Acceleration of ocean warming, salinification, deoxygenation and acidification in the surface subtropical North Atlantic Ocean

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Ocean chemical and physical conditions are changing. Here we show decadal variability and recent acceleration of surface warming, salinification, deoxygenation, carbon dioxide (CO2) and acidification in the subtropical North Atlantic Ocean (Bermuda Atlantic Time-series Study site; 1980s to present). Surface temperatures and salinity exhibited interdecadal variability, increased by ~0.85 °C (with recent warming of 1.2 °C) and 0.12, respectively, while dissolved oxygen levels decreased by ~8% (~2% per decade). Concurrently, seawater DIC, fCO2 (fugacity of CO2) and anthropogenic CO2 increased by ~8%, 22%, and 72% respectively. The winter versus summer fCO2 difference increased by 4 to 8 μatm decade−1 due to seasonally divergent thermal and alkalinity changes. Ocean pH declined by 0.07 (~17% increase in acidity) and other acidification indicators by ~10%. Over the past nearly forty years, the highest increase in ocean CO2 and ocean acidification occurred during decades of weakest atmospheric CO2 growth and vice versa.

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Recent acceleration of changes in ocean physics and chemistry since the 1980s is demonstrated at two sustained open-ocean hydrographic stations in the North Atlantic Ocean near Bermuda. Of these two time-series sites, Hydrostation S (formerly known as the Panulirus site) is the oldest, located ~25 km southeast of Bermuda at 32°10′N, 64°30′W, and consists of repeat biweekly hydrographic observations of temperature, salinity and dissolved oxygen (DO) conducted through the water column since 1954–12. The Bermuda Atlantic Time-series Study (BATS) site is located ~80 km southeast of Bermuda (31°40′N, 64°10′W)3. It consists of a comprehensive monthly sampling of the physics, chemistry and biology of the entire water column commencing in October 19884–7, and one of only a handful of ship-based biogeochemical time-series sites remaining in the global ocean7. The BATS–hydrostation S time-series represent the two longest records in the global open ocean of warming, salification, deoxygenation and changes in ocean carbon dioxide (CO2)–carbonate chemistry that drives ocean acidification (OA). Observations over the past 40 years in the subtropical gyre of the North Atlantic Ocean allow for examination of variability and the trends of ocean change which critically underpin future assessment and prediction. BATS–Hydrostation S time-series data also provide partial support for the conclusion, deoxygenation and changes in ocean carbon dioxide (CO2) since pre-industrial times estimated at 120–150 Pg C yr−1. The range of global ocean anthropogenic CO2 uptake presents, BATS), with detailed carbonate properties at the BATS site (1988–present) with earlier data (1983–1988, Hydrostation S; 1988–present, BATS), with detailed sampling protocols, analytical approaches and the computation of other parameters such as pH and pCO2 described in the “Methods” section. DIC and TA data are also normalised to a salinity of 36.6 (mean value in the Sargasso Sea) to remove the influence of evaporation and precipitation, and denoted as nDIC and nTA, respectively.

The suite of seawater CO2–carbonate parameters (i.e., fCO2, pCO2, pH, Ω_calcite and Ω_aragonite, Revelle factor) are determined for each sample using DIC and TA at situ temperature and salinity (see “Methods”). pH, Ω_calcite and Ω_aragonite in seawater are dimensionless and pH is expressed on the total scale.

Seasonally detrended anomalies of physical and chemical properties are shown to illustrate changes over time (Figs. 1 and 2) with trend analyses summarised in Table 1 for the past 40 years, and for the decades of the 1980s, 1990s, 2000s and 2010s. It is important to note that while surface data are shown and detailed here, seasonal variability and trends in the mixed layer are nearly identical to surface data. A tracer, TrOCA (Tracer combining Oxygen, inorganic Carbon and total Alkalinity)29 is used to test represents more than 450 cruises from October 1988 to the end of 2019. Sampling and analytical methods are detailed in the “Methods” section (Supplemental Materials). Measurements of seawater CO2–carbonate chemistry include those of dissolved inorganic carbon (DIC), and total alkalinity (TA) since 1983–1988, Hydrostation S; 1988–present, BATS), with detailed sampling protocols, analytical approaches and the computation of other parameters such as pH and pCO2 described in the “Methods” section. DIC and TA data are also normalised to a salinity of 36.6 (mean value in the Sargasso Sea) to remove the influence of evaporation and precipitation, and denoted as nDIC and nTA, respectively.

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Fig. 1 Hydrographic and seawater CO2–carbonate properties at the BATS site (1988–present) with earlier data (1983–1988) from Hydrostation S. Observed data (left vertical axis; open blue symbols) and anomalies (right vertical axis; orange symbols) are shown. Regression lines are from anomaly data plotted in the right vertical axis and given in Table 1. a Surface temperature and anomalies (°C). b Surface salinity and anomalies. c Surface dissolved oxygen (DO) and DO anomalies (µmol kg−1); the dashed line shows the trend from 1990. d Surface DIC and salinity normalised DIC (nDIC; µmol kg−1). e nDIC and CANT values (µmol kg−1).

Ocean sampling and methods

The Hydrostation S dataset represents more than 1381 cruises undertaken during the 1954–2020 period, while the BATS dataset
and deep water variability and trends (including warming and cooling, and freshening) is the subject of a future comprehensive treatment, with, for example, warming below the mixed layer at 300 m at −0.8 °C over the past three decades.

In the 1990s and 2000s, no significant trends in surface temperature and salinity were observed (Table 1a). In contrast, warming and salinification occurred during the 1980s and 2010s (Fig. 1a, b). In the 1980s, salinity increased by 0.27 (Table 1a) associated with the 1989–1990 transition from lower to higher salinity in the subtropical gyre (Fig. 3c, d). During the last decade (2010–2019), rapid warming of 1.18 °C and salinification of 0.14 has occurred. Temperature and salinity trends go back to 1954 (Table 1b). The warming of 1.18 °C in the last decade may at first appear inconsistent with overall warming of ~0.85 ± 0.12 °C over the past 40 years. The longer trend estimate since 1983 reflects periods of interannual/decadal warming and cooling (e.g., the decades of the 1990s and 2000s had no statistical change) with overall warming since the early 1980s.

