Decadal acidification in the water masses of the Atlantic Ocean

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Global ocean acidification is caused primarily by the ocean’s uptake of CO2 as a consequence of increasing atmospheric CO2 levels. We present observations of the oceanic decrease in pH at the basin scale (50°S–36°N) for the Atlantic Ocean over two decades (1993–2013). Changes in pH associated with the uptake of anthropogenic CO2 (ΔpHCant) and with variations caused by biological activity and ocean circulation (ΔpHNat) are evaluated for different water masses. Output from an Institut Pierre Simon Laplace climate model is used to place the results into a longer-term perspective and to elucidate the mechanisms responsible for pH change. The largest decreases in pH (ΔpH) were observed in central, mode, and intermediate waters, with a maximum ΔpH value in South Atlantic Central Waters of −0.042 ± 0.003. The ΔpH trended toward zero in deep and bottom waters. Observations and model results show that pH changes generally are dominated by the anthropogenic component, which accounts for rates between −0.0015 and −0.0020/y in the central waters. The anthropogenic and natural components are of the same order of magnitude and reinforce one another in mode and intermediate waters over the time period. Large negative ΔpHNat values observed in mode and intermediate waters are driven primarily by changes in CO2 content and are consistent with (i) a poleward shift of the formation region during the positive phase of the Southern Annular Mode in the South Atlantic and (ii) an increase in the rate of the water mass formation in the North Atlantic.

Significance

We provide the first (to our knowledge) observation-based acidification trends in the water masses of the Atlantic basin over the past two decades and compare them with climate model results. Observations and model output confirm that pH changes in surface layers are dominated by the anthropogenic component. In mode and intermediate waters, the anthropogenic and natural components are of the same order of magnitude and sign. Large changes in the natural component of newly formed mode and intermediate waters are associated with latitudinal shifts of these water masses caused by the Southern Annular Mode in the South Atlantic and by changes in the rates of water mass formation in the North Atlantic.


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Central Water; SAMW, Subantarctic Mode Water. The lowercase first letters Central Water; NADW, North Atlantic Deep Water; SACW, South Atlantic Intermediate Water; CDW, Circumpolar Deep Water; NACW, North Atlantic (36°N). Water mass acronyms: AABW, Antarctic Bottom Water; AAIW, Antarctic masses. Ocean Data View was used for mapping.

Fig. 1. (A, Left) Tracks of the OACES/CO2 (1993), CITHER-II (1994), and FICARAM-XV (2013) cruises used in this study. (Right) The vertical distribution of salinity along the FICARAM-XV cruise in 2013. (B, Left) IPSL model profile. (Right) The vertical distribution of salinity of the model. The horizontal white lines are along the FICARAM-XV cruise in 2013. (Fig. S2, Appendix). The greatest acidification was detected in the southern region from the surface to depths greater than 1,000 m. In this area, the average ΔpH reached −0.042 in the SACW, −0.037 in the SAMW, and −0.029 in the AAIW. The northern region showed relatively strong acidification with average ΔpH values of −0.035 in the uNACW, −0.040 in the lower North Atlantic Central Water (INACW), and −0.012 in the AAIW. Acidification was much weaker in the upper water column in the equatorial region. For example, ΔpH in the uCDW–uNADW layer was about two times higher in the southern and northern regions than in the equatorial region (−0.010 versus −0.005). Deeper in the water column, ΔpH values were similar along the transect, ranging from −0.006 to −0.007 in the lower Circumpolar Deep Water (ICDW)–lower North Atlantic Deep Water (INADW) layer and up to −0.008 in the AABW layer.

The ΔpH was separated into changes associated with the increase in Cant in the ocean (ΔpHCant) and those associated with a modification in total inorganic carbon (CT) resulting from the remineralization of organic matter and changes in ocean circulation (ΔpHNat). To separate the anthropogenic and natural components of ΔpH, a pH value (pH2013-ant) was computed for 2013 from thermodynamic equations that included total alkalinity (AT) and the nonanthropogenic changes in CT (ΔCant). This computation yielded the anthropogenic CT component of the pH change (ΔpHCant = pH2013 − pH2013-ant), where pH2013 is the measured pH. The natural component (ΔpHNat) equaled ΔpH − ΔpHCant over the 20-y timespan. Cant concentrations were determined by the carbon-based pCT method (21, 22), with an SD in the changes in Cant (ΔCant) of ±2.7 μmol/kg. This SD translates into an uncertainty of 0.005 in ΔpHCant and ΔpHNat (SI Appendix, SI Text). To avoid biases in ΔpHNat caused by water mass mixing, the ΔpHNat was corrected for these effects using changes in potential temperature (ΔΘ) in the water masses from 1993 to 2013 (SI Appendix, Table S3).

