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Atlantic Ocean CO_2 uptake reduced by weakening of the meridional overturning circulation

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Abstract:

Uptake of atmospheric carbon dioxide in the subpolar North Atlantic Ocean declined rapidly between 1990 and 2006. This reduction in carbon dioxide uptake was related to warming at the sea surface, which—according to model simulations—coincided with a reduction in the Atlantic meridional overturning circulation. The extent to which the slowdown of this circulation system—which transports warm surface waters to the northern high latitudes, and cool deep waters south—contributed to the reduction in carbon uptake has remained uncertain. Here, we use data on the oceanic transport of volume, heat and carbon dioxide to track carbon dioxide uptake in the subtropical and subpolar regions of the North Atlantic Ocean over the past two decades. We separate anthropogenic carbon from natural carbon by assuming that the latter corresponds to a pre-industrial atmosphere, whereas the remaining is anthropogenic. We find that the uptake of anthropogenic carbon dioxide—released by human activities—occurred almost exclusively in the subtropical gyre. In contrast, natural carbon dioxide uptake—which results from natural Earth system processes—dominated in the subpolar gyre. We attribute the weakening of contemporary carbon dioxide uptake in the subpolar North Atlantic to a reduction in the natural component. We show that the slowdown of the meridional overturning circulation was largely responsible for the reduction in carbon uptake, through a reduction of oceanic heat loss to the atmosphere, and for the concomitant decline in anthropogenic CO_2 storage in subpolar waters.

1. Introduction

Contemporary CO_2 uptake from the atmosphere by the global ocean has been estimated to be 1.6±0.9 PgC yr^{-1} from an observation-based CO_2 flux climatology[1] referenced to the year 2000. Contemporary atmospheric CO_2 consists of a mix of molecularly identical natural and anthropogenic CO_2 (C_{ANT}). The whole North Atlantic (from the Equator to the Bering Strait, including the Arctic seas) represents only 13% of the global ocean area and yet annually accounts for about one-third of the contemporary ocean CO_2 uptake (0.47 PgC yr^{-1}) and has the largest of C_{ANT} storage rates (0.49±0.04 PgC yr^{-1} referenced to 2004) of all oceans[2]. However, air–sea CO_2 uptake in the North Atlantic is not necessarily predominantly anthropogenic[3, 4]. In fact, air–sea CO_2 fluxes in the North Atlantic result from anthropogenic forcing and progressive northward cooling of the upper limb of the meridional overturning circulation

(MOC). The latter is responsible for the NA uptake of natural CO_2 (ref. 5) that would occur even in the absence of the anthropogenic forcing. This air-sea flux of natural CO_2 is driven by thermal processes[5] — not biological processes — and has been estimated in 0.31-0.39 $PgC\cdot y^{-1}$ (refs 5,6), which represents roughly three-fourths of the contemporary air-sea CO_2 uptake. The remaining uptake (0.08-0.16 $PgC\cdot y^{-1}$) comes from the anthropogenic perturbation, which alone cannot account for the C_{ANT} storage rate of the NA[2]. The additional source of C_{ANT} comes from the northward transport of C_{ANT}-laden south-latitude waters[4,7–9] by the upper limb of the MOC.

Air-sea CO_2 fluxes in the subpolar and subtropical regions have similar rates (0.27 and 0.22 $PgC\cdot y^{-1}$, referenced to 2000, respectively[1]), but the flux per unit area in the subpolar NA is twice that in the subtropical NA (2.0 vs. 1.0 $mol\text{-}C\cdot m^{-2}\cdot y^{-1}$). At multidecadal time scales, sea surface pCO_2 in these regions follow the atmospheric increase[1]. However, these two regions also have contrasting responses to different North Atlantic Oscillation (NAO) periods. Between 1993 and 2006, the CO_2 uptake rate in the western subpolar[10] and, more generally, in the subpolar gyre[11] dramatically weakened as evidenced by the rapid increase in sea-surface pCO_2 compared to atmospheric pCO_2. Changes in the NAO (the index declined from high positive values in the early 1990s to lower values in the early 2000s)[12] and the associated weakening of the northward transport of subtropical water by the North Atlantic Current (NAC) have been identified, using inverse atmospheric CO_2 and physical-biological models[13,14], as the main causes for the decrease in CO_2 uptake in the subpolar NA. In contrast, in the subtropical NA, CO_2 uptake increased during the years with low NAO index[15,16]. There are, however, few observations of C_{ANT} transport reported for different NAO conditions. In addition, numerical models have shown contrasting CO_2 uptake responses[14,17] and discrepancies with field data, suggesting that more observations are required to better understand the interactions between ocean circulation and the carbon cycle, in particular regarding the mechanisms governing the exchange, advection and accumulation of CO_2.

CO_2 transport by the meridional overturning circulation

The analysis of repeated trans-Atlantic sections at 25ºN showed that the upper limb of the MOC carries 18.7±2.1 Sv (Sv = 10^6 $m^3 \cdot s^{-1}$) northwards[18] (northward transport is considered positive). Most of this transport occurs through the Gulf Stream and, downstream, through the NAC (Fig. 1). The warm water moving northward in the upper limb of the MOC has high concentrations of C_{ANT} ([C_{ANT}]), whereas the cold, deep water moving southward[4,7] has very low [C_{ANT}]. This pattern yields net northward transports of heat[19] and C_{ANT} of 1-1.3 PW and 0.19-0.23 $PgC \cdot y^{-1}$ (refs 4, 7), respectively. The overturning and the southward transport of deep water of the MOC happen in the northern NA and Nordic seas, where high wintertime heat loss generates vertical convection and produces cold, fresh and well-ventilated deep waters[20] that are entrained in the deep western boundary current. Recent estimations of the MOC across the repeated A25 section (Greenland to Portugal; Fig. 1) showed slightly weaker mass transports[21,22] (12-18.5 Sv) than at 25ºN. The upper and lower limbs of the MOC showed contrasting temperatures and [C_{ANT}] (Fig 1b, see Methods for details on C_{ANT} computations), but both properties are positively correlated. The small westward increase in [C_{ANT}] at constant temperature indicates recent ventilation of the western side of the section. In the surface layer, [C_{ANT}] is close to saturation. East of the NAC, the low values (< 10 $\mu mol.kg^{-1}$) in deep waters create a larger vertical gradient of C_{ANT} between the surface and the deep ocean than to the west of the NAC, *i.e.* in the subpolar region, where the Labrador Sea Water (LSW), the Denmark Strait Overflow Water and the Iceland-Scotland Overflow Water show moderate [C_{ANT}].

