Convergence of atmospheric and North Atlantic CO_2 trends on multidecadal timescales

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The oceans' carbon uptake substantially reduces the rate of anthropogenic carbon accumulation in the atmosphere[1], and thus slows global climate change. Some diagnoses of trends in ocean carbon uptake have suggested a significant weakening in recent years[2-8], while others conclude that decadal variability confounds detection of long-term trends[9-11]. Here, we study trends in observed surface ocean partial pressure of CO_2 (pCO_2) in three gyre-scale biomes of the North Atlantic, considering decadal to multidecadal timescales between 1981 and 2009. Trends on decadal timescales are of variable magnitudes and depend sensitively on the precise choice of years. As more years are considered, oceanic pCO_2 trends begin to converge to the trend in atmospheric pCO_2. North of 30ºN, it takes 25 years for the influence of decadal-timescale climate variability to be overcome by a long-term trend that is consistent with the accumulation of anthropogenic carbon. In the permanently stratified subtropical gyre, warming has recently become a significant contributor to the observed increase in oceanic pCO_2. This warming, previously attributed to

both a multidecadal climate oscillation and anthropogenic climate forcing[12,13], is beginning to reduce ocean carbon uptake.

The ocean is the ultimate long-term sink for anthropogenic carbon, having taken up approximately 30% of anthropogenic emissions from preindustrial times to 1994[1]. Anthropogenic climate change may drive physical and biogeochemical shifts in the ocean that result in reduced efficiency of this sink. Detection of such "climate-carbon feedbacks" is of great interest, but is complicated by the influence of poorly-quantified decadal timescale variability[2-11,14,15].

Previous studies have estimated trends in the North Atlantic carbon sink from oceanic pCO_2 data and numerical model output for recent decades, but have not agreed as to its direction and magnitude[2-11,16]. Comparison of these studies is complicated by the different time periods, regions, and methodologies used. Distinct from previous studies, we determine trends in oceanic pCO_2 from data across three (3) large biogeographic regions ("biomes")[17] that together occupy 87% of the total area of the North Atlantic (Figure 1a). The northern seasonally stratified subpolar gyre (SP-SS) biome is cold and biologically productive, the southern permanently stratified subtropical gyre (ST-PS) biome is warm and has low productivity, and between these extremes is the seasonally stratified subtropical (ST-SS) biome. Our focus on biome-scale trends is motivated by relevance to the global scale partitioning of CO_2 between the atmosphere and the ocean.

Our methodology takes advantage of the strengths of both methods previously used to study trends in the ocean carbon uptake potential: (1) pCO_2 observations from surface seawater and air, and (2) numerical models. The results we present are based solely on analysis of the data. The suitability of the methodology used to derive these results is double-checked by taking advantage of a numerical model that is subsampled as the data, and the resulting trend estimates are then compared to trends calculated from all model points (see Methods and Supplementary Information, sections 1 and 2). Our data are 1,116,539 each for oceanic pCO_2 and sea surface temperature (SST) from 1981-2009[18], and 797 dissolved inorganic carbon (DIC), alkalinity (ALK), sea surface salinity (SSS) and SST observations along a commercial shipping route between Iceland and Newfoundland (SURATLANT, SURveillance ATLANTique) for 1993-2007[5,6], from which pCO_2 values were computed[19]. We compare trends of oceanic pCO_2 to those for atmospheric pCO_2 estimated from a global observational network[20] for each biome. Since the air-sea CO_2 flux is proportional to the sea-air pCO_2 difference, it has previously been assumed that if the rate of increase in oceanic pCO_2 is faster than the rate of increase in atmospheric pCO_2, then the ocean carbon sink of that region is declining, and vice versa; and, that if the rate of change of oceanic pCO_2 is statistically indistinguishable from that of atmospheric pCO_2, then the carbon sink in that region is steady[4-7,10]. However, this is not strictly true since both temperature change and modification of dissolved inorganic carbon and alkalinity through surface freshwater fluxes or circulation variability could change oceanic pCO_2 without CO_2 uptake from or release to the atmosphere. In this study, we do compare rates of increase of oceanic and atmospheric pCO_2, but the strength of the carbon sink is interpreted in more detail based on decomposition of oceanic pCO_2 trends into two driving components[21]. The pCO_2-T trend is the part of an oceanic pCO_2 trend driven by SST change, and thus indicates the influence of changing physics, for example surface heat fluxes and heat advection. The pCO_2-nonT trend indicates accumulation or loss of carbon in the surface ocean or other chemical changes that modify oceanic pCO_2. For only SURATLANT, more detailed chemical data allows a further decomposition of oceanic pCO_2 change into a part associated with carbon accumulation or loss (dissolved inorganic carbon is directly related to oceanic pCO_2) and a part associated with the charge balance of major ions (alkalinity is inversely related to oceanic pCO_2). All trends are presented with 1σ uncertainty bounds[2], and as in previous studies[5-7], an indistinguishable difference between trends occurs when these bounds overlap (see Methods).

