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Key Points:

- We analyzed the DIC, C_{ant}, and DIC_{nat} budgets in the eastern SPNA in the 2000s from in situ data
- The DIC storage was the result of the anthropogenic perturbation
- The DIC_{nat} budget was in steady state. The biology drove the air-sea flux of natural CO_2

Supporting Information:

- Text S1
- Table S1

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Dissolved inorganic carbon budgets in the eastern subpolar North Atlantic in the 2000s from in situ data

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Abstract The subpolar North Atlantic (SPNA) is important in the global carbon cycle because of the deep water ventilation processes that lead to both high uptake of atmospheric CO_2 and large inventories of anthropogenic CO_2 (C_{ant}). Thus, it is crucial to understand its response to increasing anthropogenic pressures. In this work, the budgets of dissolved inorganic carbon (DIC), C_{ant} and natural DIC (DIC_{nat}) in the eastern SPNA in the 2000s, are jointly analyzed using in situ data. The DIC_{nat} budget is found to be in steady state, confirming a long-standing hypothesis from in situ data for the first time. The biological activity is driving the uptake of natural CO_2 from the atmosphere. The C_{ant} increase in the ocean is solely responsible of the DIC storage rate which is explained by advection of C_{ant} from the subtropics (65%) and C_{ant} air-sea flux (35%). These results demonstrate that the C_{ant} is accumulating in the SPNA without affecting the natural carbon cycle.

1. Introduction

The ocean has taken up approximately one third of the total human emitted CO_2, or anthropogenic CO_2 (C_{ant}), from the atmosphere [*Khatiwala et al.*, 2013], causing a continuous increase in dissolved inorganic carbon (DIC) concentration. Because this increase is very small compared to the DIC concentrations in the ocean ($<$0.1% per year), most work dealing with CO_2 storage rates quantify C_{ant} storage rate rather than DIC storage rate [*Sabine et al.*, 2004; *Khatiwala et al.*, 2013; *Pérez et al.*, 2013]. The highest C_{ant} inventories are found in the subpolar North Atlantic (SPNA) [*Sabine et al.*, 2004; *Khatiwala et al.*, 2013]. It is due to the combination of (i) a large amount of C_{ant} advected by the North Atlantic Current from subtropical to subpolar latitudes [*Pérez et al.*, 2013] and (ii) convective processes that cause an important injection of C_{ant} enriched surface water into the interior ocean [*Sabine et al.*, 2004].

It is generally assumed that the increase of CO_2 due to human activities (the anthropogenic perturbation) does not alter the natural carbon cycle [*Sarmiento et al.*, 1992]. This means that there is no variation in the natural DIC (DIC_{nat}) concentration in the ocean at relatively long timescales. Recently, some works have questioned this hypothesis [*Tanhua and Keeling*, 2012; *Tanhua et al.*, 2013]. The inherent difficulty is that DIC is subject to a strong natural variability at regional scale and/or short period of time, which could cause changes in the DIC inventory and mask any possible response to the anthropogenic perturbation. For example, *Corbière et al.* [2007] detected that DIC concentration in surface waters in the SPNA remained constant from 1993 to 2003; they interpreted this signal as a compensation of the DIC increase due to the anthropogenic perturbation, with the DIC decrease due to both the enhanced primary production and the larger arrival of tropical water poorer in DIC than SPNA water. The latter process was also proposed by *Thomas et al.* [2008] after analyzing ocean physical-biological numerical simulations. Alternatively, *Tanhua and Keeling* [2012] estimated the storage rates of DIC and DIC abio (their denotation for C_{ant}) in different regions of the Atlantic Ocean; for the eastern SPNA during the period 1982–2005, they detected a storage rate of DIC twice as large as the storage rate of DIC abio, but the uncertainties were too large to conclude whether the natural variability of the marine carbon cycle was causing a change in the DIC inventory.

Previous works compared the budgets of DIC, C_{ant}, and DIC_{nat} using in situ data in different regions of the Atlantic Ocean [*Holfort et al.*, 1998; *Roson et al.*, 2003; *Macdonald et al.*, 2003]; but none of them considered the biological activity in their budgets, even though it could be important for the DIC budget. Additionally, the outputs of ocean inversion methods also provide information for analyzing the DIC, C_{ant}, and DIC_{nat} budgets. The ocean inversion methods combine DIC observations with transport and mixing from ocean models,

