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## RESEARCH LETTER

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## Key Points:

- We analyzed the DIC,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  budgets in the eastern SPNA in the 2000s from in situ data
- The DIC storage was the result of the anthropogenic perturbation
- The  $\text{DIC}_{\text{nat}}$  budget was in steady state. The biology drove the air-sea flux of natural  $\text{CO}_2$

## 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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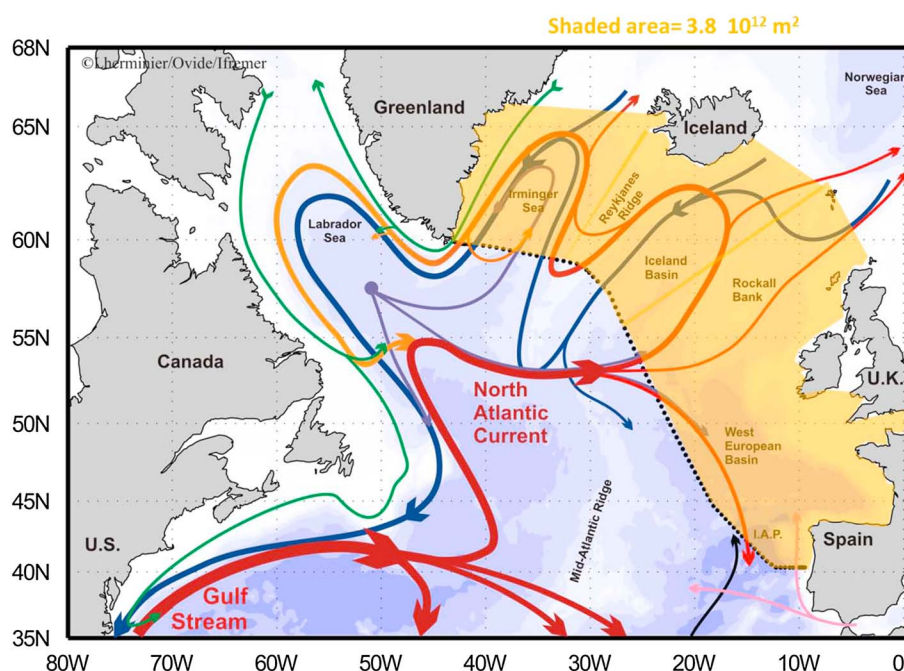
**Abstract** The subpolar North Atlantic (SPNA) is important in the global carbon cycle because of the deep water ventilation processes that lead to both high uptake of atmospheric  $\text{CO}_2$  and large inventories of anthropogenic  $\text{CO}_2$  ( $C_{\text{ant}}$ ). Thus, it is crucial to understand its response to increasing anthropogenic pressures. In this work, the budgets of dissolved inorganic carbon (DIC),  $C_{\text{ant}}$  and natural DIC ( $\text{DIC}_{\text{nat}}$ ) in the eastern SPNA in the 2000s, are jointly analyzed using in situ data. The  $\text{DIC}_{\text{nat}}$  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  $\text{CO}_2$  from the atmosphere. The  $C_{\text{ant}}$  increase in the ocean is solely responsible of the DIC storage rate which is explained by advection of  $C_{\text{ant}}$  from the subtropics (65%) and  $C_{\text{ant}}$  air-sea flux (35%). These results demonstrate that the  $C_{\text{ant}}$  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  $\text{CO}_2$ , or anthropogenic  $\text{CO}_2$  ( $C_{\text{ant}}$ ), from the atmosphere [Khaliwala 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  $\text{CO}_2$  storage rates quantify  $C_{\text{ant}}$  storage rate rather than DIC storage rate [Sabine et al., 2004; Khaliwala et al., 2013; Pérez et al., 2013]. The highest  $C_{\text{ant}}$  inventories are found in the subpolar North Atlantic (SPNA) [Sabine et al., 2004; Khaliwala et al., 2013]. It is due to the combination of (i) a large amount of  $C_{\text{ant}}$  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  $C_{\text{ant}}$  enriched surface water into the interior ocean [Sabine et al., 2004].

