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A U-turn for mercury concentrations over 20 years: how do environmental conditions affect exposure in Arctic seabirds?

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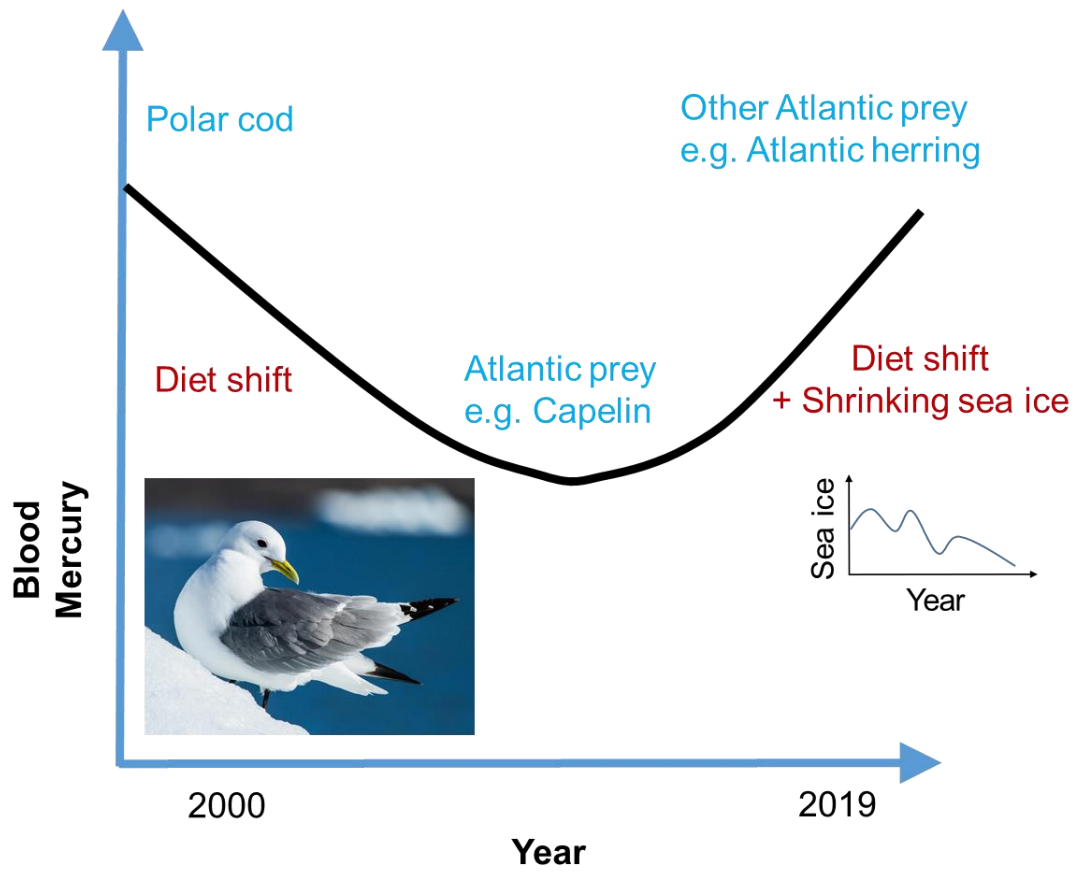
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Data sharing policy

The data that support the findings of this study are available from the corresponding author upon reasonable request.

TOC Art



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. 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* 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 of Hg concentrations until 2013. Then, a new shift in 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 rise since 2014 raises concern for Arctic seabirds.

Key-words: Global changes; Trace elements; Temporal trends; *Rissa tridactyla*; Chlorophyll *a*; Sea-ice extent; Feeding ecology

Synopsis: Prey community shifts and MeHg release from young sea-ice are counterbalancing the previously observed decline of Hg in kittiwakes.

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, sea-ice thickness and 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 period³. 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 multi-year sea-ice or millennia-old ice shelves, are at risk and some are vanishing³. 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 regions⁷. 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 methylation⁴. Methylmercury (MeHg), which is known to be the most toxic among the Hg compounds, is generated when inorganic Hg is dissolved into water⁸. While bacteria methylation is considered as the major pathway⁹, chemical reactions yielded by environmental factors such as pH and temperature contribute to make MeHg more

bioavailable in aquatic ecosystems¹⁰. 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. One of the most important degradation processes of MeHg in ocean surfaces is photo-induced demethylation. MeHg decomposes under ultraviolet and visible light¹⁴. Therefore, a reduced sea-ice extent will expose a larger ocean surface and could enhance Hg photo-demethylation. The transfers and transformations of Hg in Arctic aquatic systems are poorly understood and need more research¹⁵. Hg concentrations biomagnify throughout the food webs, with higher concentrations in higher trophic level species¹⁶. 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 uncertain¹². 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 period²². Since the 1990s, Hg exposure followed a non-linear increase in some mammals of Svalbard and have 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 kittiwake *Rissa tridactyla* (hereafter 'kittiwakes'), are generalist top predators, and can be useful indicators of Hg contamination in their habitat²³. Between 1975 and 2010, at the exception of a few²⁴, kittiwake colonies have been declining throughout their breeding range, and this coincided with an abrupt warming of sea-surface temperature in the 1990s²⁵. 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 size²⁵ may be aggravated by Hg-related breeding failure³⁰.

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 kittiwakes diet³¹, 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 non-linear increase of Hg in Svalbard kittiwakes. In addition, since fish community modifications in Kongsfjorden were driven by sea-ice extent³¹, we predict that 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 details in the **supplementary information**. Briefly, from 2000 to 2019 (excluding 2010), 1152 kittiwakes were captured during different phases of the breeding cycle in Kongsfjorden, Svalbard (78°54'N, 12°13'E), 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 calliper. Birds were captured during the breeding period, which can be separated into three stages: the pre-laying, the egg-incubation, and the chick-rearing phases. Temporal trends of total blood Hg (hence 'Hg') and associations with climate-related variables were analysed in chick-rearing kittiwakes only (n = 392), in order 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 **supplementary 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 months³²⁻³⁴, see **supplementary information** for more details. Thus, sampling conducted during the chick-rearing stage represents the feeding ecology within the two previous months (May-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*) as a proxy of primary productivity and a useful indicator of fisheries production at both regional and global scales^{35,36}, and precipitation. We also analysed the relationships between Hg concentrations and ocean acidification indicators such as pH and

aragonite saturation at the bottom of the water column (see **supplementary 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 in order 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 variables³⁷. 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 methods in details in addition to results conducted on non-detrended data in the **supplementary information**.

We first tested Hg and stable isotopes variations (detrended variables) across the breeding cycle by using linear mixed-effect models (LMMs) with breeding period and sex as independent categorical 3- and 2-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 SMI³⁸ 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 \times sex interactions) by using the ‘emmeans’ package³⁹.

Second, to investigate the temporal trends of Hg and other biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, body mass and skull size) in chick-rearing kittiwakes, over a 20-year period, we used GAMMs on non-detrended data to account for non-linear relationships between response variables and predictors, and for recaptured individuals. Year, $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ and sex were used as continuous predictors, and bird identity as a random factor to analyse temporal trends of Hg, whereas models explaining the temporal trends of $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, 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' modelling technique^{40,41} detailed in **supplementary information**.

Third, we analysed 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, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values for each sex, and performed statistical analyses on this subset of n=38 sampling units. Since Hg trends in biota often present non-linear variations⁴²⁻⁴⁴ and have been increasing non-linearly in Svalbard vertebrates^{18,19}, we tested both linear (e.g. sea-ice extent) and quadratic (e.g. sea-ice extent + sea-ice extent²) 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 analysed separately using LMs. Finally, we tested which of the strongest

climate-related variables would better explain feeding habits ($\delta^{13}\text{C}$ or $\delta^{15}\text{N}$), to do so we used LMs with detrended variables.

All models were ranked using AIC⁴⁷. 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 $e^{(-0.5(\Delta\text{AICc}))}$ (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. Model fit was assessed by using residual diagnostic plots. All analyses and figures were performed using R software v.3.6.3⁴⁸ with the following packages: car, mgcv, nlme, effects, ggplot2 and corrrplot^{49–54}. Data sharing policy is detailed in **supplementary information**.

RESULTS

Biological variables influencing individual Hg concentrations

From 2000 to 2019, Hg was analysed in 1152 kittiwake blood samples. Among these birds, biometrics (skull and body mass) were available for 1100 birds and stable isotopes were analysed in 508 birds (**Table S1**). Mercury concentrations decreased by 55.7% across the breeding cycle, ranging from 1.97 ± 0.03 $\mu\text{g/g dw}$ in pre-laying kittiwakes to 1.09 ± 0.02 $\mu\text{g/g dw}$ (values represent raw mean \pm standard error concentrations) in chick-rearing kittiwakes (**Figure 1A, 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, Table 1A**). Blood Hg concentrations in males were higher by 0.22 $\mu\text{g/g}$ (0.04; 0.39) during the pre-laying period, 0.47 $\mu\text{g/g}$ (0.33; 0.66) during the incubation period and 0.37 $\mu\text{g/g}$ (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 analysed in a large enough number of individuals for each period, we analysed $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ variations across the breeding cycle (**Figure 1B**). Stable isotope values in incubating and chick-rearing birds were lower in comparison to values in pre-laying birds (**Table 1B, Figure 1B**). In 2011, stable isotopes were not related to sex at any stage of the breeding season (males vs females, $\delta^{15}\text{N}$: pre-laying period: -0.09 (-0.54; 0.36), incubation: 0.32 (-0.21; 0.85), chick-rearing 0.29 (-0.29; 0.86); $\delta^{13}\text{C}$: pre-laying period: 0.08 (-0.14; 0.31), incubation: -0.0003 (-0.25; 0.25), chick-rearing -0.15 (-0.42; 0.12), EMMs estimates and 95% CI).

