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Abstract

In January and February 1998, when an unprecedented fourth repetition of the zonal hydrographic transect at 24.5°N in the Atlantic was undertaken, carbon measurements were obtained for the second time in less than a decade. The field of total carbon along this section is compared to that provided by 1992 cruise which followed a similar path (albeit in a different season). Consistent with the increase in atmospheric carbon levels, an increase in anthropogenic carbon concentrations of $8\pm3\,\mu\text{mol kg}^{-1}$ was found in the surface layers. Using an inverse analysis to determine estimates of absolute velocity, the flux of inorganic carbon across 24.5° is estimated to be $-0.74\pm0.91$ and $-1.31\pm0.99\,\text{Pg C yr}^{-1}$ southward in 1998 and 1992, respectively. Estimates of total inorganic carbon flux depend strongly upon the estimated mass transport, particularly of the Deep Western Boundary Current. The 1998 estimate reduces the large regional divergence in the meridional carbon transport suggested by previous studies and brings into question the idea that the tropical Atlantic constantly outgasses carbon, while the subpolar Atlantic sequesters it. Uncertainty in the carbon transports themselves, dominated by the uncertainty in the total mass transport estimates, are a hindrance to determining the “true” picture.

The flux of anthropogenic carbon ($C_{\text{ANTH}}$) across the two transects is estimated as northward at $0.20\pm0.08$ and $0.17\pm0.06\,\text{Pg C yr}^{-1}$ for the 1998 and 1992 sections, respectively. The net transport of $C_{\text{ANTH}}$ across 24.5°N is strongly affected by the difference in concentrations between the northward flowing shallow Florida Current and the mass balancing, interior return flow. The net northward transport of $C_{\text{ANTH}}$ is opposite the net flow of total carbon and suggests, as has been found by others, that the pre-industrial southward transport of carbon within the Atlantic was stronger than it is today. Combining these flux results with estimates of atmospheric and riverine inorganic carbon input, it is determined that today’s oceanic carbon system differs from the pre-industrial system in that today there is an uptake of anthropogenic carbon to the south that is advected northward and stored within the North Atlantic basin.

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1. Introduction

Throughout the last decade approximately 6.2 PgC yr\(^{-1}\) (1 PgC = 1 GtC = \(10^{15}\) g of carbon) have been released into the atmosphere through the burning of fossil fuels (IPCC, 2001; Battle et al., 2000). Meanwhile, the amount of carbon in the atmosphere has increased at a rate of only 2.8 PgC yr\(^{-1}\) (1 PgC yr\(^{-1}\) = 2642 kmol C s\(^{-1}\)). The increased partial pressure of atmospheric CO\(_2\) has caused, through air–sea exchange, the amount of carbon stored in the ocean to increase by about 2.0 ± 0.6 PgC yr\(^{-1}\) (IPCC, 2001; Battle et al., 2000; Schimel et al., 1996; Siegenthaler and Sarmiento, 1993). These estimates strongly suggest that the ocean and ocean circulation are significant players in the global carbon cycle and control, in part, the rate of the atmospheric increase in CO\(_2\).

Just where the oceanic uptake of anthropogenic CO\(_2\) is occurring and how circulation patterns affect the sources and sinks of carbon within the oceans are not well understood. Until recently direct oceanic observations have been sparse and numerical models, which have been extremely good at producing similar net ocean CO\(_2\) storage estimates, have failed to produce a quantitative consensus on where the uptake and storage of anthropogenic carbon are occurring (Wallace, 2001; Orr et al., 2001). This study seeks to understand better one piece of this puzzle, that is the uptake and transport of carbon within the North Atlantic. We focus on the carbon transport across 24.5°N. Previous estimates of the carbon transport across this line of latitude have been made. The earliest, Brewer et al. (1989) was based on carbon measurements taken at just nine stations occupied in the late fall of 1988, combined with the transport results of the full hydrographic transect made in 1981 (Hall and Bryden, 1982). The increased effort to measure carbon dioxide concentrations in seawater during the World Ocean Circulation Experiment (WOCE), the Ocean–Atmosphere Carbon Exchange Studies (OACES) and the US Joint Global Ocean Flux Study (JGOFS) programs has now provided us with a far more detailed view of the ocean carbon field. Rosón et al. (2002) presented inorganic carbon and anthropogenic carbon transport estimates based on data from the summer 1992 occupation of the Atlantic 24.5°N transect. Their carbon transport estimates along with estimates from previous studies at other latitudes suggested that to the north of 24.5°N the subpolar ocean takes up carbon from the atmosphere, whereas to the south it is released to the atmosphere. We compare their data and results to those from the winter 1998 occupation of the 24.5°N transect, with particular emphasis on the carbon and anthropogenic carbon fields. Calculated meridional transport of inorganic and anthropogenic carbon allows us to provide an updated view of the uptake and storage within the Atlantic Basin.

2. The observations

In 1998, the R.V. Ronald Brown made a high density hydrographic transect across the North Atlantic at 24.5°N (Fig. 1 in black) sponsored by OACES. It was the second transect along this line of latitude to obtain measurements of carbon and carbon related species. The second leg of the cruise, which made these measurements left Las Palmas on January 23, 1998 and arrived in Charleston S.C. 1 month later on February 24. This leg included 130 CTD stations at which oxygen, nutrient and chlorofluorocarbon (CFC) measurements were taken with a 36 position bottle rosette. LADCP data also were obtained, but are not used in this particular study. Selected stations sampled dissolved organic carbon, dissolved inorganic carbon, alkalinity and pH. In this study, total inorganic carbon and total alkalinity were used in the determination of the 1998 pre-anthropogenic component of carbon (Chen and Millero, 1979; Gruber et al., 1996).

The 1998 cruise track was similar to that followed by the 1981 Atlantis cruise (AT109) in that it included a jog to the coast of Africa and sampled the Florida Straits at 27°N. In this study, the 1998 carbon data are compared to those collected in 1992 on the WOCE A5 transect undertaken by the R/V BIO Hespérides (Fig. 1 in red). This cruise track is similar to that of the 1957 IGY transect. It lies to the south of the 1998
track near both coasts and samples the Florida Straits at 26°N. Beginning on July 12, 1992 and ending on August 15, this earlier cruise occupied 112 CTD stations and used a 24-bottle rosette to provide measurements of oxygen, various nutrients, as well as alkalinity and pH, which were used to estimate total inorganic carbon, as few dissolved inorganic carbon measurements were made. The bottle data have been interpolated in neutral density space onto the CTD 2 db depths. In the deep basin, below 5000 db the interpolation is done in pressure space which allows better vertical resolution in more homogeneous waters.

To estimate anthropogenic CO2 on the 24°5′N line, the so-called “\( \Delta C \star \)” method originally developed by Gruber et al. (1996) has been used. This method separates the anthropogenic CO2 component \( C_{\text{ANTH}} \) from the measured total inorganic carbon \( \text{TIC} \) through three corrections: biology \( \Delta C^{\text{BIO}} \), the carbon concentration that the water would have had at the surface in equilibrium with a pre-industrial atmospheric pCO2 of 280 μatm \( (\text{C}^{\text{EQ}}) \), and the air–sea disequilibrium \( \Delta C^{\text{DISEQ}} \).

\[
\begin{align*}
C_{\text{ANTH}} (\mu \text{mol kg}^{-1}) &= \Delta C \star - \Delta C^{\text{DISEQ}} \quad (1) \\
&= \text{TIC}^{\text{MEAS}} - \Delta C^{\text{BIO}} - \text{C}^{\text{EQ}} - \Delta C^{\text{DISEQ}}. \quad (2)
\end{align*}
\]

The \( \Delta C^{\text{DISEQ}} \) can be estimated using either the water mass information from transient tracers or the \( \Delta C \star \) distribution at depths thought to be unaffected by anthropogenic CO2. For direct comparison to the previous study of the 1992 data set, we estimate \( C_{\text{ANTH}} \) here using the method of Rosón et al. (2002), which differs from the one described above in that a constant disequilibrium value, \( \Delta C^{\text{DISEQ}} \), estimated from the \( \Delta C \star \) distribution in anthropogenic CO2-free waters is applied to the entire water column. The disequilibrium term for the 1992 data set is 6.9 ± 5 μmol kg\(^{-1}\) (Rosón et al., 2002) and -11.1 ± 2.0 μmol kg\(^{-1}\) for the 1998 data set. The former value is consistent with the 3.8 ± 4.8 μmol kg\(^{-1}\) suggested by Pérez et al. (2002) for the eastern North Atlantic, while the
latter value is at the upper end of the −24 to −10 μmol kg\(^{-1}\) range of Gruber (1998) for the North Atlantic. A more complete discussion of the anthropogenic carbon calculation is included in the appendix.