Based on this analysis, the uptake of Cant by the ocean has been the main contributor to ΔpH over the last two decades (Table 1). As expected, ΔpHCant and ΔCant distributions showed similar patterns (Fig. 3 and D), with the highest changes located in the upper layer in contact with the atmosphere. There is a clear south-to-north gradient, with ΔpHCant values in the southern region being about twice as large as in the equatorial and northern regions. In the surface layers, ΔpHCant reached values up to −0.039 in the southern region (SACW) and −0.030 in the equatorial and northern regions (SACW–uNACW), coinciding with the highest ΔCant values of 21 and 15 μmol/kg, respectively (Table 1). In the subsurface layer (SAMW–INACW), ΔpHCant also showed higher absolute average values in the southern region (−0.033) than in the equatorial and northern regions (0.016), in agreement with the ΔCant values of 15 and 6 μmol/kg, respectively (Fig. 3 and D and Table 1). Deeper in the ocean, ΔpHCant values dropped from around −0.018 (ΔCant ~7 μmol/kg) in the southern AAIW layer to between −0.003 and −0.010 (ΔCant ~1–4 μmol/kg) in the CDW and AABW layers. ΔpHCant and ΔCant were strongly correlated because ΔpHCant is caused directly by anthropogenic CO2 entering the ocean and because ΔpHCant is computed from ΔCant (see above). Determining ΔCant by an independent method [transit time distribution (TTD)] in which no CO2 measurements were used (23) yielded the same results (SI Appendix, Fig. S2).

Fig. 2. Vertical distributions of (A) pH at 25 °C on the seawater scale (SWS) along 2013 and (B) pHWS changes (ΔpH) between 2013 and 1993–94. Ocean Data View was used for mapping. Note that the depth scale is not linear.
Average values and SDs for each layer and region of pH change (ΔpH); the anthropogenic component of ΔpH (ΔpHCant); the natural component of ΔpH (ΔpHNat); the anthropogenic CO₂ change estimated by the fcC_8 method (Δ Cant) and by the TTD method (Δ Cant-TTD); the change in nitrate (ΔNO₃) between 1993–94 and 2013 from observations; and ΔpHCant and ΔpHNat (= 3.92×std(pHNat)) from the IPSL model. Italic indicates values that are not statistically significant (Student’s t test <0.05). Column N indicates the number of values within each layer. See Fig. 1 for water mass acronyms.

ΔpHNat includes changes in pH related to changes in ocean circulation and/or biological activity, which are driven by natural variability and climate change. The pH changes associated with climate change were estimated to be less than 0.0012/yr, i.e., <0.002 over the past two decades (9). We corrected ΔpHCant for mixing between the water masses by using the changes in potential temperature (Δθ) (SI Appendix, Table S3). ΔpHNat, therefore, primarily captured the natural interannual and decadal variability of the ocean’s circulation and carbon cycle. This interannual-to-decadal variability was particularly pronounced in the higher latitudes of the North Atlantic (12, 24) and South Atlantic (25–28). ΔpHNat was generally lower than ΔpHCant, except in mode and intermediate waters (i.e., SAMW, INACW, and AAIW), where ΔpHNat and ΔpHCant had similar magnitudes with rates up to −0.0010 ± 0.0002/yr (Fig. 4A and Table 1). ΔpHNat patterns closely matched the patterns of changes in nitrate (ΔNO₃) corrected for mixing using Δθ (Fig. 3 B and E). The correlation coefficient (r²) obtained between ΔpHNat and ΔNO₃ for all the water masses was 0.63, with a slope of −0.012 ± 0.002 and P < 0.01 (Fig. 4B). The relationship between ΔpHCant and ΔNO₃ is attributed to remineralization. The slope indicates that 0.012 units of pH change correspond to 1 μmol of NO₃. Thus a change of 0.001 units of pH is equivalent to 0.52 μmol of CO₂, from which a stoichiometric NC ratio of 6.2, close to the Redfield ratio of 6.6, was obtained (29). Central and mode waters showed the largest changes in ΔpHNat (+0.030) and ΔNO₃ (+3 μmol/kg), findings that are in agreement with the spatial and temporal variability of the ocean’s circulation and biology being stronger in the upper waters (Fig. 3 B and E). The intermediate water class with the largest change was the AA IW, with ΔpHNat up to −0.013 and ΔNO₃ up to 0.32 μmol/kg. The smallest changes occurred in the deep and bottom waters (+0.005 pH units and changes in NO₃ of ±0.5 μmol/kg) (Fig. 3 B and E and Table 1).
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by the European Station for Time Series in the Ocean in the North
similar to rates previously reported in the South Atlantic (30) and
ventilation pathways, namely, mode waters with rates up to
acidification rates also were observed in the water masses along the
winters, thus enabling a larger uptake of atmospheric CO2
mode waters have a larger exposure to the atmosphere, with a
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masses in the northeastern region of the section are expected to be
older than in the southwestern region and to have lower ΔCant
values. However, another process probably explains why ΔCant
and ΔH Cant were weaker in the North Atlantic. In the subpolar
region of the North Atlantic, a weakening in the formation of
Labrador Sea Water led to a decrease in the relative Cant
inventory of North Atlantic Deep Water (NADW) from 1997 to
2003 (32) and to a smaller change in ΔH Cant. In contrast, a
larger increase in the Cant inventory was found in the South
Atlantic compared with the North Atlantic from 1989 to 2005 (33),
possibly related to the Southern Annular Mode (SAM) affecting
the uptake of Cant in the South Atlantic and to the North Atlantic
Oscillation shift impacting ΔH Cant in the North Atlantic (34).

