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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

Table 1
Hg concentrations and isotopic ratios measured in little auks and zooplankton species samples at Kap Höegh (East Greenland; 70°44′N, 21°35′W) between 2007 and 2014. Since Hg concentrations measured in body feathers reflect bird contamination during the previous breeding season (i.e. previous year), values for body feathers collected yearly from 2007 to 2014 reflect contamination for the period 2006–2013.

Year	Hg concentrations (μg/g of dw)							δ^{15}N (‰)	δ^{13}C (‰)
	Little auk body feather	Little auk head feather	Copepods CIV*	Copepods CV**	*Themisto libellula*	*Gammarus* spp.	*Apherusa glacialis*	Little auk blood	Little auk blood
2006	1.00 ± 0.22 (20)	–	–	–	–	–	–	–	–
2007	1.13 ± 0.24 (19)	3.73 ± 1.33 (20)	0.045	0.048	0.033	0.057	0.073	11.85 ± 0.11 (15)	−22.31 ± 0.08 (15)
2008	1.04 ± 0.26 (19)	2.86 ± 0.71 (20)	–	–	–	–	–	11.11 ± 0.12 (15)	−22.33 ± 0.14 (15)
2009	1.37 ± 0.70 (45)	3.06 ± 0.99 (20)	0.069	0.032	0.036	0.046	0.056	11.57 ± 0.16 (15)	−22.26 ± 0.19 (15)
2010	1.70 ± 0.66 (17)	3.17 ± 0.82 (40)	0.045	0.048	0.037	–	–	11.17 ± 0.16 (15)	−22.14 ± 0.14 (15)
2011	1.25 ± 0.51 (20)	3.21 ± 0.99 (25)	0.048	0.046	0.037	0.051	0.081	11.43 ± 0.15 (15)	−22.07 ± 0.10 (15)
2012	1.69 ± 0.80 (20)	2.27 ± 0.41 (20)	0.042	0.052	0.052	0.052	0.060	11.61 ± 0.29 (15)	−22.58 ± 0.17 (15)
2013	2.11 ± 1.49 (20)	2.60 ± 0.56 (19)	0.058	0.062	0.042	0.060	–	12.44 ± 0.26 (15)	−22.10 ± 0.18 (15)
2014	–	3.02 ± 1.14 (16)	–	–	–	–	–	12.01 ± 0.19 (15)	−22.80 ± 0.08 (15)

* Includes *Calanus hyperboreus*, *C. glacialis* and *C. finmarchicus* copepodite stage CIV.
** Includes *Calanus hyperboreus*, *C. glacialis* and *C. finmarchicus* copepodite stage CV.

sample collection). Finally, Hg concentrations measured in zooplankton samples reflect their contamination at the time of their collection (summer) off the little auk breeding site in East Greenland.

Stable isotope analyses were performed on ~0.5 mg subsamples of homogenized, non-lipid extracted whole blood loaded into tin cups, and using an elemental analyzer (Flash EA 1112, Thermo Fisher) coupled in continuous flow mode to an isotope ratio mass spectrometer (Delta V Advantage, Thermo Fisher, Bremen, Germany). Stable isotope abundances were expressed in δ notation as the deviation from standards in parts per thousand (‰), according to the equation: $\delta X = [(R_{sample}/R_{standard}) - 1] \times 10^3$, where X is ^{13}C or ^{15}N and R is the corresponding ratio $^{13}C/^{12}C$ or $^{15}N/^{14}N$. Standard values were Vienna Pee Dee Belemnite (VPDB) for C and atmospheric N_2 (air) for N. Replicate measurements of internal laboratory standards (acetanilide) indicated that the measurement error was <0.2% for both $\delta^{15}N$ and $\delta^{13}C$ values. Obtained $\delta^{15}N$ isotopic values reflect the relative trophic position of birds and are considered as an indicator of their diet (Kelly, 2000). $\delta^{13}C$ values mainly reflect the carbon source and are considered as an indicator of their feeding habitat (Kelly, 2000).

Hg and isotopic analyses were performed at the Littoral Environnement et Sociétés laboratory (LIENSs, La Rochelle, France).