The resulting data set includes all available properties, as well as the derived C\(^{\text{\#ANTH}}\) estimates on 2 db pressure intervals. These data are used in a 1998–1992 comparison of the fields themselves and in a box inversion calculation of meridional property fluxes.

2.1. The carbon fields

To compare the data fields, the properties, already on the same vertical grid, were objectively mapped every 50 db onto the same horizontal grid: 0.5° in longitude, between 75.5°W and 16°W (Table 1). The technique used is similar to that described in Lavin (1999), which was in turn fashioned after Fukimori and Wunsch (1991). To handle the discontinuity created by the Mid-Atlantic ridge, below 2750 db the eastern and western basins were treated separately. The smoothing appears to work well everywhere, except within the abyss (below about 6000 m) where the lack of data produces large uncertainties.

2.1.1. Total inorganic carbon

Although differing somewhat in magnitude, at 24.5°N in the Atlantic, the 1998 and 1992 TIC (total inorganic carbon) fields display similar features (Fig. 2, upper panels). Carbon concentrations increase from the surface to depth. They are greater in the east than in the west. Near the surface the lower CO\(_2\) concentrations towards the west are associated with warmer temperatures. Using the 1998 field by way of example, it can been seen that although the top to bottom difference in carbon concentration is only O(280 μmol kg\(^{-1}\)) compared to a mean of O(2100 μmol kg\(^{-1}\)), the strongest vertical gradients are confined, as expected, to the waters above 1000 m. Below 1000 m, the range in concentrations is less than 60 μmol kg\(^{-1}\).

There is a relative maximum in TIC at ~1000 db in the eastern basin associated with the Mediterranean outflow. The weaker maximum in the west at the same depth is identified with the CFC minimum of Upper Circumpolar Deep Water (UCDW). Antarctic Intermediate Water (AAIW) lies above at about 850 db. The TIC minimum which appears to emanate from the western side of the basin encompasses both the Upper and Middle North Atlantic Deep Water (NADW) water masses. These include upper and classical LSW which appear as a single water mass (1200–2300 db) in the 1998 section, the CFC minimum of Iceland–Scotland Overflow Water (at ~2500 db), down to about 3400 db just above the core of Lower NADW recognized by its CFC maximum. In the abyss, particularly in the western basin, higher concentrations are associated with Antarctic Bottom Water.

The largest uncertainties in the interpolation (Fig. 2, lower left panel) occur in the regions of the strongest gradients. In the 1998 transect this is at about 100 m. In the 1992 transect, the uncertainty remains high above 100 m all the way to the surface as the concentration gradients are stronger in the near surface waters during the summer.

Using the estimated errors from the two TIC fields to determine the uncertainty in the differenced fields, it was found that the apparent differences between the 1992 and 1998 TIC concentrations (Fig. 2, lower right panel) were not significant at individual grid points. However, there are clearly patterns within the field. Since it is
likely that both properties and property differences are correlated across individual measurements, and such correlations would reduce the error, averaging was invoked. Zonal averaging of these data indicates that there was a significant increase in TIC concentrations in the upper layers over the 6-year period. This increase is a maximum at the surface, ($14 \pm 7 \, \text{µmol kg}^{-1}$, where the
uncertainty is the square root of the variance in the surface layer), falls off to zero by 600 m, and the pattern (Fig. 2) indicates that the increase is strongest in western basin near the recirculation region (70°W). Removing some of the differences associated with evaporation and precipitation through a normalization to a standard salinity, \( S = 35 \) (NTIC = TIC*\( S/35 \)), the surface increase from 1992 to 1998 is estimated at 8 ± 6 \( \mu \)mol kg\(^{-1} \). Both TIC and especially NTIC are well correlated across the basin with the seasonal decrease in temperature. Although the apparent increase in carbon concentrations in the LSW and AABW near the western boundary (70–80°W) should be noted, they do not represent enough of a pattern to be considered significant compared to the noise level.

2.1.2. Anthropogenic carbon

Anthropogenic carbon (C\( \star \)ANTH) has been calculated in a similar fashion for both the 1998 and 1992 sections (Fig. 3, upper panels). As expected, higher values of C\( \star \)ANTH are associated with more recently ventilated waters. There is a maximum from the surface to about 200 m depending upon longitude, and a decrease in C\( \star \)ANTH concentrations with depth. The highest concentrations occur at the surface in the eastern portion of the basin. Between 20°W and 70°W the average mixed-layer concentration of C\( \star \)ANTH decreases by about 0.5 \( \mu \)mol kg\(^{-1} \) for every degree of longitude. At 70°W the average mixed-layer concentration is 45 ± 5 and 51 ± 3 \( \mu \)mol kg\(^{-1} \) for the 1992 and 1998 transects, respectively. The mean mixed layer concentrations in the Florida Straits are higher: 52 ± 5 \( \mu \)mol kg\(^{-1} \) in 1992 and 70 ± 2 \( \mu \)mol kg\(^{-1} \) in 1998. In the western basin there is a slight maximum in C\( \star \)ANTH associated with the chlorofluorocarbon (CFC) maximum of LSW (Fig. 4, ~1500 db). Zonal averaging (Fig. 5) of the data suggests a change in the vertical gradient of C\( \star \)ANTH associated with a similar CFC feature at the depth of UCDW/AAIW interface at ~800 db.

We look to chlorofluorocarbons whose spatial and temporal variations have been previously scrutinized (e.g. Beining and Roether, 1996; Warner et al., 1996; Doney et al., 1998; Mecking, 2001) to understand better some of the features in the C\( \star \)ANTH fields. The 24.5°N sections of CFC11 and CFC12 are similar. Here, we concentrate on the CFC12 fields as the atmospheric partial pressure of this tracer has continued to rise over recent years, whereas CFC11 partial pressures have begun to decline, therefore CFC12 provides better diagnostics of recent water mass ventilation. CFC12 concentrations are relatively high near the surface and there are maxima at about 150–200 m and at 1500 m (Fig. 4, left panel). The former appears to emanate from the surface layers near the eastern boundary (which are also rich in C\( \star \)ANTH), but is seen across the entire basin. The latter is mainly in the western boundary (LSW) (Molinari et al., 1998).

Using equilibrium CFC partial pressures (pCFC = measured CFC concentration * CFC solubility function (Warner et al., 1996)) to remove most of the differences due to temperature variations (Fig. 4, right panel), it is found that due to greater solubility at lower temperatures, the 200-m CFC maximum does not appear in the pCFC field so the largest signal in pCFC12 is at the surface. Its disappearance suggests that this upper layer signal is seasonal in origin. This hypothesis is consistent with the findings of Doney and Bullister (1992) that a subtropical North Atlantic maximum in CFCs at depths of 100 to 200 m along a meridional line at about 20°W is caused by the sensitivity of CFC solubility to temperature, and heating and degassing of the seasonal thermocline.

