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A seventy-two-year record of diminishing deep-water oxygen in the St. Lawrence estuary: The northwest Atlantic connection

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Abstract

Oxygen concentrations in the bottom waters of the Lower St. Lawrence estuary (LSLE) decreased from 125 μmol L^{-1} (37.7% saturation) in the 1930s to an average of 65 μmol L^{-1} (20.7% saturation) for the 1984–2003 period. A concurrent 1.65°C warming of the bottom water from the 1930s to the 1980s suggests that changes in the relative proportions of cold, fresh, oxygen-rich Labrador Current Water (LCW) and warm, salty, oxygen-poor North Atlantic Central Water (NACW) in the water mass entering the Laurentian Channel probably played a role in the oxygen depletion. We estimate that about one half to two thirds of the oxygen loss in the bottom waters of the LSLE can be attributed to a decreased proportion of LCW. This leaves between one third and one half of the oxygen decrease to be explained by causes other than changes in water mass composition. An increase in the along-channel oxygen gradient from Cabot Strait to the LSLE over the past decades, combined with data from sediment cores, suggests that increased sediment oxygen demand may be partly responsible for the remainder of the oxygen decline. In July 2003, approximately 1,300 km^2 of seafloor in the LSLE was bathed in hypoxic water ($<$62.5 μmol L^{-1}).

Severe hypoxia is a condition that occurs in the water column when oxygen (O_2) falls below the 2 mg L^{-1} or 62.5 μmol L^{-1} level necessary to sustain most animal life (Diaz and Rosenberg 1995). Hypoxia occurs naturally in many coastal environments with restricted circulation, such as fjords, but hypoxia in coastal and estuarine areas with less restricted circulation appears to be on the rise due to anthropogenic nutrient loading and coastal eutrophication (Cloern 2001). Environments with a shallow, seasonally stratified water column can be ventilated by seasonal mixing events and are not generally hypoxic throughout the year (Rabalais et al. 2001). We report here on the recent emergence of persistent, year-round hypoxia ($<$62.5 μmol L^{-1}) in the Lower St. Lawrence estuary, an estuarine system that receives the second largest freshwater discharge in North America (10,900 m^3 s^{-1}; Bourgault and Koutitonsky 1999).

The dominant bathymetric feature in the Lower St. Lawrence estuary (LSLE) is the Laurentian Channel, a submarine valley with depths everywhere exceeding 250 m. The Channel extends 1,240 km landward from the continental shelf edge to Tadoussac (Fig. 1). The circulation in the Laurentian

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Channel is estuarine, with water flowing seaward in the surface layer and landward in the deep layer (Saucier et al. 2003). In the 200-m- to 300-m-deep layer, the cross-channel average of the landward along-channel flow component was estimated at 0.5 cm s^{-1} (Bugden 1988). Using this estimate, a parcel of water that enters the mouth of the Laurentian Channel at 250-m depth would reach the head of the Laurentian Channel in about 7 yr. More recent estimates of this travel time are on the order of 3 yr to 4 yr (Gilbert 2004). At 250-m depth, the water is isolated from the atmosphere throughout the year by a permanent pycnocline that begins around 100-m depth in the winter (Petrie et al. 1996). As this deep water travels landward, it gradually loses oxygen because of respiration and organic matter mineralization. More oxygen is lost along the way than can be replenished by vertical diffusion from the oxygen-rich overlying waters, causing progressively lower oxygen levels landward.

In this paper, we construct a multidecadal time series of oxygen in the bottom waters of the LSLE from all available sources of data. We also explain a major part of the observed trend in oxygen by quantifying the effect of interdecadal changes in the temperature–salinity–oxygen characteristics of the water entering the mouth of the Laurentian Channel at the edge of the continental shelf in the northwest Atlantic Ocean.

Methods

Oxygen data—From 1931 to 1938, personnel from Laval University (Québec City, Canada) operated a physical-, biological-, and chemical-sampling station at Trois-Pistoles (Fig. 1). They designed a grid of equally spaced stations in the St. Lawrence estuary, which they occupied a number of times each summer (Dugal 1934). Oxygen measurements by Winkler (1888) titration were performed by only two individuals, so that variability in end-point detection by different analysts was kept to a minimum. Their analytical method and sampling protocol were essentially the same as used today (Grasshoff et al. 1999). All temperature, salinity, and O_2 measurements from the 1932–1935 period were tabulated (Université Laval 1934, 1936). Based on the standard deviation of O_2 measurements repeated every 2 h at a fixed station (48.278°N, 69.333°W) over a 24-h period on 18–19 July 1934, we estimate the reproducibility of their measurements to be better than ± 8 μmol L^{-1}. This estimate is conservative, as we expect real O_2 variability over a semidiurnal tidal cycle at this station, located 20 km seaward of the head of the Laurentian Channel, where strong internal tides are generated (Saucier and Chassé 2000).

Several other data sets were incorporated into the analyses. To ensure a high degree of uniformity in methods and data quality in the O_2 time series, we limited the compilation to oxygen data determined by Winkler titration (Grasshoff et al. 1999). Continuous vertical profiles of O_2 measured with electronic sensors were not included in any of the time-series analyses. Most of the O_2 measurements compiled here come from the Biochem database (http://www.meds-sdmm.dfo-mpo.gc.ca/biochem/Biochem_e.htm). A second major source of oxygen data for the LSLE and the Gulf of St. Lawrence is the Maurice-Lamontagne Institute's Oceanographic Data Management System (http://www.osl.gc.ca/sgdo/en/accueil.html). Other sources of oxygen data include measurements made by the McGill University Marine Sciences Centre during the late 1960s and early 1970s (e.g., Steven et al. 1973) as well as data from various research surveys conducted between 1984 and 2003 (e.g., Gilbert et al. 1997). Given the large number of investigators and laboratories involved in the oxygen data collection, the quality of the data may not be uniform. We estimate that the reproducibility of most of the O_2 measurements is in the 0.3–5.0 μmol L^{-1} range. The complete list of references for the oxygen data as well as the entire data set of oxygen measurements from the bottom waters ($\geq$295 m) of the LSLE and their metadata are found in Web Appendix 1 at http://www.aslo.org/lo/toc/vol_50/issue_5/1654a1.html.

Temperature and salinity data—All temperature and salinity data come from the CLIMATE database (http://www.mar.dfo-mpo.gc.ca/science/ocean/database/data_query.html) developed and maintained at the Bedford Institute of Oceanography (Gregory 2004). For the Gulf of St. Lawrence, prior to 1943, all data came from bottle casts with reversing thermometers performed at discrete standard hydrographic depths (temperature accuracy $\pm$0.01°C, salinity accuracy $\pm$0.05). Starting in 1943, mechanical bathythermographs allowed measurements of temperature on a much finer vertical scale (temperature accuracy $\pm$0.1°C). A similar improvement in fine-scale salinity measurements took place in 1969, when electronic conductivity–temperature–depth (CTD) probes were first used for surveys in the Gulf of St. Lawrence (temperature accuracy $\pm$0.01°C, salinity accuracy $\pm$0.01). A very low 1934 annual mean salinity of 33.45 in the bottom waters ($\geq$295 m) of the LSLE was

Table 1. Latitude and longitude coordinates of the polygonal regions (Figs. 1A, 8A) for which we derived temperature, salinity, and oxygen statistics.