Surface ocean deoxygenation. The upper-ocean record of DO shows an overall decline of ~17.8 ± 2.4 µmoles kg⁻¹ over the past 40 years (Table 1a; Fig. 1c), with short-term year-to-year variability (Fig. 3e, f). The longer-term data represents a loss of DO of ~8.1% or ~2% per decade. The trend of DO decline in the Sargasso Sea since the 1980s is in the range shown for global ODO rates of ~0.3 to ~0.7 µmoles kg⁻¹ year⁻¹ (~3–7 µmoles kg⁻¹ decade⁻¹) in the thermocline.

Apparent oxygen utilisation (AOU) represents the difference between measured oxygen concentration and oxygen concentration at saturation in seawater with the same physical and chemical properties. AOU, as a calculated parameter, offers a few clues as it reflects changes in oxygen due to the combined effect of biological processes such as primary production and respiration, but also diffusion, circulation, and ventilation. Here, the contribution of nitrification and denitrification to AOU is considered minor in surface waters. The trend in AOU in the upper 500 m is about −0.54 µmoles kg⁻¹ year⁻¹, which is slightly higher than the contemporaneous decrease in DO (Table 1a).

The cause for deoxygenation of the Sargasso Sea over the past 40 years appears only partly due to solubility changes associated with warming. Given the ocean warming observed in the Sargasso Sea (Table 1a), we estimate that the warming impact on DO solubility would likely have contributed to about 13% of the total decline of DO over the past nearly 40 years. The remaining deoxygenation (~87%) must result from the combined effect of changes in ocean biology and physics. Recent warming in the 2010s of 1.18 °C contributed about 18% of the concurrent decline in DO in that decade. From 1988 to 2017, open-ocean chlorophyll biomass (~22% per decade) and rates of in situ primary production observed offshore at BATS increased. Coccolithophore (i.e., calcifying phytoplankton) biomass has increased at BATS and over the entire subtropical gyre of the North Atlantic Ocean, but it is typically a small (<10%) contributor to primary production. Furthermore, low nitrate values in the mixed layer have doubled (i.e., an increase of ~0.25 µmoles kg⁻¹) over the past two and a half decades while suspended particulate organic matter and dissolved organic carbon have increased by ~30% and 2% per decade, respectively. Other studies indicate that the nutrient stream supporting production in the subtropical North Atlantic has reduced. These puzzles are not addressed here, but likely includes the loss of oxygen from the surface ocean due to gas flux, and perhaps supplies of new nitrogen sources from nitrogen fixation and dissolved organic nitrogen.

**Results**

**Surface ocean warming and salinification.** Menzel and Ryther first documented seasonal cycles of the local convective mixed layer with subsequent papers improving our understanding of the year-to-year and multi-decadal variability of the physical seasonality of the subtropical gyre and relationships to climate drivers. The Sargasso Sea exhibits a seasonal sea surface temperature variation of ~9–11 °C (Fig. 3a) with higher surface salinities occurring in wintertime and freshening during the summertime, and low salinity events occasionally apparent (Fig. 3c).

Over the past nearly 40 years (1983–2019), surface temperatures have increased by 0.85 ± 0.12 °C (Table 1a; Figs. 1a and 3b). The summer surface temperatures have increased at a higher rate than winter (i.e., 0.26 ± 0.01 °C year⁻¹ compared to 0.10 ± 0.01 °C year⁻¹; Table 1a; note that winter rates are not statistically significant). During the same period, surface salinity also increased by ~0.11 ± 0.02 (Table 1a; Fig. 1b). Importantly, these data show evidence of decadal variation in Table 1. Trend analysis of temperature, salinity, oxygen and ocean CO₂-carbonate chemistry from the BATS site (1988–2018; Hydrostation S data from 1983 to 1988) is included (see also Table 1b). Intermediate
Surface ocean TA and DIC changes. The observations near Bermuda provide the longest record of seawater CO$_2$-carbonate chemistry in the global open-ocean. Surface TA shows similar seasonal patterns to salinity, and a slight increase associated with salinity increases over time (Fig. 3c). However, salinity normalised alkalinity (nTA) shows marginally statistically significant changes over time (Table 1a; p-value of 0.03).

Surface DIC exhibits typical summer to wintertime seasonality of ~30–40 µmoles kg$^{-1}$ (Fig. 4a, b; salinity normalised DIC also shows a similar pattern but with reduced seasonality$^{13,36}$). Surface DIC and nDIC have significantly increased at rates of 1.26 ± 0.04 and 1.07 ± 0.03 µmoles kg$^{-1}$ year$^{-1}$, respectively, over the past 40 years (Fig. 1d; Table 1a). The difference between DIC and nDIC trends indicate that salinity changes contributed about 20% of the changes in DIC.

Earlier studies at BATS$^{36–40}$, elsewhere off Hawai’i$^{41}$ and the Canary Islands$^{42}$, attribute the increase in DIC to the uptake of anthropogenic CO$_2$ from the atmosphere$^2$. Here, we use the TrOCA method to determine if the remaining 80% of DIC increase is due to anthropogenic CO$_2$ uptake. The TrOCA method$^{29}$ has limitations in use for anthropogenic CO$_2$ quantification$^{43}$ but nevertheless, provides a useful tracer of ocean chemical changes when used at a fixed location. In the Sargasso Sea, TrOCA derived anthropogenic carbon concentration (C$_{ANT}$) surface values exhibit seasonal ranges of ~30–40 µmoles kg$^{-1}$ (Fig. 4c, d). Importantly, C$_{ANT}$ has increased by ~38 µmoles kg$^{-1}$ (+72%) over the past 30 years, with anthropogenic CO$_2$ concentration almost doubling in the upper ocean (Fig. 1d; Table 1a). The rate of increase of C$_{ANT}$ and nDIC are very similar (1.03 ± 0.05 vs. 1.07 ± 0.03 µmoles kg$^{-1}$ year$^{-1}$; Table 1a) and indicative that ocean uptake of anthropogenic CO$_2$ uptake has substantially changed ocean carbon chemistry over the past four decades at least.

Surface ocean fCO$_2$ and Revelle factor changes. Surface seawater fCO$_2$ also exhibits substantial seasonality with typical ranges of ~40–60 µatm (Figs. 2a and 4e, f). This variability reflects the underlying seasonality of temperature and DIC (similar trends
and patterns observed for $p$CO$_2$, or partial pressure of CO$_2$, with typically less than 1 µatm difference between fCO$_2$ and pCO$_2$. In response to anthropogenic CO$_2$ uptake by surface waters and an increase in DIC, surface seawater fCO$_2$ exhibited significant gain over the 40 years with fCO$_2$ anomalies increasing by ~76 µatm (Fig. 4f; Table 1a). The rate of change of fCO$_2$ is 1.92 ± 0.08 µatm year$^{-1}$ (Table 1a), representing an increase of nearly 25% from 1980 to 2020. The winter and summer fCO$_2$ values in recent years are higher and outside the range of surface fCO$_2$ seasonality observed in the 1980s.