The C\( \star \)ANTH maximum at 200 m is either an artifact of the calculations due to remineralization ratios in the top of the thermocline that do not follow Redfield stoichiometry or it is caused by the capping of the winter mixed layer by the seasonal mixed layer containing lower concentrations of C\( \star \)ANTH. Apparent oxygen utilization in the top 100 m of the 1998 data set is ~4 ± 6 \( \mu \)mol kg\(^{-1} \), therefore a small overestimate on C\( \star \)ANTH of order 3 \( \mu \)mol kg\(^{-1} \) is possible due to oxygen oversaturation. In either case, these anomalies appear to cause relatively little bias in the section-wide integral of C\( \star \)ANTH transport outside the 200 m depth range (Fig. 5).
The estimate of $C_{\text{ANTH}}$ assumes stationarity and that increases in dissolved inorganic carbon due to organic carbon remineralization is in fixed stoichiometry with increases in nitrate (see the appendix). Although these assumptions may not hold strictly, particularly on longer timescales, they are reasonable assumptions for the present comparison. Recent basin-wide $C_{\text{ANTH}}$ calculations (Sabine et al., 1999, 2002) suggest estimation uncertainties of order 5 \( \mu \text{mol kg}^{-1} \) above 1500 m.
and 3.5 μmol kg⁻¹ below this level. Therefore, it is reasonable to use $C_{\text{ANTH}}$, which is after all the perturbation signal in which we were interested, to examine changes in carbon concentrations over time. As with TIC, although most of individual mapped features are not significant, there is a pattern to the differences between the 1998 and 1992 $C_{\text{ANTH}}$ fields (Fig. 3, lower right panel). The magnitude of the changes are of the order of 10 μmol kg⁻¹.

Assuming that the surface water increase keeps pace with the atmospheric CO₂ rise, estimates of pCO₂ along the 24°N line (calculated according to Lewis and Wallace (1998) from measured alkalinity, TIC and temperature along with the atmospheric CO₂ increase from 1992 to 1998) suggest an increase in surface water TIC of about 7 μmol kg⁻¹ between 1992 and 1998. This value compares well to other recent, but less local estimates, 6 μmol kg⁻¹ (Lee et al., 2000a) and 10 μmol kg⁻¹ (Bates et al., 1996). Consistent with these estimates and our own estimate of surface changes in TIC and NTIC, horizontal averaging across the basin reveals a surface increase in $C_{\text{ANTH}}$ from 1992 to 1998 of $8 \pm 3$ μmol kg⁻¹. The maximum increase occurs from about 150–200 m. Increases of the order of 10 μmol kg⁻¹ are apparent to about 1000 m and horizontally averaging deeper layers we find increases greater than 5 μmol kg⁻¹ down to 2000 m. These increases are concentrated near the western boundary in the DWBC and suggest an important role for this current in moving ocean $C_{\text{ANTH}}$ away from the ventilation regions to the north. Below 3000 m in the eastern basin and 4000 m in the western basin, the suggested decrease in $C_{\text{ANTH}}$ is of the same order or smaller than the uncertainty in the estimation, and is therefore not significant.

Turning again to the equilibrium CFC partial pressures, we find some similarities in the types of changes which have occurred. Between 1992 and 1998 there has been an overall increase of pCFC12 with time. In the zonal average (Fig. 5) there is a large maximum (70 ppt) at 500 m and another, lesser maximum at 1500–1800 m. The shallower maximum is greatest in the western boundary in the 200 m to 800 m range, comprised of the relatively well ventilated water masses of AAIW and UCDW. It is also evident across the entire extent of the basin. The deeper maximum is confined to the western side of the western basin in the region of LSW.
To summarize, differences in both TIC and C\%ANTH over the 6-year period between 1992 and 1998 suggest an increase in carbon concentrations in the upper layers with a near-surface maximum. The increase of both properties in the surface layers is consistent with the changes occurring in the atmosphere. The enhancement in concentrations appears to go deeper than just the surface layers with the deep changes concentrated in the far western basin. The increase in C\%ANTH that we take to be significant to about 2000 db is consistent with the increase in pCFC12 (Fig. 5). However, the spatial pattern of the increases are somewhat different. The increase in pCFC12 displays a sharp reduction between 500 and 1000 m, whereas the increase in C\%ANTH concentrations displays a much more gradual gradient with depth. Gruber (1998) averaged concentrations of anthropogenic carbon for the latitude bands 20–30°N and 30–40°N using data taken from the Transient Tracers in the Ocean North Atlantic Study program 1981. Comparing these profiles to those at 24.5°N (Fig. 5) suggests that the profile of C\%ANTH at 24.5°N, even at depth, is moving towards the higher values previously associated with waters lying further to the north (also see Gruber, 1998, Figs. 8c and d).

It should be noted that the C\%ANTH difference fields may still contain biases or systematic errors in spite of the care taken on our part to calculate this derived variable for the 1998 data set in a fashion similar to that used for the 1992 data set. One such source of error may arise from the fact that the 1998 TIC values were measured whereas the 1992 values were calculated from measured pH and total alkalinity, and although the TIC values for these two data sets have been closely compared to remove biases as part of the Atlantic synthesis effort (Wanninkhof et al., 2003), there is still the possibility of errors which change with depth.

3. Carbon transport

3.1. Method

To obtain an estimate of the absolute meridional velocity field for each of the two 24.5°N transects a simple inverse box model has been used. It is based upon the same box inverse model method introduced to the physical oceanographic community by Wunsch (1978) and later described in much greater detail in Wunsch (1996). The inverse method as used here is simply a technique for estimating ocean circulation based on hydrographic observations and a set of simple physical constraints. The observations (temperature and salinity) allow for the calculation of geostrophic velocities relative to a reference surface. The model physics (in this case a set of conservation equations) is used to constrain the possible solutions for the unknown velocities at the reference surface in a least-squares sense. The model software originates from the system created...
The models are defined by $21$ neutral density interfaces in the vertical (Fig. 6) and the observed station spacing in the horizontal. The flow is assumed to be hydrostatic and geostrophic so that the thermal wind equations apply. Relative geostrophic velocities are computed between stations pair on the CTD 2-dbar pressure intervals and are integrated vertically to produce layer transport equations. The models are made up a set of transport constraint equations of the following form.

\[
X_i X_j q_{ij} a_{ij} v_{iU} C_{ij} \left( \frac{E_{kc}}{C_0} \right) + \sum_k q_{ik} a_{ik} C_{ik} v_{ik} E_{kc} + \text{known flux into or out of a box} \ldots ,
\]

where $i,j,k$ are the station pair, layer and Ekman layer indices; $\rho$ the density; $v_R$ the relative velocity; $v_U$ the unknown reference level velocity; $v_{EK}$ the initial Ekman velocity estimate; $C$ the property concentration; $a$ the vertical layer area; $E_{kc}$ the multiplicative Ekman component correction factor.

These equations are combined to form the familiar constraint set $A b + n = y$, where, the elements of the $A$ matrix are $A_{ij} = \int_B \rho_j a_j C_{ij} dp$ and are constructed from the integrated CTD pressure values. $T$ and $B$ are the top and bottom pressures, respectively, of the layer or sets of layers used in an individual constraint. The vector $b$ represents the unknown reference level velocities ($v_{U}$) resulting from the dynamic method and the correction terms to the initial Ekman estimates. The right-hand side vector, $y$ contains the known relative velocity transports and can include Ekman transport, and freshwater inflow/outflow and other net transport estimates.