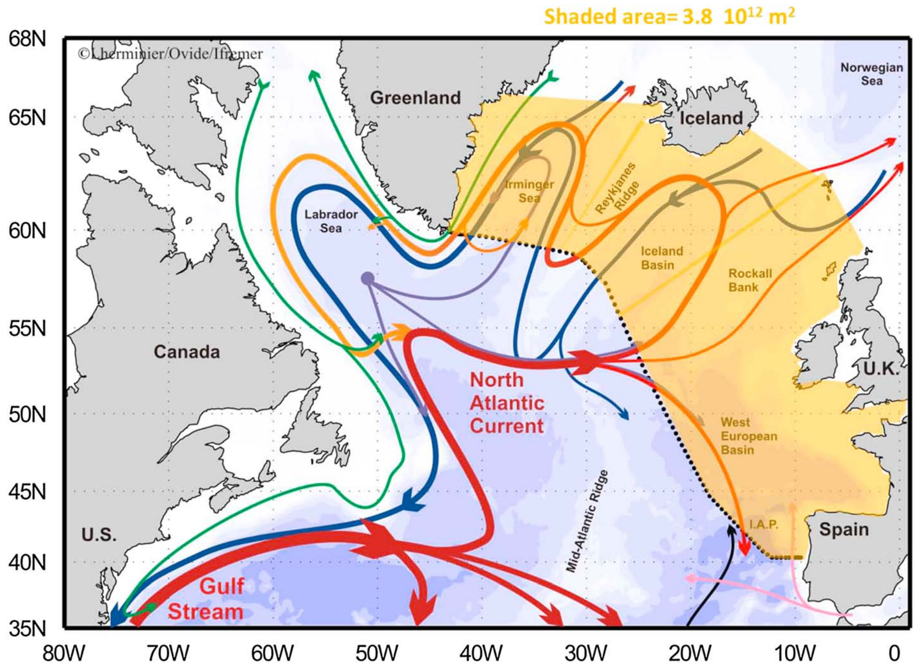

Figure 1. General circulation pattern in the subpolar North Atlantic. Dotted line indicates the location of the OVIDE section. Shaded yellow area indicates the surface of the eastern SPNA region considered in this study, which has a total area of $3.8 \cdot 10^{12}$ m^2.

assuming no sources or sinks in the ocean interior, allowing them to infer the air-sea fluxes for different regions of the ocean from the imbalance of the lateral advection [*Mikaloff Fletcher et al.*, 2006, 2007; *Gruber et al.*, 2009]. Nevertheless, some works, based on in situ estimates of the ocean circulation, have suggested that the lateral advection of C_{ant} is underestimated by ocean inverse models [*Pérez et al.*, 2013; *Zunino et al.*, 2015a]. So, if this is also true for DIC, we suspect that the air-sea fluxes inferred from ocean inverse methods may be biased.

Comparing DIC, C_{ant}, and DIC_{nat} budgets in the eastern SPNA using in situ data only, this study has two main objectives: first, quantify and evaluate the compatibility between the DIC and C_{ant} budgets in the eastern SPNA region over the period 2002–2010; second, validate the assumption that the ocean natural carbon cycle is still in steady state despite the anthropogenic perturbation. This work will also provide benchmarks from in situ data for the modeling community.

2. Data and Method

2.1. Data

The OVIDE section, from Greenland to Portugal (Figure 1), has been repeated biennially since summer 2002 (http://wwz.ifremer.fr/lpo/La-recherche/Projets-en-cours/OVIDE). In this work we analyzed data from five cruises during the spring-summer seasons of 2002, 2004, 2006, 2008, and 2010. DIC was calculated from pH and total alkalinity (A_T) with an accuracy of 4 μmol kg^{-1} following the recommendations and guidelines from *Velo et al.* [2010]. C_{ant} was determined applying the ϕCt° method [*Vázquez-Rodríguez et al.*, 2009a, 2009b] resulting in an error of ± 5.2 μmol kg^{-1}. This method assumes the following: (i) No changes in C_{ant} in the sample due to biological activities from the last contact with the atmosphere until the moment the sample was taken; for this purpose we used the relation between oxygen, A_T and nutrient concentrations to determine the natural DIC variation; (ii) no seasonal variability because all of the parameterizations were done using data from 100 to 200 m depth; and (iii) temporal variation of the CO_2 air-sea disequilibrium [*Matsumoto and Gruber*, 2005]. *Pérez et al.* [2010] already showed the C_{ant} distribution along the OVIDE section in 2002, 2004, and 2006. The DIC_{nat} concentration is estimated as the difference between DIC and C_{ant} concentrations.

The current velocities perpendicular to the OVIDE section were calculated by combining the geostrophic current with the velocities measured by the vessel-mounted acoustic Doppler current profilers using a generalized least squares inverse model [see *Lherminier et al.*, 2007, 2010; *Mercier et al.*, 2015].

Direct observations of the components of the CO_2 system and volume transport across the Greenland-Iceland-Scotland (G-I-S hereafter) sills are unfortunately not available for the same years they were measured at the OVIDE section. However, tracer concentrations and volume transports of water masses flowing over the sills are available in the literature [*Hansen et al.*, 2008; *Key et al.*, 2010; *Jeansson et al.*, 2011; *Pérez et al.*, 2013]. Table S1 in the supporting information summarizes the transports and water mass properties used in our computations, taken from *Pérez et al.* [2013] and *Jeansson et al.* [2011].

