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Baseline

Mercury concentrations in feathers of marine birds in Arctic Canada

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ABSTRACT

Mercury (Hg) concentrations are a concern in the Canadian Arctic, because they are relatively high compared to background levels and to similar species farther south, and are increasing in many wildlife species. Among marine birds breeding in the Canadian Arctic, Hg concentrations have been monitored regularly in eggs and intermittently in livers, but feathers have generally not been used as an indicator of Hg exposure or burden. We examined Hg concentrations in six marine bird species in the Canadian Arctic. Ivory gull *Pagophila eburnea*, feather Hg was exceptionally high, while glaucous gull *Larus hyperboreus* feather Hg was unexpectedly low, and ratios of feather THg to egg THg varied across species. The proportion of total Hg that was comprised of methyl Hg in ivory gull feathers was lower than in other species, and may be related to photo-demethylation or keratin breakdown in semi-opaque feather tissue.

Mercury (Hg) concentrations are elevated in many ecosystems across the Canadian Arctic (Dietz et al., 2013; Lavoie et al., 2013). Different forms of mercury (species) occur in the environment, with methylmercury (MeHg) readily accumulating in organisms due to its affinity for cellular proteins (Dietz et al., 2013), and thereby accounting for the majority of the mercury found within tissues in higher trophic levels of a system (>90%; Ackerman et al., 2013), compared to inorganic mercury species. MeHg may be detrimental to organisms because it has negative impacts on their physiology, including acting as a neurotoxin and immunotoxin (Wolfe et al., 1998). Methylmercury will biomagnify in food webs by a factor of 4–10 per trophic step (Kidd et al., 2011; Lavoie et al., 2013). Hence, organisms feeding at high trophic levels may accrue high concentrations of MeHg. This is particularly evident in Arctic marine birds (Braune et al., 2002, 2006, 2014). To date, most Hg research on marine birds in the Canadian Arctic has examined concentrations in liver, muscle or egg tissues (Mallory and Braune, 2012). However, for most seabirds in the Canadian Arctic, sampling Hg in feathers has not been undertaken to date, despite the advantages that the bird or egg does not need to be destroyed during sampling (a particular benefit in the case of rare species), shed feathers may be taken from nests, and that feathers are chemically stable (i.e., Hg concentrations do not change in feathers once they are grown; Appelquist et al., 1984). Concentrations of Hg in feathers can vary markedly depending on sex, age and molt sequence (Braune and Gaskin, 1987; Bond and Diamond, 2009), and therefore knowledge of species molt patterns and chronology is essential for interpreting Hg concentrations from feathers. Despite variation among feathers and feather groups, feather sampling has enabled determination of Hg load in certain marine bird species (reviewed in Burger, 1993), including for example northern fulmar *Fulmarus glacialis* (Thompson et al., 1992a,b), Bonaparte's gull *Larus philadelphia* (Braune and Gaskin, 1987), herring gull *Larus argentatus* (Thompson et al., 1993), and albatrosses (Tavares et al., 2013).

Recently, Bond et al. (2015) found a disconcerting pattern in MeHg in feathers from one Arctic seabird, the ivory gull (*Pagophila eburnea*), an endangered species in Canada. Using breast feathers from museum specimens, they measured a 45-fold increase in MeHg in ivory gull feathers over the period 1877–2007, and argued that Hg loads may have contributed to the decline of this species in the past three decades (Gilchrist and Mallory, 2005).

We conducted a pilot study to assess Hg variation in primary feathers across marine bird species breeding in Arctic Canada, to establish reference values for these birds, and in particular for

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two of the key gull species identified by Provencher et al. (2014) for concern with Hg levels, ivory gull and glaucous gull (*Larus hyperboreus*). If concentrations of Hg in feathers varied according to the known trophic position of each species (as they vary in eggs and livers; Provencher et al., 2014), then we predicted that: (1) across bird species, total mercury (THg) and methylmercury (MeHg) concentrations in feathers would be higher in birds occupying higher trophic positions; and (2) mercury concentrations found in feathers would be correlated with those in the literature for mercury concentrations in eggs.

Feather samples of six species of Canadian Arctic-breeding marine birds were collected from 2009 to 2011 from four locations in the Canadian Arctic (Nunavut Territory; Fig. 1), the same locations where eggs have been monitored for Hg (Braune et al., 2006; Akearok et al., 2010). While the ivory gull remains in Arctic waters year-round (Spencer et al., 2014), the other bird species likely spend the winter south of the Arctic in coastal and offshore waters of the northwest Atlantic Ocean (Mosbech et al., 2006; Mallory et al., 2008a; Gaston et al., 2011; Frederiksen et al., 2012). We wanted to compare Hg concentrations in feathers that had been grown by marine birds while in the Arctic, and presumably when relying on foods acquired in the Arctic to supply much of the nutrients for feather growth (although some Hg would reflect longer-term concentrations in the body from year-round exposure). Consequently, we referred to published guides reporting the known feather molt patterns of these species (Ginn and Melville, 1983; Gaston and Hipfner, 2000; Goudie et al., 2000; Mallory et al., 2008b, 2012a,b; Weiser and Gilchrist, 2012) to select feathers grown while the birds were in the Arctic. For all species, this meant analyzing one of their inner primary (flight) feathers, which would have been among those grown during or shortly after the previous breeding season prior to southward migration. However, for the ivory gull, a species that spends its entire year in Arctic waters, we also analyzed some body and tertial feathers, as the collection of feathers from nests was opportunistic. Because Hg content can vary with feather position and molt sequence (Braune, 1987; Head et al., 2011), we standardized to the extent possible by sampling the same feathers within and across species. Primary feathers (position 1, 2 or 3) were collected for analysis from carcasses of thick-billed murre (*Uria lomvia*; $n = 10$), northern fulmar ($n = 10$), and black-legged kittiwake (*Rissa tridactyla*; $n = 2$) at Prince Leopold Island (74°N, 90°W) in 2009 as part of an International Polar Year project (Gaston et al., 2011). Shed feathers of ivory gull (*P. eburnea*; $n = 8$) were collected in 2010 from eight different nests on Seymour Island (Mallory et al., 2012a); these were mostly primary feathers (estimated positions 2–5), although some body feathers and tertials were collected. Primary feathers (position 2, 3 of three birds, position 4, 5 of one bird) of glaucous gull (*L. hyperboreus*; $n = 4$) were sampled in 2010 from carcasses collected from Nasaruvaalik Island (75.8°N, 96.3°W) as part of a long-term study at that site (Mallory et al., 2012b). Primary feathers (position 1, 2 or 3) of common eider (*Somateria mollissima borealis*; $n = 10$) were collected in 2011 from carcasses of birds sampled near Cape Dorset (64.2°N, 76.6°W; Provencher, 2013). We analyzed THg from the third primary and MeHg from the second primary, with the exception of the one glaucous gull (above) and ivory gull feathers.