The constraints are based upon the baroclinic flow field described by the hydrographic transects,
initial order of magnitude estimates of velocity at the reference surface, and estimates of the solution and data covariances. Since these constraints (Eq. (3)) are based on the flux across the sections only, the equations for our models are particularly simple and do not include dianeutral transfer or diffusion terms. The models presented here begin with initial estimates of net mass transport (entering from the north through the Bering Straits), net surface freshwater input from evaporation minus precipitation and rivers, and Ekman transport (Table 2).

The initial uncertainty estimate of $2 \times 10^9$ kg s$^{-1}$ in the top to bottom mass transport constraint has been computed in the following manner: (1) The uncertainty on the flow through the Bering Strait ($0.3–0.6 \times 10^9$ kg s$^{-1}$) (Coachman and Aagaard, 1988; Roach et al., 1995). (2) The uncertainty in the wind field (20% or $1.1 \times 10^9$ kg s$^{-1}$) comes from the range suggested by various wind products (Hellerman and Rosenstein, 1983), the Southampton Oceanography Centre global wind stress climatology, and the NCEP-NCAR and ECMWF atmospheric reanalyses). The Hellerman and Rosenstein (1983) field was used in the model because its net Ekman transport was closest to the mean of these four products. The 20% uncertainty is consistent with the low end of the range suggested by Ganachaud and Wunsch (2003). (3) The uncertainty in the baroclinic variability taken to be order $(1 \times 10^9$ kg s$^{-1}$), which is low compared to the 5 Sv suggested by Ganachaud and Wunsch (2003). (4) The uncertainty due to measurement noise is taken as $1 \times 10^9$ kg s$^{-1}$, again low compared to the 3 Sv suggested by Ganachaud and Wunsch (2003) for mid-latitudes where it is dominated by vertical heave of density surfaces by internal waves. (5) The uncertainty due to unobserved bottom triangle transport is taken to be $O(1 \times 10^9$ kg s$^{-1}$) the same as that suggested by Ganachaud and Wunsch (2003). (6) The uncertainty in the evaporation minus precipitation plus river runoff to the north of the transects is taken conservatively as 30% or 0.2 $\times 10^9$ kg s$^{-1}$. (7) We assume that the error due to unobserved ageostrophic motions (e.g., inertial oscillations) is negligible. The net uncertainty arising from (1)–(7), assuming that they are independent of one another, is $2.08 \times 10^9$ kg s$^{-1}$, which has been rounded off to $2 \times 10^9$ kg s$^{-1}$. It should be considered an optimistic estimate of the net uncertainty in the top to bottom mass flux.

Although other constraints such as silicacean conservation have been tested, the models used here are based solely on top-to-bottom mass and salt conservation. A constraint on the expected transport in the Florida Strait is also included (Table 2). The layer interfaces for the model were based upon

Table 2
Inverse model constraints, a priori estimates of uncertainty, data sources, and a priori estimates of solution variance

<table>
<thead>
<tr>
<th>Basic model constraints</th>
<th>Geostrophic + Ekman flux$^a$ = freshwater flux$^b$ with an integration reference point at Bering Strait$^c$ $-0.78 \pm 2 \times 10^9$ kg s$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass flux</td>
<td></td>
</tr>
<tr>
<td>Salt flux</td>
<td></td>
</tr>
<tr>
<td>1992</td>
<td>Geostrophic + Ekman flux$^a$ = Bering Strait transport$^c$ $-27.7 \pm 20 \times 10^6$ kg s$^{-1}$</td>
</tr>
<tr>
<td>1998</td>
<td>Total transport in the Florida Straits at 26°N: $30 \pm 1.0 \times 10^9$ kg s$^{-1}$</td>
</tr>
<tr>
<td></td>
<td>Total transport in the Florida Straits at 27°N: $32 \pm 1.0 \times 10^9$ kg s$^{-1}$</td>
</tr>
</tbody>
</table>

Initial magnitude of unknowns

| 20–30 cm s$^{-1}$      | Reference level velocities in Florida Straits |
| 2 cm s$^{-1}$          | Reference level velocities in the interior |
| 6 cm s$^{-1}$          | Reference level velocities where reference level is the bottom |
| 20%                   | Correction factor to input Ekman flux |

Ekman fluxes are computed from annual mean winds.

$^a$ 1992 Ekman flux: Hellerman and Rosenstein (1983), $5.75 \times 10^9$ kg s$^{-1}$ $\pm$ 20% and 1998 Ekman flux: Hellerman and Rosenstein (1983), $5.60 \times 10^9$ kg s$^{-1}$ $\pm$ 20%.

$^b$ Freshwater flux: Schmitt et al. (1989), $0.07 \times 10^9$ kg s$^{-1}$ $\pm$ 30%.

$^c$ Bering Strait: Roach et al. (1995) $0.83 \pm 0.66 \times 10^6$ m$^3$ s$^{-1}$ at $-1^\circ$C and $S = 32.5$. 
the work of Ganachaud (1999), which in turn was based on Macdonald (1998) and Rintoul and Wunsch (1991). The reference levels are based on Rintoul and Wunsch (1991) and Lavkin (1999), which showed that the use of a reference surface across the interior basin of about 3000 db leads to more realistic deep property balances. We have chosen to raise the reference surface out of the DWBC to approximately 800 db (where the Abaco moorings show an average 0 velocity) and in regions where it runs into topography near the coasts. Tests have been performed on a variety of reference surface choices besides those indicated in Fig. 6. In particular, a shallow set on \( \Gamma = 27.38 \) (~800 db) in the western basin and \( \Gamma = 27.62 \) (~1000 db) in the eastern basin and a deep set on \( \Gamma = 28.072 \) (~3000 db) were tested. The results of these runs will be mentioned where relevant.

Solutions (i.e. estimates of velocities at the reference surface and corrections to the initial Ekman component estimates) are found using a tapered-weighted least-squares (Gauss–Markov type) technique (Wunsch, 1996; Macdonald, 1998). This technique weights the system only by a priori estimates of the data and solution covariance.

3.2. Meridional mass transport across 24.5°N

The models have little trouble in meeting the constraints as long as large enough reference velocities are allowed for in the Florida Straits (see Table 2). This result is to be expected as the system is extremely under-determined. Larger reference velocity adjustments were required everywhere for the 1992 section, for, as will be shown shortly, the initial imbalance was larger. The shapes of the reference level velocity curves (Fig. 7) follow the changes in the initial estimate of the magnitude of the solutions. The solution size also follows the shape of the bathymetry. Because this system has been solved using a Gauss–Markov-type formulation, there has been no attempt to weight or scale the constraints or unknowns by anything other than the initial estimates of data and solution covariance (Wunsch, 1996). That is, there has been no attempt to remove the effects of differences in bottom depth, station spacing, etc. The corrections to the initial estimates of Ekman transport were minute as would be expected given the simplicity of the model.

The model produces circulation patterns (Figs. 8 and 9) based on the initial constraints and assumptions. The initial imbalances are large because Florida Current transport is under-estimated by the initial guess. To meet the constraints, the mass transport through the Florida Straits is increased by \( 6.5 \times 10^9 \) and \( 9.4 \times 10^9 \) kg s\(^{-1}\) in the 1998 and 1992 transects, respectively. Over the interior a fairly uniform decrease in the southward transport (Fig. 8, upper panels) has occurred throughout most of the water column. An increase in northward transport of bottom water is also apparent. (Note the definition of which layers we include in our use of the terms surface, intermediate, deep and bottom waters are given in Table 3).