For 1981-2009, trends in oceanic pCO_2 are indistinguishable from trends in atmospheric pCO_2 in all biomes (Figure 1a; Figure 1c, gray bars). Trends are due to changing chemistry of the surface ocean (pCO_2-nonT) in all biomes (Figure 1c, green bars), which is consistent with a long-term oceanic equilibration with atmospheric pCO_2. Additionally, in the permanently stratified subtropical gyre (ST-PS) there is a significant contribution to the oceanic pCO_2 trend from rising temperatures (Figure 1c, blue bars).

Between the mid-1990's and mid-2000's, the North Atlantic Oscillation transitioned from a strong positive to a neutral or slightly negative phase, and at the same time, the longer-term Atlantic Multidecadal Variation transitioned from a negative to a positive phase[12,22]. A trend analysis for the period 1993-2005 is indicative of oceanic pCO_2 trends driven by such climatic transitions. Comparison of oceanic pCO_2 to atmospheric pCO_2 trends (Figure 1b) differs among the 3 biomes for this 13 year period: indistinguishable in the subpolar biome (SP-SS); oceanic pCO_2 increasing more rapidly in seasonally stratified subtropical biome (ST-SS); and oceanic pCO_2 increasing more slowly in the permanently stratified subtropical gyre biome (ST-PS).

In the subpolar biome for 1993-2005, both warming and chemistry drive the positive trend in oceanic pCO_2 (Figure 1d). For SURATLANT, contained within the subpolar biome, warming was responsible for the increase in oceanic pCO_2 and chemistry changes were negligible[2,5,6]. Alkalinity and dissolved inorganic carbon data allow further decomposition of the chemical change (Figure 1d, inset; ref. 10), which reveals that increasing sea surface salinity[22] and decreasing salinity-normalized alkalinity (sALK) drove up oceanic pCO_2. If salinity changes were only due to surface fluxes of freshwater, then the alkalinity / dissolved inorganic carbon ratio should not have changed and the impact on oceanic pCO_2 by pCO_2-SSS should have been small. The fact that the pCO_2-SSS trend is not small suggests that the alkalinity / dissolved inorganic carbon ratio of waters mixing into the area did change[6]. Salinity-normalized dissolved inorganic carbon (sDIC) does not drive a significant trend in oceanic pCO_2 in SURATLANT, which is consistent with little or no net carbon accumulation in the western subpolar gyre from 1993-2005[6,10]. Yet, the positive trend in pCO_2-nonT for the entire subpolar gyre biome is consistent with some larger-scale carbon accumulation, which is consistent with observations in the Norwegian Sea[23]. In this biome, with the North Atlantic Oscillation and Atlantic Multidecadal Variation phase transition from the mid-1990's to the mid-2000's, warming and reduced surface buoyancy loss led to reduced deep convection, less injection of cold waters to the gyre core, and thus, a slowing of the subpolar gyre's geostrophic circulation[22,24,25]. This analysis identifies the same warming trend and, at the same time, suggests a lowered rate of pCO_2-nonT increase in SURATLANT and the subpolar biome that is consistent with reduced vertical supply of dissolved inorganic carbon from the deep ocean[10]. This SURATLANT / subpolar biome comparison also highlights the fact that biome-scale, spatially-integrated trends do not preclude the existence of different trends at smaller scales[2,4-6,10,13].

In the seasonally stratified subtropical biome (ST-SS) for 1993-2005 there is a larger rate of oceanic pCO_2 increase than atmospheric pCO_2, driven by the chemistry term (Figure 1d)[4,7]. The aforementioned changes in the subpolar gyre circulation have been associated with a slowing of the surface circulation[11,22,24] and a reduced supply of low dissolved inorganic carbon waters from the subtropics along the North Atlantic Current[9]. This is consistent with increased dissolved inorganic carbon accumulation in ST-SS (Figure 1d for 1993-2005, green bar larger than in Figure 1c for 1981-2009). Finally, for 1993-2005 in the subtropical gyre biome (ST-PS, Figure 1b,d), oceanic pCO_2 went up more slowly than atmospheric pCO_2, with the oceanic increase driven by both warming and chemical change consistent with carbon accumulation.

