Environmental Pollution 172 (2013) 216–222

Contents lists available at SciVerse ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

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

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ARTICLE INFO

*Article history:*
Received 6 January 2012
Received in revised form
17 August 2012
Accepted 8 September 2012

*Keywords:*
Herring gull
Egg
Mercury
Stable isotope
Atlantic

ABSTRACT

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 ($\delta^{15}N$), these trends disappeared. Decreasing temporal trends of stable isotopes in gull eggs were observed at four sites, suggesting shifts in gull diets. 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., Aebischer 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 ($^{13}C/^{12}C$, or $\delta^{13}C$, and $^{15}N/^{14}N$, or $\delta^{15}N$) will reflect the consumer's diet at the time of tissue synthesis (Hobson, 1995; Hobson and Clark, 1992). $\delta^{13}C$ 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 $^{13}C$ (France, 1995; Goericke and Fry, 1994; Peterson and Fry, 1987; Popp et al., 1998), while $\delta^{15}N$ values increase 2–5‰ with each trophic level because $^{14}N$ 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 $^{13}C$) have altered the baseline $\delta^{13}C$ value globally, including in the North Atlantic (Quay et al., 2007), but this can be taken into account

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http://dx.doi.org/10.1016/j.envpol.2012.09.001

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 biomonitors 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 (Lavoie 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 Corossol, Québec, 50°05′N, 66°23′W), two in the Bay of Fundy (Kent Island, New Brunswick, 44°34′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 sampled at each colony beginning in 1972 (Corossol, 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 within 24 h of collection and then were shipped by overnight courier to Environment Canada's National Wildlife Research Centre (NWRC; Ottawa, ON), where egg contents were stored at −40 °C 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 3030b, AAS (Waltham, MA, USA) and Varian VGA-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; Altec Ltd., Prague, Czech Republic), which is a direct 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 ± 7.1% (mean relative standard deviation ($r_{sd}$) ± S.D.) Regression analysis showed a strong association between the CVAAS vs. AMA-254 data (CVAAS = 0.98 * AMA + 0.12, $R^2 = 0.86$).

Of the 185 total samples, 100 (54%) were analysed in duplicate or triplicate. Average variability ($r_{sd}$ ± S.D.) of these replicate samples was 1.86 ± 2.69%. Within each analytical run, Hg concentrations were corrected for recoveries of certified reference materials (mean ± S.D. recovery among all analytical runs, certified concentration): 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, $n = 14$), and TORT-2 (lobster hepatopancreas, 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 delta notation in parts per thousand (‰, per mil) relative to the above standards, and mean values are reported for samples taken in duplicate. Analytical precision of both $\delta^{13}C$ and $\delta^{15}N$, based on repeat measures of a standard (C-55), was ±0.2‰.

Lipids are naturally depleted in $^{13}C$, and because individual eggs vary in lipid content, lipids 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

$$\delta^{13}C_{normalized} = \delta^{13}C_{uncorrected} - 3.32 + 0.99 \times C:N \tag{1}$$

Herring gull C:N in our samples ranged from 5.74 to 10.89 (mean ± SD: 7.5 ± 0.5), and 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 $\delta^{13}C$ values for the Suess Effect using the post-1950 portion of their Eq. (2):

$$\delta^{13}C_{corrected_i} = \delta^{13}C_{normalized_i} - b_{his} \times (1950 - t_i) - b_{mod} \times (t_i - 1950) \tag{2}$$

where $\delta^{13}C_{normalized}$ is the $\delta^{13}C$ value normalized for lipid content, $b_{his}$ is the historical annual decline in $\delta^{13}C$ (0.007‰; Tagliabue and Bopp, 2008), and $b_{mod}$ is the modelled annual decline in $\delta^{13}C$ in North Atlantic surface waters between 1950 and 1993 (0.026‰; Körtzinger 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; Keeling et al., 1995). 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 $\delta^{15}N$ and $\delta^{13}C$. 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 $\delta^{15}N$ (or $\delta^{13}C$), we adjusted Hg concentrations for dietary influence (Braune, 2007; Weseloh et al., 2011):

$$Hg_{adj} = Hg_{measured} + A \times \left(\delta^{15}N_{average} - \delta^{15}N_{measured}\right) \quad (3)$$

where A is the regression coefficient for Hg-$\delta^{15}N$ relationships for each site, and $(\delta^{15}N_{average} - \delta^{15}N_{measured})$ calculates how each sample's $\delta^{15}N$ differed from the mean at each site (this is the mathematical equivalent of using the residual Hg values after regression of Hg and $\delta^{15}N$). 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 contaminants elsewhere (de Solla et al., 2010; Weseloh et al., 2011). Using WILDSPACE™ 3.1.0 (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 Gull Island ($\beta = -0.096$, $r^2 = 0.69$, $p = 0.00002$) 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 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 ($\beta$ 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 Gull 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 Gull 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).

