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Article

A U-Turn for Mercury Concentrations over 20 Years: How Do Environmental Conditions Affect Exposure in Arctic Seabirds?

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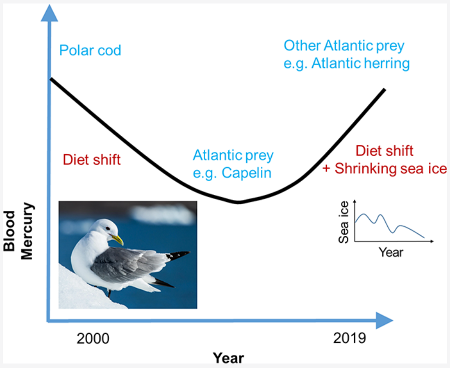

ABSTRACT: Mercury (Hg) is highly toxic in its methylated form (MeHg), and global change is likely to modify its bioavailability in the environment. However, it is unclear how top predators will be impacted. We studied blood Hg concentrations of chick-rearing black-legged kittiwakes *Rissa tridactyla* (2000−2019) in Svalbard (Norway). From 2000 to 2019, Hg concentrations followed a U-shaped trend. The trophic level, inferred from nitrogen stable isotopes, and chlorophyll *a* (Chl *a*) concentrations better predicted Hg concentrations, with positive and U-shaped associations, respectively. As strong indicators of primary productivity, Chl *a* concentrations can influence production of upper trophic levels and, thus, fish community assemblage. In the early 2000s, the high Hg concentrations were likely related to a higher proportion of Arctic prey in kittiwake's diet. The gradual input of Atlantic prey in kittiwake diet could have resulted in a decrease in Hg concentrations until 2013. Then, a new shift in the prey community, added to the shrinking sea ice-associated release of MeHg in the ocean, could explain the increasing trend of Hg observed since 2014. The present monitoring provides critical insights about the exposure of a toxic contaminant in Arctic wildlife, and the reported increase since 2014 raises concern for Arctic seabirds.

KEYWORDS: *global change, trace elements, temporal trends, Rissa tridactyla, chlorophyll a, sea-ice extent, feeding ecology*

INTRODUCTION

The Arctic is warming two to three times faster than any other region on Earth.[1,2] Key indicators, such as temperature, precipitation, snow cover, and sea-ice thickness or extent, show rapid and widespread changes underway in the Arctic. Between 1971 and 2019, the annual surface temperature changes in the Arctic (land and ocean) were three times higher than the global average during the same period.[3] The loss of sea-ice cover over extensive areas of the Arctic Ocean has allowed radiation to penetrate the water during summer, storing heat in the upper ocean. Unique ecosystems, such as those associated with multiyear sea-ice or millennia-old ice shelves, are at risk and some are vanishing.[3] These rapid changes are causing fundamental changes in ecosystems that affect biogeochemical fluxes, bottom-up processes, ecosystems, and food webs, which may lead to modifications in contaminant exposure in the Arctic biota.[5,6] Among contaminants of high concern, mercury (Hg) is a non-essential trace element from both natural and anthropogenic sources. The Arctic has been considered as a sink for Hg deposition, with the highest deposition observed in the European part of the Arctic, downwind of major anthropogenic source regions.[7] However, the fate of Hg trends in Arctic ecosystems in light of global change remains unclear.

Global warming, which will lead to an earlier onset of thawing and a later start of freezing, is likely to extend the season of Hg methylation.[4] Methylmercury (MeHg), which is known to be the most toxic among the Hg compounds, is generated when inorganic Hg is dissolved into water.[8] While bacterial methylation is considered as the major pathway,[9] chemical reactions yielded by environmental factors, such as pH and temperature, contribute to making MeHg more bioavailable in aquatic ecosystems.[10] MeHg has been identified as a priority contaminant to consider in future climate change−pollutant scenarios.[11,12] An increased period during which MeHg can be produced may then lead to enhanced MeHg exposure in marine food webs. In contrast, global warming is also likely to modify Hg demethylation processes in polar ecosystems.[13] One of the most important degradation processes of MeHg in ocean surfaces is photoinduced

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demethylation.[13] MeHg decomposes under ultraviolet and visible light.[14] Therefore, a reduced sea-ice extent will expose a larger ocean surface and could enhance Hg photodemethylation. The transfers and transformations of Hg in Arctic aquatic systems are poorly understood and need more research.[15] Hg concentrations biomagnify throughout the food webs, with higher concentrations in higher trophic level species.[16] Modifications in productivity at the lower levels of food webs may affect the amount of energy available for predators to grow and reproduce. Energy intake, as measured by body condition or body size, and feeding habits are therefore interesting variables to consider when studying Hg concentrations in higher trophic level species.

The dynamics of Hg exposure over time can be monitored through the exposure of top predators that are considered as relevant bioindicators of environmental pollution.[17−19] A review paper has suggested that, as a consequence of global warming, the temporal trends of MeHg in top predators are uncertain.[12] Increasing trends of Hg concentrations were observed in several Arctic seabird species[20,21] and have been attributed to anthropogenic emissions, climate-related variables, or changes in food web contamination. The Barents Sea is a hotspot of Arctic warming. Between 1979 and 2013, the number of sea-ice-free days around the Svalbard Archipelago (Norwegian Arctic) has increased by 41.8 days, which is far greater than what had been monitored in other parts of the Arctic during the same period.[22] Since the 1990s, Hg exposure followed a nonlinear increase in some mammals of Svalbard, which has been attributed to changes in the food web as well as re-emission of previously stored Hg from thawing sea ice, glaciers, and permafrost.[18,19]

Black-legged kittiwakes *Rissa tridactyla* (hereafter "kittiwakes") are generalist top predators and can be useful indicators of Hg contamination in their habitat.[23] Between 1975 and 2010, at the exception of a few,[24] kittiwake colonies have been declining throughout their breeding range, and this coincided with an abrupt warming of sea surface temperature in the 1990s.[25] In addition, in this species, high Hg concentrations have been associated to decreased breeding probability, lower concentrations of the parental hormone prolactin, and decreased breeding success.[23,26,27] Although research conducted on kittiwakes was based on field studies, several experimental studies conducted on other bird species have led to similar conclusions.[28,29] For instance, Hg exposure led to lower survival and hatching success in bird laboratory models.[28,29] Therefore, the observed decline in kittiwake colony size[25] may be aggravated by Hg-related breeding failure.[30]