Region	Vertex 1 (lat °N, long °W)	Vertex 2 (lat °N, long °W)	Vertex 3 (lat °N, long °W)	Vertex 4 (lat °N, long °W)
Lower St. Lawrence estuary	48.674, 69.65	49.326, 68.01	48.752, 67.691	48.177, 69.023
Cabot Strait	47.26, 60.74	48.03, 59.43	47.5, 58.7	46.52, 60.22
Laurentian Channel mouth	45.198, 57.829	44.416, 57.242	44.884, 55.863	45.666, 56.464
North Atlantic central water	35, 71	36, 71	38.3, 50	35, 50
Labrador current water (LCW)	42.840, 49.443	43.960, 48.687	44.827, 48.729	45.863, 47.554
LCW (continued)	46.003, 47.973	45.163, 49.149	43.932, 49.359	43.036, 49.946

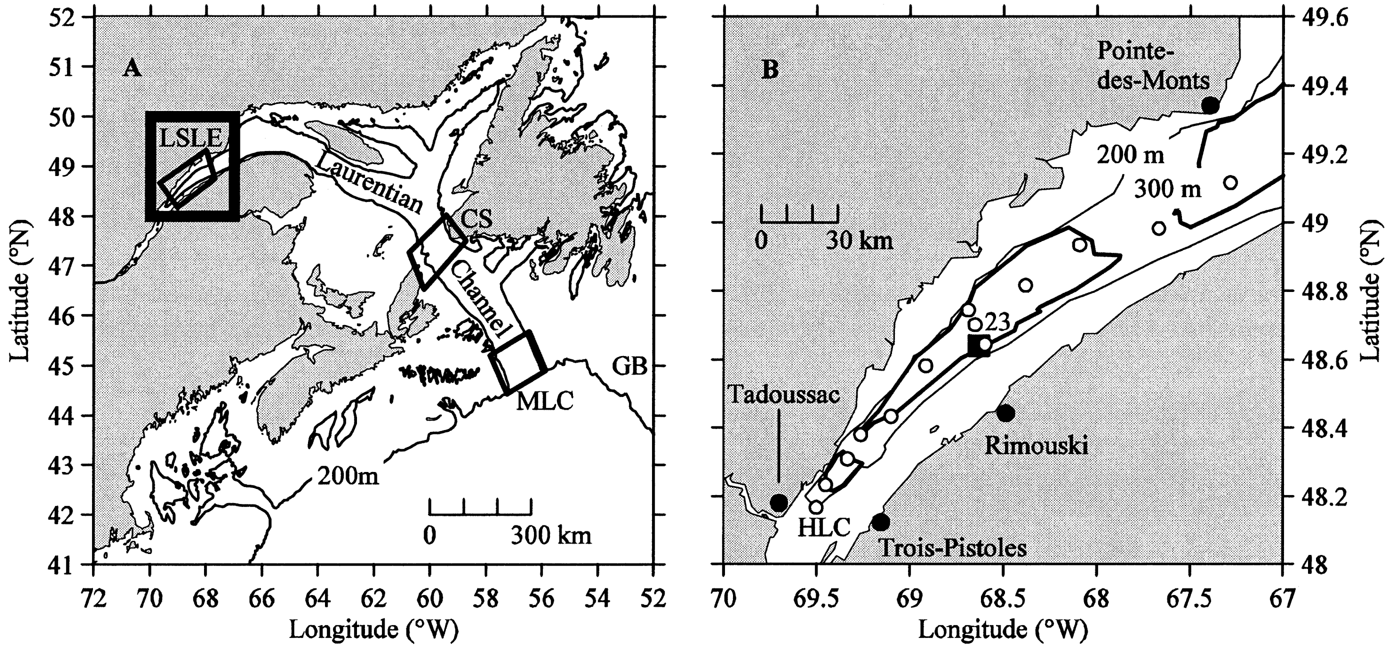

Fig. 1. (A) Map of the Gulf of St. Lawrence showing the 1240-km-long Laurentian Channel, as outlined by the 200-m isobath, and the location of its mouth (MLC) at the edge of the continental shelf to the west of the Grand Bank (GB) of Newfoundland. The thick square shows our main study area, whereas the thinner polygons show the MLC, Cabot Strait (CS), and Lower St. Lawrence estuary (LSLE) regions for which we constructed time series of temperature, salinity, density, and oxygen concentration–saturation. (B) Map of the Lower St. Lawrence estuary showing towns mentioned in the text, the head of the Laurentian Channel (HLC), Sta. 23 and the 12 other stations (open circles) at which we carried out Winkler oxygen titrations in July 2003, and the 200-m (thin line) and 300-m (thick line) isobaths. The filled black square offshore of Rimouski shows the site where a long piston core and a sediment box core were collected, in 1999 and 2000, respectively.

discarded because it was inconsistent with the local temperature–salinity relationship.

Water-column data analysis and statistics—The latitudes and longitudes of the corners of the polygons for which we derived oxygen, temperature, salinity, and density time series are given in Table 1. The LSLE polygon was designed to encompass only the central 300-m-deep basin of the estuary (Fig. 1B). We did this in order to avoid unwanted variance in the oxygen time series that might arise from spatial differences in the mean oxygen levels of the three 300-m-deep basins of the LSLE.

For the LSLE time series, where the maximum bottom depth is ~355 m, we retained all measurements from depths of 295 m or more. Over the 295–355-m depth range, the water density is nearly uniform due to the presence of a 290-m-deep sill that controls exchanges near the eastern end of the LSLE (Fig. 1B). In accordance with physical oceanographic convention, we subtracted 1,000 kg m^{-3} from all the reported potential densities (σ_θ). The long-term (1932–2003) mean potential density of the LSLE bottom waters is 27.27 kg m^{-3}, with close to 90% of the annual mean σ_θ values in the 27.10 to 27.45 kg m^{-3} range. Considering that water properties mix much more efficiently along isopycnals than across them, and considering that isopycnals in the top ~2,000 m of the ocean are found at depths differing by several hundreds of meters on either side of the Gulf Stream as a consequence of geostrophy and the tendency of surface currents to decrease with depth (Knauss 1978), we opted for an isopycnal-based rather than a depth-based analysis of the temperature–salinity–oxygen data. Vertical profiles of temperature, salinity, and oxygen at discrete depths were interpolated onto potential density surfaces by using the Matlab™ (The Mathworks Inc., Natick, Massachusetts) interp1 function with a shape-preserving, piecewise cubic Hermite interpolating polynomial (pchip) method that ensures continuous first derivatives at the data points while generally avoiding overshooting (Moler 2004).

We calculated the 1914–2003 climatological means and standard deviations of depth, temperature, salinity, oxygen concentration, and saturation on 21 equally spaced density levels from 26.75 kg m^{-3} to 27.75 kg m^{-3} from the annual mean values of these parameters. For convenience, we picked the 27.25 kg m^{-3} potential density surface for many of the analyses because it is very close to the long-term mean density of the bottom waters in the LSLE (27.27 kg m^{-3}) and corresponds nearly to the standard hydrographic depth of 250 m everywhere along the Laurentian Channel (Table 2), thereby reducing errors that might arise from vertical interpolation. Potential temperature and potential density were calculated from in situ temperature, salinity, and pressure using the Matlab™ seawater properties algorithms of Morgan (1994). Standard statistical tests were carried out with the Matlab™ statistics toolbox.

Table 2. Climatological means and standard deviations (1914–2003) of depth, temperature, salinity, and oxygen concentration and saturation on potential density surfaces for the polygons defined in Table 1. The number of years and the total number of profiles for which we have coincident temperature–salinity (T–S) data and coincident temperature–salinity–oxygen (T–S–O_2) data are indicated.

Density σ_θ (kg m^{-3})	Depth (m)	Temperature (°C)	Salinity	No. of years, No. of profiles (T–S)	O_2 (μmol L^{-1})	O_2 saturation (%)	No. of years, No. of profiles (T–S–O_2)
Lower St. Lawrence estuary							
27.00	187.5±28.5	3.44±0.53	33.95±0.06	35, 1083	104.9±25.6	32.2±7.3	16, 88
27.25	249.5±33.8	4.21±0.86	34.36±0.11	27, 441	74.6±26.3	23.4±7.6	10, 30
Cabot Strait							
27.00	190.3±19.0	4.26±0.96	34.06±0.12	61, 1322	201.4±20.8	62.7±6.1	16, 91
27.25	249.1±24.8	5.13±0.76	34.49±0.11	59, 1075	178.1±20.4	56.5±5.9	16, 80
27.50	351.6±28.8	4.93±0.46	34.77±0.07	55, 732	179.7±18.9	56.8±5.8	16, 48
Laurentian Channel mouth							
27.00	187.3±38.0	5.78±1.61	34.31±0.24	46, 460	225.7±19.9	70.8±4.9	8, 49
27.25	246.6±42.5	6.15±0.92	34.66±0.15	42, 395	199.2±19.1	63.4±4.4	8, 45
27.50	342.0±40.1	5.18±0.52	34.81±0.09	30, 285	214.2±15.6	67.8±4.2	8, 30
Labrador current water							
27.00	118.2±36.9	0.65±0.79	33.70±0.08	82, 2171	320.2±15.6	92.2±2.9	20, 101
27.10	136.5±38.2	1.05±0.82	33.85±0.08	81, 2052	311.6±18.6	90.5±3.2	20, 89
27.25	170.8±42.2	1.54±0.71	34.07±0.07	79, 1836	298.9±22.3	87.6±4.7	20, 79
27.50	263.5±46.7	2.66±0.51	34.49±0.06	77, 1466	289.0±22.8	86.8±5.5	18, 58
North Atlantic central water							
27.00	724.8±67.6	12.28±0.36	35.60±0.09	57, 1631	169.0±13.1	63.0±5.1	42, 807
27.10	772.7±64.4	11.14±0.38	35.45±0.09	57, 1617	164.2±13.1	59.7±4.8	42, 800
27.25	849.5±63.8	9.43±0.38	35.26±0.09	57, 1588	163.2±14.2	57.4±4.8	42, 795
27.50	985.9±62.5	7.00±0.46	35.11±0.10	57, 1492	189.4±13.7	62.9±4.4	42, 763