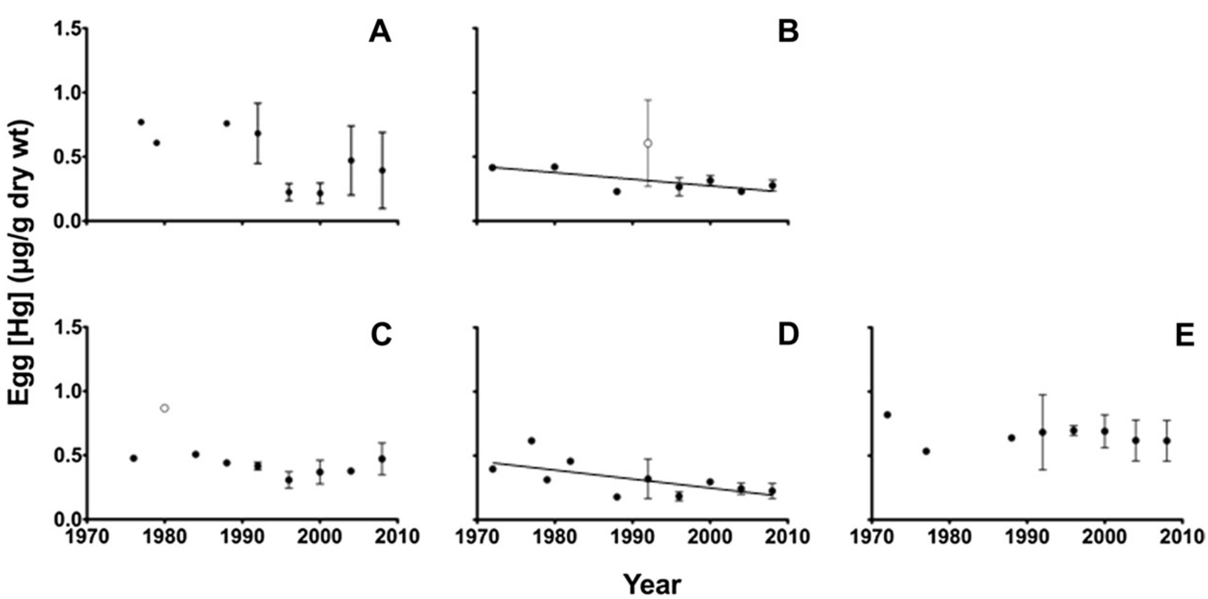


**Fig. 1.** Herring gull eggs showed significant declines in unadjusted Hg (±S.E., µg/g dry weight) over time at Île du Corossol (B) and Manawagonish Island (D). No significant temporal trends were observed at Gull Island (A), Kent Island (C) or Sable Island (E). Outliers (open circles) were not included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

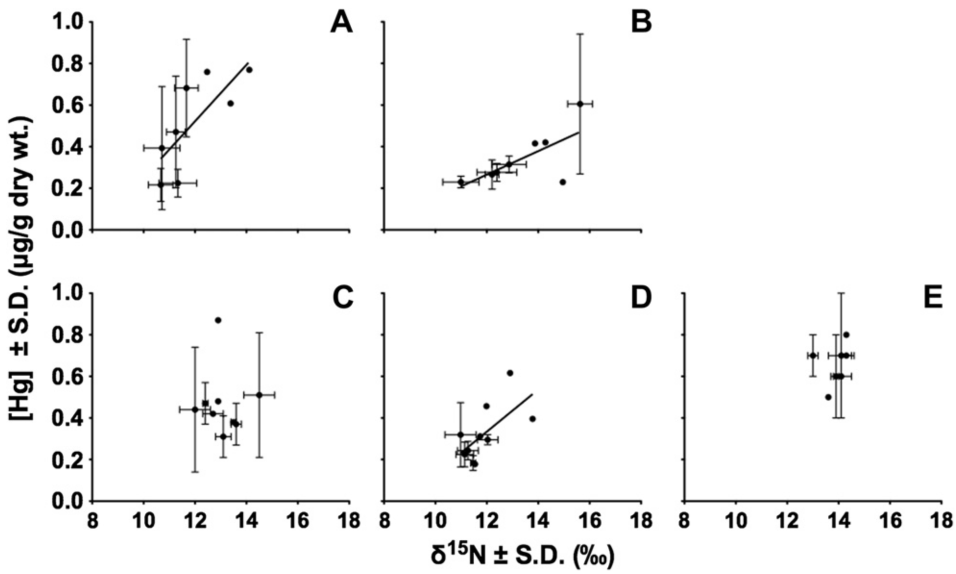


**Fig. 2.** Herring gull eggs from Gull Island (A), Île du Corossol (B) and Manawagonish Island (D) showed significant positive associations between unadjusted Hg (±S.D., µg/g dry weight) and $\delta^{15}N$ (±S.D., ‰). No relationships were observed at Kent Island (C) or Sable Island (E).

