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Changes in column inventories of carbon and oxygen in the Atlantic Ocean

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Abstract

Increasing concentrations of dissolved inorganic carbon (DIC) in the interior ocean is expected as a direct consequence of increasing concentrations of CO_2 in the atmosphere. This extra DIC is often referred to as anthropogenic carbon (C_{ant}), and its inventory, or increase rate, in the interior ocean has previously been estimated by a multitude of observational approaches. Each of these methods are associated with hard to test assumptions since C_{ant} cannot be directly observed. Results from a simpler concept with few assumptions applied to the Atlantic Ocean are reported on here using two large data collections of carbon relevant bottle data. The change in column inventory on decadal time scales, i.e. the storage rate, of DIC, respiration compensated DIC and oxygen is calculated for the Atlantic Ocean. The average storage rates for DIC and oxygen is calculated to 0.72 ± 1.22 (95 % confidence interval of the mean trend: 0.65–0.78) mol m^{-2} yr^{-1} and −0.54 ± 1.64 (95 % confidence interval of the mean trend: −0.64–(−0.45)) mol m^{-2} yr^{-1}, respectively, for the Atlantic Ocean, where the uncertainties reflect station-to-station variability and where the mean trends are non-zero at the 95 % confidence level. The standard deviation mainly reflects uncertainty due to regional variations, whereas the confidence interval reflects the mean trend. The storage rates are similar to changes found by other studies, although with large uncertainty. For the subpolar North Atlantic the storage rates show significant temporal variation of all variables. This seems to be due to variations in the prevalence of subsurface water masses with different DIC concentrations leading to sometimes different signs of storage rates for DIC and C_{ant}. This study suggest that accurate assessment of the uptake of CO_2 by the oceans will require accounting not only for processes that influence C_{ant} but also additional processes that modify CO_2 storage.

1 Introduction

The ocean has stored a large fraction of the CO_2 emitted by human activities over the last few hundred years, i.e. the anthropogenic CO_2 (C_{ant}). A major scientific challenge today is to assess the oceanic sink and storage of CO_2. It is therefore relevant to monitor the storage rate of C_{ant} in the ocean and to assess its sensitivity to (climate-induced) changes in circulation and biology. The term "anthropogenic CO_2" normally refers to the increase of CO_2 in the ocean due to increasing atmospheric CO_2 concentrations, i.e. changes related to the thermodynamic air-sea disequilibrium driven by atmospheric changes. Much of prior work has focused on determining the total oceanic uptake of C_{ant} during the industrial period. Since C_{ant} cannot be measured directly, a variety of techniques have been developed to infer C_{ant} concentrations from observed properties, such as the carbonate system parameters, oxygen, nutrients and transient tracers. These methods depend on hard-to-test assumptions and the various methods regularly infer different C_{ant} concentrations from the same data set (e.g. Vázquez-Rodríguez et al., 2009; Alvarez et al., 2009; Ríos et al., 2010; Waugh et al., 2006; Friis, 2006; Tanhua et al., 2007). The differences arise both from biases in the methods and biases/errors in the measurements of the variables. The current methods all assume steady state circulation and are biased if that assumption is violated. Furthermore, the use of transient tracers for determination of C_{ant} concentration directly from knowledge of ventilation and circulation is an intrinsic part of most C_{ant} inference schemes. Thus, measurements of biogeochemical variables are often not directly used for the C_{ant} calculation, particularly for the upper part of the water column, so that the results probably do not capture the full variability in the ocean biogeochemistry.

Decadal storage rates of C_{ant} can be assessed by comparing direct measurements of the carbonate system and related variables obtained from the same stations or along the same section; this is often referred to as "repeat hydrography". It has been shown that the storage rates calculated from repeat hydrography can be scaled to encompass the full C_{ant} inventory in the North Atlantic by assuming steady state circulation and transient steady state behavior of the anthropogenic carbon (e.g. Tanhua et al., 2007; Gammon et al., 1982), and that the C_{ant} concentration in the surface ocean is, to a first approximation, exponentially increasing. In principle, the change in DIC concentration between repeat measurements (i.e. ΔDIC) at a specific location and depth in the ocean can be assumed to represent the anthropogenic component, which can be integrated over the water column to assess the storage rate of anthropogenic carbon. However, a direct comparison of the DIC fields usually show large variability, i.e. a patchy image, of the decadal change in DIC (or any other property) concentrations due to spatial and temporal variability in the ocean such as eddies and variable location of ocean fronts (e.g. Wanninkhof et al., 2010).

A common approach to estimate C_{ant} involves combining DIC data with dissolved O_2 or nutrient data to reduce variability due to internal ocean processes such as changes in remineralization. The variable effect on DIC is assumed to be captured from other tracers assuming fixed elemental ratios (C / N, C / P, C / O, i.e. Redfield ratios). A related approach uses multiple linear regressions (MLRs) where relations between a number of relevant properties, such as nutrients, oxygen and salinity are used to determine the DIC concentration of the sample. The use of MLR has the capacity to compensate for the some of the small scale variability in the ocean and the result is usually relatively smooth fields of ΔDIC. A variation of the MLR method was suggested by Friis et al. (2005) in which the MLR coefficients for both the cruise are subtracted from each other and then directly used for the calculation of the ΔDIC. This approach is known as extended MRL (eMLR). Wanninkhof et al. (2010) finds significant biases and various amount of scatter in the ΔDIC fields depending on the method applied to a section through the Atlantic Ocean, indicating that the correct choice of methodology is critical. It is however not obvious how to decide which methodology is most reliable.

Furthermore, estimates of C_{ant} that use O_2 as a component of tracer combination are subject to an easily understood bias. The integration of the tracer combination over the column to yield the change in the inventory of C_{ant} yields a sum of terms, one for inventory of each of the components, including the O_2 inventory. Globally, the change in O_2 inventory, however, is largely controlled by air-sea exchanges of O_2 (Keeling and Garcia, 2002). Thus a combined tracer that includes O_2 will be sensitive not just to processes driving long-term uptake of CO_2 by the oceans, but also processes driving long-term changes in O_2 (Keeling, 2005; Yool et al., 2010). Recent studies suggest that the change in global ocean O_2 inventory is decreasing at the level of $\sim 50\,\text{Tmol}\,\text{yr}^{-1}$ ~ due to warming and increased ocean stratification (Keeling et al., 2010; Helm et al., 2011), which is significant compared to estimated global ocean CO_2 uptake of $\sim 200\,\text{Tmol}\,\text{yr}^{-1}$. A tracer combination that uses O_2 therefore cannot yield a reliable estimate of ocean CO_2 uptake unless it is combined with independent estimates of the changes in ocean O_2 inventory (Keeling et al., 2010). Estimates of C_{ant} that include potential temperature as a correlating variable will be similarly sensitive to changes in ocean heat content (Levitus et al., 2012).

Related problems have been documented locally. For instance, time series data from the DYFAMED site in the Western Mediterranean Sea show increasing DIC concentrations with time for almost all depths (Touratier and Goyet, 2009), but the authors conclude that the C_{ant} concentration is decreasing based on a particular method to calculate the C_{ant} concentration. Similarly, Wakita et al. (2010) present measured DIC concentrations on a time-series station from the NW Pacific Ocean (stations KNOT and K2), and conclude that the C_{ant} concentration has a significant increasing trend with time although the DIC concentrations do not show such a trend. The reason for this discrepancy is often changes in circulation, i.e. another water mass with different preformed concentrations, ventilation etc. becomes more dominate at a certain location, or the thicknesses of the water mass at one location are varying with time. Other reasons might be changes to any one of the following processes, or a combination thereof: remineralization-depth, biological production, oxygen concentrations (Keeling et al., 2010), Redfield ratios (Riebesell et al., 2007), dominating phytoplankton species (Cermeno et al., 2008), or stratification. A review of some important such "secondary" mechanisms are discussed in Sabine and Tanhua (2010). The conclusions of Touratier and Goyet (2009) and Wakita et al. (2010), for instance, demonstrate that the

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DIC inventory of a water parcel do not necessary follow the trend in the inventory of C_{ant}.