When analysing the effects of sex, body metrics and condition on kittiwake's Hg concentrations, sex over morphometrics explained Hg concentrations (**Table 1**), 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-biometrics relationships according to sex and 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 $\delta^{15}\text{N}$ (**Figure 2, Table 2**). 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, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ presented oscillating variations over time (**Figure 3A, C**). Overall, $\delta^{13}\text{C}$ values presented a decrease over time (LMM: -0.019 (-0.023; -0.010)), mostly related to $\delta^{13}\text{C}$ values which remained low after 2015 (**Figure 3A**), whereas $\delta^{15}\text{N}$ 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 overtime (LMM: 0.15 (-0.38; 0.68), **Figure 3E**), skull length overall increased by 7% per year (LMM: 0.07 (0.03; 0.10)) (**Figure 3D**). For Hg, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, 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, S4**.

Environmental variables explaining Hg concentration in kittiwakes

Among climate-related variables, we show the ten most competitive models in **Table S5** for both detrended and raw data. The best predictor of detrended Hg concentrations was Chl *a* (**Table 3**). Sea-ice extent was the top model when considering non-detrended variables (**Table S5, 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, 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 2000-2013, 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 in comparison to body size or body condition. Hg was higher in males than in females and declined through the breeding season. At the ecosystem level, Chl *a* concentration 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 differences⁵⁵. 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/excretion⁵⁶⁻⁵⁸. An experimental study has reported higher fat mass in birds exposed to higher concentrations of Hg⁵⁹. Moreover, in rodents a greater urinary excretion of Hg has been observed in females⁶⁰ and despite similar Hg intake, males attained a 20–30% significantly higher whole-body retention in males than females⁶¹.

As previously observed in kittiwakes sampled in Kongsfjorden⁶², 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 muscles⁶³. Since Hg concentrations in red blood cells reflect the exposure of the Hg ingested for a period of up to two 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 $\delta^{15}\text{N}$ values, although we cannot exclude a Hg and/or $\delta^{15}\text{N}$ baseline shift between the wintering and the 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^{23,66}. This raises a concern about Hg contamination in North Atlantic waters, the area west of the Mid-Atlantic Ridge, has been estimated as a wintering area to 80% of the 4.5 million Atlantic adult kittiwakes⁶⁷ and has already been characterised as a hotspot for Hg contamination^{68,69} with a likely main source of Hg provided from the Arctic ocean⁷⁰.

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 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values around 2013-2015, which could coincide with the rise 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^{31,71}. The increasing presence of these communities coincides with an increase in the

intrusion of Atlantic water with associated Atlantic prey into Svalbard^{71,72}. Periodic arrivals of specific species could result in the yearly oscillations of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values in kittiwake blood. The reason for 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 Hg⁷³⁻⁷⁵. Studying the stable isotopes of Hg could help better track the origin of Hg contamination in kittiwakes^{76,77}. The inclusion of sulphur stable isotopes ($\delta^{34}\text{S}$) in addition to $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in further analysis, could provide a better understanding of Hg variations, as $\delta^{34}\text{S}$ has been described as a reliable trophic marker to predict Hg concentrations in six seabird species from the Pacific Canada⁷⁸.

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 prey community as suggested by a previous study⁷⁹. For instance, kittiwake chicks from a Norwegian colony grew faster in years with a higher proportion of sandeel *Ammodytes* spp. in their diet⁷⁹, 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 then a decline^{80,81}. In contrast, an increase in feather Hg concentrations has been observed for little auks from East Greenland⁸², ivory gulls *Pagophila eburnea* from the Canadian Arctic and western Greenland²⁰, and in eggs of northern fulmars *Fulmarus glacialis* and Brunnich's guillemot *Uria lomvia* from the Canadian High Arctic²¹, for the periods of 2007-2014, 1877-2007 and 1975-2014, respectively. These discrepancies likely result from Hg integration time in the sampled tissue^{83,84} and differences in regional Hg contamination.

Environmental variables explaining Hg concentration in kittiwakes

Among the environmental variables selected to explain Hg concentrations in kittiwakes, Chl *a* concentration was 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-water⁸⁵. New primary production is directly available to mesozooplankton which in turn supports production of upper trophic levels³⁶. 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 communities⁸⁶⁻⁸⁸. For example, in the Bering and Chukchi seas, the distributions and community composition of pelagic species were dependent of temperature and chlorophyll concentrations⁸⁶. Polar cod *Boreogadus saida* were most abundant in 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 five selected environment-related variables, Chl *a* was the best predictor of trophic level and carbon source in kittiwakes' blood, this confirms the fact Chl *a* could be a reliable indicator of fish community shifts.

Kittiwakes are generalist species that can shift trophic level according to environmental conditions⁸⁹ and a shift in trophic level has been related to a modification of Hg exposure in other Arctic seabirds *Uria lomvia*⁶. As previously mentioned, since 2005, Kongsfjorden kittiwakes have been shifting their polar cod dominated diet by 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 temperatures⁹⁰. 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 capelin⁹¹. Similar results were reported in the Canadian Arctic, with total Hg in polar cods that were higher than in capelin⁹². From 2013 to 2019, the occurrence of Atlantic herring in Kongsfjorden kittiwake's diet has often been above 20% (Eulaers I., Gabrielsen G.W. et al., personal communication), and Hg concentrations are similar between polar cod and herring⁹³. Consequently, a progressive replacement of polar cod to capelin, then capelin to other more Hg contaminated species such as Atlantic cod juveniles or Atlantic herrings in Kongsfjorden kittiwakes diet could partly explain the U-shaped relationship in kittiwakes' blood Hg concentrations from 2000 to 2019.

Sea-ice, which was the strongest predictor of non-detrended 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 surface Arctic Ocean⁹⁴. In a recent study, Schartup and colleagues⁹⁵ observed that first-year ice contains more MeHg than multi-year ice and calculated that the ongoing shift from an old multi-year ice to a first-year ice dominated Arctic Ocean may cancel out the previously expected decline in MeHg contributions from sea-ice⁹⁵. Therefore, although a reduction in sea-ice extent would accelerate GEM evasion and sunlight-induced breakdown of MeHg in surface waters⁹⁶, 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 eggs²¹. Snowfall and sea-ice concentrations were also strong predictors for murre and fulmars, respectively, however these relationships

were only observed after time lags between four (NAO, summer temperature) and seven 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 Fram Strait is one of the most climatically sensitive areas in the world^{3,97}. 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 colonies' size. In more recent years some colonies seem to have recovered from the decline observed between 1970 and 2015⁹⁸. Atlantification or borealization processes in some feeding habitats may be offering a rather favourable environment to kittiwakes⁹⁸ 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*, but also 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.

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Table 1. Biological factors influencing blood Hg concentrations in black-legged kittiwakes from Kongsfjorden Svalbard. 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.

A) Hg and stable isotopes variation according to breeding phase							
Response variable	Dataset	Intercept		Incubation		Chick-rearing	
Hg ($\mu\text{g/g dw}$)	All individuals	0.56 (0.50; 0.63)		-0.40 (-0.46; -0.33)		-0.79 (-0.86; -0.72)	
	Subset only with	0.53 (0.45; 0.61)		-0.44 (-0.52; -0.36)		-0.85 (-0.93; -0.77)	
$\delta^{13}\text{C}$ (‰)	2011	0.07 (0.006; 0.150)		-0.28 (-0.38; -0.17)		-0.40 (-0.51; -0.29)	
$\delta^{15}\text{N}$ (‰)	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	AIC	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)
<i>Null model</i>	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)
SCM: size corrected mass; SMI: scaled mass index							

Table 2. Predictors of blood Hg variation in chick-rearing kittiwakes (n=392) from Kongsfjorden, Svalbard (2000-2019). Model selection and model-averaged estimates. The first part of the table shows the different candidate models and their AIC ranking, the most competitive model appears on the first line ($\Delta\text{AIC} = 0$). The second part of the table shows the estimates and 95% confidence intervals derived from GAMMs. Bold values represent significant relationships. GAMMs including year as a smoother, sex, body mass, skull size, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ and sex 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.