Kongsfjorden is a fjord located on the west coast of Svalbard, where kittiwakes breed in large colonies. In recent years, Kongsfjorden has been strongly influenced by the West Spitsbergen Current, with increasing temperatures and declining sea-ice cover inshore. Since 2005, an increasing proportion of Atlantic species has been part of kittiwake diet,[31] which may possibly influence Hg exposure. The aim of the present study was to investigate the temporal trends of blood Hg exposure in kittiwakes over two decades and identify its potential drivers. We predicted that, in concordance to what is observed in other seabird species from Canada and Greenland and in Svalbard mammals,[18,19,21] a climate-driven diet modification would lead to a nonlinear increase of Hg in Svalbard kittiwakes. In addition, since fish community modifications in Kongsfjorden were driven by sea-ice extent,[31] we predicted that the sea-ice extent, over other climate-related variables, would be the strongest predictor of Hg concentrations in Svalbard kittiwakes.

■ MATERIAL AND METHODS

Fieldwork and Blood Analyses. Captures, blood sampling and analyses, are described in detail in the Supporting Information. Briefly, from 2000 to 2019 (excluding 2010), 1152 kittiwakes were captured during different phases of the breeding cycle in Kongsfjorden (78°54′N, 12°13′E), Svalbard , Norway. A blood sample (ca. 1 mL) was collected from the alar vein. Birds were weighed to the nearest 5 g using a Pesola spring balance, and their skull length was measured to the nearest 0.5 mm with a sliding caliper. Birds were captured during the breeding period, which can be separated into three stages: the pre-laying, egg-incubation, and chick-rearing phases. Temporal trends of total blood Hg (hence "Hg") and associations with climate-related variables were analyzed in chick-rearing kittiwakes only (n = 392) to reflect local Hg exposure and to make between-year comparisons possible. Methods for QA/QC, molecular sexing, Hg, and stable isotope analyses are detailed in the Supporting Information. An overview of the samples available per year and per breeding stage is given in Table S1.

Environmental and Biological Data. Mercury, carbon, and nitrogen stable isotopes in blood reflect the diet during the previous month up to the two previous months;[32−34] see the Supporting Information for more details. Thus, sampling conducted during the chick-rearing stage integrates the feeding ecology within the two previous months (May and June). We selected 10 environmental variables known to influence Hg in the ecological niche of kittiwakes.[4,21] These variables included the North Atlantic Oscillation (NAO), the Arctic Oscillation (AO), atmospheric gaseous elemental Hg (GEM), air temperature, sea-ice extent, sea surface temperature anomaly (SSTA), chlorophyll *a* concentrations (Chl *a*) which is used as a proxy of primary productivity and a useful indicator of fishery production at both regional and global scales,[35,36] and precipitation. We also analyzed the relationships between Hg concentrations and ocean acidification indicators such as pH and aragonite saturation at the bottom of the water column (see the Supporting Information). Except for global measures (NAO and AO), all measures were taken locally during the spring period in the foraging area of kittiwakes. Climate-related variables, sampling periods, and sampling years available for each variable are summarized in Table S2. Except for pH and aragonite saturation, which will be the subject of another paper, temporal trends of the eight other variables suspected to predict Hg concentrations in kittiwakes are shown in Figure S1.

Statistical Analyses. We detrended biological- and climate-related time series to test if year-to-year variations in the response variable are driven by year-to-year variations in some explanatory variables. To do so, we extracted the residuals from independent generalized additive mixed-effect models (GAMMs) with year as the predictor, with a smoothed parameter k adapted to the shape of the trend (e.g., k = 4 for Hg and climate-related variables, k = 5 for morphometrics, and k = 8 for stable isotopes). Then, we examined the residual covariation between Hg and biotic or abiotic variables.[37] All analyses were conducted using detrended variables except those aimed specifically at testing the effect of year on a variable (e.g., temporal trends of Hg). We provide all statistical

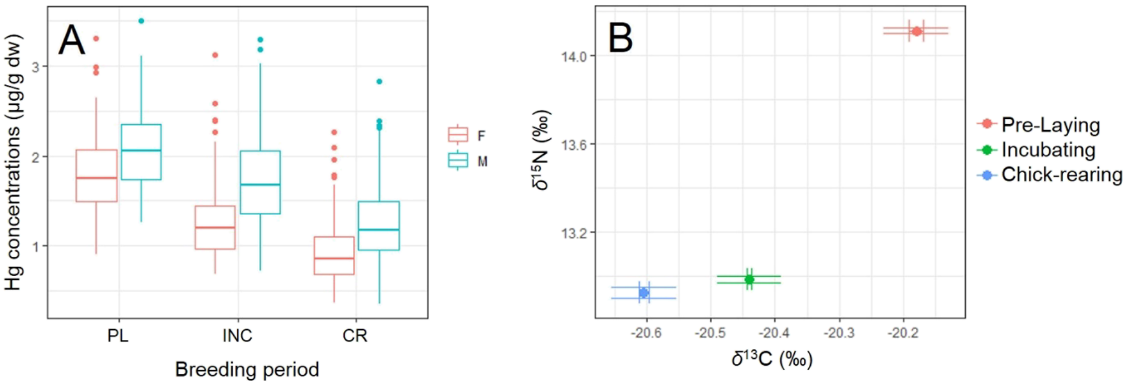

Figure 1. Blood Hg concentrations and stable isotope values across the breeding cycle in black-legged kittiwakes (Kongsfjorden, Svalbard). (A) Boxplot of non-adjusted Hg concentrations (n = 1152, 2000−2019) in males (blue) and females (pink) for each breeding period (PL, pre-laying; INC, incubation; CR, chick-rearing). The median is shown with lower and upper quartiles (box), whiskers (1.5× interquartile range from the box), and outliers (values beyond whiskers); (B) mean nitrogen (δ^{15}N) and carbon (δ^{13}C) stable isotope values with error bars (SE) in blood samples from black-legged kittiwakes (n = 77, 2011) during each breeding period.