Methods to determine C_{ant} uptake, whether from transient tracers or decadal budgets, also suffer from an additional fundamental limitation. If the ocean circulation is not steady, or other changes in biogeochemical cycling are occurring, then even a perfect determination of C_{ant} (assuming measurement limitations could be overcome), is not sufficient because C_{ant} is then no longer a complete measure of the uptake of CO_2 by the oceans. Additional contributions to the air-sea exchange of CO_2 due to non-steady biology or ocean circulation also need to be quantified. It is fundamentally a matter of the question being asked: Are we primarily interested in the change of C_{ant} with time, i.e. the excess CO_2 in the ocean that is a direct consequence of increasing atmospheric CO_2 concentrations, or are we interested to know the actual change of the inorganic carbon pool in the ocean due all processes? If the goal is to quantify the impact of the ocean on the global trend in atmospheric CO_2, then answer is clear: all processes contributing to air-sea exchange must be accounted for. To achieve this goal, it is therefore not sufficient simply to improve methods for determining C_{ant}. What is needed is a method that can determine the ocean CO_2 uptake from all processes.

Conceptually, there is actually a simple method available for determining the total uptake of CO_2 by the oceans. One simply measures DIC with sufficient accuracy and coverage to establish the total inventory of DIC in the global ocean, and then one tracks this over time through repeat hydrography. Although the DIC inventory can vary due to several processes, including any imbalance globally in the production of destruction of organic carbon, or in the global rate of the precipitation or dissolution of calcium carbonate, these processes are currently dwarfed globally by the changes caused by uptake of CO_2 from the atmosphere. A measurement of changes in the DIC inventory, with small corrections applied to account for organic carbon or carbonate effects (e.g. based on alkalinity or dissolved organic carbon measurements), would effectively determine the CO_2 uptake by all processes.

The principle difficulty with this method is that it requires detecting trends in the inventory of DIC against the background variability, not by using correlating tracers, but simply by having sufficiently high coverage. We are not aware of any attempt to date to apply this method. Over the past few decades, however, a large increase in the coverage of DIC measurements has been realized in certain ocean regions. As a first step, we focus here on the feasibility of tracking changes in the DIC column inventory of the upper 2000m of the water column in the Atlantic Ocean over the past few decades. Our study takes advantage of major new data assets including the GLODAP (Key et al., 2004) and CARINA (Key et al., 2010) datasets, which together provide an unprecedented coverage in time and space. Our study explores not just the changes in DIC inventory, but also changes in dissolved O_2, and in a combination of dissolved O_2 and DIC that compensates for changes due to photosynthesis and respiration, which we call abiological DIC, or DIC_{abio}. Changes in dissolved O_2 are of interest in relation to recent studies suggesting that O_2 levels in the ocean may be declining due to increasing ocean stratification, and DIC_{abio} is of interest because of its potential close relationship to C_{ant}.

The change in DIC column inventory is a function of the air-sea flux, the convergence or divergence of DIC, the net change in organic carbon in the column, and the net precipitation or dissolution of carbonate. Similarly, the change in oxygen column inventory is a function of the air-sea flux, convergence or divergence of oxygen, and the net change in organic carbon in the water column. The convergence and divergence terms will, obviously, become less important the larger the scale. In this study we focus on relating observed changes in column inventory of DIC and oxygen to the air-sea flux component and the convergence/divergence terms of these relations. We are assuming no change in organic carbon concentrations and $CaCO_3$ since these are presumably negligible and we are not aware of any observational evidence for the contrary.

2 Methods

2.1 Data

This study use data contained in the data products for the Atlantic Ocean within GLODAP (Global Ocean Data Analysis Project, http://cdiac.ornl.gov/oceans/glodap/index.html, Key et al., 2004) and CARINA (CARbon IN the Atlantic, (http://cdiac.ornl.gov/oceans/CARINA, Key et al., 2010). These products contain carbon relevant data from 48 and 98 cruises for the Atlantic Ocean, respectively. Both data products have gone through rigorous quality control procedures to assure the highest possible quality and internal consistency (e.g. Stendardo et al., 2009; Pierrot et al., 2010). Together these data collections form the most comprehensive and consistent data set for carbon related water properties ever gathered for the Ocean to date. The combination of these products is suitable for assessments of oceanic carbon inventories and uptake rates. However, there are a few known deficits to the GLODAP data, and a few duplications with CARINA. Thus the GLODAP data were modified in the following manner: (1) Cruise 45 (TTONAS_1–7) DIC and Alkalinity data are adjusted accordingly to (Tanhua and Wallace, 2005). (2) Cruise 23 (OACES93) was overcorrected for oxygen in GLODAP; therefore oxygen is adjusted by $-7.5\,\mu\text{mol kg}^{-1}$ as suggested by (Sabine et al., 2005). 3) Cruise 24 (3230CHITHER2_1–2) is adjusted for alkalinity by $-8\,\mu\text{mol kg}^{-1}$ (Velo et al., 2009). (4) GLODAP cruises 2, 3, and 29 are also available in both data collections, but with additional data in CARINA. To avoid using the same cruise twice, duplicate cruises are excluded from the GLODAP Atlantic data. Furthermore, we did not use any of the GEOSECS data in our calculation because of the large and variable biases in DIC for this data set, up to $27\,\mu\text{mol kg}^{-1}$ (e.g. Peng and Wanninkhof, 2010).

The property AOU (Apparent Oxygen Utilization) is calculated as the difference between measured oxygen concentration and the calculated saturation of oxygen at the temperature and salinity of the sample using the solubility of Weiss (1970). We use AOU to separate changes in oxygen concentration due to change in heat content or salinity (changes that will change the equilibrium concentration of oxygen) from changes due to ventilation/circulation or respiration. Furthermore, AOU is less sensitive to seasonal variations than dissolved O_2 because it compensates for seasonality in mixed layer temperature, and non-linearity issues in the oxygen solubility is less of an issue if AOU is used. The property DIC_{abio} refers to dissolved inorganic carbon that has been corrected for dissolution of organic matter according to:

$$DIC_{abio} = DIC - 0.67 \times AOU \tag{1}$$

The factor 0.67 is the ratio $C/-O_2$ for remineralization of organic matter. This relation neglects the effects of dissolution/formation of calcium carbonate, i.e. changes in alkalinity, assuming that changes in alkalinity over the short time periods in question are negligible. The storage rate of DIC_{abio} thus reflect changes in DIC not due to respiration of organic matter, and thus may better reflect changes in C_{ant} than the storage rate of DIC does.

