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Rapidly increasing methyl mercury in endangered ivory gull (*Pagophila eburnea*) feathers over a 130 year record

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Mercury (Hg) is increasing in marine food webs, especially at high latitudes. The bioaccumulation and biomagnification of methyl mercury (MeHg) has serious effects on wildlife, and is most evident in apex predators. The MeHg body burden in birds is the balance of ingestion and excretion, and MeHg in feathers is an effective indicator of overall MeHg burden. Ivory gulls (*Pagophila eburnea*), which consume ice-associated prey and scavenge marine mammal carcasses, have the highest egg Hg concentrations of any Arctic bird, and the species has declined by more than 80% since the 1980s in Canada. We used feathers from museum specimens from the Canadian Arctic and western Greenland to assess whether exposure to MeHg by ivory gulls increased from 1877 to 2007. Based on constant feather stable-isotope ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) values, there was no significant change in ivory gulls' diet over this period, but feather MeHg concentrations increased 45× (from 0.09 to 4.11 $\mu\text{g g}^{-1}$ in adults). This dramatic change in the absence of a dietary shift is clear evidence of the impact of anthropogenic Hg on this high-latitude threatened species. Bioavailable Hg is expected to increase in the Arctic, raising concern for continued population declines in high-latitude species that are far from sources of environmental contaminants.

1. Introduction

Anthropogenic mercury (Hg) emissions are the predominant input to the global atmospheric Hg pool [1], and is expected to continue to increase owing to both increasing emissions and global change feedbacks [2], particularly at high latitudes [3]. While most deposition of atmospheric Hg to the Earth's surface is as inorganic Hg (IHg), under specific environmental conditions, a small fraction is converted into the more toxic bioaccumulating methyl mercury (MeHg) [4]. The bioaccumulation and biomagnification of MeHg has serious effects on wildlife [5], and in addition to the documented effects of MeHg ingestion on humans, these effects are most serious and evident in apex predators such as marine mammals and birds [6].

An organisms' MeHg body burden is determined by the balance of ingestion through diet, and excretion in urine, faeces or through keratin production in hair, nails, claws and feathers, which strongly binds MeHg to disulfides. The elimination of MeHg via moulted feathers is a well-known process [7]. The direct relationship between the concentration of MeHg in feathers and the body burden of MeHg during the time over which the feather was grown makes feathers an effective and commonly used indicator of overall MeHg body burden [8], although feather type (e.g. flight versus body) and species' moult patterns affect the coupling between feather and body MeHg [7–9]. Once grown, feathers are inert, and the Hg bound within the feather is stable [10]. It is therefore possible to examine temporal trends in feather MeHg (and therefore MeHg body burden) using dated museum specimens, allowing retrospective analyses of more than 100 years [11–14]. Most Hg in feathers is MeHg [15], so total Hg (IHg + MeHg) is often measured as a proxy; however prior to the 1940s,

museum specimen preparation techniques often used IHg (usually as HgCl_2) as a preservative, introducing a source of Hg contamination [16,17]. This preservation does not interfere with the selective measurement of MeHg in feathers; as a consequence, MeHg is the only appropriate measure of a birds' Hg body burden when analysing museum specimens with unknown preservation histories [12].

Stable-isotope values in feathers are unaltered by museum preparation, and can be used to study long-term changes in foraging ecology [18]. Isotope values of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) in feathers are indicative of the foraging location ($\delta^{13}\text{C}$) and trophic position ($\delta^{15}\text{N}$) of the bird at the time the feathers were grown [19]. While contaminant and stable-isotope values in feathers are not comparable directly [20], both can be examined to provide valuable information not possible by each assay alone.

Ivory gulls (*Pagophila eburnea*) are listed as Near Threatened by the International Union for Conservation of Nature, and are endangered in Canada, where they have declined by 80–85% since the 1980s, and now number 400–500 breeding pairs [21–23]; data on population trends before 1980 are not available. The reason for this decline is not well understood, but could be related to contaminants [24], illegal harvesting in Greenland [25] and/or changing sea-ice conditions [21,22,26]. Ivory gull eggs have the highest total Hg recorded for any Arctic bird [24], probably because of their high trophic position [27] related to their habit of scavenging on marine mammal carcasses (figure 1) [28]. Canadian ivory gulls also have the highest egg Hg from throughout the species range [29]. Understanding historic trends in contaminant levels will help elucidate whether ivory gulls have been consistently exposed to naturally high concentrations of MeHg, or whether the concentrations reported in recent years [24] are a result of contemporary (and probably anthropogenic) processes. If MeHg has been consistently high, it is unlikely that it has been a cause of the population decline. If, however, MeHg has increased over the past 130 years, this could indicate a relationship between increasing Hg exposure and current declining population trends. Given this important implication, our objectives were to determine whether MeHg concentrations in ivory gull breast feathers had increased over the period from 1877 to 2007 using museum specimens, and to use $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ to establish if there have been any trophic shifts over the same period which might explain any observed changes in MeHg concentrations.

