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Article

Stable Mercury Trends Support a Long-Term Diet Shift Away from Marine Foraging in Salish Sea Glaucous-Winged Gulls over the Last Century

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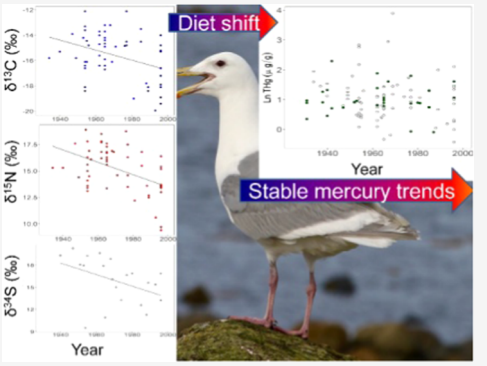
ABSTRACT: Marine predators are monitored as indicators of pollution, but such trends can be complicated by variation in diet. Glaucous-winged gulls (*Larus glaucescens*) have experienced a dietary shift over the past century, from mainly marine to including more terrestrial/freshwater inputs, with unknown impacts on mercury (Hg) trends. We examined 109-year trends in total mercury (THg) and methylmercury (MeHg) concentrations in glaucous-winged gull feathers (1887−1996) from the Salish Sea. Adult flank feathers had higher MeHg concentrations than immature feathers, and males head feathers had higher THg concentrations than females. Overall, we found no evidence of a trend in feather MeHg or THg concentrations over time from 1887 to 1996. In the same individuals, δ^{15}N, δ^{13}C, and δ^{34}S declined over time in gull feathers. In comparison, egg THg concentrations declined from 1970 to 2019 in two species of cormorants, likely reflecting decreases in local Hg sources. We conclude that diet shifts through time may have countered increased Hg deposition from long-range transport in glaucous-winged gulls. The lack of Hg trends over time in glaucous-winged gull feathers provides additional support that these gulls have decreased the amount of marine forage fish in their diet.



KEYWORDS: *mercury, seabirds, stable isotopes, glaucous-winged gulls, cormorants, North Pacific, food webs*

INTRODUCTION

Mercury (Hg) remains a persistent pollutant worldwide, as emissions have increased approximately 3- to 5-fold from the Industrial Revolution to the present.[1−3] Through microbial methylation by sulfate-reducing bacteria and related mechanisms in aquatic ecosystems, total Hg (THg) can be transformed to methylmercury (MeHg), its organic and most toxic form, capable of food web biomagnification and bioaccumulation at high concentrations in top predators. Total Hg concentrations in the North Pacific Intermediate Water mass have increased since the mid-1980s due to increased emissions from Asia.[1,4−6] It remains unclear whether THg concentrations have also increased in wildlife and ecosystems across the Pacific Ocean over time;[6] however, consequences to wildlife may be severe as seawater Hg concentrations are projected to increase by 50% by 2050 relative to the 2015 concentrations recorded in the North Pacific Ocean.[4,7]

Long-lived upper trophic predators such as seabirds are considered effective biomonitors of THg concentrations in marine ecosystems.[8−15] The Salish Sea of southern British Columbia and northern Washington is ranked among the most disturbed ecosystems in the world.[16] Glaucous-winged gulls (*Larus glaucescens*), which are generalist marine predators in the Salish Sea ecosystem, have experienced population fluctuations over the last century[17,18] along with long-term reductions in egg volume and clutch size.[19] Gull populations increased from 1900 to the mid-1980s and then declined to less than 50% of peak estimates by 2010—likely due to a cessation of hunting and egging in the early 1900s, and a gradual reduction in access to fish prey.[17−20] As glaucous-winged gulls prey on mesopelagic fish, which uptake the Hg produced via microbial activity,[21−23] they may be strong indicators of MeHg transfer in Pacific marine food webs. However, glaucous-winged gulls have also experienced a shift in diet over the past 150 years as indicated by stable isotope analyses of feathers from museum collections.[20] Feather stable nitrogen (δ^{15}N) and carbon (δ^{13}C) isotope values declined from 1860 to 2009, but remained constant in various forage fish species, suggesting gulls were either feeding at lower trophic levels or reduced their consumption of marine foods.[20] Stable sulfur (δ^{34}S) and hydrogen (δ^{2}H) isotope values have provided further evidence that glaucous-winged gulls shifted diets during the past to more terrestrial and freshwater-based

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diets, with an estimated increase of 30% in terrestrial/freshwater sources, e.g., earthworms (Lumbricidae), crop spoils, garbage.[24] As the main route of Hg transfer to seabirds is through prey,[9,25] it is unclear what effect this long-term shift in diet may have on THg concentrations in gulls.

