Uptake and spreading of anthropogenic trace gases in an eddy-permitting model of the Atlantic Ocean

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1. Introduction

The ocean is taking up some part of the trace gases that have been released into the atmosphere by human activity, such as carbon dioxide (CO2) or chlorofluorocarbons (CFCs) [Sarmiento and Gruber, 2002]. The uptake of CO2, which is on the order of a third of the total anthropogenic emissions, helps to reduce the anthropogenic part of the greenhouse effect. The present uptake is largely dominated by physical mechanisms [e.g., Sarmiento et al., 1992], namely equilibration with the upper ocean and subsequent transport into the deeper ocean with the formation of deep and intermediate waters. In the future, however, feedbacks between changes in climate, biological productivity and the carbon cycle will play an increasing role [Friedlingstein et al., 2003, and references therein].

Because of the dominant role of the North Atlantic for the formation of deep water, it is also the location of the strongest penetration of the anthropogenic signal into the deep ocean interior [Gruber, 1998; Körtzinger et al., 1998; Keir et al., 1998]. However, the processes relevant for the uptake of anthropogenic carbon C\text{ant} are not fully understood. On the basis of partial pressure measurements of CO2 (pCO2) it has recently been conjectured that the air-sea pCO2 difference in the subpolar North Atlantic may have been diminishing over the last decades [Omar et al., 2003; Lefèvre et al., 2004; Omar and Olsen, 2006], indicating a decrease in the uptake of anthropogenic carbon. Global carbon models on the other hand suggest that the North Atlantic is a region of exceptionally strong air-sea flux of C\text{ant} [e.g., Orr et al., 2001], and that the flux is still continuing to increase. The assumption of an increasing air-sea pCO2 difference has also been used in the calculation of C\text{ant} flux based on the pCO2 climatology by Takahashi et al. [2002].

Both data- and model-based estimates of anthropogenic trace gas uptake and distribution have their specific limitations: in spite of the tremendous increase in the number and quality of carbon system observations during the WOCE era [Wallace, 2001] and from ongoing volunteer observing ship programs, direct calculations of CO2 flux are still hampered by limitations in temporal and spatial data coverage. Separation of the small C\text{ant} signal from the large natural background in the ocean interior involve a number of assumptions, such as purely isopycnal transport and fixed elemental stoichiometries [e.g., Gruber et al., 1996], that introduce substantial errors [Matsumoto and Gruber, 2005].

Models have other limitations: One of the more important ones is that, because of the required long integration times, global carbon cycle models still mostly lack an explicit representation of the ocean mesoscale dynamics.

with eddies and frontal structures. To describe the large-scale implications of these motions, subgrid-scale parameterization are used in the model equations. The Ocean Carbon Model Intercomparison Project OCMIP has demonstrated that there is still a considerable scatter in model-based estimates of anthropogenic trace gas uptake and spreading [Orr et al., 2001; Dutay et al., 2002; Matsumoto et al., 2004]. The scatter can be traced back to differences in the physical circulation field caused by differing model resolutions and choices of physical parameterizations [Doney et al., 2004]. Interestingly, a realistic representation of one anthropogenic tracer (e.g., bomb radiocarbon) does not necessarily imply a good representation of others (e.g., CFCs) [Matsumoto et al., 2004], so there is not one ‘best’ model, demonstrating that the implications of the subgrid-scale characterizations are not yet fully understood.

[8] Besides a critical assessment of model results against available observational data, there is therefore a need for a better understanding of the processes that drive anthropogenic trace gas uptake and spreading in models. The model investigation presented here contributes to that aim by examining anthropogenic tracer uptake and spreading in an eddy-permitting model. The model configuration used here has extensively been used for studying various aspects of the physical circulation and its interannual to decadal variability [Beismann et al., 2002; Getzlaff et al., 2005]; simulations of CFC uptake have been used to critically assess the deep water formation in the subpolar North Atlantic [Böning et al., 2003; Beismann and Redler, 2003]. The model domain is limited to the Atlantic basin only, and we use the perturbation approach by Siegenthaler and Joos [1992] for separating the increase of dissolved inorganic carbon from its preindustrial value. Both approaches introduce errors into the model solution, but the reduced computational cost allows for a better representation of the mesoscale in its influence on water-mass formation processes.

[7] The paper is organized as follows: After a description of the model (section 2) the distribution and inventories of \( C_{\text{atm}} \) are compared to other models and data-based estimates (section 3). In section 4 we make use of the improved representation of physical processes to discuss the relative importance and spatial distribution of processes that are responsible for anthropogenic carbon uptake in the model. In section 5 we discuss how the physical circulation determines the strength and the timescales of the meridional \( C_{\text{atm}} \) transport. In the discussion (section 6) we discuss the relevance of our findings to the open questions about anthropogenic carbon cycling in the Atlantic.

### Table 1. Summary of Experiments

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<tr>
<th>Resolution</th>
<th>Wind Forcing</th>
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<tr>
<td>REF</td>
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<td>HEAT</td>
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<td>COARSE</td>
<td>4/3°</td>
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[9] The model domain covers the Atlantic from 70°S to 70°N, with open boundaries across the Drake passage and along 30°E. The 1/3° configuration of the model, which is mainly used here, has an isotropic horizontal resolution of \( 1/3^\circ \times 1/3^\circ \cos \phi \) where \( \phi \) is the geographical latitude) that results in grid sizes of about 20 km in the subpolar North Atlantic. This resolution resolves the upper part of the mesoscale spectrum, and is therefore called “eddy permitting.” In the vertical, the water column is divided into 45 levels with depth ranges from 10 m at the surface to 250 m below 2250 m depth.