2. Methods

(a) Sample collection

We sampled eight breast feathers from each of 80 ivory gulls held in collections at many North American museums (see Acknowledgements). Feathers from each bird were stored dry in individual paper envelopes until analysis. We sampled birds that were known to have been collected in the Canadian Arctic, western Greenland or those from eastern Canadian waters (assumed to be from the same population; [30]). We did not sample pre-fledged chicks or birds without a date of collection. Birds were categorized into two age classes (adult, first-winter) based on plumage [31,32].

(b) Laboratory analyses

(i) Stable isotopes

We measured $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values at the National Hydrology Research Centre (Saskatoon, Saskatchewan). Feathers were



Figure 1. Adult ivory gull feeding on a seal carcass, Resolute Bay, Nunavut, Canada, 10 June 1989. Photograph by K. A. Hobson. (Online version in colour.)

washed in a 2 : 1 chloroform : methanol solution to remove external contamination, and 1.0 mg was placed in a tin capsule, and analysed by continuous-flow isotope-ratio mass spectrometry. Results were calibrated using secondary isotopic reference materials, bowhead whale baleen ($\delta^{13}\text{C}$: -18.5‰ , $\delta^{15}\text{N}$: $+14.4\text{‰}$, $n = 7$) and porcine gelatin ($\delta^{13}\text{C}$: -13.5‰ , $\delta^{15}\text{N}$: $+4.7\text{‰}$, $n = 7$); both of which had within-run s.d. $< 0.2\text{‰}$. Stable-isotope values are presented relative to Vienna Peedee Belemnite ($\delta^{13}\text{C}$) or atmospheric nitrogen (air, $\delta^{15}\text{N}$) following Coplen [33] and Bond & Hobson [34].

We adjusted $\delta^{13}\text{C}$ values for the Suess effect [35], whereby carbon from fossil fuel combustion has resulted in a global decrease in $\delta^{13}\text{C}$, using the equation

$$\delta^{13}\text{C}_{\text{adjusted}} = \begin{cases} \delta^{13}\text{C}_{\text{raw}} - b_{\text{his}} \times (t_i - t_1), & \text{if } t_i \leq 1950 \\ \delta^{13}\text{C}_{\text{raw}} - b_{\text{his}} \times (1950 - t_1) - b_{\text{mod}} \times (t_i - 1950), & \text{if } t_i > 1950 \end{cases}$$

We used values of -0.007‰ for b_{his} , the historical global annual decline in $\delta^{13}\text{C}$ [36], as no region-specific measurements are available, and -0.026‰ for b_{mod} , the change in oceanic $\delta^{13}\text{C}$ in the North Atlantic measured from 1950 to 1993 [37], following the same rationale as Farmer & Leonard [38].

(ii) Total mercury

Total mercury was analysed in a subset of samples from prior to 1900 ($n = 8$) and after 1975 ($n = 10$) to determine whether older specimens were contaminated owing to specimen preservation with Hg, thus precluding the use of total Hg in the time-series analysis. These were the only specimen samples where sufficient feather mass for total Hg analyses remained after MeHg analyses were completed. To clean surface contamination, feather samples were placed in a centrifuge tube, covered with 40 ml of a solution of 1% Alconox Liquinox® critical cleaning liquid detergent dissolved in 18.2 MOhm deionized water, capped and shaken for 1 min approximately every 10 min for 1 h. The liquid was then completely decanted from the sample, and the sample rinsed five times with deionized water. Samples were then placed in clean, dry tubes in a fumehood and left to dry for 3 days to dry at room temperature. Total mercury was determined using A Milestone® DMA-80 direct mercury analyser [39] in accordance with Environmental Protection Agency (EPA) method 7473 [40] in an ISO 17025 accredited laboratory in the Biotron Centre for Experimental Climate Change Research at Western University, London, Ontario. A single instrument calibration with a four-point standard curve had an $r^2 = 0.9998$. A human hair certified reference material (CRM; IAEA-86, certified value $0.573 \mu\text{g g}^{-1}$ total Hg) was analysed once as part of this limited analysis of a subset of feathers for quality assurance, and had a recovery of 98% ($0.564 \mu\text{g g}^{-1}$ total Hg) of its certified value indicating satisfactory performance of the instrument.