Numerical models have shown that NAO conditions influence air-sea CO_2 uptake in the NA[13] by modulating the strength with which the NAC carries subtropical waters into the subpolar gyre[14]. However, these results have not been confronted with measurements of volume, heat and CO_2 transports due to the lack of observations during different NAO conditions. We examined several occupations of the A25 Greenland-Portugal section (Fig. 1a) conducted in August 1997 (FOUREX cruise) and in June 2002, 2004 and 2006 (OVIDE cruises). The year 1997 came after an unusually long high NAO period followed by a period of lower NAO between 2002 and 2006. The A25 cruise was specifically designed to run perpendicularly across the main NA currents (the different branches of the NAC and the boundary currents linked to the topography) in order to minimize the transports due to eddies[23]. Measurements from these cruises were used to calculate MOC_σ transport[21,22,24], taking σ_1 (density anomaly referenced to 1000 dbar) as the vertical coordinate (Fig. 2). MOC_σ, varied from 20.5±2.2 Sv in 1997 to the average value of 14.6±1.7 Sv for the 2002-2006 period (see Methods and Supplementary Information for details on the removal of the seasonal cycle and the computation of the uncertainties). When integrated from Greenland to Portugal along constant σ_1-lines, heat and C_{ANT} transports resemble the vertical profiles of the overturning circulation (Fig. 2). Volume, heat and C_{ANT} transport profiles are highly correlated ($0.92>r^2>0.89$), because the upper limb of the MOC transports warmer waters with higher [C_{ANT}] than the lower limb. On average, the net volume transport is negligible, and there is a net northward transport of heat (0.59±0.09 PW) and C_{ANT} (0.092±0.010 $PgC \cdot y^{-1}$). In 1997, the circulation showed a strong southward volume transport at intermediate levels ($32.4<\sigma_1<32.5$) that corresponds to the layer of the classical LSW (Fig. 2). On the other hand, during the lower NAO period, the southward volume transport was slightly stronger in the layer of the upper LSW ($32.2<\sigma_1<32.3$)[20]. In addition, the upper limb of MOC_σ ($\sigma_1<32.1$) showed a stronger transport in 1997 than in 2002-2006 (Fig. 2a), which is attributed to the NAC variability[24]. The heat and C_{ANT}

transports in 2002-2006 (0.41±0.06 PW and 0.074±0.009 $PgC \cdot y^{-1}$) were lower than in 1997 (0.76±0.09 PW and 0.110±0.012 $PgC \cdot y^{-1}$). Most remarkably, although the weakening of MOC_{σ} and of C_{ANT} transport were very similar (29% and 33%, respectively), heat transport underwent a more dramatic reduction (46%) between 1997 and 2002-2006. This contrasting behavior of volume and heat transports agrees with results from high-resolution circulation models[25]. We will treat the observations obtained in 1997 as a case study of circulation linked to a high NAO period, as opposed to the measurements obtained during 2002-2006 that were associated with a low/neutral NAO period.

Anthropogenic CO_2 budget of the North Atlantic

The C_{ANT} budget of any oceanic region is the result of the balance between lateral advection, air-sea fluxes and storage rates. Hereinafter, we will refer to the NA as the region extending from 25ºN to the Bering Strait. We calculated the NA C_{ANT} budget referenced to 2004 from updated datasets and for four different subregions or boxes (Fig. 3). In the subtropical box, the C_{ANT} storage rate was computed as described in the Methods section, while the estimates in other boxes were obtained from the literature (Supplementary Information). For the NA, we obtained a storage rate of 0.386±0.012 $PgC \cdot y^{-1}$ (0.95±0.05 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$) consistent with previous results (0.39±0.02 $PgC \cdot y^{-1}$, referenced to 2004; ref. 26). The C_{ANT} transports at 25°N (refs 4,7) were updated from 1992 and 1998 to 2004, resulting on a mean value of 0.25±0.05 $PgC \cdot y^{-1}$ (Methods section) that is consistent with a long term average MOC (ref. 18). Comparatively, C_{ANT} transport in the Bering Strait is low (0.008±0.003 $PgC \cdot y^{-1}$)[7,26]. Closing the C_{ANT} budget in the NA, an air-sea C_{ANT} flux of 0.13±0.05 $PgC \cdot y^{-1}$ was inferred. This estimate is compatible with the value of 0.17±0.06 $PgC \cdot y^{-1}$ (rescaled to 2004) derived from $\delta^{13}C$ observations[9]. Overall, these results indicate that the net advective transports contribute to 65±13% of the NA C_{ANT} storage rate (Fig. 3). Importantly, our observation-based estimate of the contribution of lateral transports to the C_{ANT} storage rate is larger than the 30% obtained by ocean inversions that combine C_{ANT} observations with transports and mixing from GCMs (ref. 26). By way of contrast, our result is consistent with a biogeochemical model[27] that predicted larger northward C_{ANT} transports than ocean inversions in the NA. Subtracting our estimate of air-sea C_{ANT} flux from the contemporary CO_2 uptake for the NA (0.49 $PgC \cdot y^{-1}$; ref. 1), we obtained a natural CO_2 uptake of 0.36 $PgC \cdot y^{-1}$, thereby corroborating independent estimates[5,6]. The air-sea C_{ANT} flux represents about 26% of the contemporary air-sea CO_2 uptake, which is much smaller than the 63% obtained from oceanic inversions[3]. The relevance of our result is that the air-sea C_{ANT} and natural CO_2 uptake estimates from the C_{ANT} budget are consistent with independent $^{13}C/^{12}C$ observations[9] and with other estimates of the air-sea natural CO_2 uptake[5,6].

The C_{ANT} storage rate estimated for the subtropical box is 0.280±0.011 $PgC \cdot y^{-1}$ (1.41±0.05 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$). So the subtropical box contributes 73% of the NA C_{ANT} storage rate, even though it represents only 49% of the NA area. By closing the C_{ANT} budget for this box (Fig. 3), we inferred an air-sea C_{ANT} uptake of 0.12±0.05 $PgC \cdot y^{-1}$ (0.60±0.25 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$). Here, the air-sea C_{ANT} flux is predominant in the contemporary air-sea CO_2 flux (1.0 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$). It represents 92% of the NA air-sea C_{ANT} uptake. In contrast, in the subpolar box, the C_{ANT} storage rate per unit area (0.99±0.06 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$) amounts to ~70% of that in the subtropical box[28]. To derive the C_{ANT} budget for the subpolar box, the C_{ANT} lateral transport over the Nordic sills (0.063±0.019 $PgC \cdot y^{-1}$) was calculated from available volume transports[22,29] and from [C_{ANT}] estimated from water mass ages and mixing models[30] (Supplementary Information). Then, the air-sea C_{ANT} flux was estimated at 0.016±0.012 $Pg\text{-}C \cdot y^{-1}$, which represents 35% of the C_{ANT} storage rate in this box. The air-sea C_{ANT} flux per unit area in the subpolar box (0.36±0.25 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$) is about 60% of the subtropical box which gives to the subtropical box a prevailing role in C_{ANT} uptake. Furthermore, the contemporary air-sea CO_2 uptake per unit area (2.0 mol-C·m^{-2}·y^{-1}) in the subpolar box is 5 times higher than the air-sea C_{ANT} uptake. This means that the natural component largely prevails over the anthropogenic component in the subpolar box. Interestingly, this result is in contrast with the subtropical box, where the air-sea anthropogenic flux is the major component (~60%).