The overall goal of our study was to examine Hg concentrations in glaucous-winged gull feathers to determine if gulls reflected long-term trends in Hg deposition of Pacific ecosystems. We examined Hg in head, flank, and primary feathers from museum collections, as feathers reflect the Hg body burden during growth.[25−29] We predicted that, because gulls have shifted to lower trophic and/or terrestrial/freshwater prey over time, THg trends in gull feathers would have increased at a slower rate than trends in North Pacific Ocean food webs. Mercury is also a strong indicator of marine foraging in seabirds;[30] therefore, stable Hg trends in gulls may provide further evidence of a shift to freshwater and/or terrestrial prey. Our objectives were to: (1) analyze long-term trends in THg and MeHg concentrations in gull feathers from 1887 to 1996; (2) model potential drivers of MeHg and THg exposure to gulls, such as year, biological factors (sex, age), and colony; and (3) compare THg trends in gulls to those in other seabirds [double-crested cormorants (*Nannopterum auritum*) and pelagic cormorants (*Urile pelagicus*)] in the Salish Sea ecosystem.

MATERIALS AND METHODS

Sample Collection. We sampled head (n = 171), flank (n = 65), and primary (n = 22) feathers for total mercury (THg) and methylmercury (MeHg) from glaucous-winged gull museum skins collected from 1887 to 1996 at 28 nesting colonies (48.42−49.68°N, 122.82−124.93°W) in the Salish Sea ecosystem in British Columbia, Canada, and Washington. Adult samples were taken both from feathers grown during the postbreeding molt (brown "winter" head feathers) and from those grown prior to territory establishment and breeding (white "summer" head feathers).[20,31] Winter feathers represent Hg exposure during breeding and prebreeding, whereas summer feathers represent exposure during the postbreeding period. In contrast, flank and primary feathers are molted only once per year and represent year-round exposure.[32] There were two adult samples not labeled for plumage stage; we designated feathers collected from November to February as equivalent to "winter" and from March to August as equivalent to "summer" stages (no feathers collected in September or October).[31,33,34] For primary feathers, segments of innermost primaries, ∼1 × 2 cm^2, were sampled (from P1; P2 if P1 was unavailable; occasionally P3 if neither P1 nor P2 were available); these are grown during the period immediately prior to or following egg-laying. For head feathers, we grouped first- and second-year subadults (n = 18) with juvenile (i.e., hatch-year; n = 14) birds as "immature" as they demonstrated no difference in MeHg concentrations (two-sample t-test, $t_{28.4}$ = 0.60, p = 0.55) and do not have a molt pattern. From 1970 to 2019, eggs from double-crested cormorants were collected for contaminant research at Mandarte Island (48.63°N, 123.28°W) and from pelagic cormorants at Mandarte and Mitlenatch Island (49.95°N, 125.00°W), British Columbia, Canada. Both feathers and eggs were stored in the National Wildlife Specimen Bank (NWSB) and analyzed at the National Wildlife Research Centre (NWRC, Ottawa, Ontario, Canada) following the methods presented in Miller et al.[35] and Elliott and Elliott[36] as detailed below.

Total Mercury (THg) and Methylmercury (MeHg) Analysis. We placed several (mean = 3) whole head feathers from individual gulls in a sieve and rinsed them with acetone, 0.25% Triton X-100, and then ultrapure water. Feathers were then air-dried for approximately 48 h. At NWRC, samples were analyzed for THg using a DMA-80 Direct Mercury Analyzer (Milestone, Monroe, CT) and reported in μg/g dry weight. Method detection and reporting limits were 0.028 ng and 0.141 ng, respectively, and were determined using SRM Oyster Tissue 1566b. Quality control and assurance included daily calibrations of at least four check standards of certified reference materials (BCR-463 tuna fish, NCR TORT-3 lobster hepatopancreas, IAEA-436 fish flesh, NIST 2976 mussel tissue, and IAEA-085 Human Hair). Recoveries for THg ranged from 92.1 to 109.0%. Based on 11 replicates of feathers, measurement precision for THg was estimated to be 8.46%.

We pooled and homogenized head feathers collected from 9-year subgroups between 1896 to 1948 for both THg and MeHg analysis. Several head feathers (mean = 18) were pooled from 2 to 14 individual birds collected during the same year. At NWRC, we analyzed MeHg using a Tekran Series 2700 Methyl Mercury Auto-Analysis System. First, MeHg was extracted from the samples in 5 mL of 17.5% nitric acid per 0.05 dry wt. for 16 hours at 60 °C and then stored at 4 °C until analysis. We treated extracts with sodium tetraethylborate to ethylate the Hg species, which were then moved onto a Tenax trap via argon where they were concentrated. The mercury species were desorbed from the trap by heating and transferred to a gas chromatographic column where they were separated. Each mercury species was then pyrolyzed and reduced to elementary Hg and detected via fluorescence. Quality control and assurance included daily calibrations of at least six check standards of certified reference materials (NCR TORT-3, NRC DORM-4 fish, IAEA-436, NIST 2976, and IAEA-085). Recoveries for MeHg ranged from 90.6 to 107.0%. Based on two replicates of pooled feathers, measurement precision for MeHg was estimated to be 1.49%.