(iii) Methyl mercury

Methyl mercury was determined using a modified version of the EPA method 1630 [41] in an ISO 17025 accredited laboratory in the Biotron Centre for Experimental Climate Change Research at Western University, London, Ontario. Feather samples were cleaned according to the methods outlined above. Each feather was digested intact. Samples were weighed, placed into a pre-acid cleaned 60 ml Teflon digestion vessel (Savillex®) with 8 ml of 5 M HNO₃ solution, capped, vortexed for 20 s (3000 rpm) and left overnight at room temperature. The next day, the digestion vessels were re-vortexed, placed into an 80°C oven for 8 h, and allowed to cool for 1 h. Next, 10 ml of deionized water was added to each vessel, re-capped, vortexed and left at room temperature to be run next day. For analysis, a sample aliquot was diluted with deionized water to fill the autosampler vials for the Tekran 2700 automated MeHg analyser, vortexed and the pH of each sample was adjusted with 2 M sodium acetate buffer solution to between 4.0 to 4.5 (required for proper ethylation). Finally, 1% sodium tetraethylborate was added, the vial capped, vortexed a final time and left to sit 30 min prior to loading in the autosampler.

The instrument calibration curve consisted of an eight-point standard curve with an average $r^2 = 0.9993$ (s.d. = 0.0011, RSD% = 0.0011) throughout the three separate runs. Secondary source calibration checks were run each 10 vials, using low (0.03 ng l⁻¹; average recovery 101.5%), mid-range (0.3 ng l⁻¹; average recovery 100.1%) and high (1.5 ng l⁻¹; average recovery 98.1%) checks on a rotating basis. A human hair CRM (IAEA-86, certified value 0.258 µg g⁻¹ MeHg) was analysed in triplicate every 10 samples, with an average recovery of 82% of its certified value (s.d. = 4.89, RSD% = 0.0589). As feather sample volume available was insufficient for a sample triplicate, the CRM was analysed in triplicate, and a subset of test feathers was run in duplicate every 10 samples. The sample precision for the duplicate test feather, provided an average repeatability of 95% (s.d. = 13.36, RSD% = 0.1396). The test feathers were also used for matrix spike samples (every 10 samples), with an average recovery of 110% (s.d. = 15.187, RSD% = 0.1372). The method blank (every 10 samples) had an average concentration of 0.9296 ng l⁻¹ (s.d. = 0.9701, RSD% = 1.0437). Final concentrations are reported as dry weight MeHg concentrations in feathers through back calculation from aqueous concentrations of digestate, accounting for dilutions.

(c) Statistical analysis

We first tested for normality in contaminant and isotopic data using Shapiro–Wilk’s test [42], and identified outliers as those with Cook’s distance more than 3 [43–45]. We used general linearized models (GLMs) to examine the effects of year of collection, age class and for a subset of birds, sex and we restricted our model to primary effects and two-way interactions. Analyses were conducted in R v. 3.1 [46], and terms were considered significant when $p < 0.05$.

We also performed a time-series analysis using the program PIA [47] to compare trends with the large body of literature on Hg trends in the Arctic [48,49]. This analysis uses a robust-regression and log-linear regression techniques to detect trends (both linear and nonlinear) using a running-mean smoother based on geometric means [50]. We performed separate analyses for each age class (see GLM results below), set the statistical power to detect a trend at 80%, and the minimum slope to detect at 10% over 10 years at $p < 0.05$ using a 3 year running-mean smoother. We were also able to compare these results with an analysis of Atlantic puffin (*Fratercula arctica*) and manx shearwater (*Puffinus puffinus*) using data from Thompson *et al.* [11]

