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Spatial Ecotoxicology: Migratory Arctic Seabirds Are Exposed to Mercury Contamination While Overwintering in the Northwest Atlantic

Jérôme Fort,*[,†] Gregory J. Robertson,[‡] David Grémillet,[§,∥] Gwendoline Traisnel,[†] and Paco Bustamante[†]

[†]Littoral Environnement et Sociétés, UMR 7266 CNRS-Université La Rochelle, 2 rue Olympe de Gouges, 17000 La Rochelle, France

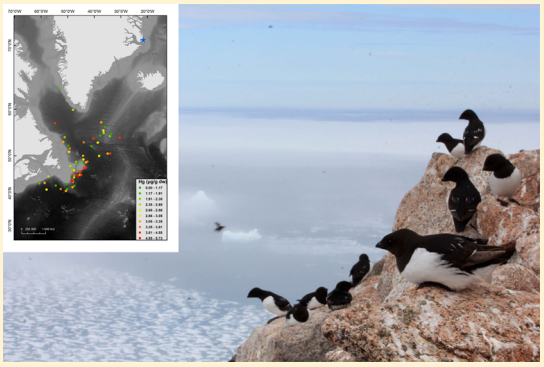
[‡]Wildlife Research Division, Environment Canada, 6 Bruce Street, Mount Pearl, NL, Canada

[§]Centre d'Ecologie Fonctionnelle et Evolutive, UMR 5175 du CNRS, 1919 route de Mende, 34293 Montpellier, France

[∥]Percy FitzPatrick Institute, DST/NRF Centre of Excellence, University of Cape Town, Rondebosch 7701, South Africa

Supporting Information

ABSTRACT: Arctic organisms are exposed to various levels of pollutants, among which mercury (Hg) has raised important environmental concerns. Previous studies examining Hg levels, trends, and effects on Arctic marine top predators have focused on the Arctic region. However, many of these top predators, such as seabirds, migrate to spend a large part of their life cycle far from the Arctic in areas where their exposure to contaminants is largely unknown. By combining biotelemetry and Hg and stable isotope analyses, we studied the seasonal Hg contamination of little auks (*Alle alle*, the most abundant Arctic seabird) in relation to their distribution and marine foraging habitat, as well as its potential impacts on bird reproduction. We show that little auks were ∼3.5 times more contaminated when outside the breeding season, and that Hg that accumulated during this nonbreeding non-Arctic period was related to egg size the following season, with females having more Hg laying smaller eggs. Our results highlight that ecotoxicological studies should be expanded to yield a comprehensive understanding of contamination risks and associated threats to top predators over their entire annual cycle. Furthermore, we show that an important nonbreeding area located in the northwest Atlantic was associated with greater Hg contamination and demonstrate the utility of bird-borne miniaturized technology for evaluating the contamination of marine systems on large spatial scales.



INTRODUCTION

Although it is far from major human industries, the Arctic region is threatened by pollution risks and pollutant levels. Transported over large distances by ocean currents and atmospheric circulation, many pollutants originating from industrialized or developing countries are deposited in the Arctic.[1,2] More recently, sea-ice cover has been greatly reduced in some areas, releasing pollutants trapped over decades and opening up new areas to human activities such as shipping and extractive industries, thereby increasing direct discharges of pollutants into Arctic ecosystems.[3,4] As a consequence of all these activities, concentrations of pollutants such as trace metals or hydrocarbons have increased in some parts of the Arctic over the past several decades.[5−7] Once in the environment, pollutants can become bioavailable, enter the food chain, and can have major impacts on organisms and biodiversity. Among pollutants that are liable to affect Arctic wildlife, mercury (Hg) has raised important environmental concerns and drawn extensive attention.[8] This nonessential metal is highly toxic, particularly in its main organic form of methylmercury, and even at low concentrations has been shown to be a powerful neurotoxicant.[9,10] In this context, defining concentrations, trends, and ecotoxicological effects of Hg on Arctic organisms is important in developing strategies for the conservation of vulnerable species and ecosystems. Recently, the Arctic Monitoring and Assessment Programme (AMAP) working group of the Arctic Council published an extensive review of the current knowledge of Hg contamination of Arctic species and the deleterious effects of Hg.[8] This group emphasized the importance of focusing on marine top predators because they are expected to be exposed to high concentrations of Hg through bioaccumulation and biomagnification.[11]

