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Convergence of atmospheric and North Atlantic carbon dioxide trends on multidecadal timescales

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Oceanic uptake of carbon dioxide substantially reduces the rate at which anthropogenic carbon accumulates in the atmosphere[1], slowing global climate change. Some studies suggest that the rate at which the oceans take up carbon has significantly decreased in recent years[2–8]. Others suggest that decadal variability confounds the detection of long-term trends[9–11]. Here, we examine trends in the partial pressure of carbon dioxide in the surface waters of three large biogeographic regions in the North Atlantic, using observational data collected between 1981 and 2009. We compare these oceanic observations with trends in atmospheric carbon dioxide levels, taken from a global observational network. We show that trends in oceanic carbon dioxide concentrations are variable on a decadal timescale, often diverging from trends in atmospheric carbon dioxide. However, when the entire 29-year period is considered, oceanic trends converge with atmospheric trends in all three regions; it takes 25 years for this long-term trend to emerge and overcome the influence of decadal-scale variability. Furthermore, in the southernmost biome, the data suggest that warming—driven by a multidecadal climate oscillation and anthropogenic forcing[12,13]—has started to reduce oceanic uptake of carbon in recent years.

The ocean is the ultimate long-term sink for anthropogenic carbon, having taken up approximately 30% of anthropogenic emissions from pre-industrial times to 1994 (ref. 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 large biogeographic regions ('biomes')[17] that together occupy 87% of the total area of the North Atlantic (Fig. 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 Sections S1 and S2). Our data are 1,116,539 each for oceanic pCO_2 and sea surface temperature (SST) from 1981 to 2009 (ref. 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 (refs 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. As 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, because 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 (Fig. 1a; Fig. 1c, grey bars). Trends are due to changing chemistry of the surface ocean (pCO_2-nonT) in all biomes (Fig. 1c, green bars), which is

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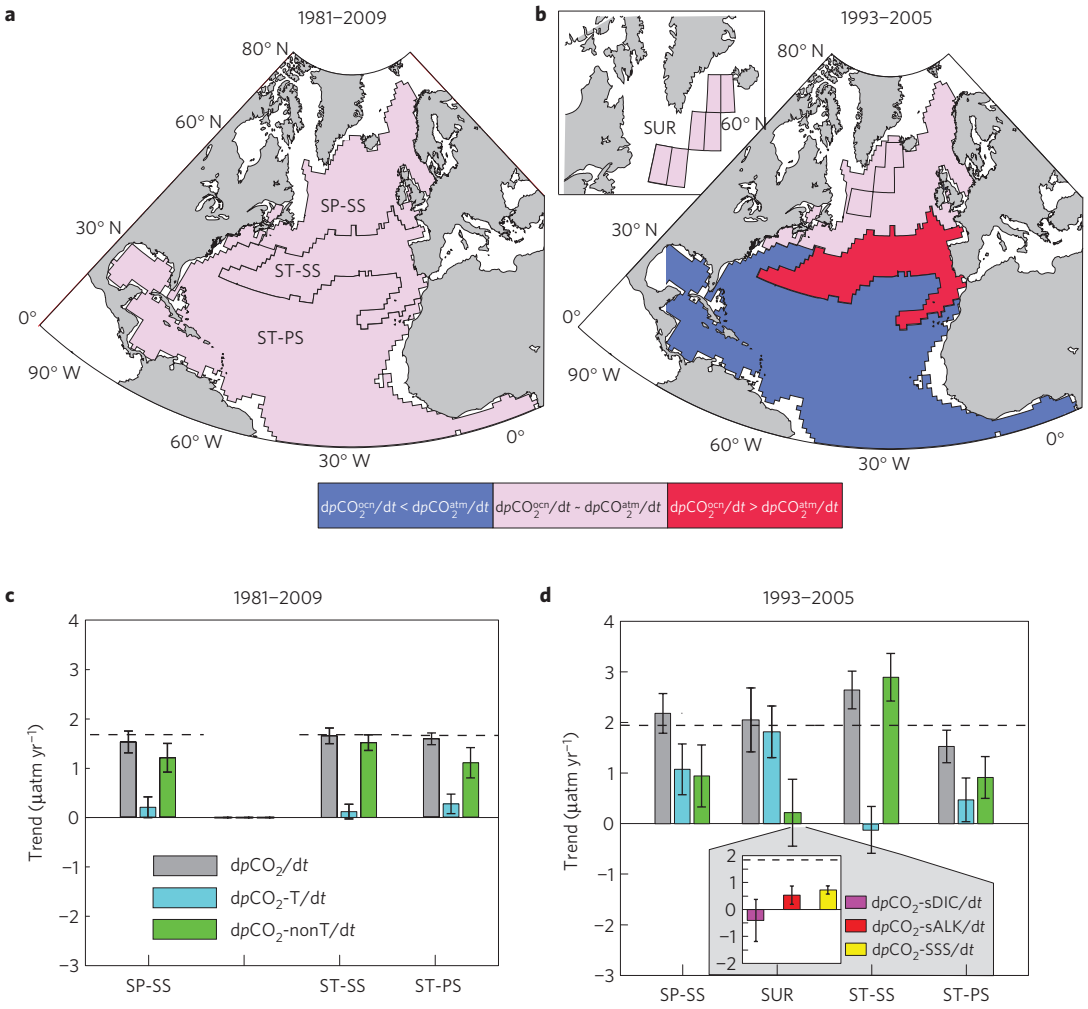