At the Biotron Analytical Services lab at the University of Western Ontario, an additional 102 individual head, flank, and primary feathers were analyzed for THg and MeHg. Whole feathers were analyzed for THg using a DMA-80 following EPA methods 7473.[37] Recoveries averaged 96% and method detection and reporting limits were 0.0643 and 0.193 ng, respectively. Whole feathers were analyzed for MeHg using a Tekran 2700 following EPA methods 1630.[38] Samples were weighed and digested using 2 mL of 25% KOH in methanol solution and placed in an 80 °C oven for 4 hours. The samples were diluted to a volume of 10 mL and ethylated using 30 μL of NaBEt41%. The final pH of the samples was adjusted to 4.0−4.5 using 500 μL of buffer acetate and was analyzed by gas chromatography coupled to a pyrolyzer and an atomic detector. For both THg and MeHg analysis, for every 10 samples, quality control included a blank, a Matrix Spike/Spike Duplicate, a precision and recovery sample, and a quality control sample (IAEA-086 CRM). Recoveries for MeHg averaged 106% and method detection and reporting limits were 0.007 and 0.022 ng, respectively.

We homogenized individual and pooled (n = 2−10) whole cormorant egg samples as described in Miller et al.[39] Samples collected from 1970 to 2011 were analyzed for THg using an AMA-254 and from 2015 to 2019 a DMA-80 mercury analyzer following methods described in Elliott and Elliott.[36]

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Stable Isotope Analysis. We obtained $\delta^{13}C$ and $\delta^{15}N$ values for individual glaucous-winged gull head feathers analyzed for THg from Blight et al.,[20] and $\delta^{34}S$ in primary feathers from Hobson et al.[24] As primary feathers and winter (postbreeding) head feathers did not differ isotopically,[20] we used individual values for primary feathers in cases where stable isotope values for head feathers were not available. For $\delta^{13}C$ and $\delta^{15}N$ values of cormorant eggs collected from 1970 to 2011, analyses were carried out using a PDZ Europa ANCA-GSL elemental analyzer interfaced to a PDZ Europa 20−20 Isotope Ratio Mass Spectrometer (IRMS; Sercon Ltd., Cheshire, U.K.) at the Stable Isotope Facility at the University of California, Davis (http://stableisotopefacility.ucdavis.edu) and described elsewhere.[35,36,39,40] The internal standards were ($\delta^{15}N$, $\delta^{13}C$ in ‰): [nylon (−9.77, −27.81), bovine liver (7.72, −21.69), USGS-41 glutamic acid (47.6, −37.63), glutamic acid (−4.26, −28.85)]. Values of $\delta^{13}C$ and $\delta^{15}N$ were calculated relative to Vienna Pee Dee Belemnite (VPDB) and atmospheric nitrogen (AIR), respectively, and normalized to internal standards calibrated to international reference materials (IAEA-600, USGS-40, USGS-41, USGS-42, USGS-43, USGS-61, USGS-64, and USGS-65) with an analytical precision for both isotopes of ±0.2‰. From 2015 to 2019, stable isotope analysis was conducted at the Ján Veizer Stable Isotope Laboratory at the University of Ottawa. Samples for $\delta^{13}C$ and $\delta^{15}N$ were freeze-dried and weighed into tin capsules and then flash combusted at 1800 °C in a Vario El Cube elemental analyzer coupled to an IRMS (Delta Advantage, Thermo Finnigan, Germany) with a Conflo interface (Conflo III). The internal standards in ‰ for $\delta^{15}N$ and $\delta^{13}C$ were: [nicotinamide (0.07, −22.95), ammonium sulfate + sucrose (16.58, −11.94), caffeine (−16.61,−34.46), glutamic acid (−3.98, −28.53)]. Values of $\delta^{13}C$ and $\delta^{15}N$ were calculated relative to VPDB and AIR and normalized to internal standards calibrated to international standards [AIR: IAEA-N1(+0.4‰), IAEA-N2(+20.3‰), USGS-40(−4.52‰) and USGS-41(47.57‰); VPDB: IAEA-CH-6(−10.4‰), NBS-22(−29.91‰), USGS-40(−26.24‰) and USGS-41(37.76‰)] with an analytical precision for both isotopes of ±0.2‰. For $\delta^{34}S$ values of cormorant eggs collected from 1970 to 2019, all analyses were conducted at the Ján Veizer Stable Isotope Laboratory. The samples were weighed into tin capsules and then flash combusted at 1800 °C in an Isotope Cube (Elementar, Germany) elemental analyzer coupled to an IRMS (Delta Plus XP, Thermo Finnigan, Germany) with a Conflo interface (Conflo IIV). The internal standard used was S-6 ($\delta^{34}S$, −0.7‰). Values for $\delta^{34}S$ were calculated relative to Vienna-Cañon Diablo Troilite (VCDT) and normalized to the internal standard calibrated to international standards [silver sulfides: IAEA-S-1 ($\delta^{34}S$, −0.3‰), IAEA-S-2 ($\delta^{34}S$, 22.7‰)] with an analytical precision of ±0.3‰. For all stable isotope analyses, duplicate assays were run every sixth sample. To ensure that data between laboratories were comparable, nine samples were compared for interlaboratory variability and reported to have a mean difference of 0.44‰ for $\delta^{13}C$ and 0.52‰ for $\delta^{15}N$; the standard deviations between laboratories were 0.45‰ for $\delta^{13}C$ and 0.39‰ for $\delta^{15}N$. Stable isotope values are reported in delta (δ) notation in parts per thousand (‰) deviation for international standards. Values of $\delta^{13}C$ in whole egg homogenates were lipid normalized using the equation from Elliott and Elliott[36]