Across the North Atlantic, biome-scale trends in oceanic pCO_2 are more similar to trends in atmospheric pCO_2 on long timescales than on short ones. How does the system transition from the shorter-timescale regime, significantly modulated by temperature changes, a proxy for the influence of decadal-timescale variability (Figure 1b,d), to the long-term regime more influenced by carbon accumulation (Figure 1a,c)? Given the sparse data, we would also like to know the sensitivity of oceanic pCO_2 trend estimates to the choice of years for a trend analysis.

Figure 2 is a comparison of oceanic pCO_2 trends to atmospheric pCO_2 trends for start years ranging from 1981 to 1993 and end years ranging from 2001 to 2009. For timeseries shorter than 25 years in the subpolar biome (SP-SS, Figure 2a), estimated trends vary significantly based on the choice of years, and pCO_2-T trends are frequently greater than zero. However, for timeseries at least 25 years long, oceanic pCO_2 trends are, with only one exception, consistent with atmospheric pCO_2. For these long timeseries, warming contributes to the oceanic pCO_2 trend only for timeseries starting in 1981; chemistry otherwise drives trends. Convergence of the oceanic pCO_2 trends to the atmospheric pCO_2 trend for timeseries longer than 25 years is a robust feature, and the fact that temperature trends are largely indistinguishable from zero suggests that carbon accumulation is the primary driver of these trends. However, a long-term waning influence of pCO_2-T is not entirely clear, given that timeseries starting in 1981 continue to be influenced by warming; and thus, multi-decadal climate variability may still be influencing subpolar biome pCO_2 trends[12,13,25] over the full period for which data is available. In the seasonally stratified subtropical biome (ST-SS, Figure 2b) oceanic pCO_2 trends are also sensitive to the choice of years for short timeseries. Beyond 25 years, oceanic pCO_2 trends are, with only one exception, indistinguishable from atmospheric pCO_2 trends. Intriguingly, warming significantly influences only one oceanic pCO_2 trend in ST-SS (Figure 2b, stippled; 1981-2001), indicating that chemical changes dominate these trends. These chemical changes are likely driven by variations in horizontal advection and vertical mixing[9,10,13]. In the permanently stratified subtropical gyre biome (ST-PS, Figure 2c), oceanic pCO_2 trends are generally the same as atmospheric pCO_2. However, in contrast to the northern biomes, the influence of warming on pCO_2 trends increases as years after 2006 are included (Supplementary Information, section 4; Supplementary Figure 8). With oceanic pCO_2 trends indistinguishable from atmospheric pCO_2, pCO_2-T trends greater than zero require pCO_2-nonT trends to be less than atmospheric pCO_2 (Figure 1c,d), which suggests that warming is damping ocean carbon uptake. The fact that this applies to almost all trends with end years 2006 to 2009, irrespective of start year, suggests that oscillatory behavior on interannual to decadal timescales is not strongly at play; instead this finding is consistent with a long-term tendency over these 29 years. The Atlantic Multidecadal Variation has a period of about 60 years, and likely explains some of this trend[6,12,13,22]. Anthropogenic forcing appears to be the other part of the explanation[12], and thus the increasing likelihood of a statistically significant influence of warming temperatures on oceanic pCO_2 trends in the subtropical gyre is consistent with a climate-carbon feedback by which anthropogenic warming reduces the ocean's ability to remove anthropogenic carbon from the atmosphere.

For both decadal and multi-decadal timescales, we find less dramatic amplitudes of recent trends in the North Atlantic surface ocean pCO_2 than others have suggested[2-7]. This is due, in part, to the fact that we estimate trends from observations across much larger, gyre-scale, regions than previously considered. Our parallel analysis with a numerical model indicates that sampling is sufficient for recovery of gyre-average oceanic pCO_2 trends, but uncertainty is still significant and will be best reduced with additional data. At the 1σ confidence level, we are able to detect short-term shifts in oceanic pCO_2, reasonably explained by climate variability[9-11], and north of 30ºN, long-term oceanic pCO_2 trends that track the rate of atmospheric pCO_2 increase. A significant role for the seasonally stratified biomes of the North Atlantic in the proposed multi-decadal increase in the atmospheric fraction of anthropogenic CO_2[8,26,27] is not distinguishable. However, in the North Atlantic permanently stratified subtropical gyre we do find an increasing influence on oceanic pCO_2 by a warming trend that is partially due to anthropogenic forcing[12]. This is evidence of a climate-carbon feedback that is beginning to limit the strength of the ocean carbon sink.

