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A changing nutrient regime in the Gulf of Maine

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ABSTRACT

Recent oceanographic observations and a retrospective analysis of nutrients and hydrography over the past five decades have revealed that the principal source of nutrients to the Gulf of Maine, the deep, nutrient-rich continental slope waters that enter at depth through the Northeast Channel, may have become less important to the Gulf's nutrient load. Since the 1970s, the deeper waters in the interior Gulf of Maine (> 100 m) have become fresher and cooler, with lower nitrate (NO_3) but higher silicate ($Si(OH)_4$) concentrations. Prior to this decade, nitrate concentrations in the Gulf normally exceeded silicate by 4–5 μM, but now silicate and nitrate are nearly equal. These changes only partially correspond with that expected from deep slope water fluxes correlated with the North Atlantic Oscillation, and are opposite to patterns in freshwater discharges from the major rivers in the region. We suggest that accelerated melting in the Arctic and concomitant freshening of the Labrador Sea in recent decades have likely increased the equatorward baroclinic transport of the inner limb of the Labrador Current that flows over the broad continental shelf from the Grand Banks of Newfoundland to the Gulf of Maine. That current system now brings a greater fraction of colder and fresher deep shelf waters into the Gulf than warmer and saltier offshore slope waters which were previously thought to dominate the flux of nutrients. Those deep shelf waters reflect nitrate losses from sediment denitrification and silicate accumulations from rivers and in situ regeneration, which together are altering the nutrient regime and potentially the structure of the planktonic ecosystem.

1. Introduction

The Gulf of Maine is a semi-enclosed continental shelf sea partially isolated from the open NW Atlantic by Georges and Browns Banks (Fig. 1). Oceanographic research in the Gulf has enjoyed a rich history, especially since the pioneering work of Henry Bryant Bigelow in the early part of the last century (Bigelow, 1926, 1927; Bigelow et al., 1940), and interest continues today stimulated in part by the Gulf's high biological productivity, with measured rates of primary production in offshore waters averaging 270 g C m^{-2} yr^{-1} (O'Reilly and Busch, 1984; O'Reilly et al., 1987). The principal source of nutrients supporting this production has been generally thought to be the influx of nutrient-rich deep slope water from beyond the Gulf through the Northeast Channel (Ramp et al., 1985; Schlitz and Cohen, 1984; Townsend, 1991, 1998). Once delivered to the Gulf, those nutrients are mixed into the surface layers by way of various physical mechanisms including winter convection and tidal mixing.

Nutrient concentrations in the source waters are, of course, important determinants of the total new primary production possible in the Gulf. Two types of slope waters are involved: Labrador Sea Slope Water (LSW), which is relatively cold and fresh, and the significantly warmer and saltier Warm Slope Water (WSW), originating from the Gulf Stream and North Atlantic Central Water (Gatien, 1976). Nutrient concentrations are higher in WSW, with nitrate concentrations greater than 24 μM, while LSW has about 15–17 μM nitrate; silicate concentrations are slightly lower in LSW than WSW but range from about 11 to 15 μM in both (Fig. 2; Drinkwater et al., 2002; Petrie and Yeats, 2000; Townsend et al., 2006; Townsend and Ellis, 2010). Changes in the predominance of either LSW or WSW in the deep waters of the Gulf of Maine have been correlated with the North Atlantic Oscillation (NAO; Smith et al., 2001; Greene and Pershing, 2003; Thomas et al., 2003; Townsend et al., 2006), which is a decadal-scale oscillation of wintertime surface atmospheric pressure over the Arctic (Icelandic Low) and the subtropical Atlantic (the Bermuda-Azores High). During low NAO index years, when north–south pressure differences are least, LSW (beyond the 200 m isobath) can be traced as far south as the New York Bight (following a 1–2-year lag), versus NAO high index years when Labrador Slope Water extends only as far south as the Laurentian Channel well east of Nova Scotia. It is thought that the "cold 1960s" (Colton, 1968; True and Wiitala, 1990; Petrie and Drinkwater, 1993; Petrie and Yeats, 2000; Loder et al., 2001), for

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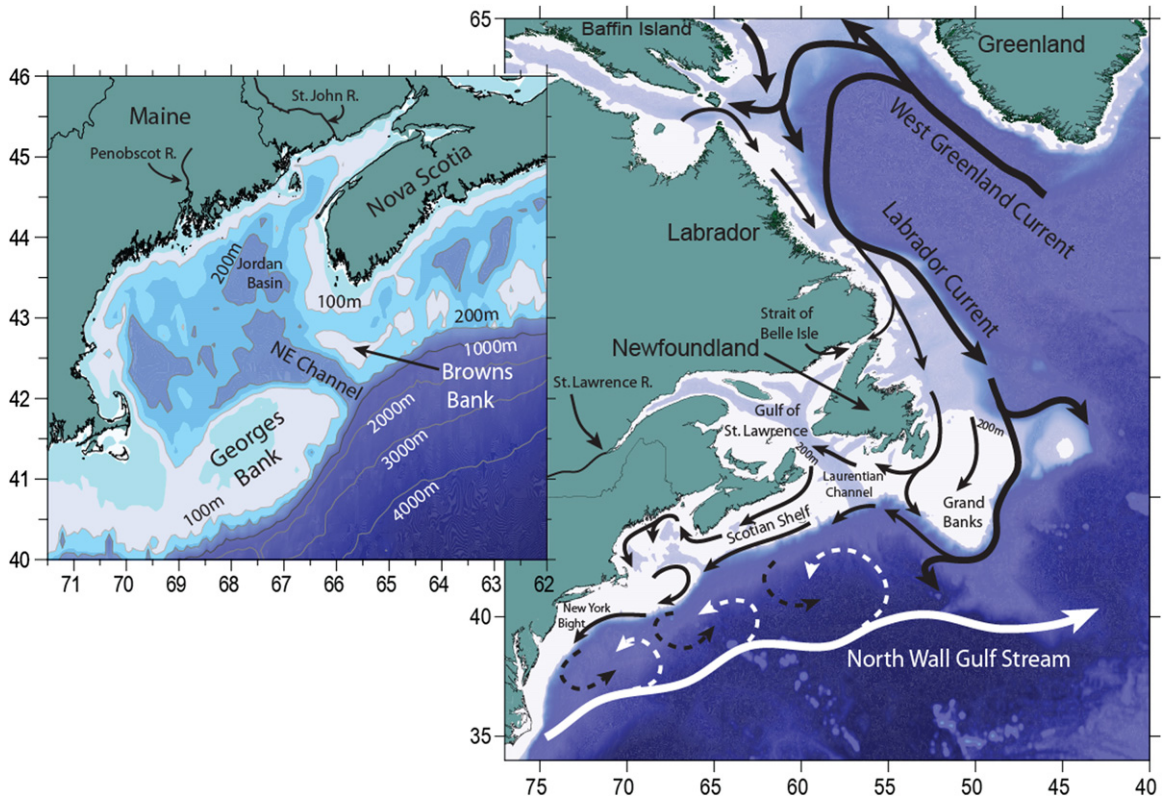

Fig. 1. Map of the NW Atlantic Ocean, Labrador Sea and Gulf of Maine, showing the major current systems (after Chapman and Beardsley, 1989; Loder et al., 1998), isobaths in m, and various features referred to in the text. Dashed arrows indicate mixing of waters (not currents) in the slope sea (Csanady and Hamilton, 1988). Inset shows location of the Northeast Channel (sill depth ca. 220 m) and the channel between Browns Bank and Nova Scotia (depth ca. 150 m).

example, resulted from a protracted NAO low which increased transport of cold LSW to the south (Smith et al., 2001; Pershing et al., 2001) where it entered the deep Gulf of Maine through the Northeast Channel. Details of how a brief NAO low in 1996 influenced shelf waters on the Scotian Shelf and in the Gulf of Maine have also been described (Pershing et al., 2001; Drinkwater et al., 2002; Greene and Pershing, 2003; Thomas et al., 2003).

The oceanographic effect of NAO is to influence the relative proportions of the two different slope water masses available for entrance to the Gulf of Maine at depth through the Northeast Channel, but regardless which water mass dominates, both slope water sources have significantly higher concentrations of nitrate than silicate (Fig. 2). This excess nitrate becomes important in determining the species composition of the phytoplankton. For example, diatoms dominate the spring phytoplankton bloom in the Gulf of Maine, and take up nitrate and silicate in roughly equal proportions. Because nutrient concentrations in deep source waters have historically been higher in nitrate than silicate, silicate is typically depleted first, thus limiting the diatom bloom and leading to seasonal changes in phytoplankton species composition in a manner consistent with post-bloom levels of residual nitrate. Exceptions, of course, are near shore and estuarine areas that receive inputs of river waters high in silicate (Schoudel, 1996; Anderson et al., 2008).

