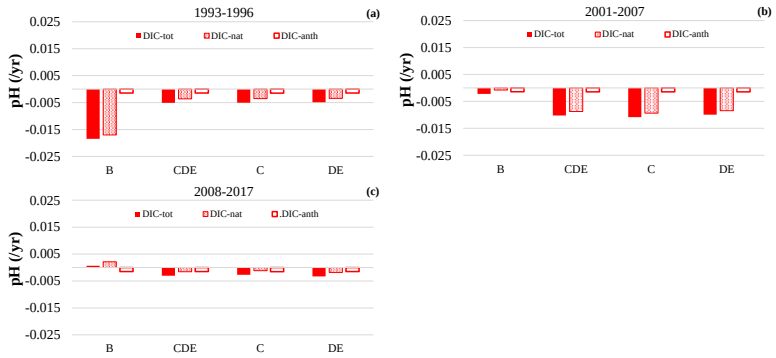

Figure 1. Effect of the changes in total (filled), anthropogenic (doted) and natural (empty) DIC to the trends in surface pH during summer for the three periods and different boxes (B, C, D-E and C-D-E).

Fig. 1.

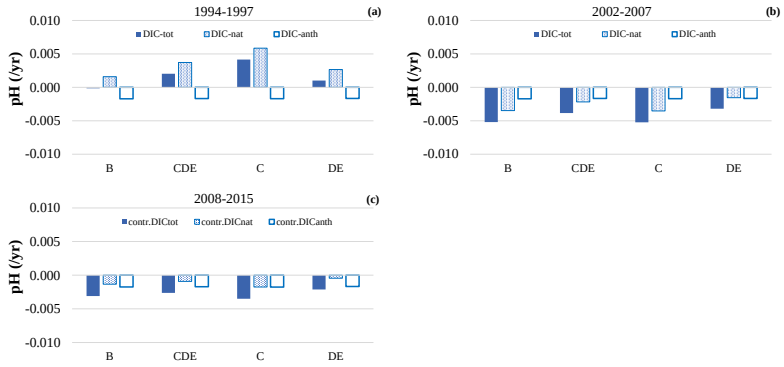

Figure 2. Effect of the changes in total (filled), anthropogenic (doted) and natural (empty) DIC to the trends in surface pH during winter for the three periods and different boxes (B, C, D-E and C-D-E).

Fig. 2.

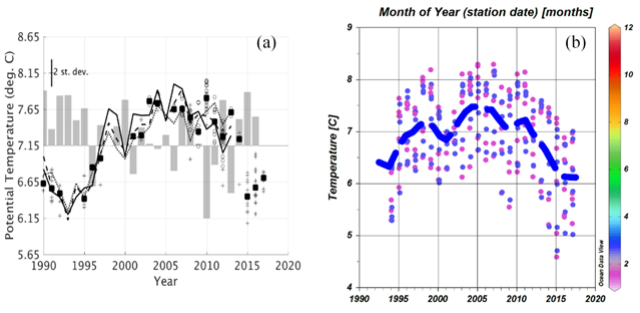

Figure 3. a) Temperature potential of Modal Water (Reykjanes Ride, cross), associated with NAO index (grey bar). Thierry et al. 2007; extended data, V. Thierry pers. comm. 19/06/2019. b) Sea Surface Temperature of C-D-E region (Jan-Fev, dot) estimate with AX2-binned (Reverdin et al. 2018b)

Fig. 3.

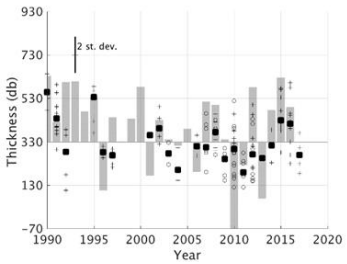

Figure 4. Index of variability of the thickness of the mode water layer (indicative of winter mixing layer (MLD), Reykjanes Rides. Thierry et al. 2007; extended data, V. Thierry pers. comm. 19/06/2019).

Fig. 4.

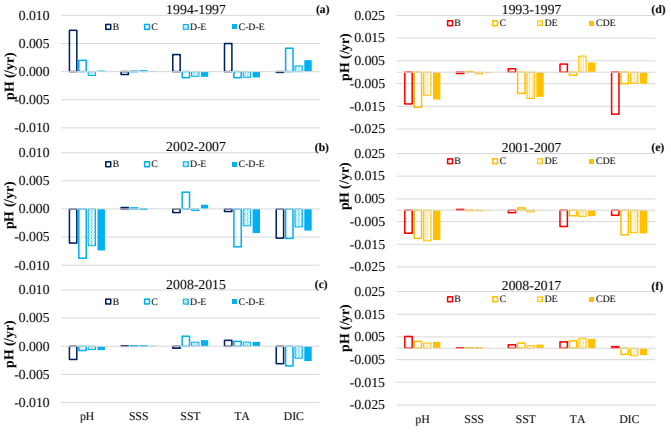

Figure 5. Decomposition of the trends in surface pH for winter (a,b,c) and summer (d,e,f). The effect of the changes in SSS, SST, TA and DIC is shown for the three periods.