Table 1. Biological Factors Influencing Blood Hg Concentrations in Black-Legged Kittiwakes from Kongsfjorden, Svalbard[a]

(A) Hg and stable isotope variation according to the breeding phase

response variable	dataset	intercept	incubation	chick-rearing
Hg (µg/g dw)	all individuals	**0.56 (0.50; 0.63)**	**−0.40 (−0.46; −0.33)**	**−0.79 (−0.86; −0.72)**
	subset only with recaptures	**0.53 (0.45; 0.61)**	**−0.44 (−0.52; −0.36)**	**−0.85 (−0.93; −0.77)**
δ^{13}C (‰)	2011	**0.07 (0.006; 0.150)**	**−0.28 (−0.38; −0.17)**	**−0.40 (−0.51; −0.29)**
δ^{15}N (‰)	2011	**0.69 (0.54; 0.85)**	**−1.13 (−1.35; −0.91)**	**−1.15 (−1.38; −0.91)**

(B) Hg variation according to sex and morphometrics

models: Hg	df	logLik	AICc	ΔAIC	weight	β (95% CI)	
sex	**4**	**−783.23**	**1574.5**	**0**	**1**	intercept	**−0.16 (−0.22; −0.11)**
mass	4	−794.27	1596.57	22.07	0	sex	**0.35 (0.27; 0.42)**
skull	4	−796.17	1600.38	25.88	0	mass	**0.004 (0.003; 0.005)**
SCM	4	−815.3	1638.64	64.14	0	skull	**0.05 (0.04; 0.06)**
null model	3	−819.71	1645.44	70.95	0	SCM	**0.002 (0.001; 0.003)**
SMI	4	−827.42	1662.87	88.38	0	SMI	0.0001 (−0.0002; 0.0004)

[a](A) Blood Hg concentration (n = 1066) and stable isotope values (n = 80) across the breeding cycle. (B) Blood Hg variations according to biometrics (n = 1066). Models are LMMs computed on detrended variables, with year and bird identity as random factors. Values are estimates and 95% confidence intervals. The subset of recaptured birds consists of 43 samples representing 20 individuals recaptured within the same year between two and three times. SCM: size-corrected mass; SMI: scaled mass index.

methods in detail, in addition to results conducted on nondetrended data in the Supporting Information.

We first tested Hg and stable isotope variations (detrended variables) across the breeding cycle by using linear mixed-effect models (LMMs) with the breeding period and sex as independent categorical three- and two-level predictors and bird identity as a random factor. Then, we investigated whether sex, body mass, size-corrected mass (SCM) (i.e., residuals from the mass−skull size regression), scaled mass index (SMI),[38] and skull size would influence individual detrended Hg concentrations using LMMs. We obtained estimated marginal means (EMMs) for specified factor combinations of the LMM (breeding period × sex interactions) by using the "emmeans" package.[39]

Second, to investigate the temporal trends of Hg and other biological variables (δ^{13}C, δ^{15}N, body mass, and skull size) in chick-rearing kittiwakes, over a 20 year period, we used GAMMs on nondetrended data to account for nonlinear relationships between response variables and predictors and for recaptured individuals. Year, δ^{13}C or δ^{15}N, and sex were used as continuous predictors and bird identity as a random factor to analyze temporal trends of Hg, whereas models explaining the temporal trends of δ^{13}C, δ^{15}N, body mass, and skull size only used sex as a predictor and bird identity as a random factor. For each model, we adapted the smoothed term (k) to avoid overfitting. Break points were identified by using a Bayesian approach ("broken stick" modeling technique,[40,41] detailed in the Supporting Information.

Third, we analyzed the relationships between detrended Hg concentrations and detrended climate-related variables. To avoid pseudoreplication due to the large, and sometimes unbalanced, number of kittiwakes caught per year, we calculated for each year the average Hg concentrations and δ^{13}C and δ^{15}N values for each sex and performed statistical analyses on this subset of n = 38 sampling units. Since Hg trends in biota often present nonlinear variations[42−44] and have been increasing nonlinearly in Svalbard vertebrates,[18,19] we tested both linear (e.g., sea-ice extent) and quadratic (e.g., sea-ice extent + sea-ice extent^2) relationships using linear models (LMs). We computed a list of 34 biologically relevant model candidates by adding the variables selected from the best model from the analysis described on step two (Hg variations in relation to morphometrics and sex), to which we added one of the eight climate-related variables without missing years, either as linear or quadratic. We then selected the best fits (linear or quadratic) for all variables included in the top 10 models to explain detrended Hg concentrations and used conditional model averaging to make inference from all of the models.[45,46] Variables with missing years (pH and aragonite saturation at the bottom of the water column) were analyzed separately using LMs. Finally, we tested which of the strongest climate-related variables would better explain feeding habits (δ^{13}C or δ^{15}N); to do so, we used LMs with detrended variables.

All models were ranked using AIC.[47] We obtained the degrees of freedom (df), the log likelihood (logLik), the difference in AIC values between the "best" model and the model at hand (ΔAIC), and a normalized weight of evidence in favor of the specific model, relative to the whole set of candidate models, derived by $e^{(-0.5(\Delta \mathrm{AICc}))}$ (weights). We obtained estimates and 95% confidence intervals (95% CI) for each parameter, and an effect was considered significant when the 95% CI did not include 0. The model fit was assessed by using residual diagnostic plots. All analyses and figures were performed using R software v.3.6.3[48] with the following packages: car, mgcv, nlme, effects, ggplot2, and corrplot.[49−54] The data sharing policy is detailed in the Supporting Information.