We present evidence in this communication, based on oceanographic observations and a retrospective analysis of nutrients and hydrography, that over the past several decades the principal source of nutrients to the Gulf of Maine, the deep, nutrient-rich continental slope waters that enter at depth through the Northeast Channel may have become less important to the Gulf's nutrient load. As a result of accelerated melting in the Arctic and freshening of the Labrador Sea in recent decades the equatorward baroclinic transport of the inner limb of the Labrador Current over the continental shelf has likely increased, which is now bringing a greater fraction of colder and fresher deep shelf waters to the Gulf of Maine than warmer and saltier offshore slope waters. More importantly, and unlike slope waters, those deep shelf waters reflect nitrate losses from sediment denitrification and silicate accumulation from rivers and *in situ* regeneration, thus altering the nutrient regime and potentially the structure of the planktonic ecosystem. Furthermore, this shelf water flow appears to overwhelm the opposing effects of positive NAO indices that should have resulted in a greater flux of WSW after the 1970s, and reduced fresh water discharges from the region's major rivers. Together these two phenomena should be making the Gulf warmer, saltier and higher in nitrate, not cooler, fresher and lower in nitrate.

2. Materials and methods

2.1. Oceanographic survey

We conducted an oceanographic survey in the northern Gulf of Maine aboard the R/V *Cape Hatteras* from June 29 to July 2, 2006, surveying an area intended to capture the distal end of the Eastern Maine Coastal Current system and its associated upwelling of deep Gulf of Maine waters (Pettigrew et al., 2005). Standard hydrocasts were made at 78 stations (Fig. 3) using a SeaBird CTD and carousel water sampler equipped with 5-l Niskin bottles. Water samples were analyzed for phytoplankton chlorophyll, concentrations of inorganic nutrients, and phytoplankton species composition and abundance.

Nutrient samples were filtered through Millipore HA filters, placed immediately in a seawater-ice bath for 5–10 min and frozen at −18 °C to be analyzed following the cruise for NO_3+NO_2, NH_4, $Si(OH)_4$ and PO_4 using a Bran Luebbe AA3 autoanalyzer and

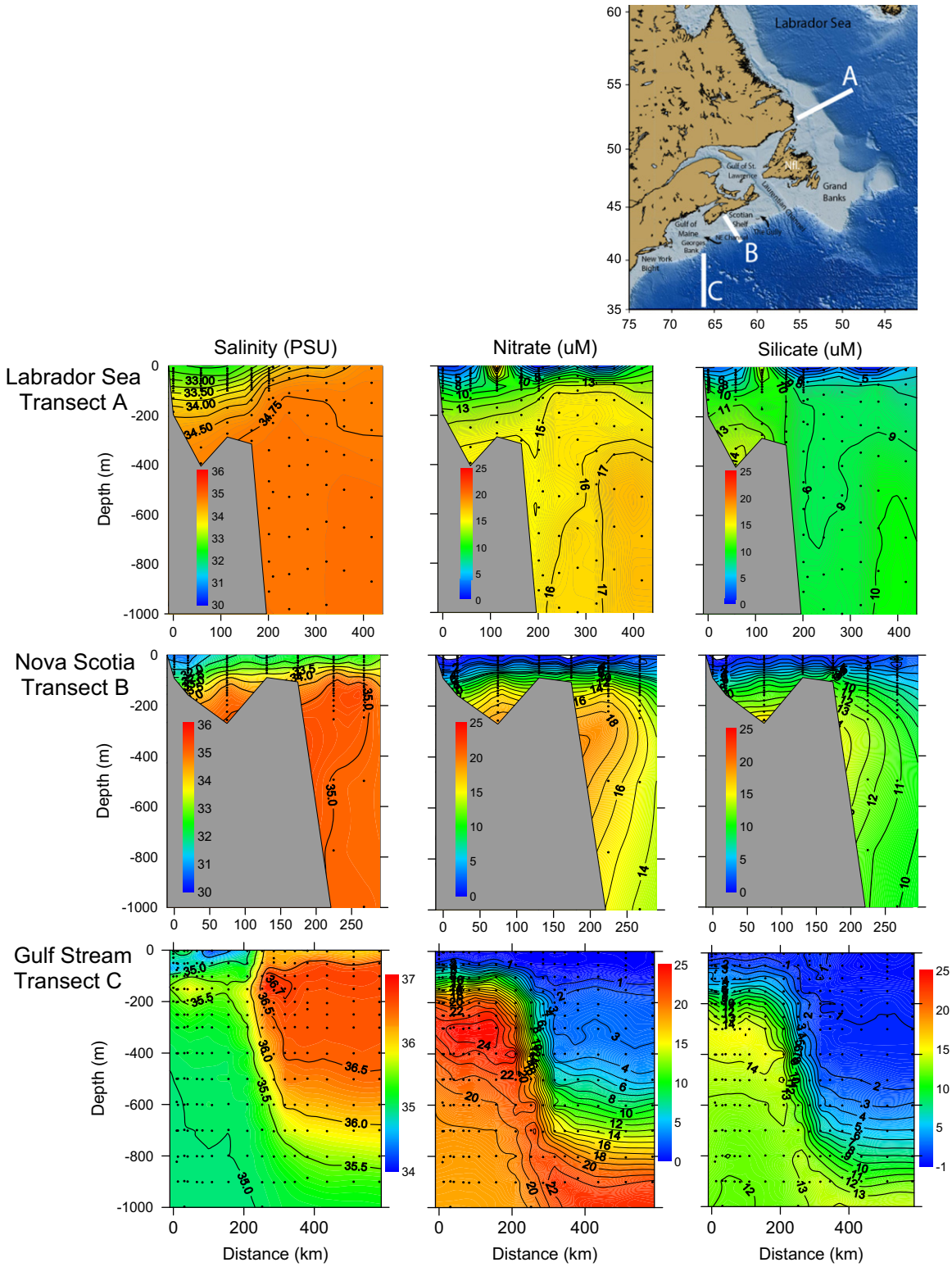

Fig. 2. After Townsend and Ellis (2010). Vertical sections of salinity, nitrate and silicate in the top 1000 m along Transects A–C in map, June 1997. Dots indicate discrete sample depths. Transect A extends from the coast out into the Labrador Sea; Transect B crosses the Nova Scotian Shelf; Transect C is off Georges Bank to the Gulf Stream. Data source: World Ocean Circulation Experiment ("eWOCE-Electronic Atlas of WOCE Data", at http://www.ewoce.org/).

standard techniques. Phytoplankton chlorophyll *a* analyses were based on fluorometric analyses of acetone extracts of particulate material collected from 100 ml on GF/F glass fiber filters (Parsons et al., 1984). Surface water samples (1 m depth, 100 ml preserved in Lugol's iodine solution) were collected at alternate stations for determination of concentrations and taxonomic composition of the phytoplankton populations. Samples were concentrated by settling 50 ml of the sample for 48 h in a graduated cylinder. The top 40 ml were siphoned off and the remaining 10 ml mixed; a 1 ml subsample of the five-fold concentrated sample was placed in a counting cell, and phytoplankton cells enumerated using an inverted compound microscope at 200 × and identified to major

