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Current concentrations and spatial and temporal trends in mercury in Great Lakes Herring Gull eggs, 1974–2009

D. V. Chip Weseloh · David J. Moore · Craig E. Hebert · Shane R. de Solla · Birgit M. Braune · Daryl J. McGoldrick

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Abstract Current concentrations and spatial and temporal trends of total mercury (Hg) were assessed in eggs of the Herring Gull (*Larus argentatus*) over the period 1974–2009 at 15 sites in the Great Lakes: 2–3 sites per lake and one site in each of 3 connecting channels. Current (2009) concentrations ranged from 0.064 μg/g (wet weight) at Chantry Island (Lake Huron) to 0.246 μg/g at Middle Island (Lake Erie). There were significant inter-colony differences in mean Hg concentrations (2005–2009). Mercury concentrations at 14 of 15 sites declined from 23 to 86% between when it was first measured (usually 1974) and 2009. Declining temporal trends over the entire period (1974–2009) were significant at 10 of the 15 sites. On the other hand, there were no significant trends in mercury over the last 15 years. In the early years, declines of Hg in Herring Gull eggs tracked those in Rainbow Smelt (*Osmerus mordax*) in most Great Lakes. More recently, declines in gull eggs were more evident than in smelt and may be partially explained by temporal changes in the gull diet. When gull Hg data were adjusted for temporal changes in the gull diet, as inferred from stable nitrogen isotope values in eggs, significant declines in egg mercury levels were found only at 4 of 15 sites. Overall, Hg concentrations have declined in Great Lakes Herring Gull eggs over the period 1974–2009 but changes in the gull diet may be contributing, in part, to those declines. Examination of contaminant temporal trends in multiple indicator species will ensure accurate inferences regarding contaminant availability in the environment.

Keywords Monitoring · Great Lakes · Herring Gull · Mercury

D. V. C. Weseloh (✉)
Canadian Wildlife Service, Environment Canada, 4905 Dufferin Street, Toronto, ON M3H 5T4, Canada
e-mail: chip.weseloh@ec.gc.ca

D. J. Moore
Canadian Wildlife Service, Environment Canada, Canada Centre for Inland Waters, Box 5050, Burlington, ON L7R 4A6, Canada

C. E. Hebert · B. M. Braune
Wildlife and Landscape Science Directorate, National Wildlife Research Centre, Environment Canada, Carleton University, Raven Road, Ottawa, ON K1A 0H3, Canada

S. R. de Solla
Wildlife and Landscape Science Directorate, Environment Canada, Canada Centre for Inland Waters, Box 5050, Burlington, ON L7R 4A6, Canada

D. J. McGoldrick
Water Science and Technology Directorate, Environment Canada, Canada Centre for Inland Waters, Box 5050, Burlington, ON L7R 4A6, Canada

Introduction

Mercury (Hg) has long been known as a serious contaminant for wildlife. There are numerous anthropogenic sources of mercury, such as metal smelting and production, chlor-alkali and pulp industries, waste treatment, and the burning of wood and fossil fuels (Morel et al. 1998). Mercury can cause brain lesions, spinal cord degeneration, and central nervous system dysfunctions (Wolfe et al. 1998). Toxicity can be manifested as tremors, posture changes, uneven gait, impaired reproductive performance (Spalding et al. 2000; Evers et al. 2007), increased mortality of embryos, altered pairing behaviour and reduced clutch size and reproductive success (Stoewsand et al. 1971; Heinz 1979; Frederick and Jayasence 2011).

Methylmercury and other forms of organic mercury are substantially more toxic and bioaccumulative than the inorganic forms of mercury (Scheuhammer 1987). While organic mercury tends to sequester in certain organs, such as the liver and kidneys (Boening 2000; Wolfe et al. 1998), it is maternally transferred to eggs. Approximately 90% of total mercury in the eggs of piscivorous birds consists of methylmercury (Scheuhammer et al. 2001).

In the Great Lakes, and world-wide, mercury is a contaminant of concern which has received intensive study (Evers et al. 1998; Mackay and Toose 2004; Weis 2004; French et al. 2006; Braune 2007; Munthe et al. 2007; Monson 2009). Anthropogenic emissions of mercury in North America increased rapidly from the 1900s to the 1970s, with approximately 1/3 released in the Great Lakes region (Pirrone et al. 1998). Spatial patterns of mercury concentrations in surficial sediments in the Great Lakes have been identified (Marvin et al. 2004). Numerous studies of Hg in fish have been carried out on the Great Lakes (e.g. Goulet et al. 2007; Hogan et al. 2007; Bhavsar et al. 2010; Gewurtz et al. 2010). These studies are relevant to Herring Gull research as the main route of exposure is through its diet (Scheuhammer et al. 2007).

Piscivorous birds tend to have relatively high mercury exposure compared to other vertebrates; body burdens tend to increase with trophic level in freshwater birds (Zillioux et al. 1993). Hence, diet composition is an important factor regulating Hg accumulation in wildlife. In Herring Gulls, resources used for egg formation are primarily exogenous (Hobson et al. 1997); therefore, egg chemical composition will reflect the gull diet over several weeks during the period of egg formation. Insights into what the female was eating during this period can be gained by measuring ecological tracers (e.g. stable nitrogen and carbon isotopes and fatty acids) in eggs (Hebert et al. 2006). From 1982 to 2004, Herring Gulls in the Great Lakes shifted their diets, and may be relying on terrestrial foods to a greater extent than in the past (Hebert et al. 2008, 2009). This will generally result in reduced exposure to biomagnifying contaminants (e.g. PCBs, Hg) because birds are consuming lower trophic level prey (Hebert and Weseloh 2006; Akearok et al. 2010). Recognizing these changes underscores the importance of considering how changes in diet composition may influence the interpretation of temporal trends for other biomagnifying contaminants such as Hg.

The Great Lakes Binational Toxics Strategy was developed jointly by Canada and the USA in 1996–1997 with the goal of 50–90% reductions in the deliberate use and/or release of mercury from anthropogenic sources by 2006 (IJC 1988). Data from biomonitoring studies allow an assessment of the level of success in achieving reduction targets. In this study, current (2009) concentrations and spatial and temporal trends in mercury in eggs of Great Lakes Herring Gulls over the period 1974–2009 are presented. Also, the impact of dietary change in Herring Gulls on temporal trends in mercury in eggs is examined.

Methods

Herring Gull eggs have become a standard medium for monitoring contaminant levels in wildlife in the Laurentian Great Lakes (Mineau et al. 1984; Pekarik and Weseloh 1998; Hebert et al. 1999a; Weseloh et al. 2006). Koster et al. (1996) summarized mercury levels from the Great Lakes Herring Gull Monitoring Program up to and including 1992.