2.2 Storage rate calculations

As the first step in the analysis, stations with measurements of DIC and oxygen from the surface to at least 2000 m depth with sufficient vertical resolution to allow for reasonable interpolation the data were identified in the combined CARINA/GLODAP data collections. For these stations we vertically interpolated the data using a piecewise hermite interpolating polynomial routine. The maximum vertical distance over which interpolation was allowed was 65 m in the top 100 m, 205 m between 90 and 300 m depth, 405 m between 300 and 750 m depth, 505 m between 750 and 1500 m depth, and 705 m below 1500 m depth. If the distance between two samples exceeds these definitions, the column inventory was not calculated. These limits represent a delicate balance between a rigorous definition that tend to exclude a large fraction of the stations due to too sparse vertical sampling and a too generous definition that risk creating bad interpolation values in sharp gradients, thus biasing the column inventory estimate. The column inventory was calculated by integrating the interpolated concentration profile from the surface down to 2000 m depth for DIC, DIC_{abio}, oxygen and AOU for each one of the stations. At this stage we converted the gravimetric units reported in CARINA/GLODAP to volumetric units so that:

$$\text{column inventory} = \int_0^{2000} c \times \text{rho}\, dz \tag{2}$$

where C is the vertically interpolated concentration in gravimetric units, and rho is the density. We then located stations sampled close to each other (see below) and calculated the difference between the column inventories of the stations in the pair. The storage rate was calculated as the difference in column inventory of a station pair divided by the time in years between the repeats of this station so that the unit of storage rate is mol m^{-2} yr^{-1}.

For most regions there are only small changes in the property fields below 2000 m depth on decadal time-scales, thus analytical uncertainties and data-biases tend to the dominate signal merely introducing noise in the data with little additional information. In addition, by always calculating the storage rate over 2000 m water column, the different stations pairs can easily be compared to each other. Changes in DIC and oxygen below 2000 m depth were therefore neglected. Another issue is changes in column inventories due to changes in season, i.e. one could expect the column inventory of oxygen and DIC should be different in summer than in winter, everything else being constant. Thus, if the repeat stations are occupied during different seasons, the storage rate calculation might be affected. In order to investigate the effect of sampling during different seasons we conducted one analysis based on the number of months difference in the sampling for the station pair, and one analysis where the top 200 m of the water column, (assuming that this is where the seasonal signal can be detected) was excluded. Sampling during different time of the year has only a small, and mostly insignificant, effect of the storage rate calculation. This seems to be mainly due to the surface mixed layer affected by seasonality being relatively small in comparison to the 2000 m of water column we are analyzing, and that the seasonal changes are relatively small. All station pairs were included in the analysis independent of the time of year the stations were sampled, realizing that this could potentially bias the analysis or at least increase the scatter in the data, but with the benefit of having significantly more data available for the analysis.

The Atlantic Ocean was divided into 7 areas in order to resolve differences in the storage rate for different areas; (A) the western basin of the subpolar north Atlantic, (B) the eastern basin of the subpolar north Atlantic, (C) the western basin in the subtropical north Atlantic, (D) the eastern basin of the subtropical north Atlantic, (E) the tropical Atlantic between 15° N/S, (F) the western basin of the south Atlantic, and (G) the eastern basin of the south Atlantic (e.g. Fig. 1). No distinction was made between subpolar and subtropical south Atlantic due to limited number of data in the south Atlantic.

3 Results

With the combined CARINA/GLODAP data set as described above, 2328 station-pairs in the Atlantic Ocean where we could calculate the storage rate from the surface to 2000 m depth were identified. If we increase the maximum allowed distance between stations to 200 km instead of 100 km, 6757 station pairs was found, but with significant more noise in the result, i.e. 200 km distance is generally too large for considering them being a repeat station. Because of the inherent uncertainty in the change in column inventory the scatter in the storage rate for any station-pairs decreases with time between the repeats, i.e. the signal to noise ratio improves with increasing time between repeats. It seems that a minimum time between repeats of a station of 6 yr was a reasonable minimum time for this study. By excluding repeats of less than 6 yr apart, the number of available repeats drops to 1204. It is interesting to note that the more than 14 000 stations available for the Atlantic Ocean in the CARINA/GLODAP data collection only results in ∼ 1200 station-pairs that qualify for being repeat stations as described above. Apart from stations being too far apart from each other, a large fraction of the stations do not have sufficient vertical resolution to make meaningful vertical interpolation of the profiles. The insufficient vertical resolution for several of the profiles (particularly in the upper ocean) will most likely also affect attempts to interpolate any property over the entire basin for calculating inventories of, for instance, anthropogenic carbon. The storage rates, their standard deviations and the 95 % confidence intervals (CI) for all regions and variables are listed in Table 1. Figures of the spatial distribution of storage rates for DIC, $\mathrm{DIC_{abio}}$, oxygen and AOU are shown in Figs. 1–4, and Fig. 5 graphically represent the information contained in Table 1. The standard deviation mainly reflects uncertainty due to regional variations and uncertainties in measurement and vertical interpolation, whereas the 95 % confidence interval reflects the uncertainty of the mean trends.

3.1 DIC and $\mathrm{DIC_{abio}}$ storage rates

Changes in the column inventory per year, i.e. the storage rate, of DIC for the Atlantic Ocean, are viewed in Fig. 1 and listed in Table 1. In this figure all data that pass the criteria for a valid repeat measurement mentioned above are plotted; bluish colors for decreasing, and green or reddish colors for increasing, column inventories of DIC. The mean rate for all areas is positive, i.e. on average, and as expected, there is an increase in the column inventory of DIC with time. Even though for almost all areas the uncertainty (one standard deviation of all estimates) encompasses zero storage rate (i.e. no change in column inventory), the 95 % confidence interval of the mean DIC storage rate indicate a positive storage rate everywhere, except region F, the southwest Atlantic. The average storage rate is generally between 0.6–0.9 $\mathrm{mol\,m^{-2}\,yr^{-1}}$ for the north Atlantic (regions A to D), somewhat lower for the tropical and southwest Atlantic, whereas the southeast Atlantic shows a large increase in DIC. For the whole Atlantic, the average storage rate is is DIC 0.72 ± 1.22 (CI: 0.65–0.78) $\mathrm{mol^{-2}\,yr^{-1}}$. A different view of the distribution of the storage rates is provided by the histograms in the right hand panel of Fig. 1, where additionally the mean value is indicated together with 95% confidence interval.

The averaged storage rates for the northern North Atlantic (regions A and B) are generally somewhat smaller than previously reported storage rates of anthropogenic carbon in this area (e.g. Sabine and Tanhua, 2010). One obvious reason for these changes is that this method only evaluates the changes in the water column above 2000 m depth whereas the published literature generally analyzes the whole water column. For the north Atlantic a significant amount of anthropogenic carbon has penetrated the water column to more than 2000 m depth (e.g. Tanhua et al., 2007; Sabine and Tanhua, 2010; Pérez et al., 2010). This bias can probably be up to about 0.5 $\text{mol m}^{-2}\,\text{yr}^{-1}$, but is obviously heavily dependent on the region, i.e. on the presence of recently ventilated deep water. Perfect agreement is not expected, however, because other published methods tend to implicitly correct for changes DIC caused by biological activity and circulation in order to calculate the "anthropogenic carbon".

Similarly for the subtropical North Atlantic (regions C and D) we find storage rates of about 0.7–1.1 $\text{mol m}^{-2}\,\text{yr}^{-1}$. This is comparable to, and within the uncertainty of, previously published results using various approaches to calculate the storage rates: In general, the eMLR based estimate by (Wanninkhof et al., 2010; Peng and Wanninkhof, 2010) are in the lower range of the storage rates in this study, whereas slightly higher storage rates are found by Tanhua et al. (2007), see also Sabine and Tanhua (2010).

