Mercury trends in herring gull (Larus argentatus) eggs from Atlantic Canada, 1972–2008: Temporal change or dietary shift?

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A B S T R A C T

Mercury (Hg) is a pervasive contaminant that can adversely affect predatory wildlife. Bird eggs provide insights into breeding females’ Hg burdens, and are easily collected and archived. We present data on Hg trends in herring gull (Larus argentatus) eggs from five sites in Atlantic Canada from 1972 to 2008. We found a significant decrease in Hg at Manawagonish Island, New Brunswick and Île du Corossol, Quebec, but after correcting Hg for dietary shifts using stable isotopes (δ15N), these trends disappeared. Decreasing temporal trends of stable isotopes in gull eggs were observed at four sites, suggesting shifts in diet. At Gull Island, Newfoundland, diet-adjusted Hg increased from 1977 to 1992, dropped sharply between 1992 and 1996, and rose again from 1996 to 2008. After adjusting Hg trends for dietary shifts of herring gulls, it appears that environmental Hg in coastal ecosystems has remained relatively constant at most sites in Atlantic Canada over the last 36 years.

1. Introduction

Mercury (Hg) is an atmospherically transported contaminant, much of which is anthropogenically generated, and deposition is increasing worldwide (Nriagu, 1989; Nriagu and Pacyna, 1988; Streets et al., 2009). As top predators in the marine environment, seabirds are used frequently as sentinel species for monitoring ecosystem contamination (Burger and Gochfeld, 2004; Goodale et al., 2008; Hebert et al., 1999; Mineau et al., 1984; Pearce et al., 1979). Their high trophic position also means that seabirds can be exposed to high concentrations of biomagnified contaminants, including Hg (Braune et al., 2005). The biologically active and toxic form, methylmercury (MeHg), is the predominant type found in seabird feathers and eggs (Bond and Diamond, 2009; Thompson and Furness, 1989). Seabirds acquire MeHg through ingestion of contaminated prey. MeHg is then deposited in body tissues, demethylated in the liver or brain, or depurated into feathers or eggs (Braune and Gaskin, 1987; Monteiro and Furness, 2001; Spalding et al., 2000).

In some marine ecosystems, Hg concentrations in seabirds have increased over time (e.g., Braune, 2007; Thompson et al., 1992). This increase could be due to two processes: first, the birds consumed a relatively constant diet over time and MeHg levels increased in that diet, or second, the birds shifted their diet over time to prey with higher Hg concentrations (see Hebert et al., 2009). The first possibility of changing Hg levels in prey could result from environmental changes, such as increased atmospheric deposition of Hg, increased bioavailability of MeHg, climatic or oceanographic changes (e.g., Aebscher et al., 1990; Drinkwater, 1996), or changes in food web structure (e.g., Carscadden et al., 2001; Montevecchi and Myers, 1996). Stable isotope analysis can be used to address the second possibility (a shift in diet) because isotope values (δ13C/12C, or δ15N/14N) will reflect the consumer’s diet at the time of tissue synthesis (Hobson, 1995; Hobson and Clark, 1992). δ13C can give an indication of the geographic foraging area, as inshore and more productive oceanic areas (including terrestrial sources, such as landfill sites) are enriched in δ13C (France, 1995; Goercke and Fry, 1994; Peterson and Fry, 1987; Popp et al., 1998), while δ15N values increase 2–5‰, with each trophic level because 14N is excreted preferentially in nitrogenous waste (Kelly, 2000; Minagawa and Wada, 1984; Steele and Daniel, 1978). However, analysing changes in carbon isotope values over time is potentially confounded by the Suess (1955) Effect, as the combustion of fossil fuels (naturally depleted in 13C) have altered the baseline δ13C value globally, including in the North Atlantic (Quay et al., 2007), but this can be taken into account...
mathematically (e.g., Farmer and Leonard, 2011). The Suess Effect is a global phenomenon affecting terrestrial, freshwater, and marine ecosystems (Keeling, 1979; Keeling et al., 1995; Quay et al., 2007; Suess, 1953, 1955).