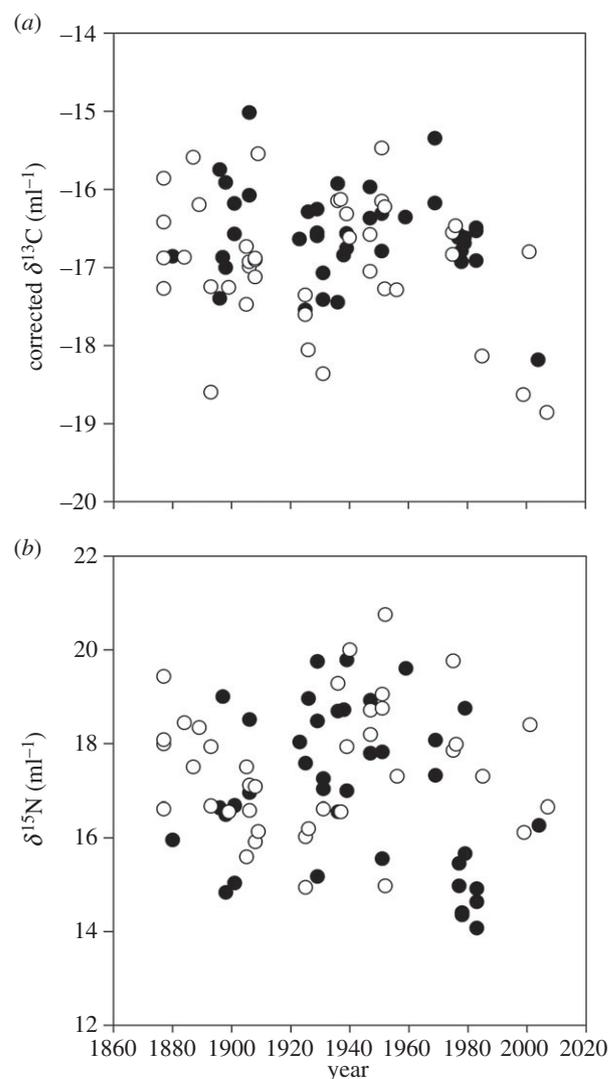


Figure 2. There was no significant change in (a) $\delta^{13}\text{C}$ or (b) $\delta^{15}\text{N}$ in ivory gull feathers from Arctic Canada and western Greenland over time, or between age classes ($n = 80$). White dots are first-winter birds; black dots are adult birds. One $\delta^{15}\text{N}$ outlier removed (see text for details).

3. Results

After removing one outlier, ivory gull feather $\delta^{15}\text{N}$ was normally distributed (Shapiro–Wilk’s $W = 0.98$, $p = 0.47$), and there were no significant differences in $\delta^{15}\text{N}$ among age classes ($F_{1,75} = 1.72$, $p = 0.19$), or over time ($F_{1,75} = 0.72$, $p = 0.40$; figure 2), and the age class \times year interaction was not significant ($F_{1,75} = 3.18$, $p = 0.08$). Using only birds of known sex ($n = 63$; 79% of our sample), there was no effect of sex ($F_{1,56} < 0.01$, $p = 0.96$), age class ($F_{1,56} = 2.02$, $p = 0.16$), year of collection ($F_{1,56} < 0.01$, $p = 0.93$) or any interaction (all $p > 0.09$). $\delta^{13}\text{C}$ in ivory gull feathers was also normally distributed (Shapiro–Wilk’s $W = 0.97$, $p = 0.07$), and while there was no change in $\delta^{13}\text{C}$ over time after correcting for the Suess effect ($F_{1,75} = 0.73$, $p = 0.40$; figure 2), adults had a slightly higher $\delta^{13}\text{C}$ than first-winter birds (-16.6‰ versus -16.9‰ ; $F_{1,75} = 5.00$, $p = 0.03$), but this difference is not biologically relevant; the age class \times year interaction was not significant ($F_{1,75} < 0.01$, $p = 0.98$). We found the same pattern using birds of known sex—no effect of year ($F_{1,56} = 0.012$, $p = 0.73$), or sex ($F_{1,56} < 0.01$, $p = 0.96$), but adults with higher $\delta^{13}\text{C}$ ($F_{1,56} = 4.92$, $p = 0.03$), and no significant interaction terms (all $p > 0.48$).