It is generally assumed that the increase of  $\text{CO}_2$  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 ( $\text{DIC}_{\text{nat}}$ ) 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  $C_{\text{ant}}$ ) 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,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  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,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  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  $3.8 \cdot 10^{12} \text{ m}^2$ .

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  $C_{\text{ant}}$  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,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  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  $C_{\text{ant}}$  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://wwwz.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  $4 \mu\text{mol kg}^{-1}$  following the recommendations and guidelines from Velo *et al.* [2010].  $C_{\text{ant}}$  was determined applying the  $\phi C_t^0$  method [Vázquez-Rodríguez *et al.*, 2009a, 2009b] resulting in an error of  $\pm 5.2 \mu\text{mol kg}^{-1}$ . This method assumes the following: (i) No changes in  $C_{\text{ant}}$  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  $\text{CO}_2$  air-sea disequilibrium [Matsumoto and Gruber, 2005]. Pérez *et al.* [2010] already showed the  $C_{\text{ant}}$  distribution along the OVIDE section in 2002, 2004, and 2006. The  $\text{DIC}_{\text{nat}}$  concentration is estimated as the difference between DIC and  $C_{\text{ant}}$  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<sub>2</sub> 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  $C_{\text{ant}}$  in an oceanic region is the balance between lateral advection of  $C_{\text{ant}}$  due to ocean circulation,  $C_{\text{ant}}$  storage rate and  $C_{\text{ant}}$  flux through the air-sea interface. The terms of the  $C_{\text{ant}}$  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  $11 \text{ kmol s}^{-1}$  ( $0.004 \text{ Pg C yr}^{-1}$ ) 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  $C_{\text{ant}}$  or DIC budget in the eastern SPNA can be written as,

$$S = A + F + B \quad (1)$$

where  $S$ ,  $A$ , and  $F$  stand for storage rate, lateral advection, and air-sea flux, respectively, of either  $C_{\text{ant}}$  ( $S_{\text{Cant}}$ ,  $A_{\text{Cant}}$ , and  $F_{\text{Cant}}$ ) or DIC ( $S_{\text{DIC}}$ ,  $A_{\text{DIC}}$ , and  $F_{\text{CO}_2}$ ), 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<sub>nat</sub> ( $S$ ,  $A$ ,  $F$ , and  $B$ ) are calculated by differences between the terms in the DIC and  $C_{\text{ant}}$  budgets.

The inventories of DIC and  $C_{\text{ant}}$  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  $C_{\text{ant}}$  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.