2.4. Statistical analyses

Analyses on body feathers were preferred to blood samples to investigate Hg temporal trends during the breeding season. Indeed, as mentioned above, little auk body feathers integrate Hg accumulated between the pre-breeding and the post-breeding moults and therefore reflect bird Hg contamination during the whole breeding season (Fort et al., 2014), assumed to be comparable between years. Conversely, blood reflects bird short-term contamination at the time of sampling and can therefore fluctuate along the breeding season (Bustamante et al. *unpublished data*). Since the timing of blood collection varied between years, from late incubation to mid-chick rearing (representing a period of about three weeks), a direct comparison of Hg levels measured in blood between years was not carried out to avoid uncertainties.

Stable isotope analyses were performed on blood samples as it is the only collected tissue that reflects bird trophic status during the breeding season. Stable isotopes in feathers indeed reflect the trophic status at the time of their synthesis, i.e. during the pre-breeding or post-breeding moults (Hobson, 1993). Stable isotope half-lives in the blood of little auks were estimated to be 12–15 days (Fort et al., 2010). We therefore assume that isotopic values measured during the breeding season in different years were comparable.

Individual little auks sampled in this study were not sexed. However, previous investigations demonstrated that there was no difference between males and females when considering Hg concentrations or stable isotope ($\delta^{15}N$ and $\delta^{13}C$) signatures measured in body and head feathers (Fort et al., 2010, 2014). Results from all birds were therefore pooled in the statistic analyses.

Statistics were computed using R version 3.0.2 (R Development Core Team 2011). Hg data measured in little auks were log-transformed to comply with parametric assumptions of normality and homoscedasticity. Because *Apherusa glacialis* is consumed by little auks during years when there is sea-ice close to their breeding site (*Apherusa glacialis* is a sea-ice associated amphipod), samples for this species were available for four years only, and were therefore excluded from the analyses. Hg concentrations for *Apherusa glacialis* are nonetheless presented in Table 1. Linear regressions were used on log-transformed Hg data to determine temporal trends in little auk contamination during both their breeding and non-breeding seasons over the 2006–2014 time period. Because the normality of isotopic data was not achieved, we used Kruskal–Wallis χ^2 tests to examine variations of isotopic ratios ($\delta^{15}N$ and $\delta^{13}C$) among years. We used general linearized models (GLMs) to investigate the role of bird trophic status ($\delta^{15}N$), foraging habitat ($\delta^{13}C$) and of prey Hg contamination driving little auk inter-annual Hg contamination changes during the breeding season in East Greenland. To this end, Hg concentrations measured each year in *T. libellula*, *Gammarus* spp., calanoid copepods stage CIV and calanoid copepods stage CV were first weighted by prey proportions in bird diet per year (Amélineau et al. *unpublished*), and then pooled to obtain an averaged zooplankton contamination value. Models were compared using Akaike's Information Criterion corrected for small sample sizes (AICc). Among models with the lowest AICc, the one showing the highest adjusted-R^2 value was selected as the best model.

3. Results

3.1. Temporal trends

Based on log-transformed Hg concentrations, exposure of little auks to Hg during their breeding season – as reflected by body feather analyses – showed a significant increase between 2006 and 2013 ($R^2 = 0.14$, $p < 0.001$, Fig. 1A) at a rate of 3.4% per year (slope = 0.0338, SE = 0.0060). Mean Hg concentrations ranged from 1.00 μg/g dw in 2007 to 2.11 μg/g dw in 2013 (Table 1).