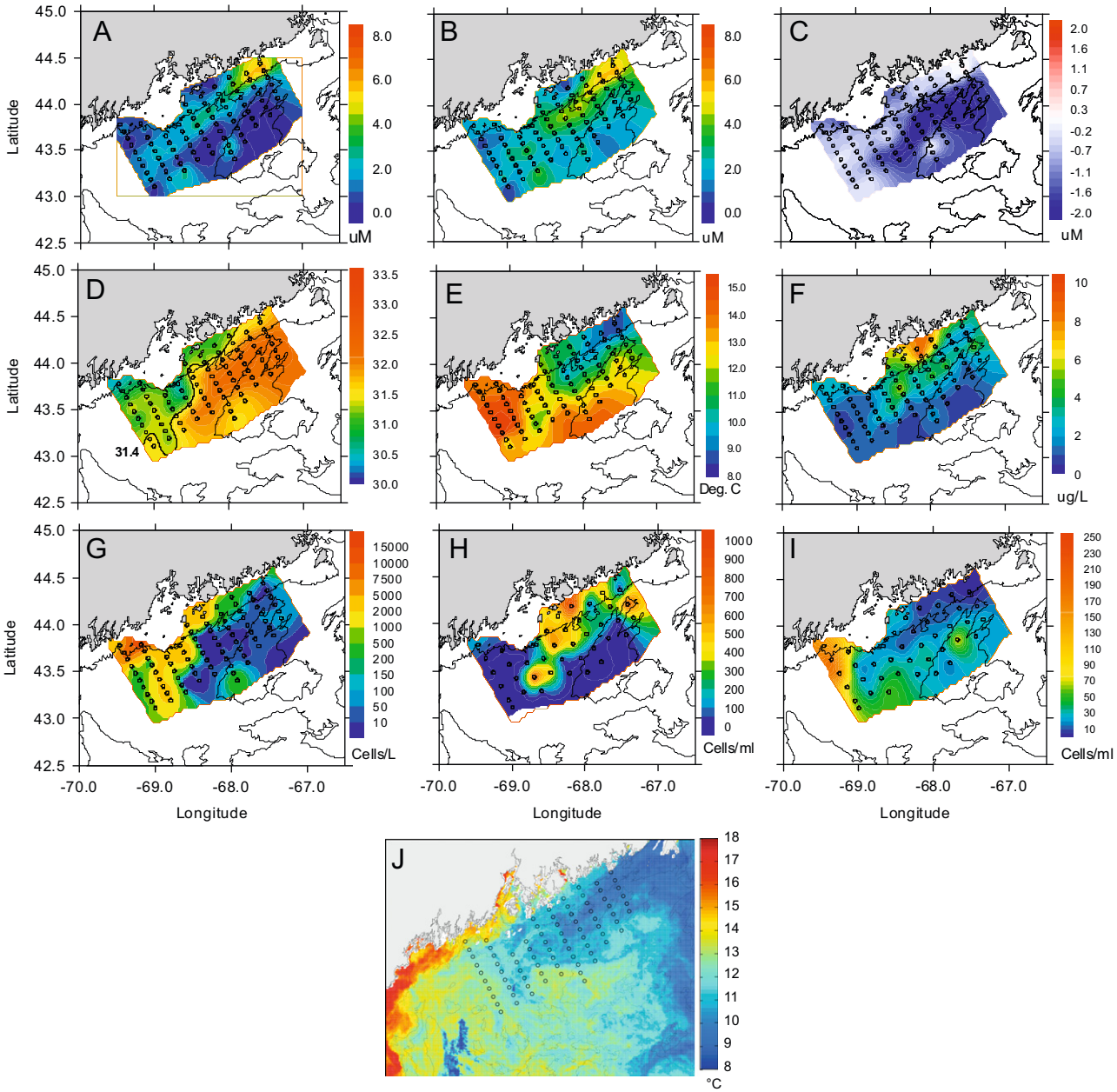

Fig. 3. Panels A–I: Areal contour plots for R/V *Cape Hatteras* cruise results, 29 June–2 July 2006, with station locations indicated: (A) concentrations in top 20 m (average of samples collected at 1, 10 and 20 m) of nitrate, assumed equal to nitrate (NO_3) plus nitrite (NO_2), (B) same as (A) but for silicate ($Si(OH)_4$), (C) residual nitrate in top 20 m (e.g., nitrate concentration minus silicate concentration); red shades are positive values, blue shades are negative values, (D) surface salinity, with the 31.4 isohaline given, (E) surface temperature, (F) surface chlorophyll *a*, (G) surface cell densities of *Alexandrium fundyense*, given in cells l^{-1}, (H) surface cell densities of diatoms, given in cells ml^{-1}, (I) surface cell densities of "other" (non-*Alexandrium*) dinoflagellates, given in cells ml^{-1}, and (J) AVHRR thermal IR satellite image of sea surface temperature on 16 July 2006 (first cloud-free image, 13 days following our survey cruise); central core of the Eastern Maine Coastal Current corresponds to cold waters extending from northeast to southwest. Sampling stations for the 29 June–2 July 2006 cruise are given. Data courtesy of A.C. Thomas, Satellite Oceanography Data Lab, University of Maine.

taxonomic group (dinoflagellates, diatoms or other flagellates). The dominant genera in those taxonomic groups were recorded.

Water samples for the enumeration of *Alexandrium fundyense*, the "red tide" dinoflagellate responsible for paralytic shellfish poisoning (PSP), were collected at all stations by sieving 2 l of water from the surface Niskin bottle (1 m) through a 20 μm mesh screen; the concentrate was preserved in a 5% formaldehyde seawater solution and stored in 20 ml vials in the dark in a refrigerator. Quantitative cell counts were performed on shore within 4 months of collection based on epifluorescence microscopy and an immunological stain specific to the genus *Alexandrium* (Adachi et al., 1993) as described in Townsend et al. (2005).

2.2. Retrospective analyses of hydrographic and nutrient data

We analyzed a geographically defined subset of a nutrient and hydrographic database for the Gulf of Maine region. The larger database (encompassing the area defined by 40–45.5°N and 71–65°W) is designed as a macronutrient dataset with dissolved inorganic nitrogen, phosphate, and silicate, but which also includes corresponding values of temperature and salinity (Rebuck et al., 2009; the entire database is available at http://grampus.umeoce.maine.edu/nutrients/). The majority of data in the Rebuck et al. database comes from two public sources: the World Ocean Database (WOD; Boyer et al., 2006; Johnson

et al., 2006; http://www.nodc.noaa.gov) maintained at the US National Oceanographic Data Center, and data compiled at the Marine Environmental Science Division at the Bedford Institute of Oceanography and the Marine Environmental Data Service in Ottawa, both of the Department of Fisheries and Oceans Canada (http://www.dfo-mpo.gc.ca/), which is updated from an earlier published version (Petrie et al., 1999). These two public sources were supplemented with additional data from various sources that have remained in provisional or unreleased status; those sources are given in Rebuck et al. (2009).

Because the nutrient database includes only historical data with matched measurements of temperature, salinity and nutrients (e.g., "bottle" data) it is not as data rich as the significantly larger body of high-resolution CTD, also available from the World Ocean Database (WOD), but which does not include nutrient data (WOD05 High-resolution Conductivity–Temperature–Depth/XCTD May 2009 update, at: http://www.nodc.noaa.gov). The total number of temperature and salinity data pairs in this much larger dataset is 215,803. No additional quality control methods were applied after downloading. Data were binned by year of collection to produce an annual mean.

2.2.1. Data analyses

Prior to the 1960s the Rebuck et al. database lacks significant numbers of matched observations of temperature, salinity, nitrate, and silicate at each bottle depth, but thereafter sufficient data exist to allow grouping into decadal averages. We computed for each decade from the 1960s to the 2000s the average temperature, salinity, nitrate, and silicate concentrations for waters deeper than 100 m in the southeastern corner of Gulf of Maine, defined as a box 42.3–43.6°N and 68.6–66.0°W (e.g., see Fig. 5F), an area we chose to represent newly entered deep waters in the Gulf (see Appendix A). Anomalies against the 48-year average (1960–2008) were calculated and plotted for temperature and salinity; we did not attempt to correct for season of collection as most of the data were collected in the warmer months of the year and we assumed that such a potentially confounding factor is minimized, although not eliminated, at depths greater than 100 m, which approximates the depth of winter convective mixing in the eastern Gulf of Maine. These data were also used to compute for each decade the residual nitrate, calculated as nitrate (μM) minus silicate (μM) concentrations on a sample-by-sample basis prior to averaging. Each parameter was compared independently using a one-way ANOVA. We also produced temperature–salinity diagrams for three depth strata (0–40, 40–100 and >100 m). The deep water nutrient data (>100 m) were also used in decadal property–property plots of silicate and nitrate by depth to illustrate temporal trends in the proportions of total nutrient loads. Finally, we tested our resulting decadal averages of temperature, salinity and nutrients for the possibility that irregularly sampled interannual variations in water properties may have aliased the result. We therefore compared the high resolution CTD temperature and salinity data with our decadal averages.

3. Results and discussion

3.1. 2006 oceanographic survey and phytoplankton cell distributions

Shipboard observations in the summer of 2006 (Fig. 3) revealed near surface (average of the surface, 10 and 20 m) silicate concentrations that exceeded nitrate by 1–2 μM, with the higher silicate concentrations associated with the colder, higher salinity waters reflecting a deep-water source. Those nutrient distributions were tied closely to the distributions and taxa of phytoplankton, with dinoflagellates (including the red tide dinoflagellate *A. fundyense*) restricted to waters low in silicate relative to nitrate, surrounding the plume of newly upwelled water high in silicate relative to nitrate, which in turn held high cell densities of diatoms.

The cell densities of diatoms, the red tide dinoflagellate *A. fundyense*, and other non-*Alexandrium* dinoflagellates during our 2006 oceanographic survey displayed overlapping distributions, but overall, the distributions of diatoms were spatially distinct from the dinoflagellates. The highest *Alexandrium* cell densities were associated with the inshore frontal edge of the cool, high nutrient waters of the southwestward-flowing Eastern Maine Coastal Current (EMCC), which is clearly evident in satellite SST images as a cold-water feature extending from the northeast to the southwest, offshore of the eastern Maine coast (Fig. 3J). These *Alexandrium* distributions are similar to those described earlier (Townsend et al., 2001, 2005) with the exception that we observed fewer cells distributed along the outer edge of the EMCC in this study. Instead, the highest cell densities (>1000 cells l^{-1}) were confined to waters inshore of the 31.4 isohaline. Within that patch of *Alexandrium* cells the highest densities did not abut the shoreline; instead they were just seaward of, and not coincident with, the lowest salinity waters adjacent to the coast, which are associated with the freshwater plume of the Penobscot River. Those strongly stratified near shore waters of the Penobscot plume are also visible as warmer surface temperatures in the SST image (Fig. 3J). In contrast to *Alexandrium* cells, diatoms were more spatially consistent with the core of the EMCC, straddling either side of its inshore frontal edge, and thus the diatoms appeared to be restricted to more recently upwelled, and hence colder, more nutrient-rich waters, showing only partial overlap with the *Alexandrium* cells. Cell densities of diatoms were two to three orders of magnitude greater than those of *Alexandrium*.