Field collections

Fresh Herring Gull eggs were collected annually at up to 15 colony sites throughout the Great Lakes and connecting channels, 1974–2009. The water bodies and sites are shown in Fig. 1. From 10 to 13 eggs were collected at each site usually during the last week of April and the first week of May; one egg was taken from each completed 3-egg clutch. Hg concentrations tend to be highest in the first egg laid and decrease with subsequent eggs (Akearok et al. 2010); since eggs were sampled randomly any bias due to egg laying order was minimized. Occasionally, if there were not enough completed clutches, an egg was taken from a 1- or 2-egg clutch. Eggs were stored cool within 2–3 days of collection. Within 3 weeks, they were transferred to Environment Canada's National Wildlife Research Centre (NWRC) in Ottawa for analysis.

Chemical analysis

Although Herring Gull eggs were available from nearly all sites for the period 1974–2009, mercury was not analyzed in all years, either originally or retrospectively. Mercury values were obtained for a minimum of 19 years and a maximum of 22 years at each site (mean = 21 years) for this study (see Appendix). Up to and including 1985, all eggs were analyzed individually; from 1986 and onwards, all eggs were analyzed as site pools. For temporal trend analysis, mean values were calculated for the years 1985 and earlier. Mercury values from a total of 309 site-years were used in this study.

For the years 1974 to 1989, eggs were analyzed by the Ontario Research Foundation (ORF; Mississauga, Ontario). Approximately 0.5 g of each gull egg homogenate was

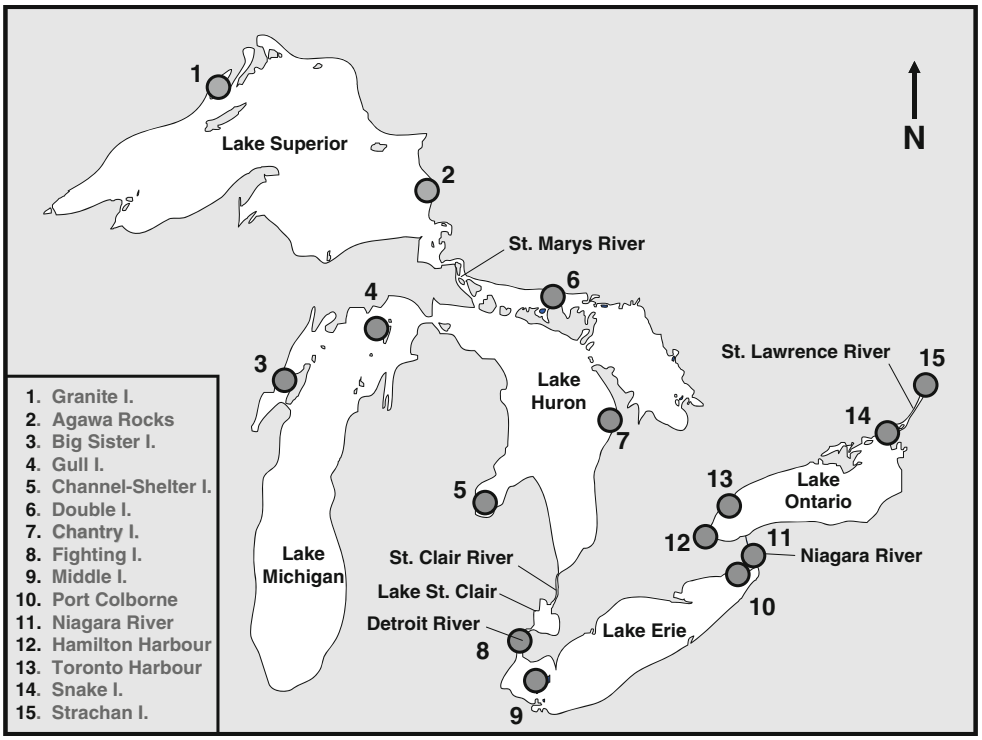

Fig. 1 Herring Gull annual monitoring colonies in the Great Lakes and connecting channels, 1974–2009. The *inset* gives the names of numbered sites from the map

dried and acid-digested. Samples were analyzed by cold vapour atomic absorption spectrophotometry (CVAAS).

From 1990 to 2001, eggs were analyzed at the NWRC using CVAAS. Approximately 0.5 g of egg homogenate was transferred into a pre-weighed acid-washed test tube and freeze dried for at least 24 h until constant weight was obtained. Dried samples were digested and analyzed for total Hg by continuous-flow CVAAS using a Perkin Elmer 3030b spectrophotometer, as described by Scheuhammer and Bond (1991).

From 2002 onward, eggs were analyzed at NWRC using an advanced mercury analyzer (AMA-254), equipped with an ASS-254 auto-sampler for solid samples. Approximately 0.5 g of egg homogenate was freeze dried, and carried by flowing oxygen to an amalgamator that selectively trapped mercury. Following flushing by O_2, the amalgamator was heated rapidly releasing mercury vapour which was carried through absorbance cells positioned in the light path of an atomic absorption spectrometer (253.7 nm). The AMA-254 software calculated the concentration of mercury in the sample based on a calibration curve and sample mass.

For all years, duplicates were analyzed to check the repeatability of Hg analyses. The nominal detection limit for total Hg was 0.05 μg/g (dry weight) for both AMA and CVAAS. Standard Reference Materials or in-house reference materials also were analyzed concurrently with Herring Gull eggs; prior to 1990 in-house Herring Gull egg reference materials were analyzed (Turle and Collins 1992), whereas after 1989 DOLT-3, TORT-2 (National Research Council of Canada) and/or 1566b oyster tissue (National Institute of Standards and Technology Standard Reference Material) were analyzed. Results are reported in μg/g wet weight. For each egg, percent moisture was recorded during the initial, post-collection preparation of samples at NWRC. There were no significant temporal trends for mean annual moisture content (%) at any of the colony sites ($p > 0.12$ in all cases, data not shown).

The comparability of the data obtained from NWRC using the two methods, CVAAS and AMA, by analyzing 33 aliquots of standard reference material (dogfish liver; DOLT-2) using both methods was evaluated. DOLT-2 measurements were significantly greater (7.6% difference, $t = 2.40$, $p = 0.0233$) using AMA than those values obtained using CVAAS (Table 1). The comparability between total mercury measured using AMA at NWRC and using CVAAS at ORF was also evaluated. Sixty-four duplicate aliquots of Herring Gull eggs from the Great Lakes that were analyzed previously using CVAAS, were reanalyzed in 2006 using AMA. These gull eggs were collected originally from 14 colony sites located across the Great Lakes during the years 1981–1983 and 1985–1986, and archived at −40°C at NWRC. Analyzes using CVAAS were performed on individual eggs ($n = 50$) from 1981 to 1985, and then on pools of eggs for each site in 1986 ($n = 14$); analyses using AMA were repeated on these

archived samples. Total mercury measurements in gull eggs were higher (5.8% difference, $t = 3.02$, $p = 0.0036$) using CVAAS than those analyzed by AMA (Table 1). There was a significant regression between the mean (or pooled) annual mercury concentrations derived from the two laboratories ($r^2 = 0.92$, $F_{1,62} = 747.9$, $p < 0.0001$), and the slope did not differ significantly from unity (slope = 0.934, 95% CI = 0.853–1.02).