Our observations show that the anthropogenic and natural
components had a similar magnitude in the MW–IW (Fig. 4A),
indicating that changes in ocean circulation and biological
activity contributed significantly to pH variability in the subsurface
waters of the Atlantic Ocean over this time period. The
observations were placed in the context of changes occurring in a
larger space and time domain by comparing them with pH
changes from the IPSL Earth System Model. This model
objectively assessed the separation of ΔpH into ΔH Cant and
ΔHNat. The distribution of ΔH Cant in IPSL, referred to as
“ΔH Cant–IPSL,” followed the distribution obtained from the
observations, with the largest changes in surface, mode, and
intermediate waters associated with water mass ventilation and
anthropogenic CO2 uptake (Fig. 4C). The good correspondence
between the model and the observations is reflected by a
correlation between ΔH Cant–IPSL and ΔH Cant of 0.92 ± 0.11
and an intercept of −0.0009 ± 0.0019 ($r^2 = 0.83, P < 0.01$).

The IPSL simulations spanned the period from 1850 to 2014 and
included the entire Atlantic Ocean, allowing the
interannual-to-decadal variability of ΔHNat determined during the
1993–94 and 2013 cruises to be placed in a larger context.
Because climate models statistically represent the natural vari-
ability of the ocean–atmosphere system but not necessarily its
effect timing (28), the variability patterns of ΔHNat–IPSL are
described in terms of the SD of the values along the cruise
transect and are compared with the amplitude of the observed
ΔHNat [abs(ΔHNat)] (Fig. 3 C and F). The comparison
between the model and the observations therefore was not exact,
because one is an SD over 164 y and the other is the difference
between two sections over 20 y. The observed changes and

Discussion
The observed changes in pH in near-surface/upper waters were in
general agreement with changes expected because of anthropogenic
CO2. However, there was an appreciable signal resulting from natural
variability that was particularly pronounced in the intermediate
waters and near-frontal systems. Surface layers showed acidification
rates up to −0.0021 ± 0.0012 y in the South Atlantic, and high
acidification rates also were observed in the water masses along the
ventilation pathways, namely, mode waters with rates up to −0.0018 ±
0.0012 y in the South Atlantic and −0.0017 ± 0.0015 y in the North
Atlantic. Intermediate waters showed changes up to −0.0014 ±
0.0007 y, whereas deep waters in the South Atlantic showed smaller
changes of approximately −0.0004 ± 0.0003 y. Although, on average,
acidification rates were higher in near-surface waters, the
highest local values were observed in subsurface mode waters
between the depths of 200 and 500 m. Higher acidification rates
(i.e., more negative pH) in subsurface waters were in agreement
with the high acidification rates observed in the subsurface waters
at the ALOHA (A Long-term Oligotrophic Habitat Assessment)
and BATS (Bermuda Atlantic Time-Series) time-series stations
(12, 14) in the South Atlantic (30) and along a North Pacific
section (7), respectively.

For the first time, to our knowledge, our observations confirm
the major role of mode and intermediate waters (MW–IW) at the
basin scale in the acidification of the ocean interior, which also was evidenced in the climate models (9). The high rates of
acidification are attributed to the lower buffering capacity of
mode waters compared with surface waters, because mode
waters have a lower temperature and AT/CT ratio and therefore are
more sensitive to increasing oceanic CT (9, 10). In addition,
mode waters have a larger exposure to the atmosphere, with a
surface area-to-volume ratio five to 10 times larger than that of
deep waters, thus enabling a larger uptake of atmospheric CO2
and, hence, stronger acidification (9).