Marine top predators, such as seabirds, can be highly mobile during their annual cycle. For instance, many seabird species leave the Arctic after their breeding season and migrate hundreds to thousands kilometres to winter areas, spending many months of the year outside of the Arctic, in boreal, temperate, or even tropical regions.[12−14] There, they could face

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equally contaminated environments or environments even more contaminated than the Arctic,[15] which could lead to the accumulation of high concentrations of Hg in the seabirds that could in turn have impacts on their physiology and behavior and ultimately reproduction, survival, and population dynamics.[16,17] Understanding exposure to pollutants, including Hg, during the season spent outside the Arctic is therefore essential for obtaining a complete view of the risk faced by these organisms in a changing environment. Nevertheless, and to the best of our knowledge, only one recent study, focused on persistent organic pollutants (POP), previously linked Arctic seabird contamination to their non-Arctic winter areas, highlighting the importance of considering the entire annual cycle in ecotoxicology studies.[18] This focus on the annual cycle has yet to be considered for Hg in Arctic seabirds. In this study, we examined and compared Hg concentrations measured in little auks (*Alle alle*), a small Arctic seabird, during different periods of the year and in relation to their winter distribution. We also investigated how Hg that accumulated during the nonbreeding period could affect their reproduction.

The little auk represents an ideal species for investigating the Hg contamination of Arctic seabirds during the breeding and nonbreeding periods. This species, like most of the alcids, moults twice per year; a complete feather moult soon after the breeding season (September) involves the replacement of the entire body plumage, while a partial prebreeding moult occurring during spring involves the replacement of throat and head feathers.[19−21] The timing of this prebreeding moult is not accurately known. However, little auks collected in March at their wintering site were all in winter plumage,[22] while birds seen at breeding colonies in May are in breeding plumage. It is therefore likely that little auks moult their head and throat feathers in April, before arriving at their colony. The feather moult, along with egg production, is a mechanism by which seabirds excrete Hg that has accumulated in their body tissues,[23] which makes concentrations measured in feathers an indicator of levels of Hg that have accumulated since the last moulting sequence.[24−26] Hence, it can be assumed that Hg concentrations measured in little auk head feathers reflect levels accumulated during the nonbreeding period while cover feathers from other parts of the body reflect levels accumulated during the breeding season.

By analyzing Hg concentrations in blood and feathers and combining these results with data about bird movement and wintering areas, obtained with biotelemetry tracking and stable isotope analysis, we tested the following hypotheses. (1) Levels of little auk Hg contamination are higher during the nonbreeding season spent outside of the Arctic. (2) Contamination of birds is related to their nonbreeding distribution and foraging habitat. (3) Levels of Hg that accumulated during the non-Arctic nonbreeding period affect the reproduction of little auks.

MATERIALS AND METHODS

Study Sites and Sample Collection. In July 2009 and 2010, a total of 135 adult little auks breeding at Kap Höegh (East Greenland; 70°44′N, 21°35′W) were equipped with a miniature geolocator data-logger (GLS; Mk14 and Mk18L, British Antarctic Survey, mass of 1.5 g, ∼1% of adult body mass) mounted on a conventional metal leg ring to track their nonbreeding movements and distribution.[27,28] The following years, birds were recaptured [in 2010, n = 47 (22 males, 24 females, and one unknown sex); in 2011, n = 35 (22 males and 13 females)], the data loggers retrieved and downloaded, and blood and feather samples collected. Blood samples (∼0.3 mL) were collected from the brachial vein, stored in 70% ethanol, and kept frozen at −20 °C. Two batches of feathers were plucked from each bird: one from the body (back or belly) and one from the head (cheek, neck, or throat). These two batches are hereafter called "body feathers" and "head feathers", respectively. All feathers were kept at ambient temperature in sealed plastic bags until they were analyzed. A few additional feathers were collected and stored at −20 °C for subsequent molecular sexing.