The distribution of phytoplankton chlorophyll was consistent with that of diatoms, indicating that, unlike *Alexandrium*, diatoms were a major component of the phytoplankton biomass. Dominant groups of diatoms sampled were species of *Chaetoceros*, *Skeletonema*, *Thalassiosira*, *Leptocylindrus* and *Pseudo-nitzschia.*

Distributions of "other" (non-*Alexandrium*) dinoflagellates were similar to *Alexandrium*, with lowest cell concentrations within the EMCC, which had both higher diatom cell concentrations than "other" dinoflagellates and high chlorophyll concentrations. Those "other" dinoflagellates include species of *Ceratium*, *Gymnodinium*, *Gyrodinium*, *Prorocentrum*, *Peridinium*, and *Scrippsiella*. Unlike diatoms and dinoflagellates, the distributions of other microflagellates (not shown), which included various cryptomonads and *Phaeocystis* sp., did not reveal any obvious spatial coherence with the hydrography or other phytoplankton taxa.

These distributions of phytoplankton taxa were clearly related to the nitrate residual (Fig. 3C) in which the dinoflagellates appeared restricted to higher nitrate residual waters (near zero values in Fig. 3C); the exception is the near shore high nitrate residual region north of the core of the EMCC (Fig. 3E and J), in which cell densities of dinoflagellates were low. We suspect that the reason is the high turbulence there as a result of vigorous tidal mixing, which completely mixes the water column to depths of 60–100 m. Diatoms were more closely associated with waters of low (negative) nitrate residual. This near-inverse distribution of diatoms and dinoflagellates has been attributed to competitive interactions in an earlier study (Townsend et al., 2005), in which regions with high cell densities or growth rates of diatoms were argued to interfere with dinoflagellates, especially *A. fundyense*.

The key finding in this survey was that there were slightly higher concentrations of silicate than nitrate in the near surface

waters of the Gulf of Maine, which was unexpected given the concentrations in the probable slope water sources. Complete nutrient profiles at the outermost, deep-water stations (> 200 m) on our survey cruise (Fig. 4) show roughly equal subsurface concentrations of nitrate and silicate all the way to the bottom, and not higher nitrate concentrations, which is inconsistent with that of the presumed slope water sources. Nutrient profiles from the same location in the 1970s and 1980s show higher concentrations of nitrate (Fig. 4) and illustrate a greater nitrate residual (nitrate minus silicate), which is more in keeping with the slope water sources. Nutrient concentrations in the two slope water types, Warm Slope Water and Labrador Slope Water, are illustrated in Fig. 2 as vertical cross sections of the top 1000 m at three locations: off southern Newfoundland and into the Labrador Sea; across the Scotian Shelf; and across the continental slope and into the Gulf Stream south of Georges Bank. Those data reveal relatively low nitrate (< 18 μM) and silicate (< 14 μM) in the Labrador Sea, slightly elevated concentrations of nitrate and silicate in slope waters off Nova Scotia, and relatively high concentrations of each in slope waters off Georges Bank (nitrate > 24 μM; silicate > 14 μM). These deep slope water nutrient loads reflect the relative ages of the deeper water masses, with recently formed Labrador Sea slope waters reflecting low nutrient concentrations of the surface waters from which they originated, while older slope waters associated with the Gulf Stream reflect nutrient loads that have accumulated over time from recycling of sinking organic matter. While nitrate concentrations are highest in Warm Slope Water, silicate concentrations in both water masses are not very different from one another; however, both water masses are significantly higher in nitrate than silicate. The absolute and relative concentrations of nitrate and silicate we observed in our brief survey cruise therefore do not reflect those of either slope water source, LSW or WSW, in that silicate concentrations were equal to, or slightly exceeded nitrate. The nature of this change in nitrate–silicate proportions is revealed in an analysis of historical nutrient and hydrographic data.

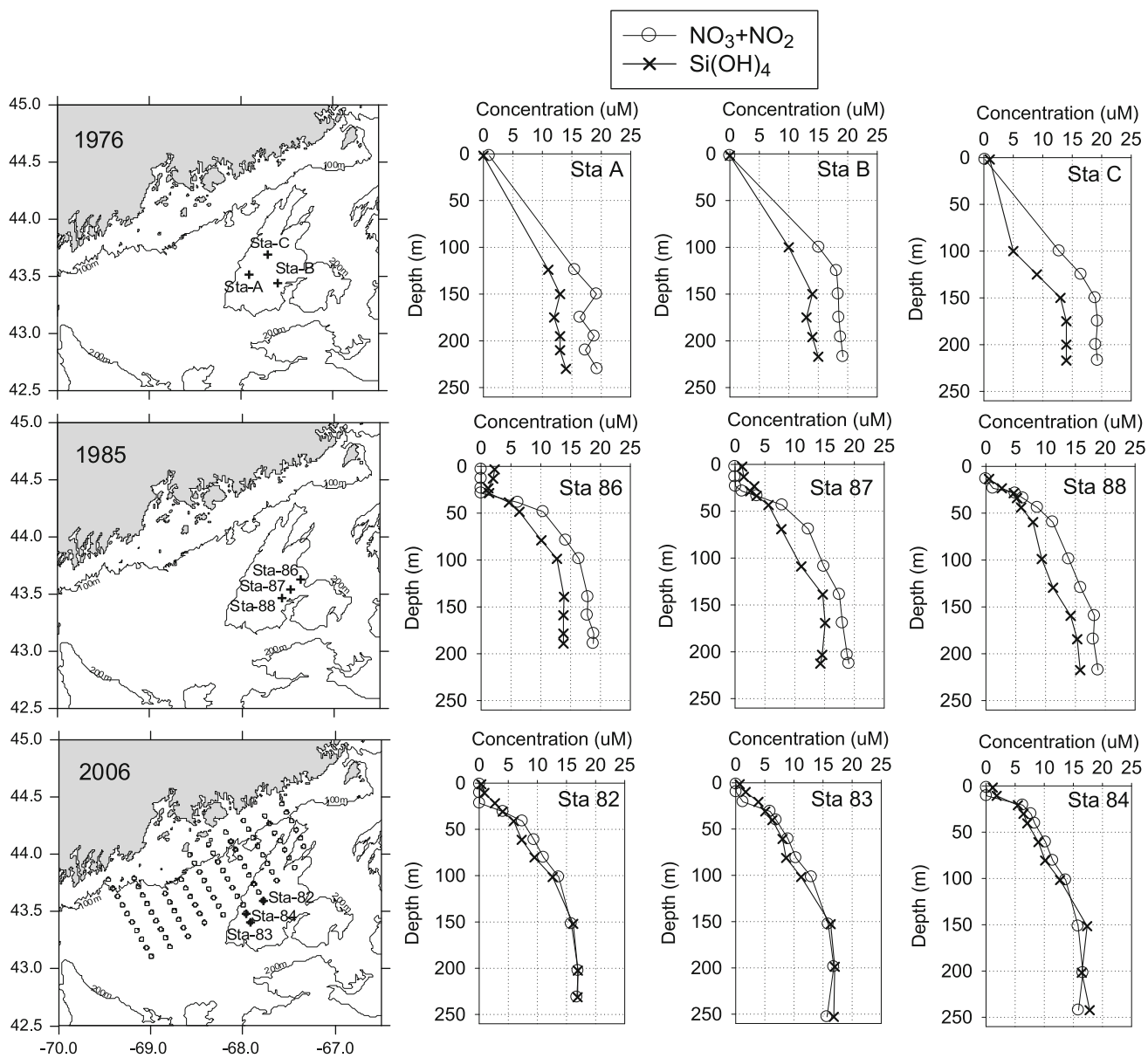

Fig. 4. Vertical profiles of nitrate and silicate concentrations in μM at stations in Jordan Basin in the Gulf of Maine as indicated in maps. Top: From May 1976 (data from World Ocean Database); Middle: From July 1986 (data from Townsend and Christensen, 1986); Bottom: From R/V *Cape Hatteras* cruise, 29 June–2 July 2006.

3.2. Retrospective analyses of hydrographic and nutrient data

Anomalies of average deep water (>100 m) temperature, salinity, nitrate and silicate, are given in Fig. 5 for each decade back to the 1960s. Those decadal averages reveal evidence of the "cold 1960s" (Colton, 1968; True and Wiitala, 1990; Petrie and Drinkwater, 1993; Petrie and Yeats, 2000; Loder et al., 2001), with a marked switch to warmer and saltier properties in the deep waters of the eastern Gulf of Maine in the 1970s. This pattern was followed by a gradual return to colder and fresher water properties by the 1990s and 2000s.