<i>AIC selection</i>											
Intercept	s(Year, k=5)	Sex	Mass	Skull	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	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
<i>Conditional model averaged estimates</i>											
Variable	β (95% CI)										
Intercept	0.42 [-2.24; 3.07]										
$\delta^{15}\text{N}$	0.09 [0.04; 0.14]										
Sex	0.29 [0.22; 0.37]										
$\delta^{13}\text{C}$	0.12 [0.04; 0.19]										
Skull	0.05 [0.03; 0.06]										
Mass	0.002 [0.001; 0.003]										
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.											

Table 3. Relationship between detrended blood Hg concentrations in chick-rearing kittiwakes and environmental factors (n=38). The table shows the 5 highest AIC ranked models, the most competitive model appears on the first line ($\Delta AIC = 0$). 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 non-detrended variables are given in the **supplementary information**.

Explanatory variables	AIC ranking					Model averaged estimates β (95% CI)		
	df	logLik	AICc	ΔAIC	weight		Linear	Quadratic
Chl <i>a</i> + Chl <i>a</i> 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 <i>a</i>	-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)
<i>Null model</i>	2	-4.26	12.87	15.28	0	Sea ice extent	-0.13 (-0.59; 0.32)	0.31 (-0.14; 0.76)

Chl *a* : Chlorophyll *a*, GEM: Gaseous Elemental Mercury, Air Temp: Air temperature, Sea Ice: Sea ice extent, SSTa: Sea surface temperature anomaly

Table 4. Relationship between feeding habits and environmental factors in chick-rearing kittiwakes (n=38). The table shows AIC ranked models, the most competitive model appears on the first line ($\Delta AIC = 0$). 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.

Explanatory variables	AIC ranking					Model averaged estimates β (95% CI)		
	df	logLik	AICc	ΔAIC	weight		Linear	Quadratic
Nitrogen stable isotopes								
Chl <i>a</i> + 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)	
<i>Null model</i>	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 <i>a</i> + 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)	
<i>Null model</i>	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)
Chl <i>a</i> : Chlorophyll <i>a</i> , GEM: Gaseous Elemental Mercury, Air Temp: Air temperature, Sea Ice: Sea ice extent, SSTa: Sea surface temperature anomaly.								

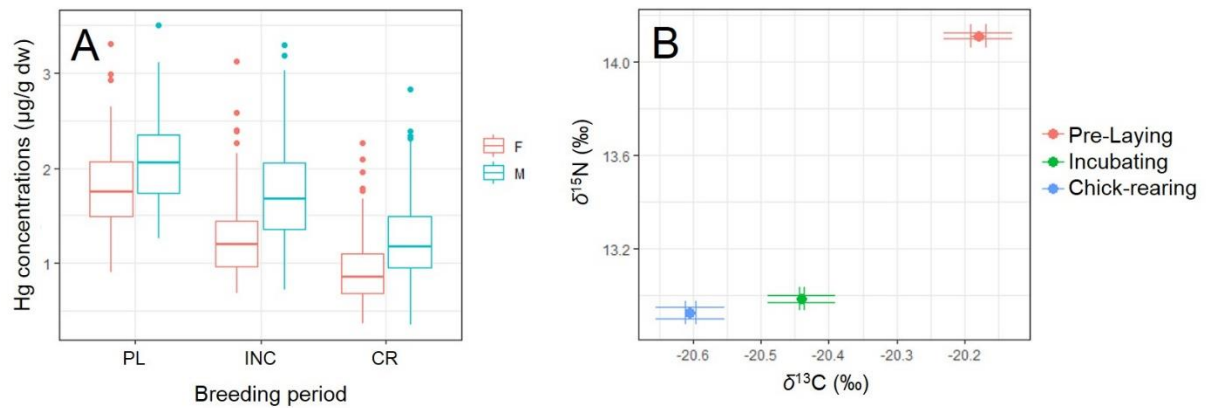


Figure 1. Blood Hg concentrations and stable isotope values across the breeding cycle in black-legged kittiwakes (Kongsfjorden Svalbard). A) Boxplot of non-adjusted Hg concentrations (n=1049, 2000-2016) in males (blue) and females (pink) for each breeding period (PL- Pre-laying, INC- Incubation, CR- Chick-rearing). Median is shown with lower and upper quartiles (box), whiskers ($1.5 \times$ interquartile range from box) and outliers (values beyond whiskers), B) Mean nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) stable isotope values with error bars (SE) in blood samples from black-legged kittiwakes (n=77, 2011) during each breeding period.

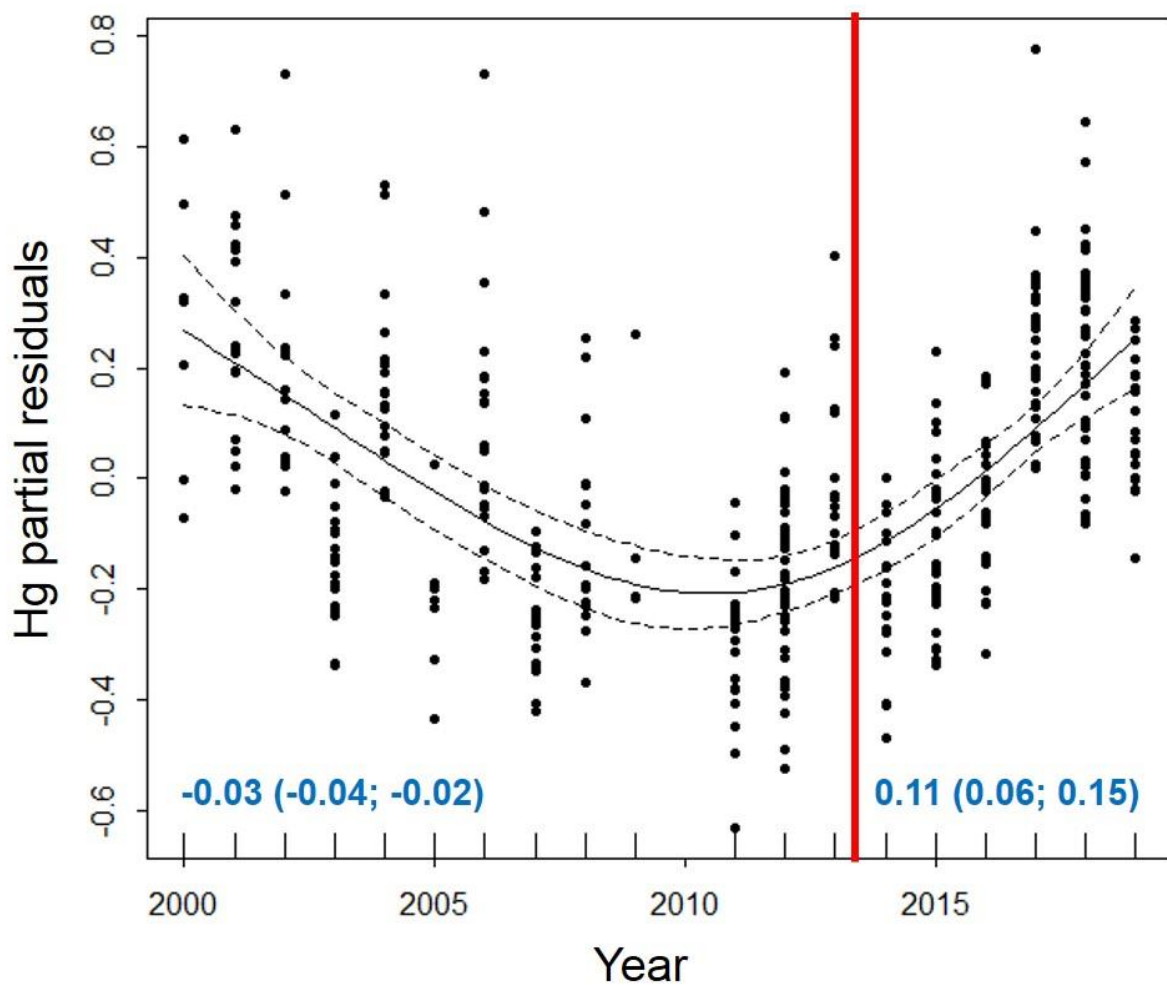


Figure 2. Temporal trends (2000-2019) of blood Hg concentrations in adult chick-rearing kittiwakes (n=392) from Kongsfjorden Svalbard. Hg partial residuals are derived from the best model selected by AIC (Table 2), Hg partial residuals are adjusted for $\delta^{15}\text{N}$, sex as fixed factors, and 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.