Herring gulls (Larus argentatus) are a common predator in the northwest Atlantic Ocean, and are found throughout Atlantic Canada (Pierotti and Good, 1994). As income breeders, herring gulls incorporate nutrients from local breeding grounds into their eggs (Drent and Daan, 1980; Hobson et al., 2000). Herring gulls have also been identified as useful biomarkers of biomagnifying contaminants, such as Hg, because they are top predators, often forage in urbanized areas, are long-lived, widely distributed, and egg samples are easy to collect (Golden and Rattner, 2003; Hebert et al., 1999). Hg concentrations in herring gulls have been studied in the Great Lakes region (Koster et al., 1996; Weseloh et al., 1990, 2011), northeastern United States (Burger and Gochfeld, 1995), Gulf of St. Lawrence (Lavioie et al., 2010a) and in Europe (Lewis et al., 1993; Rüdel et al., 2010). However, because herring gulls are opportunistic inshore predators and scavengers, they commonly shift their diet to take advantage of changes in prey availability (Pierotti and Annett, 1991; Pierotti and Good, 1994). While their diet is primarily marine fish, invertebrates and seabirds, they can also forage at garbage dumps and sewage outfalls (Pierotti and Annett, 1991; Pierotti and Good, 1994). Shifts in diet have been linked to changes in contaminant levels in herring gull eggs (Hebert et al., 2000, 2009). As a result, recent analyses of temporal trends of contaminant concentrations in herring gull eggs have adjusted for diet shifts using stable isotope data (Hebert and Weseloh, 2006; Weseloh et al., 2011).

Our objectives were to: 1) assess changes in Hg concentrations over the last 36 years in the eggs of an abundant marine predator, the herring gull, 2) examine concurrent changes in stable isotope values to detect any possible trophic shifts that might be related to simultaneous changes in Hg concentrations, and 3) to adjust the temporal changes in egg Hg concentrations for any trophic shifts observed, to yield a more accurate indicator of Hg trends in the marine ecosystems studied.

2. Methods

2.1. Sample collection

Eggs were collected from five colonies in Atlantic Canada. Colony locations were diverse and included one site in the northern Gulf of St. Lawrence (Île du Corosol, Québec, 50° 05′N, 66° 23′W), one in the Bay of Fundy (Kent Island, New Brunswick, 44°54′N, 66°44′W, and Manawagonish Island, New Brunswick, 45°12′N, 66°06′W), an offshore site 300 km off the coast of Nova Scotia (Sable Island, Nova Scotia, 43°56′N, 59°54′W), and a coastal site open to the Grand Banks (Gull Island, Newfoundland 47°13′N, 52°47′W; Fig. S-1). Eggs were collected at each colony beginning in 1972 (Corosol, Manawagonish), 1976 (Kent, Sable) or 1977 (Gull), continuing at irregular intervals until 1980, after which eggs were sampled every four years until 2008 (Table S-1). From 1972 to 1988 we collected one egg from five nests in each colony, and from 1992 to 2008 we collected one egg from 15 nests in each colony. The change in sampling design in 1992 was done to increase our ability to detect changes in contaminant levels, based on power analysis (Hebert and Weseloh, 2003). Eggs were stored in foam-lined toolboxes at 4°C in chemically cleaned glass jars prior to analysis. Our eggs had C:N in chemically cleaned glass jars prior to analysis.

To ensure comparable Hg data, archived egg samples from 1972 to 1996 were retrieved from the National Wildlife Specimen Bank at NWRC for Hg analysis. These archived samples were frozen at −40°C since their collection.

2.2. Total mercury analysis

The five eggs collected at each colony prior to 1992 were analysed individually for total Hg. The 15 eggs collected at each colony in 1992–2008 were analysed as three composite (pooled) samples of five eggs each. We used two methods to analyse total Hg in herring gull eggs. Samples collected in 2000 were analysed using cold vapour atomic absorption spectrophotometry (CVAAS) using a Perkin–Elmer 3103B, AAS (Waltham, MA, USA) and Varian VCA-76 vapour generation accessory (Agilent Technologies, Mississauga, Ontario). Detailed methods were described by Scheuhammer and Bond (1991) and Neugebauer et al. (2000). All other egg samples were analysed using an Advanced Mercury Analyser 254 (AMA-254, Altex Ltd., Prague, Czech Republic), in batch to discrete system, using EPA Method 7473 (U.S. EPA, 1998) as described by Weseloh et al. (2011).