[10] Subgrid-scale processes are parameterized by a Gent and McWilliams [1990] approach, using a rotated diffusion operator [Griffies, 1998] to ensure that mixing of tracers occurs primarily along isopycnals. For a better representation of the Nordic Seas overflows, the bottom boundary layer parameterization of Beckmann and Dösscher [1997] is used. The wind influence on the mixed layer depth is simulated explicitly by a Kraus and Turner [1967] mixed layer model.

[11] The physical model was started from a mean climatological temperature and salinity field [Boyer and Levitus, 1997] and spun up for 25 years. After that a 100-year run (1900–2001) was performed. During the first 58 years the wind stress and heat-flux forcing during were taken from a ECMWF climatology [Barnier et al., 1995]. For the years 1958–2001 (serving as the reference experiment, REF), anomalies from the NCEP reanalysis data set [Kalnay et al., 1996] relative to the ECMWF climatology were used to vary the surface forcing interannually. The formulation of the heat fluxes follows that of Eden and Willebrand [2001] in their study of the thermohaline circulation variability, allowing for some feedback of the ocean on the net atmospheric fluxes [Han, 1984; Haney, 1971]. For salinity the surface values have been restored to a monthly Levitus climatology using a timescale of 30 days.

[12] In order to help to elucidate the effects of the atmospheric forcing variability a second experiment (HEAT) was performed, where only heat flux anomalies were used in the years 1958–2001, while the ECMWF climatology without interannual variation was retained for the wind stress data. In addition, the reference model is compared to an identical (except for some resolution-dependent parameters) configuration on a non-eddy-resolving \( 4/3^\circ \times 4/3^\circ \cos \phi \) grid (COARSE). All experiments are listed in Table 1.
approach allows the model integration period to be limited to the period of major anthropogenic influence. It has successfully been used in several global models [Sarmiento et al., 1992; Caldeira and Duffy, 2000; Orr et al., 2001].

[14] At the surface spatially uniform atmospheric CO$_2$ and CFC-11 partial pressures ($p$CO$_2$ and $p$CFC) (Figure 1) are prescribed. Atmospheric $p$CO$_2$ is taken from a combination of ice-core and atmospheric measurements [Enting et al., 1994] until 1990, after that the IPCC scenario S650 is used. The CO$_2$ gas exchange is parameterized with a quadratic wind-speed–dependent gas exchange velocity following Wanninkhof [1992]. CFC-11 was initialized (by zero) in year 1958 and integrated similar to C$_{\text{ant}}$ except for different solubility, atmospheric forcing function and gas exchange velocity.

[15] Conditions for C$_{\text{ant}}$ at the boundaries to adjacent ocean basins were estimated using CFC-11 data. In the Greenland sea, the estimated profile below 750 m depth by Anderson et al. [2000] was extrapolated assuming equilibrium with the atmosphere at the surface. In the Southern Ocean, we used the C$_{\text{ant}}$ estimate by Sabine et al. [1999] and the CFC data from Archambeau et al. [1998] to estimate a linear CFC-C$_{\text{ant}}$ regression. This linear regression was then used to estimate C$_{\text{ant}}$ in Drake Passage (using CFC data from Roether et al. [1993]) and in the gap between the Sabine et al. [1999] data and Antarctica at 30$^\circ$E (using CFC data from Archambeau et al. [1998]). The lateral boundary conditions were made time-dependent by scaling them with the atmospheric $p$CO$_2$ history.

[16] Figure 1 shows the atmospheric (prescribed) and oceanic (model-calculated) $p$CO$_2$ at the Bermuda Atlantic Time-series Station site in the subtropical Atlantic. The model $p$CO$_2$, that was initialized in 1900 with no anthropogenic carbon present in the ocean, increases rapidly over the first few years, to catch up with the atmosphere. After that it follows the atmosphere with a lag caused by the finite equilibration time of CO$_2$. The annual cycle in modeled

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**Figure 1.** Prescribed atmospheric partial pressure of CO$_2$ (in ppm, dashed line) and modeled surface values (solid line) at BATS station (32$^\circ$N, 64$^\circ$W).

**Figure 2.** Mean 1990 vertically integrated C$_{\text{ant}}$ inventory (in mol m$^{-2}$) in the (a) 4/3$^\circ$ (COARSE) and in the (b) 1/3$^\circ$ model (REF).
oceanic $pCO_2$ is an effect of the annual temperature cycle, not of biological activity, as we are assuming a steady state pre-industrial carbon cycle. For the bulk of analysis we therefore concentrate on the interannual variability. Only for a detailed comparison of differences in uptake between $C_{ant}$ and CFC in respect to wintertime convection we focus on the intraseasonal timescale.

3. Basin-Scale Budget of $C_{ant}$

[17] The spatial pattern of the vertically integrated $C_{ant}$ inventory in the 4/3° model (Figure 2a) agrees with results from other coarse-resolution models [Sarmiento et al., 1992; Caldeira and Duffy, 2000; Orr et al., 2001] and data-based estimates [Sabine et al., 2004]. It shows the major storage of $C_{ant}$ in the subpolar North Atlantic and a broad southward spreading west of the Mid-Atlantic Ridge. A somewhat weaker southward spreading occurs in the eastern basin.