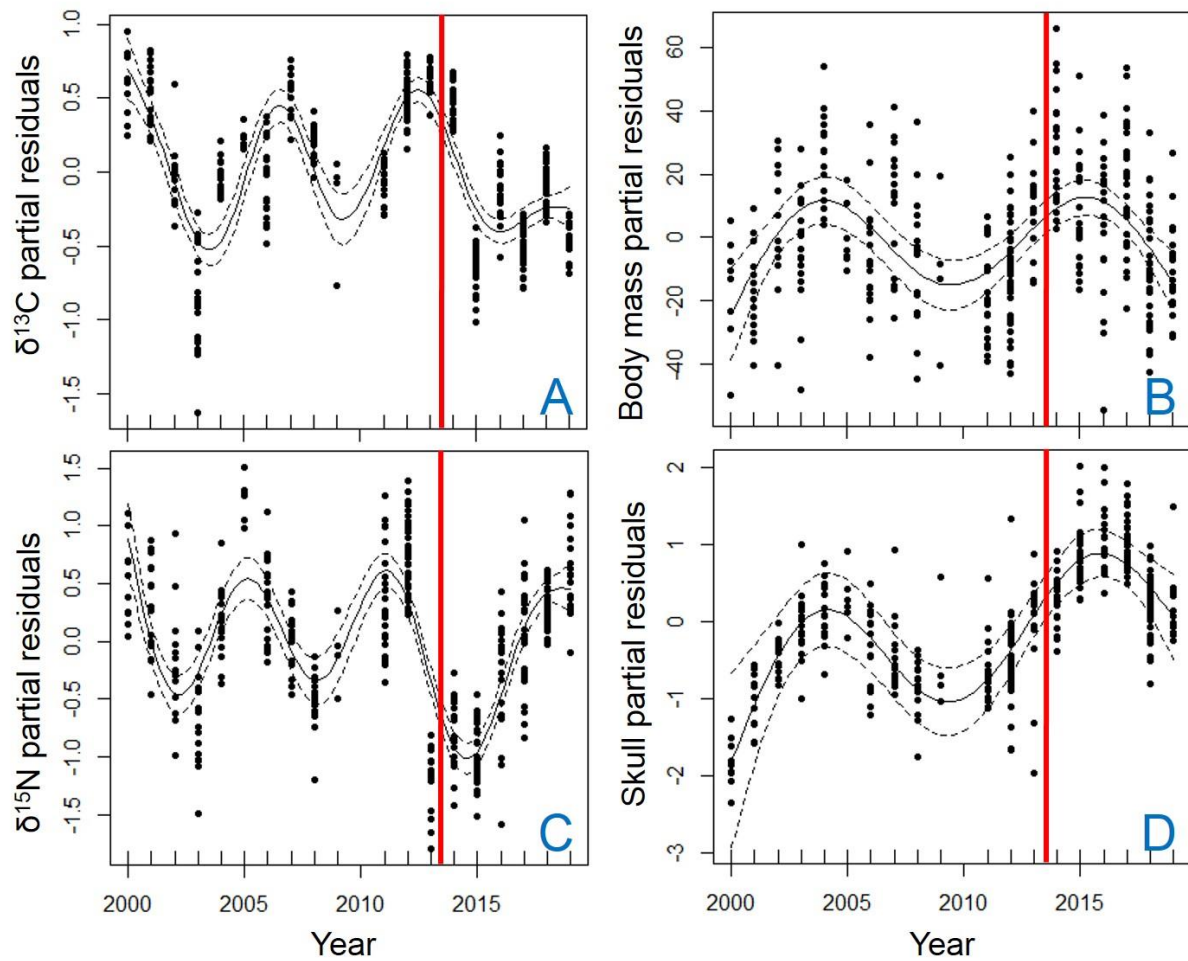


Figure 3. Temporal trends (2000-2019) of stable isotope values in red blood cells and biometrics in adult chick-rearing kittiwakes (n=392) from Kongsfjorden Svalbard. A-C) Feeding habits are inferred from nitrogen and carbon stable isotope values in blood ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, respectively), 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 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 supplementary information (Figure S1).

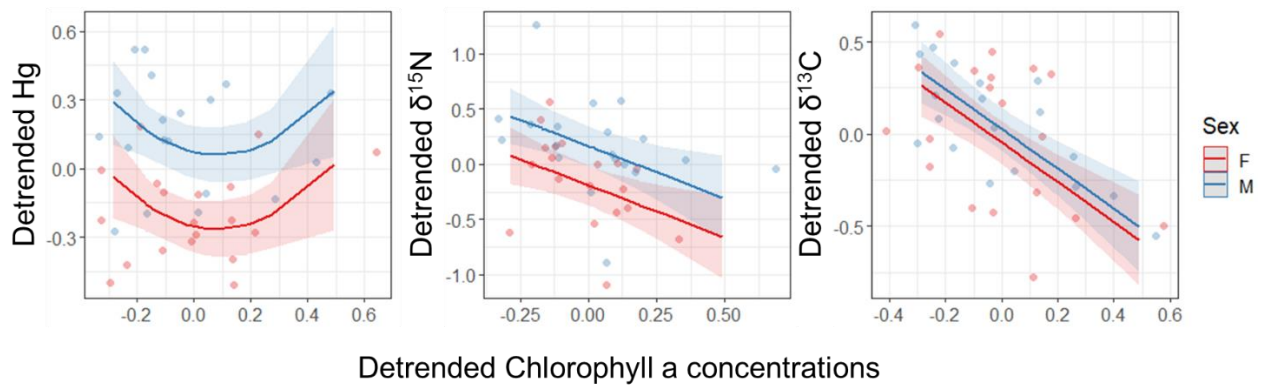


Figure 4. Influence of chlorophyll *a* on blood Hg concentrations and feeding habits in chick-rearing kittiwakes between 2000- 2019, (n=38, Kongsfjorden, Svalbard). Feeding habits are inferred from nitrogen and carbon stable isotope values in blood ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, respectively). 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 in blue.

Supporting information

MATERIAL AND METHODS

Fieldwork and blood sampling

From 2000 to 2016 (excluding 2010), 1049 kittiwakes have been captured during different phases of the breeding cycle at the Krykkjefjellet “main colony” in Kongsfjorden, Svalbard (78°54’N, 12°13’E), 7 km southeast of Ny-Ålesund, Norway. We studied kittiwakes in one plot of around 117 pairs breeding on cliff ledges at heights of 5-10 m. Kittiwakes were caught on their nests with a noose at the end of a 5 m fishing rod. A blood sample (ca. 1 mL) was collected from the alar vein with a 1 mL heparinised syringe and a 25-gauge needle. Kittiwakes were individually marked with metal rings and PVC plastic bands engraved with a three-digit code and fixed to the bird's tarsus for identification from a distance. 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 calliper, biometrics were available for 997 kittiwakes. In several years, birds were captured across the breeding period, which can be separated into three phases: the pre-laying period from end-April to early June which corresponds to the arrival to the colony, nest building and mating; the incubation period lasts from early/mid-June to mid-July and starts when the first egg is laid, then comes the chick-rearing period (~mid-July to mid-August) which starts when the first egg has hatched and lasts until all the chicks have fledged. In several occasions, within a year, individuals were captured at least during two stages (n=17), and a few times during the three stages (n=3), enabling us to study blood Hg dynamic across the breeding cycle. Temporal trends of blood Hg have been analysed in chick-rearing kittiwakes only, in order to reflect local Hg exposure and to make between year comparisons possible.

Mercury and stable isotopes in blood

In birds, MeHg storage from blood into tissues (e.g. muscles) represents 50-95% of the Hg ingested for up to a period of approximately 8 ± 2 weeks^{1,2}. When birds arrive to the breeding ground, their blood Hg concentrations therefore reflect the exposure of the wintering grounds³. The eight weeks prior to the chick-rearing period include the pre-laying period (the courtship and nest building stage), where females forage exclusively inside the fjord, whereas males forage inside and outside the fjord at the Greenland-Svalbard ridge and the Fram Strait⁴. In bird red blood cells, the turnover rate of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values (reflecting feeding habitat and trophic level, respectively) is around 30-50 days^{5,6}, thus sampling conducted during the chick-rearing stage represents the feeding habits between the pre-laying and incubating periods.

Molecular sexing, mercury and stable isotope analyses

According to the planned subsequent analyses, blood samples were centrifuged or not, therefore samples comprised either whole blood or red blood cells, which were stored at -20°C until analyses. Molecular sexing was performed at the Centre d'Etudes Biologiques de Chizé (CEBC) on red blood cells as detailed elsewhere⁷. Prior to Hg and isotopic analyses, red blood cells and whole blood samples (hereafter called blood) were freeze-dried and powdered. We measured total Hg (THg) in 1049 kittiwakes, at Littoral Environnement et Sociétés lab (LIENSs) as previously described⁸. MeHg represents more than 90% of THg in seabirds⁹. We used an Advanced Mercury Analyzer spectrophotometer (Altec AMA 254) with a detection limit of 0.1 ng Hg. At least two aliquots ranging from 5 to 10 mg were analyzed for each samples and quality assessment was measured by repeated analyses (every 10 samples) of certified reference material (CRM) TORT-2 (lobster hepatopancreas, NRCC; certified value 0.27 ± 0.06 $\mu\text{g/g}$). CRM measurements were in good agreement with the certified values and recoveries were $103.7 \pm 0.01\%$. Hg concentrations are expressed in $\mu\text{g/g}$ dry weight (dw). Hg concentrations obtained from whole blood and red blood cells were close to identical ($R^2=0.98$).