$$\delta^{13}C_{lipid-extracted} = \delta^{13}C_{non-extracted} - 5.21 + 3.94 \times Ln(C/N\ ratio) \quad (1)$$

Statistical Analyses. First, we compared differences in MeHg and THg concentrations among feather types using a paired t-test and a repeated-measures ANOVA. In the past, museums traditionally applied Hg-based preservatives to specimens.[41,42] Previous studies have excluded THg feather specimens that may have been contaminated or have relied upon MeHg concentrations, which are not affected by these preservatives.[43−45] We performed these tests on Ln(log)-transformed MeHg or THg data to meet normality. To determine the period during which museum feathers may have been contaminated with Hg-based preservatives,[41,42] we compared changes in the ratio of MeHg to THg in flank feathers by fitting broken-stick regressions to identify significant changes in slope, using the R package *SiZer*.[46] As feather Hg has been reported to be 67−100% MeHg,[26,47,48] we predicted that THg concentrations in uncontaminated feathers should be highly correlated with MeHg concentrations.[47] We also used a one-way ANOVA to compare the MeHg to THg ratio among 20-year intervals (1880−1899, 1900−1919, 1920−1939, 1940−1959, 1960−1979, 1980−1996), an approach used in other studies to identify feather mercurial contamination.[45] When appropriate, variables were Ln (log)-transformed to meet assumptions of normality.

Using MeHg and uncontaminated THg data, we tested if Hg concentrations in gulls changed over time by fitting linear models to examine factors affecting MeHg and THg concentrations in glaucous-winged gulls. We restricted our model selection to flank feathers for MeHg and head feathers for THg as they had the largest sample sizes, and removed observations with missing values. We built a global model with year (a continuous variable), sex, age (immature or adult), colony location, and their two-way interactions as predictors. We also included molt (immature, winter, or summer) as a predictor for head feathers only. We performed model selection using the dredge function in the *MuMIn* package in R[49] based on Akaike's Information Criterion adjusted for small sample size (AIC_c). The minimum adequate model within a $\Delta AIC_c < 2$ was considered the best model.[50] We calculated AIC_c weights for all available models. MeHg and THg concentrations were Ln(log)-transformed to ensure normality. Each of the top models met assumptions for normality, linearity, and homogeneity of variance. We compared trends in THg concentrations, $\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$ values over time among feathers of glaucous-winged gulls and eggs of double-crested and pelagic cormorants using linear regressions. All analyses were run using R. 3.6.3 (R Core Team, 2020), and significance was judged at $\alpha = 0.05$. All graphs were created using the *ggplot2*[51] package. Data are reported as mean ± S.E.M.

RESULTS AND DISCUSSION

Mercury Concentrations and Feather Type. There was no difference in MeHg concentrations between flank (3.11 ± 0.5 μg/g, $n = 15$) and head (3.96 ± 0.6 μg/g, $n = 15$) feathers (paired t-test, $t_{14} = -1.73$, $p = 0.11$), with MeHg in flank and head feathers positively correlated ($R^2 = 0.59$). There was a significant difference in THg concentrations among feather type (Figure 1; repeated-measures ANOVA, $F_{2,20} = 5.51$, $p = 0.012$), with higher and more variable concentrations in head

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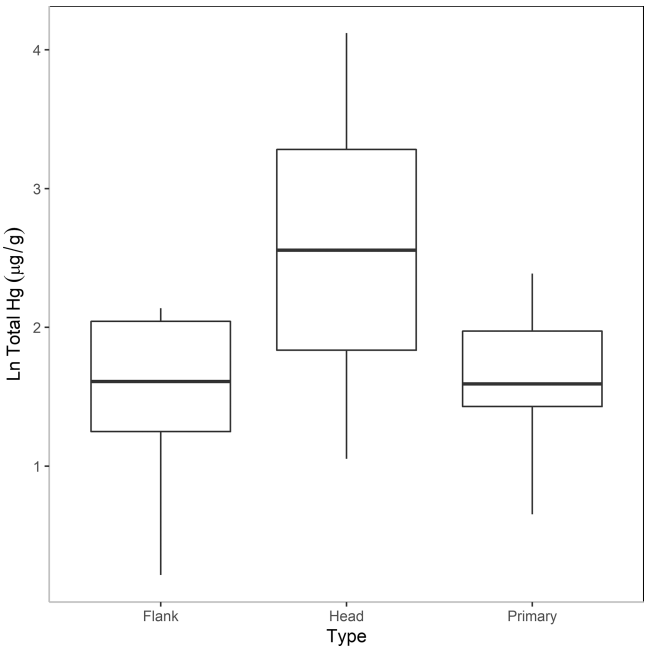

Figure 1. Boxplot of total Hg concentrations in paired flank, head, and primary feathers from glaucous-winged gulls (n = 11) collected from 1887 to 1996.