Methods

Database of $pCO_2^{s.ocean}$. Direct oceanic pCO_2 ($pCO_2^{s.ocean}$) measurements were made using air-seawater equilibration methods, and quality controlled and compiled as described in detail by Takahashi et al. (2009)[18]. We use data only within 0°N - 85°N, 100°W – 20°E. Coastal influences were eliminated by excluding data with SSS ≤20 pss. SURATLANT data[5,6] was merged to help with poor coverage in the early 2000's, resulting in 1,206,507 observations from 1981-2009, and of these, 1,117,336 points fall in our three biomes (Supplementary Table 1).

SURATLANT. Data were collected between Iceland and Newfoundland (ref. 5,6 through 2007). $pCO_2^{s.ocean}$ is calculated from measurements of DIC, SST, SSS, and ALK for 1993-1997 and 2001-2007 using accepted constants[19]. For 2001-2007, ALK was directly measured. For 1993-1997, ALK was estimated from the ALK-SSS relationship derived from 2001-2006 data (ALK = 43.857 * SSS + 773.8). We use open-ocean data from 50-64°N, 25-50°W. For comparison to previous work[2], we also study six 5°x5° regions (Figure 1b,d, Supplementary section 3, Supplementary Figure 6, Supplementary Tables 2 and 3).

Climatologies. The revised version (June 2009) of climatological mean $pCO_2^{s.ocean}$ at 4° (latitude) x 5° (longitude) resolution for reference year 2000[14] is used. We use climatological SST[28], SSS[29], DIC and ALK[30].

Trend in pCO_2^{atm}. A biome-average pCO_2^{atm} trend is calculated from the NOAA ESRL GLOBALVIEW-CO_2[20] reference marine boundary layer matrix xCO_2 using monthly mean values regridded to a 1°x1° grid, and surface pressure of 1 atm. The trend (b) is determined by a fit to y = a + b*t + c*cos(2πt + d), where t = decimal year -1990.

Biomes. Biomes[17] were assigned based on annual maximum mixed layer depth (MLD), annual mean SeaWiFS chlorophyll-a, and SST[28] at 1°x1° resolution. MLD uses a surface to depth density[29] difference of 0.125 kg/m^3. The seasonally stratified subpolar gyre biome (SP-SS) has chlorophyll ≥0.45mg/m^3 and SST 5-15°C. The seasonally stratified subtropical biome (ST-SS) biome has MLD >160m and chlorophyll <0.45 mg/m^3. The permanently stratified subtropical biome (ST-PS) has MLD ≤160m, SST ≥15°C and chlorophyll <0.2 mg/m^3. In the sea ice and low latitude upwelling biomes of ref. 17, there is insufficient data for analysis. See also Supplementary Information, section 1.

Estimation of $pCO_2^{s.ocean}$ trends for the biomes.

- i. Data are gridded to 1° x 1° spatial and then monthly temporal resolution.
- ii. Long-term mean removed to eliminate spatial aliasing
- iii. Data is averaged to the biomes, SURATLANT and its subregions.
- iv. A harmonic of the form y = a + b*t + c*cos(2πt + d), where t = decimal year - 1990, is fit. Trends reported are the value of b (in matm/yr) resulting from this fit.

Alternative trend analysis approaches were tried, but do not strongly influence results (Supplementary Information, section 1).

Trend uncertainty and trend comparisons. We present the 1σ confidence intervals (68.3%) calculated via:

$$CI_b = \pm t * RMSE * \sqrt{\frac{1}{\sum (X_i - \bar{X})^2}}$$

Where t is the two-tailed t-statistic for 68.3% confidence for N-4 degrees of freedom (DOF), with N being the number of months; RMSE is the root mean square error; X_i are the data; and $\overline{X}$ is the mean value. Distinguishability of trends determined by a student t-test with t* calculated from the data using:

$$t^* = \frac{b_{s.ocean} - b_{atm}}{\sigma_e / S_{xx}}$$

where $b_{s.ocean}$ is the surface ocean trend, b_{atm} is the atmospheric trend, σ_e is the sum of squared errors (SSE) divided by the DOF, and S_{xx} is calculated by $\sum_i^N (x_i - \bar{x})^2$. If t* is greater than $T_{(.683)}$ given the DOF, then the atmospheric and $pCO_2^{s.ocean}$ trends are significantly different. If t*< T then the trends are not significantly different (p-values are greater than 0.317).

Regional physical-biogeochemical model. Setup, forcing, ecosystem and carbon system details of the North Atlantic model at 0.5° x 0.5° horizontal resolution (MITgcm.NA) have been previously described[10], and has been extended to 1948-2009. The model compares well to physical and biogeochemical observations (Supplementary Figures 1 and 2; ref. 10). When sampling the model as the data, we do so at daily time and model spatial resolution, and then treat the sampled model as the data, using the model climatology in step (ii) of the analysis. We conclude that our methodology, applied to the available data, can capture real biome-scale trends in $pCO_2^{s.ocean}$ if trends from the model sampled as the data are within the 1σ uncertainty bounds of the trends estimated from all model points (Supplementary Information, section 2, Supplementary Figure 3).