[18] Qualitatively, the pattern in the eddy-permitting 1/3° model (Figure 2b) is very similar. However, owing to the better resolution of the western boundary structures and the resulting Deep Western Boundary Current (DWBC), a more confined spreading along the western margin transports $C_{ant}$ faster to the south and across the equator than in the coarse resolution model. The result of this spreading is a stronger connection of North and South Atlantic than in the 4/3° model. In the subpolar basin the west-east gradient of $C_{ant}$ is increased owing to a more realistic representation of bottom topography and the resulting narrower passages, such as the Gibbs Fracture Zone, leading to less transport of Labrador Sea Water (LSW) into the eastern basin. The strong meridional gradient agrees well with the strong east-west gradient in observational $C_{ant}$ estimates by Körtzinger et al. [1999]. A detailed comparison of CFC and $C_{ant}$ data with recent ship measurements in the midlatitude North Atlantic shows that the 1/3° model compares well in both structure and values [Tanhua et al., 2006]. The model showed a remarkable agreement with the increase in both CFC and total inorganic carbon over the last 20 years as inferred from a comparison of recent observations with TTO data.

[19] The latitudinal distribution of $C_{ant}$ is confronted with data-based estimates in Figure 3, averaged over 10° latitude bands and referenced to a common year. The comparison indicates qualitatively similar latitudinal dependence but shows that both model runs contain significantly less $C_{ant}$ than the estimate by Gruber [1998] (Figure 3a), except in the most southern latitude band. In the subtropical regions of both hemispheres, the inventory in the 1/3° model is higher, and thus closer to the estimate by Gruber [1998] than the 4/3° model; the converse holds in the subpolar and tropical regions. The total amount of $C_{ant}$ in the two model resolutions is almost equal.

[20] The discrepancy between the models and the more recent compilation by GLODAP [Sabine et al., 2004] is less drastic (Figure 3b). The total model inventory is only 55% of the estimate by Gruber [1998], but 80% of the more recent estimate by Sabine et al. [2004].

[21] What is the cause for the discrepancy between the model and data-based inventory estimates? One possible cause on the model side could be an underestimation of the air-sea flux of $C_{ant}$. A wide variety of conditions have been shown to influence air-sea gas exchange, among them wave breaking and bubbles, surface films, the thermal skin effect, etc. [e.g., Woolf, 1997; Zappa et al., 2001; Frew et al., 2004; McGillis and Wanninkhof, 2006]. No consensus has been reached on how best to parameterize these effects [Wanninkhof and McGillis, 1999; McGillis et al., 2000; Woolf, 2005]; when using measured distributions of $pCO_2$, global air-sea flux of CO$_2$ is quite sensitive to the choice of gas exchange parameterizations [Takahashi et al., 2002].

[22] To clarify this point, we performed three model experiments with the 4/3° model, all using the same wind speed distribution obtained from 6-hourly NCEP wind fields as in the reference run: Two experiments were conducted with doubled or halved gas exchange velocity, maintaining the quadratic wind-speed dependency. A third experiment used the cubic wind-speed dependency of Wanninkhof and McGillis [1999]. The area-integrated air-
sea-flux with halved gas exchange velocity is 18% lower compared to the reference run, 6% higher with the cubic wind speed dependency, and 13% higher for the doubled gas exchange velocity. This low sensitivity agrees with findings from earlier model studies [e.g., Sarmiento et al., 1992; England et al., 1994] and can be explained by the tendency of the air-sea disequilibrium in prognostic carbon models to become smaller for increased gas exchange velocity, partially offsetting the increased flux due to the gas exchange velocity. This cannot happen when using observed pCO2 [Takahashi et al., 2002], explaining the stronger sensitivity found in data-based flux calculations.

The average gas exchange velocity in the reference run is 18 cm hr\(^{-1}\), only slightly lower than the global average estimated by Broecker et al. [1986] of 22 cm hr\(^{-1}\). We therefore conclude that uncertainties in the gas-exchange have only a minor effect on our modeled C\(_{\text{ant}}\) inventory.

Concerning the role of STMW in the carbon uptake, it is instructive to compare the formation rates and C\(_{\text{ant}}\) inventories in the 1/3\(^{\circ}\) and 4/3\(^{\circ}\) models. The volume of STMW (density range \(\sigma = 26-26.7\)) is with \(3.6 \times 10^6 \text{ km}^3\) (total amount in the subtropical North Atlantic) correctly simulated in the 1/3\(^{\circ}\) model compared to Levitus observations (\(3.7 \times 10^6 \text{ km}^3\)), whereas the 4/3\(^{\circ}\) model under-estimates this volume by 25% (\(3.7 \times 10^6 \text{ km}^3\)). Although STMW only represents 5% of the total water mass it contains with 20% a significant amount of C\(_{\text{ant}}\). Therefore increasing STMW by only small amounts would result in an disproportionally large gain of C\(_{\text{ant}}\). Comparing the inventories between 20\(^{\circ}\)N and 40\(^{\circ}\)N in both models (Figure 3a) one notes an increased uptake the subtropics. However, comparing the density ranges of C\(_{\text{ant}}\) (Figure 4) reveals that this increase does only has little effect on the total uptake. At the same latitude there is also a partial decrease of the inventory in lighter water masses, so that the total inventory does not change significantly.