For the southeast Atlantic, region G in Fig. 1, we find the highest inventory rate of DIC of all our areas in the Atlantic. This is in contrast to the results presented by Murata et al. (2008) who found an inventory rate of only 0.43–0.49 $\text{mol m}^{-2}\,\text{yr}^{-1}$, (although for C_{ant}) partly using the same data as in this study (i.e. the repeats of WOCE section A10 in 1993 and 2003). In the southwest Atlantic (region F) we find storage rates of DIC insignificantly larger than the "no change" condition i.e. the storage rate is insignificantly different from zero, Table 1. However, (Wanninkhof et al., 2010) found find high storage rates (0.76 $\text{mol m}^{-2}\,\text{yr}^{-1}$) in the Southwest Atlantic along the WOCE section A16, i.e. in region F in Fig. 1. Similarly, Ríos et al. (2012) also finds high (0.92 ± 0.13 $\text{mol m}^{-2}\,\text{yr}^{-1}$) storage rates of C_{ant} for the southwest Atlantic Ocean.

Changes in “respiration corrected” DIC (DIC_{abio}) reflect, in principle, the increasing uptake of CO_2 at the ocean surface due to increasing atmospheric CO_2 concentrations, Fig. 2, equation 1. DIC_{abio} should be largely conserved in the ocean interior, but could change in surface water due to either air-sea exchange of CO_2 or O_2. Changes in column inventory of DIC_{abio} will therefore largely reflect a combination of the effects of long-term CO_2 and O_2 exchange with the atmosphere, with the CO_2 effect presumably dominating as a result of the uptake of anthropogenic CO_2. The well-known pattern of C_{ant} column inventory, i.e. high values in the subpolar North Atlantic (SPNA), low in the tropics and intermediate values in the subtropics of both hemispheres, are not well reflected in our maps of DIC and DIC_{abio} storage rates (Figs. 1 and 2). Interestingly, we find DIC_{abio} storage rates near zero for the subpolar North Atlantic, a region where large positive storage rates has been reported for C_{ant} (e.g. Friis et al., 2005; Pérez et al., 2008, 2010). For the western SPNA (region A) where significant storage of C_{ant} can be expected we find moderate storage rates of DIC, and neutral storage rates of DIC_{abio}. This is particularly evident for the northern portion of region B, i.e. in the Iceland Basin. In this data set, there are signs of higher than average increase of DIC and DIC_{abio} off the Iberian Peninsula, in the southeast Atlantic, off Florida and close to the Charlie-Gibbs Fracture Zone, and possibly in the northwest subtropical Atlantic.

3.2 Oxygen and AOU storage rates

For oxygen and AOU a somewhat different picture emerges (Figs. 3 and 4). For the Atlantic Ocean as a whole there is a negative storage rate of oxygen; the average of all our data is -0.54 ± 1.64 (CI: -0.64–(-0.45)) $\mathrm{mol\,m^{-2}\,yr^{-1}}$. The storage rates of oxygen and AOU are significantly different from zero for regions A and B only, as well for the average over all regions. Particularly, significant decrease in oxygen (and increase in AOU) column inventories are observed in the Labrador Sea, the Irminger Sea and in the northern part of the Iceland Basin. The change in AOU, and the confidence interval of the change, is somewhat smaller than that for oxygen indicting that some of variability is tied to changes in solubility, mostly due to changes in temperature of the water. This solubility component of the O_2 changes must be closely tied to the change in the inventory of heat. For instance, out-gassing of oxygen due to a warming ocean will not cause any directly changes in AOU, i.e. changes in AOU are indicative of air-sea O_2 fluxes driven by biology or circulation. For the regions outside of SPNA, no significant change in the column inventory of oxygen or AOU can be detected with this method.

3.3 Temporal variations

In order to identify any temporal trends in storage rates, the data on storage rates for three different time periods are evaluated. Station pairs where both repeats are conducted in the any of the time-periods 1980–1995, 1990–2000, or 1995–2005 were identified. This required discarding additional pairs done more than 15 (or 10) years apart, which further increases the uncertainty of the storage rates for each area. Since the resulting coverage is very sparse in most regions, we focus on the two northernmost regions (A – the northwestern part of the SPNA, and B – the northeastern portion of the SPNA) where more data is available, and where significant changes in deep water formation has occurred over time (e.g. Rhein et al., 2011). The data for the 3 time periods are displayed in Figs. 6–9 for DIC, DIC_{abio}, oxygen and AOU and respectively. In Fig. 10 the information for regions A and B is condensed.

It can be noted that, as expected, significant spatial variations is present, even within each region, but that some interesting patterns can be recognized. For the first time slice (1980–1995) an increase in oxygen and DIC can be observe for both regions (i.e. positive storage rates), particular for region B, although only a limited number of station pairs are available to confirm this trend. During the1990s the conditions are significantly different with negative storage rates of oxygen and close to neutral storage rates of DIC. During the last time slice (1995–2005) again a different picture emerges with different patterns for the western and the eastern domain. For region A, the storage rate for oxygen is continuously negative whereas the DIC is neutral or slightly positive; region B show positive storage rates of DIC but neutral oxygen storage rates. It is clear that there are both temporal and spatial variability in the storage rates of DIC and oxygen in the North Atlantic subpolar gyre.

4 Discussion

The most complete, quality controlled, interior ocean carbon data set for the Atlantic Ocean currently available have been used for this analysis. A method to quantify changes in DIC and oxygen in the water column using a minimum of assumptions has been applied to these data. This has resulted in a baseline estimate of decadal changes in DIC and oxygen that can constrain other, more detailed, estimates of storage rates. The significance of these results is that they provide an independent check on the storage rates of carbon and oxygen in the Atlantic Ocean. The results of this study confirms that there is a general increase in DIC in the interior ocean, just as expected from the perturbation of atmospheric CO_2 concentrations, and a small decrease of oxygen in the upper 2000 meters of the Atlantic Ocean, particularly north of about 50° N where the decreasing trend is significant.

Based on estimates of the total storage of anthropogenic carbon in the world ocean, the globally averaged storage rate for C_{ant} has been increasing from roughly $0.2\,\mathrm{mol\,m^{-2}\,yr^{-1}}$ in 1960 to $0.6\,\mathrm{mol\,m^{-2}\,yr^{-1}}$ in 2007 (Khatiwala et al., 2009). The storage rate is expected to show significant regional variability assuming that the regional pattern of storage rate is similar to that of the total storage, see for instance the Atlantic Ocean map of C_{ant} column inventory in Lee et al. (2003). However, there are a few important differences between this study and the calculation by Lee et al. (2003). Most importantly this study reports on the change in DIC (or DIC_{abio}) which is not equal to the change in C_{ant} so that temporal changes in the storage rate of DIC which is not evident by observing the total storage of C_{ant} becomes relevant. The regional pattern of storage rate of DIC in this analysis is significantly different than the well-known distribution of column inventory of C_{ant} in the Atlantic Ocean. In general, a mixed pattern of positive and negative storage rates are found in each region. The picture generally gets somewhat less patchy when considering only shorter time-periods, Figs. 6–9. Since this method of calculating storage rates does not account for small-scale temporal and spatial variability due to, for instance eddies and movements of oceanic fronts, larger variability in the storage rate is expected than from methods that do compensate for this, such as MLR based approaches. The larger scatter also reflect the additional difficulties in determining inventory changes for the total amount of DIC rather than the anthropogenic perturbation (C_{ant}), see discussion below.

It is interesting to compare our result with the results presented by Steinfedt et al. (2009) who observed only a weak increase in the C_{ant} column inventory (2 %) in the Labrador Sea and Irminger Sea during the 1997–2003 time-period based on transient tracer observations. This is roughly the same time-period (1995–2005) for which we find a weak increase in the column inventory of DIC but neutral DIC_{abio} storage rates in the same region. A water mass analysis suggest that the volume of the classic Labrador Sea Water (LSW) decreased due to decreasing deep water formation in the Labrador Sea, and as a consequence there is an increase of a lighter (i.e. less dense) version of the LSW (e.g. Rhein et al., 2011; Steinfeldt et al., 2009). Thus the deep version of the LSW only experience limited ventilated during this time, i.e. the LSW is getting “older”. This provides an explanation to the observation that the column inventory of C_{ant} remains close to constant (restricted communication with the increasing atmospheric CO_2 concentrations) and why the DIC is increasing slightly (remineralization of organic matter), DIC_{abio} remains constant and oxygen concentrations decreases during this time period. This analysis thus confirms the conclusion by (Steinfeldt et al., 2009) that the northwest subpolar gyre of the Atlantic Ocean is a region that shows significant deviation from the expected average uptake rate of anthropogenic carbon.