2.2. Methods

The volume of water enclosed between the OVIDE section and the G-I-S sills is considered here as the eastern SPNA box (Figure 1). The budget of C_{ant} in an oceanic region is the balance between lateral advection of C_{ant} due to ocean circulation, C_{ant} storage rate and C_{ant} flux through the air-sea interface. The terms of the C_{ant} budget are also included in the DIC budget. Additionally, the production or consumption of DIC due to biological activity should be included in the eastern SPNA DIC budget. The riverine input and the carbonate sedimentation could be other elements in the DIC budget. However, the river runoff is very small in our eastern SPNA box [*Berner and Berner*, 1987] allowing us to neglect the DIC riverine input in the DIC budget. Concerning the carbonate sedimentation, *Álvarez et al*. [2003] gave a value for it of 11 kmol s^{-1}(0.004 Pg C yr^{-1}) in the combined Arctic, Nordic Seas, and the eastern SPNA; this value is very small compared with the other terms of the DIC budget in the eastern SPNA, as shown in the results, and therefore neglected. The C_{ant} or DIC budget in the eastern SPNA can be written as,

$$S = A + F + B \tag{1}$$

where S, A, and F stand for storage rate, lateral advection, and air-sea flux, respectively, of either C_{ant} (S_{Cant}, A_{Cant}, and F_{Cant}) or DIC (S_{DIC}, A_{DIC}, and F_{CO2}), and B is the biological activity term. The latter is only considered in the DIC budget [*Sabine et al.*, 2004] and includes the primary production and the remineralization processes. As for the concentration, the terms of the budget of DIC_{nat} (S, A, F, and B) are calculated by differences between the terms in the DIC and C_{ant} budgets.

The inventories of DIC and C_{ant} from 2002 to 2010 are estimated using OVIDE data and the method developed by *Pérez et al*. [2010], recently improved by *Zunino et al*. [2015b]. The storage rates of DIC and C_{ant} are then computed by least squares regression from the slope of the inventory time series over 2002–2010 (Figure 2a). The storage rate errors are computed as the standard errors of the least squares fit.

The lateral advection of any property in the eastern SPNA is estimated as the balance between the property transport across the OVIDE section and the G-I-S sills. The method for calculating the transports of DIC and C_{ant} (T_{DIC} and T_{Cant}) across the OVIDE section and the G-I-S sills is explained in the supporting information.

Unlike the total air-sea CO_2 flux (F_{CO2}), the air-sea C_{ant} flux (F_{Cant}) cannot be directly estimated; therefore, it was inferred from the balance between S_{cant} and A_{cant} (equation (1), note that B equals zero in the C_{ant} budget). The F_{CO2} for the specific area of the east SPNA was estimated using the climatology of *Takahashi et al*. [2009] that is referenced to year 2000. In order to rescale F_{CO2} to year 2006, the surface seawater CO_2 fugacity (f_{CO2}) from *Takahashi et al*. [2009] was corrected to the year 2006 considering (i) a mean interannual increase in the oceanic pCO_2 of 1.75 µmol yr^{-1}, as estimated for the North Atlantic Ocean by *Takahashi et al*. [2009], and (ii) the temperature changes of the surface water by using the empirical temperature dependence of *Takahashi et al*. [1993]. Note that the F_{CO2} accounts for both the natural and the anthropogenic CO_2 fluxes across the air-sea interface.

3. Results

3.1. Storage Rates of DIC, C_{ant}, and DIC_{nat}

The DIC and C_{ant} storage rates estimated from the inventory time series (2002–2010) are 1.32 ± 0.30 mol C m^{-2} yr^{-1} (0.060 ± 0.014 Pg C yr^{-1}) and 1.21 ± 0.12 mol C m^{-2} yr^{-1}(0.055 ± 0.005 Pg C yr^{-1}),