The observed changes in pH in surface waters were largely of
anthropogenic origin (Fig. 4A). The rates associated with anthro-
pogenic forcing, ranging from −0.0015 to −0.0020 y, were found
throughout the Atlantic surface waters from 50°S–36°N and are
similar to rates previously reported in the South Atlantic (30) and
by the European Station for Time Series in the Ocean in the North
Atlantic (11). Modeling studies have confirmed the dominant role of
atmospheric anthropogenic forcing on pH changes in the surface
layers (2, 31). We found larger values of ΔH Cant, associated with larger ΔCant, in the South Atlantic than in the North Atlantic. The
larger ΔCant in the south compared with the north is attributed
partly to the southern transects of the cruises having been located
in the western Atlantic, whereas the northern transects were
located in the eastern Atlantic. Mode and deep water formation are
concentrated in the western Atlantic in both hemispheres. Water
masses in the northeastern region of the section are expected to be
older than in the southwestern region and to have lower ΔCant
values. However, another process probably explains why ΔCant
and ΔH Cant were weaker in the North Atlantic. In the subpolar
region of the North Atlantic, a weakening in the formation of
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between the model and the observations therefore was not exact,
because one is an SD over 164 y and the other is the difference
between two sections over 20 y. The observed changes and
modeled ΔpHNat variability presented similar patterns, with large changes in the surface layers and in the MW–IW of the southern and northern regions (Fig. 3 B and E). The ΔpHNat in the equatorial region was smaller in the model than in the observations. However, the observed ΔpHNat was uncertain in surface, mode, and intermediate waters in the equatorial region, because these water masses are compressed into a thin layer of about 120 m with few measurements.

The model output was used to elucidate the controls of the large ΔpHNat in the MW–IW of the northern and southern regions. The observations showed a strong relationship between ΔpHNat and ΔNO3 (Fig. 3 B and E) and between ΔpHNat and the apparent oxygen utilization (AOU) (SI Appendix, Fig. S3), which suggests that pHNat changes were mostly the result of changes in CT from biological remineralization. The strong relationship between ΔpHNat and ΔNO3 also was found in the MW–IW of the IPSL model, with 0.013 units of pH change per micromole of NO3 (r² = 0.93). The model showed that 80% of the ΔpHNat could be explained by changes in CT, whereas AΔT accounted for 15–20% of the changes, and salinity accounted for less than 10% of the changes (Fig. 5A). For the pHNat variations attributed to CT changes, 50–65% corresponded to the physical transport of CT by ocean circulation, and 15–30% corresponded to in situ biological remineralization of organic matter within the water mass (Fig. 5A).

The strong correlation between ΔpHNat and ΔNO3 supports the notion that the increase in CT controlled the observed pHNat changes but that transport of respired carbon was the main contributor, rather than local biological remineralization.

The largest observed and modeled pHNat changes in the MW–IW occurred where the water masses were formed and subjected to variability in surface forcing south of 40°S (Fig. 3C and F). In the South Atlantic, the modeled ΔpHNat in newly formed water depended on the latitude where the formation occurred (Fig. 5B). Formation at higher latitudes led to the subduction of higher-CΤ/ lower-AΔT waters, thereby decreasing ΔpHNat, with the opposite holding true if formation occurred at lower latitudes. In agreement with previous studies (35, 36), the model results suggest that the SAM might explain part of the latitudinal shift. The positive phase of the SAM shifts westerly winds and the MW–IW subduction poleward (Fig. 5B). Additionally, the positive phase of SAM increases the upwelling, northward Ekman transport, and subduction of CΤ-enriched CDW into the MW–IW (37), thereby making ΔpHNat more negative. This mechanism was supported further by the higher AOU found in association with lower pHNat in both the observations (SI Appendix, Fig. S3) and the model (Fig. 5B). It explains the large observed decrease in pHNat in the MW–IW south of 40°S between 1993 and 2013 (Fig. 3C), because in this period the SAM trended positive.

In the North Atlantic, at the mode and intermediate outcrop, ΔpHNat variability in the model depended on the volume of newly formed water (Fig. 5C). When larger volumes of MW–IW were formed, a larger proportion of the deeper CO2– and AOU-rich waters of northern origin were subducted (SI Appendix, Fig. S4), leading to more negative ΔpHNat. There is observational evidence that the winter-mixed layer depth (38), volume, and AOU content (39) of the MW–IW in the North Atlantic have increased over the past decades. This mechanism could explain the decrease in pHNat observed in the MW–IW in the North Atlantic between 1993 and 2013.