Zonally integrated solutions for the models using the deep and shallow reference surfaces mentioned earlier are not significantly different from those shown here. However, locally specific differences in the resulting circulation patterns for these test models led us to the reference surfaces chosen for the final models. For instance, the model with the deep 3000 m reference level, which began with an initial imbalance of less than
6 × 10^9 kg s^{-1} for both sections, could not produce a significant deep western boundary current (DWBC), and in fact, produced northward transport of these waters in the 1992 model. It also produced a strong deep southward transport (~ 16 × 10^9 kg s^{-1}) in the far eastern basin, which we had no reason to expect. The shallow model, which had no trouble producing recognizable DWBCs, produced no net transport of bottom water in the 1992 section and large variations in deep water transport across the basin in both sections.

Integrating from the coast east to 70°W to include local recirculation, the present model estimates the DWBC transport to be −43 ± 21 × 10^9 and −43 ± 25 × 10^9 kg s^{-1} for the 1998 and 1992 sections, respectively.
Fig. 9. A comparison of the integrated absolute mass transport (including Ekman flow) for the 1998 (dark/blue) and 1992 (light/red) transects. The boxes to the right indicate the neutral density layers included in the integration (see Fig. 6), the difference in net transport between the two transects and the direction of the subtraction. The integration is performed from west to east. The lines at the top indicate the availability of data within the layer set for each station pair. Positive values indicate northward flow.

1992 transects, respectively. These estimates are well within the mean and standard deviation (−40 ± 13 × 10⁶ m³ s⁻¹) suggested by Lee et al. (1996) from their 4.7-year time series. The uncertainties encompass the range of transport estimates obtainable using different limits of integration. The DWBC position and magnitude is extremely variable (Lee et al., 1996; Fillenbaum et al., 1997), and it should be noted that the 1998 measurements were taken 2° farther north than the 1992 measurements and had better resolution, i.e. shorter distance between stations (Fig. 1). Nevertheless, both transects were able to capture a representative portion of the DWBC transport. Integrating the 1992 data from the coast to the point of maximum transport produces an estimate of −82 ± 22 × 10⁶ kg s⁻¹ (Fig. 9, panels 3 and 4). This number includes, however, recirculating transport associated with the offshore eddy, which appears to be much stronger and deeper in the water column in 1992 than in 1998.

In general, the transport in the two sections is similar both in the vertical and in the horizontal. Some of the differences include: a southward transport in the intermediate waters (Fig. 9, panel 3) in 1998, where northward transport occurs in 1992; a compensating, strong, intermediate, southward transport in the far, eastern 1992 basin; a stronger, surface intensified offshore eddy in 1998.

<table>
<thead>
<tr>
<th>Layer type</th>
<th>Model</th>
<th>Fl. St.</th>
<th>Interior</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>1992</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Top to bottom</td>
<td>1–20</td>
<td>29.5 ± 1.0</td>
<td>−30.6 ± 1.4</td>
<td>−1.1 ± 1.0</td>
</tr>
<tr>
<td>Surface</td>
<td>1–4</td>
<td>22.9 ± 0.8</td>
<td>−11.4 ± 1.7</td>
<td>11.5 ± 1.8</td>
</tr>
<tr>
<td>Intermediate</td>
<td>5–7</td>
<td>6.5 ± 0.7</td>
<td>−3.2 ± 2.3</td>
<td>3.3 ± 2.4</td>
</tr>
<tr>
<td>Deep</td>
<td>8–17</td>
<td>0.8 ± 0.1</td>
<td>−18.8 ± 4.0</td>
<td>−18.8 ± 4.0</td>
</tr>
<tr>
<td>Bottom</td>
<td>18–20</td>
<td>2.9 ± 3.5</td>
<td>2.9 ± 3.5</td>
<td></td>
</tr>
<tr>
<td>1998</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Top to bottom</td>
<td>1–20</td>
<td>31.4 ± 1.0</td>
<td>−32.3 ± 1.3</td>
<td>−1.0 ± 0.9</td>
</tr>
<tr>
<td>Surface</td>
<td>1–4</td>
<td>25.2 ± 0.7</td>
<td>−16.8 ± 1.6</td>
<td>8.4 ± 1.7</td>
</tr>
<tr>
<td>Intermediate</td>
<td>5–7</td>
<td>6.1 ± 0.5</td>
<td>−1.9 ± 2.7</td>
<td>4.2 ± 2.7</td>
</tr>
<tr>
<td>Deep</td>
<td>8–17</td>
<td>0.1 ± 0.1</td>
<td>−18.1 ± 4.5</td>
<td>−18.1 ± 4.5</td>
</tr>
<tr>
<td>Bottom</td>
<td>18–20</td>
<td>4.4 ± 3.6</td>
<td>4.4 ± 3.6</td>
<td></td>
</tr>
</tbody>
</table>

Note the 1992 model has an initial estimate of Florida Strait mass transport which was 2 × 10⁶ kg s⁻¹ less than what was used in the 1998 model to account for the flow through the Providence Channel missed by the 26°N Florida Strait measurements. Positive values indicate northward flow.
(there appears to be no net mass transport associated with this feature in either section); and a stronger and deeper 1992 bottom transport (Fig. 8). Both transects have a small southward transport in the near surface layers, and slight northward transport between about 3000 and 3500 db (1992) and 3800–4200 db (1998).

Neither of these last two features are seen in the calculation of Lavin et al. (2003), which used the 1992 data set, similar, but not identical reference surfaces and a different technique for computing the absolute circulation field. They were not seen in the Roemmich and Wunsch (1985) analysis of the 1981 transect using an initial reference surface of $\sigma_2 = 36.8$ (~1300 m), but they were seen in their analysis of the 1957 IGY transect. They are also seen in the Rintoul and Wunsch (1991) reanalysis of the 1981 data set, which used a 3000 db initial reference level and no smoothing of the data. In the present analysis, the shallower feature is robust, appearing in all our test runs. The deeper feature is not, as its existence depends upon the chosen reference level and constraint set.

It also should be noted that the Lavin et al. (2003) DWBC estimate at $22 \times 10^6$ m$^3$ s$^{-1}$ was only half of ours. Rosón et al. (2002) in their calculation of meridional carbon transport employed the Lavin et al. circulation. Although the slightly different reference surfaces used in the two studies accounts for some of the differences in transport, the primary difference between the calculations is the velocity applied to the reference surface to obtain mass balance. Whereas the present study applies set of velocities (Fig. 7), Lavin et al. (2003) employed a single velocity within the Florida Straits and one other that was applied across the entire interior field.

3.3. Meridional TIC transport

The model also produces patterns of total inorganic carbon transport for the two cruises (Fig. 10, upper panels, and Fig. 11). The TIC transport profiles are similar in character to the complimentary mass transport figures. In the surface layers the transport through the Florida Straits is clear as is the large eddy feature near 70°W. In these upper layers, the net TIC transport across the section is northward, but as in the case of mass, there is a net interior southward return flow. In the intermediate (AAIW and UCDW) layers the net transport is again northward and much, though certainly not all, of this poleward flow occurs in the Florida Straits. The net transport of deep TIC is southward, much of it carried by the DWBC. The transport of TIC closely follows that of mass because like density, the concentration of TIC changes little over the depth of the water column or basin extent (1998, mean TIC = $2173 \pm 29$ µmol kg$^{-1}$). This correspondence between the two properties affords us an opportunity to better understand our estimates of net TIC transport in light previous estimates (Table 4).

Given the large uncertainties, the new estimates are not significantly different from previous ones. The 1998 value of $-0.74 \pm 0.91$ PgC yr$^{-1}$, does, however, suggest a smaller regional divergence in the meridional carbon transport than is suggested by either of the 1992 studies. Although the values are not significantly different, it is useful to understand why they are not identical. First, why does our 1992 estimate appear to be lower than the Rosón et al. (2002) value, second why is the 1998 estimate lower than either of the other 1992 values, and third why are the estimates of uncertainty so different?

### 3.3.1. The 1992 TIC transport estimates

Since the same data set was used to compute the 1992 estimates, any difference must be due to differing estimates of the velocity field. Approximately one-half of the difference can be attributed to the difference in net mass transport.