Deep water concentrations of nitrate and silicate in the 1960s were generally low. While our 1960s data include only 31 discrete water samples in the eastern Gulf from depths >100 m, the "cold 1960s" phenomenon is well documented in the literature and resulted from a deep water influx into the Gulf of relatively cold and fresh LSW (Smith et al., 2001; Petrie and Yeats, 2000; Loder et al., 2001). Our analysis of the higher-resolution historical CTD data in Fig. 6 (discussed further below) show this pattern of cold waters in the 1960s and warm waters in the 1970s clearly, and are thus supportive of the more limited data presented in Fig. 5. Lower nutrients are characteristic of LSW, which was present in the 1960s, in contrast to WSW which has significantly higher nitrate concentrations. In the 1970s an influx of WSW brought higher nitrate but no statistically significant change in silicate. While the following three decades (1980s, 1990s and 2000s) indicated a return

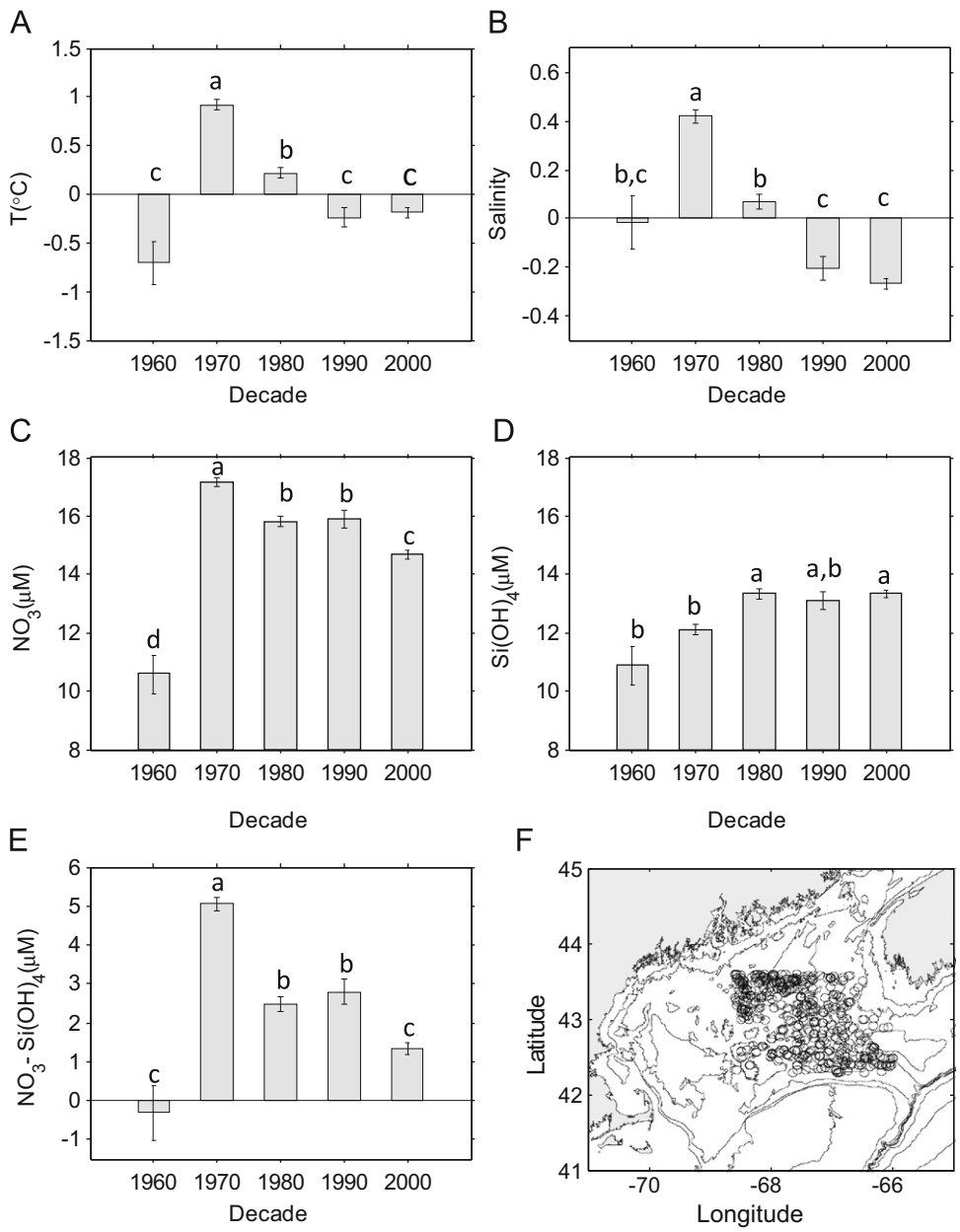

Fig. 5. Retrospective analyses of historical temperature, salinity, nitrate (NO_3) and silicate ($Si(OH)_4$) data for deep waters of the eastern Gulf of Maine restricted to waters >100 m, and within the box defined as 42.3–43.6°N and 68.6–66.0°W, by decade from the 1960s to the 2000s; station locations are given in (F). Only those temperature and salinity data with corresponding nutrient samples were used. Sample numbers: 1960s—31; 1970s—474; 1980s—401; 1990s—146; 2000s—642. (A and B): Temperature and salinity anomalies (departures from the five-decade averages, where average T=7.06 °C and average S=34.075); (C and D): Average nitrate and silicate concentrations; (E) residual nitrate, computed as nitrate minus silicate concentrations on a sample by sample basis prior to decadal averaging. Mean values for each decade are plotted with whisker plots of ±1 S.E. on each. Each parameter was compared independently using a one-way ANOVA for significance at a $p<0.01$ level. Bars are labeled with letters that identify statistically significant groups.

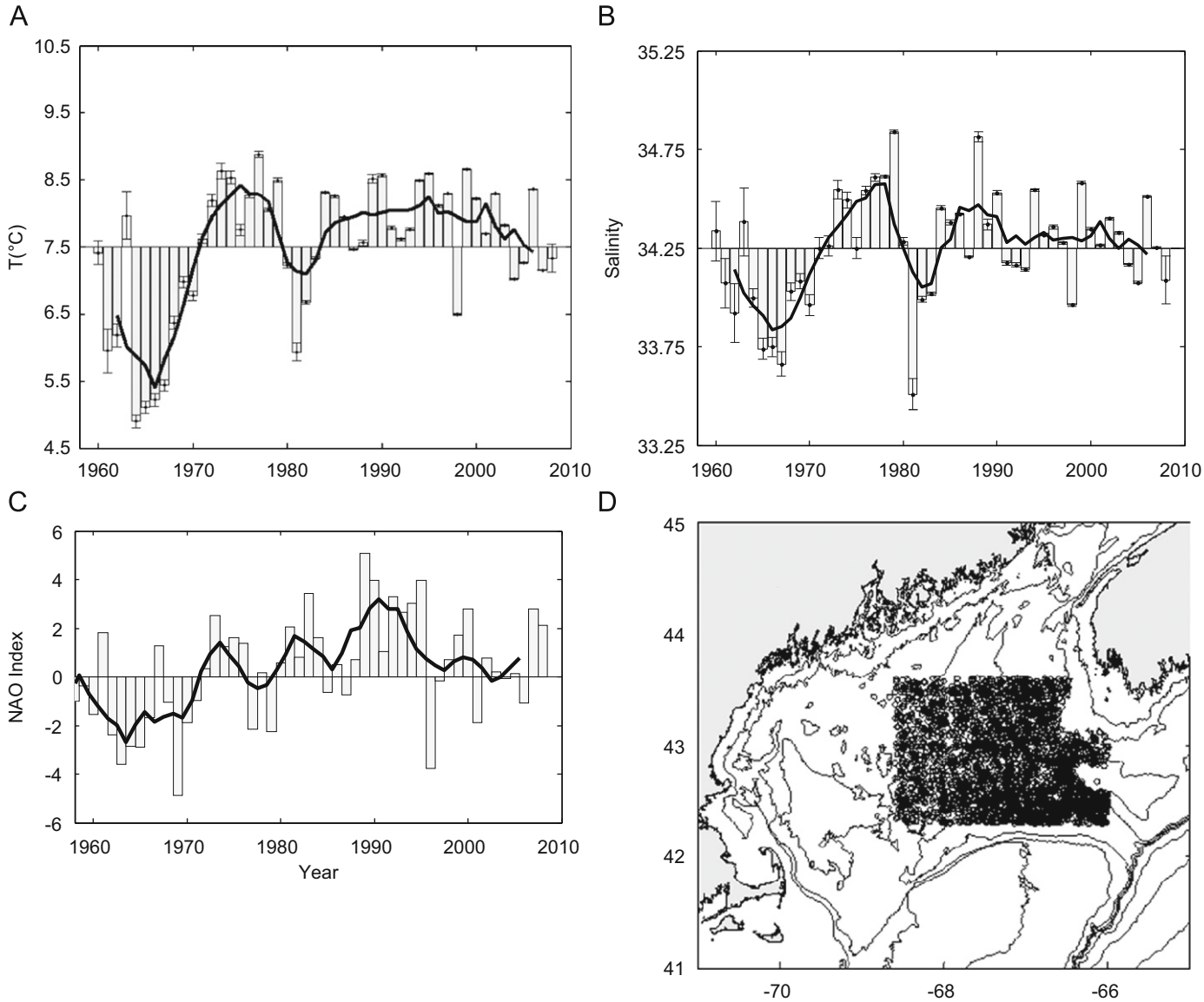

Fig. 6. (A) and (B) Bar graphs of annual mean temperature and salinity, with ±1 S.E., plotted with reference to the 48-year average, with 5-year running averages. Data are ≥100 m at the southeastern corner of the Gulf of Maine (same geographical range as the decadal analyses in Fig. 5; 42.3–43.6°N, 68.6–66°W) from 1960 to 2008. Station locations are shown on map (D) 500. Data source: WOD05 High-resolution Conductivity–Temperature–Depth/XCTD May 2009 update (http://www.nodc.noaa.gov). Total number of temperature and salinity data pairs=215,803. (C) North Atlantic Oscillation annual mean values of winter index (December–March) back to 1958 (http://www.cgd.ucar.edu/) are plotted with a 5-year running average.

to colder and fresher waters and correspondingly a decline in nitrate concentrations, silicate concentrations increased and remained relatively high into the 2000s. These decadal nutrient trends are summarized as nitrate residuals (nitrate minus silicate concentrations; Fig. 5E) which show an abrupt increase in residual nitrate from the 1960s to the 1970s, followed by a general decline in subsequent decades; the 2000s value is the lowest since the 1960s. These changes in nutrient concentrations and ratios are inconsistent with simply a dominance of one slope water source or the other, as confirmed by *T*–*S* (temperature–salinity) relationships (Fig. 7).