(mean THg: 18.94 ± 5.3 μg/g) than flank (5.24 ± 0.8 μg/g) feathers (bonferroni p_{adj} = 0.045), but not primary feathers (5.74 ± 0.9 μg/g) from the same individuals. Flank feathers are less variable in THg concentrations than primaries and are more representative of the total Hg of a bird's body burden.[25] As there was a difference in THg concentrations between flank and head feathers, we analyzed these feather types separately.

Mercury-Based Preservatives and THg Trends. There was a weak relationship between THg and lnMeHg in flank feathers (R^2 = 0.07, $F_{1,63}$ = 5.06, p = 0.028), but no relationship was found in head feathers (R^2 = 0.15, $F_{1,13}$ = 2.25, p = 0.16) from 1887 to 1996 or in pooled head feathers from 1896 to 1948 (R^2 = 0.42, $F_{1,7}$ = 5.03, p = 0.06), indicating the likelihood of some samples being contaminated with Hg-based preservatives. When we conducted a piecewise regression of the MeHg to THg ratio in flank feathers over time, a significant inflection point was identified in 1928, where the ratio increased significantly. The (ln-transformed) ratio of MeHg to THg post-1928 was significantly higher in flank feathers (mean ratio: 0.98 ± 0.06; Welch two-sample t-test, $t_{48.7}$ = 6.56, p < 0.0001) and head feathers (1.33 ± 0.36; $t_{12.9}$ = 4.03, p = 0.001) in comparison to pre-1928 ratios (flank: 0.47 ± 0.06; head: 0.29 ± 0.11). In addition, the relationship between lnMeHg and lnTHg was stronger in flank feathers after 1928 (R^2 = 0.67, $F_{1,28}$ = 55.55, p < 0.0001). We found differences in the MeHg-to-THg ratio in feathers among 20-year sampling periods (Type-III ANOVA, $F_{5,59}$ = 5.86, p = 0.0002), with ratios from 1900 to 1939 lower than from 1960 to 1996 (Table S1). Based on these results, THg concentrations prior to 1928 were removed from further analysis due to possible contamination with mercury-based preservatives.[41,42]

Drivers of MeHg Concentrations. Four models had the most support for MeHg concentrations in flank feathers, which included sex, year, and age as predictors (Table 1). Age alone was the most parsimonious model, and adults had higher MeHg concentrations than immature birds (Table 2; Figure 2). While there was support for the model year + age, year was not a significant predictor (Figure 3; t = 1.74, p = 0.088). Three models had support for THg concentrations in head feathers which included sex and molt as predictors (Table 1). Sex alone was the most parsimonious model, and males (t = 2.02, p = 0.048) had higher THg concentrations than females, a pattern observed in other birds and thought to be related to depuration via egg production.[52] In North Pacific glaucous-winged gulls, THg concentrations were higher in adult vs fledgling breast feathers, and males were found to have higher levels of feather chromium and manganese, but not THg, than females.[53]

Table 2. Comparison of Methylmercury (MeHg) in Flank Feathers and Total Mercury (THg) Concentrations in the Head Feather of Glaucous-Winged Gulls Collected from 1887 to 1996 at Colonies Near the Salish Sea in Vancouver, Canada, and Washington[a]

dependent	predictors	β	S.E.M.	t	p
LnMeHg (n = 58)	intercept	0.96	0.132	7.29	<0.0001
	age	−0.63	0.183	−3.438	0.001
LnTHg (n = 62)	intercept	0.82	0.096	8.555	<0.0001
	sex	0.29	0.146	2.024	0.0475

[a]Parameter estimates (β ± standard error) from the top linear models based on Akaike's information criterion corrected (AICc) explaining lnMeHg and lnTHg concentrations. The t and p-values are included.