Little auks nest in crevices in talus slopes and lay one egg per breeding season. When accessible, eggs of tracked birds were measured [maximal length (L) and breadth (B)] and their volumes (V) calculated using the equation $V = \pi/6 \times LB^2/1000$.[29] Nests were then checked every 2 days until the eggs had hatched to determine for each nest the hatching date. Evidence of hatching was provided by the direct sight of a chick or by the presence of egg shell fragments in the nest chamber.

Little auks were also collected in Placentia Bay (Newfoundland; 47°30′N, 54°00′W) in March 2011 and immediately frozen at −20 °C until they were dissected.[22] Newfoundland waters are known to be the main wintering quarter for little auks breeding in Greenland, including those from East Greenland.[27,28] From these carcasses, blood samples were collected (from the cardiac clot) and kept frozen at −20 °C until they were analyzed.

Hg and Stable Isotope Analyses. Prior to analyses, feathers were cleaned to remove any external contamination. Feathers were rinsed once in a 2:1 chloroform/methanol solution and twice in a methanol solution. Feathers were then dried for 48 h at 50 °C. Blood samples were dried for 72 h at ambient temperature to remove ethanol and then lyophilized for 48 h.

Total Hg (hereafter termed Hg) concentrations were measured in whole blood and feather samples collected from little auks breeding at Kap Hoëgh and in blood samples collected from little auks wintering off Newfoundland. Hg analyses were performed at the Littoral Environnement et Sociétés laboratory (LIENSs, La Rochelle, France) on ∼3 mg of homogenized whole blood or on one complete feather, using an advanced Hg analyzer spectrophotometer (Altec AMA 254).[30] Analyses were repeated two or three times for each sample until the relative standard deviation for two samples 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 was used [Lobster Hepatopancreas Tort-2; NRC, Canada; Hg concentration of 0.27 ± 0.06 μg/g of dry weight (dw)] and measured every 10 samples. The average measured value was 0.26 ± 0.01 μg/g of dw (n = 44). Additionally, blanks were run at the beginning of each sample set. The detection limit of the method was 0.005 μg/g of dw.

Stable isotope carbon ratios were measured on non-lipid-extracted whole blood samples,[31] collected from little auks wintering off Newfoundland. Analyses were performed at LIENSs on ∼0.5 mg subsamples of material loaded into tin cups, 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 $\delta^{13}C = (R_{sample}/R_{standard} - 1) \times 1000$, where R is the $^{13}C/^{12}C$ molar ratio. Standard values for C were Vienna-PeeDee Belemnite (VPDB). Replicate measurements of internal laboratory standards (acetanilide) indicated that the measurement error was ±0.06.

The $\delta^{13}C$ isotopic values mainly reflect the carbon source (i.e., foraging habitat) used by birds.[32]

Spatial Analyses. Light-level data were extracted from GLS loggers and processed following published procedures.[28] Briefly, light-level data were converted into positions using the BASTrak software package (BAS, Cambridge, U.K.). We used a threshold light intensity of 10 and an angle of sun elevation of −3.0° and applied compensation for movements. The angle of sun elevation was defined following the "Hill−Ekstrom calibration" method.[33] For each equipped bird, two positions were obtained per day (at local noon and midnight). From individual positions, we then calculated the median geographical position occupied by each tracked bird during the nonbreeding period (October 15 to February 20). Because the durations of day and night are equal during equinoxes, bird nonbreeding positions from February 20 to April 1 could not be accurately determined from light-level recordings and were therefore not included in the analysis. Spatial analyses were performed using ArcMap version 10.1 (ESRI, Redlands, CA).