The net heat and C_{ANT} transports flowing into the Nordic Seas reach 0.25±0.05 PW and 0.063±0.019 PgC·y^{-1}, respectively (Fig. 3, Supplementary Information). The C_{ANT} lateral transport almost fully accounts for the C_{ANT} storage rate in the Nordic[31] and Arctic Seas[32] meaning that air-sea C_{ANT} fluxes are practically zero (Fig. 3). Analyses based on $^{13}C/^{12}C$ measurements[33,34] have determined that the upper waters entering the Nordic Seas are saturated with C_{ANT}, preventing any further C_{ANT} uptake from the atmosphere and possibly causing outgassing due to the decline in buffering capacity. The strong air-sea heat loss in the Nordic and Arctic Seas actually drives the uptake of natural CO_2, as corroborated by observations in climatological analyses[1] that indicate a high air-sea CO_2 uptake (2.0 mol-C·m^{-2}·y^{-1}) north of 50ºN. In summary, while heat loss causes a strong natural CO_2 uptake in the Nordic and Arctic regions, the low anthropogenic component is less affected by the air-sea heat fluxes.

North Atlantic oscillation impact on CO_2 fluxes

The subpolar gyre is a remarkably rapid entrance portal for C_{ANT} into the deep ocean due to deep convection. In the early 1990s, the highly positive NAO period coincided with exceptional convection activity in the Labrador[20,35] and Irminger[36] Seas. Between 1997 and 2003, lower LSW formation rates prompted a decrease of 20 mol-C·m^{-2} in the C_{ANT} inventory, as inferred from chlorofluorocarbon data[37]. In the subpolar box, the C_{ANT} storage rate dropped from 0.083±0.008 during high NAO conditions in 1997 to 0.026±0.004 PgC·y^{-1} during the 2002-2006 low NAO period[28]. Hence, C_{ANT} storage rates per unit area were nearly three times lower during low NAO than high NAO periods

(Fig. 4). The decrease in northward C_{ANT} transport (Fig. 4) that followed the high-to-low NAO transition (from 0.110 to 0.074 $PgC \cdot y^{-1}$) is strongly related to the weakening of the intensity of the MOC (from 20.5±2.2 to 14.6±1.7 Sv). Most remarkably, the converging C_{ANT} lateral transports in the subpolar box decreased from 0.053±0.021 to 0.011±0.020 $PgC \cdot y^{-1}$. In these estimations, we assumed that the volume transport over the Nordic sills was constant, as suggested by observations[29], and [C_{ANT}] was time-rescaled using a rate of increase of 1.6% y^{-1} (Supplementary Information). After these calculations, the inferred air-sea C_{ANT} flux for the subpolar region was 0.53±0.22 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$ during the high NAO period and 0.33±0.25 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$ during the low NAO period. During the low NAO period, the C_{ANT} storage rate decreased due to the decrease in C_{ANT} lateral transport associated with the weakening of the MOC. Our results also suggest that this decrease was associated with a weakening in the air-sea C_{ANT} uptake.

The variability of the air-sea CO_2 flux in the subpolar gyre has already been described, modeled and discussed in regard to NAO variability[13,14,38,39]. In the north-western subpolar gyre, a reduction in the contemporary air-sea CO_2 flux of ~1.2 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$ was observed between 1993-94 and 2003-05 (refs 11, 38) and numerical simulation linked it to the weakening of the advection of subtropical waters with low total inorganic CO_2 (C_T) into the subpolar gyre[14]. This weakening is in agreement with our results (Fig. 4). During high NAO periods, heat loss increased[40], favouring the decrease in the surface pCO_2. The opposite is true during low NAO periods. Assuming a constant heat flux of 0.25±0.05 PW over the sills[29], we inferred, from the heat budget, a heat loss that is 1.5 to 3 times higher during high NAO than during low NAO periods (Fig. 4). Using the relationship between heat loss and natural CO_2 flux (see Methods), we inferred a decrease in the air-sea flux of natural CO_2 of 3.0±1.0 to 1.7±1.0 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$ (0.13 to 0.05 $PgC \cdot y^{-1}$, Fig. 4). This estimate is compatible with the rate of decrease in air-sea CO_2 fluxes in the subpolar gyre (2.3 to 1.0 $mol\text{-}C \cdot m^{-2} \cdot y^{-1}$) reported from surface observations[39]. Most importantly, this result strongly suggests that variability in the air-sea flux of natural CO_2 over the subpolar gyre responds to variability in the advection of subtropical waters with low $[C_T]$ and can be determined from the air-sea heat flux.

A possible explanation for the contrasting behaviour of the subtropical and subpolar regions lies in the origin of the water masses crossing the Florida Strait where ~45% of the volume transport comes from the South Atlantic as warm and intermediate waters[41] with low $[C_{ANT}]$ (ref. 4). These low $[C_{ANT}]$ waters are part of the upper limb of the MOC and reach C_{ANT} saturation levels on their path to the subpolar gyre. They incorporate about 0.08 $PgC\cdot y^{-1}$, which represents two thirds of the air-sea C_{ANT} flux in the subtropical box and contributes to the local response to anthropogenic forcing (Fig. 3). This explains why the air-sea C_{ANT} flux in the subtropical region is higher than that observed in the subpolar region. Furthermore, the intermediate water flowing through the Florida Strait is oversaturated with natural CO_2 (~30 $\mu mol\cdot kg^{-1}$) due to biological remineralization[4]. This allows the waters in the upper limb of the MOC to remain CO_2-saturated with low additional atmospheric uptake, despite the ~7ºC cooling undergone as they travel through the subtropical box, thereby explaining the low natural air-sea CO_2 flux in this box.

In summary, our results give a coherent and observation-based understanding of the CO_2 budget in NA regions. Our analysis provides evidence that the air-sea C_{ANT} flux contribution to the C_{ANT} storage and to the total air-sea CO_2 flux in the NA is lower than expected from ocean inversions. Advection is the main contribution to the C_{ANT} storage rate north of 25ºN. Practically, the entire air-sea C_{ANT} uptake in the NA occurs in the subtropical region, where the contemporary air-sea CO_2 flux is mainly anthropogenic, whereas the natural component predominates in the subpolar region. The high-to-low NAO transition was followed by a decrease in the heat and C_{ANT} transports into the subpolar region due to the weakening of the MOC and the simultaneous decrease in the C_{ANT} storage rate.

Because the anthropogenic contribution is a minor component of the contemporary air-sea CO_2 uptake in the subpolar region, we attribute the weakening of the contemporary air-sea CO_2 uptake to the decrease in natural CO_2 uptake. Our estimate of the decrease in natural CO_2 uptake inferred from the heat budget is in agreement with independent surface observations.