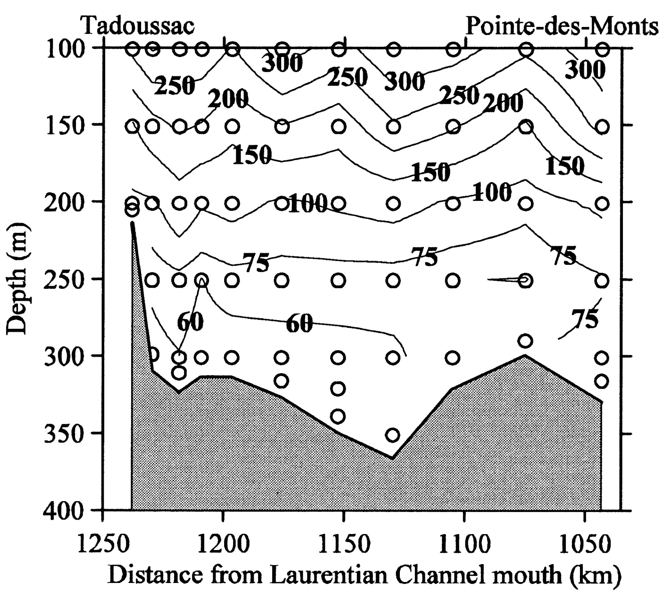

Fig. 2. Contours of oxygen concentration (μmol L^{-1}) in July 2003 in the Lower St. Lawrence estuary. The circles indicate the depths at which we performed Winkler titrations. Distance traveled from the mouth of the Laurentian Channel increases towards Tadoussac (Fig. 1).

Results

Areal extent of the hypoxic zone—In the along-channel direction, on a section from Pointe-des-Monts to Tadoussac (Fig. 1B), about 110 km of the seafloor had bottom waters with O_2 less than 60 μmol L^{-1} in July 2003 (Fig. 2). In the across-channel direction, O_2 levels were below 60 μmol L^{-1} at depths ≥300 m to the north and south of Sta. 23 in front of Rimouski (Fig. 1B). The lowest O_2 level measured during the July 2003 survey (51.2 μmol L^{-1} = 16.2% saturation at 5.21°C and a salinity of 34.53) was recorded at the station south of Sta. 23. Considering that the 60 μmol L^{-1} isoline intersected the bottom at an average depth of 275 m over most of the hypoxic zone (Fig. 2), we estimate that 1,300 ± 100 km² of the LSLE seafloor was bathed in hypoxic waters in July 2003.

Along-channel variations in oxygen—A broader perspective of along-channel variations in O_2 obtained from a CTD–O_2 section calibrated with Winkler titrations is presented in Fig. 3. Between 250-m and 300-m depth, O_2 levels drop progressively from more than 50% saturation at Cabot Strait to less than 20% saturation in the LSLE. At the mouth of the Laurentian Channel (i.e., at the edge of the continental shelf), O_2 levels are slightly higher than at Cabot Strait with a typical value of 65% saturation at 250-m depth (Table 2).

In Fig. 3B, we distinguish the presence of a middepth minimum in O_2 levels in the eastern half of the section. This

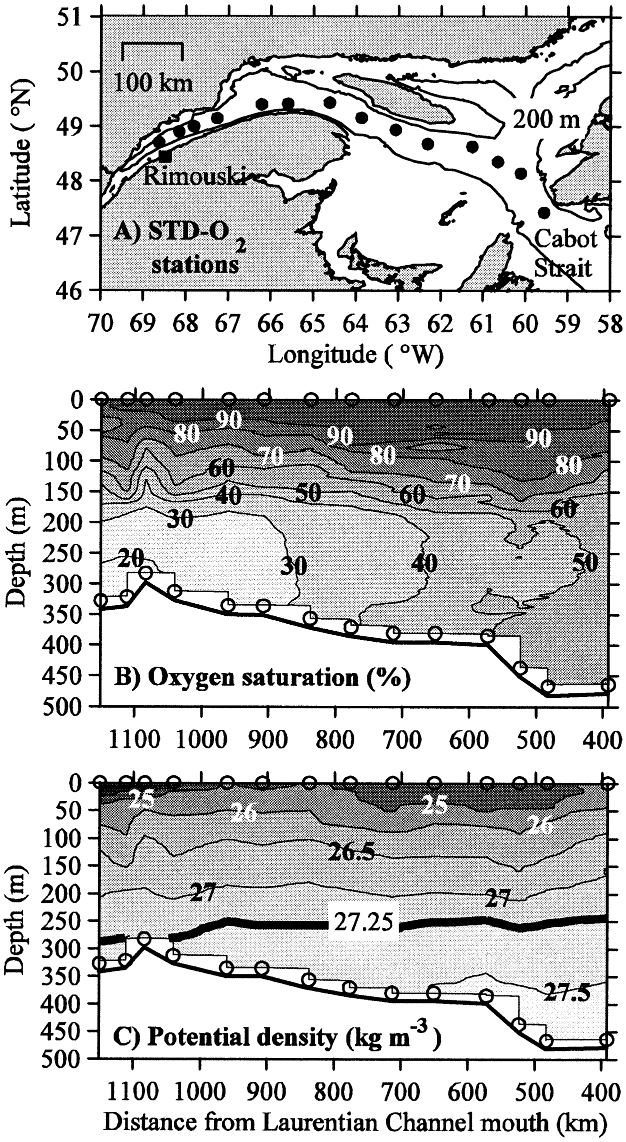

Fig. 3. (A) Map of the Gulf of St. Lawrence showing the positions of 14 stations where we collected vertical profiles of salinity, temperature, depth, and oxygen (STD-O_2) with a Seabird 911*plus* CTD equipped with a SBE-43 oxygen sensor in early November 2002. (B) Contours of oxygen saturation from Cabot Strait (right) to Rimouski (left). The circles indicate the locations of the STD-O_2 stations. (C) Contours of potential density highlighting the 27.25 kg m^{-3} density surface which roughly corresponds to the density of bottom waters in the LSLE.

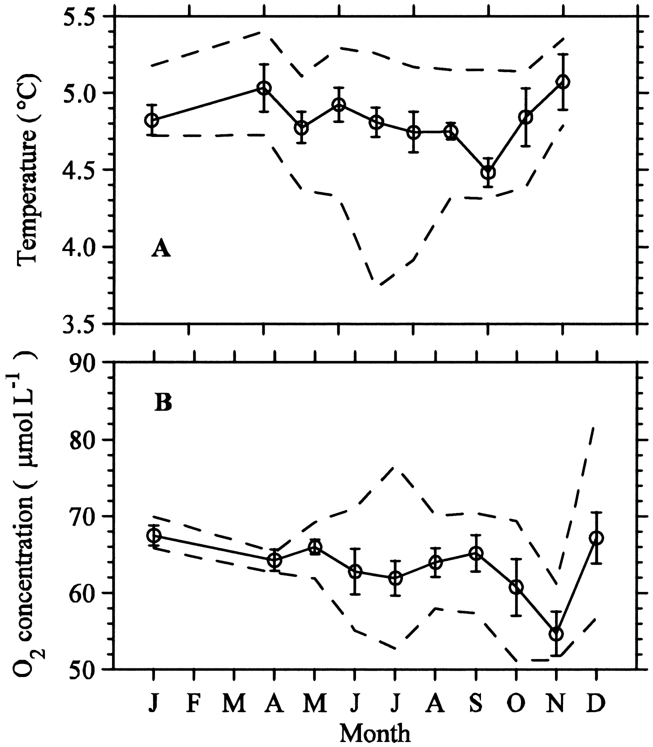

Fig. 4. Annual cycles of (A) temperature and (B) oxygen concentration in the bottom waters (≥295 m) of the LSLE for the 1984–2003 period of relatively stable temperature and oxygen conditions. The dashed lines represent the 10th and 90th percentiles of individual observations and the error bars show the 95% confidence intervals for the monthly means.

middepth O_2 minimum corresponds roughly to the 27.25 kg m^{-3} potential density surface (Fig. 3C) and serves as a tracer of the landward flow of the estuarine circulation at this density level.