Table 1. Comparison of Global Models Predicting Methylmercury (MeHg) in Flank Feathers and Total Mercury (THg) Concentrations in the Head Feather of Glaucous-Winged Gulls Collected from 1887 to 1996 at Colonies Near the Salish Sea in Vancouver, Canada, and Washington[a]

predictor	global model	best models	K	AIC_c	$\Delta_i AIC_c$	w_i	L
Ln(MeHg)	Year + Sex + Age + Colony + Sex × Age + Age × Year + Sex × Year + Colony × Sex + Colony × Age + Colony × Year	Age + Year	4	126.3	0	0.353	−58.784
		Age	**3**	**127.1**	**0.78**	**0.239**	**−60.33**
		Age + Sex + Year	5	127.2	0.88	0.227	−58.026
		Age + Sex	4	127.7	1.34	0.181	−59.453
Ln(THg)	Year + Sex + Age + Colony + Molt + Sex × Age + Age × Year + Sex × Year + Colony × Sex + Colony × Age + Colony × Year + Molt × Year + Molt × Age + Molt × Sex + Molt × Colony	Molt + Sex	5	108.2	0	0.57	−48.585
		Sex	**3**	**110.2**	**1.93**	**0.218**	**−51.877**
		Molt + Sex + Molt × Sex	7	110.2	1.97	0.213	−47.069

[a]The top linear mixed-effect models explaining lnMeHg and lnTHg after model selection with an Akaike's Information Criterion adjusted for small sample size less than 2 ($\Delta_i AIC_c$). ΔiAICc is the difference between AICc for the current model and the minimum of AICc among all of the models. l = log likelihood, K = number of parameters, ωi = Akaike's weights based on all models. Best models are bolded. If more than one model had ΔAICc < 2, the most parsimonious model was considered best.

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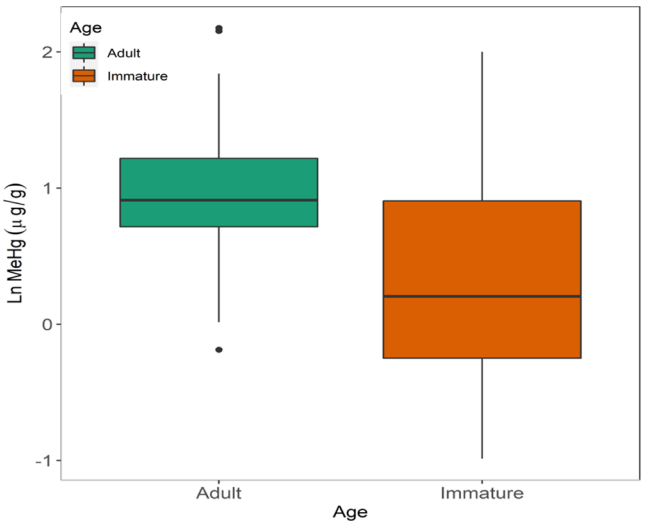

Figure 2. Boxplot of total MeHg concentrations in immature (n = 27) and adult (n = 30) flank feathers from glaucous-winged gulls collected between 1887 to 1996.

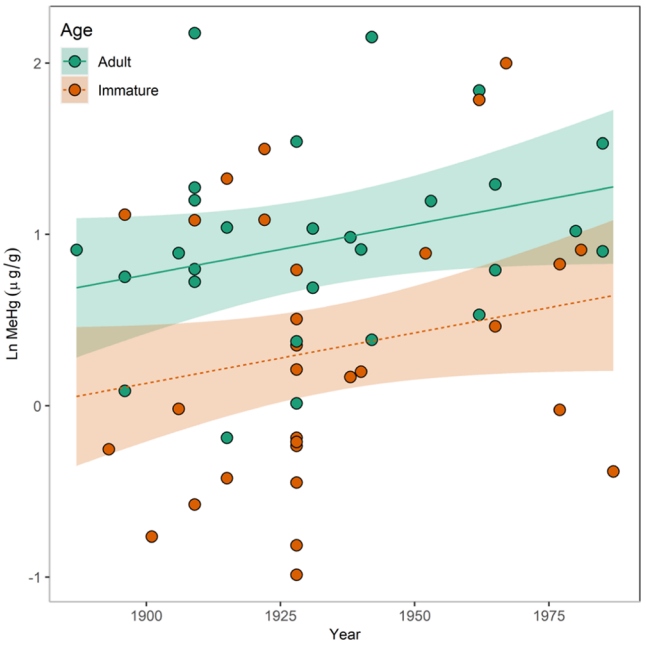

Figure 3. Linear regression of the relationship between MeHg concentrations over time in immature and adult flank feathers of glaucous-winged gulls.