Stable isotopes were analysed in 405 kittiwakes. The isotopic niche of kittiwakes was used as a proxy of their trophic niche, as blood cells is a metabolically active tissue that integrates periods of weeks before sampling^{5,10}. The isotopic method was validated in seabirds, with $\delta^{13}\text{C}$ values indicating their foraging habitats^{11,12} and their $\delta^{15}\text{N}$ values increasing with trophic levels¹³. Sub-samples of homogenized blood powder were then weighed (~0.4 mg) with a microbalance and packed in tin containers. Relative abundances of C and N isotopes were determined with a continuous flow mass spectrometer (Thermo Scientific Delta V Advantage) coupled to an elemental analyzer (Thermo Scientific Flash EA 1112). Isotopic results are presented in the δ^* notation relative to Vienna PeeDee Belemnite and atmospheric N_2 for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, respectively. Replicate measurements of internal laboratory standards (acetanilide) indicate measurement errors < 0.10 ‰ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values. The C:N mass ratios of the samples were calculated as the ratio between the mass percentages in carbon and nitrogen. The consistently low C:N values of blood cells verified that low lipid content did not necessitate lipid extraction¹⁴. The isotopic ratio of red blood cells slightly differed from that of whole blood, we therefore applied a correction factor to stable isotope values obtained from whole blood samples (red blood cell $\delta^{13}\text{C} = \text{whole blood } \delta^{13}\text{C} \times 0.985$ and red blood cell $\delta^{15}\text{N} = \text{whole blood } \delta^{15}\text{N} \times 1.004$).

Ocean acidification indicators

Aragonite saturation state is commonly used to track ocean acidification because it is a measure of carbonate ion concentration ($[\text{CO}_3^{2-}]$). At a lower pH, metals that form strong complexes with hydroxide (OH^-) and CO_3^{2-} will be more abundant in their free forms (e.g. Hg^0). These changes in speciation also increase the thermodynamic and kinetic activity of the metals (Millero, 2001a; b). As the pH of seawater decreases, metals, such as Hg that forms strong complexes with OH^- and CO_3^{2-} , will undergo significant changes in speciation^{15–18}. Adsorption of metals into organic material will also change at a lower pH. Ocean acidification measures

(pH and aragonite saturation) were measured in July in Kongsfjorden and were not available from 2000-2012¹⁹. Since pH and aragonite saturation were measured in July and the Hg concentrations in blood referred to bird exposure since May, we associated ocean acidification measures from year n-1 to blood Hg concentrations from year n, to avoid testing predictors obtained '*a posteriori*' on our response variable.

Statistical analyses

First, we investigated which biological factors (i.e. sex, body-size/-condition, or feeding habits) would influence individual Hg concentrations. To do so, we analysed the variations of Hg across the breeding cycle (pre-laying, incubation, and chick-rearing periods) and its interaction with sex, by using linear mixed-effect models (LMMs) with breeding period and sex as independent categorical 3- and 2-level predictors and bird identity as a random factor to explain detrended blood Hg concentrations. We obtained estimated marginal means (EMMs) for specified factor combinations of the LMM (breeding period \times sex interactions) by using the 'emmeans' package²⁰. To confirm the obtained trend, we analysed repeated sampling of the same individuals across the breeding cycle in a subset of the largest dataset, by including only individuals that had been captured at least twice in the same year. In addition, we analysed changes in feeding habits across the breeding cycle. To do so, we used LMMs with $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (proxies of feeding habitat and trophic level, respectively) as response variables, breeding period as a categorical 3-level predictor, and bird identity as a random factor. Sufficient data for each period of the breeding cycle were only available in 2011.

Then, we tested whether body measurements, could predict Hg concentrations in kittiwakes, using LMMs with bird identity as a random factor. Mercury concentration was set as the response variable and body mass, size corrected mass SCM (i.e. residuals from the mass ~ skull size regression), scaled mass index SMI²¹ and skull size were set as predictors in different

candidate models because most predictors were correlated. We ranked the candidate models using Akaike Information criterion (AIC)²², the best model (lowest AIC) was considered as the most reliable biological predictor of detrended Hg concentrations.

Second, to investigate the temporal trends of Hg and other biological variables in chick-rearing kittiwakes, over a 20-year period, we used GAMMs on raw data to account for non-linear relationships between response variables and predictors, and for recaptured individuals. Year, $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ and sex were used as continuous predictors, and bird identity as a random factor to analyse temporal trends of Hg. Since Hg concentrations presented non-linear variations according to year, we entered year as a smoothed term ($k=4$) using penalized regression splines and a Gaussian distribution²³. We used an identical approach (GAMM) to study the temporal trends of $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, body mass and skull size, with year and sex as continuous predictors, and bird identity as a random factor. We adapted the smoothed term to avoid overfitting. We ranked the candidate models using Akaike Information criterion (AIC)²², the best model (lowest AIC) was considered as the most reliable biological predictor of raw Hg concentrations. Break-points were identified by using a Bayesian approach ('broken stick' modelling technique^{24,25}, based on beta regression analysis with 'betareg' package in R²⁶. This method infers the 'year' where blood Hg concentration starts to translate statistically as a change-point in the distribution of blood Hg concentration versus year. To estimate the year of the change-point T_i , in blood Hg concentrations, we used a profile likelihood approach where the likelihood was computed for each year. The value of T_i that maximized the likelihood was thus evaluated, and an approximate confidence interval for T_i was computed with a Likelihood Ratio Test with 1 df.

To avoid pseudoreplication due to the large, and sometimes unbalanced, number of kittiwakes caught per year, for each year, we calculated the average Hg concentrations, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values for each sex, and performed statistical analyses on this subset of $n=38$. We tested both linear (e.g. sea ice extent) and quadratic (e.g. sea ice extent + sea ice extent²) relationships using

linear models (LMs), since Hg trends in biota often present non-linear variations^{40–42} and have been increasing non-linearly in Svalbard vertebrates^{17,18}. 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 show the ten most competitive models in **Table S3** for both detrended and raw data. 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^{43,44}. Variables with missing years (pH and aragonite saturation at the bottom of the water column) were analysed separately using LMs. Finally, we tested which of the strongest climate-related variables would better explain feeding habits ($\delta^{13}\text{C}$ or $\delta^{15}\text{N}$), to do so we used LMs with detrended variables.

All models were ranked using AIC⁴⁵. We obtained the number of parameters (K), 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 $e^{(-0.5(\Delta\text{AICc}))}$ (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. Model fit was assessed by using residual diagnostic plots. All analyses and figures were performed using R software v.3.6.3⁴⁶ with the following packages: car, mgcv, nlme, effects, ggplot2 and corrplot^{47–52}.

Data sharing policy

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Table S1. Overview of the samples available per year and per breeding stages. Values are means and standard deviation for blood Hg concentrations ($\mu\text{g/g dw}$), nitrogen $\delta^{15}\text{N}$ and carbon $\delta^{13}\text{C}$ stable isotope values (‰), body mass (g) and skull length (mm). For biometrics, in some years the number of kittiwakes weighed differed from that of measured, in that case the first number (N1) in (N1/N2) from the biometrics column represents the number of kittiwakes weighed and N2 the number of kittiwakes measured. Non-available data = na. All samples from year 2010 were missing.