Our analysis can also be compared with Perez et al. (2008), who studied a time series of biogeochemistry data in the southern Irminger Sea. They note a trend of increasing AOU for the LSW, starting in the mid-1990s, which is similar to the observations in this study. By using a data-based back-calculation method to infer the C_{ant} concentration, they conclude that there is a significantly positive storage rate of C_{ant}

for the time-period 1981 to 2006 ($1.1 \pm 0.1\,\mathrm{mol\,m^{-2}\,a^{-1}}$), and a particularly large storage rate during the time-period 1991–1997 ($2.3 \pm 0.6\,\mathrm{mol\,m^{-2}\,a^{-1}}$). The trends in C_{ant} concentration for the water masses in the SPNA is also discussed in detail by Pérez et al. (2010), where they divide the region into three areas, as opposed to the two areas in this study. They found positive storage rates for C_{ant} for the time period 1981 to 2006 for the whole SPNA. Similar to the study from 2008 (Perez et al., 2008) for the Irminger Sea, they also find large differences in storage rates with time, and correlate this to the North Atlantic Oscillation (NAO) and the formation rate of Labrador Sea Water. They find particularly high storage rates during the period 1991 to 1998 (i.e. during the time of intense formation of LSW) for the Irminger Sea and the Iceland Basin, whereas the East North Atlantic Basin seem to have a more linear increase of storage rate for C_{ant} ($0.77 \pm 0.03\,\mathrm{mol\,m^{-2}\,a^{-1}}$) (Perez et al., 2008, 2010). This overlaps with the time-period (1990–2000) where we find slightly negative storage rates for DIC and $\mathrm{DIC_{abio}}$. However, since the analysis in this study is only covering the upper 2000 m of the water column, storage changes in the deeper part of the water column remains unaccounted for. For this region with active deep water formation and significant advection of overflow water the deeper parts might indeed be important and is a source of error in this comparison. Significant temporal variations in T, S and dissolved O_2 occurred in the SPNA during the last ~60 yr are also reported by van Aken et al. (2011) who conclude that the long-term variations of the intermediate water mass properties in the SPNA are related to meteorological forcing of the Labrador Sea. Significant changes in the SPNA salinity balance has been observed during the last half century (Curry and Mauritzen, 2005) which can be consistent with varying dominance of different water masses. Based on this analysis it seems that these long-term variations also affect the inventory of DIC in the SPNA.

A detailed study of the temporal evolution of the inorganic carbon content in the SPNA is out of scope for this study. It is, however, interesting to point out the diverging trends of C_{ant} and DIC found in this region may relate to varying convection activity and water mass distribution. It seems that the inventory of DIC decreases at the same time as the inventory of C_{ant} increases. An explanation for this can be provided by changes in water mass distribution in the SPNA. For instance, the DIC concentration of LSW is in the order of 2160 µmol kg^{-1} whereas the DIC concentration of the Mediterranean Sea Overflow Water (in the Gulf of Cadiz) is in the order of 2200 µmol kg^{-1}, even though both water masses have relatively high, and somewhat similar concentration of C_{ant}. These two water masses are both present in the SPNA and variability in the relative presence of these two water masses will change the column inventory of DIC in a way that is not necessarily reflected in storage of C_{ant}.

Recently, Stendardo (2011) has used a long-term dataset for dissolved oxygen in the North Atlantic Ocean to assess any trends over the past 49 yr. She finds a complex pattern of temporal changes in oxygen concentrations; the upper water masses have generally lost oxygen, particularly in the eastern and northern Atlantic, whereas deeper layers have generally gained oxygen, particularly in the southwestern part of the North Atlantic. The results are based on observed changes in oxygen concentration for different density intervals (i.e. water masses), neglecting temporal changes in the thickness of these water masses over time. In a more detailed study focusing of repeats of the A2 sections (i.e. a zonal section across the Atlantic Ocean at $\sim 47^\circ$ N), Stendardo (2011) concludes that the oxygen concentrations show strong inter-annual variability with a tendency towards oxygen loss over time. The results presented in this study are, in general, supporting the results of Stendardo (2011) with particularly large losses of oxygen in the northern part of the North Atlantic. However, the results are difficult to compare direct as this study is focusing on the changes in column inventory rather than the concentration in various water masses.

5 Concluding remarks

A simple method associated with few assumptions to constrain the ocean storage rate of dissolved inorganic carbon (DIC), respiration corrected DIC (DIC_{abio}), oxygen, and apparent oxygen utilization (AOU) has been demonstrated. By calculating the difference in the column inventory of these properties down to 2000 m depth over the whole Atlantic Ocean for a large number of repeat stations we find generally find increase in DIC and a decrease of oxygen. The trends reported in this analysis supports other studies that have reported on increasing concentrations of inorganic carbon in the ocean based on more complicated schemes, and providing some support for the general trend of ocean deoxygenation. The degree of uncertainty in calculating trends in interior ocean properties with this method demonstrates the importance of small scale spatial and temporal variability in the ocean. One important aspect of this analysis is that variations in water mass prevalence have a large influence on inventories of interior ocean properties so that the total DIC inventory can decrease in an area even if the inventory of C_{ant} significantly increases. This has implications for balancing the global carbon budget that do not distinguish between "anthropogenic carbon" and "natural carbon". From a global perspective it is the overall increasing or decreasing inventory of carbon in the ocean that matters to balance the budget.

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Table 1. Averaged storage rates of DIC, DIC$_{abio}$, Oxygen and AOU for the 7 regions (see Fig. 1) and for the whole area. All repeats separated by at least 6 years are considered. The uncertainty is calculated as one standard deviation and only reflects the variability of estimates within a region, uncertainties due to the vertical interpolation or the analytical uncertainties are not considered. The second row in each box is the 95 % confidence interval of the estimate. The number of samples (i.e. station pairs, *N*) for each area is indicated in the left column.

Region	DIC mol m^{-2} yr^{-1}	DIC$_{abio}$ mol m^{-2} yr^{-1}	Oxygen mol m^{-2} yr^{-1}	AOU mol m^{-2} yr^{-1}
A – subpolar NW	0.59 ± 1.23	−0.14 ± 1.42	−1.63 ± 1.39	1.05 ± 0.97
N = 226	0.42–0.75	−0.33–0.05	−1.81–(−1.44)	0.93–1.18
B – subpolar NE	0.64 ± 1.29	−0.02 ± 1.45	−1.33 ± 1.63	0.97 ± 1.28
N = 256	0.48–0.80	−0.20–0.16	−1.53–(−1.13)	0.82–1.14
C – subtropical NW	0.93 ± 1.25	0.86 ± 1.41	0.19 ± 2.02	0.01 ± 1.60
N = 267	0.77–1.08	0.69–1.03	−0.05–0.44	−0.18–0.20
D – subtropical NE	1.03 ± 1.04	0.85 ± 1.03	−0.15 ± 1.42	0.23 ± 1.38
N = 240	0.89–1.17	0.72–0.99	−0.34–0.04	0.05–0.42
E – tropics	0.41 ± 0.91	0.56 ± 0.72	0.21 ± 1.06	−0.19 ± 1.11
N = 99	0.23–0.59	0.42–0.70	0.00–0.42	−0.41–0.03
F – SW Atlantic	0.24 ± 1.22	0.30 ± 1.05	−0.01 ± 0.66	−0.09 ± 0.81
N = 100	0.00–0.49	0.09–0.52	−0.14–0.12	−0.25–0.07
G – SE Atlantic	2.11 ± 1.21	2.09 ± 1.13	−0.36 ± 0.96	0.10 ± 0.78
N = 27	1.63–2.59	1.64–2.53	−0.75–0.02	−0.21–0.41
The whole Atlantic	0.75 ± 1.23	0.44 ± 1.37	−0.58 ± 1.71	0.44 ± 1.35
N = 1204	0.68–0.82	0.36–0.52	−0.68–(−0.48)	0.36–0.52