In the 1960s, waters at depths >100 m were defined by a *T*–*S* envelope with relatively cold and salty deep water properties (Fig. 7), which averaged about 6.5 °C and about 34 salinity (with no salinities exceeding 35), characteristic of modified LSW source waters (LSW end member defined as 34.90 S and 6.0 °C; Houghton and Fairbanks, 2001). The next decade (the 1970s) included a large number of *T*–*S* data pairs that indicated the addition of a volume of warmer (by 1.5–2 °C) and slightly saltier water of WSW origin (WSW end member defined as 35.20 S and 10.0 °C; Houghton and Fairbanks, 2001); in addition, there was evidence of a less dense mixture of fresher water (<33 S) producing a slight tail-like feature extending to the lower left in the *T*–*S* diagram. The same mixture of water masses below 100 m was present in the 1980s but in addition there were *T*–*S* data pairs extending in a more pronounced tail-like feature to the lower left, indicating the addition of a cooler (by more than 2 °C), fresher (<32.5 S) and less dense water mass beneath 100 m. That water mass resided above the warmer, saltier and denser WSW, which was still present, as was the cooler and still denser LSW which was on the bottom (relative depths indicated by density contours on the *T*–*S* diagram). In the 1990s the cool and fresh tail-like feature had all but disappeared, as had much of the WSW, and on average the temperatures and salinities of the center of mass of the *T*–*S* data pairs had become cooler and slightly fresher. In the 2000s the *T*–*S* data pairs occupied a broader *T*–*S* space (a wider range of *T* and *S*); that is, in this latest decade

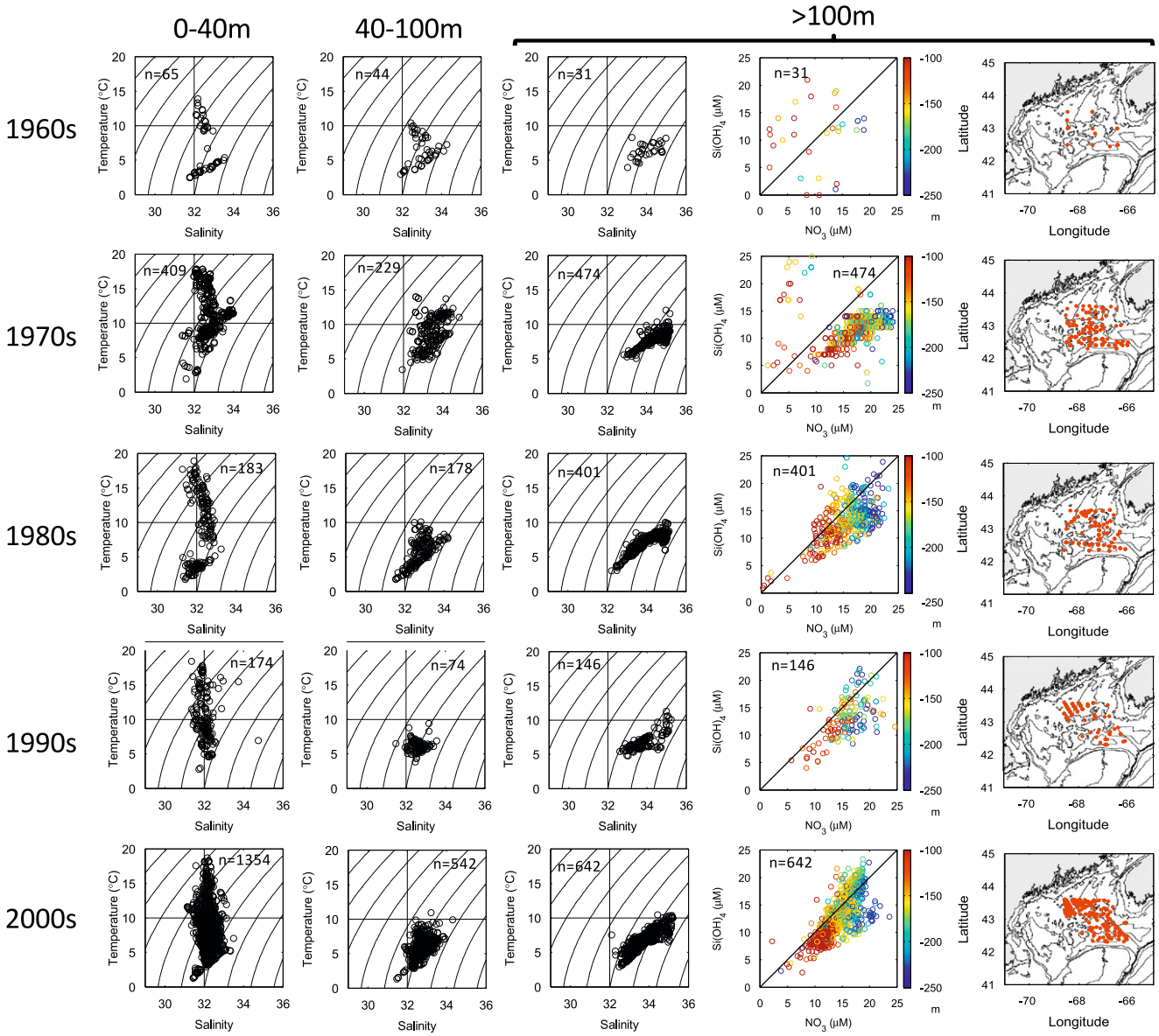

Fig. 7. *T–S* (temperature–salinity) plots, for all depths and for depths >100 m, and depth-specific property–property plots of silicate ($Si(OH)_4$) and nitrate (NO_3) concentrations at depths >100 m, along with maps of station positions, for each decade. Contour lines of constant density (sigma-t, or density anomaly) are given in the *T–S* diagrams (with increasing values toward the lower right), as are reference lines at 32 S and 10 °C. Each data point in the property–property plots of silicate and nitrate is color-coded to the depth of collection, with colour bar giving depths in m; a 1:1 reference line is given, as are numbers of samples.

there is evidence of the addition of both cooler and fresher waters resembling the tail-like feature prominent in the 1980s, in addition to the warmer and saltier WSW residing just above LSW.

Fig. 8 presents evidence that the decadal trends in deep water salinity in Fig. 5 are not the result of variations in freshwater discharge of the St. Lawrence River, which is responsible for more than half the freshwater in the Gulf of Maine (Houghton and Fairbanks, 2001), or the two largest rivers that drain directly into the Gulf of Maine, the Penobscot and the St. John (Fig. 1). In fact, the temporal trends in river discharge are very nearly opposite to our observed salinity changes in the Gulf of Maine: e.g., increased river discharges coincide with increased salinities in the Gulf of Maine – not just at depths greater than 100 m, but at all depths (Fig. 8, panels E–G). The St. Lawrence River discharge was low in the late 1960s, when the Gulf of Maine was relatively fresh, and reached a maximum discharge in the 1970s, when the Gulf of Maine was the saltiest. After the 1970s the discharge gradually decreased while the Gulf of Maine gradually freshened. Similar discharge patterns, although not as pronounced, can be seen for the St. John and Penobscot Rivers.