Decomposition of $pCO_2^{s.ocean}$. $pCO_2^{s.ocean}$ is decomposed using empirical equations[21] into the isochemical component due to temperature (pCO_2-T) and the remaining variability (pCO_2-nonT). For SUR, we can also use the full equations to determine variability in

$pCO_2^{s.ocean}$ driven individually by SSS, DIC, and ALK[10]. We determine pCO_2-sDIC and pCO_2-sALK by making the calculations with salinity normalized DIC and ALK (sDIC = 35*DIC/SSS; sALK = 35*ALK/SSS) and adding the difference from the non-normalized component (pCO_2-DIC - pCO_2-sDIC and pCO_2-ALK - pCO_2-sALK) to pCO_2-SSS, which includes salinity variation effects only in pCO_2-SSS.

References

1. Denman, K.L. *et al*. Couplings Between Changes in the Climate System and Biogeochemistry. In: *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* [Solomon, S. et al (eds.)]. Cambridge University Press, Cambridge, United Kingdom (2007).
2. Schuster, U. *et al*. Trends in North Atlantic sea-surface fCO2 from 1990 to 2006. *Deep-Sea Res. II* **56**, 620-629 (2009).
3. Watson, A. J. *et al*. Tracking the variable North Atlantic sink for atmospheric CO2. *Science* **326**, 1391-1393 (2009).
4. Le Quéré, C. *et al*. Trends in the sources and sinks of carbon dioxide. *Nature Geoscience* **2**, 831-836 (2009).
5. Corbiere, A., Metzl, N., Reverdin, G., Brunet, C. & Takahashi, T. Interannual and decadal variability of the oceanic carbon sink in the North Atlantic subpolar gyre. *Tellus B* **59**, 168-178 (2007).

6. Metzl, N. *et al*. Recent acceleration of the sea surface fCO2 growth rate in the North Atlantic subpolar gyre (1993-2008) revealed by winter observations. *Global Biogeochem.Cycles*. **24**, GB4004 (2010).
7. Le Quéré, C., Takahashi, T., Buitenhuis, E.T., Rödenbeck, C. & Sutherland, S.C. Impact of climoate change on the global oceanic sink of CO2. *Global Biogeochem.Cycles*. **24**, GB4007 (2010).
8. Canadell, J. *et al.* Contributions to accelerating atmospheric CO2 growth from economic activity, carbon intensity, and efficiency of natural sinks. *Proc. Natl. Acad. Sci.* **104**, 18886-18870 *(*2007).
9. Thomas, H. *et al*. Changes in the North Atlantic Oscillation influence CO2 uptake in the North Atlantic over the past 2 decades. *Global Biogeochem. Cycles* **22,** GB4027 (2008).
10. Ullman, D.J., McKinley, G.A., Bennington, V. & Dutkiewicz S. Trends in the North Atlantic carbon sink: 1992–2006. *Global Biogeochem. Cycles* **23**, GB4011 (2009).
11. Gruber, N. Fickle trends in the ocean. *Nature* **458**, 155-156 (2009).
12. Ting, M-F., Kushnir, Y., Seager, R., & Li, C-H. Forced and Internal Twentieth-Century SST Trends in the North Atlantic. *J. Climate* **22,** 1469-1481 (2009).
13. Löptien, U., & C. Eden. Multidecadal CO2 uptake variability of the North Atlantic. *J. Geophys. Res.* **115**, D12113 (2010).
14. Takahashi, T. *et al*. Climatological mean and decadal change in surface ocean pCO2, and net sea–air CO2 flux over the global oceans. *Deep-Sea Res. II* **56,** 554-577 (2009).

15. Bates, N.R. Interannual variability of the oceanic CO2 sink in the subtropical gyre of the North Atlantic Ocean over the last 2 decades, *J. Geophys. Res.* **112**, C09013 (2007).

16. Rödenbeck, C. *Estimating CO2 sources and sinks from atmospheric concentration measurements using a global inversion of atmospheric transport* (Tech. Rep. 6, http://www.bgc-jena.mpg.de/~christian.roedenbeck/download_CO2/ , Max-Planck-Inst. for Biogeochem. Jena, 2005).

17. Sarmiento, J. L. *et al*. Response of ocean ecosystems to climate warming. *Global Biogeochem. Cycles* **18**, GB3003 (2004).