The increasing trend in seawater fCO$_2$ observed in the North Atlantic Ocean is similar to observed changes in atmospheric fCO$_2$,$^{16,41}$ and, over shorter time-scales, similar trends in surface seawater fCO$_2$ have been reported elsewhere.$^{41,42}$ The observation that surface and atmospheric fCO$_2$ have tracked each other over time implies that the driving force for air–sea CO$_2$ gas exchange

**Fig. 3 Physical properties at BATS (1988–present) with earlier data (1983–1988) from Hydrostation S.**

- **a** Surface temperature (°C).
- **b** Surface temperature anomalies (°C).
- **c** Surface salinity.
- **d** Surface salinity anomalies.
- **e** Surface dissolved oxygen DO (µmoles kg$^{-1}$).
- **f** Surface dissolved oxygen DO anomalies (µmoles kg$^{-1}$). Please note that the occasional high and low summertime DO events observed in the 2000s occur both at hydrostation S and BATS indicating that these features are “real” surface phenomena. Dashed lines for contours are included.
The Revelle factor ($\beta$) reflects the underlying chemical state that can facilitate air–sea CO$_2$ exchange. In simple terms, the ocean’s ability to absorb CO$_2$ decreases with higher $\beta$ values and the chemical buffering capacity reduces. In the Sargasso Sea, the seasonality of $\beta$ is ~0.4, with wintertime values higher than summertime values. Lower $\beta$ conditions reflect the underlying capacity for seawater to absorb CO$_2$, and yet the lowest fCO$_2$...
occurs in winter coincide with seasonally higher $\beta$ values. This finding demonstrates the influence of other factors on $f_{CO_2}$, such as temperature. An important observation is that the Revelle factor, $\beta$, is increasing over time (~0.54 ± 0.04; Fig. 2b, Table 1a). These changes in ocean chemistry illustrate that the ocean capacity to absorb $CO_2$ from the atmosphere is gradually declining (~6% since 1980), which provides additional future feedback for assessments of the quantitative fate of anthropogenic $CO_2$ in the atmosphere and its transfer to the ocean.

Recently, Landschutzer et al. reported winter to summer $f_{CO_2}$ difference has increased from the 1980s to the 2010s by an average of 2.2 µatm per decade with a rate of 1.5 and 3.8 µatm per decade report for BATS and the Hawaii Ocean Time Series (but without details in the paper about how these rates were estimated). Such changes are attributed primarily to the long-term increase in the mean concentration of $CO_2$ in the surface ocean, $[CO_2]_{aq}$ and increase in the Revelle factor due to the reaction of added $CO_2$ with carbonate ion to reduce the capacity of the surface ocean $CO_2$–carbonate system to buffer against changes. At the BATS–Hydrostation S site, it is difficult to capture the peak maximum (summer) and minimum (winter) $f_{CO_2}$ with monthly shipboard observations (i.e., the maximum and minimum $f_{CO_2}$ may occur between cruises). Nonetheless, the mean winter to summer $f_{CO_2}$ difference increases from the 1980s to 2010s (e.g., mean and standard error; 1980s, 83.6 ± 17.2 µatm; 1990s, 87.9 ± 22.7 µatm; 2000s, 89.9 ± 14.7 µatm; 2010s, 99.2 ± 11.4 µatm; Table 1a) contemporaneously with increasing $[CO_2]_{aq}$ and Revelle factor. The increase in mean winter to summer $f_{CO_2}$ difference is 4.1 µatm per decade. Over the past nearly 40 years, the winter and summer $f_{CO_2}$ have increased at different rates of 1.62 ± 0.13 µatm, and 2.40 ± 0.19 µatm, respectively (Table 1a; total $f_{CO_2}$ change of ~65 µatm and ~96 µatm in winter and summer). The winter and summer $f_{CO_2}$ difference has increased by ~30 µatm which is ~7.9 µatm per decade. The two approaches thus indicate increases of winter and summer $f_{CO_2}$ difference in the range of 4.1–7.9 µatm per decade, and three to five times the rates reported by Landschutzer et al. (their data covers the period 1988–2014). The divergent winter and summer $f_{CO_2}$ conditions appear associated with reduction in winter length and lower rate of warming (the season with water less than 22 °C has reduced from 157.7 ± 10.8 days in the 1980s to 124.7 ± 24.7 days in the 2010s; 0.10 °C decade$^{-1}$; Table 1a), and increase in summer length and higher rate of warming (the season with water greater than 25 °C has reduced from 121.0 ± 23.4 days in the 1980s to 140.3 ± 11.3 days in the 2010s; 0.26 °C decade$^{-1}$; Table 1a). The BATS–Hydrostation S winter to summer $f_{CO_2}$ difference also substantially increases by ~10 µatm in the 2010s (due to summertime warming, Fig. 3a, and higher $f_{CO_2}$, Fig. 4e).

**OA indicators.** The BATS and Hydrostation S time-series data allow direct detection of the signal of OA in surface waters of the North Atlantic Ocean. $CO_2$ dissolved into seawater forms carbonic acid which rapidly dissociates into bicarbonate $[HCO_3^-]$ and in turn to carbonate $[CO_3^{2-}]$. Chemical equilibria reactions maintain mean contributions of $[HCO_3^-]$, $[CO_3^{2-}]$ and $[CO_2]_{aq}$ to DIC at ~88%, 11% and 1%, respectively. pH and $[CO_3^{2-}]$ decrease as seawater absorbs $CO_2$, a process termed OA. OA is potentially impactful for those organisms that secrete CaCO$_3$ skeletons, tests or shells and for marine ecosystems where calcification and pH controls on biogeochemical processes are important factors. The typical pH range of surface waters in the 1980s ranged from wintertime highs of ~8.2 to summertime lows of ~8.08–8.10 (Figs. 2c and 5a, b), with the ocean remaining mildly alkaline at present (~7.98–8.05). Concurrent with the changes in DIC, $C_{ANT}$, $f_{CO_2}$ and $\beta$ values, pH has also declined by approximately 0.07 from 1983 to 2020 (Fig. 2c; Table 1a).

