

Current Biology

Analysis of narwhal tusks reveals lifelong feeding ecology and mercury exposure

Highlights

- Male narwhal tusks provide a lifetime record of stable isotopes and mercury
- Narwhals show temporal (1962–2010) diet plasticity at individual and group levels
- Changes in diet and mercury exposure correlate with loss of sea ice
- Rise in mercury levels since the year 2000 surpasses historical accumulation rate

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In brief

The male narwhal tusk chronologically records ecological information throughout an animal's lifespan. Dietz et al. reconstruct lifetime profiles for stable isotopes (carbon and nitrogen) and mercury to reveal that diet and pollution exposure have shifted over the past decade, likely in response to climate change.



Report

Analysis of narwhal tusks reveals lifelong feeding ecology and mercury exposure

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SUMMARY

The ability of animals to respond to changes in their environment is critical to their persistence. In the Arctic, climate change and mercury exposure are two of the most important environmental threats for top predators.^{1–3} Rapid warming is causing precipitous sea-ice loss, with consequences on the distribution, composition, and dietary ecology of species^{4–7} and, thus, exposure to food-borne mercury.⁸ Current understanding of global change and pollution impacts on Arctic wildlife relies on single-time-point individual data representing a snapshot in time. These data often lack comprehensive temporal resolution and overlook the cumulative lifelong nature of stressors as well as individual variation. To overcome these challenges, we explore the unique capacity of narwhal tusks to characterize chronological lifetime biogeochemical profiles, allowing for investigations of climate-induced dietary changes and contaminant trends. Using temporal patterns of stable isotopes ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) and mercury concentrations in annually deposited dentine growth layer groups in 10 tusks from Northwest Greenland (1962–2010), we show surprising plasticity in narwhal feeding ecology likely resulting from climate-induced changes in sea-ice cover, biological communities, and narwhal migration. Dietary changes consequently impacted mercury exposure primarily through trophic magnification effects. Mercury increased log-linearly over the study period, albeit with an unexpected rise in recent years, likely caused by increased emissions and/or greater bioavailability in a warmer, ice-free Arctic. Our findings are consistent with an emerging pattern in the Arctic of reduced sea-ice leading to changes in the migration, habitat use, food web, and contaminant exposure in Arctic top predators.

RESULTS AND DISCUSSION

The Arctic is warming at a faster rate than any other area in the world, and this rapid warming is having profound effects on Arctic marine environments,⁴ with cascading effects on individuals, populations, and communities.^{5–7} For ice-associated top predators like the narwhal (*Monodon monoceros*), beluga whale (*Delphinapterus leucas*), and polar bear (*Ursus maritimus*), ongoing reductions in Arctic sea ice has implications for their distribution, diet, predator avoidance, and overall health and fitness.^{9–11} Sea-ice loss and associated ecosystem changes also influence mercury transport and fate in the Arctic, often leading to greater biological uptake of this highly neurotoxic element.^{3,8} The potential for interactions and cumulative stress impacts of climate change and mercury pollution is, therefore, cause for concern, particularly for the narwhal, which are

considered the most vulnerable of Arctic marine mammals to climate change¹⁰ and lack efficient physiological capacity to eliminate contaminants.^{12,13} Despite these known risk factors to top marine predators, our understanding of their interactive effects over time is constrained by the inherent limitations of current single-time-point assessment data (e.g., tissue samples). Using temporally sparse and aggregated (i.e., group mean) data can lead to poor characterization of population responses to environmental change, particularly for long-lived species with high individual variability.