18. Takahashi, T., Sutherland, S.C. & Kozyr, *A. Global Ocean Surface Water Partial Pressure of* CO_2 *Database: Measurements Performed during 1957-2009 (Version 2009)*. (ORNL/CDIAC-152, NDP-088r. CDIAC, ORNL, US DOE, Oak Ridge, Tennessee, doi:10.3334/CDIAC/otg.ndp088r, 2010).

19. Follows, M., Ito, T., & Dutkiewicz, S. On the solution of the carbonate chemistry system in ocean biogeochemistry models. *Ocean Model.* **12***,* 290-301 (2006).

20. GLOBALVIEW-CO2: Cooperative Atmospheric Data Integration Project - Carbon Dioxide, Reference Matrix. (CD-ROM, ftp.cmdl.noaa.gov, Path: ccg/co2/GLOBALVIEW, NOAA ESRL, Boulder, CO, 2010).

21. Takahashi, T. *et al*. Global sea-air CO_2 flux based on climatological surface ocean pCO_2, and seasonal biological and temperature effects, Deep-Sea Res. II **49**, 1601-1622 (2002).

22. Reverdin, G. North Atlantic subpolar gyre surface variability (1895-2009). *J. Climate* **23**, JCLI3493.1 (2010).

23. Skjelvan, I. Falck, E. Rey, F. & Kringstad, S.B. Inorganic carbon time series at Ocean Weather Station M in the Norwegian Sea. *Biogeosci*. **5**, 549-560 (2008).

24. Häkkinen, S. & Rhines, P.B. Shifting surface currents in the northern North Atlantic Ocean. *J. Geophys. Res*. **114**, C04005 (2009).

25. Lozier, M.S. *et al*. The spatial pattern and mechanisms of heat-content change in the North Atlantic. *Science* **319**, 800 (2008).

26. Knorr, W. Is the airborne fraction of anthropogenic CO2 emissions increasing? *Geophys. Res. Lett*. **36**, L21710 (2009).

27. Sarmiento, J.L. *et al.* Trends and regional distributions of land and ocean carbon sinks. *Biogeosci.* **7**, 2351-2367 (2010).

28. Reynolds, R.W., *et al*. Daily high-resolution-blended analyses for sea surface temperature, *J. Climate* **20**, 5473-5496 (2007).

29. *World Ocean Atlas 2005, Volume 1 and 2*. S. Levitus, Ed. (NOAA Atlas NESDIS 61, U.S. Government Printing Office, Washington, D.C., 2005).

30. Key, R. *et al.* A global ocean carbon climatology: Results from Global Data Analysis Project (GLODAP). *Global Biogeochem.Cycles* **18**, GB4031 (2004).

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Author Contributions G.A.M designed the study and wrote the manuscript. A.R.F. did the data analysis. T.T. developed the oceanic pCO_2 database. N.M. synthesized the SURATLANT data. All authors discussed and revised the manuscript.

Supplementary Information accompanies the paper at www.nature.com/naturegeoscience.

Figure Captions

Figure 1: **Trends in oceanic pCO_2 for (a) 1981-2009 and (b) 1993-2005 compared to atmospheric pCO_2 trend** [20]. (a,b) Dark blue for oceanic pCO_2 trend less than atmospheric pCO_2 trend; pink for indistinguishable; red for larger oceanic trend; (b) includes SURATLANT (SUR) and 5°x5° subregions (b, inset)[2,5,6]. (c,d) Oceanic pCO_2 trends (gray), temperature (pCO_2-T, light blue) and chemical (pCO_2-nonT, green) components, with 1σ uncertainty, and atmospheric pCO_2 trend (dash). (d, inset) Decomposition of pCO_2 for SURATLANT to salinity-normalized dissolved inorganic carbon (pCO_2-sDIC), salinity-normalized alkalinity (pCO_2-sALK), and salinity (pCO_2-SSS) components. See also Supplementary Information, section 3, Supplementary Figures 4-6 and Supplementary Tables 4-5.

Figure 2: **Trend in oceanic pCO_2 vs. atmospheric pCO_2, variable years.** (a) Seasonally stratified subpolar, SP-SS (b) Seasonally stratified subtropical, ST-SS and (c) Permanently stratified subtropical, ST-PS. Colors as Figure 1a,b. Stippling for pCO_2-T trend distinguishable from zero (dot > 0; dark < 0); and in most of these cases (86%), the pCO_2-nonT trend is also distinguishable from the atmospheric pCO_2 trend. Bold lines at timeseries of 10, 15, 20, 25 year lengths. Crosses are 1981-2009, Figure 1a,c; stars are 1993-2005, Figure 1b,d. White if sampling insufficient (Supplementary Information section 3). See also Supplementary Information, section 3, and Supplementary Figure 7.