Three methods were found for producing TIC transports from our model similar to those of Rosón et al. (2002). The first was simply to constrain the model to do so. It is given that model should be able to meet this constraint as the range provided by the uncertainty on our estimate encompasses the Rosón et al. (2002) value. The model produces this stronger southward TIC transport by decreasing (from 4.6 to $2.4 \times 10^9$ kg s$^{-1}$) the northward flux of water below 1000 db in the eastern basin which has a relatively high concentration of TIC, and by
decreasing the southward flow of deep water in the western basin which has lower TIC concentrations. Similar changes were found when a net zero or southward transport of silica was required across the section (the present model silica flux is $146 \pm 342 \text{ kmol s}^{-1}$). This balance between southward flow in the western basin, much of it carried in the DWBC, and northward flow of deep water within the eastern basin is the same as that which controls the silica transport across this transect (Lavin et al., 2003).

The third manner in which a larger net southward TIC transport can be produced is to use the deep reference model that does not capture the deep western boundary current at all. Our conclusion, therefore, is that the likely reason for the...
higher Rosón et al. (2002) TIC flux estimate is that their DBWC mass transport estimate is less than our own. The TIC flux estimates of both models could be improved through the integration of available time series data in the far western basin as it becomes apparent that, at this latitude, it is necessary to fully resolve the DWBC to get the transport of TIC correct.

3.3.2. The 1998 versus the 1992 TIC transport estimate

Our 1992 and 1998 TIC transport estimates are based on similar models. Still it appears that differences in the velocity field, here resulting from the differing data, are the main cause of the difference in TIC transport estimates. About 15% of TIC transport difference is due to the difference in net mass transport. Separating the field into overturning components equal to the layer mean TIC concentrations times the layer transports, and gyre components equal to the difference between the net layer transports and the associated overturning components, shows that two-thirds of the TIC transport difference \( (0.4 \text{ PgC yr}^{-1}) \) is due to differences in overturning. That is, although both transects carry a net overturn of \( 13–14 \times 10^9 \text{ kg s}^{-1} \), the 1992 transect carries more \( (\sim 21.6 \times 10^9 \text{ kg s}^{-1}) \) deep water rich in TIC southward than the 1998 transect \( (\sim 17.6 \times 10^9 \text{ kg s}^{-1}) \). The gyre components, which carry TIC northward, are responsible for the other third (0.2 PgC yr\(^{-1}\)).

These lower estimates of ocean TIC flux bring into question the concept that the tropical Atlantic is constantly outgassing carbon, while the subpolar Atlantic is absorbing it (Fig. 12). Seasonal variations may be important, as we have already mentioned the strong correlation between upper layer temperatures and TIC; but circulation differences likely dominate, as the TIC transports are strongly correlated with the patterns and magnitude of mass transport.

3.3.3. The TIC transport uncertainty estimates

The uncertainty in the carbon transports are an issue as they are directly related to the uncertainty in mass transport. The uncertainty estimate for the TIC transport computed here come directly from the inverse model formulation (Wunsch, 1996). The solution, \( \hat{b} \) to Eq. (3) is

\[
\hat{b} = Ky \quad \text{and} \quad P = R_{xy} - KAR_{xy},
\]  

Fig. 11. A comparison of the cumulative inorganic carbon transport (including Ekman flow) for the 1998 (dark/blue) and 1992 (light/red) transects. The boxes to the right indicate the neutral density layers included in the integration (see Fig. 6), the difference in net transport between the two transects and the direction of the subtraction. The integration is performed from west to east. The lines at the top indicate the availability of data in the layer set for each station pair. Positive values indicate northward flow.
where \( K = R_{xx} A^T (R_{nn} + A R_{xx} A^T)^{-1} \). \( R_{nn} \) is the a priori data covariance estimate, \( R_{xx} \) is the a priori solution covariance estimate, and \( P \) is the a posteriori solution covariance estimate about the unknown true values of the solution. \( P \) is used to compute the uncertainty in the property transports (MacDonald, 1995) and so includes the various components of uncertainty described in Section 3.1.

The Rosón et al. (2002) TIC transport uncertainty estimate is computed by applying a perturbation of \( \pm 2 \text{ Sv} \) to the interior transport field every 20 m and the opposite to the Florida Current to maintain the mass balance. This “transport error” is combined with a perturbation of \( \pm 4 \mu \text{mol kg}^{-1} \) in TIC to produce the \( \pm 200 \text{ kmol s}^{-1} \ (0.08 \text{ PgC yr}^{-1}) \) cited. This uncertainty is small compared to ours. The difference may arise from the Rosón et al. (2002) assumption of a perfect mass balance. Holfort et al. (1998) uses a completely different method. Taking a number of different inverse models that are solved using the SVD technique, they decide upon a range of ranks for which the carbon transport versus rank is fairly stable. The uncertainty is then taken to be the standard deviation across this range of ranks and models. Since this method is based upon the a priori choice of TIC transport range, it would appear to be self-limiting in terms of the magnitude of the a posteriori uncertainty estimate.

When our model was forced to meet the Rosón et al. (2002) estimate of TIC transport, their

### Table 4
Net transports of mass and other properties across the 24.5°N derived from the inverse model and compared to previous estimates

<table>
<thead>
<tr>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>MASS (10⁹ kg s⁻¹)</td>
<td>−1.0 ± 0.9</td>
<td>−1.1 ± 1.0</td>
<td>−1.3</td>
<td>−0.8</td>
</tr>
<tr>
<td>SALT (10⁶ kg s⁻¹)</td>
<td>−27.1 ± 30.5</td>
<td>−26.8 ± 34.0</td>
<td>−28.0 ± 1.0</td>
<td>— —</td>
</tr>
<tr>
<td>TIC (PgC yr⁻¹)</td>
<td>−0.74 ± 0.91</td>
<td>−1.32 ± 0.99</td>
<td>−1.59 ± 0.08</td>
<td>−1.21</td>
</tr>
<tr>
<td>C*ANTH (PgC yr⁻¹)</td>
<td>0.20 ± 0.08</td>
<td>0.17 ± 0.06</td>
<td>0.23 ± 0.08</td>
<td>— —</td>
</tr>
</tbody>
</table>

The Brewer et al. (1989) results are corrected to include the effect of the net mass transport from Bering Strait through flow and freshwater. Positive values are northward (1 PgC yr⁻¹ = 2642 kmol Cs⁻¹).

![North Atlantic Flux of TCO₂](image-url)

Fig. 12. Carbon transport as a function of latitude in the Atlantic (adapted from Wallace (2001) and updated with the results from this analysis).
estimate of uncertainty was also applied. This model was unable to produce the small uncertainty (0.08 PgC yr\(^{-1}\), equivalent to a mass transport uncertainty \(\sim 0.09 \times 10^9\) kg s\(^{-1}\)) suggested by these previous studies (Holfort et al., 1998; Rosón et al., 2002). That is, these very small uncertainties are inconsistent with the observations and model physics. We therefore believe that the uncertainty estimates of Holfort et al. (1998) and Rosón et al. (2002) are unrealistically small. In the final section, we discuss what improvements can be made to the model to reduce the size of the error bars.

3.4. Meridional \(C\star_{\text{ANTH}}\) transport

Our analysis suggests a northward meridional transport of \(C\star_{\text{ANTH}}\) across 24.5\(^\circ\)N of 0.20 ± 0.08 PgC yr\(^{-1}\) (1998) and 0.17 ± 0.06 PgC yr\(^{-1}\) (1992) (Table 4). These transport estimates and their uncertainties are consistent with both each other and with previous estimates, indicating that no observable temporal variation has taken place. This result would suggest that either no change has occurred or that the change over this 6-year period is too small to observe given the estimated uncertainties, and a longer period between transects may be necessary to observe \(C\star_{\text{ANTH}}\) transport variations. This issue is the same encountered with heat transport where changes in transport are too small to be observed in spite of significant warming from the surface to the deep Atlantic (Lavin, 1999).

