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ARTICLE

Mercury concentrations in multiple tissues of Arctic Iceland Gulls (*Larus glaucoides*) wintering in Newfoundland

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Abstract: Anthropogenic mercury (Hg) emissions are increasing and are potentially of concern for Arctic-nesting seabirds, particularly those that spend part of their year near dense human habitation. Iceland Gulls (*Larus glaucoides* B. Meyer, 1822) breed in the eastern Canadian Arctic and spend the majority of winter in towns and cities in eastern Newfoundland. We measured Hg in breast feathers, blood plasma, and red blood cells of Iceland Gulls wintering in and around St. John's, Newfoundland and Labrador, from 2011 to 2014. Mercury in blood plasma comprised <10% of the total blood Hg. We found no difference in red blood cell Hg between first-winter and adult birds, which likely reflects their similar feeding habits. Feather Hg in adults was significantly greater than that in first-winter birds because adults had accumulated a greater body Hg burden to excrete (up to a year, compared with a few months' accumulation in first-winter birds). Overall, concentrations were among the lowest found for *Larus* spp. and Arctic gulls, suggesting that Hg does not pose a risk to Iceland Gulls at the present.

Key words: blood, feather, *Larus glaucoides*, mercury, Newfoundland.

Résumé : Les émissions de mercure (Hg) anthropiques ne cessent d'augmenter et présentent une préoccupation potentielle pour les oiseaux de mer nidifiant en Arctique, en particulier ceux qui passent une partie de l'année près des habitats humains denses. Les goélands arctiques (*Larus glaucoides* B. Meyer, 1822) se reproduisent dans l'est de l'Arctique canadien et la plupart hivernent dans les villes de l'est de Terre-Neuve. Nous avons analysé la quantité de Hg dans les plumes de poitrine, le plasma sanguin et les globules rouges des goélands arctiques hivernant dans et autour de St. John's, Terre-Neuve et Labrador, de 2011 à 2014. Le Hg dans le plasma sanguin représente <10% du total du Hg dans le sang. Nous n'avons trouvé aucune différence de niveau de Hg dans les globules rouges entre les oiseaux adultes et ceux plus jeunes pour qui c'est le premier hiver. Ainsi, ceci reflète probablement leurs habitudes alimentaires, qui sont similaires. Le Hg dans les plumes chez les adultes était beaucoup plus élevé que chez les oiseaux plus jeunes passant leur premier hiver, car les adultes avaient accumulé une plus grande charge corporelle de Hg à excréter (jusqu'à un an, comparativement à une accumulation de quelques mois chez les jeunes oiseaux passant leur premier hiver). Dans l'ensemble, les concentrations les plus faibles trouvées étaient chez les *Larus* spp. et les goélands arctiques, ce qui semble indiquer que le Hg ne pose pas en ce moment de risque pour les goélands arctiques.

Mots-clés : sang, plume, *Larus glaucoides*, mercure, Terre-Neuve.

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Introduction

Anthropogenic mercury (Hg) is increasing globally and especially so in the Arctic (Lindberg et al. 2007; AMAP 2011; Krabbenhoft and Sunderland 2013). The biomagnifications of Hg through food webs can pose a serious risk to high-trophic predators (Lavoie et al. 2013), resulting in the potential for reproductive impairment, behavioural effects, and reduced survival (Gilman et al. 1977; Heinz 1979; Burger and Gochfeld 2002; Burgess and Meyer 2008; Erikstad et al. 2013; Tozer et al. 2013; Goutte et al. 2014).

Migratory animals can transport contaminants long distances (Blais et al. 2005), and exposure to contaminants and other stressors on the nonbreeding grounds can affect subsequent life history stages (Harrison et al. 2011; Fort et al. 2014; Fairhurst et al. 2015). Arctic wildlife that spend considerable portions of their annual cycle in more urbanized areas could therefore be important vectors for Hg to Arctic regions (Lewis et al. 1993; Gochfeld 1997; Burger and Elbin 2015*a*, 2015*b*), and the Hg they acquire during the winter could influence individuals on their Arctic breeding grounds (Fort et al. 2014; Lavoie et al. 2014).