When we examined trends in MeHg across the entire dataset, there was no relationship between MeHg over time in flank feathers from immature (Figure 3; $t_{1,28}$ = 1.33, p = 0.20) or adult ($t_{1,26}$ = 1.14, p = 0.27) birds from 1887 to 1996. In our pooled head feathers, we also found no relationship between MeHg over time from 1896 to 1948 (R^2 = 0.15, $F_{1,7}$ = 1.23, p = 0.30). Overall, there was no trend in THg concentrations in head feathers from 1928 to 1996 (Figure 4; $F_{1,77}$ = 1.79, p = 0.19) or flank feathers ($F_{1,28}$ = 0.0002, p = 0.99) from 1931 to 1996. In another Pacific seabird, black-footed albatross (*Phoebastria nigripes*), feather MeHg concentrations increased post-1940 relative to pre-1940, coinciding with increases in regional trends in THg emissions over time.[43] In gulls, post-1940 feathers were also higher than pre-1940 samples (two-sample t-test, $t_{58.3}$ = 3.16, p = 0.0025). However, while Salish Sea glaucous-winged gulls have undergone a long-term shift in diet likely associated with declines in availability of key forage fish populations such as herring (*Clupea pallasii*[20,24,56]), black-footed albatross exhibited no change in δ^{15}N values over time, and therefore, the increase in MeHg was attributed to increased Hg exposure of prey in the pelagic food web.[43] Feather THg concentrations also increased threefold in gray-headed albatross (*Thalassarche chrysotoma*) from South Georgia in the Southern Ocean from 1989 to 2014, a trend believed to be the result of a diet shift toward more contaminated prey, and an increase in MeHg exposure in foraging areas.[57] As seabirds that consume mesopelagic prey have a higher magnitude of increase in THg levels (3.5−4.8% per year) relative to those feeding on epipelagic food chains (1.1−1.9% per year[58]), any shift away from mesopelagic fish by gulls would have countered increases in THg in the marine food web. The magnitude of increase in MeHg in black-footed albatross feathers was also low relative to the increase of THg concentrations observed in the Pacific Ocean.[43] Therefore, stable feather THg concentrations in glaucous-winged gulls may be the result of a diet shift and a dampening effect of MeHg transfer in Pacific food webs.[36,43]

Trends in Total Mercury Concentrations and Stable Isotope Values in Gulls and Other Local Seabirds. While THg concentrations did not change in glaucous-winged gulls from the Salish Sea region from 1887 to 1996, THg concentrations declined from 1970 to 2019 in the eggs of both double-crested and pelagic cormorants from the same area. Mercury concentrations declined with year in double-crested (Figure 4A; R^2 = 0.19, $F_{1,115}$ = 27.25, p < 0.0001) and pelagic cormorants (R^2 = 0.43, $F_{1,124}$ = 92.66, p < 0.0001). While THg concentrations in water concentrations of the North Pacific Ocean have increased slightly with increasing THg emissions from Asia since the mid-1980s,[2,7] local efforts to mitigate pollution have reduced THg discharge into the Salish Sea ecosystem since the 1970s.[59] As such, declines in THg in cormorant eggs may reflect the local reductions in Hg emissions in the Salish Sea. Since the gull feathers were collected from a broader geographic region, THg concentrations likely also reflect a greater influence of long-range sources from Asia.

All stable isotope values (Figure 4; δ^{13}C: R^2 = 0.13, $F_{1,53}$ = 8.03, p = 0.007; δ^{15}N: R^2 = 0.22, $F_{1,53}$ = 14.61, p = 0.0003; δ^{34}S: R^2 = 0.18, $F_{1,19}$ = 4.10, p = 0.057) declined over time in glaucous-winged gulls from 1934 to 1996. In contrast, δ^{15}N values increased over time in both cormorant species (Figure 4C, double-crested R^2 = 0.11, $F_{1,76}$ = 9.04, p = 0.004; pelagic R^2 = 0.53, $F_{1,85}$ = 97.41, p < 0.0001), and δ^{13}C values increased in pelagic cormorants with year (Figure 4B, R^2 = 0.20, $F_{1,85}$ = 21.02, p < 0.0001), but not in double-crested cormorants (R^2 = 0.03, $F_{1,76}$ = 2.31, p = 0.13). Declines in δ^{15}N in gulls suggest a shift to lower trophic prey with lower THg concentrations. However, the increase in δ^{15}N and decline in THg in pelagic and double-crested cormorants suggest a reduction in THg across all trophic levels, supporting the theory that cormorants reflect reductions in local Hg sources. As δ^{13}C values are higher in marine relative to freshwater and terrestrial systems,[60] a decline in δ^{13}C over time with no change in THg in gull feathers suggests a diet shift away from marine sources. In other Pacific species, δ^{13}C values were the main driver of feather THg concentrations across seven procellariform seabird species in the Chatham Islands in the Southern Pacific