Statistical Analyses. Statistics were computed using *R* version 3.0.2 (R Development Core Team 2011). Differences in Hg concentrations between seasons, sexes, and years were tested using Student's *t* tests or Mann−Whitney tests in cases where distributions were not close to normal. We used a multiple linear regression to test for a relationship between concentrations of Hg that accumulated during the nonbreeding period (as reflected by Hg concentrations in head feathers) and the nonbreeding distribution of birds, defined as their median longitude and latitude between October 15 and February 20. Simple linear regressions were used to investigate whether Hg concentrations were linearly correlated to $\delta^{13}C$ values and whether little auk egg volumes and hatching date were correlated to Hg concentrations measured in head feathers. For these two latter relationships, only females were considered. Values are presented as means ± the standard deviation, and statistical significance was assumed at $p < 0.05$.

RESULTS

Hg concentrations were slightly but significantly higher in 2011 than in 2010 in blood samples collected during the breeding season (for the *t* test, $t = 3.51$, df = 61, and $p < 0.001$) and in body feather samples (for the Mann−Whitney test, $U = 245$ and $p < 0.01$) (see Figure S1 of the Supporting Information). Hg concentrations in head feathers were similar in 2011 and 2010 [for the *t* test, $t = 0.41$, df = 61, and $p = 0.68$ (see Figure S1 of the Supporting Information)]. No difference was found between male and female Hg concentrations in body and head feathers (for males, $t = 0.62$, df = 63, and $p = 0.54$; for females, $t = 0.20$, df = 61, and $p = 0.84$). However, breeding males had blood Hg concentrations slightly but significantly higher than those of females [for the *t* test, $t = 3.58$, df = 61, and $p < 0.001$ (Figure S2 of the Supporting Information)]. Importantly, differences observed between years and sexes did not affect the contrasting seasonal Hg contamination patterns observed in little auks (see below).

Seasonal Hg Contamination. Hg concentrations measured in blood samples were higher in nonbreeding (wintering) ($n = 14$) than in breeding ($n = 63$) little auks [for the Mann−Whitney test, $U = 0$ and $p < 0.001$ (Figure 1)]. During the nonbreeding season off Newfoundland, birds had an average

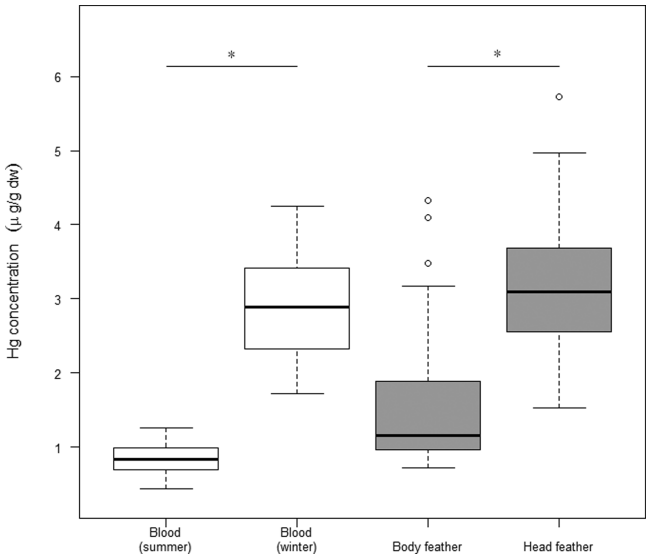

Figure 1. Hg concentrations (in micrograms per gram of dry weight) measured in little auk blood and feather samples. Body feather concentrations indicate summer contamination, while head feather concentrations indicate winter contamination (see Introduction and Materials and Methods for details). Asterisks indicate significant differences between seasons in both blood and feathers ($p < 0.001$).