The Canadian Arctic supports >12 000 pairs of Iceland Gulls (*Larus glaucoides* B. Meyer, 1822), which represents essentially the entire North American breeding population and 20%–50% of the global population (Boertmann et al. 1996; Gaston et al. 2012). Canadian Iceland Gulls (*Larus glaucoides kumlieni* Brewster, 1883) migrate south in the winter to eastern Canada and the northeastern United States (Snell 2002). In Newfoundland, large congregations representing the majority of the breeding population spend mid-November through March in mixed-species flocks with other *Larus* spp. gulls (Brown 1986; Huettmann and Diamond 2000; Huang et al. 2014; authors' personal observations). While in Newfoundland, they forage in urban areas, including sewerage outflows, landfills, and public parks. In coastal urban environments, such as the northeastern Avalon Peninsula, Newfoundland, gulls may be exposed to Hg mobilized by sediment disturbance caused by storms and wave action, particularly point sources, as often occur near long-established human settlements (Burger and Elbin 2015*a*).

There have been no previous assessments of Hg in Iceland Gulls in Canada (Mallory and Braune 2012; Provencher et al. 2014) and only one elsewhere: mean Hg in liver tissue of birds from Greenland in 1994–1995 ranged from 0.24 to 0.60 μg/g (Rigét et al. 2000). We therefore sought to determine concentrations of Hg in three tissues (breast feathers, blood plasma, and red blood cells (RBCs)) of Iceland Gulls wintering in and around St. John's, Newfoundland and Labrador, Canada, and to compare our results with Hg in other large gulls.

Methods

Sample collection

We trapped 47 Iceland Gulls in and around St. John's, Newfoundland and Labrador, between November 2011 and January 2014. A further 11 feather samples were obtained from birds obtained during authorized gull control operations at the Robin Hood Bay waste management facility in February 2014 (Table 1). Birds were identified to age class (first winter or adult; only two age classes were present in our sample) based on plumage (Olsen and Larsson 2004; Pyle 2008). We sampled four breast feathers from 21 individuals (10 live caught and 11 from control operations), which were likely moulted during the winter in adult birds (Pyle 2008) and were grown on the breeding ground in first-winter individuals. We also sampled whole blood from 25 live individuals, using a needle and syringe, from the brachial vein and centrifuged it within 6 h of collection to separate RBCs and blood plasma. Samples were then stored in sterile centrifuge tubes at −20°C prior to analyses.

Table 1. Sample sizes of Iceland Gull tissues analysed for Hg in eastern Newfoundland (sampling year refers to winter periods (November–February)).

	Sampling year			
Tissue and age class	2011–2012	2012–2013	2013–2014	Total
Feather				
Adult			6	6
First-winter			15	15
Plasma				
Adult	1	3		4
First-winter	2	12	4	18
RBCs				
Adult	3	4		7
First-winter	2	11	4	17

Mercury analysis

Samples were shipped frozen to the School of Environment and Sustainability, University of Saskatchewan (Saskatoon, Saskatchewan, Canada), for total Hg (THg) analysis by atomic absorption spectrometry using a Direct Mercury Analyzer (DMA-80, Milestone Inc., Shelton, Connecticut, USA) (Haynes et al. 2006). Mercury found in blood and feathers is predominantly (>90%) methyl mercury, the bioactive form (Bond and Diamond 2009*b*; Lavoie et al. 2010), so we analysed THg as a proxy. Two feathers from each individual were analysed as a pool to account for within-individual variation (Bond and Diamond 2008). Approximately 0.30 mg (wet mass) of RBCs and plasma were analysed for THg, but owing to low concentrations found in plasma, all samples were repeated using 0.60 mg (wet mass), and these are reported here.

Method blanks (n = 8) were all below the detection limit (0.04 ng of THg). Values were adjusted using recovery of THg from a low-concentration certified reference material, DORM-4 (fish protein homogenate; National Research Council, Ottawa, Ontario, Canada; certified concentration 0.410 ± 0.055 µg/g, recovery 98% ± 10%, n = 8), and one high-concentration certified reference material, IAEA-85 (human hair; International Atomic Energy Agency, Vienna, Austria; certified concentration 23.2 µg/g (95% CI 22.4–24.0), recovery 102% ± 1%, n = 4). Both reference materials were calibrated against TORT-3 (lobster hepatopancreas; National Research Council, Ottawa, Ontario, Canada) analysed at a range of masses (certified concentration 0.292 ± 0.022 µg/g, recovery 102 ± 9%, n = 20). Data are presented as µg/g fresh weight for feathers and wet weight for plasma and RBCs.