Here, we explore the lifetime profiles of feeding ecology and mercury exposure in response to pronounced environmental change in the high Arctic narwhal, a strongly ice-associated species expected to be highly vulnerable to climate change. To do so, we exploit the unique spiraled tusk (150–248 cm) from 10 Northwest Greenland male narwhals by characterizing mercury



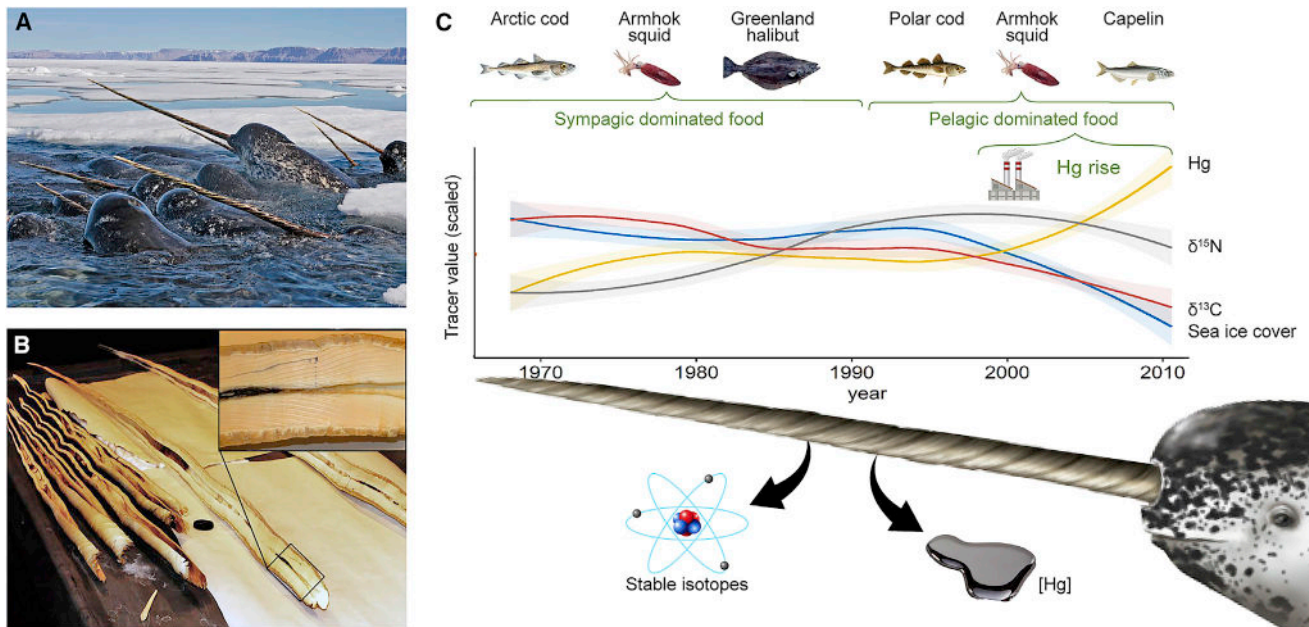


Figure 1. Conceptual diagram for the use of narwhal tusk as a tool to reconstruct lifelong profiles of biogeochemical markers to investigate historical ecological and anthropogenic pressures

(A) Narwhals with erupted tusks in their ice-associated habitat in Northwest Greenland (photo with permission, Paul Nicklen).

(B) Narwhal tusks cut in half before the subsampling showing the annual growth layer groups (GLGs) from which samples for mercury and stable isotopes were taken.

(C) Summary representation of the biogeochemical tracer profiles ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, and Hg) and summer sea-ice coverage. All variables were centered and scaled before plotting to allow for direct comparison of the temporal pattern. Shown are the smoothed conditional mean curves for each scaled variable fitted using loess functions (shaded area = standard error). Mean annual values were used for mercury and stable isotope profiles. Sea-ice coverage represents the average summer sea-ice area (in square kilometers) in Baffin Bay. (See Figure 2 for more details.) The temporal profile is divided into three periods based on biogeochemical profiles and sea-ice coverage: 1962–1990, sympagic dominated: widespread sea-ice coverage and high intake of ice-associated and benthic prey; 1990–2010, pelagic dominated: steady sea-ice cover decline and high intake of pelagic prey; 2000–2010, Hg rise: subset of the pelagic-dominated period with a particularly steep increase in tusk mercury accumulation.