Frozen egg samples were thawed at 4°C, and individual samples were homogenized using an electric mixer. For samples collected from 1992 to 2008, aliquots of equal volume were pooled in sterile Teflon vials and mixed thoroughly. Pools were generally 5 individual eggs, except in the few cases where an individual egg broke during transport to NWRC. Samples analysed using CVAAS were first freeze-dried, and then digested overnight in 70% nitric acid, followed by digestion with 95% sulphuric acid and 37% hydrochloric acid (Neugebauer et al., 2000).

Samples analysed using direct mercury analyser AMA-254 were placed in nickel boats for direct measurement of Hg, and Hg concentrations were converted to dry weight after % moisture determination. Detection limits using CVAAS were 0.02 μg/g for the analysed dry sample, and 0.006 μg/g in the dry sample for the AMA-254. The difference in detection limits was not a concern since all egg samples had Hg concentrations well above the higher detection limit.

To test the comparability of CVAAS and AMA-254 results, 24 common loon egg samples were analysed for total Hg using both methods (Bond, 2008). The mean Hg concentrations were not significantly different (Wilcoxon Sign Rank test, p = 0.2). Average variability of the paired Hg data was 8.4 ± 7.13% (mean relative standard deviation (s) ± S.D.; Regression analysis showed a strong association between the CVAAS vs. AMA-254 data (CVAAS = 0.98 * AMA – 0.32, R² = 0.96).

Of the 185 total samples, 100 (54%) were analysed in duplicate or triplicate. Average variability (sD ± S.D.) of these replicate samples was 1.86 ± 2.69%. Within each analytical run, Hg concentrations were corrected for certified reference materials (mean recovery of certified reference materials: DOLT-2 (dogfish liver, 104 ± 7% recovery, certified concentration: 2.14 μg/g, n = 13), DOLT-3 (dogfish liver, 103 ± 11% recovery, certified concentration: 3.37 μg/g, n = 8), DORM-2 (dogfish muscle, 100 ± 3% recovery, certified concentration: 4.64 μg/g, n = 5), OT-1566b (oyster tissue, 89 ± 9% recovery, certified concentration: 0.037 μg/g, n = 4), and TOT-2 (oyster tissue, 103 ± 7% recovery, certified concentration: 0.270 μg/g, n = 24). Oyster tissue was used in all analytical runs, along with one of the other four reference materials. Hg concentrations measured in the herring gull eggs were within the range of concentrations of the certified reference materials.

2.3. Stable isotope analysis

Stable nitrogen and carbon isotope analyses were conducted at the University of Ottawa’s G.G. Hatch Stable Isotope Laboratory using 1 mg (±0.2 mg) of freeze-dried egg tissue encapsulated in tin. Isotope analysis was completed using a VarioEL III Elemental Analyser (Elementar, Hanau, Germany) followed by trap and purge separation and on-line analysis by continuous-flow with a DeltaPlus Advantage isotope ratio mass spectrometer (Thermo Scientific, Waltham, USA) coupled with a ConFlo II. Data were normalized using international standards for calibration (IAEA-CH-6, IAEA-NBS22, IAEA-N1, IAEA-N2, USGS-40, USGS-41) and quality control was maintained through sample duplicates. Stable isotope values were reported in δ13C and δ15N normalized for lipid content (mod15C and mod15N) following Tagliabue and Bopp (2009). Lipids are naturally depleted in 13C, and because individual eggs vary in lipid content, lipid must be removed or adjusted for mathematically to interpret carbon isotope values (Bond and Jones, 2009; Kojadinovic et al., 2008; Logan et al., 2008; Post et al., 2007). We assumed that, as income breeders, all macronutrients incorporated into eggs would be from the breeding grounds (Oppel et al., 2010), so we adjusted for eggs’ variable lipid content mathematically using methods described in Post et al. (2007: 186) for aquatic organisms, where

\[
\delta^{13}C_{\text{normalized}} = \delta^{13}C_{\text{uncorrected}} - 3.32 + 0.99 \times C : N
\]

Herring gull C:N in our samples ranged from 5.74 to 10.89 (mean = 8.90). The equation in Post et al. (2007) was derived for C:N 3.0–7.0. Since only 11% of our eggs had C:N > 8.0 and the relationship described by Post et al. (2007) is strongly linear, we assumed that this approach was valid.