Seasonal cycle of temperature and oxygen—The bottom waters of the LSLE remain isolated from the surface waters year-round as the maximum wintertime surface mixed layer density never exceeds 26.5 kg m^{-3} in the Gulf of St. Lawrence (Petrie et al. 1996), resulting in a maximum winter surface mixed layer depth of about 125 m (Fig. 3C). Near-bottom water temperatures are therefore stable throughout the year, without a detectable seasonal cycle (Fig. 4A). Likewise, oxygen concentrations in the bottom waters of the LSLE are essentially constant throughout the year: the largest difference between any two monthly means is on the order of 10 μmol L^{-1} (Fig. 4B). Such weak seasonal variations of temperature and oxygen in the bottom waters of the LSLE allow us to pool data from all months of the year to construct interannual time series.

Temporal change in oxygen and temperature—A 72-yr time series of O_2 in the bottom waters of the LSLE is presented in Fig. 5. Despite substantial interannual variability, three distinct clusters of points indicate that bottom water O_2 decreased from 110–135 μmol L^{-1} (33–41% saturation) in the 1930s to 95–120 μmol L^{-1} in the early 1970s and then to 55–85 μmol L^{-1} (18–26% saturation) in the 1990s. The annual mean bottom water O_2 concentration measured in 2003 (58.6 μmol L^{-1}) is the second lowest ever observed during the 28 yr for which measurements are available; the lowest annual mean value (56.4 μmol L^{-1}) was recorded in 1993. We also note that 8 of the 10 lowest annual mean values occurred in the last 12 yr. The difference in O_2 concentrations between the 1932–1935 period (124.7 μmol L^{-1} = 37.7% saturation) and the 1984–2003 period (65.5 μmol L^{-1} = 20.7% saturation) amounts to 59.2 ± 9.1 μmol L^{-1} at the 95% confidence level. A least squares linear fit applied

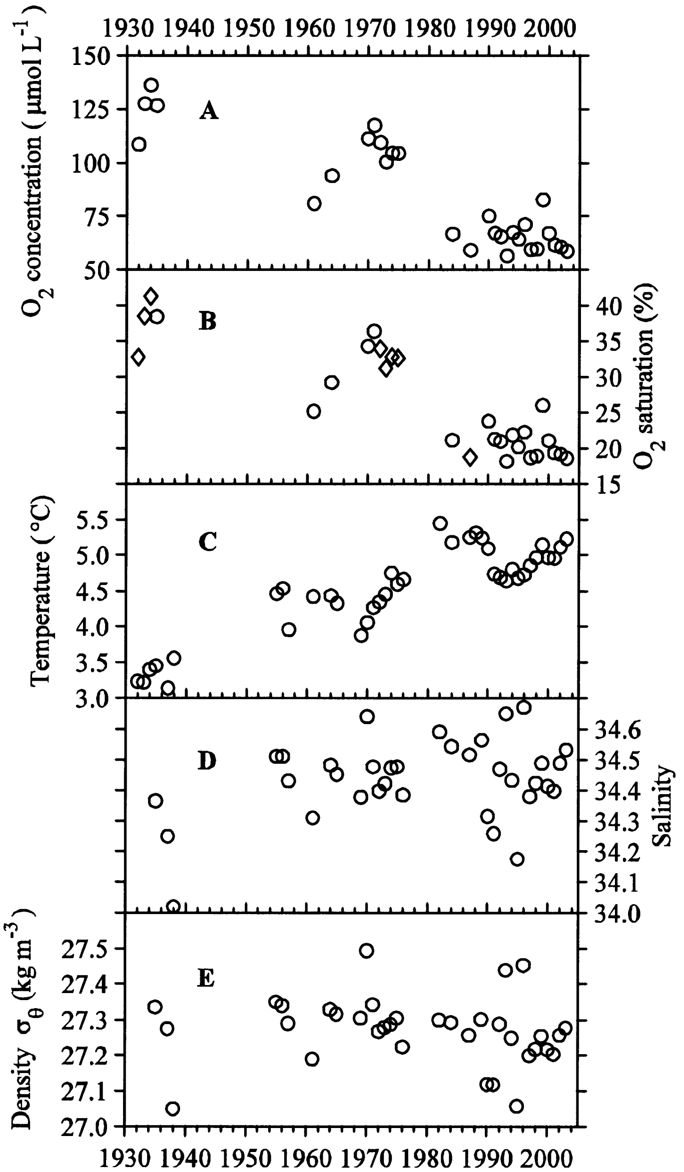

Fig. 5. Time series of (A) oxygen concentration, (B) oxygen saturation, (C) water temperature, (D) salinity, and (E) potential density between 295-m and 355-m depth in the Lower St. Lawrence estuary. Values of oxygen saturation estimated from nonsimultaneous measurements of oxygen, temperature, and salinity are indicated by diamonds in (B).

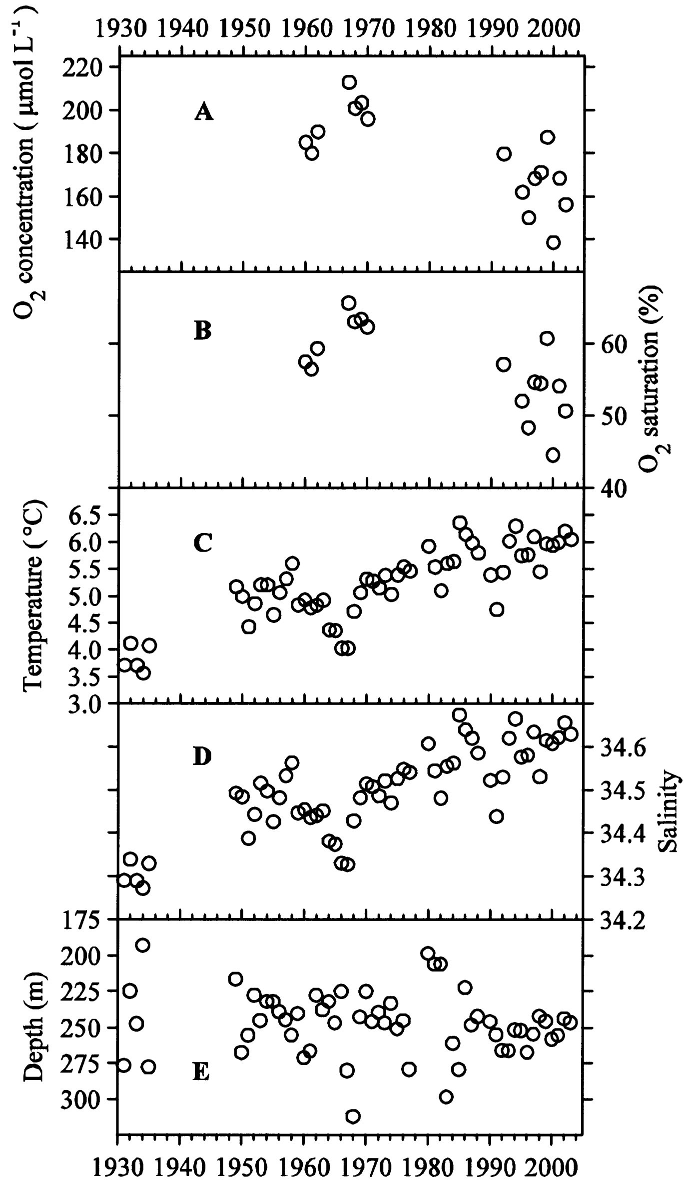

Fig. 6. Time series of (A) oxygen concentration, (B) oxygen saturation, (C) water temperature, (D) salinity, and (E) depth on the 27.25 kg m^{-3} potential density surface at Cabot Strait.

to the entire O_2 time series yields a negative slope (decreasing trend) of -1.0 ± 0.2 μmol L^{-1} yr^{-1} at the 95% confidence level. However, the oxygen regime appears to have stabilized since the mid-1980s as the slope over the 1984–2003 period is not different from zero (-0.1 ± 0.8 μmol L^{-1} yr^{-1} at the 95% confidence level).