Figure 1

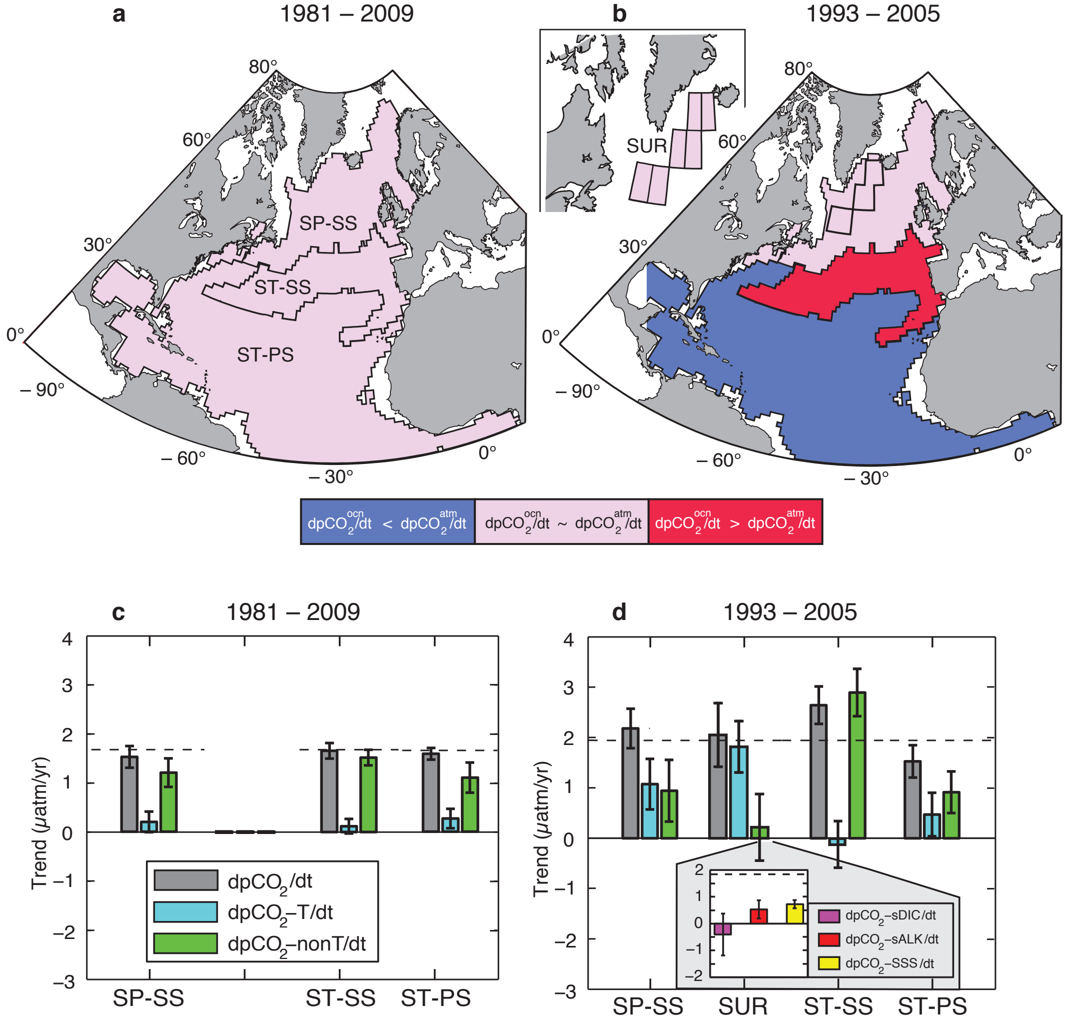
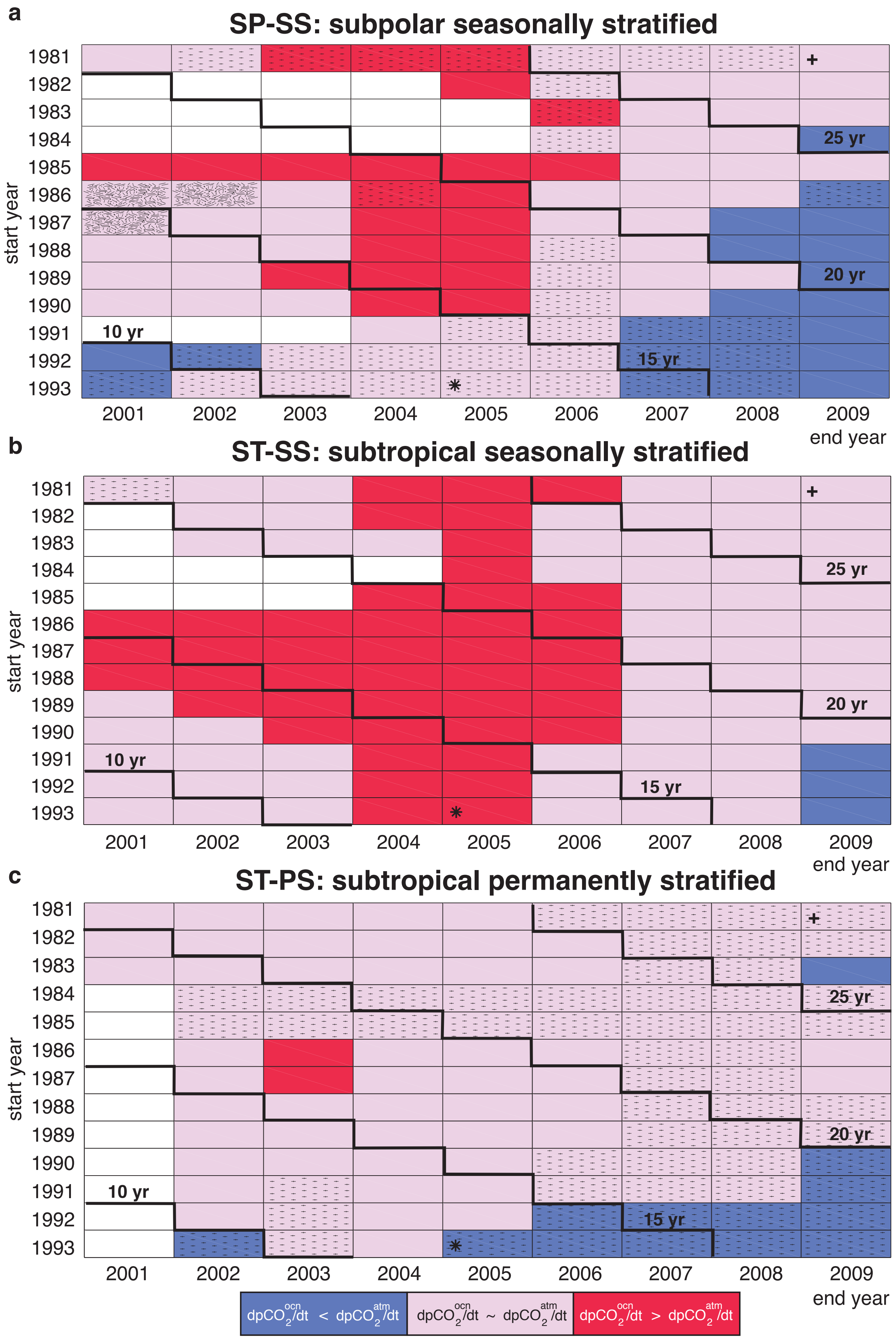