## 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 $\delta^{15}N$ and $\delta^{13}C$) over time at four colonies: Gull Island, Île du Corossol, Kent Island and Manawagonish Island (assuming baseline $\delta^{15}N$ and $\delta^{13}C$ 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 $\delta^{15}N$ 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 $\delta^{15}N$ in gull eggs over time. Gull predation on sympatric Leach's storm-petrels (*Oceanodroma leucorhoa*) and black-legged kittiwakes (*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 $\delta^{13}C$ 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 $\delta^{15}N$, 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 $\delta^{15}N$ 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

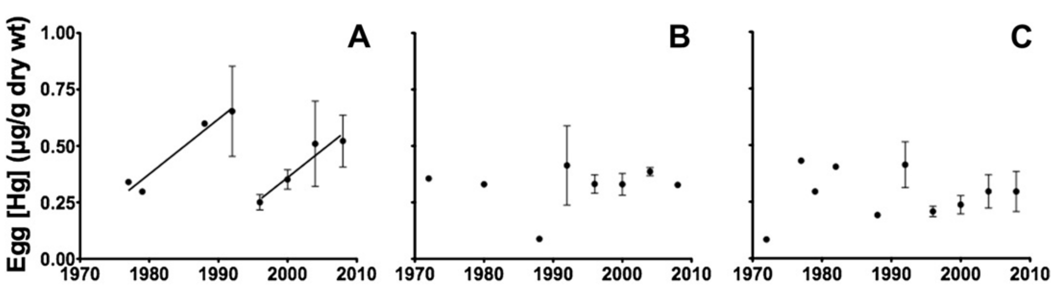


**Fig. 3.** Mean diet-adjusted Hg concentrations (±S.E., µg/g dry weight) in herring gull eggs at Gull Island (A) showed an increasing trend from 1977 to 1992, then a sharp drop between 1992 and 1996, followed by a similar increase from 1996 to 2008. No temporal trends in diet-adjusted Hg were observed at Île du Corossol (B) or Manawagonish Island (C).

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 Beukens, 2004; Boyne and McKnight, 2005). All these changes in prey availability may account for the dietary shifts suggested by the declining trends in $\delta^{15}N$ and $\delta^{13}C$ 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 Manawagonish Island and Île du Corossol, we detected a significant decrease in unadjusted Hg over time, but this trend disappeared once Hg was adjusted for changes in trophic position (using $\delta^{15}N$). Herring gull $\delta^{15}N$ 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 $\delta^{15}N$ remained unchanged), or a shift in the ecosystem's baseline $\delta^{15}N$ value. Gulls from Manawagonish began foraging at the Crane Mountain Landfill in Grand Bay-Westfield (approximately 10 km from Manawagonish Island) when the site opened in the mid 1990s (A.W. Diamond, pers. comm.). A dietary shift was not reflected in $\delta^{13}C$ because gulls likely foraged in the Saint John River outflow during the entire monitoring period, and freshwater (river) and terrestrial (landfill) $\delta^{13}C$ 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 (Durant 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) (Chapdelaine 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 $\delta^{13}C$ 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., 2012; 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.

## Acknowledgements

We thank the many people who assisted with egg collections over the years, including those from Environment Canada, New Brunswick Museum, Bowdoin College, Memorial University of Newfoundland, and Sable Island. Thanks to staff in the NWRC Tissue Preparation Lab and National Wildlife Specimen Bank. P. Dunlop assisted with the Hg analysis, and Wendy Abdi conducted the stable isotope analyses at the University of Ottawa. Environment Canada provided funding for this project. Finally, we thank J. Chardine, G. Robertson, B. Braune, and three anonymous reviewers for suggestions that improved this manuscript.

## Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2012.09.001.