To adjust all the data to be directly comparable, the data (i.e. ORF or CVAAS data) were multiplied by a constant. To adjust data from ORF (CVAAS) to be equivalent to NWRC (AMA), the ORF data were multiplied by 0.934. Similarly, data from NWRC (CVAAS) were multiplied by 1.079 to be equivalent to NWRC (AMA).

Mercury concentrations in composited whole body homogenates of Rainbow Smelt (*Osmerus mordax*) were provided by Environment Canada's Fish Contaminants Monitoring and Surveillance Program (FCMSP). The FCMSP collects fish from 10 locations annually across the 4 Canadian Great Lakes. A detailed description of monitoring locations, collection methods and laboratory processing procedures can be found in McGoldrick et al. (2010). Mercury analyses were conducted at the National Laboratory for Environmental Testing (NLET) in Burlington, Ontario, using NLET method 2801 (Environment Canada 2008), in which Hg is determined by CVAAS following acid digestion of the tissue samples. Details on the collection, processing, and storage of the fish samples are published elsewhere (Bhavsar et al. 2010).

Statistical analyses

The spatial pattern in mercury concentration among colony sites was determined by repeated-measures ANOVA (PROC GLM, Type III sums of squares; SAS Institute 2003) using data from 2005 to 2009. Post hoc contrasts among sites were performed using Student–Newman–Keuls multiple range tests (PROC GLM, SNK option; SAS Institute 2003). Data were log transformed (ln). Temporal analyses of mercury concentration, using both the full compliment of data and for retrospective analyses of discrete time periods, were performed using linear regression (PROC REG; SAS Institute 2003). All tests were performed using mean (or pooled) annual values per colony. Earlier studies have determined that pooled samples are equivalent to means calculated from the same eggs (Turle and Collins 1992). A sequential Bonferroni correction (Rice 1989) was used to assess significance when performing multiple analyses.

Change-point regression models were used to determine temporal changes in mercury concentrations, where separate trends were determined for mercury before and after a single change-point year, using a likelihood ratio test to determine the change-point year and the corresponding model of best fit (Draper and Smith 1981; for further details, see de Solla et al. 2010). All observations within 3 years from the start or finish of sampling were tested as the best possible change-point. The change point year, which corresponded to the most significant year to year change in mercury concentration and/or the year of a significant change in slope, was established. Separate slopes were calculated before and after the change point year. There were four possible models that the data could fit:

(1) a change-point model with unequal slopes (before and after the change point) and a significant change in mercury concentration at the change point year,
(2) a change-point model with equal slopes and a significant change in mercury concentration at the change point year,
(3) a change-point model with unequal slopes, but no significant change in mercury concentration at the change point, and
(4) a model with no change-point where a single slope indicates the rate of change (linear trend model).

A positive value indicates that mercury concentrations rose significantly at the change point year, and a negative value indicates that mercury concentrations declined

Table 1 Mean (variance) of mercury concentrations in dogfish liver (DOLT-2) and Herring Gull egg reference material, measured using an advanced mercury analyzer (AMA) or cold vapour atomic absorption spectrophotometry (CVAAS), at the National Wildlife Research Centre (NWRC) and Ontario Research Foundation (ORF)

Reference material	Statistic	Method (laboratory)			% Change (relative to AMA)
		AMA (NWRC)	CVAAS (NWRC)	CVAAS (ORF)	
DOLT-2	Mean	2.33	2.16		7.6
	Variance	0.066	0.02		
	n	18	15		
Herring Gull egg	Mean	0.245		0.259	−5.6
	Variance	0.019		0.02	
	n	64		64	

significantly at the change point year (Pekarik and Weseloh 1998; de Solla et al. 2010). Change point regressions were performed using an analysis module in WILDSPACE™ (Wong et al. 2003).

To characterize temporal changes in the Herring Gull diet, a Herring Gull diet index was formulated using stable carbon isotopes in eggs, trophic position (inferred from stable nitrogen isotope ratios; see Hebert et al. 1999b), and fatty acid content. Methods are described in detail in Hebert et al. (2008). Diet indices were generated for the period 1981–2005. Temporal trends in the diet index were examined using linear regression analysis (StatSoft Inc 2005). A backward, stepwise regression was performed (PROC GLM, SAS Institute 2003) to assess the partial effects of year, site and Herring Gull diet index on the Hg concentration in eggs; variables were retained in the model if $p \leq 0.10$.

To further assess the possible impact of dietary change on egg Hg trends, annual estimates of trophic position (inferred from egg δ^{15}N values) were regressed against annual $\log_e$ transformed Hg concentration data. The residuals from these regressions represented annual Hg concentrations adjusted for trophic position (see Hebert and Weseloh 2006). Temporal trends in unadjusted and adjusted Hg concentrations were compared. Temporal trends for Hg were examined at all 15 annual monitoring colonies. To examine temporal trends in egg Hg levels, a first-order linear equation was used. Annual percent change in Hg concentration was calculated as the slope (shown in Table 2) of the time/Hg regression equation.

Analysis of covariance (PROC GLM, SAS Institute 2003) was used to compare temporal trends of ln-transformed mean annual mercury concentrations in Herring Gull eggs and Rainbow Smelt tissues within individual water bodies.

Results

Current concentrations and spatial trends

Current (2009) Hg concentrations in Herring Gull eggs from the Great Lakes showed a nearly 4-fold range from 0.064 μg/g wet weight at Chantry Island to 0.246 μg/g wet weight at Middle Island (Appendix). Mean concentrations at 15 sites for the 5 year period 2005–2009 showed significant inter-site differences ($F_{14,58} = 8.87, p < 0.0001, r^2 = 0.68$; Fig. 2). Values ranged from 0.072 μg/g w.w. at Chantry

Table 2 Temporal trends in egg mercury concentrations at 15 Herring Gull monitoring colonies