The LSLE temperature time series calculated over the same depth range (295 m to the bottom) as the oxygen time series reveals an opposite trend (Fig. 5C). Over the duration of the time series (1932–2003), temperatures in the bottom waters of the LSLE have risen at the rate of 0.025 ± 0.005°C yr^{-1} at the 95% confidence level. The difference in temperature between the 1932–1935 period (3.33°C) and the 1984–2003 period (4.98°C) amounts to 1.65°C $\pm$ 0.25°C at the 95% confidence level. Like oxygen, the bottom water temperatures in the LSLE have remained stable since the mid-1980s as the trend calculated over the 1984–2003 period is not different from zero (-0.006 ± 0.021°C yr^{-1}).

Relationship between changes in temperature and oxygen—The opposite temporal trends of temperature and O_2 in the bottom waters of the LSLE (Fig. 5) suggest that part of the O_2 decline may have been caused by multidecadal changes in the physical and chemical properties of the bottom water mass. These changes do not originate in the estuary itself, but in the oceanic region adjacent to the mouth of the Laurentian Channel. The properties of this water mass are determined by mixing of other water masses present in this

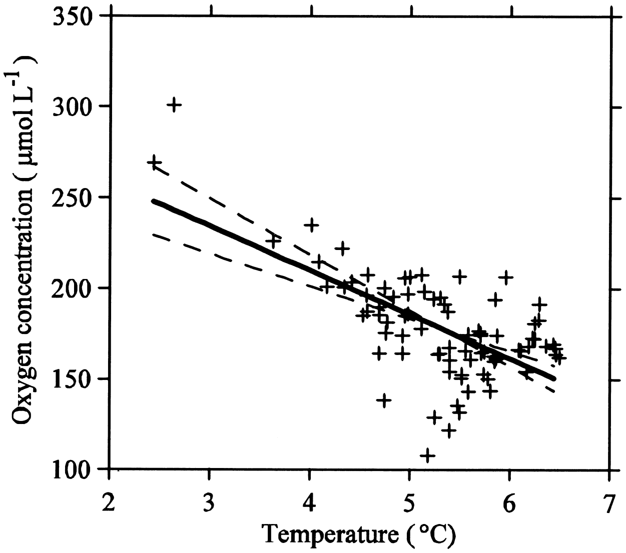

Fig. 7. Linear regression between water temperature and oxygen concentration on the 27.25 kg m^{-3} potential density surface from 80 individual vertical profiles collected between 1960 and 2002 in Cabot Strait. The best fit line, given by $O_2 = (-24.4\ \mu\text{mol L}^{-1}\ °\text{C}^{-1}) \times T + 308\ \mu\text{mol L}^{-1}$, is represented by the continuous line. The 95% confidence intervals for the slope ($\pm 6.5\ \mu$mol L^{-1} °C^{-1}) and intercept ($\pm 35\ \mu$mol L^{-1}) are shown as dashed lines.

region of the Northwest Atlantic Ocean and are therefore subjected to ocean climate variations.

To determine how much of the O_2 depletion in the estuary may be attributed to remote ocean climate variability, we focus on Cabot Strait where temperature, salinity, and oxygen data for the 27.25 kg m^{-3} isopycnal are relatively abundant (Table 2). Compared with the LSLE, the oxygen budget at Cabot Strait is much less likely to be affected by human-influenced factors such as the flux of terrigenous particulate organic matter from the St. Lawrence River (Lucotte et al. 1991). A more "pristine" temperature–oxygen relationship, less influenced by confounding factors, can thus be derived at Cabot Strait.

The LSLE warming trend from the 1930s to the 1980s–1990s was also observed at Cabot Strait (Fig. 6C). On the 27.25 kg m^{-3} isopycnal, temperature warmed by 1.95°C and salinity increased by 0.28 since the 1930s (Fig. 6D). There were no O_2 measurements from Cabot Strait in the 1930s (Fig. 6A,B), which excludes the possibility of a direct estimate of the effect of water mass changes on the declining oxygen levels observed in the LSLE. Nevertheless, over the 1960–2003 period, for which 16 yr have coincident temperature–salinity–oxygen data in Cabot Strait, high O_2 values are generally associated with cold and fresh conditions, whereas low O_2 values nearly all coincide with warm and salty conditions (Fig. 6). The relationship between O_2 and temperature, presented in Fig. 7, uses 1960–2003 data from 80 individual profiles with coincident temperature–salinity–oxygen measurements interpolated on the 27.25 kg m^{-3} isopycnal at Cabot Strait (Table 2). The slope of the least squares regression line is $-24.4 \pm 6.5\ \mu$mol L^{-1} °C^{-1} at the 95% confidence level, of which at most one-third ($-8.0\ \mu$mol L^{-1} °C^{-1}) can be attributed to the change in oxygen solubility over the range of temperatures and salinities covered by the regression (Garcia and Gordon 1992). Because Cabot Strait is close to the source of the deep Laurentian Channel water, we argue that the temperature–oxygen (T-O_2) relationship at this location reflects reasonably well the T-O_2 relationship at the mouth of the channel. We can therefore use the Cabot Strait T-O_2 relationship to estimate the change in the deep-water oxygen concentration in the LSLE that can be attributed to changes in ocean climate.

Multiplying the slope of the regression line in Fig. 7 by the 1.65°C warming observed at the LSLE from the 1930s to the 1984–2003 period gives $-40.3 \pm 10.7\ \mu$mol L^{-1} as our best estimate for the contribution of ocean climate change to the overall decline in oxygen concentration in the LSLE. Similar calculations for O_2 saturation yield a slope of -6.0 ± 2.0% O_2 saturation °C^{-1} at the 95% confidence level, so that the 1.65°C warming translates into a drop of 9.9 ± 3.3% O_2 saturation from the 1930s to the 1980s.

Temperature, salinity, and oxygen in the Northwest Atlantic—For insight into the oceanographic processes responsible for the inverse relation between O_2 and temperature on the 27.25 kg m^{-3} isopycnal, we turn to the Northwest Atlantic where the Gulf Stream and the Labrador Current meet (Fig. 8A). The depth of the 27.25 kg m^{-3} isopycnal (Fig. 8B) is relatively shallow (0–300 m deep) in the subpolar gyre, where surface waters are denser than in the subtropical gyre. The isopycnal takes a first dip downward at the Shelf/Slope sea surface temperature front, and then takes an additional dive of a few hundred meters across the Gulf Stream to depths of 700–1,000 m in the northern part of the subtropical gyre (Fig. 8B) in a water mass known as the North Atlantic Central Water (NACW; Pershing et al. 2001). The steep slope of the isopycnals across the Gulf Stream is responsible for the decreased speed of the Gulf Stream with depth (Margule's equation; Knauss 1978). On the 27.25 kg m^{-3} isopycnal, the contrasts in temperature, salinity, and oxygen properties of NACW and LCW are striking (Fig. 8). Temperature differences are as large as 10°C (Fig. 8C), salinities differ by about 1.5 (Fig. 8D), and O_2 differences of more than 100 μmol L^{-1} (Fig. 8E) and 30% saturation (Fig. 8F) are observed. The progressive landward depletion of O_2 along the three deep channels of the Gulf of St. Lawrence is clearly seen on Fig. 8E,F.