## References

Aebischer, N.J., Coulson, J.C., Colebrook, J.M., 1990. Parallel long-term trends across four marine trophic levels and weather. Nature 347, 753–755.
AMAP, 2011. AMAP Assessment 2011: Mercury in the Arctic. Arctic Monitoring and Assessment Programme, Oslo.
Bond, D., 2008. A Comparison of Samples Analyzed by CVTASS and the AMA-254 Direct Hg Analyzer: Hg in Various Tissues, Report -METRES-07-01. National Wildlife Research Centre, Environment Canada, Ottawa, ON.
Bond, A.L., Diamond, A.W., 2009. Total and methyl mercury concentrations in seabird feathers and eggs. Archives of Environmental Contamination and Toxicology 56, 286–291.
Bond, A.L., Jones, I.L., 2009. A practical introduction to stable-isotope analysis for seabird biologists: approaches, cautions and caveats. Marine Ornithology 37, 183–188.
Boyne, A.W., Beukens, J.T., 2004. Census of Gulls and Other Seabirds along the Coast of Mainland Nova Scotia. Canadian Wildlife Service, Atlantic Region, Dartmouth, NS. Ms. report.
Boyne, A.W., McKnight, J.L., 2005. Census of Terns and Gulls in Prince Edward Island, 2004. Canadian Wildlife Service, Atlantic Region, Dartmouth, NS. Ms. report.
Braune, B.M., 2007. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975–2003. Environmental Pollution 148, 599–613.
Braune, B.M., Donaldson, G.M., Hobson, K.A., 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. Environmental Pollution 117, 133–145.
Braune, B.M., Gaskin, D.E., 1987. A mercury budget for the Bonaparte's Gull during autumn moult. Ornis Scandinavica 18, 244–250.
Braune, B.M., Outridge, P.M., Fisk, A.T., Muir, D.C.G., Helm, P.A., Hobbs, K., Hoekstra, P.F., Kuzyk, Z.A., Kwan, M., Letcher, R.J., Lockhart, W.L., Norstrom, R.J., Stern, G.A., Stirling, I., 2005. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: an overview of spatial and temporal trends. Science of the Total Environment 351–352, 4–56.
Burger, J., Gochfeld, M., 1995. Heavy metal and selenium concentrations in eggs of Herring Gulls (*Larus argentatus*): temporal differences from 1989 to 1994. Archives of Environmental Contamination and Toxicology 29, 192–197.
Burger, J., Gochfeld, M., 2004. Marine birds as sentinels of environmental pollution. EcoHealth 1, 263–274.
Campbell, L., Verburg, P., Dixon, D.G., Hecky, R.E., 2008. Mercury biomagnification in the food web of Lake Tanganyika (Tanzania, East Africa). Science of the Total Environment 402, 184–191.
Carscadden, J.E., Frank, K.T., Leggett, W.C., 2001. Ecosystem changes and the effects on capelin (*Mallotus villosus*), a major forage species. Canadian Journal of Fisheries and Aquatic Sciences 58, 73–85.
Carscadden, J.E., Montevecchi, W.A., Davoren, G.K., Nakashima, B.S., 2002. Trophic relationships among capelin (*Mallotus villosus*) and seabirds in a changing ecosystem. ICES Journal of Marine Science 59, 1027–1033.
Chalmers, A.T., Argue, D.M., Gay, D.A., Brigham, M.E., Schmidt, C.J., Lorenz, D.L., 2011. Mercury trends in fish from rivers and lakes in the United States, 1969–2005. Environmental Monitoring and Assessment 175, 175–191.
Chapdelaine, G., Rail, J.-F., 1997. Relationship between cod fishery activities and the population of herring gulls on the North Shore of the Gulf of St Lawrence, Québec, Canada. ICES Journal of Marine Science 54, 708–713.
Day, R.D., Roseneau, D.G., Vander Pol, S.S., Hobson, K.A., Donard, O.F.X., Pugh, R.S., Moors, A.J., Becker, P.R., 2012. Regional, temporal, and species patterns of mercury in Alaskan seabird eggs: mercury sources and cycling or food web effects? Environmental Pollution 166, 226–232.
de Solla, S.R., Weseloh, D.V.C., Hebert, C.E., Pekarik, C., 2010. Impact of changes in analytical techniques for the measurement of polychlorinated biphenyls and organochlorine pesticides on temporal trends in Herring Gull eggs. Environmental Toxicology and Chemistry 29, 1476–1483.
Dietz, R., Rigét, F., Hobson, K.A., Heide-Jørgensen, Møller, P., Cleemann, M., De Boer, J., Glasius, M., 2004. Regional and inter annual patterns of heavy metals, organochlorines and stable isotopes in narwhals (*Monodon monoceros*) from West Greenland. Science of the Total Environment 331, 88–105.
Drent, R., Daan, S., 1980. The prudent parent: energetic adjustments in avian breeding. Ardea 68, 225–252.
Drinkwater, K.F., 1996. Atmospheric and oceanic variability in the northwest Atlantic during the 1980s and early 1990s. Journal of Northwest Atlantic Fisheries Science 18, 77–97.
Durant, J.M., Hjermann, D.Ø., Frederiksen, M., Charraissin, J.B., Le Maho, Y., Sabarros, P.S., Crawford, R.J.M., Stenseth, N.C., 2009. Pros and cons of using seabirds as ecological indicators. Climate Research 39, 115–129.
Elliott, J.E., Scheuhammer, A.M., Leighton, F.A., Pearce, P.A., 1992. Heavy metal and metallothionein concentrations in Atlantic Canadian seabirds. Archives of Environmental Contamination and Toxicology 22, 63–73.
Farmer, R.G., Leonard, M.L., 2011. Long-term feeding ecology of Great Black-backed Gulls (*Larus marinus*) in the northwest Atlantic: 110 years of feather isotope data. Canadian Journal of Zoology 89, 123–133.
France, R.L., 1995. Carbon-13 enrichment in benthic compared to planktonic algae: foodweb implications. Marine Ecology Progress Series 124, 307–312.