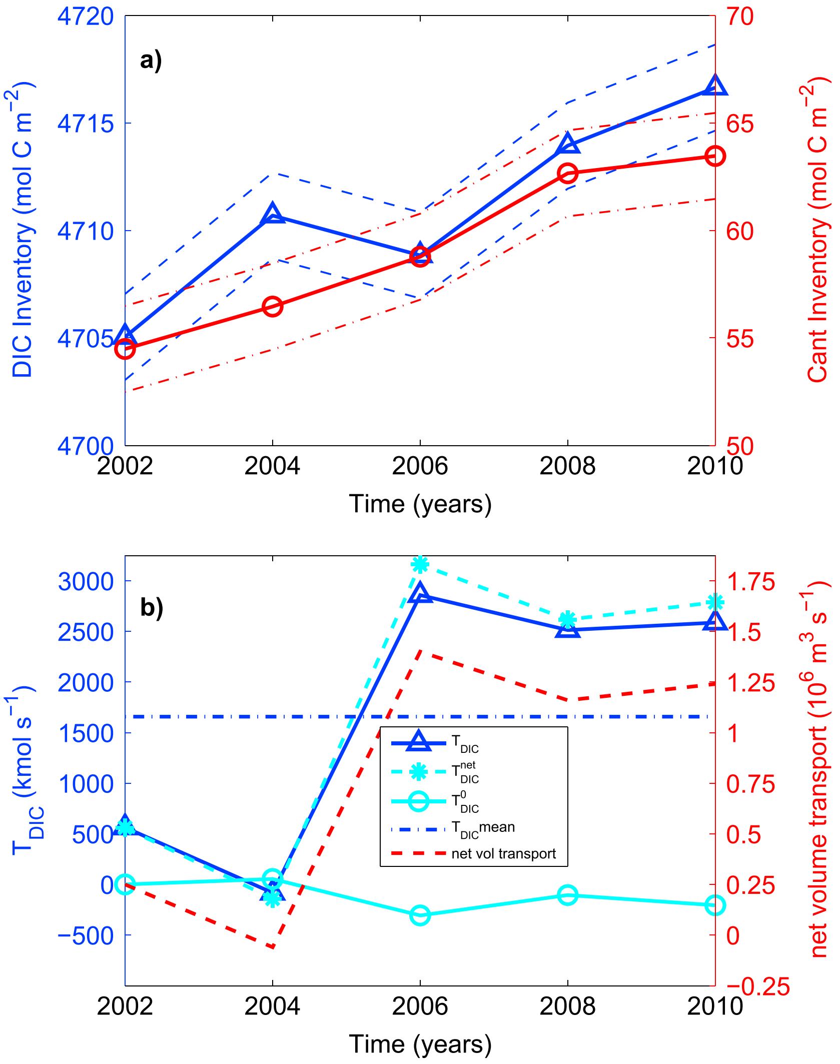

Figure 2. (a) Evolution of DIC inventory (blue line with triangles, left axis) and C_{ant} inventory (red line with circles, right axis) in the eastern SPNA. Discontinuous blue (red) lines are the DIC (C_{ant}) inventory error bars. (b) Left axes: time evolution of (1) DIC transport across the OVIDE section obtained from in situ data (T_{DIC}, blue line with triangle), (2) DIC transported by the net volume transport (T^{net}_{DIC}, cyan dashed line with stars), (3) DIC transported by the zero net volume transport (T^{0}_{DIC}, cyan dashed line with circles), and (4) mean value of DIC transport over 2002–2010 (dashed blue line). Red dashed line on the right axis is time evolution of net volume transport across the OVIDE section.

respectively. They are not statistically different. DIC and C_{ant} inventories are plotted as a function of time in Figure 2a. They both show a similar overall increase in time. The rise in DIC inventory is more variable than the increase in C_{ant} inventory, but the irregularities are not large enough to be interpreted as significant (the inventory error is 2 mol C m^{-2}). Subtracting S_{Cant} to S_{DIC}, we find that the storage rate of DIC_{nat} is not statistically different from zero. This result will be discussed in section 4.

3.2. Lateral Advection of DIC, C_{ant}, and DIC_{nat}

The temporal variability of T_{DIC} across the OVIDE section from 2002 to 2010 is shown in Figure 2b (left axis, blue line). The T_{DIC} is positive (northeastward) in all years except 2004. The interannual variability of T_{DIC} depends mainly on the variability of the circulation across the section since the interannual variability of DIC concentration is negligible in relation to its order of magnitude (~2100 μmol kg^{-1}). The transport of a tracer across a transoceanic section can be split in the component associated with the net transport of volume ($T_{\mathrm{vol}}^{\mathrm{net}}$) and a remaining component for which the net volume transport is zero [*Bacon*, 1997]. Following this decomposition, T_{DIC} can be written as

$$T_{\mathrm{DIC}} = T_{\mathrm{DIC}}^{\mathrm{net}} + T_{\mathrm{DIC}}^{0}, \tag{2}$$

with

$$T_{\mathrm{DIC}}^{\mathrm{net}} = T_{\mathrm{vol}}^{\mathrm{net}}\,\overline{[\mathrm{DIC}]}, \tag{3}$$

where T_{DIC}^{0} is the DIC transport at zero net volume transport and $\overline{[\mathrm{DIC}]}$ is the mean concentration of DIC across the section.