Figure 1 | Trend in oceanic pCO_2 compared to atmospheric pCO_2 trend[20] for a multidecadal and a decadal period. a,**c**, 1981–2009 and **b**,**d**, 1993–2005. **a**,**b**, Dark blue for oceanic pCO_2 trend less than atmospheric pCO_2 trend; pink for indistinguishable; red for larger oceanic trend. **b**, Includes SURATLANT (SUR) and 5° × 5° subregions (**b**, inset)[2,5,6]. **c**,**d**, Oceanic pCO_2 trends (grey), 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 Section S3, Figs S4–S6 and Tables S4–S5.

consistent with a long-term oceanic equilibration with atmospheric pCO_2. Furthermore, in the permanently stratified subtropical gyre (ST-PS) there is a significant contribution to the oceanic pCO_2 trend from rising temperatures (Fig. 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 (Fig. 1b) differs among the three 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 (Fig. 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 (Fig. 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 to 2005 (refs 6,10). However, 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 (Fig. 1d; refs 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 (Fig. 1d for 1993–2005, green bar larger than in Fig. 1c for 1981–2009). Finally, for 1993–2005 in the subtropical gyre biome (ST-PS, Fig. 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 (Fig. 1b,d), to the long-term regime more influenced by carbon accumulation (Fig. 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 time series shorter than 25 years in the subpolar biome (SP-SS, Fig. 2a), estimated trends vary significantly based on the choice of years, and pCO_2-T trends are frequently greater than zero. However, for time series at least 25 years long, oceanic pCO_2 trends are, with only one exception, consistent with atmospheric pCO_2. For these long time series, warming contributes to the oceanic pCO_2 trend only for time series starting in 1981; chemistry otherwise drives trends. Convergence of the oceanic pCO_2 trends to the atmospheric pCO_2 trend for time series 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 time series starting in 1981 continue to be influenced by warming; 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, Fig. 2b) oceanic pCO_2 trends are also sensitive to the choice of years for short time series. 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 (Fig. 2b, stippled; 1981–2001), indicating that chemical changes dominate these trends. These chemical changes are probably driven by variations in horizontal advection and vertical mixing[9,10,13]. In the permanently stratified subtropical gyre biome (ST-PS, Fig. 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 Section S4 and Fig. S8). A pCO_2-T trend greater than zero indicates a reduction in ocean carbon uptake potential, and as such, is almost always associated with a pCO_2-nonT trend less than atmospheric pCO_2 (98% in ST-PS, 86% in all biomes, Fig. 1c,d). In ST-PS, multidecadal convergence of oceanic pCO_2 trends to atmospheric pCO_2 trends can be partially attributed to warming; and thus less carbon uptake has occurred than would have occurred absent the warming. The fact that this applies to almost all trends with