RESULTS

Biological Variables Influencing Individual Hg Concentrations. From 2000 to 2019, Hg was analyzed in 1152 kittiwake blood samples. Among these birds, biometrics (skull and body mass) were available for 1100 birds and stable isotopes were analyzed in 508 birds (Table S1). Mercury concentrations decreased by 55.7% across the breeding cycle, ranging from 1.97 ± 0.03 μg/g dw in pre-laying kittiwakes to 1.09 ± 0.02 μg/g dw (values represent raw mean ± standard error concentrations) in chick-rearing kittiwakes (Figure 1A and Table 1A). This decrease of Hg across the breeding cycle was further supported by analyses conducted on a subset of birds, which were recaptured during different breeding phases within a year (Figure S2 and Table 1A). Blood Hg concentrations in males were higher by 0.22 μg/g (0.04; 0.39) during the pre-laying period, 0.47 μg/g (0.33; 0.66) during the incubation period, and 0.37 μg/g (0.21; 0.44) during the chick-rearing period, in comparison to females (values are estimates from EMMs and 95% CIs in parentheses). In 2011, which was the only year where stable isotopes were analyzed in a large enough number of individuals for each period, we analyzed δ^{13}C and δ^{15}N variations across the breeding cycle (Figure 1B). Stable isotope values in incubating and chick-rearing birds were lower than values in pre-laying birds (Table 1B and Figure 1B). In 2011, stable isotopes were not related to sex at any stage of the breeding season (males vs females; δ^{15}N: pre-laying period: −0.09 (−0.54; 0.36), incubation: 0.32 (−0.21; 0.85), chick-rearing: 0.29 (−0.29; 0.86); δ^{13}C: pre-laying period: 0.08 (−0.14; 0.31), incubation: −0.0003 (−0.25; 0.25), chick-rearing: −0.15 (−0.42; 0.12), EMM estimates and 95% CI).

When analyzing the effects of sex, body metrics, and conditions on kittiwake's Hg concentrations, sex over morphometrics better explained Hg concentrations (Table 1), and we therefore included sex as a fixed factor in further models. Among body metrics and condition calculations, body mass, skull size, and SCM were positively related to detrended Hg concentrations (Table 1). However, those relationships were sex- and breeding period-dependent. For instance, in females, Hg and body mass were unrelated in pre-laying birds, negatively related in incubating birds, and positively related in chick-rearing birds, whereas for males, Hg and body mass were unrelated when considering breeding periods separately (Table S3). For all Hg-biometric relationships according to sex and the breeding period, see Table S3.

Temporal Trends of Blood Hg Concentrations in Chick-Rearing Kittiwakes. In chick-rearing kittiwakes (n = 392), the best ranked model to explain temporal variations of blood Hg included year, sex, and δ^{15}N (Figure 2 and Table 2).

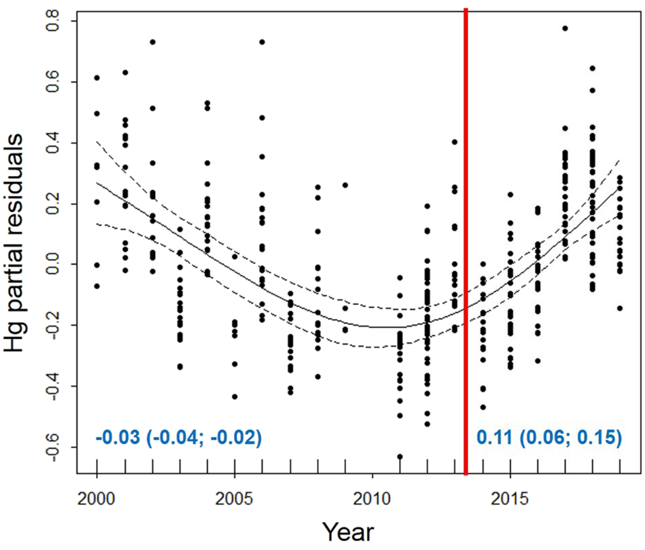

Figure 2. Temporal trends (2000−2019) of blood Hg concentrations in adult chick-rearing kittiwakes (n = 392) from Kongsfjorden, Svalbard. Hg partial residuals are derived from the best model selected by AIC (Table 2); Hg partial residuals are adjusted for δ^{15}N, sex as fixed factors, and bird identity as a random factor. The red line represents the selected change point in Hg concentrations, which corresponds to 2014. Values are estimates and 95% CI obtained from LMMs for the period of 2000−2013 on the left and 2014−2019 on the right.

Blood Hg concentrations followed a U-shaped relationship according to year, with an identified change point in 2014 (Figure 2). Mercury concentrations decreased by 3% per year until 2013 (LMM: −0.03 (−0.04; −0.02)) and increased by 11% per year after 2014 (LMM: 0.11 (0.06; 0.15)).

Over the 20 year period, δ^{15}N and δ^{13}C presented oscillating variations over time (Figure 3A,C). Overall, δ^{13}C values presented a decrease over time (LMM: −0.019 (−0.023; −0.010)), mostly related to δ^{13}C values, which remained low after 2015 (Figure 3A), whereas δ^{15}N values presented a drop from 2013 to 2015 but no general trend overtime (LMM: −0.004 (−0.017; 0.009); Figure 3C). Body mass and skull size temporal trends were bimodal, with the highest values in 2004 and 2014 for body mass (Figure 3B) and 2004 and 2016 for skull length (Figure 3D). While body mass did not follow a general trend over time (LMM: 0.15 (−0.38; 0.68); Figure 3E), the skull length overall increased by 7% per year (LMM: 0.07 (0.03; 0.10)) (Figure 3D). For Hg, δ^{15}N, δ^{13}C, body mass, and skull size, we give estimates and 95% CI for the fixed terms and effective degrees of freedom of the smoothers in Table S4 and diagnostic plots in Figures S3 and S4.

Table 2. Predictors of Blood Hg Variation in Chick-Rearing Kittiwakes (n = 392) from Kongsfjorden, Svalbard (2000−2019)[a]

AIC selection

intercept	s(year, k = 5)	sex	mass	skull	δ^{15}N	δ^{13}C	df	logLik	AIC	ΔAIC	weight
−0.142	+	+			**0.09078**		7	**−142.955**	**299.9**	**0**	**0.817**
3.447	+	+				0.1193	7	−144.661	303.3	3.41	0.148
0.9943	+	+					6	−147.175	306.3	6.44	0.033
−4.252	+			0.04553	0.09819		7	−148.938	311.9	11.97	0.002
−0.6652	+			0.04935		0.1312	7	−150.556	315.1	15.2	0
−3.364	+			0.0494			6	−153.966	319.9	20.02	0
−1.351	+		0.002474		0.1232		7	−163.333	340.7	40.76	0
2.537	+		0.002289			0.11	7	−170.93	355.9	55.95	0
0.2107	+		0.002465				6	−172.255	356.5	56.6	0
1.154	+						5	−174.943	359.9	59.98	0
1.164							3	−199.323	404.6	104.74	0

conditional model-averaged estimates

variable	β (95% CI)
intercept	0.42 [−2.24; 3.07]
δ^{15}N	**0.09 [0.04; 0.14]**
sex	**0.29 [0.22; 0.37]**
δ^{13}C	**0.12 [0.04; 0.19]**
skull	**0.05 [0.03; 0.06]**
mass	**0.002 [0.001; 0.003]**