Water body	Colony	Unadjusted data (1974–2009)[a]			Unadjusted data (1981–2009)[b]			Diet-adjusted data (1981–2005)[c]			% Rate
		r^2	p	Slope	r^2	p	Slope	r^2	p	Slope	
Lake Superior	**Granite Island**	0.17	<0.0001	−0.029	0.55	0.0006	−0.028	0.28	0.07	−0.0211	74.9
	Agawa Rocks	0.59	<0.0001	−0.025	0.46	0.0014	−0.024	0.27	0.05	−0.0173	71.6
Lake Michigan	Gull Island	0.13	0.11	−0.015	0.07	0.28	−0.012	0.03	0.56	−0.0078	NS
	Big Sister Island	0.49	0.0009	−0.032	0.33	0.0126	−0.024	0.02	0.60	−0.0064	26.2
Lake Huron	**Double Island**	0.55	<0.0001	−0.021	0.63	<0.0001	−0.026	0.02	0.59	−0.0039	14.8
	Chantry Island	0.58	<0.0001	−0.029	0.47	0.0011	−0.029	0.07	0.34	−0.0096	33.1
	Channel Shelter Island	<0.01	0.78	−0.002	0.005	0.78	−0.002	0.17	0.14	0.0131	NS
Detroit River	Fighting Island	<0.01	0.83	−0.001	0.00	0.83	−0.001	0.02	0.64	−0.0036	NS
Lake Erie	Middle Island	0.01	0.65	−0.002	0.04	0.44	0.005	0.01	0.77	0.002	NS
	Port Colborne	0.59	<0.0001	−0.031	0.63	<0.0001	−0.039	0.57	0.002	−0.0424	109.3
Niagara River	**Niagara River**	0.48	0.001	−0.236	0.48	0.001	−0.024	0.47	0.005	−0.0274	116.3
Lake Ontario	**Hamilton Harbour**	0.58	0.0002	−0.038	0.58	0.0002	−0.038	0.41	0.05	−0.0264	70.2
	Toronto Harbour	0.79	<0.0001	−0.047	0.69	<0.0001	−0.043	0.43	0.011	−0.0296	69.4
	Snake Island	0.51	0.0003	−0.030	0.39	0.0043	−0.028	0.46	0.005	−0.0373	131.8
St. Lawrence River	Strachan Island	0.08	0.21	−0.009	0.08	0.21	−0.009	0.02	0.55	−0.0059	NS

Trends were calculated using both unadjusted data (1974–2009 and 1981–2009 time periods) and mercury concentrations adjusted for temporal changes in gull diet (1981–2009; see Methods for details). All analyses were performed using ln-transformed values. % rate is the relative rate of temporal decline of the diet-adjusted data relative to the unadjusted data. NS indicates sites at which there were no significant temporal trends in Hg concentrations. Colonies in bold were those that exhibited a significant temporal decline (1981–2009) in unadjusted Hg concentrations

[a] $N = 17–21$

[b] $N = 17–21$

[c] $N = 9–14$

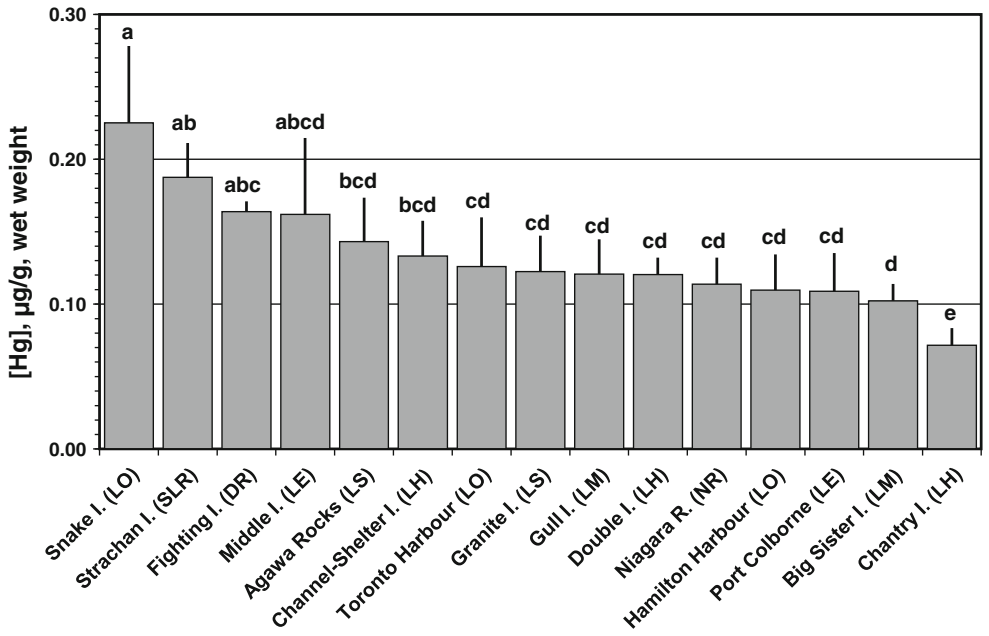

Fig. 2 Mean concentration of mercury (µg/g, wet weight, SD) in Herring Gull eggs, 2005–2009. No eggs were available from Fighting in 2009 and 11 eggs were collected per site in 4 years (Gull Island in 2005 and 2009, Fighting Island in 2007 and 2008); otherwise, 13 eggs were collected per site per year. Overall, there were significant differences in ln-transformed mercury concentrations among sites ($F_{14,58} = 8.87$, $p < 0.0001$, $r^2 = 0.68$). Similar letters above error bars denote no significant difference in post hoc comparisons among sites. Water body abbreviations are as follows: *LS* Lake Superior, *LM* Lake Michigan, *LH* Lake Huron, *DR* Detroit River, *LE* Lake Erie, *NR* Niagara River, *LO* Lake Ontario, *SLR* St. Lawrence River. See Table 4 for details of analysis

Island to 0.225 µg/g w.w. at Snake Island (Fig. 2); sites from lakes Erie, Superior and Michigan were intermediate.

Temporal patterns and trends

Mercury values at 14 of 15 sites declined from 22.6 to 85.8% between the first year of analysis (usually 1974) and the most recent year (2009) (Fig. 3). Only one site (Middle Island) showed an increase (10.5%). The average decline in mercury concentration in gull eggs over all sites was 55.0%. For the overall period, 10 of 15 sites showed significant declining trends ($p \leq 0.004$) (Table 2).

For temporal trends in mercury in recent years, linear regressions were performed on increasingly longer time

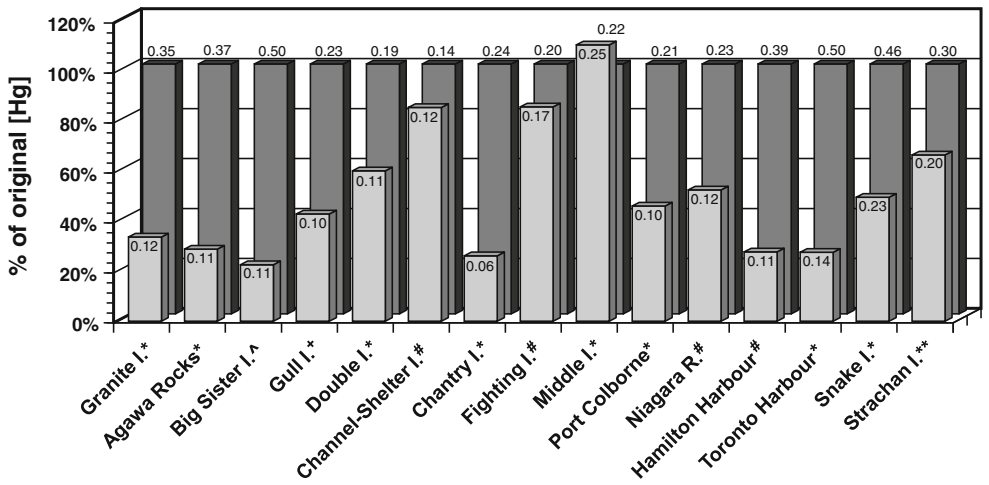

Fig. 3 Change in mercury concentration (µg/g, wet weight) in Great Lakes Herring Gull eggs from year of first measurement (*black bars*) compared to most recent measurement (2009, *grey bars*). Values in first year of measurement have been set to 100%. Years of first measurement are indicated as follows: *1974, ^1976, +1978, #1981, **1986. No eggs were available from Fighting in 2009, so the 2008 value has been used. Values at the *top* of the bars are the actual mercury values (µg/g, wet weight)