A potential source of error in these decadal averages, apart from station location which we discuss in Appendix A, is the possibility of interannual variations in water properties that when subsampled at irregular intervals, as is the case in our nutrient database, can produce aliasing and therefore false decadal-averaged patterns. Comparing those decadal changes to the larger CTD database (N=215,803 temperature and salinity data pairs over the five decades; Fig. 7) we see a similar pattern. The "cold 1960s" and "warm 1970s" are clearly present in the CTD data. The CTD data also show a drop in temperature and salinity in the early 1980s, which is seen as the "tail-like feature" in the *T–S* diagrams

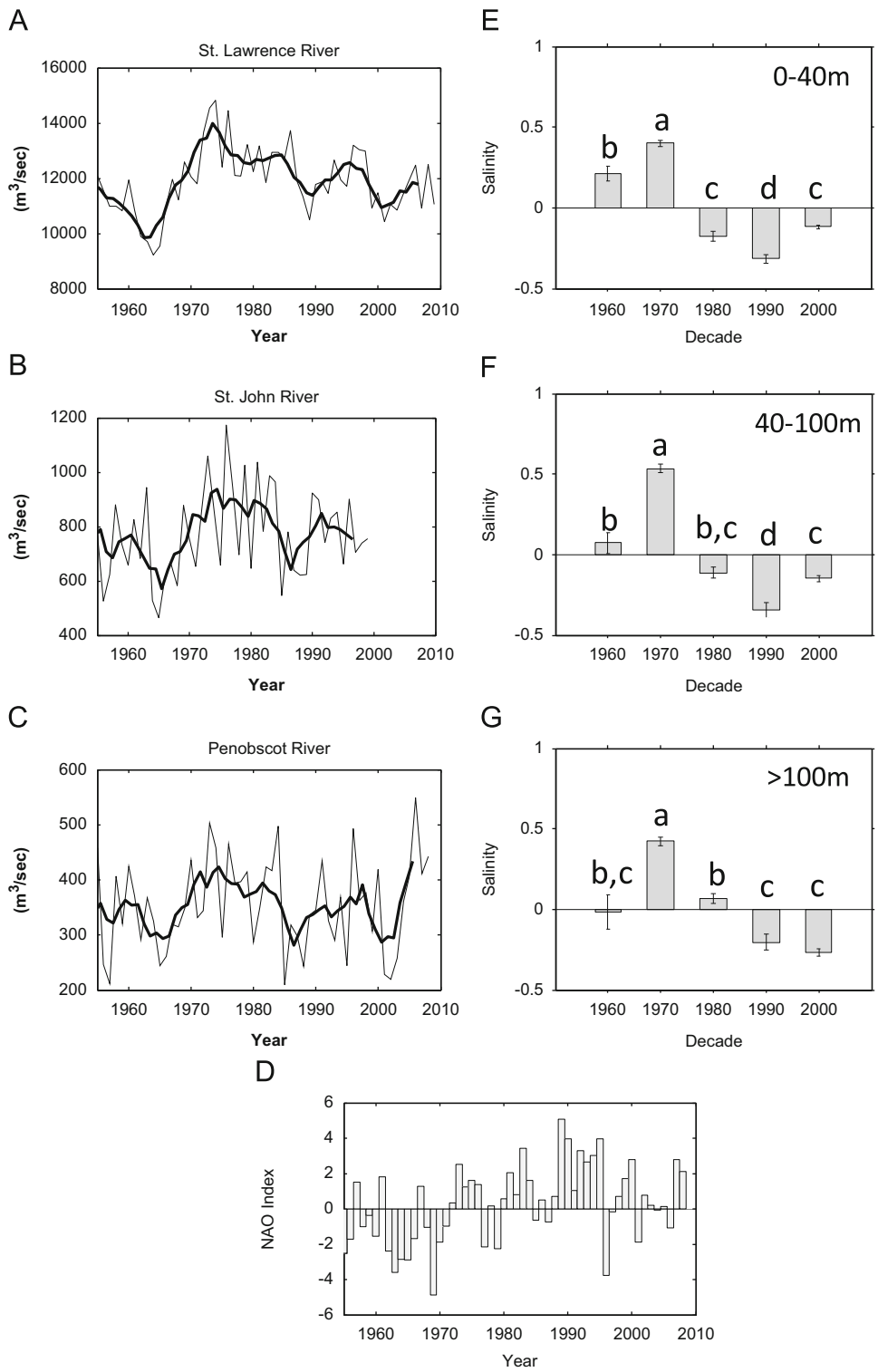

Fig. 8. (A–C) River gauge data for the St. Lawrence River (http://www.meds-sdmm.dfo-mpo.gc.ca/), the St. John River (http://www.meds-sdmm.dfo-mpo.gc.ca/) and the Penobscot River (http://waterdata.usgs.gov/nwis/) back to 1955, as annual totals and as 5-year running averages. Discharge is given as m^3 per second. (D) North Atlantic Oscillation annual mean values of Winter Index (December–March) back to 1955 (http://www.cgd.ucar.edu/). (E–G) Decadal salinity anomalies in the sample domain in Fig. 5 for the top 40 m (five-decade average salinity = 32.360), 40–100 m (five-decade average salinity = 32.995) and between 100 m and the bottom (panel G same as in Fig. 5; five-decade average = 34.075). Mean values for each decade are plotted with whisker plots of ± 1 S.E. on each. Each parameter was compared independently using a one-way ANOVA for significance at a $p < 0.01$ level. Bars are labeled with letters that identify statistically significant groups.

discussed above, followed by higher values later that decade. Apart from that dip, the CTD data support the general decline in temperature (by about 1 °C) and salinity (by about 0.7 S) after the 1970s, as also shown in Fig. 5 for the much smaller dataset.

These analyses indicate that from the 1960s to the 1970s the deep waters of the eastern Gulf of Maine became saltier and warmer, but from the 1970s to the 1990s and 2000s they became significantly fresher and cooler (Fig. 5; there were no significant

differences between the 1990s and 2000s). More importantly, the freshening and cooling of the deep waters after the 1970s was accompanied by a change in the nutrient loads.

Details of changes in both the absolute concentrations and relative proportions of nitrate and silicate at depths >100 m in the Gulf of Maine over the past five decades are more clearly brought out in the property–property plots alongside the *T–S* diagrams in Fig. 7. Nutrient data for the 1960s, although sparse, show variable nitrate–silicate ratios. The 1970s data were generally clustered below the 1:1 line, with highest silicate concentrations (e.g., corresponding to deepest depths) that, with few exceptions, did not exceed 15 μM, and much higher nitrate concentrations approaching 24 μM. While outlying high silicate–low nitrate values in the 1960s and 1970s plots may be inaccurate, the bulk of the 1970s nitrate and silicate values were characteristic of slope waters in general and were most likely a mixture of both LSW and WSW, dominated by the latter. In the 1980s maximal silicate concentrations had increased above the 1970s level while nitrate decreased. This trend of decreasing nitrate continued into the 1990s and 2000s. The overall decrease in deep water nitrate and increase in silicate after the 1970s appears to be related to changes in proportions of deep slope water sources, but only partly. That is, apparent in the 2000s, and perhaps in the 1980s and 1990s, is a bifurcation in the data (Fig. 7), producing a fork at the highest nutrient concentrations (and deepest depths) that most likely resulted from addition of another water mass to the deep slope waters, above the 1:1 line — a water mass with a lower nitrate-to-silicate ratio (in contrast to slope waters that have higher N:Si, most obvious in the 1970s), and corresponding to the cooling and freshening of that deep layer. This additional water mass is most likely deep Nova Scotian Shelf water.

The Gulf of Maine is located in a region of sharp latitudinal temperature gradients, created by the confluence of the cold, equatorward-flowing Labrador Current and the poleward flowing Gulf Stream. The two current systems and water masses associated with them (LSW and WSW) mix in the slope sea region at the offing of the Northeast Channel at the entrance to the Gulf of Maine (Csanady and Hamilton, 1988). As already discussed, changes in the predominance of either LSW or WSW in the slope sea have been correlated with the North Atlantic Oscillation (NAO; Pershing et al., 2001; Smith et al., 2001; Drinkwater et al., 2002; Greene and Pershing, 2003). The "cold 1960s" was most likely the result of a protracted NAO low that increased transport of relatively cold and fresh LSW to the south (Pershing et al., 2001; Smith et al., 2001) where it entered the deep Gulf of Maine through the Northeast Channel. The overall trend in NAO winter indices after the "cold 1960s" has been predominantly positive (Fig. 6C), which would favor the transport of WSW into the deep waters of the Gulf of Maine rather than LSW, making the deep waters warmer, saltier and higher in nitrate. This appears to have been the case in the 1970s. After that, however, our analyses indicate that the deep waters of the Gulf of Maine became slightly colder and fresher, with lower nitrate and higher silicate, which is inconsistent with an NAO-forced influx of more WSW to the Gulf. The most plausible explanation for this discrepancy is a shift in the relative importance of Nova Scotian Shelf waters in recent decades, not just to the surface layers of the Gulf, but to depths beneath 100 m, which may in turn be related to changes in the dynamics of the Labrador Current that are independent of either cycles in the NAO or in discharges from the region's major rivers.