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The limited model domain could also influence inventories through the prescribed C\(_{\text{ant}}\) values at the lateral boundaries in the Southern Ocean. The values were obtained by using a relation between CFCs and C\(_{\text{ant}}\) estimated from present-day data. Because C\(_{\text{ant}}\) has a longer atmospheric history than CFCs it is possible that older water masses contain C\(_{\text{ant}}\) but no CFCs; however, our estimate at 30\(^{\circ}\)E agrees well with the more recent estimate of C\(_{\text{ant}}\) along that latitude by Lo Monaco et al. [2005b, 2005a]. If anything, we expect the boundary condition to lead to a slight overestimation of C\(_{\text{ant}}\) in earlier periods of the model integration, when our estimate overestimates the penetration depth of the signal. Moreover, most of the signal entering in Drake Passage leaves the model domain again over the boundary to the Indian Ocean.

The initialization of the model in 1900 with no C\(_{\text{ant}}\) present clearly is an underestimate; however, most of the initial deficit is compensated by a rapid initial equilibration in the first few years of the model run (Figure 1). For the 4/3\(^{\circ}\) model we estimate the remaining deficit in the year 1900 to be less than 1 Pg C or 4%, using a fit to the Green function
for the model inventory by two exponentials. The assumption of an unchanged strength of the biological pump might be wrong. However, the predicted feedbacks of climate change on the biological pump in the ocean at least for the present are much too weak to account for the inventory difference here, and also generally in the opposite direction [Friedlingstein et al., 2003].

[27] The assumptions of (1) a preindustrial equilibrium between ocean and atmosphere $pCO_2$ at (2) constant alkalinity in the perturbation approach are both wrong. However, the effects of these two errors on equilibrium $C_{ant}$ are small and have opposing signs for the North Atlantic.

[28] In summary, we cannot identify one dominating cause that can explain the lower $C_{ant}$ inventory in our model compared to the data-based inventories of Gruber [1998] and Sabine et al. [2004]. Instead there are a number of uncertainties that possibly each contribute an uncertainty of a few percent. We would argue therefore that our model estimate, while clearly low, is not completely unreasonable and probably has an uncertainty of about the same magnitude as the data-based inventories. It is within the quoted 20% error limit [Gruber et al., 1996] of the data-based inventory by Sabine et al. [2004], but clearly outside the error limits of the Gruber [1998] inventory. The accuracy of the data-based estimates is still a matter of much debate [Hall et al., 2004; Waugh et al., 2004; Matsumoto and Gruber, 2005; Lo Monaco et al., 2005b]. Quite recently there are other studies calculating the amount of $C_{ant}$ by

Figure 5. Average 1989 atmosphere-ocean flux of $C_{ant}$ (in mol m$^{-2}$ year$^{-1}$) in the (a) $1/3\degree$ and in the (b) $4/3\degree$ model.

Figure 6. Zonal section at 55°N of $C_{ant}$ in $\mu$mol l$^{-1}$ for different months during winter 1994/1995. The isolines show the density structure in sigma units, the atmosphere-ocean flux of $C_{ant}$ (in mol m$^{-2}$ yr$^{-1}$) is shown on top.
using transit time distributions constrained by other data (usually CFC). By allowing mixing, Hall et al. [2004] and Waugh et al. [2004] calculated 20–30% lower values compared to the Gruber method. An additional air-sea disequilibrium would account for another 6%.

Despite the as yet unexplained difference in total inventory we argue that the increased physical resolution of the model allows a better understanding of processes involved in the physical transport of C\textsubscript{ant}. The horizontal distribution of C\textsubscript{ant} (Figure 2) and of C\textsubscript{ant} air-sea flux (Figure 5) both show much more detailed structures under the effect of mesoscale eddies and frontal structures than in the coarse resolution simulation. The remainder of the paper concentrates on the physical effects that cause the horizontal structure and the temporal variability. The focus is on the subpolar North Atlantic because this is a key region in terms of deep water formation.

4. Spatial Structure of the North Atlantic Air-Sea Flux of C\textsubscript{ant}

4.1. Role of Wintertime on the Anthropogenic Gas Uptake

The air-sea flux of C\textsubscript{ant} in our model shows maximum rates over large parts of the subpolar gyre in the North Atlantic (Figure 5), consistent both in strength and pattern with the coarser-resolution global models of Orr et al. [2001]. While the 4/3° model has a broad, uniform uptake over large areas, the uptake flux in the 1/3° model shows a more complex pattern, suggestive of a relation to the circulation structure in the subpolar gyre, which is substantially refined in the higher-resolution model.

What are the main processes controlling this pattern? A first assumption would be that C\textsubscript{ant} fluxes should be affected the formation of deep water masses through wintertime convection. Cooling through strong winds at the end of winter leads to an unstable water column, causing denser surface water to mix down to a depth where it is neutrally stable with respect to the surrounding water masses. Figure 6 demonstrates how this process affects C\textsubscript{ant} uptake along a section at 55°N: In fall (Oct) the water column is stable with a shallow mixed layer. The surface waters are highly saturated with C\textsubscript{ant}, and the flux from the atmosphere into the ocean is low. Later in fall (Dec) the mixed layer deepens, C\textsubscript{ant} is mixed down, and the surface mixed layer is able to take up additional C\textsubscript{ant} which causes the atmosphere-ocean flux to increase more or less uniformly. In late winter (Feb) local deep-reaching convection occurs, and C\textsubscript{ant} enriched surface water mixes with deep water with a very low C\textsubscript{ant} content, causing high C\textsubscript{ant} fluxes over small scales. An imprint of small-scale wintertime process remains visible in the annual average (Figure 5b).