The rate of pH change is ~0.0019 ± 0.0001 year$^{-1}$, which is a more negative rate than previously reported, and represents a 20% increase in hydrogen ion concentration since 1983. Both $\Omega_{calcite}$ and $\Omega_{aragonite}$ exhibit seasonal changes of ~0.3–0.5 (Fig. 5c, e). $\Omega_{calcite}$ and $\Omega_{aragonite}$ have decreased by 0.52 and 0.35, the past 40 years (Figs. 2d, e and 5d, f; Table 1a) at rates of 0.014 ± 0.001 year$^{-1}$ and 0.009 ± 0.001 year$^{-1}$, respectively.

**Present ocean carbon chemistry is now outside of 1980s values.** In forty years, seawater $CO_2$–carbonate chemistry conditions are now altered beyond the seasonal chemical ranges observed in the 1980s (Table 2). For DIC and $f_{CO_2}$, the low winter values in 2020 were the high summer values in the 1980s. Similarly, for pH, $\Omega_{calcite}$ and $\Omega_{aragonite}$, the high summer values are at present the low winter values. The statistically significant time-scale of emergence for carbonate parameters in the Sargasso Sea is typically less than 10 years (Table 2). The modification of seawater $CO_2$–carbonate chemistry will continue with future anthropogenic $CO_2$ emissions. Current chemical trends extended to 2050 indicate the following: Mean DO will be less than 200 µmoles kg$^{-1}$, mean DIC > 2100 µmoles kg$^{-1}$, and $f_{CO_2}$ and pH approaching 450 µatm and 8.000, respectively (Table 2). If current rates of anthropogenic $CO_2$ emissions continue until the end of the century, $f_{CO_2}$ will likely be at least 535 µatm, $C_{ANT}$ at >138 µmoles kg$^{-1}$ (nearly five times the 1983 concentration) and pH and $\Omega_{aragonite}$ close to 7.9 and below 3, respectively (Table 2). This end-of-century scenario is hypothetical given that $CO_2$ emissions are likely to peak and decline with future climate policy instruments, with the result that ocean carbonate chemistry is not expected to reach such levels.

**Discussion**

Decadal variability of sargasso sea ocean carbon sinks. The Sargasso Sea observations off Bermuda reveal the substantial decadal variations. As with temperature and salinity, the 1990s and 2000s were periods of modest changes with low rates of oxygen decrease. In the decade of the 1980s and 2010s, the decline of oxygen was about 8% (Table 1a), but with potentially different causes. In the 1980s, warming of ~0.26 °C contributed perhaps 5% to lower oxygen concentration. During the 2010s, oxygen concentration decreased by ~8%. Approximately, 18% of that change was due to warming and the influence of temperature on oxygen solubility. A combination of changes in gyre circulation, wind-driven ventilation and gas exchange may partly

| Year   | DO      | DIC     | $C_{ANT}$ | $f_{CO_2}$ | $\Omega_{calcite}$ | $\Omega_{aragonite}$ |
|--------|---------|---------|-----------|------------|---------------------|-----------------------|
| 1983   | 228     | 2031    | 25        | 310        | 6.18                | 3.86                  |
| 1990   | 212     | 2077    | 61        | 381        | 5.5                 | 3.62                  |
| 2000   | 199     | 2115    | 90        | 439        | 9.0                 | 3.34                  |
| 2050   | 177     | 2177    | 138       | 7.9        | 4.29                | 2.88                  |

Note: DO, DIC and C are expressed units of µmoles kg$^{-1}$, while $f_{CO_2}$ is expressed in units of µatm. The Revelle ($\beta$) factor, pH, $\Omega_{calcite}$ and $\Omega_{aragonite}$ are dimensionless and do not have units.
explain such decade to decade variation on DO loss. The slow-
down of Atlantic Meridional Overturning Circulation remains unproven at present, and it is challenging to link loss of oxygen in the subtropical gyre to basin-scale changes. The underlying dri-
vers for loss of oxygen in the subtropical gyre are the scope of future work.

An important additional finding from the BATS–Hydrostation S time-series is that although long-term trends of DIC result from anthropogenic CO2 uptake, there is a substantial decade to decade variation in changes. In the 1980s; associated with salini-
fication and an 8% DO decrease, both nDIC and nTA also declined by 11.8 and 7.3 µmoles kg\(^{-1}\), respectively (Table 1a). Prior studies of nDIC and nTA show that both decrease meridionally by 15–30 µmoles kg\(^{-1}\), from subpolar waters into the subtropical gyres of the Atlantic, Pacific and Indian Oceans\(^{49}\). The salini-
fication event at the end of the 1980s may mark a transition in North Atlantic subtropical gyre circulation and wind-driven ventilation, and perhaps a change to water masses with a more “southern” subtropical gyre physical and biogeochemical character (i.e., warmer, higher salinity and lower oxygen contents). If we assume

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**Fig. 5** Seawater CO2-carbonate properties at BATS (1988–present) with earlier data (1983–1988) from Hydrostation S. a Surface pH. b Surface pH anomalies. c Surface \(\Omega_{\text{calcite}}\). d Surface \(\Omega_{\text{calcite}}\) anomalies. e Surface \(\Omega_{\text{aragonite}}\). f Surface \(\Omega_{\text{aragonite}}\) anomalies. Dashed lines for contours are included.
an increase of DIC due to uptake of anthropogenic CO₂ (−9 µmoles kg⁻¹) for the decade of the 1980s in the Sargasso Sea, the nDIC content of a more “southern” water mass would likely have been ~20 µmoles kg⁻¹ lower.

Since the 1990s, the three decades have exhibited different DIC, nDIC, TA and nTA changes (Table 3). The largest increase in nDIC occurred in the 2000s. The finding of such significant decade to decade variation in seawater CO₂–carbonate chemistry has implications for determination of trends from shorter duration observations which likely have interannual to decadal change. Trend analysis of seawater CO₂–carbonate chemistry, as with other ocean physical and biogeochemical properties, thus requires long-term data.

