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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 1 (anonymous):

We thank the reviewer for her/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.

"General Comments: This short manuscript describes the different trends in CO2 fu-

gacity (fCO2) in the North Atlantic Subpolar Gyre (50°N-64°N) considering three different periods based on the observations of the long-term monitoring program SURATLANT. Reverdin et al. (2018) have previously described these observations in an article in ESSD. Changes in pH and aragonite saturation, variables quasilinear dependent on CO2 fugacity, are also described. As shown in the manuscript itself (p.2 l.25) is this an extension of the previous analyses made by Corbière et al., 2007; Metzl et al., 2010, there was practically nothing new. The new data given in figure 3 only represents 1/3 of the complete serie. A large part of the data and the half of figures come from the article by Reverdin et al. (2018)".

Response:

The reviewer seems disappointed with the new data and results that would offer "practically nothing new" regarding previous work. In previous studies we used data from 1993-2003 (Corbière et al., 2007) to investigate for the first time the interannual to decadal changes observed in the NASPG with the SURATLANT data. This first analysis was then complemented by Metzl et al. (2010) with new data (2004-2008) that present significant variations compared to 1993-2003 with a winter focus in order to avoid to potential biases due to biological activity in the summer season. In particular, Metzl et al. (2010) indicate very rapid change of fCO2 (up to 7 μatm/yr in 2001-2008) highlighting "the need for continued longâĂŘterm sea surface ocean observations of carbon properties (DIC, TA and fCO2)". The SURATLANT data for 1993-2007 (or 1993-2010) were also associated with other observations to better evaluate pCO2 and air-sea flux variability at regional and larger scale in the North Atlantic (Schuster et al 2009; 2013; Watson et al., 2009; Mc Kinley et al 2011; Fay and Mc Kinley, 2013) or to describe the seasonality of sea surface δ13CDIC (Racapé et al., 2014).

In the present study, we added ten years of data (2008-2017) which offers new and complementary results. The recent paper by Reverdin et al (ESSD, 2018a) aimed at describing in detail the methods, accuracy and data for the full period 1993-2017 (with some data revisited and corrected), including some preliminary results of seasonal cy-

cles and long-term trends for a few properties; however, especially for ESSD Journal, Reverdin et al (2018a) did not investigate internal processes or external forcing resulting in the observed temporal changes of the properties (SST, SSS, nutrients, TA and DIC, δ18O and δ13C). This ESSD publication was accompanied with files of the data publicly available (at SeaNoe and OCADS). The present paper is aimed at better understanding the changes in the carbonate system over the whole period (1993-2017) and in different regions and periods, including summer-time (not evaluated in Metzl et al., 2010) as well as pH, Omega trends not described in previous work. Concerning the figures, as we used the same Box definition selected by Reverdin et al (2018a), we used the same figures for simplicity. In the revised manuscript we will prepare a new single figure (probably showing all data collection for DIC/TA, boxes boundaries and the main circulation.

"The description of the seasonal cycles shown by Reverdin et al. (2018) are different from those shown here without showing the reason for it. This puts in doubt that the interannual changes given for winter and summer cannot be affected by a poor quantification of the annual cycle in part due to low temporal coverage that can generate aliasing problems".

Response:

In Reverdin et al (2018a) the seasonal cycles were presented for all Boxes (including the southern box A) and for salinity normalized DIC and TA. Here, we have chosen to present in more detail the seasonality for boxes B, C, D, E and for all properties related to the carbonate system (including fCO2, pH and Omega not shown in Reverdin et al 2018a) and properties involved in the analysis of trends and drivers (SST, SSS, DIC, TA). For TA and DIC we show the mean seasonal cycle but not normalized as in Reverdin et al. Also, as we have no observations for all years in February (winter) or July (summer), the climatological seasonal cycles are used to group data of different months (such as JFM and JJA) in order to best estimate the seasonal trend (e.g. projecting observed January anomaly in February). Thus, we briefly introduce and de-

scribe the seasonality in section 4.1. The figure is required to indicate the seasonal variability that we have to correct for when combining data of different months. This is not so well known for the carbonate properties (model evaluations used for interannual to decadal analysis do not seem yet to correctly represent the processes involved in its seasonality, Pilcher et al., 2015, Mc Kinley et al., 2017). In addition, the seasonal cycles presented for the boxes B, C, D, and E contributed to decide to merge boxes C, D, E for the trend analysis. Of course, we fully agree with the reviewer that the seasonal cycle is not the main aim of the study, which is the estimation of the trends and the analysis of key drivers that is discussed in the core of the paper.

