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Does temporal variation of mercury levels in Arctic seabirds reflect changes in global environmental contamination, or a modification of Arctic marine food web functioning?☆

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ABSTRACT

Studying long-term trends of contaminants in Arctic biota is essential to better understand impacts of anthropogenic activities and climate change on the exposure of sensitive species and marine ecosystems. We concurrently measured temporal changes (2006–2014) in mercury (Hg) contamination of little auks (*Alle alle*; the most abundant Arctic seabird) and in their major zooplankton prey species (*Calanoid* copepods, *Themisto libellula, Gammarus* spp.). We found an increasing contamination of the food-chain in East Greenland during summer over the last decade. More specifically, bird contamination (determined by body feather analyses) has increased at a rate of 3.4% per year. Conversely, bird exposure to Hg during winter in the northwest Atlantic (determined by head feather analyses) decreased over the study period (at a rate of 1.5% per year), although winter concentrations remained consistently higher than during summer. By combining mercury levels measured in birds and zooplankton to isotopic analyses, our results demonstrate that inter-annual variations of Hg levels in little auks reflect changes in food-chain contamination, rather than a reorganization of the food web and a modification of seabird trophic ecology. They therefore underline the value of little auks, and Arctic seabirds in general, as bio-indicators of long-term changes in environmental contamination.

1. Introduction

Current and projected changes of the Arctic cryosphere, combined with extensive human industries, are modifying mercury (Hg) levels present in the Arctic environment (e.g. Macdonald et al., 2005; Rydberg et al., 2010; Fisher et al., 2013). Once deposited, this Hg is methylated by bacterial activity, becomes readily bioavailable to living organisms, and therefore enters and biomagnifies through food webs, thereby potentially affecting Arctic biodiversity and ecosystems (Dietz et al., 2013). In this context, studying long-term trends of contaminants in Arctic biota, particularly of Hg, has been declared a research priority by the Arctic Council, to better understand impacts of anthropogenic activities and climate change on the exposure of Arctic species and humans to pollutants (AMAP, 2011, 2012). Such long-term monitoring programs also provide essential information about the effectiveness of strategies set-up to mitigate emissions of Hg and thereby exposure of arctic systems. However, the Arctic is a remote region characterized by extreme climatic conditions, an extensive sea-ice cover, and is therefore hardly accessible a large part of the year. Detailed and long-term at-sea investigations are thus extremely challenging and costly. As a consequence, most long-term monitoring programs focus on large marine top-predators which spend a part of their life cycle on land, where they become easier to observe and to sample (e.g. seabirds and polar bears; Dietz et al., 2006; Braune, 2007; Mallory and Braune, 2012) or which can be sampled through hunted or stranded individuals (e.g. whales and other marine mammals; Braune et al., 2005; Gaden et al., 2009). In contrast, Hg contamination studies of lower trophic levels such as invertebrates or fish

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are extremely difficult and therefore particularly limited (Rigét et al., 2011; Foster et al., 2012; Ruus et al., 2015). In this context, it is essential to identify marine predators that reflect the contamination of lower trophic levels, and can therefore be used as bio-indicators of long-term changes in global environmental contamination.

Several studies have previously suggested the use of seabirds as bio-indicators to monitor short-term environmental contamination in various regions of the world, including the Arctic (Furness and Camphuysen, 1997; Goodale et al., 2008; Verreault et al., 2010). However, their use as bio-indicators in longer term studies has been limited by the difficulty to understand the drivers of measured trends, as long-term changes of contaminant level in Arctic seabirds might reflect a general change in food web contamination, or might rather reflect a modification of the food web structure impacting bird trophic ecology. Indeed, some contaminants, as Hg, biomagnify (i.e. increase in concentration along the food chain). Hence, a change of feeding preferences of a predator across time, following a modification of food availability, may modify its exposure to contaminants, even if the contamination of the food web itself remains unchanged (Cabana and Rasmussen, 1994; McKinney et al., 2009). Discriminating, and evaluating the role of underlying ecological drivers in observed contamination trends is therefore essential, to clarify whether seabirds do indeed function as bio-indicators of long-term changes in environmental contamination. Recently, a few studies confirmed that observed Hg trends in Arctic seabirds reflect changes in environmental contamination, although bird trophic status might affect these trends according to the species considered (Burgess et al., 2013; Braune et al., 2014; Bond et al., 2015; see Braune et al., 2015 for other contaminants).

In the present study, we propose to go further, by concurrently measuring temporal (2006–2014) changes in the Hg contamination of both little auks (*Alle alle*) – the most abundant Arctic seabird – and of their major zooplankton prey species in East Greenland. Changes in bird isotopic niche (trophic status and feeding habitat) are also assessed. The objectives are (1) to describe temporal trends in little auk exposure to Hg during both their breeding and non-breeding periods over the last eight years, (2) to investigate for the first time trends of Hg contamination in several Arctic zooplankton species, and (3) to determine if little auks can be used as bio-indicators of long-term changes in environmental contamination by Hg.