Table 3 The number of non-significant regressions of mercury concentrations in Herring Gull eggs from 15 colony sites in the Great Lakes during increasingly more recent periods of analysis and 10 year standard periods

Analysis period	Length of analysis period from present (years)	Number of sites where regression was non-significant (after Bonferroni correction)
1974–2009	35	5
1979–2009	30	5
1981–2009	28	5
1982–2009	27	6
1983–2009	26	9
1984–2009	25	11
1989–2009	20	12
1994–2009	15	15
1999–2009	10	15

periods working backwards from 2009 (Table 3). At all 15 sites, for the periods 1999–2009 and 1994–2009, i.e. for the last 10 and 15 years, there were no significant trends. For 1989–2009, the last 20 years, there were three significant declining regressions. Egg mercury concentrations at most sampled nesting sites have shown little decline over the last 15–20 years.

A significant change-point year was found using change-point regression analysis for all sites except Fighting Island and Strachan Island. The most frequent change-point years were 1983 and 2004 (three sites each) followed by 1985 and 1992 (two sites). Six of 15 sites had equal slopes before and after their change-point, indicating a constant rate of decline but with a positive or negative inflection (displacement) in the change-point year. Four sites showed faster rates of decline after their change-points and three showed slower rates of decline. Two sites showed no trend (Fig. 4; Table 4).

Eleven of the 15 Herring Gull colonies showed a significant temporal change in the gull diet index. In a stepwise regression model ($F_{2,211} = 26.96$, $p < 0.0001$, $r^2 = 0.20$), year ($F = 53.11$, $p < 0.0001$) was the strongest predictor of the Hg concentration of eggs. Diet index ($F = 5.50$, $p = 0.02$) also had a significant negative effect on egg mercury; generally, Hg increased with more negative d13C values, greater trophic position, and a fatty acid index with greater proportions of omega 3 fatty acids.

Using the diet-adjusted mercury data, significant declines were found at only six colonies, as compared to 10 sites for unadjusted data (Table 2). In addition, diet-adjusted Hg levels showed a less pronounced temporal decline (estimated using the slope of the regression equations). Hg concentrations adjusted for changes in gull diet declined at a mean rate of 72% of the unadjusted Hg data (Table 2).

Discussion

In this study, the current concentrations and spatial and temporal trends for mercury in Herring Gull eggs from 15 sites in the Great Lakes over a 35 year period have been documented. Spatially, for the period 2005–2009, there were significant differences in mercury among sites. Temporal analyses showed that concentrations declined at 14 of the 15 sites. However, after adjusting concentrations for dietary change, most colonies no longer exhibited statistically significant temporal declines. Also, there were no significant regressions (increasing or decreasing trends) at any of the sites for mercury over at least the last 15 years. Overall these data suggest that while Hg concentrations in Herring Gull eggs have decreased since the 1970s, only part of this long-term decline may be the result of reductions in Hg levels in the environment; dietary change may have also been important.

The greatest source of mercury to Herring Gulls comes from the food they eat. Therefore, similar spatial distributions should be found in the fish species that Herring Gulls feed upon or in the predatory fish which feed on the same or similar species as Herring Gulls. Spatial patterns for mercury in Herring Gull eggs differ from those found in the two main species of predatory fish used as biomonitors in the Great Lakes: Lake Trout (*Salvelinus namaycush*) and Walleye (*Sander vitreus*). Historical spatial patterns in those fish showed that the lowest concentrations were in Lake Erie and the highest were in Lake Superior; more recently (2000–2007) that pattern was only true for Lake Trout (Bhavsar et al. 2010). In an analysis of contaminants in fish from Canadian Areas of Concern, Weis (2004) found that predatory fish (walleye, pike, bass, etc.) from Cornwall and/or Lake Superior had the highest mercury levels. Those from Lake Huron and some species from Lake Ontario had the lowest. Over the last decade, mean mercury levels in Rainbow Smelt, one of the fish species that Herring Gulls are known to feed on, were Lake Superior > Lake Huron > Lake Ontario > Lake Erie (D. McGoldrick, unpublished data).

Further information on the spatial distribution of contaminants can also be obtained from the analysis of sediments (Thomas 1972; Kemp and Thomas 1976; Painter et al. 2001). For example, Marvin et al. (2004) found that lakes Huron, Michigan and Superior exhibited the lowest concentrations (<0.089 μg/g). The highest concentrations