Finally, our study suggests that the long-term prediction of a reduction in the intensity of the MOC would be a positive climate-carbon feedback leading to a decrease in the C_{ANT} storage. Concomitant air-sea heat loss reduction may lead to a decrease in the abiotic component of the natural CO_2 uptake, which would be an even more important feedback.

Methods

C_{ANT} estimations. [C_{ANT}] was computed using the back-calculation φC_T^o method[42,43] with an overall uncertainty of ±5.2 μmol kg^{-1}. [C_{ANT}] in the subtropical region was estimated using the gridded CARINA dataset [44] and applying the φC_T^o, TrOCA[45] and TTD[46] methods. C_{ANT} storage rates obtained from each of these methods were in good agreement. The final C_{ANT} storage rate and its uncertainty for the subtropical region were calculated as the mean and the standard deviation of C_{ANT} storage rates obtained from each method. For the subpolar, Nordic and Arctic boxes, the storage rates were from refs 28, 31 and 47, respectively. Additional details are provided in the Supplementary Information.

Transport computations across A25. Absolute geostrophic currents were estimated using an inverse model constrained by subsurface ADCP (Acoustic Doppler Current Profiler) measurements and an overall mass conservation constraint[21,22,24]. The absolute velocity field is consistent with independent altimetry measurements[24] and estimates of the western boundary current transport[48] at the time of the OVIDE cruises. They are representative of the month of the cruise[23] and the seasonal variability was removed as explained in the Supplementary Information. Heat and C_{ANT} transports were calculated from current velocities perpendicular to the sections and from the potential temperature and C_{ANT} fields, respectively. The uncertainties of the MOC, heat and C_{ANT} transports were estimated to be ±2 Sv, 0.05 PW and 0.014 $PgC \cdot y^{-1}$, respectively (see online Supplementary Information for full calculation details).

The errors of the mean transports (volume, heat or C_{ANT}) across the A25 section were calculated as the standard deviation of the transport values divided by the square root of the number of transport values included in the estimate. Since only one transport estimate was available for the high NAO conditions, the error equals the standard deviation of the transports between 1997 and 2006, after removing a linear trend.

C_{ANT} transport at 25°N. We used the estimates of C_{ANT} transports across 25ºN reported in refs 4 and 7 that were respectively obtained from hydrographic cruises carried out in 1992 and 1998 and from C_{ANT} estimates based on a classic back-calculation method and on the C* method. We rescaled both estimates to year 2004 by removing the effect of the inter-annual variability of the MOC in C_{ANT} transports along 25ºN. In addition, we corrected the MOC estimates for their intra-annual variability. The resulting value obtained after the rescaling was 0.25±0.05 $PgC \cdot y^{-1}$. Details on these computations and the uncertainty estimates are given in the Supplementary Information.

Relationship between air-sea fluxes of heat and natural CO_2. The linear regression of natural C_T transports versus heat transports reported in Supplementary Table 4 for the A25 line has a slope of -0.56±0.10 $PgC \cdot y^{-1}$ per PW ($p < 0.05$). Assuming that the variability of heat and natural C_T transports over the sills and of accumulative terms are negligible[29,49], this slope can be interpreted as a relationship between the air-sea flux of natural CO_2 and the air-sea heat loss in the subpolar box. In the Nordic seas, a similar relationship is found between air-sea flux of natural CO_2 and air-sea heat loss. Using the mean value of the observed air-sea CO_2 uptake (0.09 ± 0.01 and 0.11 ± 0.06 $PgC \cdot y^{-1}$ as reported in refs 1 and 50, respectively) and the heat loss given in Fig. 3, we obtained a value of -0.5±0.1 PgC·yr^{-1} of air-sea flux per PW of heat loss in the Nordic seas. This relationship can also be applied to the natural CO_2 air-sea fluxes of the Nordic Seas, since here the C_{ANT} air-sea flux is negligible, as shown in Fig. 3.

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Author Contributions

All authors contributed extensively to the work presented in this paper. F.F.P., H.M. and A.F.R. designed the research. F.F.P., H.M., M.V-R, A.V., P.L. and A.F.R. analysed the physical and chemical data. H.M. and P.L. estimated the currents and thermohaline fields. F.F.P., M.V-R, A.V. and G.R. determined the anthropogenic CO_2 concentrations and storage rates. H.M., F.F.P., P.L. and A.F.R. estimated the uncertainties. F.F.P., H.M., M.V-R., P.C.P. and A.F.R wrote the paper. All authors discussed the results and implications and commented on the manuscript at all stages.

Author Information

The authors declare no competing financial interests. Supplementary Information is linked to the online version of the paper at www.nature.com/nature. Reprints and permissions information is available online at http://www.nature.com/nature/reprints. Correspondence and request for materials should be addressed to F.F.P.

Figure Legends

Figure 1 | Circulation and C_{ANT} in the North Atlantic. a) C_{ANT} storage rates (mol-C·m^{-2}·y^{-1}) and the main currents and water masses participating in the MOC (black line: North Atlantic Current (NAC), Gulf Stream (GS); grey line: Labrador Sea Water –LSW-, white lines: Denmark Strait and Iceland-Scotland Overflow Waters -DSOW and ISOW). The 25ºN, FOUREX and OVIDE section tracks are indicated (blue dotted lines); **b)** Vertical distribution of [C_{ANT}] (µmol·kg^{-1}) during the OVIDE 2004 cruise. Potential temperature (ºC; white lines) and the isopycnal σ_1 =32.10 (solid black line) separating the upper and lower limbs of MOC are also shown.

Figure 2 | Integrated transports of volume, heat and C_{ANT} across the A25 section (Greenland - Portugal) in 0.01 density bins. a) Volume transport (10^6 m^3 s^{-1}); **b)** Heat transport (PW); **c)** C_{ANT} transport (kmol s^{-1}). Color lines refer to years 1997 (grey), 2002 (yellow), 2004 (red) and 2006 (blue). The σ_1 = 32.10 horizon (solid black horizontal lines) represents the boundary between the upper and lower limbs of the MOC. NAC = North Atlantic Current, uLSW = upper Labrador Sea Water, cLSW = classical Labrador Sea Water.

Figure 3 | C_{ANT} budget in the North Atlantic referred to 2004. The upper box represents the NA and the lower boxes represent the four sub-regions. The horizontal arrows show the lateral transports of C_{ANT} in PgC·y^{-1} (blue font) and heat transports in PW (maroon font). The black numbers in the boxes are the C_{ANT} storage rates in PgC·y^{-1}. The vertical arrows show the anthropogenic (numbers in blue font) and contemporary (red font) air-sea CO_2 fluxes in PgC·y^{-1}. Errors appear in grey font. The surface area (m^2) of each region and the latitudinal boundaries between them are shown.

Figure 4 | Variability of the C_{ANT} budget in the subpolar box during high NAO (1997) and low NAO (2002-2006). Arrow and number formats are the same as in Figure 3, except for the numbers in green font that are the natural air-sea CO_2 fluxes in $PgC \cdot y^{-1}$, and in maroon font that are the air-sea heat flux in PW. Areal C_{ANT} storage rates ($mol\text{-}C \cdot m^{-2} \cdot y^{-1}$) are also given. For 1997, the heat budget includes a heat accumulation rate of 0.10±0.05 PW.