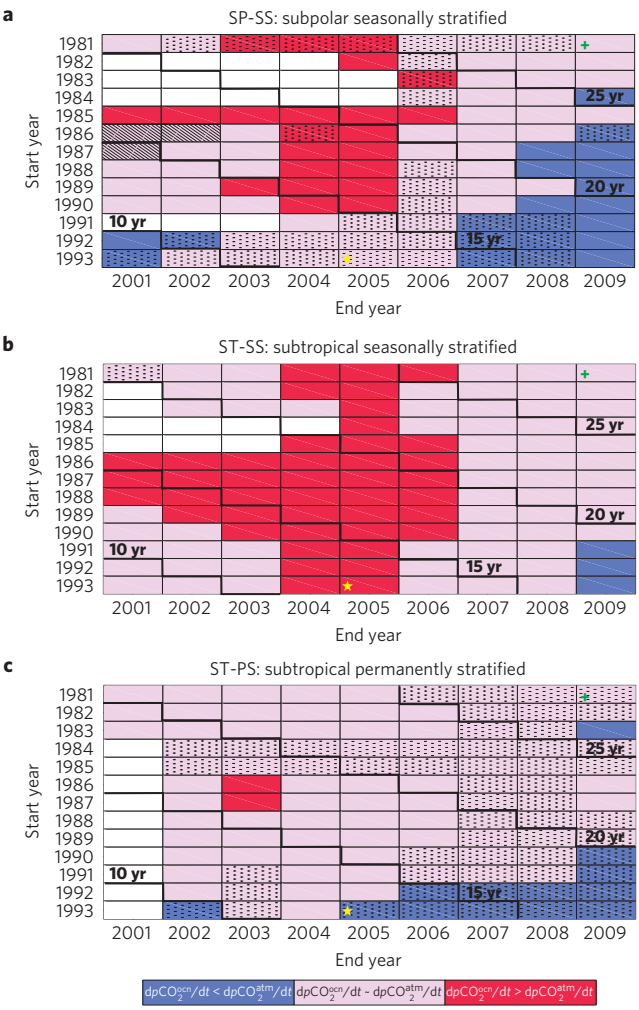

Figure 2 | Trend in oceanic pCO_2 versus atmospheric pCO_2, variable years. a, Seasonally stratified subpolar, SP-SS. **b**, Seasonally stratified subtropical, ST-SS. **c**, Permanently stratified subtropical, ST-PS. Colours as Fig. 1a,b. Stippling for pCO_2-T trend distinguishable from zero (dotted boxes > 0; diagonally lined boxes < 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 time series of 10, 15, 20, 25 years in length. Green crosses are 1981–2009 in Fig. 1a,c; yellow stars are 1993–2005 in Fig. 1b,d. White indicates sampling is insufficient (Supplementary Section S3). See also Supplementary Section S3 and Fig. S7.

end years 2006 to 2009, irrespective of start year, suggests that oscillatory behaviour 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 probably explains some of this trend[6,12,13,22]. Anthropogenic forcing seems 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 pronounced 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 further 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 30° N, 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 (refs 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; ref. 18). We use data only within 0° N–85° N, 100° W–20° E. Coastal influences were eliminated by excluding data with SSS $\leq$ 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 to 2009, and of these, 1,117,336 points fall in our three biomes (Supplementary Table S1).

SURATLANT. Data were collected between Iceland and Newfoundland (refs 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 to SSS relationship derived from 2001 to 2006 data ($ALK = 43.857 * SSS + 773.8$). We use open-ocean data from 50° to 64° N, 25° to 50° W. For comparison to previous work[2], we also study six 5° × 5° regions (Fig. 1b,d, Supplementary Section S3, Fig. S6 and Tables S2 and S3).

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

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

Biomes. Biomes[17] were assigned on the basis of annual maximum mixed layer depth (MLD), annual mean SeaWiFS chlorophyll-*a*, and SST (ref. 28) at 1° × 1° 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 $\geq$0.45 mg m^{-3} and SST 5 °C–15 °C. The seasonally stratified subtropical biome (ST-SS) biome has MLD >160 m and chlorophyll <0.45 mg m^{-3}. The permanently stratified subtropical biome (ST-PS) has MLD $\leq$ 160 m, SST $\geq$ 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 Section S1.

Estimation of $pCO_2^{s.ocean}$ trends for the biomes. (1) Data are gridded to 1° × 1° spatial and then monthly temporal resolution. (2) Long-term mean removed to eliminate spatial aliasing. (3) Data is averaged to the biomes, SURATLANT and its subregions. (4) A harmonic of the form $y = a + b*t + c*\cos(2\pi t + d)$, where t = decimal year − 1990, is fit. Trends reported are the value of b (in µatm yr^{-1}) resulting from this fit.

Alternative trend analysis approaches were tried, but do not strongly influence the results (Supplementary Section S1).

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

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

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. Set-up, forcing, ecosystem and carbon system details of the North Atlantic model at 0.5° × 0.5° horizontal resolution (MITgcm.NA) have been previously described[10], and have been extended to 1948–2009. The model compares well to physical and biogeochemical observations (Supplementary Figs and S1 and S2; 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 (2) 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 Section S2 and Fig. S3).

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 (ref. 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.

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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.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturegeoscience. Reprints and permissions information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to G.A.M.