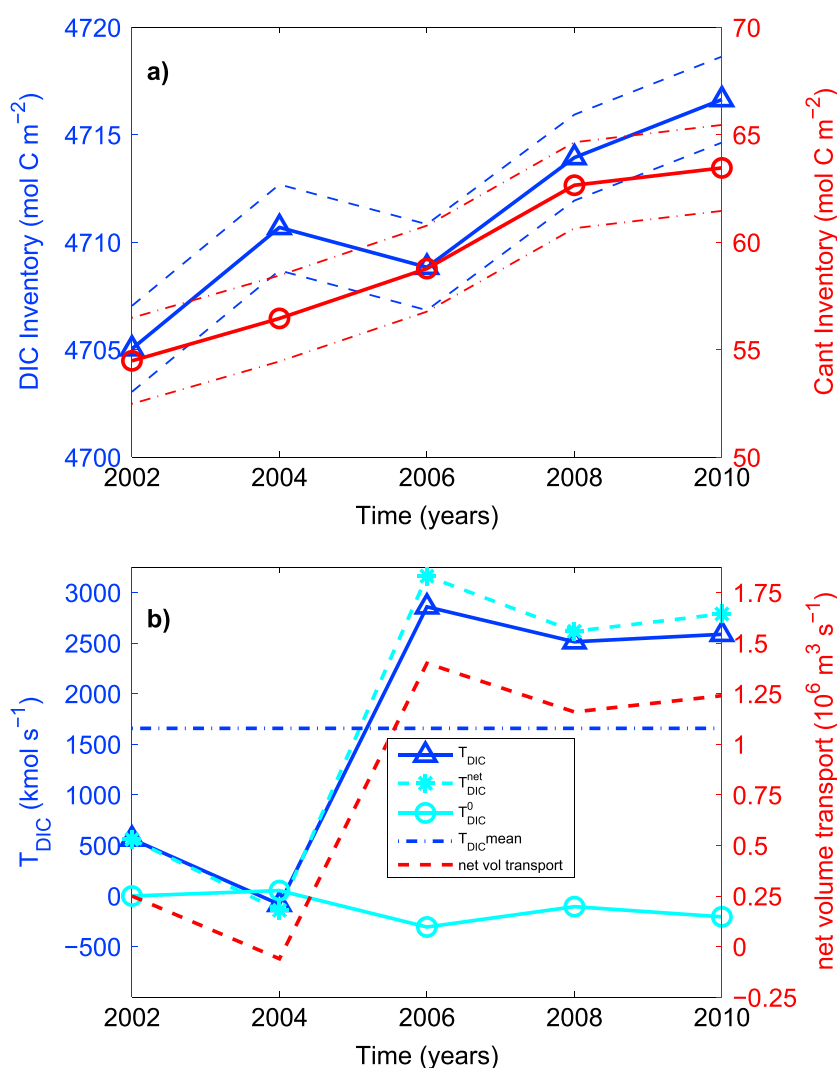
The lateral advection of any property in the eastern SPNA is estimated as the balance between the property transport across the OVIDE section and the G-I-S sills. The method for calculating the transports of DIC and  $C_{\text{ant}}$  ( $T_{\text{DIC}}$  and  $T_{\text{Cant}}$ ) across the OVIDE section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

Unlike the total air-sea CO<sub>2</sub> flux ( $F_{\text{CO}_2}$ ), the air-sea  $C_{\text{ant}}$  flux ( $F_{\text{Cant}}$ ) cannot be directly estimated; therefore, it was inferred from the balance between  $S_{\text{cant}}$  and  $A_{\text{cant}}$  (equation (1), note that  $B$  equals zero in the  $C_{\text{ant}}$  budget). The  $F_{\text{CO}_2}$  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  $F_{\text{CO}_2}$  to year 2006, the surface seawater CO<sub>2</sub> fugacity ( $f_{\text{CO}_2}$ ) from *Takahashi et al.* [2009] was corrected to the year 2006 considering (i) a mean interannual increase in the oceanic  $p\text{CO}_2$  of  $1.75 \mu\text{mol yr}^{-1}$ , 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  $F_{\text{CO}_2}$  accounts for both the natural and the anthropogenic CO<sub>2</sub> fluxes across the air-sea interface.

## 3. Results

### 3.1. Storage Rates of DIC, $C_{\text{ant}}$ , and DIC<sub>nat</sub>

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



**Figure 2.** (a) Evolution of DIC inventory (blue line with triangles, left axis) and  $C_{\text{ant}}$  inventory (red line with circles, right axis) in the eastern SPNA. Discontinuous blue (red) lines are the DIC ( $C_{\text{ant}}$ ) inventory error bars. (b) Left axes: time evolution of (1) DIC transport across the OVIDE section obtained from in situ data ( $T_{\text{DIC}}$ , blue line with triangle), (2) DIC transported by the net volume transport ( $T_{\text{DIC}}^{\text{net}}$ , cyan dashed line with stars), (3) DIC transported by the zero net volume transport ( $T_{\text{DIC}}^0$ , 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  $C_{\text{ant}}$  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  $C_{\text{ant}}$  inventory, but the irregularities are not large enough to be interpreted as significant (the inventory error is  $2 \text{ mol C m}^{-2}$ ). Subtracting  $S_{C_{\text{ant}}}$  to  $S_{\text{DIC}}$ , we find that the storage rate of  $\text{DIC}_{\text{nat}}$  is not statistically different from zero. This result will be discussed in section 4.

