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Temporal trends of mercury in eggs of five sympatrically breeding seabird species in the Canadian Arctic☆

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ABSTRACT

We compared temporal trends of total mercury (Hg) in eggs of five seabird species breeding at Prince Leopold Island in the Canadian high Arctic. As changes in trophic position over time have the potential to influence contaminant temporal trends, Hg concentrations were adjusted for trophic position (measured as $\delta^{15}N$). Adjusted Hg concentrations in eggs of thick-billed murres (*Uria lomvia*) and northern fulmars (*Fulmarus glacialis*) increased from 1975 to the 1990s, followed by a plateauing of levels from the 1990s to 2014. Trends of adjusted Hg concentrations in eggs of murres, fulmars, black guillemots (*Cepphus grylle*) and black-legged kittiwakes (*Rissa tridactyla*) had negative slopes between 1993 and 2013. Adjusted Hg concentrations in glaucous gull (*Larus hyperboreus*) eggs decreased by 50% from 1993 to 2003 before starting to increase again. Glaucous gull eggs had the highest Hg concentrations followed by black guillemot eggs, and black-legged kittiwake eggs had the lowest concentrations consistently in the five years compared between 1993 and 2013. Based on published toxicological thresholds for Hg in eggs, there is little concern for adverse reproductive effects due to Hg exposure in these birds, although the levels in glaucous gull eggs warrant future scrutiny given the increase in Hg concentrations observed in recent years. There is evidence that the Hg trends observed reflect changing anthropogenic Hg emissions. It remains unclear, however, to what extent exposure to Hg on the overwintering grounds influences the Hg trends observed in the seabird eggs at Prince Leopold Island. Future research should focus on determining the extent to which Hg exposure on the breeding grounds versus the overwintering areas contribute to the trends observed in the eggs.

1. Introduction

Anthropogenic emissions of mercury (Hg) have increased over the past century (Amos et al., 2015) and have continued to increase over recent decades (AMAP, 2011). Due to its highly volatile nature, elemental Hg partitions readily into air where it can undergo long-range atmospheric transport to remote regions, such as the Arctic, where an increase in Hg has been documented since the Industrial Era in both abiotic and biotic matrices (Beal et al., 2015; Bond et al., 2015; Dietz et al., 2009). Mercury has continued to increase in Arctic biota over more recent decades (Rigét et al., 2011). Mercury biomagnifies through the food chain making those species feeding at high trophic positions especially vulnerable to Hg exposure via their diet (Atwell et al., 1998; Campbell et al., 2005; Jaeger et al., 2009). This is the case for seabirds, which feed at relatively high trophic positions in Arctic marine food webs (Atwell et al., 1998; Hobson et al., 1994, 2002; Jaeger et al., 2009).

The most bioavailable and toxic form of Hg is methylmercury (MeHg), and nearly 100% of the Hg transferred to avian eggs is in the form of MeHg (Wiener et al., 2003). Dietary MeHg is efficiently transferred to eggs in a dose-dependent manner (Wolfe et al., 1998), making eggs an ideal and relatively non-intrusive matrix for monitoring Hg (Ackerman et al., 2013).

In the Canadian Arctic, seabird populations sampled from colonies at higher latitudes generally have greater concentrations of Hg than those at lower latitudes (Braune et al., 2002, 2014a). Since 1975, Hg concentrations have increased in eggs of thick-billed murres (*Uria lomvia*) and northern fulmars (*Fulmarus glacialis*) breeding at Prince Leopold Island in Lancaster Sound in the Canadian high Arctic (Braune, 2007), although Hg concentrations in the

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murre eggs appear to have declined in recent years (Braune et al., 2014b).

In this study, we examine temporal trends of Hg in eggs of five seabird species breeding at Prince Leopold Island in order to determine if the trends in Hg exposure among species are similar over a given time period. Four of the species, the thick-billed murre (a.k.a. Brünnich's guillemot), northern fulmar, black-legged kittiwake (*Rissa tridactyla*) and black guillemot (*Cepphus grylle*), feed in the marine environment, while the glaucous gull (*Larus hyperboreus*) feeds in both marine and terrestrial environments, although the birds at Prince Leopold Island probably feed principally on marine prey during the breeding season. Northern fulmars and black-legged kittiwakes are surface feeders with kittiwakes consuming primarily fish and zooplankton (Hatch et al., 2009), and fulmars feeding primarily on zooplankton, cephalopods and fish (e.g. Arctic cod *Boreogadus saida*) in the Canadian Arctic, although fulmars are also known to scavenge (Mallory et al., 2010, 2012). Both fulmars and kittiwakes have been observed feeding on Arctic cod in the Lancaster Sound area (Matley et al., 2012). Thick-billed murres are pursuit divers and the adult diet at Prince Leopold Island is dominated by Arctic cod (Bradstreet, 1980; Gaston and Nettleship, 1981; Provencher et al., 2012). Black guillemots are also pursuit divers and during the breeding season, they prefer shallow, inshore waters where they feed primarily on benthic fish and invertebrates (Butler and Buckley, 2002), with Arctic cod figuring prominently in the diet of guillemots in Barrow Strait west of Lancaster Sound (Bradstreet, 1980). Glaucous gulls prey on marine invertebrates, fish, birds (including eggs and chicks) and small mammals, as well as scavenging fish, carrion (e.g. marine mammals) and human refuse (Weiser and Gilchrist, 2012). At Prince Leopold Island, glaucous gull chicks are fed marine prey including fish as well as the eggs and nestlings of other seabirds (Nettleship et al., 1990). Adults have been observed feeding on Arctic cod in the Lancaster Sound area (Matley et al., 2012). Upon arrival at the breeding colonies at Prince Leopold Island, all five species feed in the surrounding area for several weeks to a month prior to egg-laying (Gaston and Nettleship, 1981; Gaston et al., 2005; McLaren, 1982), although the northern fulmar may forage hundreds of kilometers from the colony (McLaren, 1982).