Year	Breeding period	Mercury ($\mu\text{g/g dw}$)			$\delta^{15}\text{N}$ (‰) $\delta^{13}\text{C}$ (‰)					Biometrics		Body mass (g)				Skull length (mm)			
		Both sex			Both sex					Females	Males	Females		Males		Females		Males	
		N	Mean	SD	N	Mean	SD	Mean	SD	N	N	Mean	SD	Mean	SD	Mean	SD	Mean	SD
2000	Chick-rearing	17	1.51	0.50	17	13.0	0.4	-20.0	0.2	10	7	367	27.9	403	11.5	87.4	1.9	93.0	2.7
2001	Chick-rearing	30	1.59	0.32	19	13.1	0.5	-20.0	0.2	13/15	13/15	347	21.3	384	32.9	88.4	2.6	94.4	1.7
2002	Chick-rearing	28	1.24	0.37	20	12.4	0.6	-20.6	0.2	15/10	13/9	368	25.6	408	29.8	88.7	1.3	93.4	1.5
2003	Chick-rearing	20	0.78	0.25	20	11.9	0.6	-21.5	0.4	10	10	357	18.9	379	44.2	88.9	1.1	92.7	1.9
2004	Chick-rearing	30	1.44	0.37	20	12.8	0.3	-20.5	0.1	15	15/14	392	24.2	425	22.2	89.5	1.5	94.0	2.1
2005	Chick-rearing	31	0.99	0.24	7	14.0	0.3	-20.2	0.1	11/7	20/16	353	15.1	407	22.4	90.6	1.6	94.2	1.5
2006	Chick-rearing	29	1.38	0.42	20	12.9	0.6	-20.7	0.3	13	16/15	347	29.9	383	17.4	88.5	1.1	93.6	1.5
2007	Chick-rearing	30	0.86	0.21	20	12.7	0.3	-20.0	0.2	15/13	15/13	381	28.8	432	25.9	89.4	1.9	93.2	1.8
2008	Pre-laying	69	2.03	0.44	0	na	na	na	na	31/28	38/34	396	31.7	423	32.1	88.9	1.2	93.6	2.3
2008	Incubation	38	1.54	0.46	20	11.8	0.5	-20.3	0.1	15	23/22	395	16.4	440	27.7	89.7	1.5	93.3	1.9
2008	Chick-rearing	34	0.90	0.31	16	12.1	0.4	-20.3	0.2	20/18	14/13	354	16.8	411	22.4	89.2	1.5	93.0	1.9
2009	Pre-laying	46	2.18	0.51	0	na	na	na	na	20/15	26/21	388	36.6	427	31.6	89.2	1.9	94.0	1.9
2009	Incubation	2	2.00	0.60	0	na	na	na	na	na	na	na	na	na	na	na	na	na	na
2009	Chick-rearing	17	0.96	0.45	10	12.5	0.6	-20.9	0.5	9	8	346	17.5	385	24.8	89.3	2.0	94.2	1.9
2010	NA	0	na	na	0	na	na	na	na	na	na	na	na	na	na	na	na	na	na
2011	Pre-laying	93	1.85	0.46	30	14.1	0.3	-20.2	0.3	39	53/54	394	36.3	413	25.6	88.8	1.8	93.9	1.7
2011	Incubation	52	1.58	0.62	28	13.0	0.4	-20.4	0.1	24	29	382	25.6	423	28.1	89.1	1.8	93.0	2.1
2011	Chick-rearing	34	0.87	0.24	22	12.9	0.6	-20.6	0.2	18	16	338	18.2	376	24.8	88.5	1.7	92.8	1.9
2012	Pre-laying	3	1.80	0.30	0	na	na	na	na	na	na	na	na	na	na	na	na	na	na
2012	Incubation	102	1.52	0.51	0	na	na	na	na	51	51	384	22.3	429	28.0	88.0	1.9	92.9	1.8
2012	Chick-rearing	51	1.09	0.37	43	13.4	0.5	-20.0	0.2	26	25	348	25.8	394	22.8	88.1	1.4	93.1	1.5
2013	Incubation	80	1.39	0.34	2	11.5	0.9	-20.1	0.2	23	57	385	20.3	429	25.2	88.7	1.3	93.7	1.7
2013	Chick-rearing	53	1.22	0.33	18	11.3	0.4	-19.9	0.1	11	42/41	374	27.3	414	27.2	87.3	1.6	93.7	1.9
2014	Incubation	4	1.49	0.43	0	na	na	na	na	2	2	388	10.6	453	38.9	87.9	0.1	94.5	0.1
2014	Chick-rearing	34	0.77	0.31	20	11.9	0.4	-20.0	0.1	15	19	402	19.5	451	27.1	89.4	2.1	93.7	1.6
2015	Incubation	68	1.69	0.58	0	na	na	na	na	34	34	378	25.2	409	17.2	89.0	1.7	93.8	1.8
2015	Chick-rearing	31	0.88	0.33	31	11.7	0.3	-21.3	0.2	16	15	371	19.7	402	31.0	90.0	2.4	94.3	1.7
2016	Pre-laying	0	na	na	0	na	na	na	na	na	50	na	na	417	22.8	na	na	93.9	1.6
2016	Incubation	0	na	na	0	na	na	na	na	24	18	380	24.2	412	18.5	88.6	2.2	93.2	2.1
2016	Chick-rearing	22	1.04	0.29	22	12.4	0.6	-20.6	0.2	11	11	360	25.4	409	21.3	90.7	1.2	95.1	1.8
2017	Chick-rearing	33	1.45	0.32	33	12.7	0.6	-21.1	0.2	22	15	378	27.6	433	31.5	90.6	1.2	95.3	1.7
2018	Chick-rearing	46	1.48	0.35	46	12.9	0.1	-20.6	0.1	23/22	24	351	20.3	386	18.6	89.1	1.4	93.3	1.5
2019	Chick-rearing	24	1.16	0.27	24	13.2	0.5	-21.0	0.1	10	9	353	13.2	394	27.4	89.9	1.0	94.1	2.0

Table S2. Overview of environmental factors used to predict blood Hg concentrations in black-legged kittiwakes from Kongsfjorden. To allow the different factors to be representative of Hg in blood in July/August we averaged monthly values from April, May and June named under “Spring average”. Fram Strait*: data were calculated from a box delimited by latitudes 70°N and 82°N and longitudes 20°W and 15°E.

Environmental variable	Period	Area	Missing year(s)	Unit	Distance to colony (km)	Data supplier	Source
North-Atlantic Oscillation	Spring average	Global		none		UEA	https://crudata.uea.ac.uk/
Arctic Oscillation	Spring average	Arctic		none		NOAA	https://www.cpc.ncep.noaa.gov
Sea Ice extent	April	Fram Strait		10 ³ km ²	346	NPI	mosj.no
Chlorophyll a concentration	Spring average	Fram Strait*		mg/m ³	346	NASA/GSFC/OBPG	oceandata.sci.gsfc.nasa.gov
Sea surface temperature anomaly	Spring average	Fram Strait*		a.u.	346	LDEO	http://iridl.ldeo.columbia.edu/
Air temperature	Spring average	Svalbard (Ny-Ålesund)		°C	7.1	MET Norway	https://seklima.met.no/
Precipitation	Spring average	Svalbard (Ny-Ålesund)		mm	7.1	MET Norway	https://seklima.met.no/
Gaseous Elemental Hg	Spring average	Svalbard (Zeppelin mountain)		ng/m ³	7.1	NILU	NILU ebas
pH	July (year n-1)	Kongsfjorden (KB3)	2000-2012	none	4.5	NPI/IMR	Fransson and Chierici, unpublished
Aragonite Saturation	July (year n-1)	Kongsfjorden (KB3)	2000-2012	none	4.5	NPI/IMR	Fransson and Chierici, unpublished

Table S3. Relationships between morphometrics and blood Hg concentrations according to sex and breeding period. Values are estimates and 95% confidence intervals obtained from LMMs with bird identity as a random factor.

	Pre-laying	Incubating	Chick-rearing
<i>Females</i>			
Body mass	-0.003 (-0.006; 0.001)	-0.006 (-0.009; -0.003)	0.002 (0.001; 0.004)
Skull length	0.015 (-0.053; 0.083)	0.023 (-0.018; 0.065)	0.011 (-0.018; 0.04)
SCM	-0.003 (-0.0063; 0.0003)	-0.006 (-0.009; -0.003)	0.002 (0.001; 0.004)
SMI	-0.0005 (-0.002; 0.001)	-0.0005 (-0.001; 0.0001)	0.0003 (-0.0002; 0.0007)
<i>Males</i>			
Body mass	0.002 (-0.001; 0.005)	-0.002 (-0.0049; 0.0004)	0.001 (-0.001; 0.002)
Skull length	0.017 (-0.032; 0.066)	0.031 (-0.011; 0.074)	0.037 (0.007; 0.067)
SCM	0.002 (-0.001; 0.005)	-0.003 (-0.0054; -0.0003)	-0.0002 (-0.0017; 0.0014)
SMI	-0.0001 (-0.001; 0.001)	-0.0004 (-0.0009; 0.0001)	-0.0004 (-0.0008; 0.00004)

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, Svalbard. For intercept and parametric coefficients, values are estimates and standard errors, for smoother (year), values are effective degrees of freedom. Number in bold represent significant relationships, female was the reference level.

Response variable	Intercept	Parametric coefficients				Approximate significance		
		Sex	p-value	$\delta^{15}\text{N}$	p-value	k	s(Year)	p-value
Hg	-0.14 ± 0.3	0.29 ± 0.04	<0.0001	0.09 ± 0.02	0.0002	5	3.256	<0.0001
$\delta^{15}\text{N}$	12.48 ± 0.04	0.29 ± 0.06	<0.0001			8	6.924	<0.0001
$\delta^{13}\text{C}$	-20.56 ± 0.03	0.06 ± 0.04	0.0957			9	7.851	<0.0001
Body mass	363.8 ± 2.16	37.6 ± 3.04	<0.0001			5	3.909	<0.0001
Skull size	89.22 ± 0.14	4.42 ± 0.2	<0.0001			5	3.845	<0.0001

Table S5. Effects of environmental variables on Hg concentrations in Svalbard chick-rearing kittiwakes (n=38). The table shows the ten most competitive linear models (ranked using AIC) among a list of 34 for both raw (upper part) and detrended variables (lower part).