However, the process of deep water formation is restricted to the Labrador and Irminger Seas (as evidenced by the late winter mixed layer depth, Figure 7a), and some other process must be invoked to explain the extended flux pattern across the subpolar gyre: a conspicuous feature to be noted here is the apparent correspondence of the C\textsubscript{ant} flux with the upper ocean current system, particularly along the path of the North Atlantic Current and the recirculation within the Irminger Basin (Figure 5b).

Since CFC-11 has a much shorter equilibration time with the atmosphere compared to C\textsubscript{ant} we might expect a different role of the short-term, deep-winter convection processes on its uptake. The annual averaged flux pattern (Figure 5b) is indeed much more concentrated than the C\textsubscript{ant} flux. Comparing Figure 5b with the annual mean air-sea flux of CFC-11 from the model (Figure 7b), we clearly see a good match with the spots of wintertime deep convection (Figure 7a) in the Irminger and Labrador Seas in Figure 7a.

Although the atmospheric input for both tracers does not feature a seasonal cycle, Figure 6 already indicated that the oceanic response has a clear variation within one year. Figure 8 shows the variations of C\textsubscript{ant} and CFC-11 atmosphere-ocean fluxes in the subpolar North Atlantic where one clearly identifies an increased uptake in winter. In summer the uptake for both tracers shows a minimum; but while the CFC uptake decreases down to zero, C\textsubscript{ant} flux is still present throughout the whole year. A direct comparison of normalized fluxes (Figure 8, bottom) illustrates the difference between the equilibration timescales of C\textsubscript{ant} and CFC-11: For CFC-11, the uptake is mainly restricted to the winter season. In summer the surface mixed layer becomes almost completely saturated and the flux is basically zero. C\textsubscript{ant}, in contrast, has a 10 times longer equilibration time than CFCs, and in addition the atmospheric $p\text{CO}_2$ continues
to increase, while $p$CFC-11 already has stopped increasing in the 1990s. Therefore the upper ocean is basically never in equilibrium with the atmosphere for $C_{ant}$, and atmosphere-ocean flux pertains over the whole year. This also implies that $C_{ant}$ flux is not as limited to regions of wintertime deep convection as that of CFC-11 (or any other trace gas that also has a fast equilibration time).

4.2. Quantitative Assessment of the $C_{ant}$ Air-Sea Flux

[35] Direct validation of the modeled $C_{ant}$ fluxes using inverse estimates of $C_{ant}$ uptake [Gloor et al., 2003; Mikaloff Fletcher et al., 2006] is difficult, as these contain a number of assumptions, and resolve only patterns on scale of whole ocean gyres. In models, however, we can gain a better mechanistic understanding how the uptake of $C_{ant}$ is driven, and validate whether these mechanisms are represented correctly in the model.

[36] The spatial and temporal distribution of the air-sea flux of $C_{ant}$ can be understood as the result of the interplay between three driving processes:

[37] 1. First, there is a continuing increase of anthropogenic carbon in the atmosphere. In equilibrium, this increase would be accompanied by a corresponding increase of $C_{ant}$ in the surface mixed layer of the ocean. The discrepancy between the 'new' equilibrium and the actual $C_{ant}$ concentration drives air-sea flux.

[38] 2. Second, water within the mixed layer experiences temperature changes while being moved about. Owing to the temperature dependency of the carbonate chemistry in seawater, the equilibrium concentration of $C_{ant}$ is positively correlated with temperature; that is, an increase in temperature leads to an increase in $C_{ant}$ content in equilibrium for a given $p$CO$_2$ [Wallace, 2001; Völker et al., 2002]. This is in contrast to the behavior of total CO$_2$ which, for a given $p$CO$_2$, increases with decreasing temperature. As before, the difference between equilibrium and the actual $C_{ant}$ concentration drives air-sea flux.

[39] 3. Third is the entrainment into the mixed layer of older water from below containing different, usually less or no $C_{ant}$. This entrainment can be caused by the annual cycle of mixed-layer depth or by lateral or vertical advection into the mixed layer.

[40] To investigate more quantitatively the role of the three driving processes of $C_{ant}$ uptake enumerated in the beginning of this section, we analyze the Lagrangian rate of change of $C_{ant}$ of a water column that moves horizontally in the mixed layer of the ocean.

[41] Following Follows and Williams [2004], we define $C$ as the mixed-layer disequilibrium in $C_{ant}$; that is, the difference between the value that would be in equilibrium for a given $p$CO$_2$ and the actual $C_{ant}$ concentration. From the mass balance for $C_{ant}$ averaged over the mixed layer depth $h$ one obtains

$$\frac{DC'}{Dt} + \frac{F}{h} = E_1 + E_2 + E_3.$$  \hspace{1cm} (1)

where $F$ is the air-sea flux of $C_{ant}$ and the three terms on the right hand side stem isolate the forcing by the three processes mentioned in the begin of the section in the order that they appear there. They can be analyzed using an annual cycle of monthly average velocity and tracer fields from the model run and then forming an annual average.
$E_1$ corresponds to increasing $p\text{CO}_2$ and is defined by:

$$E_1 = \frac{\partial C_{\text{ant}}^{eq}}{\partial p\text{CO}_2} \frac{\partial p\text{CO}_2}{\partial t}$$

(2)

where $C_{\text{ant}}^{eq}$ is the concentration of $C_{\text{ant}}$ in equilibrium with the instantaneous $p\text{CO}_2$. As we are using the Siegenthaler-Sarmiento approximation for anthropogenic carbon, the first term on the right-hand side can be calculated from mixed-layer temperature and atmospheric $p\text{CO}_2$. The result is shown in Figure 9a. The spatial structure of that term mainly reflects that of the sea-surface temperature. Highest values are found where temperature is high, reflecting the temperature dependency of the Revelle or buffer factor.