### 3.2. Lateral Advection of DIC, $C_{\text{ant}}$ , and $\text{DIC}_{\text{nat}}$

The temporal variability of  $T_{\text{DIC}}$  across the OVIDE section from 2002 to 2010 is shown in Figure 2b (left axis, blue line). The  $T_{\text{DIC}}$  is positive (northeastward) in all years except 2004. The interannual variability of  $T_{\text{DIC}}$  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 ( $\sim 2100 \mu\text{mol kg}^{-1}$ ). The transport of a tracer across a transoceanic section can be split in the component associated with the



net transport of volume ( $T_{vol}^{net}$ ) and a remaining component for which the net volume transport is zero [Bacon, 1997]. Following this decomposition,  $T_{DIC}$  can be written as

$$T_{DIC} = T_{DIC}^{net} + T_{DIC}^0, \quad (2)$$

with

$$T_{DIC}^{net} = T_{vol}^{net} [\overline{DIC}], \quad (3)$$

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

The variability of  $T_{DIC}^{net}$  and  $T_{DIC}^0$  is shown in Figure 2b. We find that the  $T_{DIC}$  is mainly explained by  $T_{DIC}^{net}$ ; therefore, the interannual variability of  $T_{DIC}$  depends mainly on the variability of the  $T_{vol}^{net}$  ( $r^2 = 0.99$ ). The 2002–2010 mean values of DIC and volume transport across the OVIDE section are  $1688 \pm 602 \text{ kmol s}^{-1}$  ( $0.639 \pm 0.228 \text{ Pg C yr}^{-1}$ ) and  $0.8 \pm 2 \text{ Sv}$  ( $\text{Sv} = 10^6 \text{ m}^3 \text{ s}^{-1}$ ), 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  $1580 \pm 36 \text{ kmol s}^{-1}$  using Table S1 data, with a net volume transport of  $0.8 \pm 2 \text{ Sv}$ .

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  $108 \pm 82 \text{ kmol s}^{-1}$  ( $0.041 \pm 0.031 \text{ Pg yr}^{-1}$ ). 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.

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

In equation (4),  $[\overline{DIC}]^{Ov}$  and  $[\overline{DIC}]^{Sills}$  are the mean DIC concentrations at the OVIDE section and the G-I-S sills, respectively;  $\text{error } T_{vol}^{net}$  is the error of net volume transport across both lateral boundaries (2 Sv) [Lherminier *et al.*, 2010; Pérez *et al.*, 2013];  $\text{error } T_{DIC}^0 \text{ Ov}$  and  $\text{error } T_{DIC}^0 \text{ Sills}$  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  $\frac{\text{std}(T_{DIC}^0 \text{ Ov})}{\sqrt{n}}$  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  $C_{ant}$  lateral advection, the 2002–2010 average of  $T_{Cant}$  across the OVIDE section is  $248 \pm 35 \text{ kmol s}^{-1}$  ( $0.094 \pm 0.013 \text{ Pg C yr}^{-1}$ ) and the  $T_{Cant}$  across the G-I-S sills normalized to 2006 is  $162 \pm 17 \text{ kmol s}^{-1}$  ( $0.061 \pm 0.007 \text{ Pg C yr}^{-1}$ ). The lateral advection causes a net input of  $C_{ant}$  of  $86 \pm 21 \text{ kmol s}^{-1}$  ( $0.033 \pm 0.008 \text{ Pg C yr}^{-1}$ ) in the eastern SPNA box. Note that  $1 \text{ Pg C yr}^{-1}$  equals  $2642 \text{ kmol s}^{-1}$ .

Finally, we find that the  $A_{DIC}$  and  $A_{Cant}$   $0.041 \pm 0.031 \text{ Pg yr}^{-1}$  and  $0.033 \pm 0.008 \text{ Pg C yr}^{-1}$ , respectively, are not statistically different. Therefore, the  $DIC_{nat}$  convergence due to the lateral advection,  $0.008 \pm 0.032 \text{ Pg yr}^{-1}$ , 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 CO<sub>2</sub> Flux

The  $F_{CO_2}$  over the eastern SPNA was estimated at  $0.062 \pm 0.02 \text{ Pg C yr}^{-1}$  using Takahashi *et al.* [2009] climatology rescaled to 2006. Positive  $F_{CO_2}$  means that the ocean is taking up CO<sub>2</sub> from the atmosphere. The total area of the eastern SPNA is  $3.8 \cdot 10^{12} \text{ m}^2$ . Landschützer *et al.* [2013] estimated air-sea CO<sub>2</sub> fluxes for the Atlantic Ocean using data from 1998 to 2007 from the Surface Ocean CO<sub>2</sub> 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 CO<sub>2</sub> fluxes in our region during the period 2002–2010.