2. Material and methods

2.1. Sample collection

Every summer (July–August) between 2007 and 2014, adult little auks breeding at Kap Höegh (East Greenland; 70°44′N, 21°35′W) were captured by hand at the nest (little auks nest in crevices under boulders). From each bird, two batches of feathers were plucked: one from the back (hereafter called “body feathers”) and one from the throat (hereafter called “head feathers”). Feathers were kept at ambient temperature in sealed plastic bags until they were processed for Hg analysis. A small blood sample (~0.3 mL) was also collected from each individual (from the brachial vein), stored in 70% ethanol, and kept frozen at −20 °C pending stable isotope analysis. This blood preservation method was shown to have no significant effect on isotope results (Hobson et al., 1997). Birds were released into their nest within five minutes of handling. Each bird was captured and sampled only once during the entire study period. Sample sizes for each sampling year are provided in Table 1.

Concurrently, additional birds were captured each year to collect prey samples (2007–2013). Adult little auks forage at sea and bring fresh food back to their offspring in a sublingual pouch. Birds with a full pouch were caught on rocks using noose carpets. Food loads were gently scooped out of the sublingual pouch and the bird released within five minutes of capture. Prey samples were preserved in 70% ethanol, and kept frozen at −20 °C pending Hg analysis. This preservation method is believed to have no effect on measured Hg concentrations (Chouvelon et al. unpublished). Little auks primarily feed on large calanoid copepods (Harding et al., 2008). When available, they also collect other species, such as the amphipods *Themisto libellula*, *Gammarus* spp. and *Apherusa glacialis* (Harding et al., 2008; Fort et al., 2010).

2.2. Sample preparation

Prior to the analyses, feather samples were cleaned to remove dirt and chemical external contamination. Feathers were plunged into a 2:1 chloroform:methanol solution in an ultrasonic bath for two minutes, rinsed twice in a methanol solution and dried for 48 h at 50 °C. Blood samples were dried for 72 h at ambient temperature to remove ethanol, lyophilized for 48 h and ground to powder. Prey items were identified from food loads at the species or copepodite stage (for copepods only) levels. Because zooplankters are too small to be analyzed for contaminant concentrations at the individual level, they were then pooled by year and by species (*T. libellula*, *Gammarus* spp., *Apherusa glacialis*), except for the three calanoid copepods *Calanus hyperboreus*, *Calanus glacialis* and *Calanus finmarchicus* that were pooled together by copepodite stages. Each group was then dried for 72 h at ambient temperature to remove ethanol, lyophilized for 48 h and ground to powder for homogenization.

2.3. Sample analysis

Total Hg (hereafter termed Hg) concentrations were measured in head and body feathers and in zooplankton samples using an advanced Hg analyzer spectrophotometer (Altec AMA 254) as described in Bustamante et al. (2006). Each Hg analysis was performed on one complete feather or on ~1 mg of zooplankton. Analyses were repeated two or three times for each sample until the relative standard deviation for the aliquots was <10%; samples not meeting this criterion were excluded from the analysis. The mean Hg concentrations for those two measurements were then considered for statistical analyses. To ensure the accuracy of measurements, a certified reference material (CRM) was used [Lobster Hepatopancreas Tort-2; NRC, Canada; Hg concentration of 0.27 ± 0.06 μg/g of dry weight (dw)]. The CRM was measured every 10 samples and the average measured value was 0.26 ± 0.01 μg/g dw (n = 113). Additionally, blanks were run at the beginning of each sample set. The detection limit of the method was 0.005 μg/g dw.

Feather growth constitutes a major excretion route in seabirds during which >70% of the body burden of Hg is eliminated to growing feathers (Honda et al., 1986, Braune and Gaskin, 1987; Agusa et al., 2005). Hg concentrations measured in feathers is therefore believed to reflect the amount of Hg that has accumulated in body tissues since the last moult sequence (Furness et al., 1986). Little auks have two moult sequences per year: one partial pre-breeding moult of head feathers in April, and one complete post-breeding moult in September (Gaston and Jones, 1998; Mosbech et al., 2012). Therefore, Hg concentrations measured in little auk head feathers reflect the amount of Hg that has accumulated during the last non breeding season (same year as sample collection) spent in the northwest Atlantic, mainly off Newfoundland (Fort et al., 2013, 2014). Hg concentrations measured in little auk body feathers reflect the amount of Hg that has accumulated during the previous breeding season spent in East Greenland (year preceding