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Q1 Do concentrations in eggs and liver tissue tell the same story of temporal trends of mercury in high Arctic seabirds?

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ABSTRACT

Mercury (Hg) remains a key contaminant of concern in Arctic biota, and monitoring of Hg concentrations in seabird tissues will be an effective approach to track the effects of implementing the Minamata Convention. We examined trends in total Hg (THg) in liver and egg tissues of two Arctic seabirds, thick-billed murres (*Uria lomvia*) and northern fulmars (*Fulmarus glacialis*), between 1976 and 2013 to assess whether both tissues showed similar patterns of Hg change. Hepatic THg was consistently higher than egg THg, and both species had similar egg THg concentrations, but fulmars had higher hepatic THg than murres. Murre THg concentrations showed more relative variation through time than fulmars. We suggest that egg THg better reflects exposure of birds to THg in local, Arctic prey, whereas liver THg may incorporate longer term, year-round THg exposure. Additional analysis of THg distribution in Arctic seabirds post-laying would help inform interpretation of long-term trends.

Introduction

Mercury (Hg) is a naturally occurring element with no known biological function. Environmental concentrations of Hg are elevated in many ecosystems due to its release from anthropogenic activities and long-range transport (Bidleman et al., 2003; Schroeder and Munthe, 1998). It occurs in several forms, and as methylmercury, it bioaccumulates in biological tissues and biomagnifies in food chains to concentrations that can cause neurological and physiological impairment in organisms (Dietz et al., 2013; Wiener and Spry, 1996). This is of particular concern for human consumption, as many animals (notably fish; Clarkson and Magos, 2006; Scheuhammer et al., 2007) may accumulate high levels of mercury. Mercury concentrations in wildlife are a key concern for indigenous peoples in many countries, notably Arctic Canada, because wild foods continue to form a large portion of their diet (Ford, 2009). Thus, given the human health implications, efforts were undertaken to develop international guidelines on acceptable concentrations of Hg in foods (Lowenstein et al., 2010), and in August 2017, the Minamata Convention on Mercury came into force to reduce Hg entering the environment (http://mercuryconvention.org/). One of the challenges for countries now is to implement Hg controls, and to monitor effectiveness (Evers et al., 2016).

Seabirds are effective monitors of marine environmental conditions (Furness and Camphuysen, 1997), and have been used to assess and monitor levels of contaminants including Hg in marine food webs (Monteiro and Furness, 1995). This has been particularly evident in the north (Braune, 2007; Dietz et al., 1996; Savinov et al., 2003) where seabirds have formed Q4 a key component of long-term environmental monitoring

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programs for Hg in the circumpolar Arctic (Rigét et al., 2011), in part because seabirds (eggs or birds) are important wild foods for Inuit (Priest and Usher, 2004). The evidence suggests that Hg has been increasing for over a century in some Arctic species (based on museum specimens; Bond et al., 2015), and certainly during the past four decades (Braune, 2007; Braune et al., 2014, 2016). Moreover, the health effects of Hg on Arctic wildlife have been reviewed recently and suggest that some species have mercury levels above known toxicological thresholds (Dietz et al., 2013; Kirk et al., 2012).

In previous studies using Hg concentrations in Arctic seabird eggs, we found that Hg had been increasing since 1975, but that this pattern was nonlinear and that Hg trends have been generally negative since 1993 (Braune, 2007; Braune et al., 2014, 2016). However, as our knowledge on the ecology of seabirds in the Canadian Arctic increased (*e.g.*, Gaston et al., 2005, 2011; Mallory et al., 2008, 2010), notably in relation to changing climatic conditions (Ferguson et al., 2012; Gaston et al., 2005), we argued that it was unclear where migratory seabirds in the Canadian Arctic were experiencing the greatest exposure to Hg, during breeding or during overwintering (Braune et al., 2016). As sea ice conditions change, and birds adjust breeding schedules (Gaston et al., 2009), the traditional timing and locations where birds might be exposed to Hg are changing. Clearly dietary factors and trophic position influence Hg uptake (Atwell et al., 1998; Campbell et al., 2005), but different regions of the Arctic, and the wintering grounds, may be hotspots for Hg as well (Pomerleau et al., 2016; Semeniuk and Dastoor, 2017). Collectively, that uncertainty makes it difficult to interpret factors influencing Hg trends (Braune et al., 2016).