Concentrations of \(C\star_{\text{ANTH}}\) are surface intensified, and although surface values are generally higher to the north of 24.5\(^\circ\)N than to the south (Gruber, 1998), the surface to depth difference dominates. The transport weighted mean concentrations of \(C\star_{\text{ANTH}}\) are 56.7 and 38.6 \(\mu\)mol kg\(^{-1}\) for the 1998 Florida Current and interior data, respectively. Therefore, the net transport of \(C\star_{\text{ANTH}}\) across 24.5\(^\circ\)N is strongly affected by the difference in concentrations between the northward flowing, relatively shallow Florida Current (carrying 0.7 PgC yr\(^{-1}\)) and the mass balancing interior return flow (carrying −0.4 PgC yr\(^{-1}\) in the western basin and −0.1 PgC yr\(^{-1}\) in the eastern basin). The high transport weighted mean concentration of \(C\star_{\text{ANTH}}\) in the interior (high relative to the area averaged mean) indicates that higher concentrations of \(C\star_{\text{ANTH}}\) tend to be associated with higher transports, as in the DWBC for example.

It is of course important to be aware when looking at net meridional transports across basinwide sections, such as this one in the subtropical North Atlantic, that small net values are often the result of subtracting one very large number from another very large number. This is particularly true of TIC in the subtropical Atlantic, but is less true of \(C\star_{\text{ANTH}}\). Here in the 1998 transect, for example, this subtraction can be thought of as occurring:

- vertically within the meridional overturning cell: 13 \(\times 10^9\) kg s\(^{-1}\) mass transport associated with surface and intermediate waters traveling northward minus 18 \(\times 10^9\) kg s\(^{-1}\) deep southward flow plus 4 \(\times 10^9\) kg s\(^{-1}\) of bottom water moving to the north, resulting in a small southward net mass transport of only −1 \(\times 10^9\) kg s\(^{-1}\). The TIC overturning cell is the sum of 10.1 PgC yr\(^{-1}\) northward flow of surface and intermediate water minus 14.5 PgC yr\(^{-1}\) deep flow plus 3.7 PgC yr\(^{-1}\) bottom water transport, resulting in a net transport of only −0.7 PgC yr\(^{-1}\). The net flux is only 5% of the overturning components. For \(C\star_{\text{ANTH}}\) the overturning is composed of 0.3 PgC yr\(^{-1}\) in the upper layers balanced by 0.1 PgC yr\(^{-1}\) in the deep layers (there is no significant bottom water transport) for a net transport of 0.2 PgC yr\(^{-1}\), a value that is of the same order of magnitude as the upper and lower limb components;
- or horizontally within the gyre: a strong surface intensified Florida Current carrying 31.4 \(\times 10^9\) kg s\(^{-1}\) northward, nearly balanced by an interior return flow of −32.3 \(\times 10^9\) kg s\(^{-1}\) dominated by the wind-driven upper layers (Fig. 9, layers 1–4) and the DWBC (Fig. 9, layers 8–17). The associated carbon system shows 25 PgC yr\(^{-1}\) moving northward in the Florida Current, nearly balancing a return flow of 25.7 PgC yr\(^{-1}\). The net flux value is only 3% of the component fluxes. Again for \(C\star_{\text{ANTH}}\) the story is different 0.7 PgC yr\(^{-1}\) flowing
northward in the boundary current and \(-0.5\ \text{PgC yr}^{-1}\) returning in the interior.

Therefore, even as we estimate mean meridional carbon transports based on the previous success of such methods in estimating heat fluxes, serious consideration must be given to possible temporal variability in the large flows that we integrate across. Such variability could adversely affect our ability to accurately predict mean present day values, as well as possible future values. The \(C_{\text{ANTH}}\) estimates should be less affected by this problem and, therefore, although \(C_{\text{ANTH}}\) can be calculated only indirectly, it may be more appropriate to use than the directly measured TIC when looking for temporal variations.

4. Discussion

The net northward transport of \(C_{\text{ANTH}}\) is opposite the net flow of total carbon and suggests, as has been found by others (Holfort et al., 1998; Rosón et al., 2002; Wallace, 2001), that in spite of today’s greater atmospheric carbon levels, the pre-industrial southward transport of carbon within the Atlantic was stronger than it is today.

Taking the average of the 1998 and 1992 estimates\(^{1}\) for the transport of TIC and \(C_{\text{ANTH}}\) at 24\(^{\circ}\)N, and values from the literature at 78\(^{\circ}\)N (Lundberg and Haugan, 1996) and in the South Atlantic (Holfort et al., 1998), and keeping in mind the magnitude of the uncertainties, we explore how these estimates balance with the uptake from the atmosphere (Takahashi et al., 2002; Wanninkhof et al., 2001) and rivers, and provide estimates of carbon storage based on this balance (Fig. 13). The Takahashi et al. (2002) pCO\(_2\) climatology used along with the NCEP 6-h winds as outlined in Wanninkhof et al. (2001) and rivers, and provide estimates of carbon storage based on this balance (Fig. 13). The Takahashi et al. (2002) pCO\(_2\) climatology used along with the NCEP 6-h winds as outlined in Wanninkhof et al. (2001) and rivers, and provide estimates of carbon storage based on this balance (Fig. 13).

\(^{1}\)Unless specifically indicated otherwise, 1992 refers to the present analysis of the 1992 transect.

Fig. 13. (a) Estimates of present day total inorganic carbon transport (solid arrows) based on the average of the 1998 and 1992 model results, air/sea exchange (dark/blue dashed arrows) from monthly pCO\(_2\) values (Takahashi et al., 2002) combined with gas exchange wind speed relationships using the monthly average and second moment to take the wind speed variability into account (Wanninkhof et al., 2001), river input (light/green dashed arrows) based on the river discharge estimates of Perry et al. (1996), taking the inorganic carbon concentration within the rivers to be order 1000 mmol kg\(^{-1}\) and assuming burial, respiration and outgassing of organic carbon occurs close to the ocean boundaries (Wallace, 2001; Aumont et al., 2001) and finally storage (in parentheses) which is computed as the residual of the other terms. The transport values across 10\(^{\circ}\)S and 30\(^{\circ}\)S are from Holfort et al. (1998). Those across 78\(^{\circ}\)N are from Lundberg and Haugan (1996). (b) The same for present day \(C_{\text{ANTH}}\). Each air/sea flux estimate is the residual of the other values in the particular box. (c) The same for the pre-industrial Atlantic Ocean. The light grey/red portions of the oceanic transport arrows indicate the estimated uncertainty. Units are \(\text{PgC yr}^{-1}\).
the north of 10°S in spite of the decrease in $C_{\text{ANTH}}$ uptake with latitude. Uptake of $C_{\text{ANTH}}$ per unit area between 78°N and 24°N is estimated at 1 gCyr$^{-1}$m$^{-2}$, between 24°N and 10°S at 6 gCyr$^{-1}$m$^{-2}$ and between 10°S and 20°S at 14 gCyr$^{-1}$m$^{-2}$.

The pre-industrial system is taken as the difference between the present day TIC and $C_{\text{ANTH}}$ patterns, assuming that the biological conditions, riverine input of carbon and circulation have not changed. Prior to the industrial revolution, uptake of TIC to the north of 24°N was similar to that occurring today; however, there appears to have been a marked decrease in outgassing between 24°N and 30°S brought on by the uptake of anthropogenic carbon (Fig. 13c). The results suggest that the difference between today’s carbon system and that which occurred prior to the burning of fossil fuels is brought about by today’s uptake of anthropogenic carbon, which is stronger to the south and which is transported northward to be stored within the northern basin.