[a] Model selection and model-averaged estimates. The first part of the table shows the different candidate models and their AIC ranking, and the most competitive model appears on the first line (ΔAIC = 0). The second part of the table shows the estimates and 95% confidence intervals derived from GAMMs. Bold values represent significant relationships. GAMMs include year as a smoother, sex, body mass, skull size, δ^{15}N, and δ^{13}C as fixed factors, and bird identity as a random factor. Sex was included in all models except for those with body measurements, which are already sex-dependent. The "+" sign in the upper part of the table indicates that a smoothed term "s(year, k = 5)" or categorical term "sex" was included in the model.

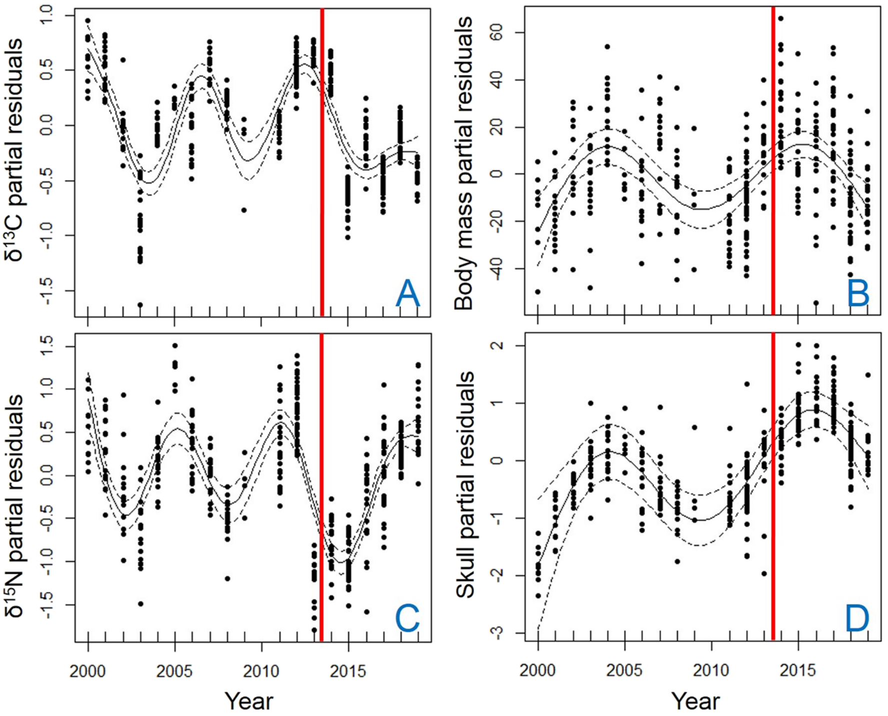

Figure 3. Temporal trends (2000−2019) of stable isotope values in red blood cells and biometrics in adult chick-rearing kittiwakes (n = 392) from Kongsfjorden, Svalbard. (A, C) Feeding habits are inferred from nitrogen and carbon stable isotope values in blood (δ^{15}N and δ^{13}C, respectively), and biometrics include (B) body mass and (D) skull length. The red line represents the year selected as a change point for temporal variations of Hg in the blood of kittiwakes. Dots are partial residuals derived from a GAMM including year as a smoother, sex as a fixed factor, and bird identity as a random factor. Diagnostic plots are given in the Supporting Information (Figure S1).

Environmental Variables Explaining Hg Concentration in Kittiwakes. Among climate-related variables, we show the 10 most competitive models in Table S5 for both detrended and raw data. The best predictor of detrended Hg

Table 3. Relationship between Detrended Blood Hg Concentrations in Chick-Rearing Kittiwakes and Environmental Factors (n = 38)[a]

	AIC ranking					model averaged estimates β (95% CI)		
explanatory variables	df	logLik	AICc	ΔAIC	weight		linear	quadratic
Chl a + Chl a^2 + sex	5	7.14	−2.41	0	0.28	intercept	**−0.19 (−0.29; −0.08)**	
sex	3	4.16	−1.62	0.8	0.18	Chl a	−0.07 (−0.5; 0.36)	**0.5 (0.07; 0.94)**
SSTa + sex	4	5.25	−1.29	1.13	0.16	sex	**0.32 (0.18; 0.47)**	
precipitation + sex	4	4.96	−0.71	1.7	0.12	SSTa	0.26 (−0.11; 0.64)	
NAO + sex	4	4.73	−0.26	2.15	0.09	precipitation	0.001 (−0.0007; 0.0027)	
GEM + GEM^2 + sex	5	5.74	0.4	2.81	0.07	NAO	0.08 (−0.07; 0.23)	
air temp + air temp^2 + sex	5	5.52	0.84	3.25	0.05	GEM	−0.28 (−0.73; 0.16)	−0.25 (−0.7; 0.2)
sea ice + sea ice^2 + sex	5	5.43	1.01	3.42	0.05	air temp	−0.08 (−0.53; 0.37)	−0.34 (−0.79; 0.11)
null model	2	−4.26	12.87	15.28	0	sea-ice extent	−0.13 (−0.59; 0.32)	0.31 (−0.14; 0.76)

[a] The table shows the variables included in the ten highest AIC-ranked models, and the most competitive model appears on the first line (ΔAIC = 0). We give the model-averaged estimates and 95% confidence intervals derived from LMs with Hg concentrations as a response variable. Bold values represent significant relationships. The complete list of candidate models and results from nondetrended variables are given in the Supporting Information. Chl a: chlorophyll a; GEM: gaseous elemental mercury; air temp: air temperature; sea ice: sea-ice extent; SSTa: sea surface temperature anomaly.