Model	Intercept	Sex	poly (Sea Ice, 2)	poly (Chl a, 2)	poly (SSTA, 2)	poly (Air Temp, 2)	poly (Precip, 2)	poly (GEM, 2)	NAO	Sea Ice	Chl a	SST A	Precip	GEM	df	logLik	AIC	ΔAIC	weight
<i>Raw variables</i>																			
13	0.9776	+	+												5	4.99	0	0	0.995
17	0.9776	+					+								5	-1.89	13.8	13.8	0.001
30	0.6937	+									0.3542				4	-3.48	15	15.0	0.001
5	1.139		+												4	-3.65	15.3	15.3	0
31	0.9621	+										0.3677			4	-3.76	15.5	15.5	0
15	0.9776	+			+										5	-2.96	15.9	15.9	0
33	0.8306	+											0.00166		4	-4.02	16	16.0	0
34	1.906	+												-0.6416	4	-4.16	16.3	16.3	0
14	0.9776	+		+											5	-3.27	16.5	16.5	0
1	0.9776	+													3	-5.42	16.8	16.8	0
<i>Detrended variables</i>																			
14	-0.1884	+		+											5	7.14	-4.3	0	0.231
31	-0.1828	+										0.2643			4	5.25	-2.5	1.8	0.094
1	-0.1884	+													3	4.16	-2.3	2.0	0.086
34	-0.1987	+												-0.6063	4	5.03	-2.1	2.2	0.076
33	-0.1802	+											0.00101		4	4.96	-1.9	2.4	0.071
18	-0.1884	+						+							5	5.74	-1.5	2.8	0.057
27	-0.186	+							0.07604						4	4.73	-1.5	2.8	0.056
16	-0.1884	+				+									5	5.52	-1	3.3	0.045
13	-0.1884	+	+												5	5.43	-0.9	3.4	0.042
29	-0.1875	+								-6E-04					4	4.36	-0.7	3.6	0.039

Table S6. Relationship between non-detrended blood Hg concentrations in chick-rearing kittiwakes and environmental factors (n=38). The table shows the 5 highest AIC ranked models, the most competitive model appears on the first line ($\Delta AIC = 0$). 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.

Explanatory variables	AIC ranking					Model averaged estimates β (95% CI)		
	df	logLik	AICc	ΔAIC	weight		Linear	Quadratic
Sea Ice + Sea Ice ² + Sex	5	4.99	1.89	0	1	Intercept	0.98 (0.86; 1.09)	
Precipitation + Precipitation ² + Sex	5	-1.89	15.66	13.77	0	Sea ice extent	0.15 (-0.31; 0.6)	1.11 (0.65; 1.56)
Chl <i>a</i> + Sex	4	-3.48	16.18	14.29	0	Sex	0.32 (0.17; 0.47)	
GEM + Sex	4	-4.16	17.53	15.64	0	Precipitation	0.46 (-0.09; 1.01)	-0.54 (-1.09; 0.01)
Sex	3	-5.42	17.54	15.65	0	Chl <i>a</i>	0.35 (-0.02; 0.73)	
SSTa + SSTa ² + Sex	5	-2.96	17.79	15.9	0	GEM	-0.64 (-1.48; 0.2)	
<i>Null model</i>	2	-10.88	26.1	24.2	0	SSTa	0.5 (-0.06; 1.06)	0.33 (-0.23; 0.9)

Chl *a* : Chlorophyll *a*, GEM: Gaseous Elemental Mercury, Air Temp: Air temperature, Sea Ice: Sea ice extent, SSTa: Sea surface temperature anomaly

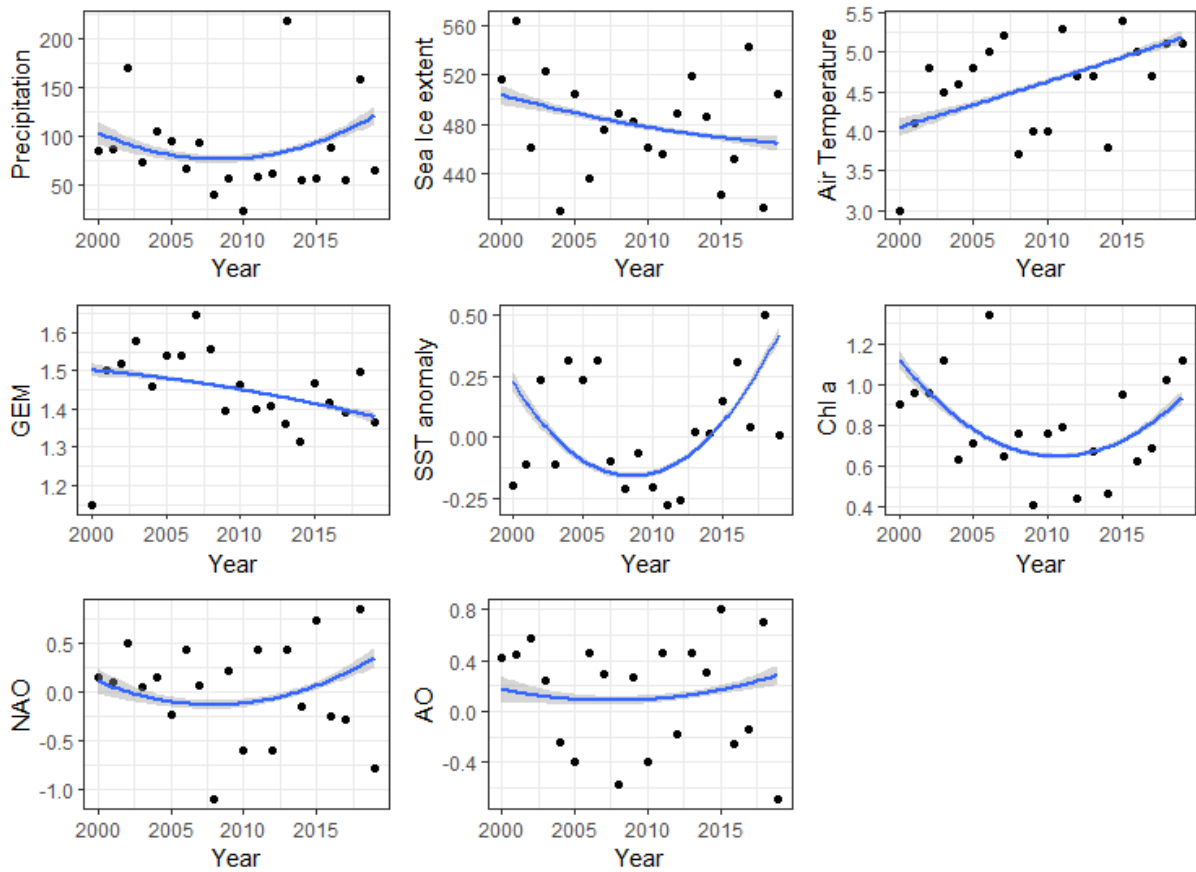


Figure S1. Temporal variations of climate-related variables measured in spring in Svalbard or the Fram Strait. The blue line represents a quadratic relationship between the environmental variables and year, and the grey shaded area the standard error.

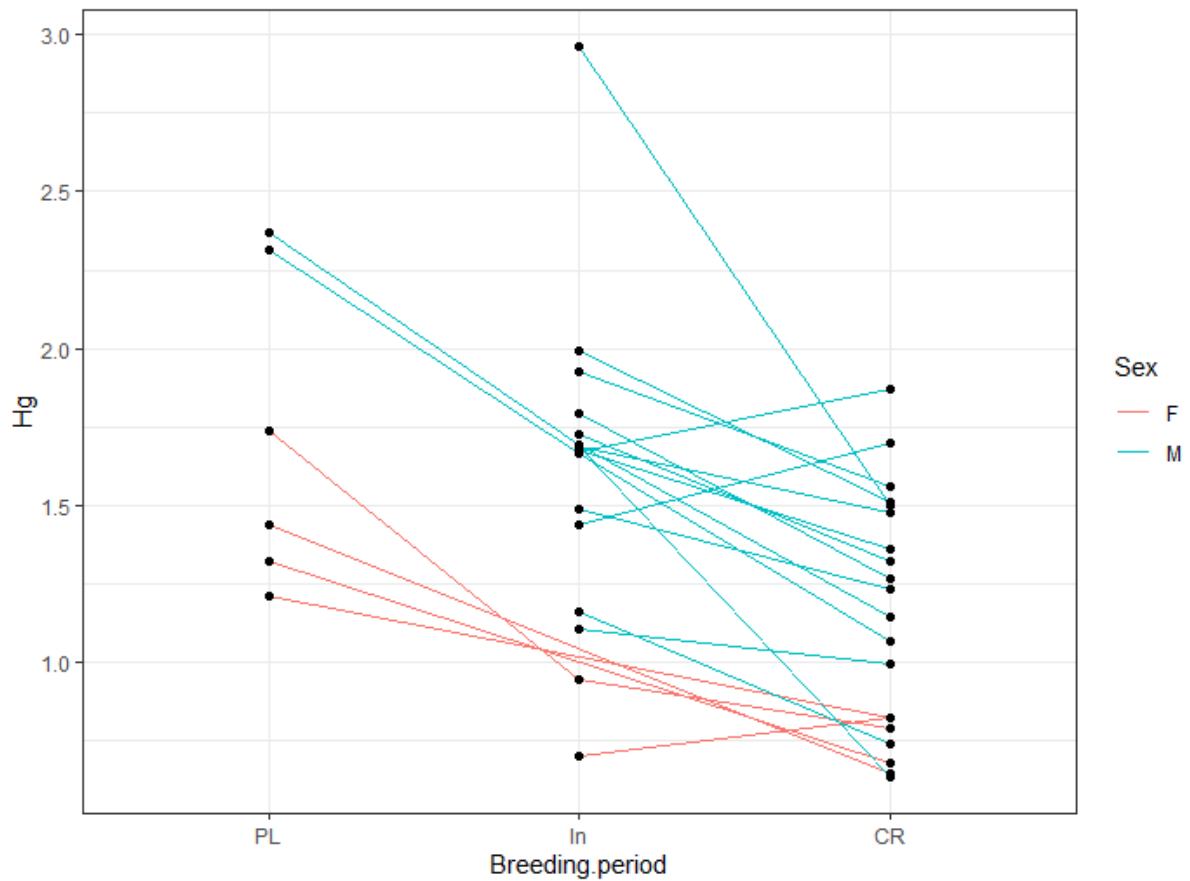


Figure S2. Blood Hg variations across the breeding cycle in kittiwakes captured several time. Each dot connected by a line represents Hg variation across the cycle, males are in green and females in pink (LMM: intercept 1.97 (1.59; 2.35), incubating birds -0.43 (-0.77; -0.10), $p=0.021$, chick-rearing -0.81 (-1.12; -0.50), $p<0.001$).