"The manuscript tries to describe the main drivers associated with CO2 chemistry using the same methodology shown in Metzl et al. (2010) for fCO2 and also García-Ibañez et al (2016) for pH. However, the relationship between the drivers and the main processes occurring in North Subpolar gyre is unfortunately very poorly developed. Partly because the authors seem to be unaware of key articles that have demonstrated the main patterns of variation linked to the NAO (Thomas et al. 2008, Keller et al. 2012, Schuster et al. 2013 and Pérez et al. 2013). In terms of acidification rates, the article by Garcia-Ibañez et al. (2016) is also ignored".

Thank you for recalling these references and suggestion to add a more in-depth discussion on the link with NAO (also suggested by reviewer 2).

Response about articles:

We were aware of these studies as they used in part SURATLANT data for 1993-2004 (or 1993-2005) to validate ocean models or reconstructed pCO2 fields (Thomas et al 2008; Ullman et al., 2009; Keller et al., 2012; Signorini et al., 2012; Rodenbeck et al., 2013, 2014). The SURATLANT data for 1993-2005 or 1993-2007 (fCO2 calculated from TA/DIC) were also used to complement fCO2 underway observations in the North Atlantic to better evaluate seasonal and decadal variations of pCO2 and air-sea CO2 fluxes in this region (Watson et al., 2009; Schuster et al 2009, 2013; Mc Kinley et

al. 2011; Fay and Mc Kinley, 2013). We will add some of these references (including Pérez et al., 2010, 2013; Garcia-Ibanez et al., 2016) in the introduction as well as in the discussion of the results we obtained in the NASPG for the period 1993-2017.

Response 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 the reviewer to highlight this issue in her/his comment.

“Very imprecise in the description of the processes involved, mixing anthropogenic factor with natural processes without a clear target. The manuscript cites the well-known key processes of water mass transformations. Besides, the drivers are disjointed way with any change with the water column chemistry. It continuously mixes the anthropogenic and non-anthropogenic changes or flows of CO2. Methodology poorly described. For interannual estimates the seasonal variability is not eliminated, which calls into question whether rates of change can be affected by changes in the annual cycle or biases in the relatively low frequency of observations”.

Response: Thank you to highlight the anthropogenic versus non-anthropogenic signal issue (same comment by reviewer 2, Are Olsen). 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.

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.

Specific remarks:

"P.1 Line 25 "As a consequence, the future evolution of air-sea CO2 fluxes, pH and the saturation state of surface waters with regards to aragonite and calcite remain highly uncertain in this region". This is a very weak point of the manuscript, since despite analyzing the drivers does not allow them to make future evolutions".

Response: we agree that this was not very clear. Although we found significant trends over 24 years, the variability we observed between different periods suggest that one cannot extrapolate the results in the future. This will be revised accordingly.

"P.2 Lines 3-4 "covering 5% of the global surface ocean, is responsible for 20% of the

oceanic uptake of anthropogenic CO2 (Khatiwala et al., 2013), with a mean annual air-sea CO2 flux estimated at 0.27 PgC/yr (Takahashi et al., 2009)." This is misleading. North Atlantic accumulates 20% anthropogenic CO2. The uptake can be produced in the subtropical regions and transported northwards. The rate given by Takahashi et al. 2009 included a big component of natural CO2 mostly due to the cooling of northward advected subtropical water and by biological carbon fixation in the subpolar gyre (Thomas et al. 2008; Perez et al. 2013)".

Response: Reviewer is correct. Sentence will be revised: "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 an integrated flux over the domain).

"P.5 Line 25 "The seasonal changes in fCO2 and pH are anticorrelated" This a consequence of the marine carbonic system when the alkalinity variability is so low as occurs in the North Atlantic".

Response: This is correct: Revised following: "As the alkalinity seasonality is low, the seasonal changes in fCO2 and pH are anti-correlated". "P.6 Line 27 "But it is due to a large increase in DIC rather than warming (Table 2), and as a consequence, it is accompanied by a rapid decrease in Omega. Why? Reduction of the vertical winter missing by cooling typical of the Irminger?"