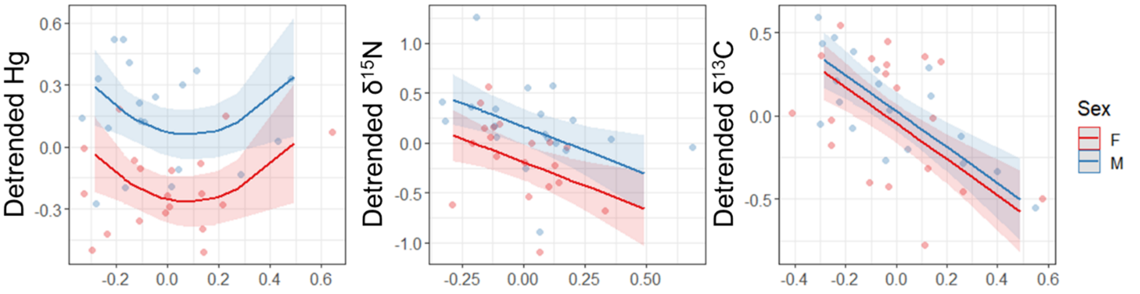

Figure 4. Influence of chlorophyll a on blood Hg concentrations and feeding habits in chick-rearing kittiwakes between 2000 and 2019 (n = 38, Kongsfjorden, Svalbard). Feeding habits are inferred from nitrogen and carbon stable isotope values in blood ($\delta^{15}N$ and $\delta^{13}C$, respectively). Dots are year average values for each sex (females in red and males in blue), the lines are the quadratic and linear fits to the predicted values, and the shaded areas represent their 95% confidence interval derived from LMs on detrended variables. Females are in red, and males are in blue.

Table 4. Relationship between Feeding Habits and Environmental Factors in Chick-Rearing Kittiwakes (n = 38)[a]

	AIC ranking					model averaged estimates β (95% CI)		
explanatory variables	df	logLik	AICc	ΔAIC	weight		linear	quadratic
				nitrogen stable isotopes				
Chl a + sex	4	−15.41	40.1	0	0.57	intercept	−0.18 (−0.38; 0.01)	
precipitation + sex	4	−16.04	41.37	1.27	0.3	Chl a	**−0.96 (−1.62; −0.29)**	
NAO + sex	4	−17.61	44.5	4.4	0.06	sex	**0.37 (0.1; 0.63)**	
sex	3	−19.54	45.83	5.73	0.03	precipitation	**−0.004 (−0.007; −0.0009)**	
GEM + sex	4	−19.12	47.52	7.42	0.01	NAO	−0.26 (−0.54; 0.01)	
null model	2	−22.72	49.8	9.7	0	GEM	−1.09 (−3.58; 1.4)	
SSTa + SSTa^2 + sex	5	−19.34	50.69	10.58	0	SSTa	0.05 (−0.85; 0.94)	0.26 (−0.64; 1.15)
air temp + air temp^2 + sex	5	−19.44	50.88	10.78	0	air temp	−0.09 (−0.98; 0.81)	−0.16 (−1.06; 0.73)
sea ice + sea ice^2 + sex	5	−19.44	50.89	10.79	0	sea-ice extent	−0.05 (−0.95; 0.85)	0.18 (−0.72; 1.07)
				carbon stable isotopes				
Chl a + sex	4	0.44	8.4	0	1	intercept	−0.03 (−0.15; 0.09)	
precipitation + sex	4	−6.32	21.93	13.52	0	Chl a	**−1.08 (−1.51; −0.66)**	
null model	2	−10.41	25.19	16.79	0	sex	0.08 (−0.09; 0.25)	
sex	3	−10.16	27.07	18.67	0	precipitation	**0.003 (0.0009; 0.0055)**	
SSTa + SSTa^2 + sex	5	−7.72	27.44	19.03	0	SSTa	0.56 (−0.09; 1.2)	−0.4 (−1.05; 0.25)
air temp + air temp^2 + sex	5	−7.85	27.7	19.29	0	air temp	−0.64 (−1.29; 0.01)	−0.21 (−0.86; 0.44)
NAO + sex	4	−9.51	28.31	19.9	0	NAO	−0.12 (−0.34; 0.1)	
GEM + sex	4	−10.14	29.58	21.18	0	GEM	−0.16 (−2.1; 1.79)	
sea ice + sea ice^2 + sex	5	−9.58	31.16	22.76	0	sea-ice extent	−0.17 (−0.85; 0.52)	−0.3 (−0.98; 0.38)

[a] The table shows AIC-ranked models, and the most competitive model appears on the first line (ΔAIC = 0). We give the model-averaged estimates and 95% confidence intervals derived from LMs with $\delta^{15}N$ and $\delta^{13}C$ values as response variables. Bold values represent significant relationships. Chl a: chlorophyll a; GEM: gaseous elemental mercury; air temp: air temperature; sea ice: sea-ice extent; SSTa: sea surface temperature anomaly.

concentrations was Chl *a* (Table 3). Sea-ice extent was the top model when considering nondetrended variables (Tables S5 and S6). Hg concentrations followed a U-shaped relationship with Chl *a* (Figure 4) and sea-ice extent. Nitrogen stable isotopes in blood were better explained by Chl *a*, sex, and precipitation, whereas carbon stable isotopes were better explained by Chl *a* and precipitation (Table 4). Nitrogen and carbon stable isotope ratios were negatively related to Chl *a* (Figure 4 and Table 4). Ocean acidification measures, such as pH and aragonite saturation at the bottom of the water column in Kongsfjorden, were unrelated to detrended Hg concentrations in kittiwakes (aragonite: 0.90 (−0.96; 2.76), pH: −2.34 (−11.8; 7.10); values are estimates and 95% CIs obtained from LMMs).

■ DISCUSSION

The present study documents the temporal trends of blood Hg in chick-rearing kittiwakes sampled in Svalbard (2000−2019). Hg showed a U-shaped temporal trend over the 20 year period, declining by 3% per year from 2000 to 2013 and then increasing by 11% per year from 2014 to 2019. At the organismal level, we show that sex is a better predictor of Hg concentrations than body size or body conditions. Hg was higher in males than in females and declined through the breeding season. At the ecosystem level, Chl *a* concentrations predicted Hg concentrations, probably through an effect on kittiwake diet as depicted by the strong relationships between Chl *a* and feeding habits gauged by stable isotope values.