The variability of $T_{\mathrm{DIC}}^{\mathrm{net}}$ and T_{DIC}^{0} is shown in Figure 2b. We find that the T_{DIC} is mainly explained by $T_{\mathrm{DIC}}^{\mathrm{net}}$; therefore, the interannual variability of T_{DIC} depends mainly on the variability of the $T_{\mathrm{vol}}^{\mathrm{net}}$ ($r^2 = 0.99$). The 2002–2010 mean values of DIC and volume transport across the OVIDE section are $1688 \pm 602\ \mathrm{kmol\ s^{-1}}$ ($0.639 \pm 0.228\ \mathrm{Pg\ C\ yr^{-1}}$) and 0.8 ± 2 Sv ($\mathrm{Sv} = 10^6\ \mathrm{m^3\ s^{-1}}$), respectively.

The circulation across the G-I-S sills is less variable than across the OVIDE section because of bathymetry restrictions. The T_{DIC} across the G-I-S sills is estimated to be $1580 \pm 36\ \mathrm{kmol\ s^{-1}}$ using Table S1 data, with a net volume transport of 0.8 ± 2 Sv.

Important for the DIC budget is the convergence of DIC due to the lateral advection, e.g., the difference between the T_{DIC} across the OVIDE section and the G-I-S sills (while the volume in the region is conserved). The lateral advection causes a convergence of DIC amounting to $108 \pm 82\ \mathrm{kmol\ s^{-1}}$ ($0.041 \pm 0.031\ \mathrm{Pg\ yr^{-1}}$). The error of the convergence of DIC due to the lateral advection was estimated as indicated in equation (4), noting that because of the volume conservation, the variations in the net volume transport across OVIDE and the G-I-S sills are correlated.

$$\mathrm{error} = \sqrt{\left(\left(\overline{[\mathrm{DIC}]}^{\mathrm{Ov}} - \overline{[\mathrm{DIC}]}^{\mathrm{Sills}}\right) \times \mathrm{error}\ T_{\mathrm{vol}}^{\mathrm{net}}\right)^2 + \mathrm{error}\ {T_{\mathrm{DIC\ Ov}}^{0}}^2 + \mathrm{error}\ {T_{\mathrm{DIC\ Sills}}^{0}}^2}, \tag{4}$$

In equation (4), $\overline{[\mathrm{DIC}]}^{\mathrm{ov}}$ and $\overline{[\mathrm{DIC}]}^{\mathrm{Sills}}$ are the mean DIC concentrations at the OVIDE section and the G-I-S sills, respectively; error $T_{\mathrm{vol}}^{\mathrm{net}}$ is the error of net volume transport across both lateral boundaries (2 Sv) [*Lherminier et al.*, 2010; *Pérez et al.*, 2013]; error $T_{\mathrm{DIC\ Ov}}^{0}$ and error $T_{\mathrm{DIC\ Sills}}^{0}$ are the error of T_{DIC} associated with the zero net volume transports across the OVIDE section and the sills, respectively; the former was computed as $\frac{\mathrm{std}\left(T_{\mathrm{DIC\ Ov}}^{0}\right)}{\sqrt{n}}$ from the $n = 5$ repetitions of the OVIDE section and the latter was estimated by random perturbation of volume transports in Table S1.

Concerning the $\mathrm{C_{ant}}$ lateral advection, the 2002–2010 average of T_{Cant} across the OVIDE section is $248 \pm 35\ \mathrm{kmol\ s^{-1}}$ ($0.094 \pm 0.013\ \mathrm{Pg\ C\ yr^{-1}}$) and the T_{Cant} across the G-I-S sills normalized to 2006 is $162 \pm 17\ \mathrm{kmol\ s^{-1}}$($0.061 \pm 0.007\ \mathrm{Pg\ C\ yr^{-1}}$). The lateral advection causes a net input of $\mathrm{C_{ant}}$ of $86 \pm 21\ \mathrm{kmol\ s^{-1}}$ ($0.033 \pm 0.008\ \mathrm{Pg\ C\ yr^{-1}}$) in the eastern SPNA box. Note that $1\ \mathrm{Pg\ C\ yr^{-1}}$ equals $2642\ \mathrm{kmol\ s^{-1}}$.

Finally, we find that the A_{DIC} and A_{Cant}, $0.041 \pm 0.031\ \mathrm{Pg\ yr^{-1}}$ and $0.033 \pm 0.008\ \mathrm{Pg\ C\ yr^{-1}}$, respectively, are not statistically different. Therefore, the $\mathrm{DIC_{nat}}$ convergence due to the lateral advection, $0.008 \pm 0.032\ \mathrm{Pg\ yr^{-1}}$, is not statistically different from zero. Consequently, the DIC convergence due to the lateral advection in the eastern SPNA is exclusively anthropogenic.