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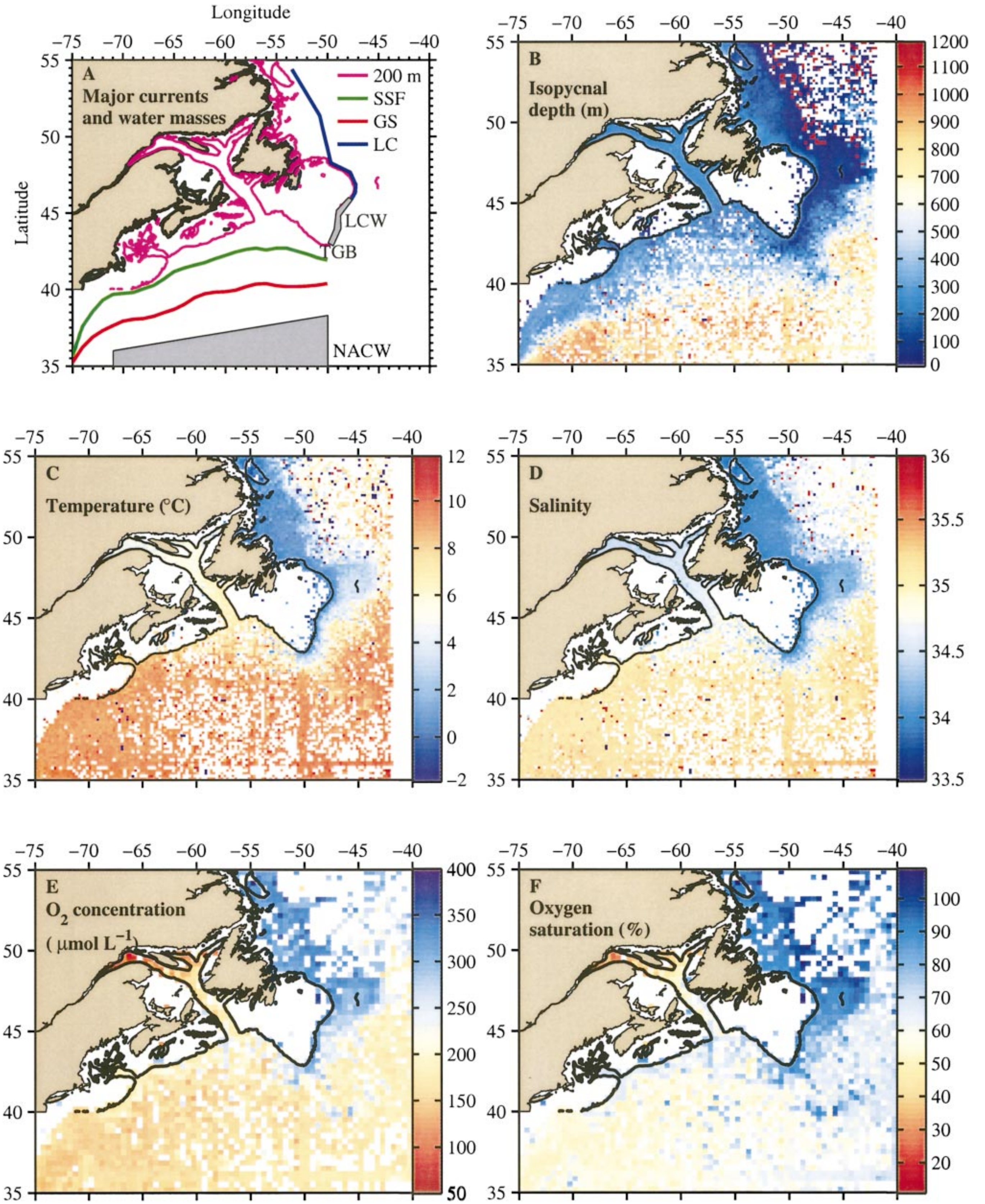
Fig. 8. (A) Map of the northwest Atlantic showing the 1973–2001 mean position of the northern edge of the Gulf Stream (red line) and the Shelf/Slope front (green line), the position of the Labrador Current (blue line), the Tail of the Grand Banks (TGB), and two polygons (LCW, Labrador Current Water; NACW, North Atlantic Central Water) for which we constructed long-term mean vertical profiles of temperature, salinity, and oxygen. Projections onto the 27.25 kg m^{-3} potential density surface of the 1914–2003 median values of (B)



depth, (C) temperature, and (D) salinity are presented for each ⅙° latitude × ¼° longitude grid square with data from the CLIMATE database. The 1914–2003 median values of (E) oxygen concentration and (F) saturation are presented for each ⅓° latitude × ½° longitude grid square with data from the BIOCHEM database.

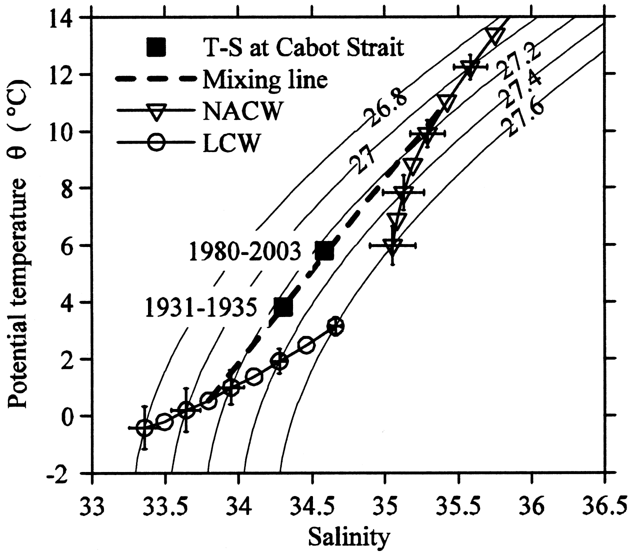


Fig. 9. Temperature–salinity (T-S) diagram of the two parent water masses, Labrador Current Water (LCW) and North Atlantic Central Water (NACW). On each water-mass T-S curve, we superimpose the interannual standard deviations of temperature and salinity at potential density intervals of 0.20 kg m^{-3}. The dashed line represents a T-S mixing line joining LCW and NACW source water types at the initial potential density of 27.10 kg m^{-3}.

Temperature–salinity diagram analysis—A temperature–salinity (T-S) diagram of the NACW and LCW water masses is presented in Fig. 9. As a result of cabbeling, mixing of NACW and LCW parent water types with a density of 27.10 kg m^{-3} produces a water mass with a density of about 27.27 kg m^{-3}, in the range of temperatures and salinities found along the T-S mixing line at Cabot Strait (Fig. 9). As this density (27.27 kg m^{-3}) equals the long-term mean density of bottom waters in the LSLE, we choose this particular T-S mixing line to estimate the likely impact of a 1.65°C warming from the 1930s to the 1984–2003 period on O_2 concentration and saturation.

Given the 10.09°C difference between NACW and LCW on the 27.10 kg m^{-3} isopycnal (Table 2), a 1.65°C warming implies that the proportion of LCW in the bottom waters of the LSLE declined by 16.4% from the 1930s to 1984–2003. To translate this into a change in O_2, we neglect biogeochemical processes and assume that oxygen mixes conservatively. Under this assumption, and given the long-term mean differences of 147.4 μmol L^{-1} O_2 concentration and 30.8% O_2 saturation between the NACW and LCW water types on the 27.10 kg m^{-3} isopycnal (Table 2; Fig. 10), a 16.4% decline in the proportion of LCW would account for a decrease of 24.2 μmol L^{-1} in O_2 concentration and 5.1% in O_2 saturation. If we further assume that the properties of the NACW and LCW vary independently, the same calculations using the long-term mean O_2 levels ± 1 standard deviation (STD, Table 2) give a measure of the uncertainty (i.e., $\mathrm{STD}(O_2) = (18.6^2 + 13.1^2)^{1/2}/2^{1/2} = 16.1$ μmol L^{-1}, and $\mathrm{STD}(O_2 \text{ saturation}) = (3.2^2 + 4.8^2)^{1/2}/2^{1/2} = 4.1\%$). We