Recently, DeVries et al.⁸ and McKinley et al.³⁰ reported substantial decadal variation in the global ocean carbon sink with climate variability a highly influential driver of ocean carbon uptake. In the 1990s, enhanced accumulation of CO₂ in the atmosphere relative to CO₂ emissions⁸ was accompanied by a period of reduced global ocean carbon uptake (~8 to ~28%) from beginning to end of the decade; Table 3. In contrast, in the 2000s, the atmospheric CO₂ growth rate slowed relative to higher anthropogenic CO₂ emissions⁸ while the global ocean carbon sink increased (~1.24 to +69%) from beginning to end of the decade; Table 3, with indications of a strengthening terrestrial carbon sink⁵. The Sargasso Sea C₅ANT data adds regional weight to these findings if we use C₅ANT as a proxy for ocean carbon uptake. In the 1990s, the rate of C₅ANT increase was lower by 16% (i.e., +0.87 ± 0.50 µmoles kg⁻¹ year⁻¹; Table 3), compared to the 1983–2020 change of +1.03 µmoles kg⁻¹ year⁻¹, indicative of lower anthropogenic CO₂ accumulation (and ocean uptake) during the decade. In contrast, in the 2000s the rate of C₅ANT increase was higher by 74% (i.e., +1.81 ± 0.53 µmoles kg⁻¹ year⁻¹; Table 3) were indicative of doubling of CO₂ accumulation (and ocean uptake) during the decade. Higher C₅ANT accumulation in the Sargasso Sea (i.e., +1.50 ± 0.58 µmoles kg⁻¹ year⁻¹; Table 3) may reflect the continuation of higher ocean carbon uptake in the 2010s from the 2000s.

The decade to decade variability in C₅ANT also covaries with rates of fCO₂ change and winter–summer fCO₂ difference. For example, the highest C₅ANT increase occurs in the 2000s along with higher fCO₂ increases and winter–summer fCO₂ difference (Table 3). This finding adds regional evidence to the study of Landschützer et al.⁴⁶ that winter–summer fCO₂ difference is increases. Following the methods of Landschützer et al.⁴⁶ that derive the thermal and non-thermal components that drive the differential winter and summer changes, the winter and summer DIC changes at BATS have a near identical influence on fCO₂. The thermal changes have a one-third and two thirds influence on fCO₂ in winter and summer, respectively. The non-thermal changes of fCO₂ are driven by an increase in nTA (0.29 µmoles kg⁻¹ year⁻¹) in winter and decrease in nTA in summer (−0.13 µmoles kg⁻¹ year⁻¹). At BATS, the differential changes in winter and summer temperature, and increase/decrease in alkalinity drive the 4–8 µatm per decade increase in winter–summer fCO₂ difference over the past forty years. In contrast, Landschützer et al.⁴⁶ reported a change of 1.5 µatm per decade, but critically, ignored the effect of alkalinity changes in their analysis. Here, we demonstrate that observed changes in alkalinity do make a substantial difference for evaluating changes in winter–summer fCO₂ difference over time and the need to incorporate TA for global synthesis⁶⁶.

Nearly, 40 years of data at BATS–hydrostation S also indicate that the ocean is not uniformly changing over time, and there is a substantial decade to decade variation in CO₂–carbonate chemistry. And that the ocean carbon sink is not stable over recent time and varies from decade to decade. If the coupling of atmospheric CO₂ accumulation, ocean carbon sink and climate variability⁸,³⁰ is proven, the Sargasso Sea appears to act in concert with global decade to decade changes in ocean carbon sinks.

In summary, these data reveal recent warming in the last decade, associated with salinification and substantial deoxygenation. These physical changes were accompanied with significant increases of DIC and fCO₂, and 20–40% decreases in pH, Ω calcite and Ω aragonite, respectively, since the 1980s. Apparent reduced ocean carbon uptake in the decades of the 1990s and higher CO₂ uptake 2000s/2010s in the Sargasso Sea agree with the findings of decadal variability in “weaker”/”stronger” global ocean carbon uptake⁶,³⁴ and points to reduced ocean uptake in the Sargasso Sea in the 2010s. Such data on ocean physics and chemistry provide critically needed and unparalleled observational tests of coupled ocean–atmosphere models and future refinement of drivers for a model-data decade to decade assessment of the global carbon cycle.

Methods

Sampling methods. Ocean sampling at hydrostation S and BATS started in 1954 and 1988, respectively. Hydrostation S began with the pioneering efforts of Hank Stommel (Woods Hole Oceanographic Institution) and colleagues⁶⁸ at a site approximately 26 km southeast of Bermuda (32° 10’N, 64° 30’W, Fig. 1). The first

| Table 3 Comparison of atmospheric CO₂ increase, global ocean carbon sinks and Sargasso Sea carbon sink for recent decades. |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|
| **Change over period**          | 1980s           | 1990s           | 2000s           | 2010s           |
| Atmospheric CO₂ increase        | [De Vries et al.]⁸ | n/a             | Higher³        | Lower²          | n/a             |
| Global ocean carbon sinks       | [De Vries et al.]³⁰ | n/a             | Weak (−8 to −16%) | Strong (+35 up to +43%) | n/a             |
| Sargasso Sea carbon sink        | [This study: CANT] | n/a             | Weak (−17 to −28%) | Strong (+24 to +69%) | n/a             |
| [CANT increase]                 | −1.94 ± 1.23     | +0.19 ± 0.08    | +2.06 ± 0.08    | +3.21 ± 0.08    | +1.84 ± 0.08    |
| [CO₂ increase]                  | 83.5 ± 17.2      | 87.9 ± 22.7     | 89.9 ± 14.7     | 99.9 ± 30.5     |
| [Winter-summer (CO₂)]          |                 |                 |                 |                 |

Note: the change is given in units of µmoles kg⁻¹ year⁻¹, while CO₂ increase is µatm year⁻¹. The winter–summer fCO₂ difference is given units of µatm. The accumulation of CO₂ in the atmosphere relative to emissions is higher in the 1990s compared to the 2000s. In Table 3, the designation of ‘weak’ and ‘strong’ global ocean uptake is taken from DeVries et al.⁸ and McKinley et al.³⁰, and represents the change in ocean sink between the beginning and end of the decade, and the range determined from the various approaches used in each of their studies. The change in C₅ANT in the anthropogenic CO₂ relative to the long-term increase of ~103 ± 0.05 µmoles kg⁻¹. Positive increases denoted in % indicate that the C₅ANT has increased at a higher rate relative to the long-term trend and thus indicate increase ocean uptake of anthropogenic CO₂.
water–column sampling occurred on the 7th June 1954 from the 61° R. V. Panularis with more than 1381 cruises conducted up to the present time from the R. V. Panularis (1986–1989); R. V. Weatherbird II (1989–2006) and R. V. Atlantic Explorer (2006–present).