Following lipid adjustment, we used the approach of Farmer and Leonard (2011: 126) to adjust δ15C values for the Suess Effect using the post-1950 portion of their Eq. (2):

\[
\delta^{15}C_{\text{corrected}} = \delta^{15}C_{\text{normalized}} - \delta^{15}C_{\text{infinite}} \times (1950 - t) - b_{mod} \times (t - 1950)
\]

where δ15Ccorrected is the δ15C value normalized for lipid content, δ15Cinfinite is the historical annual decline in δ15C (≈ 10.07‰/yr), taglub is an adjustment described by Tagliabue and Bopp (2008), and bmod is the modelled annual decline in δ15C in North Atlantic surface waters between 1950 and 1993 (0.025‰/yr; Kortzinger and Quay, 2003). We used a Suess correction based on the marine environment as a majority of herring gulls in Atlantic Canada consume marine prey (Pierotti and Annett, 1991), and the Suess effect in the world’s oceans is of a similar magnitude to that in the atmosphere (Gruber et al., 1999; Kelling et al., 1993).
We have assumed that, after applying the correction for the Suess Effect, baseline $\delta^{13}C$ values remained constant over time at each colony. There were no recorded changes in zooplankton $\delta^{15}N$ in central California from 1951 to 2001 (Rau et al., 2003), and we have no reason to suspect changes in ecosystem baseline $\delta^{15}N$ in Atlantic Canada, so $\delta^{15}N$ values were not adjusted and were assumed to have remained constant over the period of this study. All statistical analyses were conducted using $\delta^{13}C$ values adjusted for lipid content, and the adjustment for the Suess Effect was applied as indicated in the Results.

2.4. Statistical analysis

Our dataset included Hg values for five individual eggs from each colony from 1972 to 1988 and three pooled samples of five eggs each from 1992 to 2008. Since the variance of Hg values in individual eggs was greater than in pooled samples, we mathematically pooled the pre-1992 data by using the arithmetic mean Hg for the five eggs from each colony. This improved the homogeneity of variances, since all data points were mean values or pooled samples of 5 eggs.

We first examined trends in Hg over time using linear regressions, using Hg data unadjusted for dietary change. We did the same to assess temporal trends in $\delta^{13}N$ and $\delta^{15}C$. We then assessed relationships between Hg and both stable isotope values at each colony using linear regressions in SYSTAT 13 (SYSTAT Software Inc., Chicago, IL). When we found a significant relationship between Hg and $\delta^{13}N$ (or $\delta^{15}C$), we adjusted Hg concentrations for dietary influence (Braune, 2007; Weseloh et al., 2011):

$$H_{\text{adjusted}} = H_{\text{measured}} + A \times (\delta^{13}N_{\text{average}} - \delta^{13}N_{\text{measured}})$$

where A is the regression coefficient for Hg-$\delta^{13}N$ relationships for each site, and $(\delta^{13}N_{\text{average}} - \delta^{13}N_{\text{measured}})$ calculates how each sample’s $\delta^{13}N$ differed from the mean at each site (this is the mathematical equivalent of using the residual Hg values after regression of Hg and $\delta^{13}N$). Diet-adjusted Hg concentrations were used in subsequent temporal analyses. In all regressions, outliers were identified as those points with absolute values of Studentized residuals > 3.00 (Rousseeuw and Leroy, 1987).

To examine changes in diet-adjusted Hg over time at each island, we used change-point regression (also called break-point, segmented, or piece-wise regression; Gujarati, 1988; Pekarik and Weseloh, 1998). This technique has been useful in examining temporal trends in seabird egg contaminant levels elsewhere (de Solla et al., 2010; Weseloh et al., 2011). Using WILDSPEC™ 3.10 (Environment Canada, Ottawa, ON), we constructed a series of regression models to test whether Hg trends differed before and after a change-point year. Change-point year and best-fitting model were determined using a likelihood ratio test. For each colony, possible change-point years were those with at least 3 sampling periods after the start or before the end of the dataset.

3. Results

Unadjusted Hg concentrations in herring gull eggs decreased over time at Manawagonish Island ($\beta = -0.007, r^2 = 0.34, p = 0.007$) in the Bay of Fundy and at Île du Corossol ($\beta = -0.005, r^2 = 0.24, p = 0.046$, one outlier removed) in the Gulf of St. Lawrence (Fig. 1). There were no significant Hg trends at the other three colonies (all $r^2 < 0.2$, all $p > 0.06$). Mean unadjusted Hg concentrations for all colonies and years are found in Table S-1 in the supplementary material.