Hg concentration in blood of 2.86 ± 0.78 μg/g of dw (minimum of 1.72, maximum of 4.25), while this concentration was 0.84 ± 0.20 μg/g of dw (minimum of 0.44, maximum of 1.25) for individuals breeding in East Greenland. A similar trend was found in feather samples; Hg concentrations in head feathers were significantly higher than in body feathers [for the *t* test, $t = 11.07$, df = 126, and $p < 0.001$ (Figure 1)]. Head feather concentrations (average of 3.17 ± 0.83 μg/g of dw, minimum of 1.53, maximum of 5.73, $n = 81$) reflect contamination during the nonbreeding period, while body feather concentrations (average of 1.53 ± 0.84 μg/g of dw, minimum of 0.71, maximum of 4.33, $n = 78$) reflect contamination during the breeding season (see Introduction for details).

Hg Contamination versus Nonbreeding Distribution. Hg concentrations were negatively and linearly correlated with $\delta^{13}C$ values in blood samples collected during the winter [$F_{1,12} = 7.92$, $p = 0.02$, and $R^2 = 0.35$ (Figure 2)], indicating that nonbreeding Hg contamination of little auks is linked to their foraging habitat. The multiple-regression analysis showed a significant relationship between bird median position during the nonbreeding season and Hg concentration measured in head feathers ($F_{2,46} = 3.78$, $p = 0.03$, and $R^2 = 0.10$), with Hg concentrations being higher in birds wintering at more southerly ($p = 0.02$) and westerly ($p = 0.03$) positions. More specifically, little auks with a median nonbreeding position located along the eastern slopes of the Grand Banks of Newfoundland and the Flemish Cap were globally more contaminated than little auks from other areas (Figure 3).

Relationship between Female Hg Contamination and Their Reproduction. A negative linear relationship between Hg concentrations in female head feathers (i.e., nonbreeding Hg contamination) and their egg size was observed [$R^2 = 0.18$, $F_{1,16} = 4.83$, and $p = 0.04$ (Figure 4)]. There was no

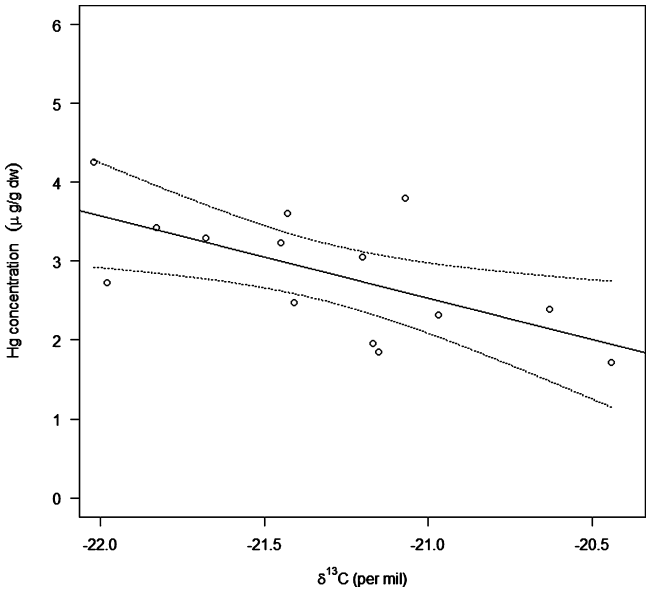

Figure 2. Blood Hg concentrations (in micrograms per gram of dry weight) in relation to the $\delta^{13}C$ isotopic values of little auks wintering in the northwest Atlantic. Higher (less negative) $\delta^{13}C$ values indicate a more inshore diet and lower (more negative) $\delta^{13}C$ values a more offshore diet.[43,44]

relationship between Hg concentrations in female head feathers and hatch dates (R^2 = 0.1, $F_{1,9}$ = 0.007, and p = 0.94). Furthermore, there was no relationship between female blood Hg concentrations and their body mass at recapture or between female head feather Hg concentrations and their median nonbreeding position (all $p > 0.2$).