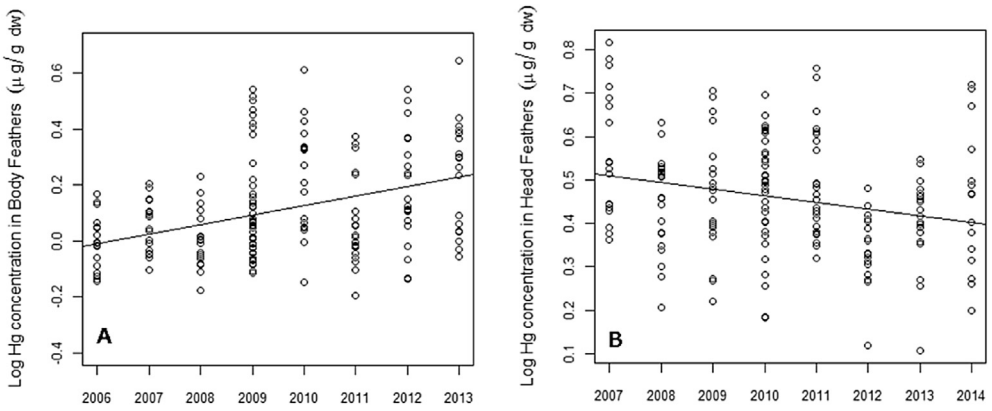

Fig. 1. Changes in little auk Hg contamination (in µg/g dw) between 2006 and 2014, during (A) the breeding season – reflected by concentrations in body feathers and (B) the non-breeding season – reflected by concentrations in head feathers (see Methods for details). Since Hg concentrations measured in body feathers reflect bird contamination during the previous breeding season (i.e. previous year), values for body feathers collected yearly from 2007 to 2014 reflect contamination for the period 2006–2013.

Conversely, bird exposure during the non-breeding season – as reflected by head feather analyses – showed a significant decrease between 2007 and 2014 ($R^2 = 0.05$, $p = 0.001$, Fig. 1B), at a rate of 1.5% per year (slope = −0.0153, SE = 0.0046). Mean Hg concentrations ranged from 3.73 µg/g dw in 2007 to 3.02 µg/g dw in 2014 with a minimum of 2.27 µg/g dw reached in 2012 (Table 1).

The contamination of zooplankton species during summer in East Greenland showed a general increase between 2007 and 2013 (Fig. 2). More specifically, Hg concentrations in *T. libellula* showed a slight, but continuous increase from 2007 to 2013, ranging from 0.036 to 0.052 µg/g dw. After a decrease between 2007 and 2009, Hg concentrations in *Gammarus* spp. and calanoid copepods (stage CV) increased to reach a maximum in 2013 (concentrations ranging from 0.046 (2009) to 0.060 (2013) µg/g dw in *Gammarus* spp. and from 0.032 (2009) to 0.062 (2013) µg/g dw in copepods). Hg concentrations in calanoid copepods (stage CIV) showed no clear pattern between 2007 and 2013.

Little auks breeding in East Greenland showed significant variation in $\delta^{15}N$ isotopic values during the 2007–2014 period (Kruskal–Wallis, $\chi^2 = 99.6$, $p < 0.001$), with an increasing trend between 2008 and 2014 ($R^2 = 0.53$, $p < 0.001$; Fig. 3). $\delta^{13}C$ values also showed significant variations between years ($\chi^2 = 81.0$, $p < 0.001$), but no trend emerged during the study period (Fig. 3).

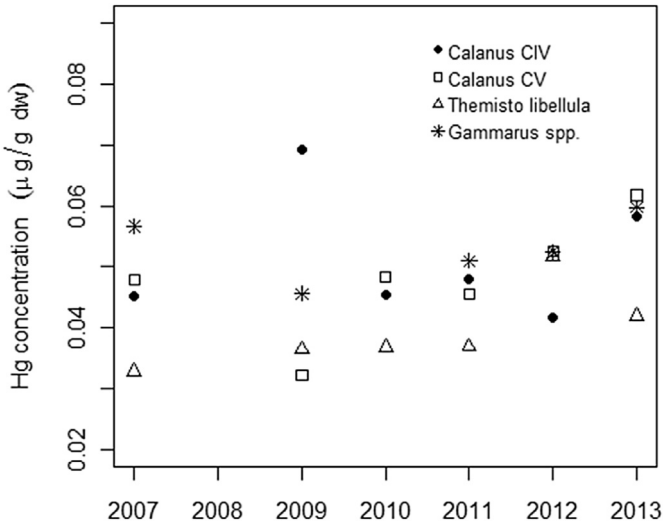

Fig. 2. Annual change of Hg concentration (in µg/g dw) in 4 groups of zooplankton consumed by little auks in East Greenland. Calanus CIV and CV groups correspond to *Calanus hyperboreus, C. glacialis* and *C. finmarchicus* copepodite stages CIV and CV, respectively.