a
1981 – 2009
80°
60°
30°
0°
– 90°
– 60°
– 30°
0°
SP-SS
ST-SS
ST-PS
b
1993 – 2005
SUR
60°
80°
30°
0°
– 90°
– 60°
– 30°
0°
$dpCO_2^{ocn}/dt < dpCO_2^{atm}/dt$
$dpCO_2^{ocn}/dt \sim dpCO_2^{atm}/dt$
$dpCO_2^{ocn}/dt > dpCO_2^{atm}/dt$
c
1981 – 2009
Trend (μatm/yr)
4
3
2
1
0
−1
−2
−3
$dpCO_2/dt$
$dpCO_2$–T/dt
$dpCO_2$–nonT/dt
SP-SS
ST-SS
ST-PS
d
1993 – 2005
Trend (μatm/yr)
4
3
2
1
0
−1
−2
−3
2
1
0
−1
−2
$dpCO_2$–sDIC/dt
$dpCO_2$–sALK/dt
$dpCO_2$–SSS/dt
SP-SS
SUR
ST-SS
ST-PS

Figure 2


a
SP-SS: subpolar seasonally stratified
b
ST-SS: subtropical seasonally stratified
c
ST-PS: subtropical permanently stratified
start year
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
2001
2002
2003
2004
2005
2006
2007
2008
2009
end year
10 yr
15 yr
20 yr
25 yr
+
*
$dpCO_2^{ocn}/dt < dpCO_2^{atm}/dt$
$dpCO_2^{ocn}/dt \sim dpCO_2^{atm}/dt$
$dpCO_2^{ocn}/dt > dpCO_2^{atm}/dt$