DISCUSSION

Hg concentrations have previously been measured in a large variety of Arctic seabirds and other top predators.[8,34] These studies provided essential information about Hg concentrations in organisms,[34] about their trends over past decades or centuries,[6] and about the ecotoxicological effects of this contaminant.[10] This work has greatly improved our understanding of the threat posed by mercury to the Arctic wildlife and of the risks associated with increases in levels of of Hg in the marine environment. Nevertheless, all of this work focused on the Arctic region, mainly during the summer when seabirds are breeding, which excludes a large part of the annual cycle. Consequently, and to the best of our knowledge, no study previously investigated the intraindividual seasonal Hg contamination of Arctic seabirds. This is unfortunate because seabirds can travel large distances and spend the nonbreeding period far from their breeding site in non-Arctic areas where their exposure to contaminants remains largely unknown. Taking advantage of the specific moulting pattern of little auks and by combining biotelemetry and Hg and stable isotope analyses, we demonstrate that ecotoxicological studies should be extended to yield a comprehensive understanding of contamination risks and associated threats to seabirds and other top predators over their entire annual cycle. Indeed, we show by analyzing blood samples that little auks breeding in East Greenland and wintering in the northwest Atlantic off Newfoundland were 3.5 times more contaminated during the winter period. This difference was smaller in feather samples, but contamination over the nonbreeding period was still twice

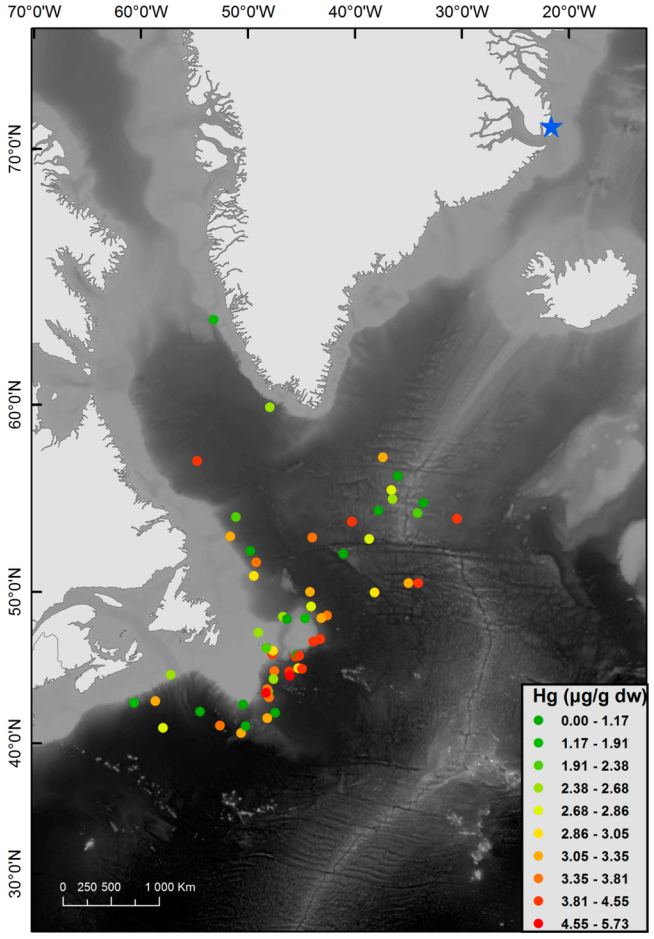

Figure 3. Hg concentrations (in micrograms per gram of dry weight) measured in head feathers of little auks breeding in East Greenland, in relation to their nonbreeding distribution in the northwest Atlantic. Each point represents the median distribution of individually tracked little auks during the nonbreeding period (October 15 to February 20). The blue star represents the breeding colony where birds were equipped.

as high. Hg concentrations measured in feathers reflect the amount of Hg that accumulated in body tissues since the last moult,[24−26] which corresponds to a period extending from approximately April to September for little auk body feathers.[20,21] Although we considered this period as the breeding season, it also includes a period of postbreeding movements (between the breeding site in East Greenland and a moulting area in the Greenland Sea)[21,28] as well as prebreeding movements, as it is unlikely that little auks moult their summer plumage at their breeding site.[19,28] During these short periods, little auks were in areas where their exposure to Hg was possibly different than that at their breeding site,[8,35] leading to slightly higher Hg concentrations in body feathers when compared to blood samples.