Biological Variables Influencing Individual Hg Concentrations. Hg concentrations in kittiwakes were better predicted by sex. In birds, three ancillary hypotheses are proposed to explain higher Hg concentrations in males than in females: maternal transfer to eggs, body-size dimorphism (i.e., the larger sex consumes larger prey, which are more contaminated), and foraging guild differences.[55] In kittiwakes, those three hypotheses could partly explain the higher Hg concentrations observed in males as they are larger and, at least during the chick-rearing period, males feed at a higher trophic level and on more oceanic prey, as suggested by the higher nitrogen and carbon stable isotope values. However, the higher Hg concentrations observed in males could also result from sex-related physiological differences in Hg retention/excretion.[56−58] An experimental study has reported higher fat mass in birds exposed to higher concentrations of Hg.[59] Moreover, in rodents, a greater urinary excretion of Hg has been observed in females,[60] and despite similar Hg intake, males attained a 20−30% significantly higher whole-body retention in comparison to females.[61]

As previously observed in kittiwakes sampled in Kongsfjorden,[62] Hg concentrations decreased as the breeding cycle progressed, and this was not due to sampling bias, as evidenced by repeated captures of the same individual within a breeding season (Figure S4). This decline was observed in females and males, which suggests that female excretion of Hg into eggs does not fully explain the temporal within-season trend. The continuous decrease of Hg is probably related to temporal shifts in prey over the breeding season, coupled to blood Hg sequestration in protein-rich tissues like muscles.[63] Since Hg concentrations in red blood cells reflect the exposure of the Hg ingested for a period of up to 2 months,[33,34] blood Hg concentrations measured during the pre-laying phase partly correspond to exposure in the wintering ground. Kittiwakes probably feed on higher trophic level more Hg-contaminated prey during winter, as suggested by δ^{15}N values, although we cannot exclude a Hg and/or δ^{15}N baseline shift between the wintering and breeding grounds. Still, it should be noted that kittiwakes are exposed to higher levels of Hg on their wintering grounds in comparison to that in their reproduction area, as observed in other Arctic seabird species,[64,65] and those levels are high enough to disrupt reproduction and parental care in this species.[23,27] This raises a concern about the Hg contamination in North Atlantic waters, the area west of the Mid-Atlantic Ridge, which has been estimated as a wintering area to 80% of the 4.5 million Atlantic adult kittiwakes[66] and has already been characterized as a hotspot for Hg contamination[67,68] with a likely main source of Hg provided from the Arctic Ocean.[69]

Temporal Trends of Hg in Kongsfjorden Kittiwakes. In adult kittiwakes sampled during the chick-rearing period in Kongsfjorden (Svalbard), blood Hg concentrations decreased by 3% per year from 2000 to 2013 and increased by 11% per year from 2014 to 2019. Stable isotope values showed oscillating trends over the study period. We observed a general drop in the values of both δ^{15}N and δ^{13}C around 2013−2015, which could coincide with the increase in Hg concentrations in the blood. Since the 2000s, there has been an expansion of the distributional ranges of many northern Atlantic fish communities into the Arctic.[31,70] The increasing presence of these communities coincides with an increase in the intrusion of Atlantic water with associated Atlantic prey into Svalbard.[70,71] Periodic arrivals of different species could result in the yearly oscillations of δ^{15}N and δ^{13}C values in kittiwake blood. The oscillating pattern of nitrogen and carbon stable isotope temporal trends could result from the fact that stable isotope baseline levels can vary globally and might have more diverse sources than Hg.[72−74] Studying the stable isotopes of Hg could help better track the origin of Hg contamination in kittiwakes.[75,76] The inclusion of sulfur stable isotopes (δ^{34}S) in addition to δ^{13}C and δ^{15}N in further analysis could provide a better understanding of Hg variations as δ^{34}S has been described as a reliable trophic marker to predict Hg concentrations in six seabird species from the Pacific Canada.[77]

Whereas body mass does not follow a particular trend, body size shows a slight increase over time. This relationship could also result from a change in the prey community as suggested by a previous study.[78] For instance, kittiwake chicks from a Norwegian colony grew faster in years with a higher proportion of sandeel *Ammodytes* spp. in their diet,[78] and they were therefore larger at fledging.

Temporal trends of Hg in Arctic seabird communities are inconsistent. Mercury time series obtained in Canadian Arctic seabirds (eggs and liver) from the 1970s to the 2010s show an increase, followed by a plateauing around 2000 and then a decline.[79,80] In contrast, an increase in feather Hg concentrations has been observed for little auks from East Greenland,[81] for ivory gulls *Pagophila eburnea* from the Canadian Arctic and western Greenland,[20] and in eggs of northern fulmars *Fulmarus glacialis* and Brunnich's guillemot *Uria lomvia* from the Canadian High Arctic[21] for the periods of 2007−2014, 1877−2007, and 1975−2014, respectively. These discrepancies likely result from Hg integration time in the sampled tissue[82,83] and differences in regional Hg contamination.

Environmental Variables Explaining the Hg Concentration in Kittiwakes. Among the environmental variables selected to explain Hg concentrations in kittiwakes, Chl *a* concentrations were the strongest predictor. Extreme Chl *a* concentrations (i.e., lowest and highest events) were associated with higher Hg exposure in kittiwakes. Chl *a* concentrations remain a reliable predictor of phytoplankton biomass and, thus, primary productivity in open water.[84] New primary production is directly available to mesozooplankton, which in turn supports production of upper trophic levels.[36] This suggests that extremely high productivity and extremely low productivity would result in high Hg concentrations in kittiwakes. Interestingly, Chl *a* and water temperature are strong predictors of shifts in fish communities.[85−87] For example, in the Bering and Chukchi seas, the distributions and community composition of pelagic species were dependent of temperature and chlorophyll concentrations.[85] Polar cod *Boreogadus saida* were the most abundant in the warm Alaska Coastal Water of the central Chukchi Sea, characterized by low chlorophyll, low nutrients, and small zooplankton taxa. In contrast, adult Pacific herring *Clupea pallasii* were more abundant in the less stratified Bering Strait waters and in the colder Bering Shelf Water of the northern Bering and southern Chukchi seas, characterized by high chlorophyll, high nutrients, and larger zooplankton taxa. Among the seven selected environment-related variables, Chl *a* was the best predictor of the trophic level and carbon source in kittiwakes' blood; this confirms the fact that Chl *a* could be a reliable indicator of fish community shifts.