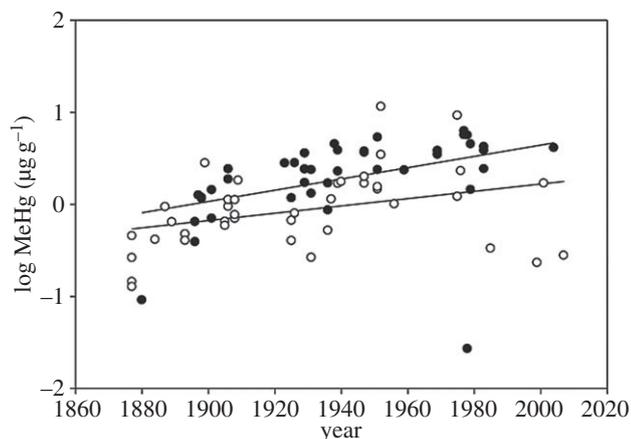


Figure 3. Feather MeHg increased in both adult ($n = 40$) and first-winter ivory gulls ($n = 40$) from Arctic Canada and western Greenland between 1877 and 2007. White dots are first-winter birds; black dots are adult birds.

Mean total Hg concentrations in feathers from the museum specimens from pre-1900 ($47.1 \pm 29.1 \mu\text{g g}^{-1}$) were dramatically higher in total Hg (THg) than those from post-1975 ($3.84 \pm 2.02 \mu\text{g g}^{-1}$). Using the mean MeHg concentrations from the same museum specimen (but not the identical feathers), the %THg as MeHg was 1.7% and 61.8%, respectively.

MeHg concentrations in ivory gull feathers were not normally distributed (Shapiro–Wilk's $W = 0.80$, $p < 0.001$), so data were log-transformed to achieve normality ($W = 0.97$, $p = 0.05$). Adult ivory gulls had higher MeHg than first-winter birds ($F_{1,76} = 9.08$, $p = 0.004$), and MeHg increased over time ($F_{1,76} = 17.53$, $p < 0.001$; adults $\beta \pm \text{s.e.} = 0.014 \pm 0.005$, first winter $\beta \pm \text{s.e.} = 0.009 \pm 0.004$ on a log scale); the age class \times year interaction was not significant ($F_{1,76} = 0.64$, $p = 0.43$; figure 3). Using birds of known sex, we found significant effects of age class (adults had higher MeHg; $F_{1,56} = 7.18$, $p = 0.010$), and MeHg increased over time ($F_{1,56} = 14.75$, $p < 0.001$). There was no difference between sexes ($F_{1,56} = 0.19$, $p = 0.67$), and all interactions were not statistically significant (all $p > 0.18$).

(a) Time-series analysis

Adult ivory gulls had an annual +1.6% log-linear increase in MeHg (95% CI: 0.6–2.7%), which was statistically significant ($r^2 = 0.33$, $p = 0.004$), whereas in first-winter birds, MeHg did not increase significantly over time (annual change: +0.3%, 95% CI: -0.7 to 1.2%, $r^2 = 0.01$, $p = 0.58$). Both datasets had high power to detect a log-linear trend over the entire period, comparable to previous assessments of seabird MeHg over decadal or centennial scales (table 1).

4. Discussion

Feathers from museum specimens collected prior to 1900 were considerably contaminated with Hg-bearing compounds during handling, preservation or proximal exposure to other samples preserved in this manner. Much lower total Hg in more recent samples is consistent with the biologically expected dominant fraction of total Hg as MeHg [15]. Changes in feather MeHg concentrations over the 130 year record

therefore reflect changes in the dietary exposure of the birds to MeHg. The widespread use of IHg for preservation means that we cannot assess the stability of MeHg in feather keratin over time [16,17].

After accounting for biogeochemical processes that influence stable-isotope values in foodwebs, we found no evidence for a change in ivory gull foraging behaviour over 130 years. This contrasts with other seabirds over similar time frames in the North Atlantic [38,51], North Pacific [18,52], and Indian and southern Oceans [53–55]. While we found that ivory gull stable-isotope values remained constant, their diet could have changed to different prey with similar isotopic values. Ivory gulls' diet consists of scavenging marine mammals, and ice-associated marine fishes [56], and they tend to occupy a high trophic position in the Arctic food web year-round [27], but there are few quantitative assessments. Karnovsky *et al.* [56] examined the stomach contents of five birds, and found Arctic cod (*Boreogadus saida*) otoliths, an unidentified liparid fish, and bones from a star-nosed mole (*Condylura cristata*). Scavenged meat and blubber from marine mammals would not leave hard parts, and would be under-represented, or missing entirely in stomach content analysis.