To assess if diets of the herring gulls changed over time, we looked at temporal trends in egg $\delta^{15}N$ and $\delta^{13}C$. $\delta^{15}N$ values decreased significantly over time in gull eggs at Gulf Island ($\beta = -0.096, r = 0.69, p = 0.00002$) in Newfoundland, Manawagonish Island ($\beta = -0.044, r^2 = 0.38, p = 0.004$) and Île du Corossol ($\beta = -0.100, r^2 = 0.35, p = 0.009$) (Fig. S-2). Lipid-adjusted $\delta^{13}C$ values decreased significantly over time at only at Kent Island ($\beta = -0.068, r^2 = 0.36, p = 0.007$) in the Bay of Fundy (Fig S-3). For the other four colonies, temporal trends in $\delta^{13}C$ were not significant ($p > 0.08$), yet the slopes (β values) were all negative (from −0.025 to −0.013). Since these negative slopes were consistent with the Suess Effect, we adjusted $\delta^{13}C$ values for the Suess Effect (see Eq. (2) in Methods) in all subsequent analyses.

To account for the influence of dietary shifts on egg Hg concentrations, we assessed the relationships between Hg and $\delta^{13}C$ and $\delta^{15}N$. We found significant positive associations between Hg and $\delta^{15}N$ at Gulf Island ($\beta = 0.164, t = 3.03, p = 0.009$), Île du Corossol ($\beta = 0.075, t = 3.55, p = 0.003$), and Manawagonish Island ($\beta = 0.145, t = 2.80, p = 0.012$) (Fig. 2). These associations indicated that some of the variation in egg Hg concentrations was related to changes in the trophic position ($\delta^{15}N$) of prey comprising the gulls’ diet. Once an outlier was removed (Kent Island, 1980), there were no significant relationships between Hg and $\delta^{13}C$ at any site ($p > 0.13$) (Fig. S-4).

After adjusting the Hg concentrations in the herring gull eggs for the observed diet shifts (using $\delta^{15}N$ values) at three colonies, there was a significant increase in Hg at Gulf Island, Newfoundland from 1977 to 1992 ($\beta \pm S.E. = 0.0243 \pm 0.005, r^2 = 0.95, p = 0.003$), followed by a significant drop between 1992 and 1996 ($p < 0.001$) and an increase from 1996 to 2008 ($p = 0.01$); the slope from 1977 to 1992 was no different from that between 1996 and 2008 ($p = 0.99$; Fig. 3A). In contrast, there were no significant trends in diet-adjusted Hg over time at Manawagonish Island or Île du Corossol ($p > 0.6$) (Fig. 3B and C).

Fig. 1. Herring gull eggs showed significant declines in unadjusted Hg ($\pm S.E., \mu g/g$ dry weight) over time at Île du Corossol (B) and Manawagonish Island (D). No significant temporal trends were observed at Gulf Island (A), Kent Island (C) or Sable Island (E). Outliers (open circles) were not included in the regression analyses.
4. Discussion

In this study, we documented Hg concentrations in herring gull eggs over a 36-year period (1972–2008) from five sites in Atlantic Canada. Two colonies (Manawagonish Island and Île du Corossol) exhibited decreasing trends in unadjusted Hg over time, but these trends became non-significant once Hg concentrations were adjusted for diet shifts. We found only one colony where diet-adjusted Hg had any significant temporal trend: Gull Island, Newfoundland, where Hg increased from 1977 to 1992, dropped sharply, and then increased at a similar rate from 1996 to 2008 (back to levels seen at the previous peak in 1992). We also detected significant trophic shifts (as indicated by decreasing trends in δ¹⁵N and δ¹³C) over time at four colonies: Gull Island, Île du Corossol, Kent Island and Manawagonish Island (assuming baseline δ¹⁵N and δ¹³C remained relatively constant at each site over the study period).