The AMAP 2011 Impact Assessment reviewed several studies that investigated the nonbreeding distribution and Hg contamination of various seabird species.[36−41] From their results, AMAP suggested that the lack of a significant temporal trend in Hg concentrations of Arctic seabirds that winter at low latitudes, outside of the Arctic, could be explained by a lower level of exposure to Hg during their nonbreeding period, in contrast to species wintering at higher latitudes that showed increasing Hg levels.[8] Here we demonstrate that this

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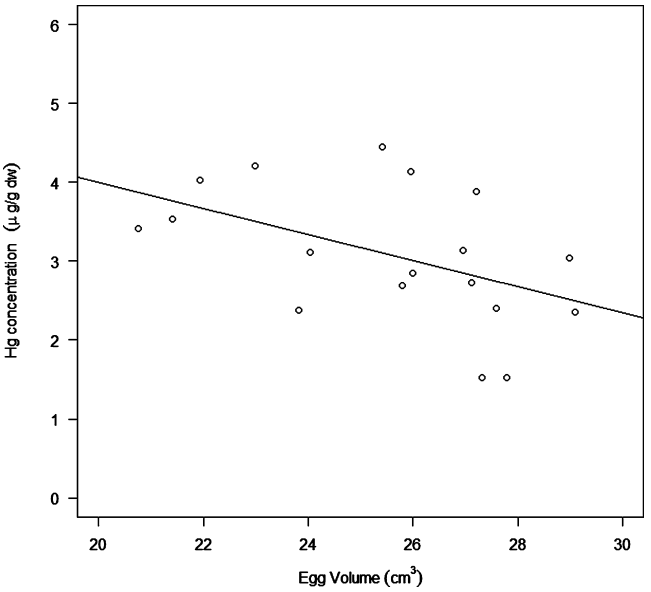

Figure 4. Egg volume (cubic centimeters) of female little auks breeding in East Greenland in relation the Hg concentrations (in micrograms per gram of dry weight) measured in their head feathers [i.e., accumulated during the nonbreeding period (see Introduction for details)].

explanation should not be generalized to all species wintering in the northwest Atlantic, which may actually be exposed to higher levels of Hg contamination when not breeding. An alternative hypothesis for explaining the lack of a trend reported by AMAP is that species and populations considered in their study did not winter in the northwest Atlantic as supposed, but in other regions where Hg concentrations are lower. Our results indeed confirm that the geographical position as well as the foraging habitat used by birds can affect their Hg contamination. In the northwest Atlantic, we found that the level of little auk Hg contamination was higher in birds that had median nonbreeding positions to the south and west. Within the Arctic, Mallory and Braune[42] also highlighted a relationship between seabird breeding distribution in the Canadian Arctic and their Hg concentrations, with birds breeding in the high Arctic being more contaminated than those from the low Arctic.[42] Isotopic results showed that wintering birds with enriched carbon ratios were less contaminated. The $\delta^{13}C$ ratio is an indicator of the source of primary production in marine systems and can also be used to indicate inshore versus offshore contribution to feeding habitats.[43,44] Because enriched carbon values in marine predators have been shown to correspond to a more inshore diet,[43,44] this suggests that little auks foraging closer to the shore were less exposed to Hg contamination than birds that had recently foraged offshore. This difference could either reflect contrasting baseline Hg contamination of these habitats or be the result of a different diet when birds are foraging offshore and inshore with contrasting contamination levels of consumed prey items.[45] Our results therefore highlight the need to extend seasonal contamination studies and focus on additional populations and species that spend the nonbreeding season in different regions and habitats where Hg exposure is different.[35] Studies combining contaminant analyses, biotelemetry, and an isotopic approach will provide essential knowledge about the seasonal contamination of Arctic seabirds and confirm the pollution risk that regions south of the Arctic represent for this community (see also ref 18 for persistent organic pollutants). For instance, little auks breeding in Spitsbergen winter at latitudes higher than those of Greenlandic breeders.[28] Other species such as kittiwakes (*Rissa tridactyla*), guillemots (*Uria* spp.), and razorbill (*Alca torda*) overwinter in different areas of the North Atlantic,[13,46,47] whereas long-tailed skua (*Stercorarius longicaudus*) and Sabine's gulls (*Larus sabini*) migrate to the South Atlantic Ocean along the West African coasts.[14,48] Moreover, future studies could also help identify, on large spatial scales, sensitive areas where contamination risks are higher for Arctic wildlife. In this context, our results suggest that long-lived top predators such as seabirds can be efficient indicators of the state of contamination of entire marine systems, by integrating contaminant concentrations on a large spatial scale and across marine food webs. Accordingly, our results indicate that the area located along the eastern slopes of the Grand Banks of Newfoundland and the Flemish Cap was associated with higher levels of Hg contamination and merits further attention.