Statistical analysis

We tested for normality using Shapiro–Wilk's test (Shapiro and Wilk 1965), but finding no improvement after log-transforming Hg (see Results), we used untransformed values for further analysis. Because feathers were all sampled within the same winter (2013–2014) (Table 1), we constructed a general linear model to determine whether there were differences in feather Hg among age classes. Plasma and RBCs were collected in multiple years, but often with low sample sizes in any given year. We therefore treated "year" (defined as the winter of collection) (Table 1) as a random factor in a general linear mixed model and age class as a fixed factor. We then compared the general linear mixed model with a null model containing only the random factor (i.e., different intercepts for each year) using a likelihood ratio test to assess the significance of the age effect. All analyses were done in

Table 2. Mercury concentrations in feathers of other *Larus* spp. and Arctic gulls (values are presented as μg/g dry weight).

Species	Location	Year	Age class	Mean ± SD (*n*)	Reference
Herring Gull *Larus argentatus* Pontoppidan, 1763	New York	1993	Adult	0.38 ± 0.27 (45)	Burger 1995
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2014**	**Adult**	**1.90 ± 1.00 (6)**	**This study**
Common Gull *Larus canus* L., 1758	Caspian Sea	2008	Adult	2.90 ± 1.40 (3)	Rajaei et al. 2010
Siberian Gull *Larus heuglini* Bree, 1876	Persian Gulf	2011	Adult	2.99 ± 0.27 (15)	Majidi et al. 2015
Herring Gull *Larus argentatus* Pontoppidan 1763	Poland	2009–2010	Adult	3.02 (6)	Szumiło et al. 2013
Lesser Black-backed Gull *Larus fuscus* L., 1758	Scotland	1986	Adult	3.40 ± 2.90 (15)	Thompson et al. 1990
Herring Gull *Larus argentatus* Pontoppidan, 1763	Scotland	1986	Adult	3.40 ± 3.90 (15)	Thompson et al. 1990
Glaucous-winged Gull *Larus glaucescens* J.F. Naumann, 1840	Aleutian Islands	2004	Adult	3.68 ± 0.37 (63)	Burger and Gochfeld 2009
Ivory Gull *Pagophila eburnea* (Phipps, 1774)	Arctic Canada	2004	Adult	4.11 (1)	Bond et al. 2015
Herring Gull *Larus argentatus* Pontoppidan, 1763	Wadden Sea	1990	Adult	4.87 ± 2.04 (23)	Lewis et al. 1993
Herring Gull *Larus argentatus* Pontoppidan, 1763	Wadden Sea	1990	Adult	6.41 ± 2.39 (9)	Lewis et al. 1993
Ivory Gull *Pagophila eburnea* (Phipps, 1774)	Arctic Canada	2007	First-winter	0.28 (1)	Bond et al. 2015
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2014**	**First-winter**	**1.08 ± 0.30 (15)**	**This study**
Herring Gull *Larus argentatus* Pontoppidan, 1763	Poland	2009–2010	Immature	3.30 (7)	Szumiło et al. 2013

R 3.1.2 (R Core Team 2014) and using the package *lme4* (Bates et al. 2014). Effects were considered significant when $p < 0.05$ and the data are presented as mean ± SD.

Results

Mercury was not normally distributed (Shapiro–Wilk's $W = 0.879$, $p < 0.001$), and log-transforming data did not produce normally distributed data ($W = 0.583$, $p < 0.001$), so we used untransformed data for subsequent analyses.

Feather Hg was significantly higher in adult birds (1.90 ± 1.00 μg/g) than in first-winter birds (1.08 ± 0.30 μg/g; $F_{1,19} = 8.72$, $p = 0.008$; range for both age classes 0.64–3.36 μg/g) (Table 2). Mercury was also higher in adults' plasma (0.10 ± 0.12 μg/g) than in the plasma of first-winter birds (0.03 ± 0.04 μg/g; likelihood ratio test $\chi^2 = 5.14$, $p = 0.023$; range of both age classes 0.01–0.26 μg/g) (Table 3), while there was no difference between age classes

Table 3. Mercury concentrations in blood plasma and RBCs of other *Larus* spp. and Arctic gulls. Values are presented as μg/g. wet weight.

Species	Location	Year	Tissue (age class)	Mean ± SD (*n*)	Reference	Notes
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2011–2014**	**Plasma (adult)**	**0.10 ± 0.12 (4)**	**This study**	
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2011–2014**	**Plasma (first-winter)**	**0.03 ± 0.04 (18)**	**This study**	
Herring Gull *Larus argentatus* Pontoppidan, 1763	Gulf of St. Lawrence	2006	RBCs (adult)	0.48 ± 0.05 (20)	Lavoie et al. (2010)	66.5% moisture
Great Black-backed Gull *Larus marinus* L., 1758	Gulf of St. Lawrence	2006	RBCs (adult)	1.04 ± 0.14 (20)	Lavoie et al. 2010	67.4% moisture
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2011–2014**	**RBCs (adult)**	**1.14 ± 0.37 (7)**	**This study**	
Iceland Gull *Larus glaucoides* B. Meyer, 1822	**Newfoundland**	**2011–2014**	**RBCs (first-winter)**	**1.15 ± 0.32 (17)**	**This study**	

in RBC Hg (adult 1.14 ± 0.37 μg/g, first winter 1.15 ± 0.32 μg/g; likelihood ratio test $\chi^2 = 0.024$, $p = 0.88$; range for both age classes 0.37–1.76 μg/g) (Table 3).