**Table 1.** Estimates of the Elements of the DIC,  $C_{\text{ant}}$  and  $\text{DIC}_{\text{nat}}$  Budgets in the Eastern SPNA Box<sup>a</sup>

	DIC ( $\text{Pg C yr}^{-1}$ )	$C_{\text{ant}}$ ( $\text{Pg C yr}^{-1}$ )	$\text{DIC}_{\text{nat}}$ ( $\text{Pg C yr}^{-1}$ )
Storage rate ( $S$ )	$0.060 \pm 0.014$	$0.055 \pm 0.005$	<i><math>0.005 \pm 0.015</math></i>
Lateral advection ( $A$ )	$0.041 \pm 0.031$	$0.033 \pm 0.008$	<i><math>0.008 \pm 0.032</math></i>
Air-sea flux ( $F$ )	$0.062 \pm 0.020$	<i><math>0.022 \pm 0.009</math></i>	<i><math>0.040 \pm 0.022</math></i>
Biological activity ( $B$ )	$-0.043 \pm 0.039$	-----	$-0.043 \pm 0.039$

<sup>a</sup>Deduced values from the other elements of the DIC or  $C_{\text{ant}}$  budget are represented in italic font.

### 3.4. Closing the Budgets of DIC, $C_{\text{ant}}$ and $\text{DIC}_{\text{nat}}$ in the Eastern SPNA

Table 1 summarizes our estimates of  $S_{\text{DIC}}$ ,  $A_{\text{DIC}}$  and  $F_{\text{CO}_2}$  for the 2002–2010 period. Using these estimates and applying a simple box model, we infer a biological DIC consumption of  $0.043 \pm 0.039 \text{ Pg C yr}^{-1}$ , accounting for primary production and remineralization processes. Concerning the  $C_{\text{ant}}$  budget, we inferred from the imbalance between  $S_{\text{Cant}}$  and  $A_{\text{Cant}}$  that  $F_{\text{Cant}}$  equals  $0.022 \pm 0.009 \text{ Pg C yr}^{-1}$  (Table 1). Finally, for the budget of  $\text{DIC}_{\text{nat}}$ : (i) the flux of natural  $\text{CO}_2$  is  $0.040 \pm 0.022 \text{ Pg C yr}^{-1}$ , deduced from the difference between  $F_{\text{CO}_2}$  and  $F_{\text{Cant}}$ ; (ii) the storage rate and the lateral advection of  $\text{DIC}_{\text{nat}}$  are not statistically different from zero, and (iii) the biological activity inferred from the DIC budget is  $-0.043 \pm 0.039 \text{ Pg C yr}^{-1}$ . From this budget we find that the biological activity consuming DIC is in balance with the air-sea flux of natural  $\text{CO}_2$ .