As changes in trophic position over time have the potential to influence contaminant trends (Braune et al., 2014b; Hebert and Weseloh, 2006; Hebert et al., 2000), we determined trophic position of the egg contents by measuring naturally occurring stable isotopes of nitrogen ($^{15}N/^{14}N$, expressed as $\delta^{15}N$) (Hebert et al., 1999; Hobson and Welch, 1992; Hobson et al., 1994), which reflect the diet of the female prior to, or during, egg-laying (Hebert et al., 1999; Hobson, 1995). We hypothesized that a change in $\delta^{15}N$ values in the eggs reflected a change in seabird diet rather than a baseline change in the isotopic composition of primary production as a previous study in Lancaster Sound suggested that there had been little baseline change in $\delta^{15}N$ over the last three decades (Moody et al., 2012).

2. Materials and methods

2.1. Sample collection and preparation

Eggs of thick-billed murres, northern fulmars, black-legged kittiwakes, black guillemots, and glaucous gulls were collected from the Prince Leopold Island Migratory Bird Sanctuary (74°02′N, 90°05′W) in Lancaster Sound, Nunavut, Canada (Fig. S1). Murre and fulmar eggs were sampled in 1975, 1976, 1977, 1987, 1988 (murre eggs only), 1993, 1998, 2003, and annually from 2005 to 2014. Kittiwake eggs were sampled in 1975, 1976, 1987, 1993, 2003, 2008, and 2013; black guillemot eggs in 1993, 1998, 2004, 2008, and 2013; and glaucous gull eggs in 1993, 1998, 2003, 2008, and 2013. Thick-billed murres and northern fulmars lay a single egg, whereas black-legged kittiwakes and glaucous gulls may lay up to three eggs (Gaston et al., 2005). Black guillemots generally lay one or two eggs (Butler and Buckley, 2002). Eggs were sampled randomly on the basis of one egg per nest as soon after laying as was possible. Eggs were collected by hand or using a small cup attached to the end of an extension pole. All eggs were taken under appropriate research and collection permits.

Eggs were kept cool in the field and shipped to the National Wildlife Research Centre (NWRC), Ottawa, Ontario, for processing and chemical analyses. Egg contents were homogenized and stored frozen (−40 °C) in acid-rinsed polyethylene vials for Hg analysis. The validity of this storage method has been previously discussed (Braune, 2007). 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 2014 were analyzed within six months of collection. Three to 15 eggs per species per year were analyzed (see Table S1 for sample sizes). As per standardized protocol at the time, egg homogenates from 1975 to 2005 were analyzed for total Hg and for $\delta^{15}N$ as pooled (composite) samples with each pool comprising equal aliquots of three individual eggs, with the exception of glaucous gull eggs sampled in 1993, which were analyzed as pools of two eggs each. Eggs sampled during 2006–2014 were individually analyzed.

2.2. Mercury analysis

Mercury analyses were carried out at NWRC. Samples collected during 1975–1998 were thawed, freeze-dried and digested in mineral acids prior to analysis. Those samples were then analyzed for total Hg 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 as described elsewhere (Neugebauer et al., 2000). Samples collected during 2003–2014 were homogenized, freeze-dried, homogenized again and weighed into nickel combustion boats. Those samples were then analyzed for total Hg by direct combustion of the solid sample in an oxygen-rich atmosphere (see Salvato and Pirola, 1996; EPA, 2007) using either an AMA-254 advanced mercury analyzer (Altec) equipped with an ASS-254 autosampler (2003–2013 samples), or a DMA-80 direct mercury analyzer (Milestone) with integrated autosampler (2014 samples). Moisture was determined by weight loss upon freeze-drying.

Analytical accuracy for total Hg was determined by analyzing one or two blank samples with each sample set, as well as standard reference materials (SRMs). For the 1975–1998 samples, SRMs DOLT-2 and DORM-1 or -2 obtained from the Canadian National Research Council (CNRC) were analyzed, and for the 2003–2014 samples, SRMs DORM-2, DOLT-2, -3 or -4, and TORT-2 or -3 from CNRC, as well as Oyster Tissue 1566b from the National Institute of Standards and Technology (NIST), were analyzed. Recoveries of SRMs were within the certified range of values for both methodologies. For the 1975–98 samples analyzed by CVAAS, average recoveries for SRMs ranged from 92.2% to 103.4% ($n = 8$), and for the 2003–14 samples analyzed by direct combustion using an advanced or direct mercury analyzer, average recoveries for SRMs ranged from 93.7% to 106.8% ($n = 225$) (Table S2). Total Hg concentrations for sample duplicates ($n = 24$) using the two methodologies were not significantly different ($p > 0.05$). Analytical precision was checked by analyzing replicate samples, averaging one replicate sample for every seven samples analyzed. Relative standard deviations for replicate readings ($n = 68$) averaged 2.8% (range 0–13%). The practical detection limits were 0.2 μg g^{-1} dry weight sample for the 1975–2003 samples, 0.08 dry weight sample