Discussion

The Hg concentrations that we found in Iceland Gull tissues are below those thought to have any adverse effects on individuals, estimated as 5–40 μg/g for feathers (Burger and Gochfeld 1997; Cristol et al. 2012; Evers et al. 2014) and 3–4 μg/g for whole blood (Burgess et al. 2005; Evers et al. 2008). Iceland Gulls' feather Hg therefore does not likely represent a toxicological risk to individuals.

Importantly, we found that the vast majority of Hg in blood was associated with the cellular fraction rather than with plasma (Table 3). In mammals, 90% of blood Hg is found in RBCs and 10% in plasma (Aihara and Sharma 1986; Wolfe et al. 1998). We found, on average, 2.5%–8.8% of blood Hg in plasma (Table 3). Given the high moisture content of plasma (>90%), the low concentrations of Hg, and the mass required for detection with current instruments, plasma is not a suitable matrix in which to regularly measure Hg, and whole blood or the cellular fraction is recommended. This is convenient, as other assays requiring plasma samples, such as hormones, diseases, or stable isotopes (Hobson and Clark 1993; Buck et al. 2007; Huang et al. 2014), can be paired with assays of RBCs, such as Hg, stable isotopes, or genetics (Norris et al. 2005; Robinson et al. 2013), maximizing the information available from a single sample.

We found that blood Hg was similar between age classes, which likely reflects the fast turnover of blood and the lack of age-class-based segregation during the winter. Mercury in blood represents recent dietary exposure before Hg is demethylated in the liver or mobilized from body stores before depuration in feathers or guano (Kim et al. 1996; Monteiro and Furness 2001*a*). Flocks of Iceland Gulls in Newfoundland in the winter comprise all age classes and likely have similar diets and therefore dietary Hg exposure.

Adults' feather Hg, however, was considerably higher than that of first-winter birds. Because feather Hg is the result of excretion from the Hg body burden accumulated between moults (often annual or subannual) (Monteiro and Furness 2001*a*) and first-winter birds grew their contour feathers on their Arctic breeding ground, they would only be

eliminating Hg for a short period and from a place with little anthropogenic inputs (from hatching to feather growth or no more than 3 months) (Snell 2002). In contrast, when adults grew their contour feathers, they would have accumulated 1 year's worth of Hg in their body pool and consequently eliminating more through feather replacement (Monteiro and Furness 2001*a*, 2001*b*). Lower Hg in first-winter birds' feathers has been found in other gulls (Burger 1995; Burger and Gochfeld 1999; Bond et al. 2015) and in seabirds generally (Becker et al. 1994; Becker et al. 2002; Bond and Diamond 2009*a*).

In comparison with other large *Larus* spp. or Arctic-breeding gulls, Iceland Gulls tend to have Hg concentrations among the lowest recorded (Tables 2 and 3). Their diet has not been well studied and in Newfoundland is unknown. Elsewhere, they consume a variety of marine invertebrates, particularly mussels (Ingolfsson 1967), but are known to exploit anthropogenic food sources, namely human refuse (Renaud et al. 1981). Mercury is spatially heterogeneous in marine and freshwater environments, and although eastern Newfoundland has had significant human habitation for several hundred years, industrial point sources of Hg are few. Gulls from the Wadden Sea and New York Bight, where contamination is much higher, experience a higher Hg burden (Lewis et al. 1993; Burger and Gochfeld 2003; Burger and Elbin 2015*a*), and even within eastern Canada, Hg in gulls is spatially variable (Goodale et al. 2008; Burgess et al. 2013).

Although Iceland Gulls winter in urbanized areas of eastern Canada and could be exposed to significant concentrations of dietary Hg and transport it to the Arctic, they have among the lowest concentrations of Hg of any gull and are not likely affected by Hg based on the concentrations that we observed.

Data availability

Data are available on figshare: http://dx.doi.org/10.6084/m9.figshare.1333601.

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