Fig. 5.

Table 1. Trends (per year) evaluated from data presented in the manuscript (with some corrections). Here we show results for boxes B, C-D-E (as in the sub. paper) plus boxes C and D-E (as suggested by the reviewer). In bold are represented significant trends.

		box	SST (°C/yr)	SSS (/yr)	TA (µmol/kg/yr)	DIC (µmol/kg/yr)	pH (/yr)	fCO_2 (µatm/yr)	Ωca (/yr)	Ωar (/yr)
1993-1996	summer	B	-0.10 ±0.16	0.041 ±0.034	1.8 ±1.6	**9.0 ±2**	**-0.0143 ±0.0031**	**12.0 ±2.6**	**-0.107 ±0.015**	**-0.069 ±0.01**
		C.D.E	**0.68 ±0.06**	**0.032 ±0.028**	2.2 ±1.3	2.4 ±2.3	**-0.0123 ±0.0031**	**10.6 ±2.4**	0.003 ±0.025	0.005 ±0.016
		C	**0.58 ±0.06**	-0.020 ±0.015	-0.6 ±0.7	2.3 ±2	**-0.0155 ±0.003**	**13.6 ±2.6**	-0.038 ±0.023	-0.022 ±0.015
		D.E	**0.72 ±0.08**	0.059 ±0.04	3.6 ±1.9	2.4 ±3.5	**-0.0103 ±0.0046**	**8.8 ±3.5**	0.026 ±0.038	0.020 ±0.024
1994-1997	winter	B	-0.19 ±0.3	**0.045 ±0.032**	**2.2 ±1.5**	0.1 ±1.8	0.0075 ±0.0037	-7.2 ±3.6	0.028 ±0.022	0.017 ±0.015
		C.D.E	0.06 ±0.1	-0.009 ±0.015	**-0.5 ±0.6**	**-0.9 ±0.5**	0.0002 ±0.001	-0.4 ±1	0.006 ±0.01	0.004 ±0.006
		C	0.07 ±0.17	-0.003 ±0.021	-0.5 ±1	**-1.7 ±1.3**	**0.0021 ±0.0019**	**-2.3 ±1.9**	0.017 ±0.015	0.011 ±0.01
		D.E	0.05 ±0.1	**-0.012 ±0.013**	**-0.5 ±0.6**	**-0.4 ±0.5**	-0.0007 ±0.0011	0.5 ±1.2	0.000 ±0.01	0.000 ±0.007
2001-2007	summer	B	0.07 ±0.09	**-0.030 ±0.013**	**-4.3 ±0.7**	**0.2 ±1.2**	**-0.0105 ±0.0012**	**8.5 ±1**	**-0.068 ±0.015**	**-0.043 ±0.01**
		C. D. E	0.01 ±0.04	**0.017 ±0.008**	**-1.3 ±0.5**	**4.9 ±0.7**	**-0.0130 ±0.0013**	**11.5 ±1.1**	**-0.093 ±0.011**	**-0.059 ±0.007**
		C	-0.07 ±0.08	**0.012 ±0.009**	**-1.2 ±0.6**	**5.1 ±1**	**-0.0125 ±0.0015**	**11.2 ±1.3**	**-0.095 ±0.015**	**-0.061 ±0.01**
		D.E	**0.05 ±0.04**	**0.019 ±0.008**	**-1.4 ±0.7**	**4.8 ±0.9**	**-0.0133 ±0.0019**	**11.6 ±1.5**	**-0.092 ±0.015**	**-0.059 ±0.01**
2002-2007	winter	B	0.04 ±0.11	**-0.020 ±0.008**	-0.2 ±0.4	**2.1 ±1.2**	**-0.0061 ±0.0014**	**6.2 ±1.3**	**-0.031 ±0.018**	**-0.020 ±0.011**
		C. D. E	-0.05 ±0.07	-0.002 ±0.012	**-1.9 ±0.7**	**1.6 ±0.4**	**-0.0074 ±0.0009**	**7.0 ±0.9**	**-0.051 ±0.006**	**-0.032 ±0.004**
		C	**-0.19 ±0.09**	**-0.015 ±0.011**	**-2.9 ±0.9**	**2.2 ±0.5**	**-0.0088 ±0.0013**	**7.9 ±1.3**	**-0.074 ±0.01**	**-0.047 ±0.006**
		D.E	0.02 ±0.07	0.004 ±0.016	-1.3 ±0.8	**1.4 ±0.6**	**-0.0065 ±0.0011**	**6.3 ±1.2**	**-0.039 ±0.005**	**-0.024 ±0.003**