Since the arrival of the R. V. Weatherbird, Hydrostation S has been occupied at near biweekly intervals, with multiple CTD–hydrocasts through the water-column to ~2600 m, and to 4500 m at BATS (more than 450 cruises to the site). Nansen bottles were used for water sampling at first, then 5 L Niskin bottles were used and additionally, they are also compared with the downcast CTD profiling. In contrast, the CTD sampling system has predominately been a Seabird 24-place rosette using 12 L Ocean Test bottles. Before profiling, the CTD is allowed to stabilise at 10 m and once stable, the CTD returns to the surface to start the profile with typical descent rates of 0.5–1.0 m s⁻¹, depending on weather conditions.

Water samples are collected on the upcast, whereby the OTE bottles are closed at the interface box and PC software. The salinometer drift during and between conductivity, as well as dissociation constants for KHSO₄ and the chemical description of alkalinity of seawater (TA) is defined as TA = [HCO₃⁻] + [CO₂(aq)] + [H₂CO₃] + [OH⁻] + [H₂PO₄⁻] + 2[HPO₄²⁻] + [SiO(OH)₃][equation S1].

In this second chemical definition, the summed concentrations of HCO₃⁻ + 2 CO₂ (aq) + [B(OH)₄⁻] represent the major chemical components of TA and the potential balance of seawater. Other chemical components to alkalinity are typically considered minor constituents in seawater44–46. Given DIC and TA are both expressed as µmoles kg⁻¹ following long-established guidelines (Dickson et al., 200742 and references therein).

Initially, at hydrostation S, samples for DIC and TA were collected into 1 l Pyrex bottles with a change to 500 ml bottles in the early 1990s, poisoned with HgCl₂ sealed with ground glass stoppers and then shipped to Scripps Institution of Oceanography (SIO) for analysis56,1973–1988). Storage time for samples before analysis ranged from a few months to several years. Similar sampling protocols were established at BIOS for sampling at the BATS54–57, but in the early 2000s, smaller Pyrex bottles (350 ml) were used. Samples for DIC and TA have been typically analysed within a few months of collection at BIOS since 1988.

The sampling frequency of the combined dataset from hydrostation S and BATS was not uniform in time. In the 1980s, samples were collected 9–12 times a year, while since 1992, sampling increased to 14–15 times a year. The increase in sampling since the middle 1990s was due to supplemental BATS bloom cruises (1–4 in number) conducted during the January to April period in addition to BATS core cruises. The increase in sampling frequency over time weights the time-series to springtime when determining non-seasonally aliased trend40.

Potentiometric titration methods were also used for determination of TA at BIOS. At the beginning of the 1990s, a manual alkalinity titrator was used for determination of TA at BIOS. This was replaced by an automated VINDTA 2S (Versatile Instrument for the Determination of Titration Alkalinity) in the early 2000s40. For both manual and automated TA systems, 15–20 titration points past the carbonic acid endpoint were determined for each sample, with TA computed from the concentration data using nonlinear algebraic methods. Determination of seawater composition and alkalinity of seawater in the Sargasso Sea were also analysed each day before sample analyses, and certified reference material (CRMs) were used routinely to calibrate the TA measurements at BIOS. At BIOS, DIC was determined using coulometric methods with a SOMMA system24. During the first 2 years of sampling, DIC samples were analysed in WHOI (e.g., R.V. Marine cruise 1–21), and subsequently at BIOS. DIC measurements were calibrated with known volumes of pure CO₂ gas while CRM's were routinely analysed each day of analysis from 1991. Hydrostation S samples were analysed for DIC at SIO from 1983 to 1988 using manometric methods66. Potentiometric titration methods were used for determination of TA at SIO, and after that at BIOS. No significant bias has been noted between these earlier methods (also analysed by A.G. Dickson to present)60. Analytical precision for DIC and TA at BIOS was typically ≤0.03% and ≤0.05%, respectively for within the bottle and between bottle replicate analyses of more than 5,000 samples. Analytical accuracy for both DIC and TA using CRM analyses is better than 0.1%.

Seawater CO₂-carbonate chemistry computations and errors. The suite of seawater CO₂-carbonate parameters (i.e., [CO₂], [H₂CO₃], [H₂CO₃], [CO₂⁻], [H⁺]) for each sample can be determined from the measurement of any two parameters (i.e., DIC, TA, pCO₂, pCO₂ and pH), along with temperature and salinity52,54,55. SeaCO₂ (fugacity of CO₂) is used here rather than the partial pressure of CO₂ (i.e., pCO₂), since this parameter was also computed in data files. The units of µmoles kg⁻¹ in seawater are dimensionless and pH expressed on the total scale53. All parameters are computed in situ temperature and salinity.

Here, seawater CO₂, pH, [CO₂⁻], [H⁺], mineral saturation states for Ωcalcite and Ωaragonite, the Revelle factor (β) were computed from DIC, TA, temperature and salinity data using the program CO2sys. Calcium carbonate compensation depths (i.e., pKᵢ and pKᵢ), as refit by Dickson and Millero (1987)62 were used for the computation, as well as dissociation constants for KHCO₃, H₂CO₃, and H₂CO₃.
The computation error estimates for calculation of $pCO_2$, pH, $\Omega_{aragonite}$ and $\Omega_{calcite}$ were undertaken using standard procedures for the propagation of uncertainty\(^1\). Given the analytical uncertainty of the two seawater carbonate chemistry parameters (i.e., DIC and TA; ± 1 mmol kg\(^{-1}\)) from replicate measurements of CRM’s, the error of the computation is estimated at 3 μmol, 0.003, 0.018, and 0.012, for $pCO_2$, pH, and $\Omega_{calcite}$ and $\Omega_{aragonite}$ for surface waters, respectively\(^2\).