MeHg in ivory gull feathers increased significantly over the past 130 years, despite the lack of evidence of a shift in diet. We attribute this increase to increases in the amount of Hg in the environment that has been observed post-industrially and attributed to human activity [57]. The increase in MeHg was consistent in adults using both analytical methods (a GLM, and a robust regression), but only the GLM approach found a significant increase in first-winter birds. We had high statistical power to detect a temporal trend using robust regression (99%; table 1), and the increase found for first-winter birds using a GLM ($\beta = 0.009 \pm 0.004$), nearly bound zero, and was small. First-winter ivory gulls' MeHg burden has therefore increased over time, but at a lower rate than adults. This could be because first-winter ivory gulls' feathers were grown less than six months prior to collection on the breeding grounds, whereas adult feathers contain MeHg accumulated over a longer period in the body and depurated in feathers. There could also be dietary (but not isotopic) differences between the age classes that resulted in different MeHg exposure.

We had excellent statistical power for detecting changes in adult ivory gull feather MeHg (0.97; table 1), and both analyses produced similar results, namely an increase of 1.6% per year. This corresponds with a $45\times$ increase from our earliest (1880; $0.09 \mu\text{g g}^{-1}$) and most recent adult sample (2004; $4.11 \mu\text{g g}^{-1}$; figure 3). Based on these projections, we anticipate that ivory gulls will show MeHg concentrations of $20 \mu\text{g g}^{-1}$, the concentration believed to cause deleterious effects in piscivores [14], by 2105, though oceanic Hg is expected to increase much more rapidly, increasing fourfold between 2005 and 2050 [58,59], meaning we could expect deleterious concentrations in approximately 50 years. The recently signed Minamata Convention, and changes in global climatic patterns, however, will affect Hg distribution in the Arctic, and consequently its uptake by ivory gulls, though to an unknown extent [60,61].

The concentration of Hg in birds (as indicated by feather Hg) that causes deleterious effects, however, is subjected to numerous factors identified in the literature, including dietary composition, moult duration, phylogeny and ability to demethylate Hg in the liver [62–64]. In a review, Burger & Gochfeld [65] suggested that feather Hg between 5 and $40 \mu\text{g g}^{-1}$ dry weight

Table 1. Robust regression analysis of MeHg time series in seabirds. Reported values are those used in the Arctic Monitoring and Assessment Programme (AMAP).

species/group	<i>n</i>	number of years (range)	% increase per year (95% CI)	years required ^a	lowest detectable change ^b (%)	power of time series ^c
ivory gull, adult	40	23 (1880–2004)	+1.6% (+0.6 to +2.7%)	20	32	0.97
ivory gull, first winter	40	27 (1877–2007)	+0.3% (–0.7 to +1.2%)	21	36	0.99
Atlantic puffin ^d	114	24 (1859–1989)	+1.2% (+0.5 to +1.9%)	16	22	1.00
manx shearwater, NW UK/Ireland ^d	92	11 (1854–1987)	+0.3% (–1.2 to +1.7%)	19	30	0.17
manx shearwater, SW UK/Ireland ^d	43	15 (1866–1989)	+1.4% (+0.7 to +2.1%)	14	17	0.84

^aThe length of time over which a total change of 10% can be detected with 80% statistical power.

^bThe overall detectable change in a 10 year period with 80% power.

^cPower to detect a log-linear trend of 10% over the entire time series.

^dData from Thompson *et al.* [11].

(dw) would be indicative of body burdens of Hg that would have adverse effects, whereas feather Hg concentrations of 10–15 $\mu\text{g g}^{-1}$ are thought to be harmful to bald eagles (*Haliaeetus leucocephalus*) [66]. We therefore used 20 $\mu\text{g g}^{-1}$ as our threshold for adverse effects in ivory gulls, as it is roughly the mid-point of the range given by Burger & Gochfeld [65], a more conservative value than the 10–15 $\mu\text{g g}^{-1}$ range of Cristol *et al.* [66], and is given as the concentration where adverse effects are expected in piscivorous loons [14].