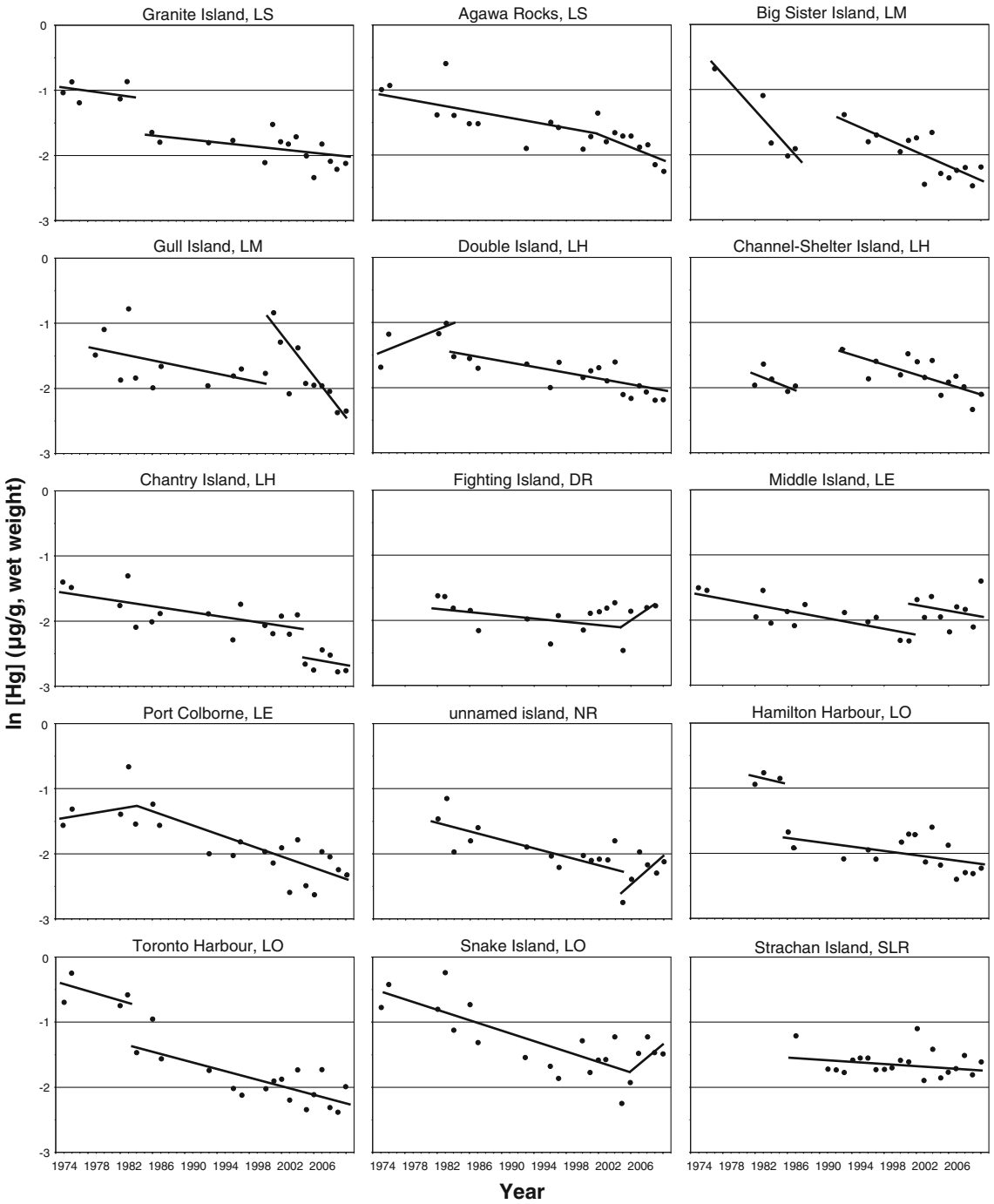

Fig. 4 Change-point regression analyses for 15 annual monitoring sites on the Great Lakes and connecting channels using annual mean, ln-transformed, mercury concentrations (µg/g, wet weight) in Herring Gull eggs, 1974–2009

were found in western Lake Erie and Lake Ontario (>0.401 µg/g). Elevated Hg sediment concentrations in western Lake Erie had been noted previously by Painter et al. (2001). Spatial patterns observed in sediment mercury concentrations are more reflective of those inferred from gull eggs than predatory fish.

Temporal trends in mercury concentrations in various media over different time frames are readily available. Many of them are declining, though there are examples of increasing concentrations in Arctic and boreal wildlife (Braune et al. 2006; Braune 2007; Hebert et al. 2011). Sediment levels declined from 25 to 80% in the Great

Table 4 Summary of change-point analyses on mercury concentrations (ln-transformed, μg/g, wet weight) from 15 monitoring colonies on the Great Lakes and connecting channels (1974–2009)

Water body	Colony	Overall model					Model parameters				
		Model selected[a]	df	F	*p*	r^2	$\beta1$	Year of change point	$\beta3$	$\beta2$	Decline in $\beta2$ relative to $\beta1$
Lake Superior	Granite Island	2	2,17	31.18	≤0.0001	0.76	Negative*	1985	Decrease*	Negative*	Constant[#]
	Agawa Rocks	3	2,18	15.54	0.0001	0.63	Negative*	2001	No change	Negative*	Faster*
Lake Michigan	Big Sister Island	1	3,15	17.03	≤0.0001	0.77	Negative***	1992	Increase***	Negative***	Slower*
	Gull Island	1	3,17	5.84	0.0062	0.51	NS	2000	Increase*	Negative**	Faster**
Lake Huron	Double Island	1	3,17	12.9	0.0001	0.69	NS	1983	Decrease*	Negative**	Faster*
	Channel Shelter Island	2	2,16	11.92	0.0007	0.60	Negative*	1992	Increase***	Negative*	Constant[#]
	Chantry Island	2	2,18	30.33	≤0.0001	0.77	Negative*	2004	Decrease**	Negative*	Constant[#]
Detroit River	Fighting Island	3	2,14	0.77	0.48	0.10	NS	2004	No change	NS	Slower*
Lake Erie	Middle Island	2	2,18	2.94	0.079	0.25	Negative*	2001	Increase*	Negative*	Constant[#]
	Port Colborne	3	2,18	16.31	≤0.0001	0.64	NS	1983	No change	Negative[^]	Faster*
Niagara River	Niagara River	1	3,15	8.61	0.0015	0.63	Negative*	2004	Decrease*	NS	Slower*
Lake Ontario	Hamilton Harbour	2	2,16	28.17	≤0.0001	0.78	NS	1985	Decrease**	NS	Constant[#]
	Toronto Harbour	2	2,18	57.1	≤0.0001	0.85	Negative***	1983	Decrease**	Negative***	Constant[#]
	Snake Island	3	2,18	14.22	0.0002	0.60	Negative[^]	2004	No change	NS	Slower*
St. Lawrence River	Strachan Island	4	1,19	1.72	0.20	0.08	NS	n/a	No change	NS	No trend

The following parameters are included in model equations: $\beta0$ = intercept, $\beta1$ = slope before change point, $\beta2$ = slope after change point, and $\beta3$ = change in value at change point

n/a not applicable

p values: * $0.05 \geq p > 0.01$; ** $0.01 \geq p > 0.001$; *** $0.001 \geq 0 > 0.0001$; [^] ≤0.0001; [#] Slopes not significantly different

[a] Models tested: (1) $y = \beta0 + \beta1 \times x1 + \beta2 \times x2 + \beta3$; (2) $y = \beta0 + \beta(x1 + x2) + \beta3$; (3) $y = \beta0 + \beta1 \times x1 + \beta2 \times x2$; (4) $y = \beta0 + \beta \times x$ (linear model)

Lakes from the 1960s/1970s to the 1990s/2000s (Painter et al. 2001; Marvin et al. 2004). In fish from Canadian Great Lakes Areas of Concern, Weis (2004) showed declines in mercury in four predatory species from Lake St. Clair from the 1970s up to 1990. During the 1990s, concentrations in Northern Pike (*Esox lucius*) continued to decline, those in Yellow Perch (*Perca flavescens*) levelled off, while those in Walleye and Smallmouth Bass (*Micropterus dolomieu*) began to increase. Common Loons (*Gavia immer*) in the north-eastern US and south-eastern Canada, showed declines in mercury levels in blood from 1999 to 2005 (Evers et al. 2007). This latter study suggested that reductions in mercury emissions from local sources led to rapid reductions in mercury in biota. Mercury concentrations in the liver of mink and otter from New York declined significantly (from 25 to 38%) during the period 1982–2003 (Yates et al. 2005).