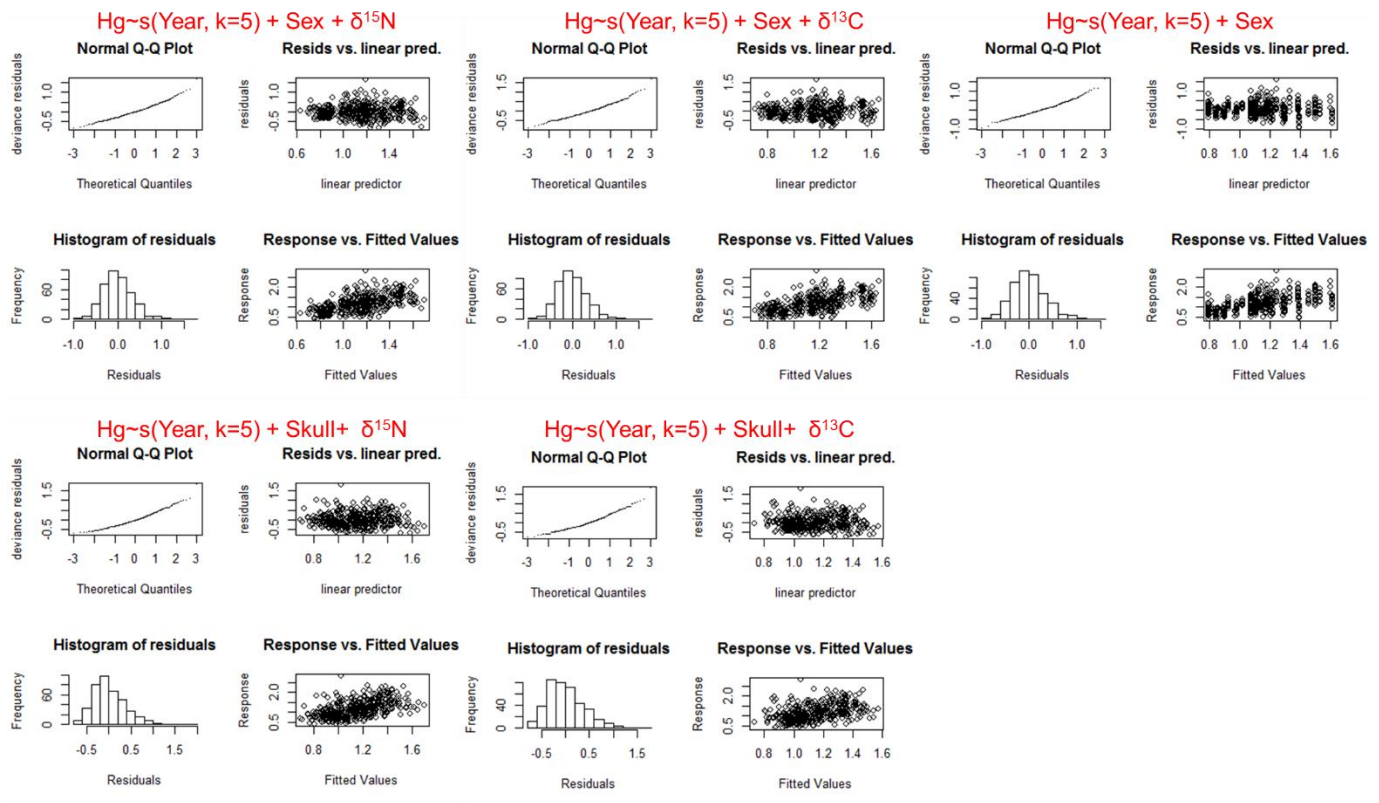


Figure S3. Diagnostic plots for the five GAM models investigating the biological variables which better explain temporal trends of Hg in kittiwakes. Diagnostic plots determine the goodness of the fit of the model to the data. For each model is produced a quantile-quantile plot of residuals (Q-Q plot), a histogram of residuals, a ‘residuals vs linear predictor’ plot, and an ‘observed vs fitted values’ plot.

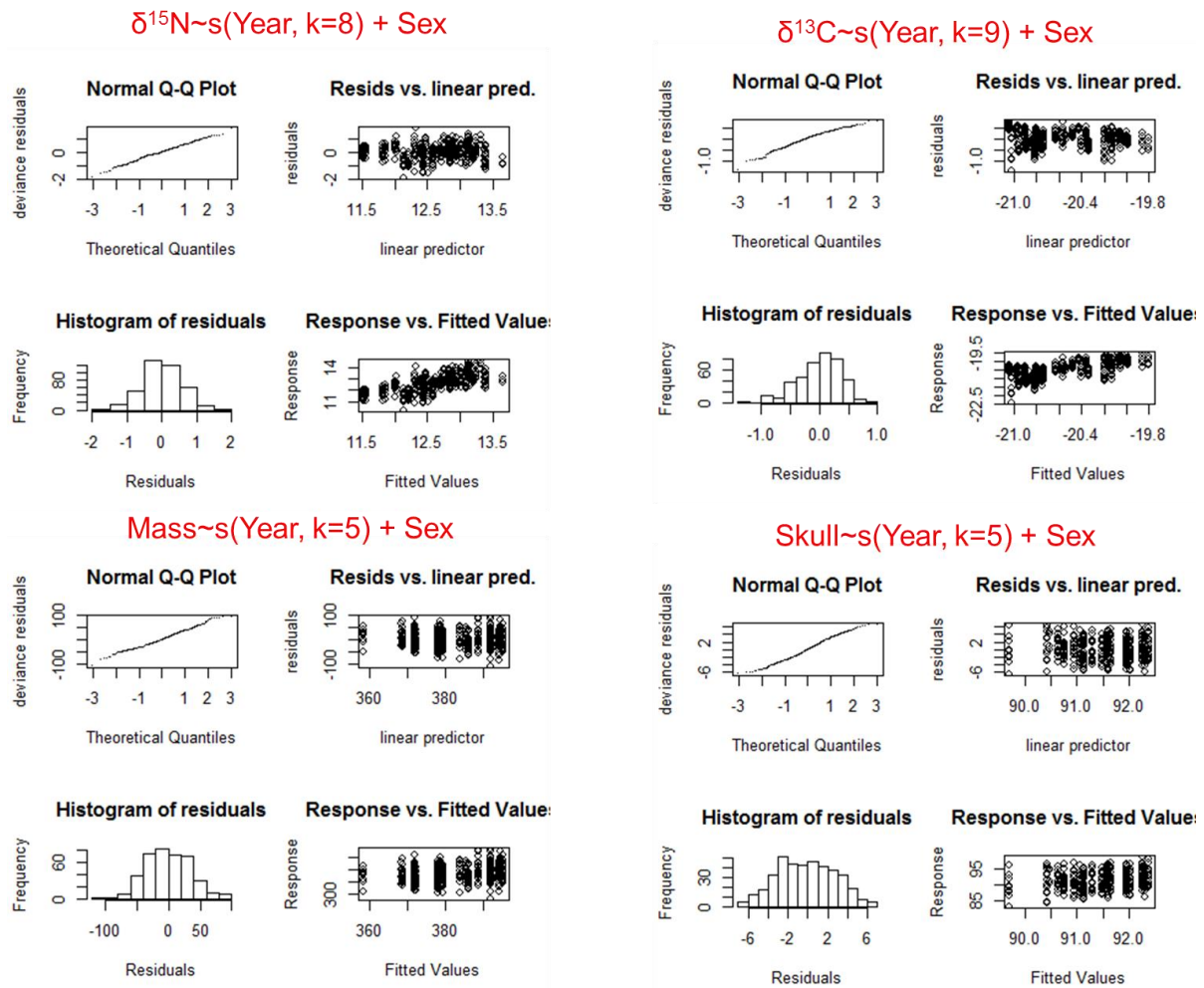


Figure S4. Diagnostic plots for GAM models investigating the temporal trends of stable isotopes and biometrics in kittiwakes. Diagnostic plots determine the goodness of the fit of the model to the data. For each model is produced a quantile-quantile plot of residuals (Q-Q plot), a histogram of residuals, a ‘residuals vs linear predictor’ plot, and an ‘observed vs fitted values’ plot.

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