concentrations and stable isotope ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) values in annually deposited dentine growth layer groups (GLGs), representing a time span of 1962–2010 (Figure 1; Table 1). Like other metabolically inert and continuously growing tissue, such as hair, baleen, earplugs, and teeth,^{14,15} the erupted narwhal tusk chronologically preserves physiological information at the time of its growth, thus preserving an invaluable archive of ecological information across the full lifespan of individual animals (>50 years) (Figures 1A and 1B).^{16,17} Biogeochemical tracers, including trace elements and their stable isotopes, provide invaluable insight into aspects of animal physiology, movement, habitat use, and feeding ecology.^{14,15,18} This usefulness stems from the fact that environmental and dietary information stored in these markers is preserved from prey to predator. In general, $\delta^{13}\text{C}$ is used as a tracer of dietary carbon source and has been found to differ between near-shore and offshore marine food webs as well as between pelagic and sympagic ecosystems.^{14,19} $\delta^{15}\text{N}$ is used primarily as a marker of trophic position, as it tends to increase somewhat predictively from prey to predator.^{14,19} Similarly, mercury accumulation in aquatic organisms occurs primarily through dietary intake, and tissue concentrations increase throughout the food web via biomagnification processes.¹² Thus, by pairing annually reconstructed biogeochemical tracer profiles with temporal changes in regional sea-ice cover, we

can explore rarely captured fine-scale, individual-level responses to environmental change (Figure 1C).

Lifetime changes in feeding ecology of high Arctic narwhal

Using the reconstructed stable isotope profiles from ten narwhal tusks, we found marked intra-population differences in isotopic (feeding) niches (Figure 2A) as well as large intra-individual variability in lifetime profiles of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ (Figure 2B; Figure S1). Lifetime isotopic profiles clustered tightly for each individual, and the temporal variation did not follow any clear pattern with narwhal age/time (Figures 2A and 2B). For both isotopes, the annual inter-individual range of values was similar to the intra-individual range throughout a lifetime, revealing marked individual and group variability in feeding ecology. For example, the range of $\delta^{15}\text{N}$ values across individuals in the same year ($1.83\text{‰} \pm 1.1\text{‰}$) was similar to the range within an individual across its lifespan ($2.08\text{‰} \pm 0.5\text{‰}$). We found contrasting temporal profiles for both isotopes at the individual level (Figure S1). Four of the 10 tusks revealed significant linear temporal trends in $\delta^{15}\text{N}$ (-0.092‰ to 0.031‰ per year) and for the grouped dataset, $\delta^{15}\text{N}$ increased significantly by 0.018‰ per year (Table S1). Significant linear trends in $\delta^{13}\text{C}$ were found for seven tusks (-0.026‰ to 0.071‰ per year) (Table S1), with the grouped

Table 1. Sampling details for narwhal tusks sampled in Northwest Greenland

ID	Sampling year	Tusk length (cm)	No. of GLGs (estimated age in years)	Samples analyzed (n)	Tusk tip
N1	2010	151.0	16	16	intact
N2	2010	64.0	7	7	intact
N3	2010	249.0	21	21	intact
N4	2010	166.0	40+	40	broken (5–6 years)
N5	2010	220.5	49+	49	broken (3–4 years)
N6	2010	165.5	18	18	intact
N7	2010	228.0	29+	29	broken (2–3 years)
N8	2008	251.0	45	23	intact
N9	2007	195.0	24	12	intact
N10	2007	179.0	24	12	intact

Individual N9 was sampled from the Uummanaq wintering stock (Niaqornat), and all other individuals were sampled from the Inglefield Breeding summering stock (Thule). Growth layer groups (GLGs) were used to estimate individual age. Every other GLG was sampled for individuals N8–N10, while all GLGs were sampled for individuals N1–N7.

data indicating a significant decline of 0.006‰ per year. The lack of consistent temporal trends at the individual level is likely the result of the non-linear temporal change in values for both isotopes (Figure S1).