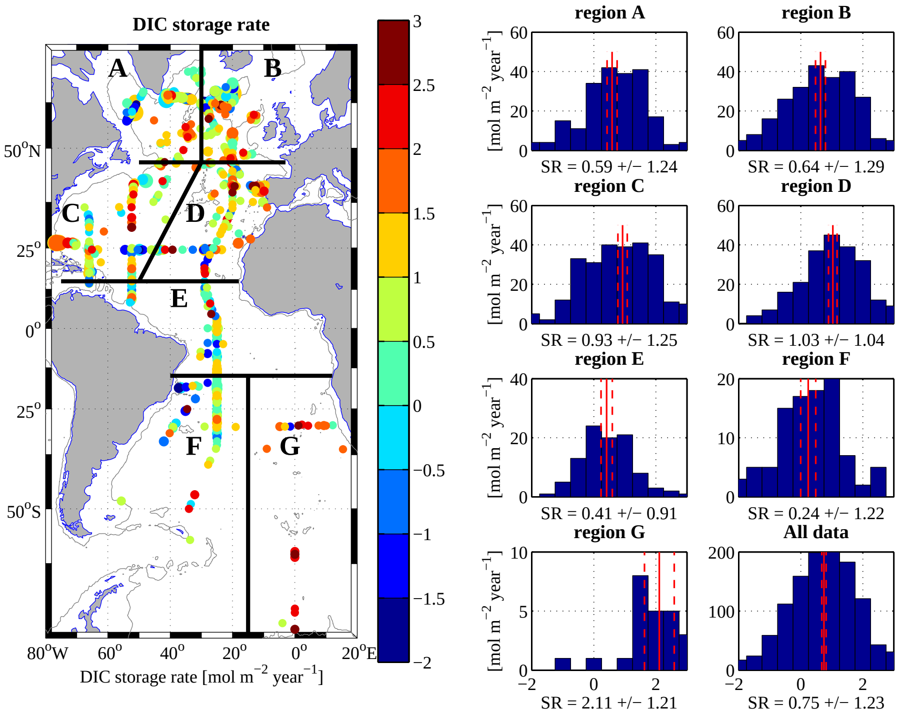

Fig. 1. Change in column inventory between two repeats of the same position, i.e. the storage rate (mol m^{-2} yr^{-1}) for DIC in the Atlantic Ocean. Left side panel: The average of storage rates for each location is shown with the color-coded markers, the sizes of the markers are made proportionally larger depending on the number of repeats at each position. The 2000 m isobath is marked with a gray thin line. Right hand panels: Histograms of the distribution of storage rates for the 7 regions and for the sum of all the regions. The average value and the 95 % confidence intervals are marked with red vertical lines. The numbers under each panel is the average and standard deviation of all the repeats for that region (SR = Storage Rate).

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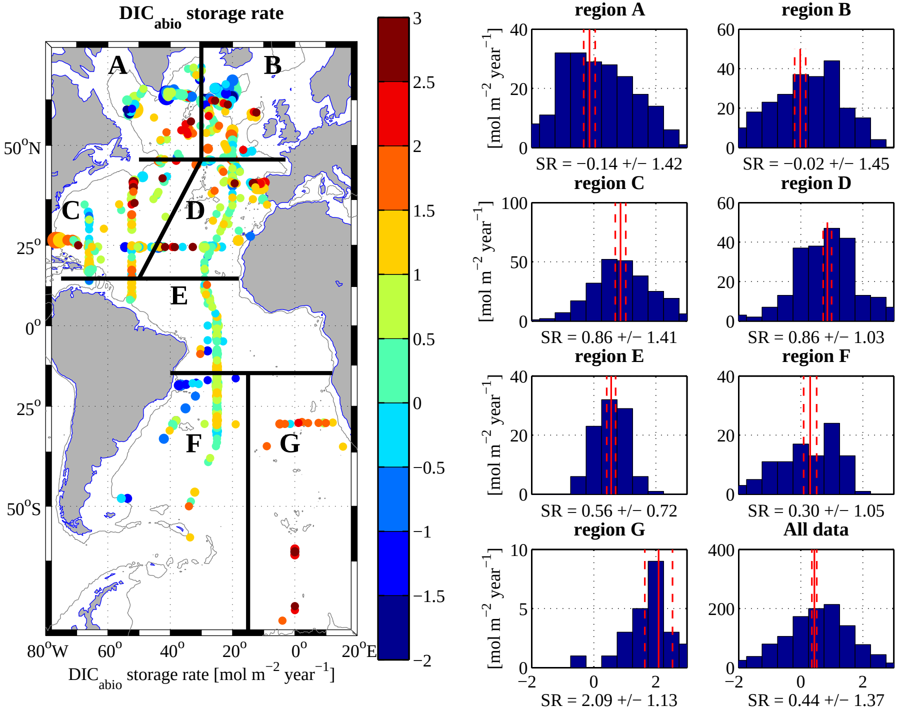

Fig. 2. Same as Fig. 1 but for storage rates (mol m^{-2} yr^{-1}) of DIC$_{abio}$.

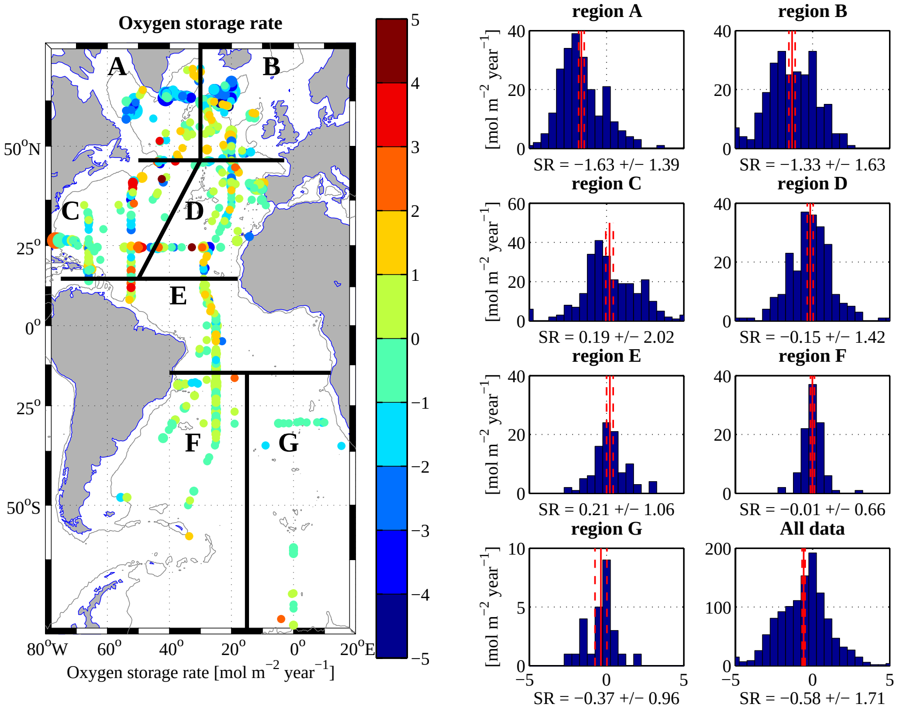

Fig. 3. Same as Fig. 1 but for storage rates (mol m^{-2} yr^{-1}) of oxygen.