The storage of anthropogenic carbon is defined as the change over time of the depth integrated concentration of $C_{\text{ANTH}}$.

\[
\text{Storage of } C_{\text{ANTH}} = \frac{d}{dt} \int C_{\text{ANTH}} \, dz.
\]

Using our $C_{\text{ANTH}}$ estimates from the 1998 and 1992 transects to perform the integration, we obtain average specific inventories of $45 \pm 11$ and $65 \pm 8$ molC m$^{-2}$ at 24.5°N for the 1992 and 1998 data sets, respectively. As would be expected, these values tend to be larger than the $34 \pm 7$ molC m$^{-2}$ estimated by Gruber (1998) based on a 1982 data set for the latitude band 18–27°N. In spite of the fairly large uncertainties, the three values taken together suggest a continued uptake and an increased storage of $C_{\text{ANTH}}$ within the basin. Differencing the 98/92 mean specific inventories suggests a change over the 6-year period in the $C_{\text{ANTH}}$ inventory at 24.5°N of $4.4 \pm 3.0 \times 10^{-14}$ PgC yr$^{-1}$ m$^{-2}$. It is known that the specific inventory within the Atlantic increases with latitude (Gruber, 1998). Therefore, to obtain an estimate of the storage within the basin based upon these changes would require similar calculations performed at a number of different locations. Such work is part of an ongoing analysis.

Using the 1998 and 1992 repeat transects at 24.5°N in the Atlantic, we have investigated the details of total inorganic carbon and anthropogenic carbon fields and transports, and have looked at how they have changed over the period of time between the observations. We have determined a rate of increase in the surface CO$_2$ concentration from both fields of $1.5 \pm 0.6$ mmol kg$^{-1}$ yr$^{-1}$, which is consistent with the estimate based on pCO$_2$ measurements. The transport of $C_{\text{ANTH}}$ is northward and is of similar magnitude in the two sections. It is dominated by the difference in concentrations between the northward flowing shallow Florida Current and the interior return flow. The transport of TIC is southward. The TIC transport estimates are sensitive to the estimated mass transport of the DWBC.

One issue with the present model, which uses only a single transect and therefore cannot take full advantage of divergence and anomaly constraints, is the large uncertainty in the TIC transports. The uncertainties computed here can be decreased in future models by including early and late 1990s data from 48°N and 57°N, which will allow the use of constraints on phosphate/oxygen and nitrate/oxygen combinations, as well as the use of anomaly constraints that together should provide a stronger constraint set (Ganachaud, 1999) for producing direct estimates of TIC and $C_{\text{ANTH}}$ transport and divergence. Although error bars on TIC transports as small as those suggested by previous authors appear unlikely, ongoing work suggests that a factor of two reduction in the magnitude of the uncertainties presented here can be expected. The new models will also allow direct estimations of storage and a residual calculation of air–sea CO$_2$ fluxes that can be compared to the estimates of Takahashi et al. (2002) used here.

The increased effort during the WOCE, OACES and JGOFS programs to directly measure carbon dioxide concentrations in seawater makes it possible to combine information from several basinwide transects in the North Atlantic to look at the large-scale transport and storage of CO$_2$. By
combining these modern, high resolution, hydrographic sections, we can gain insight into the global distribution of sources and sinks of CO$_2$, make an educated inference as to how these distributions separate into an historical background and an anthropogenic signal, and continue the compelling task of estimating how the buildup of fossil fuels in the atmosphere is reflected in the oceanic concentrations of CO$_2$.

Acknowledgements

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Appendix A. A method for calculation the anthropogenic CO$_2$

The TIC in the oceans is controlled by biological processes such as the production and remineralization of organic matter and calcium carbonate, and air–sea CO$_2$ exchange including anthropogenic CO$_2$. The technique to determine the anthropogenic CO$_2$ component is based on separating the influence of these processes on TIC concentrations. The method used here is a simplified version of the one originally developed by Gruber et al. (1996), and is based on the same general concepts as earlier methods. This method introduces a quasi-conservative tracer, $\Delta C^*$, defined as

$$\Delta C^* = \text{TIC}^\text{MEAS} + R_{\text{CO}}(O_2^\text{EQ} - O_2^\text{MEAS}) - 0.5[A_T^\text{MEAS} - A_T^0 - R_{\text{NO}}(O_2^\text{EQ} - O_2^\text{MEAS})] - C^\text{EQ}(f_{\text{CO2}} = 280 \, \mu\text{atm, } A_T^0, \Theta, S), \quad (A.1)$$

where superscript “MEAS” refers to measured concentrations in water samples, $O_2^\text{EQ}$ and $O_2^\text{MEAS}$ are the saturation and in situ concentrations of oxygen, respectively, $A_T^0$ is the preformed alkalinity, $A_T^0$ that a subsurface water parcel had when it was last at the surface ocean, $C^\text{EQ}$ is the TIC in equilibrium with the preindustrial atmosphere CO$_2$ fugacity of 280 µatm for the sample’s salinity ($S$), potential temperature ($\Theta$), and $A_T^0$; and $R_{\text{CO}}$ and $R_{\text{NO}}$ are the stoichiometric ratios relating inorganic carbon and nitrate changes to dissolved oxygen changes, respectively.

The $A_T^0$ of a subsurface water parcel is estimated from multilinear regression using the conservation tracers S and PO as independent variables (here PO = $O_2 - R_{O_3}P \times P$). The measured alkalinity data used by Gruber et al. (1996) gave the following relationship

$$A_T^0(\mu\text{mol kg}^{-1}) = 367.5 + 54.9S + 0.0074PO, \quad (A.2)$$

where $S$ is on a practical salinity scale, and PO is in µmol kg$^{-1}$. The standard error of the estimated $A_T^0$ is ±11 µmol kg$^{-1}$. The use of an additional variable such as $\Theta$ to fit surface alkalinity data does not reduce the uncertainty in Eq. (A.2).

The $C^\text{EQ}$ was calculated from the estimated $A_T^0$ and $f_{\text{CO2}} = 280 \, \mu\text{atm}$ using the dissociation constants for carbonic acid of Mehrbach et al. (1973) and other ancillary constants as recommended by Lee et al. (2000b) and linearized using a least-squares linear fit:

$$C^\text{EQ} = 2082.8 - 9.0(\Theta - 9) - 5.1(S - 35) + 0.78(A_T^0 - 2320). \quad (A.3)$$

The sum of the first three terms in Eq. (A.1) is the preformed TIC of a sample ($C^0$) when the
sample was at the surface. $\Delta C^*$ is the difference between $C^0$ and $C^{EQ}$. If the ocean takes up more anthropogenic $CO_2$, values of $\Delta C^*$ would increase. This $\Delta C^*$ reflects the anthropogenic $CO_2$ ($C^{ANTH}$) and air–sea disequilibrium ($\Delta C^{DISEQ}$) at the time when the sample lost contact with the atmosphere ($\Delta C^* = C^{ANTH} + \Delta C^{DISEQ}$).

To separate $C^{ANTH}$ from $\Delta C^*$, $\Delta C^{DISEQ}$ must be estimated. Determining the air–sea $CO_2$ disequilibrium has been problematic in estimating anthropogenic $CO_2$ in the ocean (Sabine et al., 2002). The method used here is to assume that for deep-ocean density surfaces where one can safely assume that there is no anthropogenic $CO_2$, the air–sea disequilibrium is equivalent to $\Delta C^*$. Therefore, in this study, the mean value of $\Delta C^*$ below 3500 db has been added to all samples to correct for air–sea disequilibrium.

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