Supplementary Information

Atlantic Ocean CO_2 uptake reduced by weakening of the meridional overturning circulation

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1. Anthropogenic CO_2 computation and inventory estimate

We used three methods for computing anthropogenic CO_2 (C_{ANT}) from in situ measurements: the TTD (Transit Time Distribution[1]), TrOCA[2] and φC_T^o (refs 3,4) methods. On the basis of the variables needed to compute C_{ANT}, these methods can be classified into two groups: The carbon-based methods (TrOCA and φC_T^o), which typically require measurements of C_T, A_T, oxygen, temperature, salinity and eventually nutrients, and the TTD method that uses CFC-11 or CFC-12 measurements as proxies of the anthropogenic CO_2 signal. A summary presentation of those methods of C_{ANT} computation has been given in ref. 3. We applied the TrOCA and φC_T^o methods to the GLODAP and CARINA databases. The C_{ANT} estimates obtained by applying the TTD method to the GLODAP dataset[1] were downloaded from the following website: https://jshare.johnshopkins.edu/dwaugh1/public_html/Cant/. The uncertainties in C_{ANT} are ±6.2, ±5.2, and ±5.0 μmol kg^{-1} for the TrOCA, φC_T^o and TTD methods, respectively[3]. They depend on the specific assumptions of each methodology and on the corresponding analytical errors of the variables involved.

To compute the C_{ANT} inventories in the subtropical box, we first adapted the fields of [C_{ANT}] obtained from the CARINA/ GLODAP database to the WOA09 grid using a multi-parametric interpolation algorithm[5]. Second, we computed the specific (per unit area) inventories (Fig 1a) by vertically integrating the [C_{ANT}] on the WOA09 grid and, finally, we did a spatial (surface) integration to determine the inventories. The uncertainties in these inventories were calculated by randomly propagating over depth[3] a 5 μmol kg^{-1} standard error of the C_{ANT} estimate. The inventory uncertainties were equal to ±1 mol-C m^{-2} and ±2 mol-C m^{-2} when the vertical integration went down to 3000 m and 6000 m depths, respectively. For the subtropical box (surface area = $16.6\cdot10^{12}$ m^2), the estimated inventories are 17.3±0.3, 18.7±0.3 and 16.8±0.4 PgC for the TrOCA, φC_T^o and TTD methods respectively, that yield an average value of 17.6±0.6 PgC. In the subpolar box we relied on the C_{ANT} inventories computed in Perez et al. (2010; ref. 6).

2. C_{ANT} storage rate in the subtropical box

Based on previous works[7–9] we considered a transient steady state of the C_{ANT} distribution in the subtropical NA. Accordingly, the storage rate of C_{ANT} was computed from the inventory multiplied by the annual rate of increase k_t (C_{ANT} storage rate = k_t * C_{ANT} inventory). The value of k_t (0.016±0.001 y^{-1}) was calculated as the rate of increase of [C_{ANT}] in the mixed layer divided by [C_{ANT}] in the mixed layer considering that the evolution of [C_{ANT}] in the winter mixed layer follows the exponential increase of atmospheric CO_2 (refs 8, 9). The storage rates in the subtropical box were estimated in 0.299±0.0045, 0.277±0.005 and 0.269±0.006 PgC·y^{-1} by the TrOCA, φC_T^o, and TTD methods, respectively. The value in the budget presented in Fig.3 was obtained as the mean value and standard error (0.280±0.011 PgC·y^{-1}) of the ensemble of storage rates from all three C_{ANT} methods together. Also, k_t was used as a rescaling factor of C_{ANT} storage rates and C_{ANT} transports to years 2004 and 1997, whenever these were reported for other years.

3. C_{ANT} Storage rate in the subpolar box

Because the subpolar box includes areas of water mass formation[10,11], the assumption of a steady transient tracer distribution is not valid[9,12] there. This is mostly due to the fact that the thickness of the main water mass (LSW) in this region has a strong variability associated with the

NAO[10,11]. This variability drives strong changes in the C_{ANT} storage rates[6,8,12] due to the formation of LSW during the period of low NAO compared to the exceptional convection activity during the period of high NAO[8]. For the budget presented in Fig. 4, we relied on ref. 6 who indicated a drop in the storage rate in the "OVIDE Box" from 0.054±0.006 $PgC \cdot y^{-1}$ during the high NAO period (1991-1997) to 0.026±0.004 $PgC \cdot y^{-1}$ during the low NAO period (1998-2006). These results are in agreement with those inferred from CFC data[8]. On average, the C_{ANT} storage rate for the subpolar box referred to 2004 is 0.045±0.004 $PgC \cdot y^{-1}$ (Fig 3). The budget for the high NAO period in Fig. 4a was calculated by re-computing the storage rate in ref. 6 using the area of a subpolar box south-bounded by the FOUREX track. The storage rate was estimated at 0.083±0.008 $PgC \cdot y^{-1}$.

4. C_{ANT} transports through the sills

The isopycnal $\sigma_0 = 27.80$ separates the northward flowing NA water masses entering the Nordic Seas [Eastern North Atlantic Central Water (ENACW), Modified North Atlantic Central Water (MNACW), Greenland-Iceland Inflow Water (GIIW)] in the upper layers from the southward flowing water masses [Denmark Strait Overflow Water (DSOW), Iceland Scotland Overflow Water (ISOW) and East Greenland Current (EGC)] in the lower layer (Supplementary Table 1). The volume transports and associated errors were taken from the literature[15].

5. C_{ANT} storage in the Nordic Seas

Given the average C_{ANT} inventory of 1.2 PgC estimated from chlorofluorocarbon data[13], a storage rate of C_{ANT} of 0.018±0.004 $PgC \cdot y^{-1}$ was obtained using a k_t of 0.016±0.001 y^{-1} (refs 8, 9). This storage rate value is in agreement with a recent estimation[14].

Supplementary Table 1 - C_{ANT} transports through the sills.

Water Mass	Volume Transport (Sv)	[C_{ANT}] (μmol kg^{-1})	C_{ANT} Transport referred to 2004 (kmol s^{-1})
DSOW	-3±1	30±3	-89
ISOW	-3±1	28±3	-84
EGC	-1.8±0.2	37±3	-66
ENACW	3.85±1	49±4	189
MNACW	3.85±1	51±4	196
GIIW	0.8±0.2	40±3	32
Total	0.7±2.0		166±51 kmol s^{-1} 0.063±0.019 PgC·y^{-1}

EGC (East Greeland Current), ENACW (Eastern North Atlantic Central Water) MNACW (Modified North Atlantic Central Water) and GIIW (Greenland-Iceland Intermediate Water)

The [C_{ANT}] in the upper layer were estimated assuming that surface waters are saturated with CO_2 (ref. 16). The [C_{ANT}] for the DSOW was taken from the literature[13] and [C_{ANT}] for the ISOW was estimated from water mass decomposition (Supplementary Table 2; refs 17, 18). The [C_{ANT}] data for the rest of the water masses flowing over the sills were taken from previous studies[13]. Additionally, since our study is referenced to years 1997 (high NAO) or 2004 (low NAO), C_{ANT} transports over the sills were rescaled by applying the previously derived k_t factor of 0.016 ±0.001 y^{-1}, and we obtained transports of 0.057±0.018 PgC·y^{-1} and 0.063±0.019 PgC·y^{-1} for 1997 and 2004, respectively. These results fully corroborate recent transport estimates (0.062±0.014 PgC·y^{-1} for 2002; ref. 14).