3.3. Air-Sea $\mathrm{CO_2}$ Flux

The F_{CO2} over the eastern SPNA was estimated at $0.062 \pm 0.02\ \mathrm{Pg\ C\ yr^{-1}}$ using *Takahashi et al.* [2009] climatology rescaled to 2006. Positive F_{CO2} means that the ocean is taking up $\mathrm{CO_2}$ from the atmosphere. The total area of the eastern SPNA is $3.8 \cdot 10^{12}\ \mathrm{m^2}$. *Landschützer et al.* [2013] estimated air-sea $\mathrm{CO_2}$ fluxes for the Atlantic Ocean using data from 1998 to 2007 from the Surface Ocean $\mathrm{CO_2}$ Atlas (www.socat.info) database. They found good agreement between the *Takahashi et al.* [2009] climatology and their estimates over the east SPNA region, all referenced to year 2000. Their findings justify the use of Takahashi climatology for the computation of air-sea $\mathrm{CO_2}$ fluxes in our region during the period 2002–2010.

Table 1. Estimates of the Elements of the DIC, C_{ant}, and DIC_{nat} Budgets in the Eastern SPNA Box[a]

	DIC (Pg C yr^{-1})	C_{ant} (Pg C yr^{-1})	DIC_{nat} (Pg C yr^{-1})
Storage rate (*S*)	0.060 ± 0.014	0.055 ± 0.005	*0.005 ± 0.015*
Lateral advection (*A*)	0.041 ± 0.031	0.033 ± 0.008	*0.008 ± 0.032*
Air-sea flux (*F*)	0.062 ± 0.020	*0.022 ± 0.009*	*0.040 ± 0.022*
Biological activity (*B*)	*−0.043 ± 0.039*	-----------	*−0.043 ± 0.039*

[a] Deduced values from the other elements of the DIC or C_{ant} budget are represented in italic font.

3.4. Closing the Budgets of DIC, C_{ant}, and DIC_{nat} in the Eastern SPNA

Table 1 summarizes our estimates of S_{DIC}, A_{DICv} and F_{CO2} for the 2002–2010 period. Using these estimates and applying a simple box model, we infer a biological DIC consumption of 0.043 ± 0.039 Pg C yr^{-1}, accounting for primary production and remineralization processes. Concerning the C_{ant} budget, we inferred from the imbalance between S_{Cant} and A_{Cant} that F_{Cant} equals 0.022 ± 0.009 Pg C yr^{-1} (Table 1). Finally, for the budget of DIC_{nat}: (i) the flux of natural CO_2 is 0.040 ± 0.022 Pg C yr^{-1}, deduced from the difference between F_{CO2} and F_{Cant}, (ii) the storage rate and the lateral advection of DIC_{nat} are not statistically different from zero, and (iii) the biological activity inferred from the DIC budget is −0.043 ± 0.039 Pg C yr^{-1}. From this budget we find that the biological activity consuming DIC is in balance with the air-sea flux of natural CO_2.

4. Discussion and Conclusions

Our estimate of the DIC storage rate for the eastern SPNA, 1.32 ± 0.30 mol C m^{-2} yr^{-1} (0.060 ± 0.014 Pg C yr^{-1}), is larger but not statistically different from the 0.91 ± 0.28 mol C m^{-2} yr^{-1} computed by *Tanhua and Keeling* [2012] for a similar area and the period 1995–2005. Concerning the S_{Cant}, our estimate of 1.21 ± 0.12 mol C m^{-2} yr^{-1} (0.055 ± 0.005 Pg C yr^{-1}) is statistically larger than *Tanhua and Keeling* [2012]'s estimate, 0.40 ± 0.33 mol C m^{-2} yr^{-1} (0.44 ± 0.36 mol C m^{-2} yr^{-1} when referenced to 2006). The fact that *Tanhua and Keeling* [2012] computed the inventories only for the upper 2000 m of the water column may explain part of this difference. Our result of S_{Cant} is also larger than the estimate of *Pérez et al*. [2013] using 2002–2006 OVIDE data (0.027 ± 0.004 Pg C yr^{-1} when referenced to 2006). Note that the S_{Cant} computed here for the 2000s decade is still lower than the S_{Cant} computed for 1997 by *Pérez et al*. [2013], 0.083 ± 0.004 Pg C yr^{-1} (0.100 ± 0.008 Pg C yr^{-1} when rescaled to 2006). On the one hand, *Pérez et al*. [2013] suggested a relationship between the S_{Cant} and the intensity of MOC in the SPNA. On the other hand, *Mercier et al*. [2015] observed an increase in the MOC intensity during the second half of the 2000s decade, yet it remains less intense than at the beginning of the 1990s. Consequently, the variability in the MOC intensity could explain the highest S_{Cant} obtained for 1997 and the higher S_{Cant} obtained for the period 2002–2010 than for previous periods (1995–2005 and 2002–2006). Our results reinforce the Pérez et al.'s hypothesis about the relationship between C_{ant} storage rate and MOC intensity in the SPNA.

Comparing S_{DIC} and S_{Cant}, 0.060 ± 0.014 Pg C yr^{-1} and 0.055 ± 0.005 Pg C yr^{-1}, respectively, we find that they are not statistically different. Therefore, our results suggest that over 2002–2010, DIC inventory in the east SPNA increased because of the anthropogenic perturbation, and the variability of the DIC_{nat} had a negligible contribution.