2008-2017	summer	B	-0.10 ±0.06	-0.012 ±0.009	**1.4 ±0.7**	-0.3 ±0.8	**0.0052 ±0.0013**	**-4.3 ±1.1**	**0.026 ±0.011**	**0.016 ±0.007**
		C. D. E	**-0.10 ±0.03**	**-0.012 ±0.004**	**2.0 ±0.3**	**1.4 ±0.5**	0.0027 ±0.001	-2.0 ±0.8	0.010 ±0.008	0.006 ±0.005
		C	**-0.14 ±0.05**	**-0.014 ±0.005**	**1.6 ±0.4**	**1.2 ±0.7**	0.0030 ±0.0012	-2.5 ±1.1	0.005 ±0.01	0.003 ±0.006
		D.E	**-0.07 ±0.05**	**-0.013 ±0.004**	**2.2 ±0.3**	**1.5 ±0.7**	0.0021 ±0.0014	-1.5 ±1.2	0.011 ±0.013	0.007 ±0.008
2008-2015	winter	B	0.02 ±0.07	-0.004 ±0.009	0.4 ±0.5	1.3 ±0.9	-0.0023 ±0.0012	2.2 ±1.2	-0.011 ±0.01	-0.007 ±0.006
		C. D. E	-0.07 ±0.04	-0.007 ±0.005	0.3 ±0.4	**1.1 ±0.4**	-0.0007 ±0.0005	0.6 ±0.5	-0.011 ±0.005	-0.007 ±0.003
		C	-0.11 ±0.05	-0.006 ±0.008	0.4 ±0.6	1.4 ±0.6	-0.0008 ±0.0008	0.6 ±0.8	-0.015 ±0.006	-0.010 ±0.004
		D.E	-0.05 ±0.03	**-0.007 ±0.003**	0.3 ±0.4	**0.9 ±0.5**	-0.0006 ±0.0006	0.5 ±0.6	**-0.008 ±0.005**	**-0.005 ±0.003**
1993-2017	summer	B	**0.05 ±0.02**	**-0.003 ±0.002**	0.1 ±0.2	**0.5 ±0.2**	**-0.0017 ±0.0004**	**1.5 ±0.3**	-0.005 ±0.003	-0.003 ±0.002
		C	0.02 ±0.01	**0.003 ±0.001**	**0.4 ±0.1**	**1.0 ±0.2**	**-0.0019 ±0.0003**	**1.7 ±0.3**	**-0.009 ±0.003**	**-0.005 ±0.002**
		D.E	**0.04 ±0.01**	**0.003 ±0.002**	**0.3 ±0.1**	**0.9 ±0.2**	**-0.0020 ±0.0004**	**1.9 ±0.3**	**-0.009 ±0.003**	**-0.005 ±0.002**
		C.D.E	**0.03 ±0.01**	**0.003 ±0.001**	**0.3 ±0.1**	**0.9 ±0.1**	**-0.0020 ±0.0003**	**1.8 ±0.2**	**-0.009 ±0.002**	**-0.005 ±0.001**
		B. C. D. E	**0.04 ±0.01**	0.000 ±0.002	**0.2 ±0.1**	**0.7 ±0.1**	**-0.0018 ±0.0002**	**1.7 ±0.2**	**-0.007 ±0.002**	**-0.004 ±0.001**
1993-2015	winter	B	**0.02 ±0.02**	0.003 ±0.002	0.1 ±0.1	**0.6 ±0.2**	**-0.0016 ±0.0003**	**1.5 ±0.3**	**-0.006 ±0.002**	**-0.004 ±0.001**
		C	-0.02 ±0.01	**0.005 ±0.002**	0.0 ±0.1	**0.9 ±0.1**	**-0.0018 ±0.0002**	**1.7 ±0.2**	**-0.012 ±0.002**	**-0.007 ±0.001**
		D.E	**0.02 ±0.01**	**0.005 ±0.001**	**0.2 ±0.1**	**0.5 ±0.1**	**-0.0013 ±0.0001**	**1.3 ±0.1**	**-0.005 ±0.001**	**-0.003 ±0.001**
		C.D.E	**0.01 ±0.01**	**0.005 ±0.001**	**0.1 ±0.1**	**0.6 ±0.1**	**-0.0014 ±0.0001**	**1.4 ±0.1**	**-0.007 ±0.001**	**-0.004 ±0.001**
		B. C. D. E	**0.01 ±0.01**	**0.004 ±0.001**	**0.1 ±0.1**	**0.6 ±0.1**	**-0.0015 ±0.0001**	**1.5 ±0.1**	**-0.007 ±0.001**	**-0.004 ±0.001**

Fig. 6.