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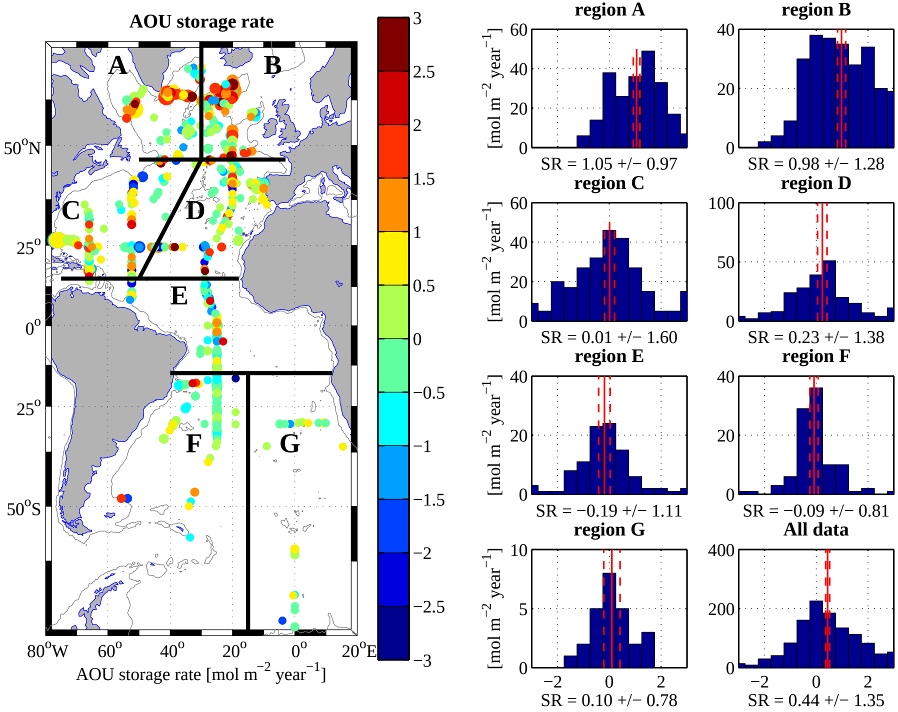

Fig. 4. Same as Fig. 1 but for storage rates (mol m^{-2} yr^{-1}) of AOU.

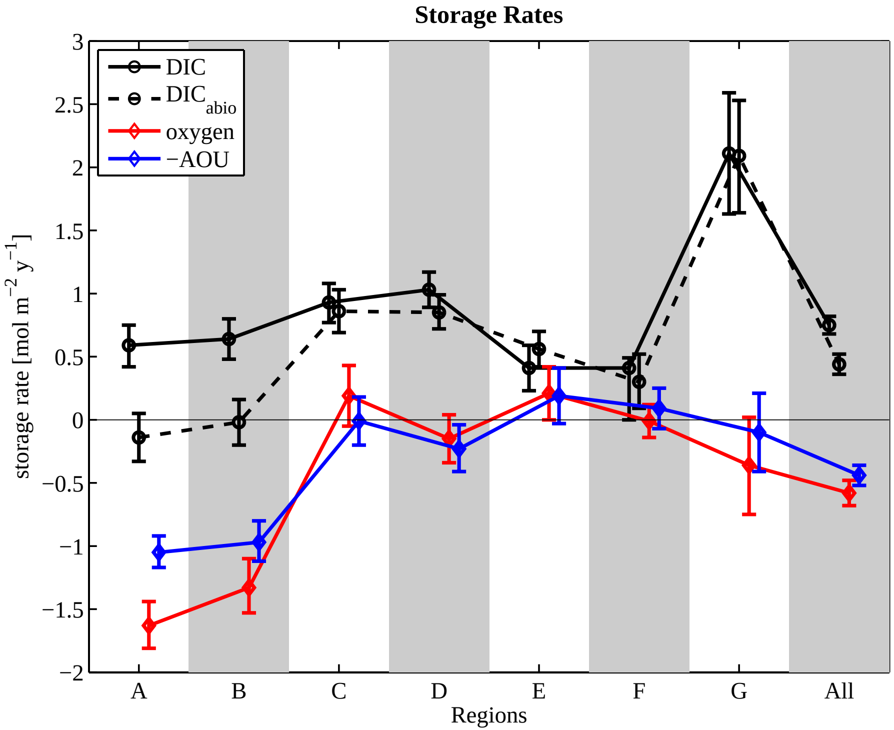

Fig. 5. Graphical representation of the storage rates for DIC, DIC$_{abio}$, oxygen and AOU for the 7 regions (A–G) as well as the whole Atlantic Ocean. The vertical error bars represent the 95 % confidence interval of the mean trend.

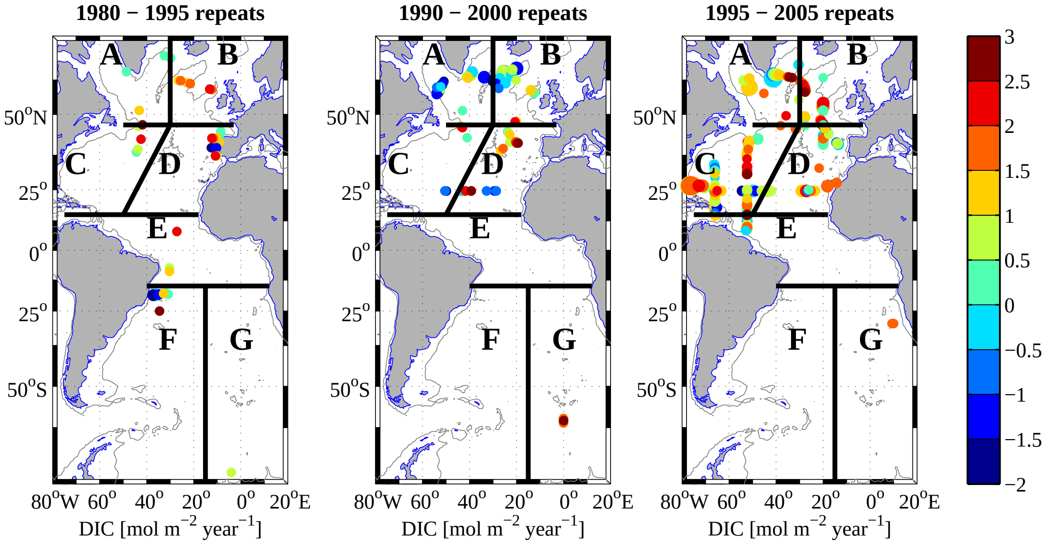

Fig. 6. Storage rates for DIC for three different time periods. Left panel – storage rates for repeats where both cruises were conducted between 1980 and 1995; Middle panel – both repeats were conducted between 1990 and 2000; Right panel – both repeats were conducted between 1995 and 2005. The 2000 meter isobath is marked with a gray thin line.