We suggest here that the addition of freshwaters from recent melting in the Arctic (Abdalati and Steffen, 2001; Smedsrud et al., 2008; Steele et al., 2004) is not only freshening the Labrador Sea, as alluded to by others (Smith et al., 2001; Greene and Pershing, 2003, 2007; Häkkinen, 2002) but is likely enhancing the baroclinic transport of the coastal limb of the Labrador Current over the continental shelf, which is leading to both fresher water on the shelf as well as a greater transport of modified LSW and Labrador Shelf Water as far south as the Gulf of Maine. The Labrador Current off Labrador extends across isobaths from the narrow continental shelf over the continental slope and rise (Fig. 1). The major flow branches into two currents, with most of the transport occurring over the continental slope beyond the outer edge of the shallow Grand Banks, with perhaps 10% (Chapman and Beardsley, 1989) flowing across the Banks. The cold and fresh admixture of shelf and slope waters continues southwestward to the Laurentian Channel, where a significant volume of fresh water from the St. Lawrence River is added, and on to the Nova Scotian Shelf (as the Nova Scotian Current). Some of that low salinity shelf current enters the Gulf of Maine between Browns Bank and Nova Scotia (Smith et al., 2001; Fig. 1). Should the baroclinic transport of this inner limb of the Labrador Current and Nova Scotia Current systems (e.g., Smith, 1983) be enhanced as a result of additional freshwater fluxes in the Arctic, the Gulf of Maine would receive a greater proportion of deep shelf waters than offshore slope waters, which in turn would exhibit dissolved inorganic nutrient loads significantly altered from their source in the Labrador Sea. That is, in transit along the continental shelf from the Grand Banks of Newfoundland to the Nova Scotian Shelf, as well as through the Gulf of St. Lawrence via the Strait of Belle Isle, sediment denitrification would reduce near-bottom nitrate concentrations (e.g., Christensen et al., 1987; Seitzinger and Giblin, 1996). In addition, we would expect to see those shelf waters accumulate remineralized biogenic silica in addition to ongoing fluxes from rivers, including the St. Lawrence River, and the Gulf of St. Lawrence. A similar phenomenon has been reported for the deep and bottom waters in the Gulf of Maine (Christensen et al., 1996), where nitrate is significantly reduced by sediment denitrification and silicate is enriched as those waters flow in a general counterclockwise pattern from the eastern Gulf to the western Gulf.

Changes in the Arctic and their associated effects on the transport of shelf waters to the south appear to overwhelm the opposing effects of local river discharge and NAO-driven changes in slope water fluxes to the Gulf of Maine. The patterns of temperature and salinity from high resolution CTD data in Fig. 6 are initially coherent with the NAO winter index, whereby positive NAO indices are reflected in higher salinities and warmer temperatures below 100 m in the Gulf, and low indices correspond with colder, lower salinity water; Fig. 6 also shows a brief NAO low period from 1978 to 1980. But in the later decades, the temperature and salinity responses are muted, and not nearly as obvious as in the 1960s and 1970s. For example, the period of highest NAO indices in the early 1990s correspond with gradually decreasing temperatures and salinities, which is opposite to that expected if NAO were correlated with a greater flux of WSW to the Gulf. The CTD data in Fig. 6, like the dataset in Fig. 5 show a temperature decrease of about 1 °C between the 1970 and the 2000s, and a decrease in salinity of about 0.7 S. Therefore, an increased baroclinic transport of shelf waters to the Gulf of Maine at depths below 100 m (between Browns Bank and Nova Scotia as well as through the NE Channel), driven by additional melt waters in the Arctic, is a better explanation for the slight cooling and freshening, and especially the reduction in nitrate and increase in silicate over the past several decades than NAO-forced fluctuations in the proportions of LSW and/or WSW, or variations in river discharges.

The resulting altered nutrient regime in the Gulf of Maine may be forcing changes in the structure of the planktonic ecosystem with respect to relative abundances of diatoms and dinoflagellates

in the offshore waters of the Gulf. Our oceanographic observations in 2006 revealed an inverse relationship between cell densities and distributions of diatoms and dinoflagellates, including the red tide dinoflagellate *A. fundyense*. When there were more diatoms there were fewer dinoflagellates. Predictably, those distributions corresponded with near-surface nitrate and silicate fields, reflecting the elevated silicate and depleted nitrate in the deep source waters. As such we might have in these analyses a contributing factor for the apparent onset of *A. fundyense* bloom phenomena in the 1970s (Anderson, 1997), which is when there was a shift from unfavorable *A. fundyense* growth conditions (a low nitrate residual) to favorable growth conditions (high nitrate residual). It is tempting to speculate that with increased freshening of the Labrador Current the trends we describe are likely to continue into the future, perhaps reversing the predominance of *A. fundyense* red tides that began in the 1970s. Similar cycles in commercially exploited species in the Gulf of Maine (northern shrimp and ground fish) also varied with these changes (Koeller et al., 2009), perhaps related to good and bad periods of diatom production. Testing of such speculations will, of course, depend on further, more detailed studies of the circulation and nutrient dynamics of shelf and slope waters of this region, the nature of ecological interactions among phytoplankton taxa, and the potential indirect impacts of a warming global climate.

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Appendix A. Testing for bias as a function of station locations

In selecting our sample domain in Fig. 5, which is intended to sample newly entered deep waters in the Gulf of Maine, we may have introduced a bias in our analyses of changes in deep water nutrients and water properties. For example, the station maps in Fig. 7 reveal variable station locations, the average of which might differ from one decade to the next. We analyze here how such geographical variability in station location within our sample domain might have influenced our decadal analyses.

As deep waters enter the Gulf of Maine, either through the Northeast Channel or from the Nova Scotian Shelf between Nova Scotia and Browns Bank, they can be expected to continue to lose nitrate by way of sediment denitrification, and to conserve or enrich silicate by way of remineralization. In addition, those waters will mix with the resident deep waters already inside the Gulf of Maine, which could alter the average salinity and either enrich or dilute the nutrient loads in the newly arrived deep source waters. Another compounding effect of variable station location, of course, is simply variable bottom depth of stations sampled. Therefore, source waters, upon entering and spreading throughout the Gulf of Maine, would be expected to exhibit a decrease in salinity, a decrease in nitrate concentrations, and an increase in silicate concentrations as a function of distance from a reference point at the entrance to the Gulf. If the mean distance of stations from that reference point varies among decades, then it is probable that some fraction of our observed differences in average decadal salinities and nutrient concentrations may be the result of such bias in the locations of stations in our historical database. To test this, we performed linear regressions of salinity, nitrate concentrations and silicate concentrations in waters deeper than 100 m as a function of linear distance from the lower right corner (southeast corner) of our sample domain (Fig. 5).

Results of that analysis are summarized in Table 1. The average station distances from the southeast corner of the domain in the 1960s, 1970s and 1980s were nearly identical (137, 135 and 137 km) and thus there was no station location effect on the values we report. The mean station locations did change in the 1990s and 2000s (e.g., they were 169 and 185 km, respectively), and it is these two decades that could have been biased, and which we analyzed in Table 1. When the slope of the linear regression equation was significantly different from zero, we used it and the change in station location (in km) from the 1980s value to compute an adjusted value of salinity, nitrate and silicate for the 1990s and 2000s. Those computed corrections were then compared with the variance explained by the regression equation (the coefficient of determination, or R^2). For salinity, the adjusted value in the 1990s was higher than the data mean by 0.19, and in

Table 1
Results of regression analyses of deep water (> 100 m) salinity and concentrations of nitrate and silicate by decade as a function of linear distance of sample station locations from the southeast corner of the sample domain (in Fig. 5). Number of samples and mean values for each decade of temperature, salinity, nitrate, silicate are given, as are the mean linear distances from the southeast corner. Adjusted decadal values of the mean salinity, nitrate and silicate were computed using the regression slope and the change in distance from the average of the 1960s, 1970s and 1980s (e.g., using 137 km).

Decade	*N*	Mean Dist. From SE Corner (km)	ΔDistance from 1980s (km)	Regression analyses											
				Salinity				Nitrate				Silicate			
				Mean salinity	Regress. slope (adj. factor)	Adj. salinity	R^2	Mean NO_3 (μM)	Regress. slope (adj. factor)	Adj. nitrate (μM)	R^2	Mean $Si(OH)_4$ (μM)	Regress. slope (adj. factor)	Adj. silicate (μM)	R^2
1960s	31	137	N/A	34.059	N/A	N/A	N/A	10.57	N/A	N/A	N/A	10.88	N/A	N/A	N/A
1970s	474	135	N/A	34.497	N/A	N/A	N/A	17.16	N/A	N/A	N/A	12.10	N/A	N/A	N/A
1980s	401	137	N/A	34.144	N/A	N/A	N/A	15.81	N/A	N/A	N/A	13.33	N/A	N/A	N/A
1990s	146	169	32	33.871	−0.006**	34.063	0.400	15.88	−0.002(ns)	N/A	N/A	13.09	0.021**	12.42	0.14
2000s	642	185	17	33.807	−0.006**	34.097	0.357	14.66	−0.012**	15.24	0.055	13.33	0.019**	12.41	0.08

**Significant at the 0.01 level.
R^2=coefficient of determination.
N/A=not applicable.
ns=not significant.

the 2000s it was higher by 0.29. Applying these adjustments to the salinity anomaly plot in Fig. 5 would likely mean that salinity did not change from the 1980s to the 2000s. However, while regression slopes were significantly different from zero ($P<0.01$), the R^2 was 0.40 in the 1990s and 0.36 in the 2000s, and therefore a salinity correction may not be warranted. For nitrate the regression slope in the 1990s was not significant (P=0.6), but the slope was significant in the 2000s, for which we computed a nitrate adjustment upward of 0.58 μM. However, the R^2 was only 0.055. In the case of silicate, the regression slopes in the 1990s and 2000s were both significant ($P<0.01$) but the R^2 was 0.14 in the 1990s and 0.08 in the 2000s. We conclude from these analyses that the effect of station location bias was negligible relative to the magnitude of the observed decadal changes in nutrient concentrations.

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