$E_2$ corresponds to heat flux forcing and is:

$$E_2 = \frac{\partial C_{\text{ant}}^{eq}}{\partial T} \frac{DT}{Dt}$$

(3)

where $DT/dt$ is the Lagrangian derivative of the mixed-layer temperature. Again, the first term on the rhs. can be evaluated from mixed-layer temperature and atmospheric $p\text{CO}_2$, using the Siegenthaler-Sarmiento approach. The second term can be evaluated from the Lagrangian form of the internal energy balance for the mixed layer:

$$\frac{DT}{Dt} = \frac{Q}{\rho c_p h} + E_T.$$  

(4)

where $Q$ is the surface heat flux, and $E_T$ is the rate of change of temperature due to the entrainment of water of different temperature from below. The result of the calculation using model fields is shown in Figure 9b. Note that the range of values is much larger than in Figure 9a. The largest values are reached around the equator and in the southern end of the subpolar gyre where heat fluxes are strongly positive. The pattern differs quite strongly from the pattern of annually averaged heat-flux alone: over the large areas of the subtropical and subpolar North Atlantic the values of $E_2$ are positive although heat fluxes are negative. This is explained by the rectifying effect of the temporal covariance between heat flux and mixed layer depth, as shown in
The annual average of $Q/(\rho C_p h)$ can be positive even when the average heat flux $Q$ is strongly negative if, over the annual cycle, positive values of $Q$ are more likely to occur during times of shallow mixed layer than negative values. Advective contributions to $E_2$ are generally small. 

The term $E_3$ finally describes the change of $C_{\text{ant}}$ by vertical entrainment. Concentrations within the mixed layer change only when the mixed layer is deepening, entraining water from below with a different $C_{\text{ant}}$, not when it is shallowing. This leads to:

$$E_3 = \begin{cases} 0 & \text{if } \frac{Dh}{Dt} \leq 0 \\ \frac{Dh}{Dt} \frac{C_{\text{ant}}^b - C_{\text{ant}}}{C_{\text{ant}}/C_0/C_1} & \text{if } \frac{Dh}{Dt} > 0 \end{cases}$$

where $Dh/Dt$ is the Lagrangian time derivative of mixed layer depth, and $C_{\text{ant}}^b$ is the anthropogenic carbon concentration in the entrained water below the mixed layer. Note that this definition of entrainment corresponds to the definition of 'detrainment' as used in studies of mode water ventilation [e.g., Marshall et al., 1993]. The distribution of $E_3$ (Figure 9c) is dominated by the influence of the annual mixed layer cycle over most of the subpolar domain and by lateral entrainment over large parts of the Gulf stream region. The values of $E_3$ are high throughout the entire subpolar North Atlantic, not only over the main convection regions in the Labrador and Irminger seas.

The sum of all three forcing terms $E_1 + E_3 + E_3$ is shown in Figure 9d. It exhibits maximum values over the Labrador and Irminger seas related to vertical exchange, but another, very pronounced maximum at the southern end of the subpolar gyre, off Newfoundland, that is caused by the ocean heat gain. For an infinitely fast gas exchange, $E_1 + E_3 + E_3$ should be equal to the annual mean of $F/h$, the $C_{\text{ant}}$ air-sea flux divided by the mixed layer depth (equation (1)). Owing to the long equilibration time of $C_{\text{ant}}$, however, the maximum values of $F/h$ are shifted downstream relative to those of $E_1 + E_3 + E_3$. We can therefore attribute the increased $C_{\text{ant}}$ flux rates along the path of the North Atlantic Current and its recirculation in the Irminger Basin, shown in Figure 5b, primarily to the heat-flux forcing off Newfoundland.

Over most of the subpolar gyre, the entrainment terms dominates the forcing of air-sea flux, while the temperature-related forcing dominates over the subtropics (Figure 11). The term caused by the temporal increase in $pCO_2$ is only a minor Contribution; that is, even if the increase of anthropogenic carbon in the atmosphere was
stopped, the ocean would continue taking up \( \text{C}_{\text{ant}} \) at a similar rate for some time.

[47] One could make a similar calculation also with respect to \( \text{CFC} \) flux. However, because the equilibrium concentration of \( \text{CFC} \) decreases with increasing temperature while the converse hold for \( \text{C}_{\text{ant}} \), one can immediately infer, that the forcing term corresponding to \( E_2 \) (i.e., the heat-flux related forcing) must have the opposite sign from that of \( \text{C}_{\text{ant}} \). We therefore do not expect a similar forcing of uptake off Newfoundland, and thus no enhanced uptake along the path of the North Atlantic current downstream.