The variability of T_{DIC} across the OVIDE section depends mainly on the variability of the net volume transport. Conversely, the variability of net volume transport causes a minor variability in the T_{Cant} that depends mainly on the variability of the MOC intensity [*Zunino et al*., 2014]. The variability of T_{DIC} and T_{Cant} are controlled by the variability of different components of the circulation because the vertical gradients of DIC and C_{ant} are different. Specifically, the upper and lower branches of the MOC transport almost the same quantity of DIC northeastward and southwestward, respectively, since the DIC concentration minimally varies with depth (roughly 4%). By contrast, the upper branch of the MOC brings much more C_{ant} northeastward than its lower branch does southwestward because the surface waters are enriched in C_{ant} by a factor of 2 or more compared to deep waters. The T_{DIC} is an order of magnitude larger than T_{Cant} since DIC concentration is much larger than C_{ant} concentration (e.g., DIC and C_{ant} concentration averages in 2010 at the OVIDE section are 2161 μmol kg^{-1} and 28 μmol kg^{-1}, respectively). In spite of these differences between T_{DIC} and T_{Cant}, the convergences of DIC and C_{ant} due to the lateral advection are not statistically different: 0.041 ± 0.031 and 0.036 ± 0.008 Pg yr^{-1}, respectively. Consequently, we found that the convergence of DIC_{nat} due to lateral advection is not significantly different from zero. This means that the convergence of DIC in the eastern SPNA resulting from the lateral advection is almost entirely anthropogenic.

The air-sea flux, F_{CO2}, was estimated at 0.062 ± 0.02 Pg C yr^{-1}. It is smaller than the 0.09 Pg C yr^{-1} given in *Pérez et al.* [2013] for the same region. This difference is because their value was based on the F_{CO2} given in *Takahashi et al.* [2009] for latitudes > 50°N in the Atlantic Ocean, which include areas with stronger F_{CO2} than the eastern SPNA. The F_{Cant} was deduced from the C_{ant} budget at 0.022 ± 0.009 Pg C yr^{-1}. Considering our estimates of F_{CO2} and F_{Cant} (Table 1), we estimated the air-sea flux of natural CO_2 to be 0.040 ± 0.022 Pg C yr^{-1}. This result is in agreement with the 0.042 Pg C yr^{-1} computed from the natural CO_2 fluxes for the North Atlantic high latitudes by *Mikaloff Fletcher et al.* [2007] using ocean inverse methods. Our results also confirm those obtained by *Pérez et al.* [2013] who demonstrated that the natural component dominates the CO_2 fluxes in the SPNA.

The biological consumption/production of DIC inferred from the other terms of the DIC budget (Table 1) gives -0.043 ± 0.039 Pg C yr^{-1}. This is in agreement with the estimate of *Maze et al.* [2012] obtained from nutrient and oxygen budgets for the same region: -0.040 ± 0.023 Pg C yr^{-1}. Note that their value comes from the sum of their estimates of biological consumption in their Irminger and northeastern European Basin boxes, -0.019 ± 0.016 Pg C yr^{-1} and -0.021 ± 0.016 Pg C yr^{-1}, respectively. Using this estimate of biological activity in the budget of natural CO_2, we find that it is in balance with the air-sea transfer of natural CO_2 that was deduced from the $F_{CO2} - F_{Cant}$ difference. Because the lateral advection and the storage rate of natural CO_2 are not significantly different from zero, our result suggests that the biological activity drives the air-sea flux of natural CO_2. *Körtzinger et al.* [2008] and *Takahashi et al.* [2002] found that the biological carbon drawdown during spring/summer mediates the CO_2 sink in the Labrador Sea and in the 40°N–60°N region, respectively. Here we show that this balance concerns only the natural CO_2 cycle, without any noticeable influence of the anthropogenic perturbation. Finally, our study uses in situ data to show that the anthropogenic perturbation does not affect the cycle of natural CO_2 and supports the general assumption of a steady state of the carbon cycle [*Sarmiento et al.*, 1992] at regional scale.

The ocean inversion methods assume the steady state hypothesis. However, these methods do not include any sources or sinks in the interior ocean; by contrast, in the present study, we find that the biological activity acts as a DIC sink in the eastern SPNA. *Mikaloff Fletcher et al.* [2007] and *Gruber et al.* [2009], using ocean inverse methods, found a net convergence of DIC_{nat} in the SPNA due to the lateral advection, which they balanced with the uptake of natural CO_2 from the atmosphere without considering biological activity. Surprisingly, their estimates of natural air-sea CO_2 flux are in agreement with ours. Therefore, we suspect that for this specific region, the ocean inverse methods present deficiencies in the estimation of the divergence/convergence of DIC forms, in agreement with *Pérez et al.* [2013], likely due to an improper representation of the ocean circulation [*Zunino et al.*, 2015a]. While the ocean inversion methods are tremendously important for covering areas where in situ data are not available, the analysis of budgets of DIC (natural, anthropogenic, and contemporary) using only in situ data are needed to provide benchmarks for validation of ocean inversion methods as well as for specifying constraints at regional scale.