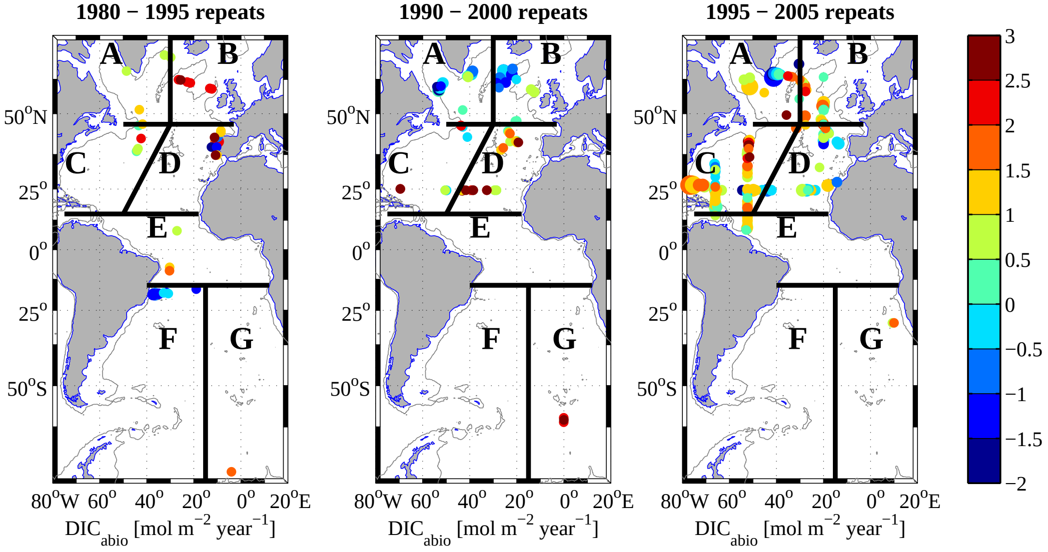

Fig. 7. Same as Fig. 5 but for storage rates (mol m^{-2} a^{-1}) of DIC$_{abio}$.

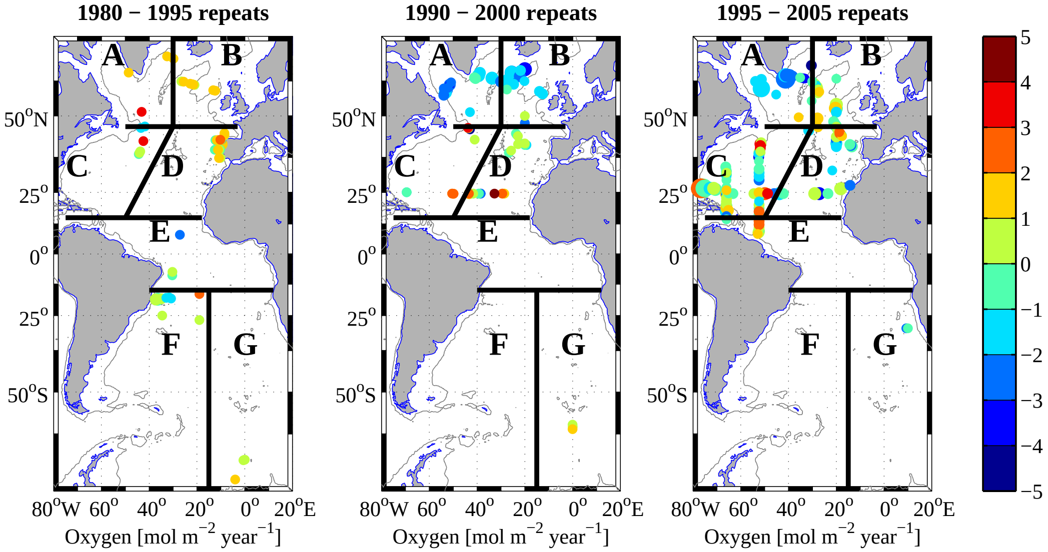

Fig. 8. Same as Fig. 5 but for storage rates (mol m^{-2} a^{-1}) of AOU.

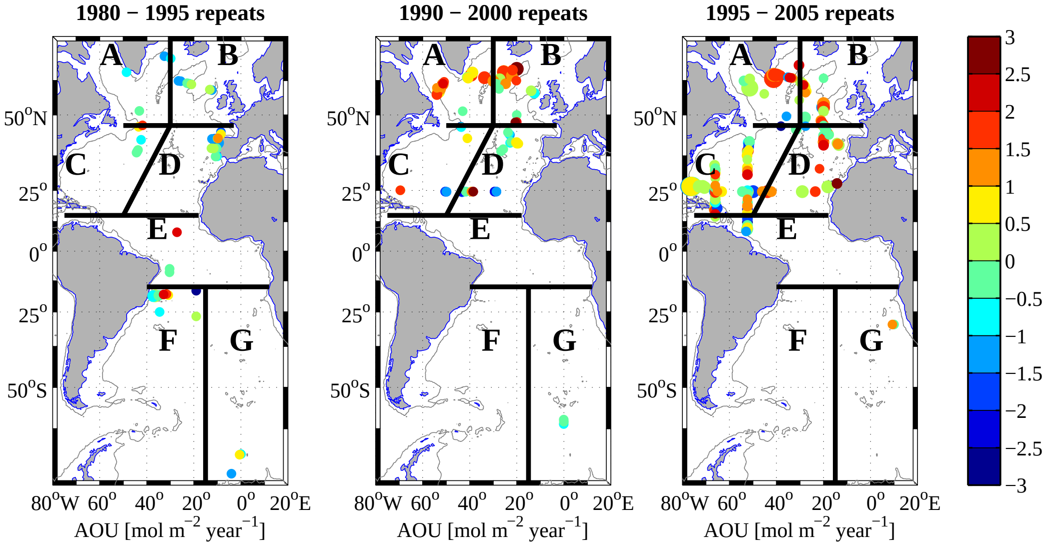

Fig. 9. Same as Fig. 5 but for storage rates (mol m^{-2} a^{-1}) of oxygen.

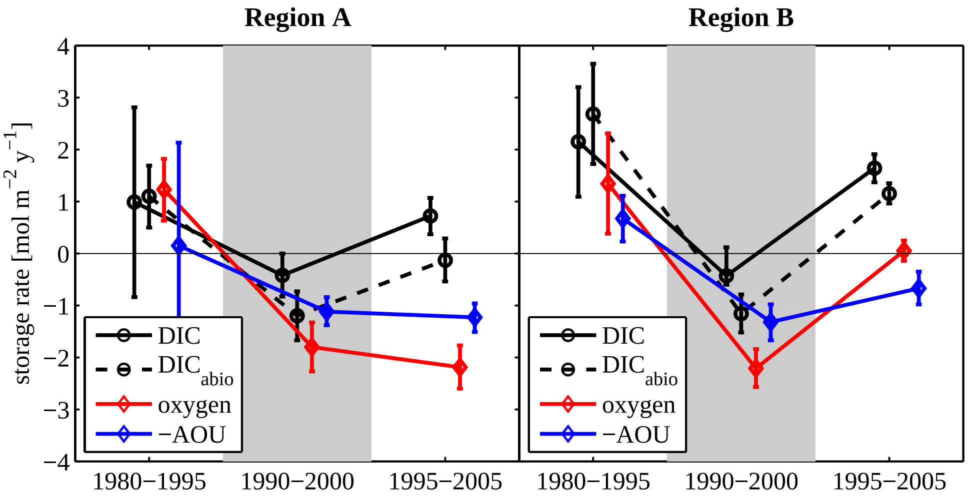

Fig. 10. Storage rates for DIC, DIC$_{abio}$, AOU and oxygen for regions A and B for three time periods (1980–1995, 1990–2000, 1995–2005), see Figs. 5–8. Note that negative AOU is plotted and that the markers are slightly offset for clarity. The error-bars represent the 95 % confidence interval of the mean trend.