In our current study, we sought to use information on Hg concentrations in different tissues of two Arctic seabird species, thick-billed murres (*Uria lomvia*) and northern fulmars (*Fulmarus glacialis*), to assess whether this helped us distinguish when and where seabirds might be experiencing altered exposure to Hg. Our specific goal was to compare what trends we saw in Hg from two seabird tissues, liver and egg, collected from birds at the same colony in the same years, to evaluate whether those tissues showed the same Hg trends through time. We reasoned that egg Hg should reflect proportionally more of the local, Arctic environment because murres and fulmars are income breeders, relying on local, exogenous food supplies to form their eggs after they arrive at the breeding grounds (Bond and Diamond, 2010; Mallory et al., 2008). In contrast, we expected that liver taken from nesting birds (post egg-laying) would reflect more of the long-term exposure to Hg, including the wintering grounds.

1. Methods

We conducted our study at the Prince Leopold Island Migratory Bird Sanctuary (74°02′N, 90°05′W) in Lancaster Sound, Nunavut, Canada (Fig. 1), the site of long-term seabird population (*e.g.*, Gaston et al., 2005) and contaminant monitoring (*e.g.*, Braune et al., 2016; Mallory and Braune, 2012). We took samples of eggs and adult thick-billed murres as follows: 1976 (9 eggs measured as 3 pools of 3 eggs, 5 adults), 1993 (5 × 3-egg pools, 5 adults), 2008 (5 × 3-egg pools, 5 adults), 2013 (5 × 3-egg pools, 10 adults). Fulmar collections were similar but included an extra year: 1976 (4 × 3-egg pools, 4 adults), 1987 (2 × 3-egg pools, 4 adults), 1993 (5 × 3-egg pools, 5 adults), 2008 (5 × 3-egg pools, 5 adults), 2013 (5 × 3-egg pools, 10 adults). The egg samples are a subset of data presented previously by Braune et al. (2016). We did not include sex as a factor because our earlier work showed that there was no difference in hepatic Hg between sexes for these species and sampling locations (Braune et al., 2014). Samples were taken by scientists who descended the cliffs by rope to the breeding ledges, and then collected eggs by hand or with a small cup attached to the end of an extension pole. Both murres and fulmars lay only a single egg, so we did not have to consider possible influences of egg-laying sequence on Hg concentrations (*e.g.*, Akearok et al., 2010). We captured adults by noose pole while they were at their nests during incubation (Gaston et al., 2005). Adults were immediately euthanized using approved procedures, and all collections were made under appropriate annual animal care (*e.g.*, EC-PN-13-020), land use (N2003J0014), and research permits (*e.g.*, NUN-MBS-12-03, NUN-MBS-SCI-04, WL 2014-040, EP4151936).

We kept eggs cool in the field and then shipped them to the National Wildlife Research Centre (NWRC), Environment Canada, for processing and chemical analyses. Each egg was cut open and the contents homogenized for 10–20 sec using either a stainless steel Sorvall Omni Mixer or Brinkmann Polytron homogenizer. Homogenates were then stored frozen (–40°C) in acid-rinsed polyethylene vials until Hg analysis. Archived samples collected prior to 1993 were retrieved from the National Wildlife Specimen Bank at NWRC and analyzed retrospectively, whereas samples collected from 1993 to 2013 were analyzed within six months of collection. Egg homogenates were analyzed for total Hg as pooled (composite) samples with each pool comprising equal aliquots of three individual eggs (see also Braune et al., 2014). However, we conducted Hg analyses on livers from individual birds.

Egg samples collected during 1975 to 1998 were thawed, freeze-dried and digested in mineral acids, and then analyzed for total Hg (THg) using cold vapor atomic absorption spectrophotometry (CVAAS) with a 3030b-AAS (Perkin-Elmer) equipped with VGA (Varian) vapor generation system and PSC-55 (Varian) autosampler (Neugebauer et al., 2000). Samples collected during 2003–2013 were homogenized, freeze–dried, homogenized again and weighed into nickel combustion boats, and then analyzed for THg by direct combustion of the solid sample in an oxygen-rich atmosphere (see Salvato and Pirola, 1996; EPA, 2007) using an AMA-254 advanced mercury analyzer (Altec) equipped with an ASS-254 autosampler. We determined moisture content by weight loss during freeze-drying. The practical detection limits were 0.2 μg/g dry weight for the 1976, 1987 and 1993 egg samples, 0.006 μg/g dry weight for the 2008 samples, and 0.004 μg/g dry weight for the 2013 samples. All Hg concentrations measured in eggs were above detection limits (Braune et al., 2016 provides more quality assurance details).

Liver samples came from archived tissues which were homogenized with equipment (as above), and then sent to RPC Science and Engineering, Inc. laboratories in Fredericton, Q5 New Brunswick where chemical analyses were undertaken. Portions of samples were prepared by microwave-assisted

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