Though some previous works have calculated DIC budgets in the ocean based on in situ data, the comparison of their results with our results presents some difficulties. The DIC and C_{ant} budgets in the North Atlantic and Arctic Ocean (from 24°N to the Bering Strait) have been evaluated by *Roson et al.* [2003] and *Macdonald et al.* [2003] using in situ data. Their results are not directly comparable to our results since theirs dealt with subtropical, subpolar, and polar areas combined, despite those areas having differing heterotrophic/autotrophic activity and/or differing natural-anthropogenic contributions to the CO_2 air-sea fluxes. *Jeansson et al.* [2011] evaluated the DIC and C_{ant} budget in the Nordic Sea. They closed the DIC budget considering the C_{ant} storage rate in the DIC balance and inferred the air-sea CO_2 flux in the Nordic Seas to be 0.19 ± 0.06 Pg C yr^{-1}, comparable to the 0.11 ± 0.06 Pg C yr^{-1} estimated using the *Takahashi et al.* [2009] climatology. *Jeansson et al.* [2011] concluded that there is a negligible uptake of C_{ant} from the atmosphere in the Nordic Seas. None of these studies included the biological activity for closing the DIC budget. *Álvarez et al.* [2003] analyzed the DIC, C_{ant}, and DIC_{nat} budgets in a region accounting for the Arctic Ocean and the eastern SPNA region, taking into account the biological activity. Nevertheless, the comparison with our results is not possible for several reasons. First, the region analyzed is not exactly the same. Second, it has been demonstrated that the velocity field used in their work for the FOUREX section, which results in a southward net transport (−0.4 Sv), could be improved, particularly in the Irminger Sea [*Lherminier et al.*, 2007]. Finally, they considered a DIC biological production computed from an estimate of nitrate biological production [*Álvarez et al.*, 2002], which also depended on the velocity field via the nitrate budget.

We are aware that some of the elements of the DIC budget can be seasonally biased since some of the data analyzed here were measured in spring-summer time. However, the seasonal variability does not affect either the S_{DIC} estimate, since it was computed as the time derivative of data measured approximately at the same period of the year, or the F_{CO2}, since we used the annual accumulated F_{CO2} to close our DIC budget. Concerning the lateral advection, the seasonal variability of T_{DIC} may depend on the seasonal variability of both the DIC concentration and the circulation. The DIC concentration fluctuates seasonally due to the primary production (40–80 μmol kg^{-1}) [*Corbière et al.*, 2007] in the first 100 m of the water column. We calculated the T_{DIC} error associated with these DIC changes as the standard deviation of T_{DIC} obtained from DIC surface concentrations that were randomly perturbed between 40 and 80 μmol kg^{-1}. This error, 0.015 Pg C yr^{-1}, is smaller than the T_{DIC} error. Furthermore, the DIC concentration variability may affect the DIC transports at both the OVIDE section and the sills in the same way, implying that the seasonal cycle of DIC concentration may not result in a seasonal bias in the net input of DIC. However, there is little information on the seasonal variability of the circulation across both lateral limits of the eastern SPNA. Concerning the variability of the yet unknown net volume transport, we suspect that it may not affect the DIC budget, since the net volume transports across both boundaries are compensated. However, a seasonal variability of 10% has been identified for the East Greenland Current [*Daniault et al.*, 2011] and for the MOC intensity across the OVIDE section [*Mercier et al.*, 2015], which could affect the zero net transport of DIC. This possible variability of the circulation results in a very small T_{DIC} variability (11 kmol s^{-1} or 0.004 Pg C yr^{-1}) since T_{DIC} mainly depends on the variability of the net volume transport (see Figure 2b). Consequently, we conclude the DIC budget presented in this work is not significantly biased by the seasonal cycle.

Summarizing, we present in this work the budgets of DIC, C_{ant}, and DIC_{nat} in the eastern SPNA using in situ data. Comparing them we find that the anthropogenic perturbation appears as a DIC accumulation in the DIC budget which is explained by the C_{ant} lateral advection (65%) and the air-sea flux of C_{ant} (35%). Further, we find there is a sink in the DIC budget due to the biological activity, which is in balance with the air-sea flux of natural CO_2. Our results verify for the first time the generally accepted assumption that the CO_2 increase in the ocean due to anthropogenic activities does not affect the natural carbon cycle. These results will be important for the modeling community, acting as benchmarks for improving model configuration, and ultimately, model forecasts.

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