**Use of the trace TrOCA.** TrOCA and TA is a method first proposed by Touratier and Goyet (2004a, b\(^3\)) and, and updated by Touratier et al. (2007)\(^4\) to quantify anthropogenic carbon (i.e., $C_{ANT}$) in seawater. The quasi-conservative tracer, TrOCA in water masses accounts for changes in carbonate chemistry due to biological influences (e.g., remineralization of organic matter) and abiotic processes (e.g., dissolution of CaCO\(_3\)), as such, is also useful for following ocean chemistry changes in the surface ocean. Here, the formulation proposed by Touratier et al. (2007)\(^4\); their equation 11) is used as a water tracer: $C_{ANT} = ([O_2] + 1.279 \times (DIC – TA/2) – EXP (7.511 – 1.087 \times 10^{-2} \times \Phi – 7.81 \times 10^{-3} \times \Phi^2))$ (equation S3) where $\Phi$ is the potential temperature. The uncertainty of the estimate is ~6 mmol kg\(^{-1}\)\(^5\). $C_{ANT}$ estimates determined using the TrOCA method for the mixed layer are modified by euphotic zone biological activity and air–sea gas exchange\(^6\), but, over time, provide additional evidence for changes in ocean chemistry.

**Trend analyses.** Trend analyses were conducted of the time-series of surface temperature and salinity, seawater carbonate chemistry (DIC, TA, $pCO_2$ and Revelle factor) and OA indicators (pH, $[CO_3^{2-}]$, $\Omega_{aragonite}$ and $\Omega_{calcite}$). Here, trend analyses were performed using linear regression methods with the number of samples and bottle data with both dataset available to the end of 2019. The surface BATS and Hydrostation S data sampled on a monthly and twice-monthly basis, respectively, since October 1988. Trend analysis for the period from 1983 to 2020, and for each decade (i.e., the 1980s, 1990s, 2000s, 2010s) was determined using linear regression methods with recent global warming. Annu. Rev. Mar. Sci. 768 (2015).