Gaston, A.J., Bertram, D.F., Boyne, A.W., Chardine, J.W., Davoren, G.K., Diamond, A.W., Hedd, A., Montevecchi, W.A., Hipfner, J.M., Lemon, M.J.F., Mallory, M.L., Rail, J.-F., Robertson, G.J., 2009. Changes in Canadian seabird populations and ecology since 1970 in relation to changes in oceanography and food webs. Environmental Reviews 17, 267–286.
Gebbink, W.A., Letcher, R.J., Burgess, N.M., Champoux, L., Elliott, J.E., Hebert, C.E., Martin, P., Wayland, M., Weseloh, D.V.C., WIlson, L., 2011. Perfluoroalkyl carboxylates and sulfonates and precursors in relation to dietary source tracers in the eggs of four species of gulls (Larids) from breeding sites spanning Atlantic to Pacific Canada. Environment International 37, 1175–1182.
Goericke, R., Fry, B., 1994. Variations of marine plankton $\delta^{13}C$ with latitude, temperature and dissolved $CO_2$ in the world ocean. Global Biogeochemical Cycles 8, 85–90.
Golden, N.H., Rattner, B.A., 2003. Ranking terrestrial vertebrate species for utility in biomonitoring and vulnerability to environmental contaminants. Reviews in Environmental Toxicology 176, 67–136.
Goodale, M.W., Evers, D.C., Meirzykowski, S.E., Bond, A.L., Burgess, N.M., Otorowski, C.I., Welch, L.J., Hall, C.S., Ellis, J.C., Allen, R.B., Diamond, A.W., Kress, S.W., Taylor, R.J., 2008. Marine foraging birds as bioindicators of mercury in the Gulf of Maine. EcoHealth 5, 409–425.
Gruber, N., Keeling, C.D., Bacastow, R.B., Guenther, P.R., Lueker, T.J., Wahlen, M., Meijer, H.A.J., Mook, W.G., Stocker, T.F., 1999. Spatiotemporal patterns of carbon-13 in the global surface oceans and the oceanic Suess effect. Global Biogeochemical Cycles 13, 307–335.
Gujarati, D.N., 1988. Basic Econometrics. McGraw-Hill, New York, NY.
Hebert, C.E., Hobson, K.A., Shutt, J.L., 2000. Changes in food web structure affect rate of PCB decline in herring gull (*Larus argentatus*) eggs. Environmental Science & Technology 34, 1609–1614.
Hebert, C.E., Norstrom, R.J., Weseloh, D.V., 1999. A quarter century of environmental surveillance: the Canadian wildlife Service's Great Lakes Herring Gull monitoring Program. Environmental Reviews 7, 147–166.
Hebert, C.E., Weseloh, D.V.C., 2003. Assessing temporal trends in contaminants from long-term avian monitoring programs: the influence of sampling frequency. Ecotoxicology 12, 141–151.
Hebert, C.E., Weseloh, D.V., 2006. Adjusting for temporal change in trophic position results in reduced rates of contaminant decline. Environmental Science & Technology 40, 5624–5628.
Hebert, C.E., Weseloh, D.V., Gauthier, L.T., Arts, M.T., Letcher, R.J., 2009. Biochemical tracers reveal intra-specific differences in the food webs utilized by individual seabirds. Oecologia 160, 15–23.
Heinz, G.H., Hoffman, D.J., Klimstra, J.D., Stebbins, K.R., Kondrad, S.L., Erwin, C.A., 2009. Species differences in the sensitivity of avian embryos to methylmercury. Archives of Environmental Contamination and Toxicology 56, 129–138.
Heinz, G.H., Hoffman, D.J., Kondrad, S.L., Erwin, C.A., 2006. Factors affecting the toxicity of methylmercury injected into eggs. Archives of Environmental Contamination and Toxicology 50, 264–279.
Hobson, K.A., 1995. Reconstructing avian diets using stable-carbon and nitrogen isotope analysis of egg components: patterns of isotopic fractionation and turnover. Condor 97, 752–762.
Hobson, K.A., Clark, R.G., 1992. Assessing avian diets using stable isotopes I: turnover of $^{13}C$ in tissues. Condor 94, 181–188.
Hobson, K.A., Sirois, J., Gloutney, M.L., 2000. Tracing nutrient allocation to reproduction with stable isotopes: a preliminary investigation using colonial waterbirds of Great Slave Lake. Auk 117, 760–774.
Jaeger, I., Hop, H., Gabrielsen, G.W., 2009. Biomagnification of mercury in selected species from an Arctic marine food web in Svalbard. Science of the Total Environment 407, 4744–4751.
Keeling, C.D., 1979. The suess effect: $^{13}$carbon and $^{14}$carbon interactions. Environment International 2, 229–300.
Keeling, C.D., Whorf, T.P., Wahlen, M., van der Plicht, J., 1995. Interannual extremes in the rate of rise of atmospheric carbon dioxide since 1980. Nature 375, 666–670.
Kelly, J.F., 2000. Stable isotopes of carbon and nitrogen in the study of avian and mammalian trophic ecology. Canadian Journal of Zoology 78, 1–27.
Kimbrough, K.L., Johnson, W.E., Lauenstein, G.G., Christensen, J.D., Apeti, D.A., 2008. An Assessment of Two Decades of Contaminant Monitoring in the Nation's Coastal Zone, vol. 74. NOAA Technical Memorandum NOS NCCOS, Silver Spring, MD.
Kojadinovic, J., Richard, P., Le Corre, M., Cosson, R.P., Bustamante, P., 2008. Effects of lipid extraction on $\delta^{13}C$ and $\delta^{15}N$ values in seabird muscle, liver and feathers. Waterbirds 31, 169–178.
Körtzinger, A., Quay, P.D., 2003. Relationship between anthropogenic $CO_2$ and the $^{13}C$ Suess effect in the North Atlantic Ocean. Global Biogeochemical Cycles 17, GB1005. http://dx.doi.org/10.1029/2001GB001427.
Koster, M.D., Ryckman, D.P., Weseloh, D.V.C., Struger, J., 1996. Mercury levels in Great Lakes Herring Gull (*Larus argentatus*) eggs, 1972–1992. Environmental Pollution 93, 261–270.
Lavoie, R.A., Champoux, L., Rail, J.-F., Lean, D.R.S., 2010a. Organochlorines, brominated flame retardants and mercury levels in six seabird species from the Gulf of St. Lawrence (Canada): relationships with feeding ecology, migration and molt. Environmental Pollution 158, 2189–2199.