5. Temporal Variability of the \( \text{C}_{\text{ant}} \) Transport

[48] Observational studies agree that the transport of \( \text{C}_{\text{ant}} \) is directed northward, opposite to the transport of total dissolved inorganic carbon, throughout the tropical and subtropical Atlantic [Holfort et al., 1998; MacDonald et al., 2003; Roson et al., 2003]. Nevertheless, some controversy still exists on the size of that transport. This has led to contradicting inferences about the magnitude and even the sign of the air-sea flux of \( \text{C}_{\text{ant}} \) within the North Atlantic basin [Wallace, 2001; Völker et al., 2002; MacDonald et al., 2003; Roson et al., 2003; Gloor et al., 2003].

[49] In agreement with these estimates, the transport of anthropogenic \( \text{C}_{\text{ant}} \) in this model is directed northward (Figure 12) over the tropical and subtropical Atlantic. While increasing in time, the transport is subject to substantial variations on a monthly timescale that can be in the same order of magnitude as the annual mean transport. The annual mean transport at 25°N is 10–20% lower than the estimate of MacDonald et al. [2003], albeit still within their error estimate (see mean and error bars in Figure 12). This corresponds well to the fact that the maximum northward heat transport in the model is also lower than data-based estimates by about the same factor (1 PW, compared to 1.2 ± 0.3 PW given by Ganachaud and Wunsch [2000]).

[50] Why does the meridional transport of \( \text{C}_{\text{ant}} \) agree so well with the data-based estimates in contrast to the total inventory? The total inventory depends strongly on small concentrations of \( \text{C}_{\text{ant}} \) over the intermediate and deep waters that depend on the formation, spreading, and mixing rates of the source water masses. These dependencies affect both the data-based estimates and our model results. The transport of \( \text{C}_{\text{ant}} \), on the other hand, is dominated by large surface values of \( \text{C}_{\text{ant}} \) that coincide with the strongest meridional velocities. Surface concentrations of \( \text{C}_{\text{ant}} \) are almost in equilibrium with the atmosphere and thus not very dependent on possible model errors. Since the upper branch of the thermohaline circulation, the Florida and the Antilles Currents, are also within the range of observations, the \( \text{C}_{\text{ant}} \) transport is well constrained.

[51] There is a good temporal correlation between the transports of \( \text{C}_{\text{ant}} \) and heat at 25°N (Figure 12a), in contrast to the situation at 45°N (Figure 12b). The correlation coefficient between the two transports is shown in Figure 13 as a function of latitude. Whereas monthly \( \text{C}_{\text{ant}} \) and heat transport are highly correlated with values around 0.9 over the whole tropical and subtropical gyres the correlation breaks down sharply near 40°N, and is almost zero in the subpolar gyre. This is explained by analyzing the correlations of the two individual transports with the maximum of the meridional overturning cell (MOC, in Sv, at the given latitude). In the subtropical North Atlantic the meridional overturning determines both transports: the differences in temperature and \( \text{C}_{\text{ant}} \) between the upper and lower branches of the MOC are much larger than their zonal gradients,
leading to an only small contribution to the net northward transport by the horizontal gyre circulation. This MOC dominance does not hold in the subpolar gyre, where east-west property gradients become much more important. However, while the heat transport-overturning correlation totally breaks down owing to the zonal contrast of warm water flowing on the eastern side (the North Atlantic Current) and cold water flowing southward on the western side (the Labrador Current), the C\textsubscript{ant} contrast of both current regimes is much smaller than for temperature, still leaving room for a strong northward transport of C\textsubscript{ant} at the surface that is correlated to the upper branch of the meridional overturning.

The almost perfect correlation at midlatitudes between the C\textsubscript{ant} and heat transports also holds on interannual-decadal timescales. Figure 14a shows both transports smoothed by a 2-year filter removing the (intra-)seasonal variability in both time series. Although variations are now on a smaller scale (±0.1 PgC/year), the heat and C\textsubscript{ant} transports are still are still corresponding to each other. This implies that the ongoing efforts in establishing a monitoring system for the heat transport in the North Atlantic [Hirschi et al., 2003] will also be useful for estimating the variability of C\textsubscript{ant} transports.

It has already been shown that the seasonal to interannual heat transport variability at 25\textdegree N is primarily governed by the wind-driven (Ekman) transports and their compensation by deep return flows [Jayne and Marotzke, 2001]. Figure 14b shows that this is similar for the transport of anthropogenic carbon. In the experiment HEAT, forced by interannual heat fluxes but climatological wind stress, the year-to-year variability of C\textsubscript{ant} transport is lowered by interannual heat fluxes but climatological wind stress, of anthropogenic carbon. In the experiment HEAT, forced by interannual heat fluxes but climatological wind stress, the year-to-year variability of C\textsubscript{ant} transport is lowered by interannual heat fluxes but climatological wind stress, of anthropogenic carbon.

\[ \text{Figure 14. As in Figure 12a but for interannual transports for (a) Experiment REF and (b) Experiment HEAT. Note that C}_{\text{ant}} \text{ transports are detrended to remove the atmospheric increase.} \]

of an eddy-permitting general circulation model. The model outcomes show patterns of C\textsubscript{ant} distribution, uptake and transport that are broadly in agreement with earlier studies using less highly resolved physics. However, they also show much more spatial structure, which results, for example, in a better representation on the observed zonal gradients of C\textsubscript{ant} in the deep North Atlantic. The total inventory appears low compared to estimations by Gruber [1998] and Lee et al. [2003]. It has, however, been argued that, especially in deep waters, the back-calculation techniques for estimating C\textsubscript{ant} might be fought with large uncertainties [Matsumoto and Gruber, 2005], although quantifying these uncertainties is difficult. The lower bias of the model inventory for C\textsubscript{ant} in the deep ocean appears in marked contrast to the fact that our model reproduces observations of the Labrador Sea Water inventory of CFC-11 quite well [Böning et al., 2003]. A similarly large underestimation of C\textsubscript{ant} inventory with respect to data-based estimates has also been found in another global model simulation (see the IPSL simulation discussed by Orr et al. [2001]) of C\textsubscript{ant} that uses the perturbation approach by Siegenthaler and Joos [1992].