1. Menzel, D. W. & Ryther, J. H. The annual cycle of primary production in the Sargasso Sea off Bermuda. Deep Sea Res. 6, 351–367 (1960).
2. Phillips, H. E. & Joyce, T. M. Bermuda’s tale of two time series: Hydrostation ‘S’ and BATS. J. Phys. Oceanogr. 37, 554–571 (2007).
3. Michaels, A. F. & Knap, A. H. Overview of the U.S. JGOFS Bermuda Atlantic Time-series study and the hydrostation S program. Deep Sea Res. II 43, 157–198 (1996).
4. Steinberg, D. K. et al. Overview of the U.S JGOFS Bermuda Atlantic Time-series Study (BATS): a decade-scale look at ocean biology and biogeochemistry. Deep Sea Res. II 48, 1405–1447 (2001).
5. Gruber, N., Bates, N. R. & Keeling, C. D. Interannual variability in the North Atlantic Ocean carbon sink. Science 298, 2374–2378, (2002).
6. Longhurst, A., Long, M. C. & Rounsevell, M. D. P. Two decades and counting: 24 years of sustained open ocean biogeochemical measurements in the Sargasso Sea. Deep Sea Res. II 93, 16–32 (2013).
7. Bates, N. R. et al. Changing ocean chemistry: a time-series view of ocean uptake of anthropogenic CO2 and ocean acidification. Oceanography 27, 121–146 (2014).
8. DeVries, T. et al. Decadal trends in the ocean carbon sink. Proc. Natl Acad. Sci. USA 116, 11646–11651 (2019).
9. Le Quéré, C. et al. (2017). Global Carbon Budget 2017. Earth Syst. Sci. Data https://doi.org/10.5194/essd-2017-123 (2017).
10. Friedlingstein, P. et al. Global carbon budget, 2019. Earth Syst. Sci. Data 11, 1399–1438 (2019).
11. Takahashi, T. et al. Climatological mean and decadal change in surface ocean $pCO_2$ and net-sea–air CO2 flux over the global oceans. Deep Sea Res. II 56, 554–577 (2009).
12. Sabine, C. L. et al. The oceanic sink for anthropogenic CO2. Science 305, 367–371 (2004).
13. Khatiwala, S. et al. Global ocean storage of anthropogenic CO2. Biogeosciences 10, 2169–2191 (2013).
14. Gruber, N. et al. The oceanic sink for anthropogenic CO2; from 1994 to 2007. Science 363, 1193–1199 (2019).
15. Caldeira, K. & Wickett, M. F. Anthropogenic carbon and ocean pH. Nature 435, 325–330 (2005).
16. Doney, S. C., Bopp, L. & Long, M. C. Historical and future trends in ocean climate and biogeochemistry. Oceanography 27, 108–119 (2014).
17. Keeling, R. & Garcia, H. E. The change in oceanic $C_{ANT}$ inventory associated with recent global warming. Proc. Natl Acad. Sci. USA 99, 7848–7853 (2002).
18. Keeling, R. F., Körtzinger, A. & Gruber, N. Ocean deoxygenation in a warming world. Annu. Rev. Mar. Sci. 2, 199–229 (2010).
19. Stramma, L. and Schmidtko, S. Global evidence of deoxygenation. In Ocean Deoxygenation: Everyone’s Problem. (eds Laffoley, D., & Baxter, J. M.) 25–36 (IUCN, 2019).
20. Gruber, N. Warming up, turning sour, losing breath: ocean biogeochemistry under global change. Philos. Trans. R. Soc. Ser. A 369, 1980–1996 (2011).
21. Doney, S. C., Bopp, L. & Long, M. C. Historical and future trends in ocean climate and biogeochemistry. Oceanography 27, 108–119 (2014).
22. García, H. E. & Gordon, L. I. Oxygen solubility in seawater: better fitting equations. Limnol. Oceanogr. 37, 1307–1312 (1992).
23. Levin, L. A. & Le Bris, N. The deep ocean under climate change. Science 350, 766–768 (2015).
24. Lemburg, C., et al. Temperature dependence of prokaryotic production. Front. Mar. Sci. https://doi.org/10.3389/fmars.2016.00090 (2016).
25. Bednaršek, N., Harvey, C. J., Kaplan, I. C., Feely, R. A. & Mőzsa, J. Pteropods on the edge: Cumulative effects of ocean acidification, warming and deoxygenation. Proc. Oceanogr. 145, 1–24 (2016).
26. Stramma, L., Schmidtko, S., Levin, L. A. & Johnson, G. C. Oxygen minimum expansions and their biological impacts. Deep Sea Res. Part I 57, 587–595 (2010).
27. Rabalais, N. N. et al. Eutrophication-driven deoxygenation in the coastal ocean. Oceanography 27, 172–183 (2014).
28. Duarte, C. M. et al. Reconsidering ocean calorimetrics. Bioscience 65, 130–139 (2015).
29. Touratier, F., Azoouli, L. & Goyet, C. CFC-11, 14\(^{C}\) and 3\(^{He}\)tracers as a means to assess anthropogenic CO2 concentrations in the ocean. Tellus 59B, 318–325 (2007).
30. McKinley, G. A., Fay, A. R., Edeebbar, Y. A., Gloege, L. and Lovenduski, N. S. External forcing explains recent decadal variability of the ocean carbon sink. AGU Adv. https://doi.org/10.1002/essoar.10501723.2 (2020).
31. Stendardo, I. & Gruber, N. Oxygen trends over five decades in the North Atlantic. J. Geophys. Res. 117, C11004 (2012).
32. Montes, E. Decadal variability in the oxygen inventory of North Atlantic subtropical underwater captured by sustained, long-term oceanographic time series observations. Glob. Biogeochem. Cycles 30, 460–478 (2016).
33. Bates, N. R. Twenty years of marine carbon cycle observations at Devils Hole Bermuda provides insight into seasonal hypoxia, coral reef calcification and
ocean acidification. Front. Mar. Sci. 4, https://doi.org/10.3389/fmars.2017.00036 (2017).
34. Krumhardt, K. M., Lovenduski, N. S., Freeman, N. M. & Bates, N. R. Increasing coccolithophore abundance in the subtropical North Atlantic from 1994 to 2014. Biogeosciences 13, 1163–1177 (2016).
35. Tagkis, F., Ito, T. & Bracco, A. Modulation of the North Atlantic deexygenation by the slowdown of the nutrient stream. Biogeosciences 17, 231–244 (2020).
36. Bates, N. R. Interannual variability of oceanic CO2 and biogeochemical properties in the Western North Atlantic subtropical gyre. Deep Sea Res. II 48, 1507–1528 (2001).
37. Bates, N. R. Interannual variability of the oceanic CO2 sink in the subtropical gyre of the North Atlantic Ocean over the last two decades. J. Geophys. Res. (Oceans) 112, C07013 (2007).
38. Bates, N. R. & Peters, A. J. The contribution of atmospheric acid deposition to ocean acidification in the subtropical North Atlantic Ocean. Mar. Chem. 107, 547–558 (2007).
39. Bates, N. R., Pequignet, A. C., Johnson, R. J. & Gruber, N. A short-term sink for atmospheric CO2 in subtropical mode water of the North Atlantic Ocean. Nature 420, 489–493 (2002).
40. Bates, N. R. et al. Indicators of anthropogenic carbon dioxide uptake and ocean acidification in the North Atlantic Ocean. Biogeosciences 9, 2509–2532 (2012).
41. Dore, J. E., Lukas, R., Sadler, D. W., Church, M. J. & Karl, D. M. Physical and biogeochemical modulation of ocean acidification in the central North Pacific. Prog. Ocean. Sci. 106, 1235–1240 (2009).
42. González-Dávila, M., Santana-Casiano, J. M., Rueda, M. J. & Llinas, O. The cation in the North Atlantic Ocean. Biogeosciences 17, 727–751 (2020).
43. Robbins, L. L., Hansen, M. E., Kleypas, J. A., and Meylan, S. C., (2010). CO2calc: a user-friendly seawater carbon calculator for Windows, Max OS X, and iOS (iPhone). U.S. Geological Survey Open-File Report, 2010–1280, 1–17, http://pubs.usgs.gov/of/2010/1280/, last access date 15 October 2018, 2010.
44. Mehrbach, C., Culberson, C. H., Hawley, J. E. & Pytkowicz, R. M. Measurement of the apparent dissociation constants of carbonic acid in seawater at atmospheric pressure. Limnol. Oceanogr. 18, 897–907 (1973).
45. Dickson, A. G. & Millero, F. J. A comparison of the equilibrium constants for the dissociation of carbonic acid in seawater. Deep Sea Res. Part A 34, 1733–1743 (1987).
46. Dickson, A. G. Standard potential of the reaction: AgCl(s)+12H2(g) = Ag(s) + HCl(g) and the standard acidity constant of the ion HSO4- in synthetic seawater from 273.15 to 318.15 K. J. Chem. Thermodyn. 22, 113–127 (1990).
47. Fabry, V. J., Seibel, B. A., Feely, R. A. & Orr, J. C. Impacts of ocean CO2 on marine fauna and ecosystem processes. Biogeosciences 15, 3472–3486 (2018).
48. Henson, S. A. et al. Rapid emergence of climate change in environmental model simulations off Bermuda? Geophys. Res. Lett. 40, 3081 (2013).
49. Knap, A. H. et al. BATS Methods Manual, Version 4 Woods Hole, MA, US. U. S. JGOFS Planning Of S. JGOFS Planning Of, 136 pp (1997).
50. Schroeder, E. & Stommel, H. How representative is the series of Panulirus stations off Bermuda? ICES J. Mar. Sci. 65, 103–107 (2008).
51. Knap, A. H. et al. BATS Methods Manual, Version 4 Woods Hole, MA, US. U. S. JGOFS Planning Of, 136 pp (1997).
52. Strickland, J. D. H., and Parsons, T. R. A Practical Handbook of Seawater Analysis. (Fisheries Board, Canada, Ottawa, 1968).
53. Zeebe, R. E. and Wolf-Gladrow, D. CO2 in Seawater: Equilibrium, Kinetics, isotopes. (Elsevier Oceanography Series, 2001).
54. Dickson, A. G., Sabine, C. L., and Christian, J. R. Guide to Best Practices for Ocean CO2 Measurements. (North Pacific Marine Science Organization, Sidney, British Columbia, 2007) PICES Special Publication 3.
55. Miller, F. J. Chemical Oceanography. Fourth Edition. 552 (CRC Press, 2013).
56. Keeling, C. D. Surface ocean CO2 in: Global Carbon Cycle, (ed Heimann, M.) 22–29 (Kluwer Publishers, 1993).
57. Brix, H., Gruber, N. & Keeling, C. D. Interannual variability of the upper carbon cycle at station ALOHA near Hawaii. Glob. Biogeochem. Cycles 18, GB4019 (2004).
58. Bates, N. R., Michaels, A. F. & Knap, A. H. Seasonal and interannual variability of oceanic carbon dioxide species at the US JGOFS Bermuda Atlantic Time-series Study (BATS) site. Deep Sea Res. Part II 43, 347–383 (1996).
59. Bates, N. R., Michaels, A. F. & Knap, A. H. Alkalinity changes in the Sargasso Sea: geochemical evidence of calcification? Mar. Chem. 51, 347–358 (1996).