Lavoie, R.A., Hebert, C.E., Rail, J.-F., Braune, B.M., Yumvihoze, E., Hill, L.G., Lean, D.R.S., 2010b. Trophic structure and mercury distribution in a Gulf of St. Lawrence (Canada) food web using stable isotope analysis. Science of the Total Environment 408, 5529–5539.

Lewis, S.A., Becker, P.H., Furness, R.W., 1993. Mercury levels in eggs, tissues, and feathers of Herring Gulls *Larus argentatus* from the German Wadden Sea coast. Environmental Pollution 80, 293–299.

Logan, J.M., Jardine, T.D., Miller, T.J., Bunn, S.E., Cunjak, R.A., Lutcavage, M.E., 2008. Lipid corrections in carbon and nitrogen stable isotope analyses: comparison of chemical extraction and modelling methods. Journal of Animal Ecology 77, 838–846.

Massaro, M., Chardine, J.W., Jones, I.L., Robertson, G.J., 2000. Delayed capelin (*Mallotus villosus*) availability influences predatory behaviour of large gulls on Black-legged Kittiwakes (*Rissa tridactyla*), causing a reduction in kittiwake breeding success. Canadian Journal of Zoology 78, 1588–1596.

Minagawa, M., Wada, E., 1984. Stepwise enrichment of $^{15}$N along food chains: further evidence and the relation between $\delta^{15}$N and animal age. Geochimica et Cosmochimica Acta 48, 1135–1140.

Mineau, P., Fox, A., Norstrom, R.J., Weseloh, D.V., Hallett, D., Ellenton, J.A., 1984. Using the herring gull to monitor levels of organochlorine contamination in the Canadian Great Lakes. In: Nriagu, J.O., Simmons, M.S. (Eds.), Toxic Contaminants in the Great Lakes. John Wiley and Sons, New York, pp. 426–452.

Monteiro, L.R., Furness, R.W., 2001. Kinetics, dose-response, and excretion of methylmercury in free-living adult Cory's Shearwaters. Environmental Science & Technology 35, 739–746.

Montevecchi, W.A., 2001. Interactions between fisheries and seabirds. In: Schreiber, E.A., Burger, J. (Eds.), Biology of Marine Birds. CRC Press, Boca Raton, pp. 527–557.

Montevecchi, W.A., Myers, R.A., 1996. Dietary changes of seabirds indicate shifts in pelagic food webs. Sarsia 80, 313–322.

Neugebauer, E.A., Sans Cartier, G.L., Wakeford, B.J., 2000. Methods for the determination of metals in wildlife tissues using various atomic absorption spectrophotometry techniques. Canadian Wildlife Service Technical Report 337, 3.1–3.12.