Supplementary Table 2.- Water masses properties and [C_{ANT}] over the Nordic sills**.**

Water Mass	Mixing %	θ range (ºC)	θ avg. (ºC)	Salinity	[C_{ANT}] referred to 2004 (μmol kg^{-1})
NSAIW+NSDW	50	<0.4	0	34.885	10.9
MEIW	25	<3	2	34.80	40.4
MNAW	25	8	7.75	35.15	49.2
ISOW	100		2.44	34.93	28.0

NSAIW (Norwegian Sea Arctic Intermediate Water), NSDW (Norwegian Sea Deep Water), MEIW (Modified East IcelandicWater) MNAW (Modified North Atlantic Water)

6. Arctic Seas C_{ANT} storage and transports

Based on earlier estimates[19], we considered an average value of 2.9±0.4 PgC for the C_{ANT} inventory in the Arctic Seas referred to 2005. By applying the same k_t factor of 0.016±0.001 y^{-1}, we estimated a C_{ANT} storage rate of 0.043 $PgC \cdot y^{-1}$ referenced to 2004. The C_{ANT} transport between the Arctic and Nordic Seas in 1991 was estimated to be 0.031±0.004 $PgC \cdot y^{-1}$ northward[20]. Rescaling this value to 2004, a C_{ANT} transport of 0.039±0.008 $PgC \cdot y^{-1}$ was obtained. This result agrees with recent transport estimates of 0.040±0.019 $PgC \cdot y^{-1}$ referenced to 2002 (ref. 14). The C_{ANT} transport through Davis Strait was neglected. The net C_{ANT} transport from the Pacific to the Atlantic Ocean through the Bering Strait was obtained from previous works[21,22].

7. Transports and uncertainties at 25ºN

The seasonal variability of the MOC at 25°N has been recently evaluated[23] on the basis of the RAPID measurements. It has shown that the seasonal variability of the MOC is forced by the wind stress curl variability at the eastern boundary and affects the upper mid-ocean transport (T_{UMO}) in a narrow band close to the eastern boundary. The $[C_{ANT}]$ in this region ($[C_{ANT}]_{TUMO}$, Supplementary Table 3) was obtained from previous works[24,25] and the seasonal correction of T_{CANT} was modeled as $\Delta T_{UMO}*[C_{ANT}]_{TUMO}$, where ΔT_{UMO} is seasonal transport anomaly. ΔT_{UMO} was estimated[23] at 0.9±0.9 and -2±0.9 Sv for the 1992 and 1998 cruises, respectively. The rescaled C_{ANT} transports were hence computed applying the following equation:

$$T_{CANT}(2004) = (T_{CANT}(1992/1998) - \Delta T_{UMO} \,.[C_{ANT}]_{TUMO}) \cdot (1+k_t)^{\Delta y} \cdot MOC_{RAPID}/MOC_{corr}$$

where MOC_{corr} and MOC_{RAPID} are the de-seasonalized and long-term averaged MOC (18.7±2.1 Sv) as given by ref. 23. Δy is the time lapse (in years) between 2004 and 1992 or 1998. The final uncertainties were computed as the standard deviation of an ensemble generated by random perturbations of the 1992/1998 C_{ANT} transports, ΔT_{UMO}, $[C_{ANT}]_{\Delta TUMO}$ and k_t. The value of 0.25±0.05 $PgC \cdot y^{-1}$ for C_{ANT} transport at 25ºN is obtained as the mean between the 1992 and 1998 estimates, rescaled to 2004 and de-aliased from the seasonal variability. The most important contributions to the uncertainties are the initial uncertainties[24,25], while the rescaling of the MOC is practically negligible.

Supplementary Table 3.- Deseasonalized C_{ANT} transport**. (**1 $PgC \cdot yr^{-1}$ = 2642 kmol/s)

Year	**C_{ANT} transport (kmol/s)**	**2004 C_{ANT} transport (kmol/s)**	**ΔT_{UMO} (Sv)**	**C_{ANT} T_{UMO} (µmol·kg^{-1})**	**MOC_{cor} (Sv)**	**$(1+ k_t)^{\Delta y}$**	**Long term 2004 C_{ANT} transport ($PgC \cdot y^{-1}$)**
1992	630±200	725±200	0.9±0.9	45±3	18.5	1.213±0.015	0.28±0.08
1998	449±159	610±160	-2±0.9	51±3	18.1	1.066±0.005	0.23±0.06

8. Transports and uncertainties at A25

The MOC, heat, C_{ANT} and natural C_T transports across the A25 section are given in Supplementary Table 4. The natural C_T was computed as the difference between measured (total) C_T and C_{ANT}. The natural C_T transports were used to establish a relationship between air sea fluxes of heat and natural CO_2 for the subpolar box (see Methods). The associated uncertainties in Supplementary Table 4 are the standard deviations of an ensemble of 100 tracer transport estimates obtained by random perturbations of the volume transports and tracer fields scaled using the error covariance matrix of the velocity field given by the inverse model[15] and the uncertainties in natural C_T and C_{ANT} concentrations (6 µmol·kg^{-1} each). The natural C_T and C_{ANT} transport uncertainties were equal to 0.026 and 0.014 $PgC \cdot yr^{-1}$, respectively. These uncertainties are very similar to those that would be obtained using the approximate method proposed by ref. 26.

The seasonal variability of the MOC along A25 was evaluated using the high-resolution DRAKKAR ocean general circulation model[27]. During the OVIDE cruises the seasonal anomaly was not significant (0.0±0.5) because these cruises were conducted in June, when the seasonal anomaly is at its minimum. On the contrary, the FOUREX occupation in September 1997 did need a seasonal correction of +2.0±0.5 Sv. The vertical gradient of the transport of [C_{ANT}] is not affected by the seasonal cycle[28] and we assumed that the seasonal variability of the vertical gradient of the transport of heat and natural C_T is also negligible. So, we corrected for the seasonal variability of MOC transports from refs 29 and 30 by linearly rescaling the transports by the ratio <MOC>/MOC obtained in the model, where <MOC> is the annual value and MOC the monthly value (Supplementary Table 4).