3.2. Drivers of little auk contamination change during the breeding season

The best model contains Hg concentration in zooplankton only as explanatory variable, and explains 72% of the measured inter-annual variability in little auk Hg contamination during the breeding season (Table 2). $\delta^{15}N$ and $\delta^{13}C$ values were not retained during model selection process.

4. Discussion

Defining long-term changes in organism exposure to Hg in the Arctic is essential, and recently became a major research field (Gaden and Stern, 2010; Dietz et al., 2011; Braune et al., 2014; Bond et al., 2015). However, while most of these investigations focused on large marine top-predators, studies on lower trophic levels have remained limited (Rigét et al., 2011). In this study, we measured temporal (2006–2014) changes in the Hg contamination of both little auks and their major zooplankton prey species (*Calanoid* copepods, *T. libellula, Gammarus* spp.) in East Greenland. Although we are aware that our time-series are limited to a decade and should now be extended to confirm obtained trends, they nonetheless provide essential information to better understand impacts of a rapidly changing environment for the conservation of vulnerable Arctic marine ecosystems. Our results also demonstrate that inter-annual variations of Hg levels in little auks reflect changes in the food-chain contamination rather than a reorganization of the food web, and therefore underline the value of little auks as bio-indicators of long-term changes in environmental contamination.

Specifically, our results demonstrate that during summer (July–August) in East Greenland, exposure to Hg of little auks and zooplankton species have generally increased over the study period (Figs. 1A and 2). Seabird contamination to Hg shows contrasting trends according to their breeding site in the Arctic, with an absence of temporal trend observed in northern Norway (Helgason et al., 2008) and increasing trends found in Canada (e.g. Braune, 2007; Bond et al., 2015). There was, however and to the best of our knowledge, no previous long-term investigation performed on Greenlandic seabirds. With an increasing contamination of 3.4% per year found in little auks (see Fig. 1A), our results confirm the enhanced vulnerability of this species and of the seabird

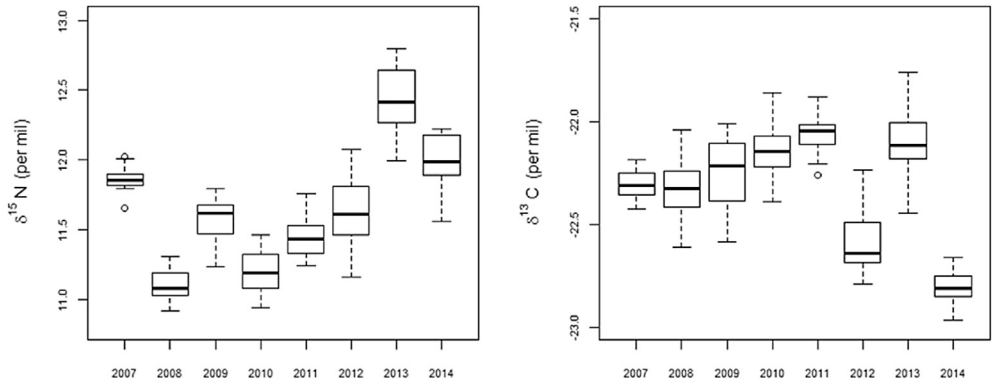

Fig. 3. $\delta^{15}N$ and $\delta^{13}C$ isotope ratios (in per mil) measured in little auk blood samples during the 2007–2014 study period, reflecting birds' trophic status and foraging habitat, respectively, during the breeding season.

Table 2
Model selection for the general linearized models (GLMs) used to define drivers of inter-annual changes in little auk Hg contamination during the breeding season. The selected model (showing the lowest AICc value and the highest R^2 value within comparable AICc) is in bold. $Hg_{zooplankton}$ corresponds to the averaged Hg contamination of the 4 zooplankton species/categories consumed by little auks (*Themisto libellula*, *Gammarus* spp., calanoid copepods stage CIV and calanoid copepods stage CV) weighted by prey proportions in bird diet. Calanoid copepods correspond to *Calanus hyperboreus, C. glacialis* and *C. finmarchicus*. $\delta^{15}N$ and $\delta^{13}C$ correspond to isotopic values measured in little auk blood samples.