Nriagu, J.O., 1989. A global assessment of natural sources of atmospheric trace metals. Nature 338, 47–49.

Nriagu, J.O., Pacyna, J.M., 1988. Quantitative assessment of worldwide contamination of air, water and soils by trace metals. Nature 333, 134–139.

Oppel, S., Federer, R.N., O'Brien, D.M., Powell, A.N., Hollmén, T.E., 2010. Effect of lipid extraction on stable isotope ratios in avian egg yolk: is arithmetic correction a reliable alternative? Auk 127, 72–78.

Pearce, P.A., Peakall, D.B., Reynolds, L.M., 1979. Shell thinning and residues of organochlorines and mercury in seabird eggs, Eastern Canada, 1970–1976. Pesticides Monitoring Journal 13, 61–68.

Pekarik, C., Weseloh, D.V., 1998. Organochlorine contaminants in Herring Gull eggs from the Great Lakes, 1974–1995: change point regression analysis and short-term regression. Environmental Monitoring and Assessment 53, 77–115.

Pereira, M.G., Walker, L.A., Best, J., Shore, R.F., 2009. Long-term trends in mercury and PCB congener concentrations in gannet (*Morus bassanus*) eggs in Britain. Environmental Pollution 157, 155–163.

Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. Annual Review of Ecology and Systematics 18, 293–320.

Pierotti, R., Annett, C.A., 1991. Diet choice in the Herring Gull: constraints imposed by reproductive and ecological factors. Ecology 72, 319–328.

Pierotti, R.J., Good, T.P., 1994. Herring Gull (*Larus argentatus*). In: Poole, A., Gill, F. (Eds.), The Birds of North America, vol. 124. Cornell Lab of Ornithology, Ithaca, NY. Retrieved from the Birds of North America Online: http://bna.birds.cornell.edu/bna/species/124.

Popp, B.N., Laws, E.A., Bridigare, R.R., Dore, J.E., Hanson, K.L., Wakeham, S.G., 1998. Effect of phytoplankton cell geometry on carbon isotopic fractionation. Geochimica et Cosmochimica Acta 62, 69–77.

Post, D.M., Layman, C.A., Arrington, D.A., Takimoto, G., Quattrochi, J., Montaña, C.G., 2007. Getting to the fat of the matter: models, methods and assumptions for dealing with lipids in stable isotope analysis. Oecologia 152, 179–189.

Quay, P., Sonnerup, R., Stutsman, J., Maurer, J., Körtzinger, A., Padin, X.A., Robinson, C., 2007. Anthropogenic $CO_2$ accumulation rates in the North Atlantic Ocean from changes in the $^{13}C/^{12}C$ of dissolved inorganic carbon. Global Biogeochemical Cycles 21, GB1009. http://dx.doi.org/10.1029/2006GB002761.

Rau, G.H., Ohman, M.D., Pierrot-Bults, A., 2003. Linking nitrogen dynamics to climate variability off central California: a 51 year record based on $^{15}N/^{14}N$ in CalCOFI zooplankton. Deep-Sea Research Part II Topical Studies in Oceanography 40, 2431–2447.

Regehr, H.M., Rodway, M.S., 1999. Seabird breeding performance during two years of delayed capelin arrival in the northwest Atlantic: a multi-species comparison. Waterbirds 22, 60–67.

Rigét, F., Dietz, R., Born, E.W., Sonne, C., Hobson, K.A., 2007. Temporal trends of mercury in marine biota of west and northwest Greenland. Marine Pollution Bulletin 54, 72–80.

Robertson, G.J., Fifield, D.A., Massaro, M., Chardine, J.W., 2001. Changes in nesting-habitat use of large gulls breeding in Witless Bay, Newfoundland. Canadian Journal of Zoology 79, 2159–2167.

Rousseeuw, P.J., Leroy, A.M., 1987. Robust Regression and Outlier Detection. J. Wiley and Sons, New York.

Rüdel, H., Fliedner, A., Kösters, J., Schröter-Kermani, C., 2010. Twenty years of elemental analysis of marine biota within the German Environmental Specimen Bank – a thorough look at the data. Environmental Science and Pollution Research 17, 1025–1034.

Scheuhammer, A.M., Bond, D.E., 1991. Factors affecting the determination of total mercury in biological samples by continuous-flow cold vapor atomic absorption spectrophotometry. Biological Trace Element Research 31, 119–139.

Shore, R.F., Pereira, M.G., Walker, L.A., Thompson, D.R., 2011. Mercury in nonmarine birds and mammals. In: Beyer, W.N., Meador, J.P. (Eds.), Environmental Contaminants in Biota: Interpreting Tissue Concentrations, second ed. CRC Press, Boca Raton, FL, pp. 609–624.