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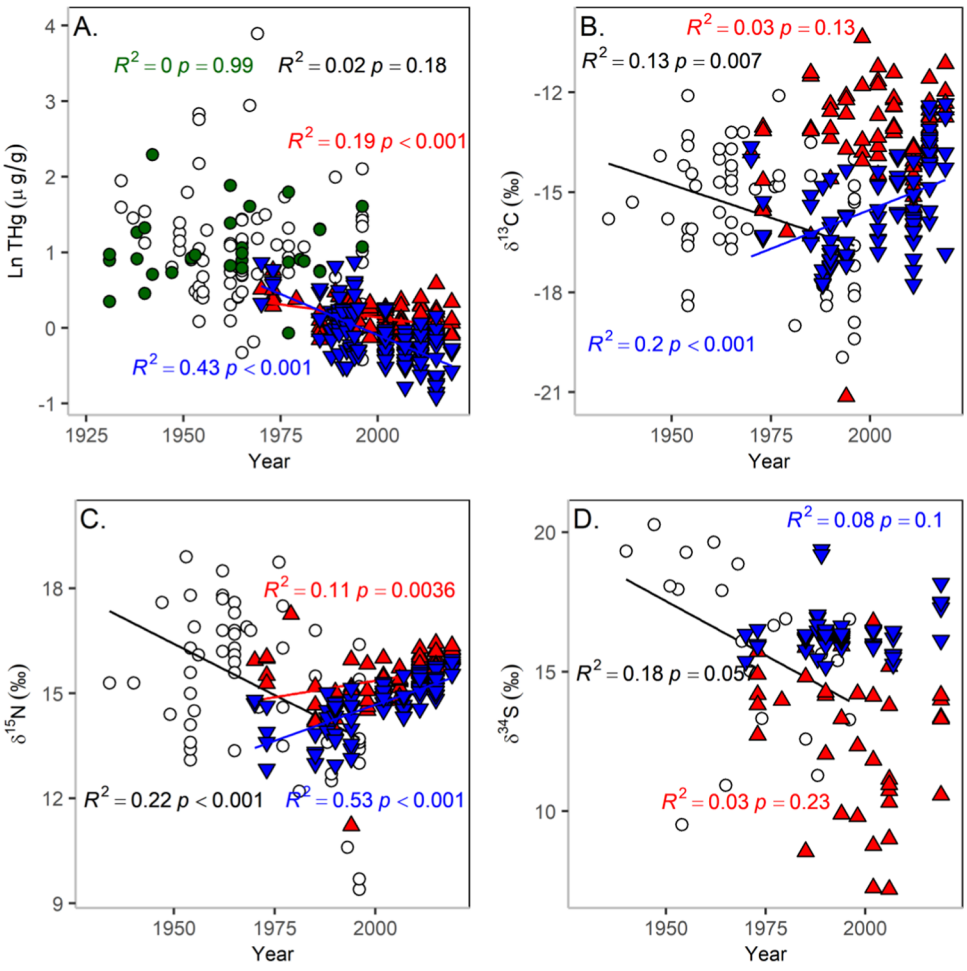

Figure 4. Linear regression of the relationship between (A) lnTHg, (B) δ^{13}C, (C) δ^{15}N, and (D) δ^{34}S in head feathers (open circles, black text) and flank feathers (green circles, green text) of glaucous-winged gulls and eggs from double-crested (red triangles and text) and pelagic cormorants (blue inverted triangles and text) from Pacific Canada over time.

Ocean,[61] and 20 species of albatross across the Southern Ocean.[62] In these studies, variation in δ^{13}C values reflected differences in foraging habitat, with species that foraged in warmer subtropical waters having higher THg concentrations than those foraging in colder waters.[57,61,62] In telemetry studies of other gull species, blood THg concentrations were higher in western gulls (*L. occidentalis*) from the Northeast Pacific and herring gulls (*L. argentatus*) in the Northeastern U.S. that foraged in ocean habitats relative to gulls feeding inland in terrestrial, freshwater habitats from urban colonies,[30,55] suggesting that Hg is a strong indicator of marine foraging.[30]

Considering there has been a long-term decline in δ^{13}C, δ^{15}N, and δ^{34}S values in gulls but not in cormorants, our results suggest that the gradual shift from marine to greater terrestrial foraging may explain stable THg concentrations in glaucous-winged gulls over time. As a result of this shift in diet, THg concentrations in glaucous-winged gulls may not accurately reflect regional trends of Hg exposure in the North Pacific. The shift in diet over time and its effect on reproductive output, as well as increases in bald eagle (*Haiaeetus leucocephalus*) predation, are believed to have resulted in population declines in gulls;[18] however, pollutants have also been suggested as an alternative contributing factor.[19] While THg concentrations have remained relatively stable, approximately 13% of glaucous-winged gulls had head feather THg concentrations that exceeded the minimum toxicological benchmark for reproductive effects documented in other birds (5 μg/g dw[63,64]). There was also a high variation in feather THg concentrations among individual gulls, with THg concentrations ranging from 0.66 to 8.23 μg/g dw (mean: 3.11 ± 2.19 μg/g) in 11 gulls from the same colony (Mandarte Island) sampled in 1996. As high variation in diet and stable isotope values among individual gulls was observed in colonies closer to urban areas,[54,65] future research assessing factors affecting individual variation in THg concentrations may provide insights into the sources and dynamics of THg in gulls and other marine birds. Overall, the lack of MeHg and THg trends over time in two different feather types of glaucous-winged gulls provides additional support that these gulls have decreased the amount of forage fish in their diet.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c03760.

Tukey HSD post hoc comparison of the ratio of MeHg to THg in flank feathers among 2-year collection periods from 1880 to 1996 (Table S1) (PDF)

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Notes

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