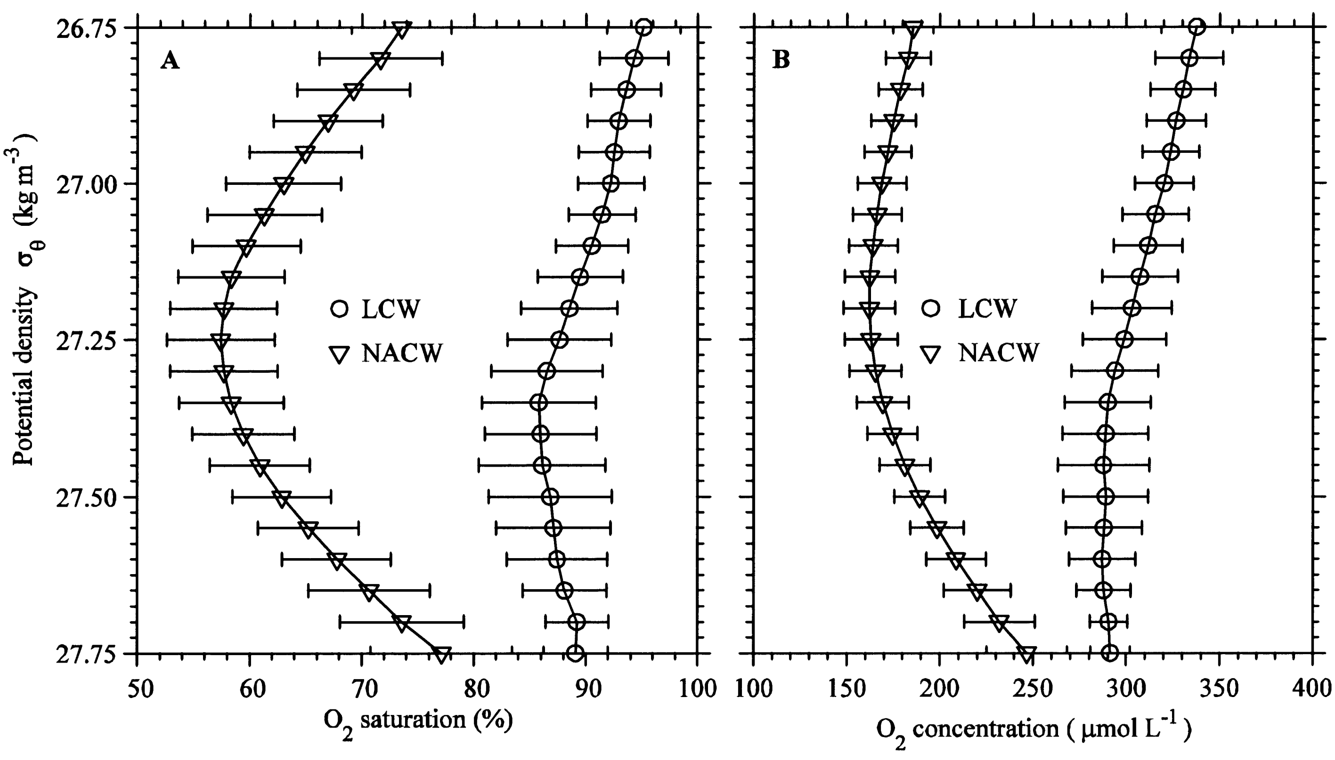


Fig. 10. (A) Oxygen saturation and (B) concentration as a function of potential density for Labrador Current Water (LCW) and North Atlantic Central Water (NACW). The error bars represent ±1 standard deviation from the mean.

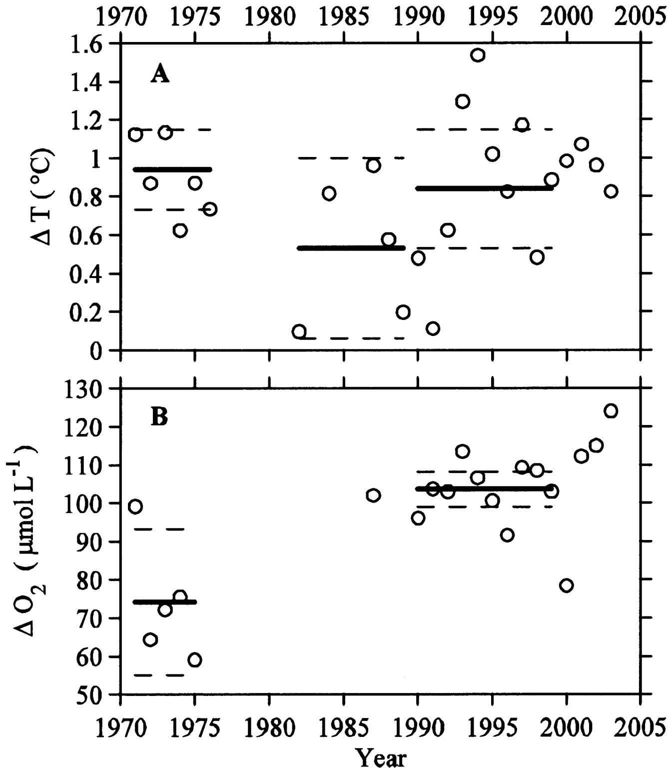


Fig. 11. Difference in (A) temperature and (B) oxygen between Cabot Strait (250-m depth) and the bottom waters of the LSLE. For each decade, we show the decadal mean value (thick line) and its 95% confidence interval (dashed lines).

thus obtain 24.2 ± 16.1 μmol L^{-1} and 5.1 ± 4.1% for the declines in O_2 concentration and O_2 saturation associated with the 1.65°C warming. Given the uncertainties, these estimates are not different from what we obtained with the linear regression analysis of the Cabot Strait temperature and O_2 data. As the differences in O_2 concentration and saturation of the LCW and NACW source waters do not vary much over the 27.0–27.25 kg m^{-3} σ_θ range (Fig. 10; Table 2), a choice of parent source water types with density other than 27.10 kg m^{-3} would have only minor effects on the calculations.

Proportions of parent water types in the mixed water type—A fundamental part of water mass analysis involves determining the proportions of each parent water type in the mixed water type (Tomczak 1999). We perform this calculation for Cabot Strait where salinity data from the 1930s are more abundant (Table 2; Fig. 6) and of better quality than in the LSLE. For the 27.25 kg m^{-3} potential density surface we conclude that in the 1930s, waters with T-S values of 3.84°C and 34.304 (Fig. 9) originated from a mixture of 72% LCW and 28% NACW. Over the 1980–2003 period, waters at the same density were 1.95°C warmer and 0.28 saltier and originated from a mixture of 53% LCW and 47% NACW.

These calculations assume constant T-S values for the parent water types. Over the entire 1930–1939 decade, average T-S properties of LCW and NACW on the 27.10 kg m^{-3} isopycnal were the same as those reported in Table 2 to within 0.01°C and 0.01. Over the 1980–2003 period, the average T-S properties of LCW (0.98°C, 33.84) and NACW (11.10°C, 35.44) did not differ from the climatological mean values of Table 2 by more than 0.07°C and 0.01. Therefore, our estimates of the proportions of LCW and NACW on the 27.25 kg m^{-3} isopycnal at Cabot Strait in the 1930s and over the 1980–2003 period are essentially unaffected by interannual variability of the parent water mass properties.

Bugden (1988) performed similar calculations and concluded that the proportion of LCW at Cabot Strait was 66% in a cold year (1966) and 43% in a warm year (1985) for parent water types with a density of 27.10 kg m^{-3}. His definition of LCW comprised waters from the Labrador Shelf and Slope, about 1,000 km to the north of the LCW polygon used in the present study (Table 1), which are about 0.8°C colder and 0.07 fresher. Using our definition of LCW, his estimates would have been 73% LCW for the cold 1966 and 48% LCW for the warm 1985, very close to our own estimates based on a larger data set and wider time windows.

Discussion

Several factors may have contributed to the decrease of the oxygen concentration in the bottom waters of the Lower St. Lawrence estuary since the 1930s. These include (1) a change in water mass properties at the mouth of the Laurentian Channel, (2) an increase in water column and/or sediment oxygen demand, and (3) a decrease in the landward advection velocity of the bottom water with consequent longer residence time in the Laurentian Channel. A lagged correlation analysis of temperature time series from several locations along the Laurentian Channel (Gilbert 2004) suggests that over the last decades, the mean landward advection speed of waters at 250-m depth did not undergo changes that would significantly affect the oxygen budget of the bottom waters in the LSLE (Benoit et al. in press). Hence we only address the first two factors.

Ocean climate variability—The relationship between temperature and O_2 on the 27.25 kg m^{-3} isopycnal at Cabot Strait suggests that the 1.65°C temperature increase in the bottom waters of the LSLE over the last 72 yr is associated with an oxygen loss of 40.3 ± 10.7 μmol L^{-1}, leaving unexplained 8–30 μmol L^{-1} of the observed oxygen decrease (59.2 μmol L^{-1}). Alternatively, a T-S diagram analysis, together with the assumption that oxygen mixes conservatively, suggests that changes in the proportions of LCW and NACW in the water entering the Laurentian Channel were associated with an oxygen loss of 24.2 ± 16.1 μmol L^{-1}, leaving unexplained 19–51 μmol L^{-1} of the oxygen decrease. In spite of the uncertainties in these estimates, it appears that changes in ocean climate have had a profound effect on the oxygen regime in the LSLE. Objective maps of pentadally averaged thermal anomalies from bathythermograph data show that, between 100-m and 500-m depth, the Northwest Atlantic Slope Water region was characterized by mostly below-normal temperatures from 1956 to 1970 and by mostly above-normal temperatures from 1970 to

1994 (Grey et al. 2000). But what drives this ocean climate variability?