Model	k	AIC_c	ΔAIC_c	R^2adj	w_i
$Hg_{zooplankton}$	**3**	**12.91**	**0.00**	**0.72**	**0.46**
NULL	2	13.03	0.12	0.00	0.43
$\delta^{15}N$	3	16.38	3.47	0.21	0.08
$\delta^{13}C$	3	19.15	6.24	−0.17	0.02
$\delta^{15}N + \delta^{13}C$	4	30.33	17.42	0.02	0.00
$\delta^{15}N + Hg_{zooplankton}$	4	42.55	29.64	0.64	0.00
$\delta^{13}C + Hg_{zooplankton}$	4	42.67	29.77	0.64	0.00

community to contaminants in Greenland, as it is in Canada. Recent studies indeed showed that Hg can significantly impact reproductive seabirds by affecting bird investment in reproduction (e.g. Tartu et al., 2013), breeding success (Goutte et al., 2014b), hatching and fledging success (Wolfe et al., 1998; Goutte et al., 2014a). An increase in bird exposure during the breeding season therefore likely has long-term effects on populations (Goutte et al., 2014a).

In contrast to our results, a recent study showed a decreasing trend of Hg contamination in polar bears from East Greenland (Dietz et al., 2006). Although the two time series performed on little auks (2006–2014, this study) and polar bears (1973–2001, Dietz et al., 2006) do not overlap, such contrasting observed trends should be investigated. For instance, long-term changes in polar bear foraging habitat (more pelagic/open-water) and diet (less high trophic position ringed seals) associated to a decrease of the sea-ice extent in East Greenland, might explain contrasting trends between the two species (McKinney et al., 2013). Moreover, polar bears were shown to be less exposed to Hg than other top-predators considering their trophic position (Atwell et al., 1998), potentially affecting biomagnification processes.

Over the study period and during the breeding season, little auks also showed a change in their trophic status as reflected by $\delta^{15}N$ isotopic values. This change is rather small as the difference between lowest (2008) and highest (2013) values is 1.33‰, reflecting a dietary change of <0.5 trophic level (Hobson and Welch, 1992). We speculate that this change might be linked to a decreased availability of the ice-associated amphipod *Apherusa glacialis* following the recent decrease of sea-ice cover in the vincinity of the little auk colony (Amélineau et al. *submitted*). Among prey consumed by little auks, *Apherusa glacialis* indeed has the lowest nitrogen isotopic signature (Tamelander et al., 2006).

Contrary to the breeding season, non-breeding little auks showed a decreasing exposure to Hg over the last decade. Little auks breeding in East Greenland were found to mainly spend the non-breeding season in the northwest Atlantic, off Newfoundland (Fort et al., 2012, 2013, 2014). There is no other published trend available for this offshore region, and further investigations should be performed to define if decreasing emissions of Hg to air from North America and Europe over the last decades (UNEP, 2013) have contributed to this decreasing exposure of marine organisms through lower deposition, and if so, why Hg concentrations are still increasing in East Greenland. In this context, a recent study suggested that the non-breeding season could be a major source of contamination for little auks breeding in East Greenland, potentially affecting their subsequent reproductive period (Fort et al., 2014). Our temporal investigation validates these former results; with Hg concentrations measured in head feathers (non-breeding contamination) being consistently higher (1.4–3.7 times higher) than concentrations measured in body feathers (breeding contamination), for samples collected the same year (Table 1). This confirms that studies aiming at determining ecotoxicological impacts of Hg and other contaminants on migratory Arctic wildlife should consider contamination of organisms over their entire annual cycle, as well as their spatial movements and migration chronology. However, because of the opposite trends observed during the breeding and the non-breeding periods (Fig. 1), the role played by the former season on little auk contamination might increase and ultimately become prominent. Indeed, and as illustrated in Fig. 4, the difference of Hg concentrations between head and body feathers continuously decreased over the study period.