Kittiwakes are generalist species that can shift the trophic level according to environmental conditions,[88] and a shift in the trophic level has been related to a modification of Hg exposure in other Arctic seabirds *U. lomvia*.[6] As previously mentioned, since 2005, Kongsfjorden kittiwakes have been shifting their polar cod-dominated diet to a more generalist diet composed of species from sub-Arctic and Atlantic origin, such as capelin (*Mallotus villosus*), Atlantic herring (*Clupea harengus*), young Atlantic cod (*Gadus morhua*), and haddock (*Melanogrammus aeglefinus*).[31] Sub-Arctic and Atlantic pelagic fish have higher temperature tolerances and generally benefit from warmer water temperatures.[89] Interestingly, Hg concentrations in polar cod muscles sampled in the east of the Barents Sea were 2.8-fold higher than in capelin, whereas in Atlantic cod, Hg concentrations were 3.2-fold higher than in capelin.[90] Similar results were reported in the Canadian Arctic, with total Hg in polar cods that was higher than in capelin.[91] From 2013 to 2019, the occurrence of Atlantic herring in Kongsfjorden kittiwake diet has often been above 20% (I. Eulaers and G.W. Gabrielsen, personal communication), and Hg concentrations are similar between polar cod and herring.[92] Consequently, a progressive replacement of polar cod to capelin and then capelin to other more Hg-contaminated species such as Atlantic cod juveniles or Atlantic herrings in Kongsfjorden kittiwake diet could partly explain the U-shaped relationship in kittiwakes' blood Hg concentrations from 2000 to 2019.

Sea-ice extent, which was the strongest predictor of nondetrended Hg concentrations in kittiwakes, plays a large role in the accumulation of GEM and acts as a barrier for GEM air-sea exchange; thus, it has been proposed that melting sea-ice in a warming climate would lead to increased GEM evasion, resulting in decreased Hg levels in the Arctic Ocean's surface.[93] In a recent study, Schartup and colleagues[94] observed that first-year sea-ice contains more MeHg than multiyear sea-ice and calculated that the ongoing shift from an old multiyear ice to a first-year ice-dominated Arctic Ocean may cancel out the previously expected decline in MeHg contributions from sea-ice.[94] Therefore, although a reduction in sea-ice extent would accelerate GEM evasion and sunlight-induced breakdown of MeHg in surface waters,[95] these mechanisms could be counterbalanced by the release of MeHg from young sea-ice.

In a study on two other seabird species, the thick-billed murre and the northern fulmar in the Canadian High Arctic, a 40 year time series showed that NAO and surface air temperature were the strongest predictors of Hg concentrations in eggs.[21] Snowfall and sea-ice concentrations were also strong predictors for murres and fulmars, respectively; however, these relationships were only observed after time lags between 4 (NAO, summer temperature) and 7 years (sea-ice concentrations). It is interesting to see that, in Svalbard, environmental conditions in the preceding spring can immediately influence blood Hg concentrations in kittiwakes. This could result from the labile characteristic of blood and the numerous biogeochemical exchanges that occur on the western coast of Spitsbergen. The area around Spitsbergen and the Fram Strait is one of the most climatically sensitive areas in the world.[3,96] Therefore, in comparison to the high Canadian Arctic, Hg exchanges between the environment and biota could be more dynamic and thus accelerated.

We have actually no evidence that those increased concentrations of Hg in kittiwake blood will result in deleterious effects on the colonies' size. In more recent years, some colonies seem to have recovered from the decline observed between 1970 and 2015.[97] Atlantification or borealization processes in some feeding habitats may be offering a rather favorable environment to kittiwakes,[97] and the toxic effects of Hg could be counterbalanced by a higher nutritive value of Atlantic prey.

To conclude, from 2000 to 2013, blood Hg in breeding kittiwakes from a Svalbard colony decreased by 3% per year, and from 2014 to 2019, blood Hg increased by 11% per year. Sex was the strongest biological predictor of Hg concentrations in kittiwakes, whereas the strongest environmental predictor of Hg in kittiwakes was Chl *a*, with extreme values associated to high Hg concentrations. Chl *a* and sea-ice extent are reliable predictors of fish communities. The blood Hg U-shaped trend is likely due to a shift from polar cod to Atlantic fish species in kittiwake diet, likely added in the later years to the input of MeHg from Arctic shrinking sea ice. Moreover, our results raise concern for Hg contamination in the wintering ground of kittiwakes because the concentrations measured in pre-laying birds have been associated to decreased breeding probability and disrupted parental care in this species.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c07633.

> Detailed methods for fieldwork and blood sampling, molecular sexing, mercury and stable isotope analyses, ocean acidification indicators, and statistical analyses; Table S1: overview of the samples available per year and per breeding stages; Table S2: overview of environmental factors used to predict blood Hg concentrations in black-legged kittiwakes from Kongsfjorden; Table S3: relationships between morphometrics and blood Hg concentrations according to sex and the breeding period; Table S4: outputs from GAM models explaining Hg, stable isotopes, body mass, and skull length temporal variation (2000−2019) in chick-rearing kittiwakes from

Kongsfjorden; Table S5: effects of environmental variables on Hg concentrations in Svalbard chick-rearing kittiwakes (n = 38); Table S6: relationship between nondetrended blood Hg concentrations in chick-rearing kittiwakes and environmental factors (n = 38); Figure S1: temporal variations of climate-related variables measured in spring in Svalbard or the Fram Strait; Figure S2: blood Hg variations across the breeding cycle in kittiwakes captured several times; Figure S3: diagnostic plots for the five GAM models investigating the biological variables that better explain temporal trends of Hg in kittiwakes; Figure S4: diagnostic plots for GAM models investigating the temporal trends of stable isotopes and biometrics in kittiwakes (PDF)

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Notes

The authors declare no competing financial interest.
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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