Spalding, M.G., Frederick, P.C., McGill, H.C., Bouton, S.N., McDowell, L.R., 2000. Methylmercury accumulation in tissues and its effects on growth and appetite in captive Great Egrets. Journal of Wildlife Diseases 36, 411–422.

Steele, K.W., Daniel, R.M., 1978. Fractionation of nitrogen isotopes by animals: a further complication to the use of variations in the natural abundance of $^{15}$N for tracer studies. Journal of Agricultural Science 90, 7–9.

Stenhouse, I.J., Montevecchi, W.A., 1999. Indirect effects of the availability of capelin and fishery discards: gull predation on breeding storm-petrels. Marine Ecology Progress Series 184, 303–307.

Stenhouse, I.J., Robertson, G.J., Montevecchi, W.A., 2000. Herring Gull *Larus argentatus* predation on Leach's Storm-petrel *Oceanodroma leucorhoa* breeding on Great Island, Newfoundland. Atlantic Seabirds 2, 35–44.

Streets, D.G., Zhang, Q., Wu, Y., 2009. Projection of global mercury emissions in 2050. Environmental Science & Technology 43, 2983–2988.

Suess, H.E., 1953. Natural radiocarbon and the rate of exchange of carbon dioxide between the atmosphere and the sea. In: National Research Council Committee on Nuclear Science (Ed.), Nuclear Processes in Geologic Settings. National Academy of Sciences, Washington, D.C, pp. 52–56.

Suess, H.E., 1955. Radiocarbon concentration in modern wood. Science 122, 415–417.

Sunderland, E.M., Chmura, G.L., 2000a. The history of mercury emissions from fuel combustion in Maritime Canada. Environmental Pollution 110, 297–306.

Sunderland, E.M., Chmura, G.L., 2000b. An inventory of historical mercury emissions in Maritime Canada: implications for present and future contamination. Science of the Total Environment 256, 39–57.

Sunderland, E.M., Cohen, M.D., Selin, N.E., Chmura, G.L., 2008. Reconciling models and measurements to assess trends in atmospheric mercury deposition. Environmental Pollution 156, 526–535.

Sunderland, E., Amirbahman, A., Burgess, N.M., Dalziel, J., Harding, G., Jones, S.H., Kamai, E., Karagas, M.R., Shi, X., Chen, C.Y., 2012. Mercury sources and fate in the Gulf of Maine. Environmental Research. http://dx.doi.org/10.1016/j.envres.2012.03.011.

Tagliabue, A., Bopp, L., 2008. Towards understanding global variability in ocean carbon-13. Global Biogeochemical Cycles 22, GB1025. http://dx.doi.org/10.1029/2007GB003037.

Thompson, D.R., 1996. Mercury in birds and terrestrial mammals. In: Beyer, W.N., Heinz, G.H., Redmon-Norwood, A.W. (Eds.), Environmental Contaminants in Wildlife: Interpreting Tissue Concentrations. CRC Press, New York.

Thompson, D.R., Furness, R.W., 1989. The chemical form of mercury stored in south Atlantic seabirds. Environmental Pollution 60, 305–317.

Thompson, D.R., Furness, R.W., Walsh, P.M., 1992. Historical changes in mercury concentration in the marine ecosystem of the north and north-east Atlantic Ocean as indicated by seabird feathers. Journal of Applied Ecology 29, 79–84.

U.S. EPA, 1998. EPA Method 7473: Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry, Revision 1998. U. S. Environmental Protection Agency, Washington, DC.

U.S. EPA, 2005. Regulatory Impacts Analysis of the Clean Air Act Mercury Rule. Office of Air Quality PLanning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, NC. Final Report. Vol. EPA-452/R-05-003.

Votier, S.C., Furness, R.W., Bearhop, S., Crane, J.E., Caldow, R.W.G., Catry, P., Ensor, K., Hamer, K.C., Hudson, A.V., Kalmbach, E., Klomp, N.I., Pfeiffer, S., Phillips, R.A., Prieto, I., Thompson, D.R., 2004. Changes in fisheries discard rates and seabird communities. Nature 427, 727–730.

Weiser, E.L., Powell, A.N., 2011. Reduction of garbage in the diet of nonbreeding Glaucous Gulls corresponding to a change in waste management. Arctic 64, 220–226.

Weseloh, D.V., Mineau, P., Struger, J., 1990. Geographical distribution of contaminants and productivity measures of Herring Gulls in the Great Lakes: Lake Erie and the connecting channels 1978/79. Science of the Total Environment 91, 141–159.

Weseloh, D.V.C., Moore, D.J., Hebert, C.E., de Solla, S.R., Braune, B.M., McGoldrick, D.J., 2011. Current concentrations and spatial and temporal trends in mercury in Great Lakes Herring Gull eggs, 1974–2009. Ecotoxicology 20, 1644–1658.