Concurrently to bird measurements, our study documents the first time-series of Hg concentrations measured in different Arctic zooplankton species, and highlights a general increase in their contamination over the last decade. The three studied Arctic calanoid copepod species are herbivorous (Falk-Petersen et al., 2009) and considered to represent the second trophic level of Arctic marine food-webs (Hobson et al., 1995, 2002). This is therefore unlikely that temporal changes of Hg levels measured in copepods result themselves from modifications of their diet but rather from changes in environmental exposure. East Greenland Hg concentrations were in the range of values measured in zooplankton from the Canadian Arctic (Chételat and Braune, 2012; Foster et al., 2012), but were about 10 times higher than for zooplankton from Spitsbergen (Ruus et al., 2015). Calanoid copepods (*C. hyperboreus, C. glacialis* and to a lesser extent *C. finmarchicus*) constitute the most important biomass species in Arctic marine food webs, and

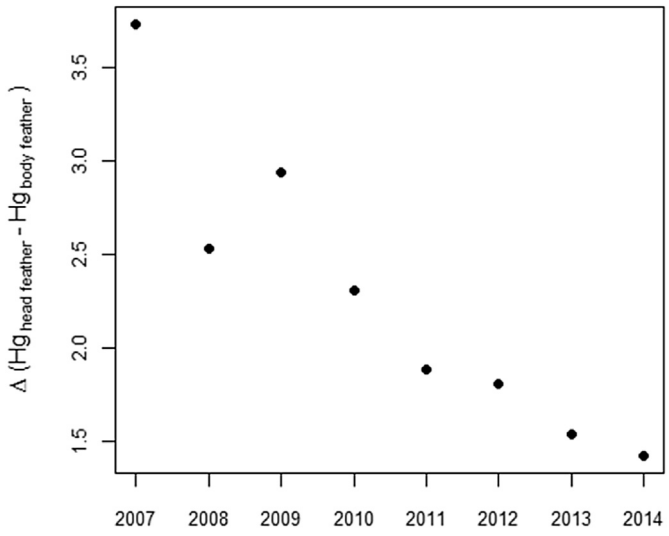

Fig. 4. Difference of Hg concentrations measured in head feathers (contamination during the non-breeding period) and body feathers (contamination during the breeding season) collected the same year.

therefore play a key role in the energy flux within these ecosystems, both as consumers of phytoplankton and as an essential source of food for large stocks of predators, from carnivorous zooplankton to fish, seabirds and marine mammals (Lynch et al., 2001; Falk-Petersen et al., 2004; Hop et al., 2006; Karnovsky et al., 2008). Consequently, an increased contamination of these species might impact, through biomagnification processes, Hg contamination and thereby the health of the entire marine food webs up to top-predators. This is illustrated, for instance, by the positive relationship which we found between increases of Hg levels in zooplankton and in little auks.

In this context, it appears essential to develop long-term programs throughout the Arctic, aiming at monitoring changes of environmental contamination. These should favor the use of selected bio-indicators, easy to sample and reflecting specific compartments of marine Arctic ecosystems. In this study, we showed that little auks, which occupy the top level of a short food chain (trophic position comprised between 3.2 and 3.4 according to regions; Hobson et al., 2002, Jæger et al., 2009, Linnebjerg et al. *unpublished data*), constitute an appropriate bio-indicator of temporal trends in environmental contamination within Arctic pelagic systems. Indeed, we found that variations of breeding little auks Hg contamination between 2006 and 2014 were primarily explained by changes of Hg concentrations in their main prey (zooplankton). These results confirm previous studies which demonstrated that Arctic seabirds reflect local environmental Hg contamination, both at a specific time and over long-term periods, and are therefore good indicators of the state of contamination of marine food-webs (e.g. Verreault et al., 2010; Bond et al., 2015). In addition, the little auk has important features. It is the most abundant seabird in the Arctic, has a wide breeding distribution ranging from West Greenland to the Russian Arctic (Gaston and Jones, 1998), and different populations overwintering in contrasting areas from the Northwest Atlantic to the Kara Sea (Fort et al., 2013; *unpublished data*), allowing the study of various Arctic regions. Moreover, thanks to a specific moulting sequence (two distinct moults per year in approx. April and September; Gaston and Jones, 1998, Mosbech et al., 2012), little auk feather samples collected during summer integrate information about bird and thereby environmental contamination for two distinct periods: summer (breeding) and winter (non-breeding). We therefore recommend the use of little auks as a key bio-indicator species to track spatio-temporal changes in exposure of Arctic marine food-webs to Hg.

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