In summary, the observations and model show that decreases in the pH of near-surface waters are dominated by the contribution of Cant. In the MW–IW, anthropogenic and natural components are of the same order of magnitude and are in the same direction from 1993 to 2013, with natural changes reinforcing the anthropogenic acidification signal. The observations and model output confirm that, although carbonate chemistry and geochemical properties of the water masses are crucial for explaining long-term changes in pH and oxygen levels (3, 9), changes in ocean circulation and biological production (13, 14, 40) also exert a large impact on pH on interannual-to-decadal time scales in subsurface waters. Based on projections of future increases in atmospheric CO2 and the associated penetration of Cant into intermediate and deep waters, the contribution of anthropogenic forcing to the acidification of subsurface waters will increase with time and eventually will exceed natural variability and trends.

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Fig. 5. ΔpHNat in mode and intermediate waters (MW–IW) over the Atlantic Ocean (80°S–65°N) in the IPSL model. (A) Contribution to ΔpHNat associated with changes in salinity (S, cyan), changes in normalized AΔT (nAΔT, red), and changes in normalized CT (nCT) caused by physical transport (blue) and biological activity (green). (B and C) ΔpHNat time-series in newly formed MW–IW in the South Atlantic (B) and North Atlantic (C). Anomalies in latitude, volume of formed waters, AOU, SAM index, and North Atlantic Oscillation (NAO) index also are shown with their correlation coefficient to ΔpHNat.
Materials and Methods

Data Processing. To obtain the changes in the variables (pH, Cant, Cant-TTD, θ, and NO3) between 1993-94 and 2013, we used the position and depth of the 100 stations of the FICARAM-XV line as a base. Data from the 94 stations of OACES/CO2 that coincided with the FICARAM-XV line were interpolated to the station positions and depths of FICARAM-XV using a Delaunay triangulation to obtain an interpolated dataset with the same grid for both periods. This interpolated dataset allowed us to determine differences in the variables over the 20-y time period. For Table 1, we determined an average and SD for all of the values in a particular water mass ("layer") for each region. More details on the datasets can be found in SI Appendix, Table 51.

Uncertainties. The uncertainty of ΔpH amounted to 0.0035 based on the analytical uncertainty of each pH dataset (0.001, 0.003, and 0.0015 for OACES/CO2, CITHER-II, and FICARAM-XV, respectively). The uncertainty of Cant was based on a propagation of error for each of the variables used in the calculation that yielded an SD of a Cant of 2.7 μmol/L. This calculation translates to an uncertainty of 0.005 in ΔpHCant and ΔCantNat, which is the maximum error for ΔpHCant as shown in SI Appendix, SI Text.

The IPSL Climate Model. The low-resolution (LR) IPSL climate model (IPSL-CM5A-LR), which participated in the Coupled Model Intercomparison Project Phase 5 (10) was used. The IPSL simulations covered the period from 1850 to 2014. They included sensitivity experiments separating anthropogenic perturbations from natural variability and climate-induced feedbacks. Four IPSL simulations were investigated: (i) the historical run from 1850 to 2005, with increasing CO2 concentrations and changes in radiative forcing such as volcanoes and aerosols; (ii) an "intermediate" representative concentration pathway (RCP4.5) for the period 2006-2014, characterized by an additional radiative forcing of 4.5 Wm-2 and corresponding to a CO2 atmospheric concentration of 538 ppm in 2100; (iii) a sensitivity experiment (esmfbk2) from 1850 to 2014 that included the increased radiative forcing of model sensitivity experiments separating anthropogenic perturbations from natural variability and climate-induced feedbacks.

The models were run with the ocean model ECHAM6/MPI-OM (12) and the IPSL Climate Model.

To obtain the changes in the variables (pH, Cant, Cant-TTD, θ, and NO3) from the historical run, the pHCant-IPSL was computed from the difference between 2013 and 1993.5 at the 111 profiles of the FICARAM-XV section using the model pixel closest to the station location (Fig. 18). The ΔpHCant-IPSL was estimated using the interannual anomaly of pHNat over the 164 y of the simulation and ΔpHCant-IPSL was based on the 95% confidence interval of ΔpHCant-IPSL (±1.96 × SD, i.e., 3.92 × SD).

Although the model results were compared with observations along the cruise transect (Fig. 18), the processes regulating ΔpHCant in the MW–IW were computed for the entire Atlantic basin (80°S–65°N). The SAM index was computed based on sea-level pressure.

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