Major oceanographic changes, and their effects on forage fish, around Newfoundland in the early-mid 1990s are well documented (Carscadden et al., 2001; Regehr and Rodway, 1999). These changes may be related to the temporal trends observed in Hg and δ¹⁵N in the Gull Island herring gull eggs. Fisheries discards from the Atlantic cod (Gadus morhua) fishery likely comprised a significant portion of gulls’ diet (Pierotti and Annett, 1991), but decreasing size of fish in discards, and the total closure of the cod fishery in Newfoundland in July 1992 likely resulted in an abrupt change in the gulls’ diet (Montevecchi, 2001) and a significant decrease in δ¹⁵N in gull eggs over time. Gull predation on sympatric Leach’s storm-petrels (Oceanodroma leucorhoa) and black-legged kitiwakes (Rissa tridactyla) has increased since the 1970s, as the gulls’ reliance on fisheries discards during egg formation in April/May decreased (Robertson et al., 2001; Stenhouse and Montevecchi, 1999; Stenhouse et al., 2000). Delays in capelin (Mallotus villosus) spawning have also caused changes in the composition of gull diets in Newfoundland (Massaro et al., 2000; Carscadden et al., 2002). Hg levels in Leach’s storm-petrels are higher than in herring gulls in Atlantic Canada (Burgess, unpubl. data; Elliott et al., 1992). Importantly, the high variation found in δ¹³C in 1996 (Fig. S-3) at Gull Island could represent diverse foraging tactics (Pierotti and Annett, 1991) used by individual gulls represented in the pooled egg samples. While the Hg levels in the eggs at Gull Island were adjusted for changes in δ¹⁵N, the abrupt change in Hg levels at Gull Island between 1992 and 1996 may be related to possible diet shifts associated with the closure of the cod fishery that are not fully reflected by the δ¹⁵N data. In the Gulf of St. Lawrence, including Île du Corossol, an influx of cold oceanic water in 1990–1991 negatively affected some seabird populations through prey limitation, or shifts to lower trophic level prey (Gaston et al., 2009). Seabirds in
the Bay of Fundy have also experienced significant bottom-up influences on diet and foraging. The decline of Atlantic herring (Clupea harengus) in the Bay of Fundy area has likely contributed to decreased reproductive success of several seabird species (Gaston et al., 2009). The reduction of high-trophic-level fisheries discards has also negatively affected the abundance of gulls in Atlantic Canada (Boyne and Beukenk, 2004; Boyne and McKnight, 2005). All these changes in prey availability may account for the dietary shifts suggested by the declining trends in δ^{15}N and δ^{13}C seen in eggs at four herring gull colonies.

The change-point trend in diet-adjusted Hg in eggs from Gull Island contrasts with Hg trends in herring gull eggs from the Great Lakes region, where the majority of sites showed significant decreases in Hg over a similar time period (Weseloh et al., 2011), although the Hg concentrations in Atlantic herring gull eggs were generally lower than those recorded in the Great Lakes. None of the Great Lakes sites showed an increase in herring gull egg Hg over time. Increasing egg Hg levels have been observed in several species of seabirds in the Canadian Arctic (Braune, 2007). Importantly, correcting Hg for diet variability using stable isotopes allows insight into environmental Hg, and reduces the possibility that a diet shift caused a change in Hg concentrations in a predator.

At Manawanish Island and Île du Corosol, we detected a significant decrease in unadjusted Hg over time, but this trend disappeared once Hg was adjusted for changes in trophic position (using δ^{15}N). Herring gull δ^{15}N values decreased over the same period by about 2.5‰ (Table S-1), which is the result of either a shift to lower trophic level prey (assuming that prey δ^{15}N remained unchanged), or a shift in the ecosystem's baseline δ^{15}N value. Gulls from Manawanish began foraging at the Crane Mountain Landfill in Grand Bay-Westfield (approximately 10 km from Manawanish Island) when the site opened in the mid 1990s (A.W. Diamond, pers. comm.). A dietary shift was not reflected in δ^{13}C because gulls likely foraged in the Saint John River outflow during the entire monitoring period, and freshwater (river) and terrestrial (landfill) δ^{13}C values may be similar (Peterson and Fry, 1987).

The unadjusted egg Hg concentrations observed in herring gulls in Atlantic Canada were similar to those observed in Long Island, NY in 1989–1994 (0.12–0.46 µg/g, dry weight) (Burger and Gochfeld, 1995), in the Gulf of St. Lawrence in 2006–2007 (0.43 µg/g, dry weight) (Lavoie et al., 2010a), and were similar to the lower concentrations observed in the Great Lakes in 2009 (0.26–0.98 µg/g, converted to dry weight assuming 75% moisture) (Weseloh et al., 2011), while they were lower than most herring gull eggs collected in the North and Baltic Seas of Germany in 2007 (0.6–1.5 µg/g, converted to dry weight) (Rüdel et al., 2010).