## 4. Discussion and Conclusions

Our estimate of the DIC storage rate for the eastern SPNA,  $1.32 \pm 0.30 \text{ mol C m}^{-2} \text{ yr}^{-1}$  ( $0.060 \pm 0.014 \text{ Pg C yr}^{-1}$ ), is larger but not statistically different from the  $0.91 \pm 0.28 \text{ mol C m}^{-2} \text{ yr}^{-1}$  computed by *Tanhua and Keeling* [2012] for a similar area and the period 1995–2005. Concerning the  $S_{\text{Cant}}$ , our estimate of  $1.21 \pm 0.12 \text{ mol C m}^{-2} \text{ yr}^{-1}$  ( $0.055 \pm 0.005 \text{ Pg C yr}^{-1}$ ) is statistically larger than *Tanhua and Keeling* [2012]'s estimate,  $0.40 \pm 0.33 \text{ mol C m}^{-2} \text{ yr}^{-1}$  ( $0.44 \pm 0.36 \text{ mol C m}^{-2} \text{ yr}^{-1}$  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  $S_{\text{Cant}}$  is also larger than the estimate of *Pérez et al.* [2013] using 2002–2006 OVIDE data ( $0.027 \pm 0.004 \text{ Pg C yr}^{-1}$  when referenced to 2006). Note that the  $S_{\text{Cant}}$  computed here for the 2000s decade is still lower than the  $S_{\text{Cant}}$  computed for 1997 by *Pérez et al.* [2013],  $0.083 \pm 0.004 \text{ Pg C yr}^{-1}$  ( $0.100 \pm 0.008 \text{ Pg C yr}^{-1}$  when rescaled to 2006). On the one hand, *Pérez et al.* [2013] suggested a relationship between the  $S_{\text{Cant}}$  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  $S_{\text{Cant}}$  obtained for 1997 and the higher  $S_{\text{Cant}}$  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  $C_{\text{ant}}$  storage rate and MOC intensity in the SPNA.

Comparing  $S_{\text{DIC}}$  and  $S_{\text{Cant}}$ ,  $0.060 \pm 0.014 \text{ Pg C yr}^{-1}$  and  $0.055 \pm 0.005 \text{ Pg C yr}^{-1}$ , 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  $\text{DIC}_{\text{nat}}$  had a negligible contribution.

The variability of  $T_{\text{DIC}}$  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  $T_{\text{Cant}}$  that depends mainly on the variability of the MOC intensity [Zunino et al., 2014]. The variability of  $T_{\text{DIC}}$  and  $T_{\text{Cant}}$  are controlled by the variability of different components of the circulation because the vertical gradients of DIC and  $C_{\text{ant}}$  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  $C_{\text{ant}}$  northeastward than its lower branch does southwestward because the surface waters are enriched in  $C_{\text{ant}}$  by a factor of 2 or more compared to deep waters. The  $T_{\text{DIC}}$  is an order of magnitude larger than  $T_{\text{Cant}}$  since DIC concentration is much larger than  $C_{\text{ant}}$  concentration (e.g., DIC and  $C_{\text{ant}}$  concentration averages in 2010 at the OVIDE section are  $2161 \mu\text{mol kg}^{-1}$  and  $28 \mu\text{mol kg}^{-1}$ , respectively). In spite of these differences between  $T_{\text{DIC}}$  and  $T_{\text{Cant}}$ , the

convergences of DIC and  $C_{\text{ant}}$  due to the lateral advection are not statistically different:  $0.041 \pm 0.031$  and  $0.036 \pm 0.008 \text{ Pg yr}^{-1}$ , respectively. Consequently, we found that the convergence of  $\text{DIC}_{\text{nat}}$  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,  $F_{\text{CO}_2}$ , was estimated at  $0.062 \pm 0.02 \text{ Pg C yr}^{-1}$ . It is smaller than the  $0.09 \text{ Pg C yr}^{-1}$  given in Pérez *et al.* [2013] for the same region. This difference is because their value was based on the  $F_{\text{CO}_2}$  given in Takahashi *et al.* [2009] for latitudes  $> 50^\circ\text{N}$  in the Atlantic Ocean, which include areas with stronger  $F_{\text{CO}_2}$  than the eastern SPNA. The  $F_{\text{Cant}}$  was deduced from the  $C_{\text{ant}}$  budget at  $0.022 \pm 0.009 \text{ Pg C yr}^{-1}$ . Considering our estimates of  $F_{\text{CO}_2}$  and  $F_{\text{Cant}}$  (Table 1), we estimated the air-sea flux of natural  $\text{CO}_2$  to be  $0.040 \pm 0.022 \text{ Pg C yr}^{-1}$ . This result is in agreement with the  $0.042 \text{ Pg C yr}^{-1}$  computed from the natural  $\text{CO}_2$  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  $\text{CO}_2$  fluxes in the SPNA.