There was no significant trend in total Hg in ivory gull eggs from Seymour Island, Nunavut, collected in 1976, 1987 and 2004 [24], but eggs collected in 2004 had the highest Hg concentrations recorded in any Arctic seabird (6.37 $\mu\text{g g}^{-1}$ dw, 1.61 $\mu\text{g g}^{-1}$ wet weight (ww); 74.6% moisture, B. Braune 2006, unpublished data). Furthermore, egg Hg concentrations in Canadian ivory gulls were an order of magnitude greater than those in Eurasia (range: 0.15–0.26 $\mu\text{g g}^{-1}$ ww; 0.59–1.02 $\mu\text{g g}^{-1}$ dw assuming the same moisture content) [67]. The processes for Hg deposition in feathers and eggs differ, so the concentrations are not directly comparable. More important, however, is that birds' exposure to Hg has obviously increased since the nineteenth century, and whereas concentrations in different tissue types, or even different feather tracts, are not comparable directly [7], our measurements within a single feather type show an increasingly rapid rise in Hg exposure that is mirrored in egg concentrations [24]. Egg sampling, however, is destructive which is an important consideration for sampling tissues for this declining species, and as a consequence, is unlikely to provide large sample sizes, especially for any given year. Further monitoring of Hg in ivory gulls should focus on feathers in live-caught birds.

The increase in ivory gulls' MeHg, particularly that of adults, was much greater than the other species and locations analysed (table 1), and twice the median annual log-linear change in a review of 83 Arctic Hg time series [+0.8%; 48]. Of the 45 Arctic datasets that met minimum statistical power requirements, 10 (22%) had an increase more than 1% per year, seven (16% overall) of which were in the Canadian Arctic or western Greenland [48]. The increase in adult ivory gulls' MeHg was similar to mesopelagic-feeding seabirds from the Azores, (*Calonectris diomedea*: +1.4% per year), but

less than mesopelagic specialists (*Bulweira bulwerii*: +2.9% per year, *Oceanodroma castro*: +4.1% per year) [68]. Procellariiformes (albatrosses, shearwaters, petrels and storm-petrels), however, tend to have higher feather Hg concentrations than other species owing to their protracted moulting periods [69,70].

In Canada, total Hg in thick-billed murre (*Uria lomvia*) and northern fulmar (*Fulmarus glacialis*) eggs increased from 1975 to 2003 (+2.9% and +1.8%, respectively), but has been stable in fulmars from 2003 to 2009, and has remained constant in black-legged kittiwakes (*Rissa tridactyla*) from 1975 to 2009 [71]. Hg temporal trends in other marine mammal predators varied by location, with some increasing and others decreasing [71].

Hg can have wide-ranging deleterious effects on individuals, populations and communities of birds. Individual wandering albatrosses (*Diomedea exulans*) with higher blood Hg were less likely to breed, their eggs less likely to hatch and their chicks less likely to fledge [72], and clapper rails' (*Rallus longirostris*) body condition was negatively related to blood and feather Hg concentrations [73]. Declining populations of rusty blackbirds (*Euphagus carolinus*) tend to have higher blood and feather Hg than other populations [74], and several species within a songbird community more than 100 km downstream from an historical Hg point source had Hg concentrations of concern [75]. Hg in eggs of Arctic-breeding shorebirds, however, did not seem to have an effect on population trends [76].

Ivory gull populations have declined precipitously in Canada since the 1980s [21], and we posit that this could be, in part, the result of significant increases in MeHg in both adult and first-winter birds. We attribute this increase in MeHg to increases in anthropogenically derived environmental MeHg, not changes in diet as shown by the lack of change in stable-isotope values. An increase in total Hg in ivory gull eggs from 1976 to 2004 supports our finding [24], and would suggest that the population decline may be, at least in part, owing to declines in reproductive success from Hg exposure [77]. Bioavailable Hg, transported long distances in the atmosphere from emission sources in Asia, is expected to continue increasing in the Arctic [2], raising alarm for continued population declines in ivory gulls and other high-latitude

species at risk that are otherwise far from sources of environmental contaminants.

Ethics statement. Feathers were collected with the permission of museum curators, and under Canadian Wildlife Service permits 12-SK-SC004, POS-399, SP-2781 and CWS98-S004.

Data accessibility. Specimen collection data, stable isotope values ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), total and methyl mercury concentrations and museum catalogue numbers. Figshare <http://dx.doi.org/10.6084/m9.figshare.1285424>.

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Authors' contributions. A.B. conceived and helped design the study, collected samples, performed statistical analyses and led the manuscript writing. K.H. helped design the study, and performed stable isotope analyses. B.B. performed mercury analyses. All authors edited the manuscript considerably and gave their final approval for publication.

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