Petrie and Drinkwater (1993) have shown that the westward transport of LCW along the continental shelf edge to the south of the Grand Banks of Newfoundland (Fig. 8A) can vary by about a factor of 4 and that it is the most important factor driving interannual variability of temperature and salinity in the Laurentian Channel as well as in the deep basins of the Scotian Shelf and the Gulf of Maine. They demonstrated that despite slightly warmer and saltier than normal LCW properties in the 1960s, the mid-1960s were characterized by much colder than normal water temperatures in the 100-m to 300-m depth range in the Laurentian Channel, Emerald Basin, and Gulf of Maine. Based on geostrophic transport calculations, they estimated that the volume transport of the Labrador Current in the 1960s was larger than in the 1970s by $\sim$1–4 $\times$ 10^6 m^3 s^{-1}. This finding was further substantiated by the modeling study of Loder et al. (2001), who showed that the cold conditions of the mid-1960s were linked to a 1–2 $\times$ 10^6 m^3 s^{-1} increase in the westward shelf-edge flow of LCW during the winter.

Another possible driver of interdecadal climate variability involves large-scale shifts in atmospheric pressure systems and wind patterns, as exemplified by the North Atlantic Oscillation (NAO) index derived from the sea level pressure difference between Lisbon, Portugal, and Stykkisholmur/Reykjavik, Iceland (Hurrell 1995). However, taking into account the effects of serial correlation (Ebisuzaki 1997), the correlations between the NAO index and temperature time series on the 27.25 kg m^{-3} isopycnal at the three polygons along the Laurentian Channel (Fig. 1) were not statistically different from zero. Other attempts to link the NAO with temperature indices between 100-m and 300-m depth further west along the edge of the Scotian Shelf have also been unsuccessful (Pershing et al. 2001).

Excursions in the north–south direction of both the Shelf/Slope sea surface temperature (SST) front and the northern edge of the Gulf Stream (Fig. 8A) may also influence water properties in the Laurentian Channel, as suggested by Bugden (1988). Gilbert (2004) computed lagged correlations with the temperature time series at the mouth of the Laurentian Channel on the 27.25 kg m^{-3} density surface using 1973 to 2001 time series of the latitudinal position of these two SST fronts at intervals of 1° longitude from 75°W to 50°W (Drinkwater et al. 1994). For lags of 0 yr to 4 yr and at longitudes between 65°W and 50°W, he found a dominant pattern of weak ($r < 0.5$) but positive correlations between the latitudinal position of the northern edge of the Gulf Stream and temperature time series from the mouth of the Laurentian Channel. This could imply that warm, salty, oxygen-poor Gulf Stream waters have a higher likelihood of mixing with the Slope and Shelf Waters to the north when the Gulf Stream moves further north than its normal position (Fig. 8A). This would promote conditions that generate water at the mouth of the Laurentian Channel that is warmer, saltier, and more oxygen depleted than usual.

Oxygen variation not related to ocean climate variability—A comparison between changes over time of the along-channel gradients of temperature and oxygen concentration reveals that recent variations in oxygen are not entirely driven by ocean climate variability. Figure 11A shows that the difference in temperature between waters at 250 $\pm$ 10-m depth at Cabot Strait and the bottom waters of the LSLE was stable from the 1970s (0.94 $\pm$ 0.21°C) to the 1990s (0.84 $\pm$ 0.31°C). The along-channel temperature gradient ($\sim$0.9°C 800 km^{-1} = 1.13 $\times$ 10^{-5}°C km^{-1}) on the 27.25 kg m^{-3} isopycnal results from vertical mixing of colder overlying waters during the landward estuarine flow and is density compensated by a salinity gradient of the same sign as the temperature gradient (Bugden 1988).

In contrast, the corresponding along-channel O_2 gradient increased from 74.1 $\pm$ 19.1 μmol L^{-1} in the 1970s to 103.6 $\pm$ 4.6 μmol L^{-1} in the 1990s. The difference is 29.5 $\pm$ 11.9 μmol L^{-1} at the 95% confidence level. The fact that the along-channel gradient of O_2 increased while the along-channel gradient of temperature remained unchanged suggests that perhaps as much as 18–41 μmol L^{-1} of the O_2 decline may be attributable to factors other than ocean climate variability. This result is consistent with our earlier result that changes in ocean climate could not explain between 8 μmol L^{-1} and 51 μmol L^{-1} of the observed oxygen decrease. What other factor(s) could be responsible for the remaining decline in O_2? The answer may reside in the sediment record.

Changes in organic matter fluxes and sediment oxygen demand—A composite profile of two sediment cores recovered at a station situated across from Rimouski in the LSLE (Fig. 1B) revealed that the organic carbon (C_{ORG}) content of the sediment increased following European settlement in the area around the 17th century (St-Onge et al. 2003). Concomitantly, the C_{ORG} : N ratio of the preserved sedimentary organic carbon increased from $\sim$8 to 14 (A. Mucci unpubl. data), and the $\delta^{13}C$ ratio (St-Onge et al. 2003, Background Dataset, Fig. 1) decreased from −24.1‰ in the 1600s to −24.7‰ around 1970, as a greater proportion of terrigenous organic matter was accumulating in the sediment (Lucotte et al. 1991). Deforestation and accompanying soil erosion, which began in the 17th century and continues to this day, is a potentially important source of terrigenous organic carbon to the LSLE system. Other possible sources of terrigenous organic matter include municipal sewage and pulp and paper mill wastewaters. Since about 1970, the carbon isotopic trend has reversed, and the $\delta^{13}C$ values have increased to −24.3‰ (St-Onge et al. 2003). The recent $\delta^{13}C$ increase suggests that the proportion of marine organic matter delivered to the sediment may have become more important. The shift in $\delta^{13}C$ is accompanied by a significant increase in the abundance of dinocysts and organic linings of benthic foraminifera (B. Thibodeau et al. pers. comm.). Whereas the former could be interpreted as a proxy of increased primary production in the surface waters, the latter may reflect a concomitant increase of benthic production (B. Thibodeau et al. pers. comm.). These preliminary results are consistent and suggest strongly that an increase in primary production, possibly in relation to coastal eutrophication (Cloern 2001), may have taken place over the last two to three decades in the St. Lawrence estuary.

Irrespective of source, any increase in the delivery of metabolizable organic matter to the deep waters and sediments of the Laurentian Channel will translate into a greater oxygen demand. The results of a numerical model that couples water transport with a diagenetic model of oxic organic matter mineralization in the sediment reveal that, for realistic values of mean advection velocity and vertical and horizontal eddy diffusivities, the present flux of organic carbon to the sediment is sufficient to generate hypoxic waters in the bottom waters of the LSLE (Benoit et al. in press). Given the warming trends presented here, future diagenetic model studies could examine the possibility that higher bottom water temperatures may cause greater sediment oxygen demand due to higher bacterial decomposition rates.

Oxygen in the bottom waters of the LSLE declined by 60 μmol L^{-1} between the 1930s and the 1990s. Over the same period of time, the temperature of the bottom waters warmed by 1.65°C. A linear regression analysis of the T-O_2 relationship suggests that this warming is associated with 30–50 μmol L^{-1} of the oxygen decrease. Alternatively, a T-S-O_2 diagram analysis suggests that a reduction in the proportion of LCW in the bottom waters of the LSLE explains 8–40 μmol L^{-1} of the oxygen decrease. We therefore conclude that about one half to two thirds of the observed historical oxygen loss in the LSLE can be attributed to a decreasing proportion of oxygen-rich LCW in the water mass entering the Laurentian Channel, leaving between one third and one half of the oxygen decrease unexplained.

Further evidence that factors other than changes in water mass composition contributed to the oxygen decline comes from an analysis of the temporal evolution of along-channel gradients of temperature and oxygen in the Laurentian Channel. Whereas the temperature difference between the bottom waters of the LSLE and waters at 250-m depth in Cabot Strait did not change from the 1970s to the 1990s, the oxygen gradient between these two locations increased by 29.5 $\pm$ 11.9 μmol L^{-1} over the same period. Preliminary data from the sediment suggest that increases in sediment oxygen demand in the estuary may be partly responsible for this change in along-channel oxygen gradient.

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