The formation of subtropical mode waters is another important pathway for the sequestration of C\textsubscript{ant} [Follows et al., 2002]. Representing their formation and structure correctly requires even higher model resolutions, which would make longer global carbon model studies computationally expensive. Interestingly however, in our study the volume of subtropical and subpolar modewaters differs quite substantially between the eddy-permitting and the coarse-resolution model versions. Nevertheless, this still results in similar total inventories of C\textsubscript{ant} in both the model versions. The misrepresentation of one water mass in the coarse resolution model is compensated by a shift in the C\textsubscript{ant} distribution over density classes. However, if one is interested in changes of the ocean circulation and its effect on the C\textsubscript{ant} sequestration an explicit and correct representation of the individual water masses is essential. The better representation of the upper layer current structure, for example, of the energetic western boundary currents and its branches of the North Atlantic Current in the eddy-permitting simulation offers some new perspectives on the relevant mechanisms of the C\textsubscript{ant} uptake.

The gross spatial pattern and magnitudes of the air-sea flux of C\textsubscript{ant} in the eddy-permitting model are similar to the coarse-resolution model runs, and consistent with the results from an ensemble of global carbon model integra-
tions [Orr et al., 2001]. All simulations show maximum rates of uptake in the North Atlantic subpolar gyre, where deep convection brings old water poor in C\textsubscript{ant} to the surface. However, in the eddy-permitting model, the association between the relatively localized convection events and air-sea flux is much weaker for C\textsubscript{ant} than for CFC-11. The C\textsubscript{ant} uptake does not only extend over a much larger area in the eastern portion of the gyre, but also shows a clear association to the structure of the North Atlantic surface currents in the model.

[57] A novel Lagrangian analysis [after Follows and Williams, 2004] of the driving forces behind the C\textsubscript{ant} air-sea flux is able to separate the role of vertical mixing, heating or cooling, and the temporal evolution of pCO\textsubscript{2} on the flux. It shows that vertical mixing contributes most to the air-sea flux over the central subpolar gyre but that there is also a dominant contribution from cooling over the southern portion of the gyre: the latter mechanism contributes to the bulk of the uptake of C\textsubscript{ant} along the path of the North Atlantic Current. While the contribution by vertical mixing is highest in the regions of deep convection in the Labrador and Irminger Seas, it is also significant over parts of the subtropical North Atlantic, associated with the seasonal cycle of the mixed layer deepening. Although in the annual mean, the heat flux over large parts of the midlatitude North Atlantic is directed into the ocean, the effect on C\textsubscript{ant} fluxes is confounded by the temporal correlation between heat fluxes and mixed layer depth, leading to a net forcing of ocean uptake in spite of a net cooling. Finally, the continuing increase of pCO\textsubscript{2} is contributing little to the forcing of air-sea flux of C\textsubscript{ant} within most of the subpolar ocean. It is dominant only in the tropical ocean outside a band around the equatorial upwelling.

[58] Like the meridional heat transport, the transport of C\textsubscript{ant} by the ocean is directed northward throughout the subtropical North Atlantic and strongly correlated to variations in the overturning component of the oceanic circulation. Unlike heat transport, however, some correlation with the overturning continues to hold over parts of the subpolar regime, where heat transport if dominated by the gyre part of the circulation. The correlation holds over a large range of timescales from monthly over interannual, and perhaps even in the mean. The reason for the strong coupling of the C\textsubscript{ant} transport to the overturning is the large concentration difference between northward flow of surface waters that carry the imprint of the current pCO\textsubscript{2} and southward flow of older waters that have not been in contact with the atmosphere recently. This contrast is exacerbated by the fact that C\textsubscript{ant} values tend to be higher in warmer waters than in colder, because of differences in the buffer factor. A further implication of the correlation between the transports of heat and C\textsubscript{ant} is that coarse-resolution models, which tend to have a too sluggish overturning, and may underestimate the northward transport of C\textsubscript{ant} in the North Atlantic, and therefore perhaps overestimate the effect of the local air-sea flux [Wallace, 2001].

[59] The strong coupling between overturning and C\textsubscript{ant} transport implies that a reduction of the intensity of the Atlantic overturning by about 30% during the 21st century as prognosticated by climate models [Gregory et al., 2005] should lead to a significant decrease in C\textsubscript{ant} transport. One might speculate, however, whether such a reduction in transport might lead to enhanced or reduced air-sea flux of C\textsubscript{ant} north of that latitude. It would be interesting to know whether such a purely physical mechanism might be behind the inferred reduction in air-sea flux of C\textsubscript{ant} in recent studies by Omar et al. [2003], Lefèvre et al. [2004] and Omar and Olsen [2006]. However, because of the setup of our model (i.e., the perturbation approach for C\textsubscript{ant}), we are not able to discuss the potentially large feedbacks between variability in the physical circulation and the biological carbon pump [e.g., Follows and Dutkiewicz, 2002] and their repercussions on the uptake of C\textsubscript{ant}.

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