As mentioned above, Hg contamination of organisms is also closely related to their diet, and a seasonal change in diet could affect ingested Hg concentrations.[45] Little auks breeding in East Greenland are known to mainly forage on copepods during the breeding season.[49] However, isotopic data suggested a change in diet soon after breeding, toward krill and fish larvae.[50] Stomach content analyses performed on the same individuals that were used for our study also showed a diet mainly composed of krill during late winter (March).[22] Although we do not know if these different diets reflect different trophic positions, the higher level of contamination observed during the nonbreeding period could also partially result from a change in diet. Such a change could also be a second alternative explanation to trends interpreted by AMAP. Like Brünnich's guillemots (*Uria lomvia*)[51] or northern fulmars (*Fulmarus glacialis*),[52] which switch from a fish-based to a zooplankton-based diet during winter, other species could feed at lower trophic levels during the nonbreeding period, resulting in lower levels of Hg exposure during the nonbreeding period.

Moreover, our results suggested that female little auks that had higher head feather concentrations and so had accumulated more Hg during the nonbreeding season laid smaller eggs. It is not possible from this study to determine if Hg has a direct impact on little auk reproduction or if it rather reflects general contamination by trace metals or other contaminants. Hg concentrations measured in this study ranged from 0.4 to 5.7 μg/g of dw (1.5−5.7 μg/g of dw in head feathers). These values are similar or below concentrations measured in similar tissues in other seabird species,[45,53,54] so a direct relationship between Hg concentration and female investment in reproduction may seem surprising. Hg contamination thresholds that lead to adverse effects are poorly known in seabirds, especially for feather and blood matrices (see refs 55 and 56 for other matrices). However, studies performed on other aquatic birds suggested that Hg concentrations of 5 μg/g of dw in feathers can cause reproductive impairment.[57] A recent Arctic study suggested that low Hg concentrations in blood samples (approximately 3 μg/g of dw) could affect the reproductive success of kittiwakes by disrupting their endocrine system.[16] Therefore, further studies are required to confirm the adverse effect found in our study and to understand the direct or indirect physiological link between Hg concentrations and egg synthesis in seabirds. Among others, it will be important to further consider selenium (Se) concentrations and the Hg:Se ratio. Indeed, Se interacts, through specific chemical mecha-

nisms, with Hg to form nontoxic Hg−Se complexes and therefore acts as a form of protection against methyl-Hg toxicity.[58] Hence, high Hg levels would not necessarily induce adverse toxicological effects if Se is abundant compared to Hg and if the Hg:Se ratio is low. Many seabird species are known to overwinter in huge numbers in the northwest Atlantic[13,59−61] and, like little auks, could be exposed to high levels of Hg contamination with the associated threats to their reproduction. Such studies would improve our understanding of the threat that Hg exposure represents for these vulnerable species, within and outside of the Arctic.

■ ASSOCIATED CONTENT

Supporting Information

Annual and sex differences in Hg concentrations measured in little auk blood and feather samples (Figures S1 and S2, respectively). This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: fort.jerome@gmail.com. Telephone: +33 546458388.

Notes

The authors declare no competing financial interest.

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