Unadjusted temporal trends in Herring Gull eggs were compared with unadjusted temporal trends in Rainbow Smelt (D. McGoldrick unpublished data) in each of the Canadian Great Lakes (Fig. 5). In all four cases, the slopes tracked one another and there were no differences between the slopes of gulls versus smelt in each lake. This strongly suggests a similar rate of decline between these two components of the aquatic food web (Table 5). However, it appears as if rates of decline of Hg in gull and smelt may be diverging in the most recent years. This could be the result of slowing rates of decline of Hg in smelt or could be a result of dietary change in gulls as discussed below.

Adjusting Hg trends for dietary change

Over the last two decades, there have been significant changes in the trophic structure of Great Lakes food webs.

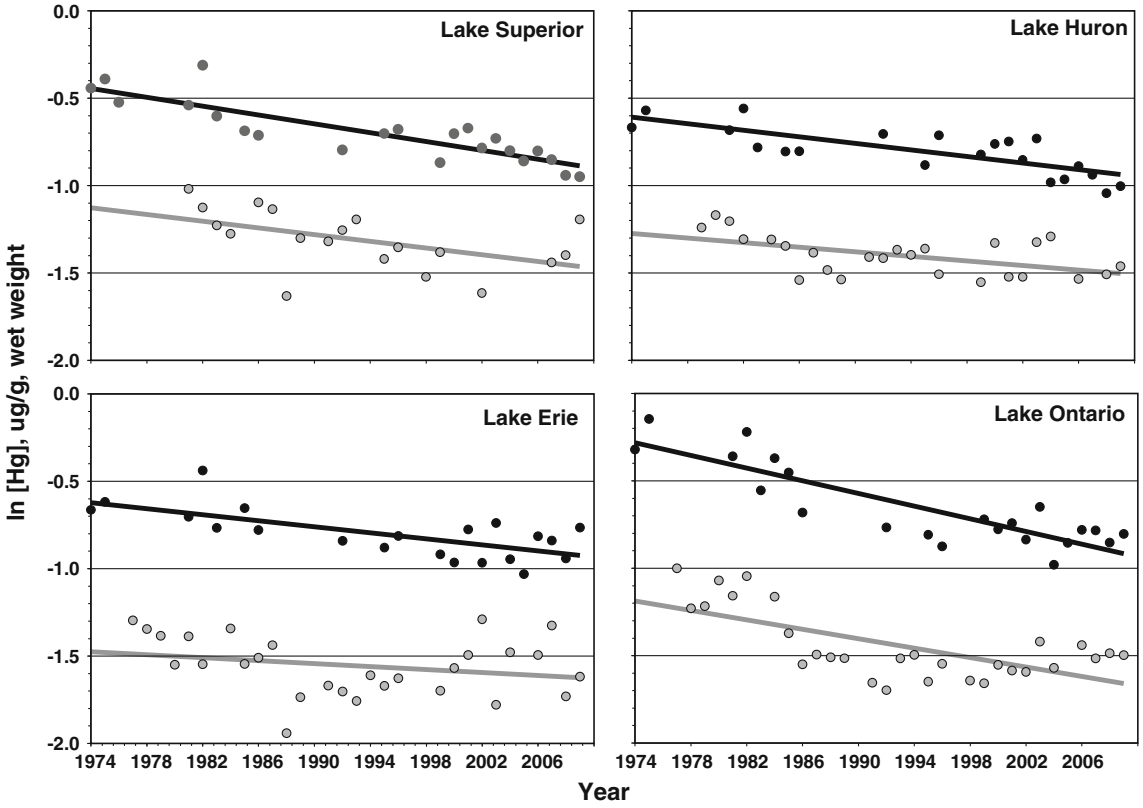

Fig. 5 Temporal patterns in annual mean mercury concentrations (ln-transformed, μg/g, wet weight) in Herring Gull eggs (*black*) and Rainbow Smelt (*grey*) from Lakes Superior, Huron, Erie and Ontario. Values for Herring Gulls represent the average of the 2–3 colonies sampled on each water body. See Table 5 for details of analysis

Table 5 Results of ANCOVA assessing changes in mean mercury concentration (μg/g, wet weight; log 10-transformed) in Herring Gull eggs and Rainbow Smelt (tissue) from 1974 to 2009 by water body

Water body	Overall model			Tissue			Year		
	df	F	*p*	df	F	*p*	df	F	*p*
Lake Erie	2,46	172.97	<0.0001	1,47	344.18	<0.0001	1,47	10.57	0.0022
Lake Huron	2,43	250.02	<0.0001	1,44	482.46	<0.0001	1,44	34.49	<0.0001
Lake Ontario	2,48	233.78	<0.0001	1,49	421.40	<0.0001	1,49	68.37	<0.0001
Lake Superior	2,38	152.23	<0.0001	1,39	272.76	<0.0001	1,39	40.18	<0.0001

For all tissue × year interactions, $p \geq 0.27$

Levels of phosphorous and other nutrients have been reduced significantly; populations of pelagic prey fish also have been diminished greatly and there has been the accidental introduction of bottom-dwelling invasive species such as Dressenid mussels (*Dreissena polymorpha, D. bugensis*) and the Round Goby (*Neogobius melanostomus)* (Hogan et al. 2007; Stewart et al. 2009). Research involving food web tracers identified changes in the diets of Herring Gulls nesting on the Great Lakes (Hebert et al. 2006, 2008, 2009). These dietary changes may have important implications for the interpretation of contaminant trends data.

Both year and diet were important in regulating Hg concentrations in eggs. Eggs having less negative $\delta^{13}C$ values and fatty acid profiles with lower proportions of omega 3 fatty acids are indicative of consumption of less

aquatic food, i.e. fish (Hebert et al. 2008), in recent years. Prey fish occupy higher trophic levels than other foods that gulls consume (Hebert et al. 1999a); therefore, reductions in fish consumption result in declines in gull trophic position. Comparing trends in egg mercury data with and without trophic position adjustment allowed for the examination of the effect of changes in the food of Herring Gulls on temporal changes in Hg concentrations in their eggs.

It is useful to compare temporal trends in Hg in gull eggs with other aquatic species in the Great Lakes. For example, Hg in lake trout (Bhavsar et al. 2010) showed declines in all of the Canadian Great Lakes, similar to the trends inferred from the unadjusted Herring Gull egg Hg data. Hg trends in walleye (Bhavsar et al. 2010) exhibited declines in lakes Superior and Huron but not in lakes Erie or Ontario. This more limited indication of temporal declines in Hg levels is more similar to the trends inferred from the adjusted gull egg Hg data. The possible influence of dietary change on contaminant trends data is not limited to Herring Gulls. The broad-scale changes in food web structure observed in the Great Lakes are also impacting other important biomonitoring species such as the lake trout (Paterson et al. 2009). Obviously, more research is required to understand the implications of food web change and other factors on contaminant exposure and accumulation in important biomonitoring species. Through such research the apparent differences in Hg trends across datasets may be reconciled and a more accurate picture of Hg availability in the Great Lakes obtained.

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Appendix

See Table 6.