Sample feathers were cleaned for analysis by washing three times with Milli-Q water and were then oven-dried overnight at 60 °C. The weight of the dry feathers was measured to the nearest 0.1 mg and recorded. Those feathers weighing $\leqslant$50 mg were digested in 10 mL of 25% KOH/MeOH solution, while those >50 mg were digested in 40 mL of 25% KOH/MeOH solution. A sample aliquot (20 μL) of the digested sample was transferred to a reaction bubbler and analyzed for MeHg and inorganic Hg content through ethylation with $NaB(C_2H_5)_4$ and purge-and-trap gas chromatography prior to detection by atomic fluorescence spectroscopy (Brooks Rand Model III) by Florida Department of Environmental Protection method HG-003-2.10 (Edmonds et al., 2012). Concurrent calibration for MeHg and Hg(II) was performed and THg was determined by addition of inorganic and methylmercury values. Internal quality control included analytical sample replication and certified reference material (DOLT-4, National Research Council of Canada). The mean relative percent difference (standard deviation [SD] /mean) for analytical sample replication was 10.0% for MeHg, 9.5% for Hg(II), and 7.1% for THg. The mean recoveries for the certified reference material ($n = 4$) was 99.9% for MeHg, 107.9% for Hg(II), and 103.7% for THg. Analytical detection limits ($3 * SD$ of reagent blanks) were 0.29 pg for MeHg and 2.41 pg for Hg(II). Method detection limits ($4 * SD$ of method blanks) were 1.31 pg for MeHg and 23.01 pg for Hg(II). All samples were well above detection limits.

Kolmogorov–Smirnov tests indicated that data distributions for some species did not approximate normality, so we used conservative, non-parametric Kruskal–Wallis (KW) tests to assess whether there were overall significant differences ($p < 0.05$) in MeHg and THg concentrations among species. We followed this with Dunn's Multiple Comparison test if the KW test was significant, to determine which species had MeHg or THg values significantly different from each other. All tests were conducted with InStat (GraphPad Software, 2009).

There was considerable interspecific variation in Hg concentrations derived from primary feathers, which led to significant differences in median values among the six species for both THg ($KW = 31.1$, $n = 44$, $p < 0.001$) and MeHg ($KW = 27.0$, $n = 44$, $p < 0.001$). For both types of Hg, highest concentrations were found in ivory gull and lowest were in common eider (Table 1). Ivory gull also had the largest range in values and the highest coefficient of variation for both THg and MeHg. Comparing THg and MeHg medians among species, ivory gull had significantly higher Hg values than common eider (Dunn's Multiple Comparisons Test, $p < 0.001$) and thick-billed murre ($p < 0.05$), while northern fulmar had higher Hg feather concentrations than common eider ($p < 0.01$). In fact, the minimum THg in all three types of ivory gull feathers was higher than the maximum THg in most other species. The highest measured THg was 43.66 μg/g dw in primary feathers from one ivory gull. Although body and tertial feathers of ivory gulls had lower median THg and MeHg than primary feathers (Table 1), these differences were not significant ($KW = 1.1$, $n = 15$, $p > 0.6$), presumably due to the small sample sizes and high variation among feather samples.

There was also a significant difference in the proportion of MeHg/THg in primary feathers among sample species ($KW = 28.3$, $n = 44$, $p < 0.001$). The lowest MeHg/THg ratio was that of ivory gull (67%), which was 20% lower than the next lowest proportion in common eiders (Table 1).

Our data on Hg in feathers of marine birds in the Canadian Arctic were consistent with previous studies on other avian tissues, and assuming that much of the Hg comes from local Arctic food supplies, these data suggest that contamination of Arctic marine birds by long-range transport of Hg emitted from locations in temperate and tropical regions continues to be a serious environmental concern (Braune et al., 2006, 2010). Based on available tracking data or information on arrival and nesting dates, the six species in our pilot study use Arctic food webs as their main source of nutrients to form their eggs or to grow the feathers we analyzed (Gaston and Hipfner, 2000; Goudie et al., 2000; Mosbech et al., 2006; Mallory et al., 2008c; Frederiksen et al., 2012; Sénéchal et al., 2011; Spencer et al., 2014). We found that MeHg concentrations in feathers of Canadian Arctic marine birds (Table 1) spanned much of the range reported in studies from tropical to polar regions (from various feather types; means of 0.43–28.0 μg/g dw;