The biological consumption/production of DIC inferred from the other terms of the DIC budget (Table 1) gives  $-0.043 \pm 0.039 \text{ Pg C yr}^{-1}$ . This is in agreement with the estimate of Maze *et al.* [2012] obtained from nutrient and oxygen budgets for the same region:  $-0.040 \pm 0.023 \text{ Pg C yr}^{-1}$ . Note that their value comes from the sum of their estimates of biological consumption in their Irminger and northeastern European Basin boxes,  $-0.019 \pm 0.016 \text{ Pg C yr}^{-1}$  and  $-0.021 \pm 0.016 \text{ Pg C yr}^{-1}$ , respectively. Using this estimate of biological activity in the budget of natural  $\text{CO}_2$ , we find that it is in balance with the air-sea transfer of natural  $\text{CO}_2$  that was deduced from the  $F_{\text{CO}_2} - F_{\text{Cant}}$  difference. Because the lateral advection and the storage rate of natural  $\text{CO}_2$  are not significantly different from zero, our result suggests that the biological activity drives the air-sea flux of natural  $\text{CO}_2$ . Körtzinger *et al.* [2008] and Takahashi *et al.* [2002] found that the biological carbon drawdown during spring/summer mediates the  $\text{CO}_2$  sink in the Labrador Sea and in the  $40^\circ\text{N}$ – $60^\circ\text{N}$  region, respectively. Here we show that this balance concerns only the natural  $\text{CO}_2$  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  $\text{CO}_2$  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  $\text{DIC}_{\text{nat}}$  in the SPNA due to the lateral advection, which they balanced with the uptake of natural  $\text{CO}_2$  from the atmosphere without considering biological activity. Surprisingly, their estimates of natural air-sea  $\text{CO}_2$  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  $C_{\text{ant}}$  budgets in the North Atlantic and Arctic Ocean (from  $24^\circ\text{N}$  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  $\text{CO}_2$  air-sea fluxes. Jeansson *et al.* [2011] evaluated the DIC and  $C_{\text{ant}}$  budget in the Nordic Sea. They closed the DIC budget considering the  $C_{\text{ant}}$  storage rate in the DIC balance and inferred the air-sea  $\text{CO}_2$  flux in the Nordic Seas to be  $0.19 \pm 0.06 \text{ Pg C yr}^{-1}$ , comparable to the  $0.11 \pm 0.06 \text{ Pg C yr}^{-1}$  estimated using the Takahashi *et al.* [2009] climatology. Jeansson *et al.* [2011] concluded that there is a negligible uptake of  $C_{\text{ant}}$  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,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  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 ( $-0.4 \text{ Sv}$ ), 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  $S_{\text{DIC}}$  estimate, since it was computed as the time derivative of data measured approximately at the same period of the year, or the  $F_{\text{CO}_2}$ , since we used the annual accumulated  $F_{\text{CO}_2}$  to close our DIC budget. Concerning the lateral advection, the seasonal variability of  $T_{\text{DIC}}$  may depend on the seasonal variability of both the DIC concentration and the circulation. The DIC concentration fluctuates seasonally due to the primary production ( $40\text{--}80 \mu\text{mol kg}^{-1}$ ) [Corbière *et al.*, 2007] in the first 100 m of the water column. We calculated the  $T_{\text{DIC}}$  error associated with these DIC changes as the standard deviation of  $T_{\text{DIC}}$  obtained from DIC surface concentrations that were randomly perturbed between 40 and  $80 \mu\text{mol kg}^{-1}$ . This error,  $0.015 \text{ Pg C yr}^{-1}$ , is smaller than the  $T_{\text{DIC}}$  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  $T_{\text{DIC}}$  variability ( $11 \text{ kmol s}^{-1}$  or  $0.004 \text{ Pg C yr}^{-1}$ ) since  $T_{\text{DIC}}$  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,  $C_{\text{ant}}$ , and  $\text{DIC}_{\text{nat}}$  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  $C_{\text{ant}}$  lateral advection (65%) and the air-sea flux of  $C_{\text{ant}}$  (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  $\text{CO}_2$ . Our results verify for the first time the generally accepted assumption that the  $\text{CO}_2$  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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