Effect levels of Hg in eggs vary by species, and by the criteria used to assess effects. Estimates include broad ranges across multiple taxa (0.5–2.0 µg/g, wet weight for birds in general, Thompson, 1996), to more precise estimates (>0.60 µg/g, wet weight for non-marine birds; Shore et al., 2011). In herring gulls, the LC_{50} (median lethal concentration) determined experimentally by injecting MeHg into eggs, was 0.28 µg/g (wet weight; approximately 1.1 µg/g, dry weight assuming 75% moisture) (Heinz et al., 2009). When this herring gull egg-injection study was repeated with control hatching success that was more typical of wild populations, the LC_{50} was 0.56 µg/g (wet weight; approximately 2.2 µg/g, dry weight) (Burgess, unpubl. data). Using the LC_{50} from injection studies is overly protective when applied to wild eggs because injected MeHg is more toxic than maternally-transferred MeHg (Heinz et al., 2009, 2006). Even with this conservative effect threshold, none of the eggs in our study exceeded the lowest estimate of herring gulls' LC_{50}, and in only one case was measured Hg > 1.0 µg/g, dry weight. Thus, the toxicological risk of reproductive impairment associated with current levels of Hg appears lower for herring gulls than for other seabirds in Atlantic Canada (Burgess, unpubl. data).

Interpreting time-series data of biomagnified contaminants requires an understanding of concurrent trophic dynamics. Dietary shifts in top predators that often accompany large-scale climatic variability (Duran et al., 2009) can affect Hg deposited into seabird eggs. For generalist predators like gulls that are adapted to using human-dominated landscapes, dietary changes may also stem from changing land use practices (e.g., landfill management) or resource extraction industries (e.g., fisheries) (Chapadelaine and Rail, 1997; Votier et al., 2004; Weiser and Powell, 2011). Stable nitrogen isotopes offer a continuous scale with which to determine trophic position, and there are now methods to correct for Suess Effect-related changes in δ^{13}C in consumers over time (Farmer and Leonard, 2011) allowing greater insight into actual changes in environmental Hg on decadal scales. Stable C and N isotopes are now commonly used to aid in the interpretation of contaminant concentrations in fish and wildlife: including studies of biomagnification (Campbell et al., 2008; Jaeger et al., 2009; Lavoie et al., 2010b), spatial patterns (Braune et al., 2002; Day et al., 2005; Dietz et al., 2004; Gebbink et al., 2011), and temporal trends (Braune, 2007; Hebert and Weseloh, 2006; Rigét et al., 2007).

Considering Hg inputs into Atlantic Canada from human sources, anthropogenic Hg emissions in Canada have declined by more than 90% since the 1970s and by more than 60% in the United States since 1990 (Sunderland and Chmura, 2000a, 2000b; U.S. EPA, 2005). However, the consequent reductions in atmospheric Hg deposition from North American sources have been largely offset by increased deposition from global Hg sources (Sunderland et al., 2008). Overall, Sunderland et al. (2008) showed a small decline in atmospheric Hg deposition rates from the early 1990s to the early 2000s in the Gulf of Maine. Monitoring of Hg trends (since 1990) in blue mussels from 15 sites around the Gulf of Maine revealed steady Hg levels at 12 sites and decreasing Hg trends at three sites (Kimbrough et al., 2008). In their review of Hg sources and fate in the Gulf of Maine, Sunderland et al. (2012) concluded that there was little indication of Hg declines in blue mussels and seabird eggs, except in areas where industrial point-source Hg inputs had been reduced in recent decades. These conclusions are supported by the findings of our study.

Multi-site or multi-species assessments of long-term Hg trends in top predators indicate that temporal trends differ greatly across ecosystems (AMAP, 2011; Chalmers et al., 2011; Pereira et al., 2009). Here we have shown that after adjusting Hg concentrations for dietary shifts, significant temporal trends were found in Hg in herring gull eggs at only one of five colonies studied. This suggests that environmental Hg in inshore coastal ecosystems has remained relatively constant at most sites in Atlantic Canada over the last 36 years.

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