Response: We are not sure to understand the reviewer question but maybe the reviewer was confused with numbers listed in the table (we apologize, there was a mistake in our original Table for CDE Omega-Ar trend, should read 0.005, not -0.069). In this section we show that in summer 1993-1997 (not in winter) the fCO2 increase is particularly fast (10 to 12 μatm/yr) in both regions, box B and CDE, but for a different reason: For CDE (north) this was related to a warming, whereas for box B (south) this was explained by DIC increase. Contrary to Box CDE, Omega decreases in box B

in summer 1993-1997. The interpretation is however limited to a short period 1993-1997. The DIC increase in Box B may be related to change in advection or productivity, but with data in hand we cannot separate these processes (and there were too few nutrients data for this period). We thus only conclude on the contrasting effect in the north (warming) and the south (DIC) to explain the rapid fCO2 trend in summer and the north/south trends in Omega. We will rewrite the section to clarify.

"P.6 Line 30 "a need to further investigate the drivers of TA variability, which seem partially decoupled from surface": However, Reverdin et al. 2018 show a perfect linear regression between TA and salinity, and the manuscript used this relationship to fill the gap of many observations without a second carbonic system variable".

Response: The reviewer is correct. We used the TA/S relation established by Reverdin et al (2018a) when TA data were not available in 1993-1997 and occasionally when TA data were flagged "doubt or bad" for other years. For a regional view (as described in Reverdin et al), the TA/S relation was first 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 requires a specific analysis. In addition, one cannot exclude the impact of blooms of coccolithophorids observed in the NASPG as variability of such blooms may also change TA concentrations with resulting deviations 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 spatio-temporal distribution and drivers of TA variability should be performed, but this is beyond the scope of the present analysis. We will rewrite the section.

"P.8 Line 20 "saturation with respect to calcium carbonate ()". This is a speculative addendum given the strong uncertainties and possible aliasing due to the seasonal coverture of the data".

Response: we agree that the future evolution of saturation is somewhat speculative. Based on the trends observed for different periods, one can get very different projections for future scenario and caution should be taken when using specific periods to extrapolate the results. Based on the observed trends we only suggest a range of years to reach undersaturation. We will revise this section.

"Fig 1 and Fig 2 come from Reverdin et al. 2018". Response: this is correct: as we used the same Box definition selected by Reverdin et al (2018), we used the same figures for simplicity. In the revised manuscript we will prepare a new figure (probably showing all data collection for DIC/TA, boxes boundaries and the main circulation).

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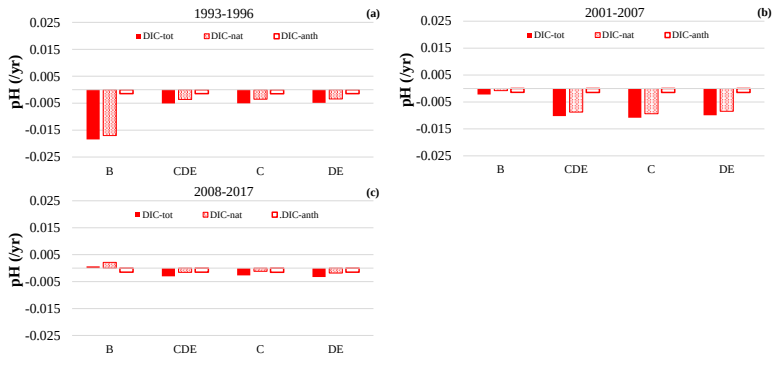

Figure 1. Effect of the changes in total (filled), anthropogenic (doted) and natural (empty) DIC to the trends in surface pH during summer for the three periods and different boxes (B, C, D-E and C-D-E).

Fig. 1.

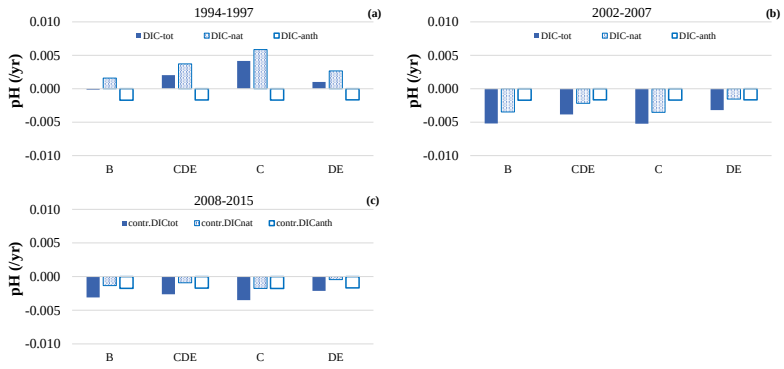

Figure 2. Effect of the changes in total (filled), anthropogenic (doted) and natural (empty) DIC to the trends in surface pH during winter for the three periods and different boxes (B, C, D-E and C-D-E).

Fig. 2.