Table 6 Mean (±SD) mercury concentration (μg/g, wet weight), adjusted for analytical technique, for each of 15 annual monitoring colonies in the Great Lakes and connecting channels, 1974–2009

Year[a]	Lake Superior		Lake Michigan		Lake Huron			Detroit River
	Granite Island (48.722, −88.462)	Agawa Rocks (47.359, −84.693)	Big Sister Island (45.221, −87.131)	Gull Island (45.516, −86.724)	Double Island (46.173, −82.865)	Channel-Shelter Island (43.677, −83.829)	Chantry Island (44.493, −81.403)	Fighting Island (42.216, −83.117)
1974	0.331 ± 0.120	0.344 ± 0.073			0.174 ± 0.053		0.228 ± 0.067	
1975	0.389 ± 0.119	0.370 ± 0.139			0.290 ± 0.118		0.213 ± 0.077	
1976	0.280 ± 0.066		0.467 ± 0.030					
1978				0.211 ± 0.053				
1979				0.314 ± 0.073				
1981	0.305 ± 0.079	0.234 ± 0.070		0.142	0.290 ± 0.065	0.133 ± 0.046	0.159 ± 0.076	0.185 ± 0.088
1982	0.393 ± 0.109	0.518 ± 0.133	0.311 ± 0.163	0.431 ± 0.115	0.337 ± 0.059	0.182 ± 0.044	0.254 ± 0.081	0.181 ± 0.099
1983		0.233 ± 0.188	0.153 ± 0.039	0.148 ± 0.031	0.204 ± 0.070	0.144 ± 0.059	0.114 ± 0.068	0.153 ± 0.072
1984								
1985	0.179 ± 0.044	0.205 ± 0.073	0.124 ± 0.037	0.128 ± 0.020	0.195 ± 0.038	0.121 ± 0.015	0.123 ± 0.048	0.149 ± 0.075
1986	0.155	0.207	0.139	0.176	0.168	0.130	0.141	0.109

Table 6 continued

Year[a]	Lake Superior		Lake Michigan		Lake Huron			Detroit River
	Granite Island (48.722, −88.462)	Agawa Rocks (47.359, −84.693)	Big Sister Island (45.221, −87.131)	Gull Island (45.516, −86.724)	Double Island (46.173, −82.865)	Channel-Shelter Island (43.677, −83.829)	Chantry Island (44.493, −81.403)	Fighting Island (42.216, −83.117)
1990								
1991								
1992	0.181	0.165	0.272	0.151	0.212	0.264	0.163	0.149
1993								
1994								
1995	0.183	0.246	0.177	0.178	0.148	0.168	0.109	0.102
1996		0.227	0.200	0.197	0.217	0.219	0.192	0.158
1997								
1998								
1999	0.131	0.162	0.156	0.184	0.172	0.181	0.135	0.127
2000	0.233	0.194	0.181	0.469	0.192	0.249	0.119	0.163
2001	0.181	0.279	0.190	0.299	0.200	0.220	0.158	0.168
2002	0.160	0.167	0.086	0.125	0.150	0.161	0.110	0.163
2003	0.180	0.192	0.190	0.250	0.201	0.206	0.150	0.178
2004	0.136	0.180	0.102	0.146	0.122	0.121	0.071	0.085
2005	0.096	0.181	0.095	0.143	0.114	0.147	0.064	0.156
2006	0.163	0.152	0.107	0.141	0.138	0.161	0.088	
2007	0.124	0.158	0.111	0.130	0.126	0.139	0.080	0.165
2008	0.110	0.119	0.085	0.093	0.111	0.098	0.063	0.170
2009	0.119	0.106	0.113	0.097	0.112	0.122	0.064	

Year[a]	Lake Erie		Niagara River	Lake Ontario			St. Lawrence River
	Middle Island (41.682, −82.682)	Port Colborne Breakwall (42.868, −79.258)	Unnamed Island (43.070, −79.072)	Hamilton Harbour (43.278, −79.791)	Toronto Harbour (43.623, −79.330)	Snake Island (44.190, −76.543)	Strachan Island (45.015, −74.812)
1974	0.208 ± 0.065	0.197 ± 0.036			0.466 ± 0.098	0.428 ± 0.184	
1975	0.200 ± 0.077	0.250 ± 0.047			0.724 ± 0.372	0.614 ± 0.261	
1976							
1978							
1979							
1981	0.134 ± 0.027	0.236 ± 0.072	0.216 ± 0.078	0.362 ± 0.094	0.443 ± 0.125	0.421 ± 0.119	
1982	0.196 ± 0.040	0.486 ± 0.028	0.296 ± 0.047	0.435 ± 0.076	0.521 ± 0.052	0.735 ± 0.179	

Table 6 continued

Year[a]	Lake Erie		Niagara River	Lake Ontario			St. Lawrence River
	Middle Island (41.682, −82.682)	Port Colborne Breakwall (42.868, −79.258)	Unnamed Island (43.070, −79.072)	Hamilton Harbour (43.278, −79.791)	Toronto Harbour (43.623, −79.330)	Snake Island (44.190, −76.543)	Strachan Island (45.015, −74.812)
1983	0.121 ± 0.027	0.200 ± 0.096	0.131 ± 0.039		0.215 ± 0.043	0.306 ± 0.077	
1984				0.398 ± 0.046			
1985	0.143 ± 0.031	0.271 ± 0.109	0.154 ± 0.060	0.178	0.360 ± 0.197	0.453 ± 0.097	
1986	0.116	0.195	0.190 ± 0.128	0.137	0.196 ± 0.154	0.252 ± 0.101	0.280
1990							0.195
1991							0.193
1992	0.164	0.147	0.162	0.135	0.189	0.231	0.181
1993							0.220
1994							0.231
1995	0.143	0.142	0.142	0.155	0.144	0.203	0.229
1996	0.155	0.177	0.118	0.135	0.130	0.167	0.194
1997							0.193
1998							0.200
1999	0.108	0.153	0.141	0.175	0.142	0.299	0.223
2000	0.107	0.127	0.133	0.195	0.162	0.183	0.216
2001	0.201	0.161	0.135	0.195	0.167	0.224	0.356
2002	0.141	0.075	0.124	0.118	0.112	0.209	0.150
2003	0.196	0.169	0.165	0.201	0.177	0.296	0.246
2004	0.142	0.083	0.065	0.112	0.096	0.105	0.156
2005	0.114	0.072	0.092	0.152	0.122	0.145	0.171
2006	0.168	0.139	0.139	0.090	0.178	0.229	0.182
2007	0.160	0.129	0.115	0.099	0.100	0.295	0.222
2008	0.122	0.107	0.101	0.099	0.092	0.229	0.164
2009	0.246	0.097	0.122	0.107	0.137	0.227	0.200

The latitude and longitude of each colony is indicated below the site name. From 1986 onwards, all eggs were analyzed as site pools (no associated variance)

[a] No data exist for 1977, 1980, 1987–1989

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