Supplementary Table 4.- C_{ANT} and natural C_T transports through A25 section

Cruise	**MOC (Sv)**	**Heat (PW)**	**C_{ANT} ($PgC \cdot yr^{-1}$)**	**Natural C_T ($PgC \cdot yr^{-1}$)**
4X 1997	20.5	0.76±0.09	0.110±0.014	-0.352±0.026
Ov 2002	16.2	0.44±0.05	0.077±0.014	-0.207±0.026
Ov 2004	16.4	0.50±0.05	0.087±0.014	-0.265±0.026
Ov 2006	11.2	0.29±0.05	0.058±0.014	-0.079±0.026

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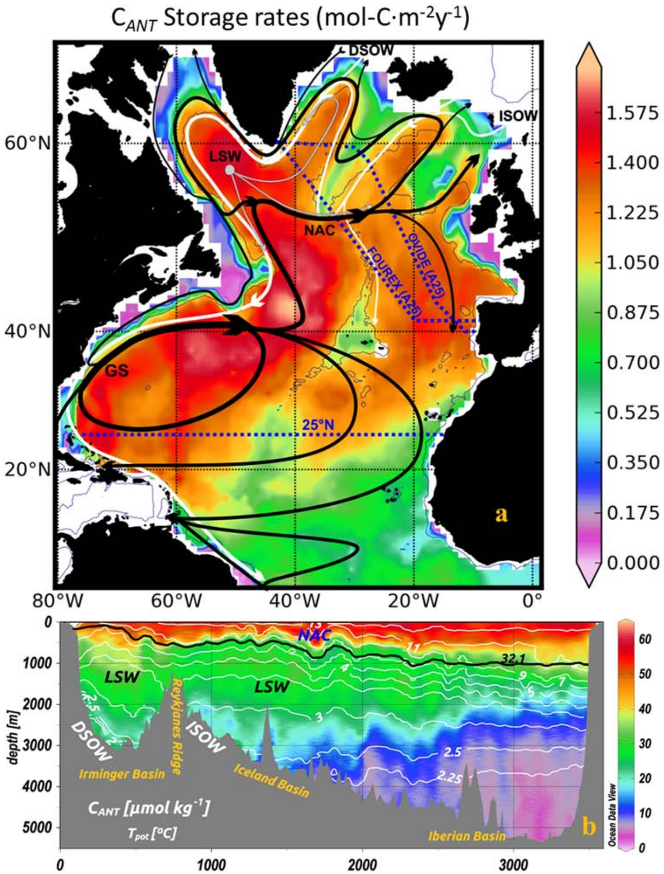
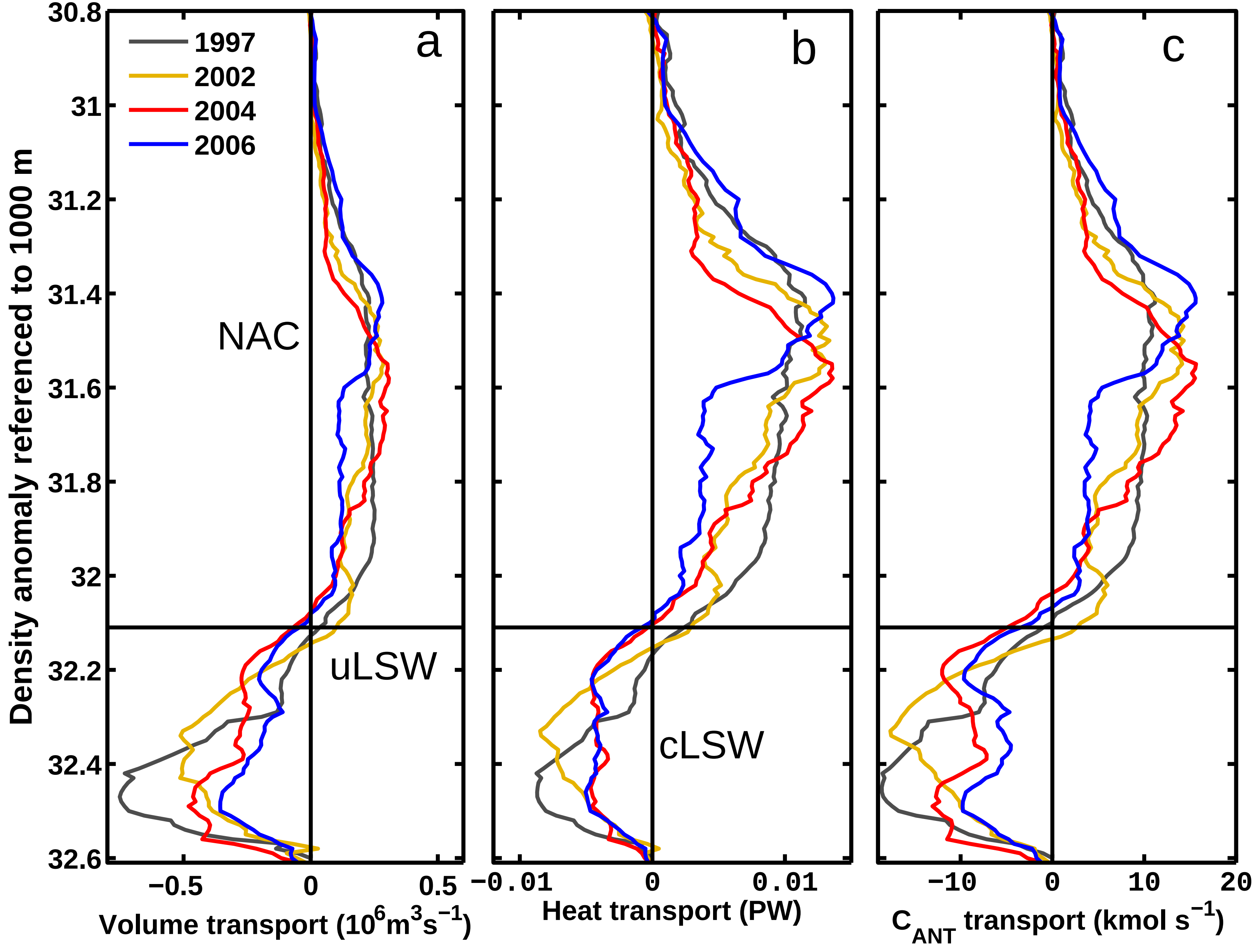
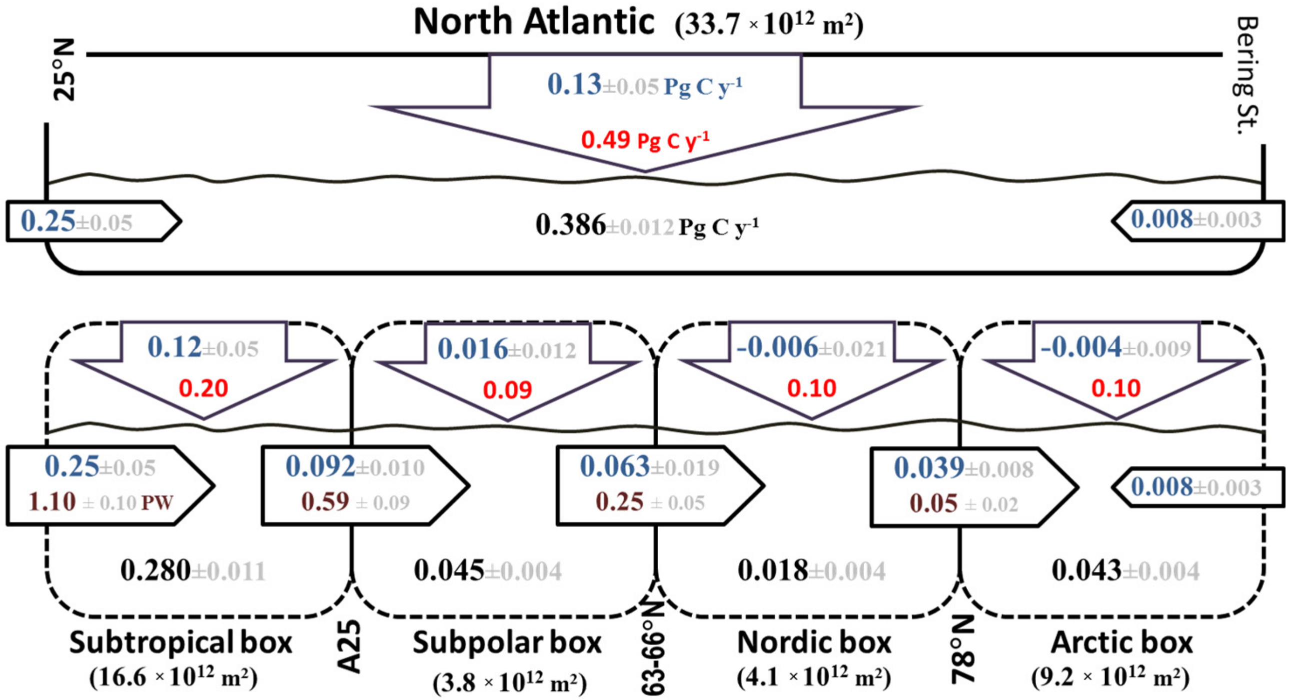


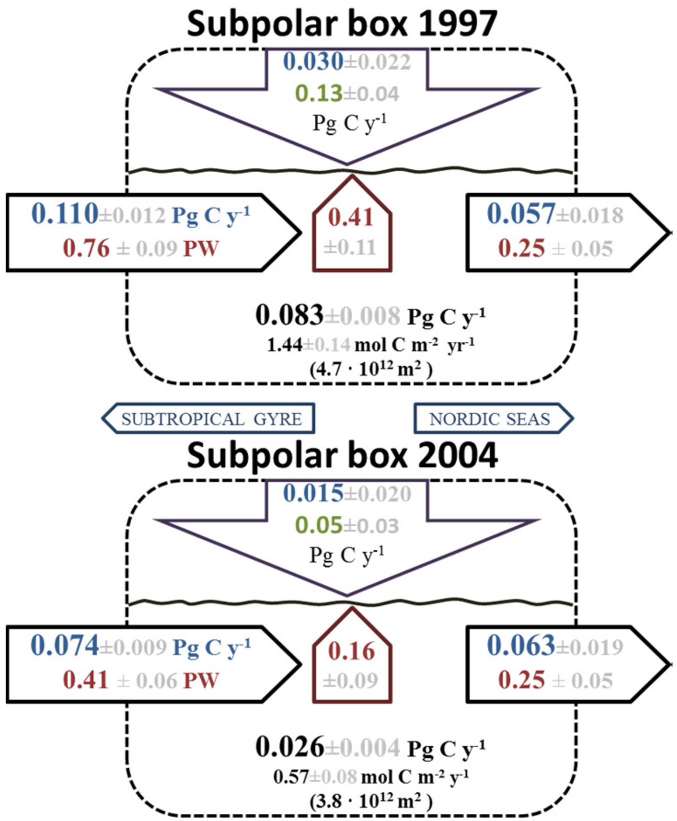
C_ANT Storage rates (mol-C·m-2y-1)
DSOW
ISOW
LSW
NAC
GS
25°N
FOUREX (A25)
OVIDE (A25)
60°N
40°N
20°N
80°W
60°W
40°W
20°W
0°
1.575
1.400
1.225
1.050
0.875
0.700
0.525
0.350
0.175
0.000
a
NAC
LSW
LSW
32.1
DSOW
ISOW
Reykjanes Ridge
Irminger Basin
Iceland Basin
Iberian Basin
C_ANT [µmol kg-1]
Tpot [°C]
depth [m]
0
1000
2000
3000
4000
5000
2.5
2.25
60
50
40
30
20
10
0
Ocean Data View
b



1997
2002
2004
2006
a
b
c
NAC
uLSW
cLSW
Density anomaly referenced to 1000 m
30.8
31
31.2
31.4
31.6
31.8
32
32.2
32.4
32.6
−0.5
0
0.5
Volume transport ($10^6 m^3 s^{-1}$)
−0.01
0
0.01
Heat transport (PW)
−10
0
10
20
C_{ANT} transport (kmol s^{-1})



North Atlantic (33.7 × 10¹² m²)
25°N
Bering St.
0.13±0.05 Pg C y⁻¹
0.49 Pg C y⁻¹
0.25±0.05
0.386±0.012 Pg C y⁻¹
0.008±0.003
0.12±0.05
0.20
0.016±0.012
0.09
-0.006±0.021
0.10
-0.004±0.009
0.10
0.25±0.05
1.10 ± 0.10 PW
0.092±0.010
0.59 ± 0.09
0.063±0.019
0.25 ± 0.05
0.039±0.008
0.05 ± 0.02
0.008±0.003
0.280±0.011
0.045±0.004
0.018±0.004
0.043±0.004
Subtropical box
(16.6 × 10¹² m²)
A25
Subpolar box
(3.8 × 10¹² m²)
63-66°N
Nordic box
(4.1 × 10¹² m²)
78°N
Arctic box
(9.2 × 10¹² m²)


Subpolar box 1997
0.030±0.022
0.13±0.04
Pg C y-1
0.110±0.012 Pg C y-1
0.76 ± 0.09 PW
0.41
±0.11
0.057±0.018
0.25 ± 0.05
0.083±0.008 Pg C y-1
1.44±0.14 mol C m-2 yr-1
(4.7 · 10^12 m^2)
SUBTROPICAL GYRE
NORDIC SEAS
Subpolar box 2004
0.015±0.020
0.05±0.03
Pg C y-1
0.074±0.009 Pg C y-1
0.41 ± 0.06 PW
0.16
±0.09
0.063±0.019
0.25 ± 0.05
0.026±0.004 Pg C y-1
0.57±0.08 mol C m-2 y-1
(3.8 · 10^12 m^2)