Using mean annual values, $\delta^{15}\text{N}$ increased linearly by 0.0244‰/year over the study period ($R^2 = 0.37$, $p < 0.001$; $n = 48$ years) (Figure 2C). We found no significant linear temporal trend in mean annual $\delta^{13}\text{C}$ values ($R^2 = 0.04$, $p = 0.17$; $n = 48$ years), likely due to non-linear increases and decreases within an individual lifetime over the study period (1962–2010) (Figures 2D and S1). To account for the influence and interaction of multiple covariates, we assess a suite of linear mixed-effect models to evaluate which variables most strongly predicted stable isotope values in narwhal tusks over time, never combining age and year in the same model due to their strong covariation. For models predicting $\delta^{13}\text{C}$, averaged coefficient estimates for the top performing models (Table S2) showed a significant positive effect of $\delta^{15}\text{N}$ and summer sea-ice coverage and negative effects of year and age (Figure 2F; Table S3). Top selected models for $\delta^{15}\text{N}$ (Table S2) indicated significant positive effects of year, age, $\delta^{13}\text{C}$, and summer sea ice coverage (Figure 2G; Table S4). Interaction terms between $\delta^{13}\text{C}$ and year or age were kept in top selected models, although effect sizes were low, and confidence intervals overlapped zero, signifying that these variables were relatively uninformative.

These results reveal evidence for a temporal change in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in the narwhals of Northwest Greenland from the 1960s to 2010, broadly associated with a sympagic (ice-associated) dietary signal in the first half of the study period and a shift to an increasingly pelagic dominated signal in the second half (Figure 1C). The change in isotopic values could represent a change in the foraging ecology of narwhals over time or a change in the baseline signal of the ecosystem.¹⁴ A fraction of the observed decline in average $\delta^{13}\text{C}$ may be due to anthropogenic inputs of carbon dioxide (Suess effect: approximately -0.011‰ per year),²¹ but this alone cannot explain the rate of change in this study ($\sim 0.015\text{‰}$ per year). Given that individuals did not always follow the same isotopic pattern over time, we posit that observed temporal changes more likely represent a change in forage ecology rather than a baseline shift, likely in response to

changing environmental conditions. Summer sea-ice cover (in square kilometers) in Baffin Bay has declined by 11.4% per decade since 1968,²² with implications for salinity,²³ vertical stratification of ocean water, and overall irradiance.²⁴ As a result of these climate-driven environmental changes, notable changes have been documented in phytoplankton biomass, production, and community composition.²⁴ Bottom-up processes are likely to extend observed changes in the base of the food web to top predators like narwhal. The spread of sub-Arctic species into Arctic ecosystems is increasing in a process called “borealization.”⁷ Changes in sea-ice dynamics will also influence the spatiotemporal movement patterns of narwhal.²⁵ Thus, environmentally driven changes in the temporal feeding ecology of narwhal can occur through either changes in food web composition or accessibility to different prey through changes in migration patterns. It is unclear which mechanisms are the primary drivers of the observed variability in feeding ecology of narwhal in our study, yet most processes are likely to be strongly influenced by climate change.

The mean isotopic profiles parallel closely the record of summer sea-ice coverage in the region, characterized by a shift from variable but high sea-ice coverage until ~ 1990 to steadily declining levels thereafter (Figure 2E). Further, the positive relationship between stable isotope values and sea-ice coverage in narwhal GLGs in mixed-effect models suggests that increased intake of sympagic prey (more positive $\delta^{13}\text{C}$ values) is associated with a higher trophic level (Figures 2F and 2G; Tables S3 and S4). The observed temporal pattern in $\delta^{13}\text{C}$ values indicates a diet rich in sympagic carbon in the 1970s, followed by a period of mixed sympagic and pelagic diet until ~ 1990 , and a precipitous decline in sympagic prey thereafter (Figure 1C). The temporal pattern in $\delta^{15}\text{N}$ values indicates an increasing intake of high trophic prey until ~ 1990 , followed by an increasing proportion of low trophic prey thereafter (Figure 1C). These patterns follow broadly the summer sea-ice cover trend in the area, which was characterized by relatively high coverage until ~ 1990 , followed by a precipitous decline thereafter (Figures 1C and 2D). These results are consistent with a shift in known narwhal prey items from primarily Greenland halibut (*Reinhardtius hippoglossoides*) and Arctic cod (*Boreogadus*

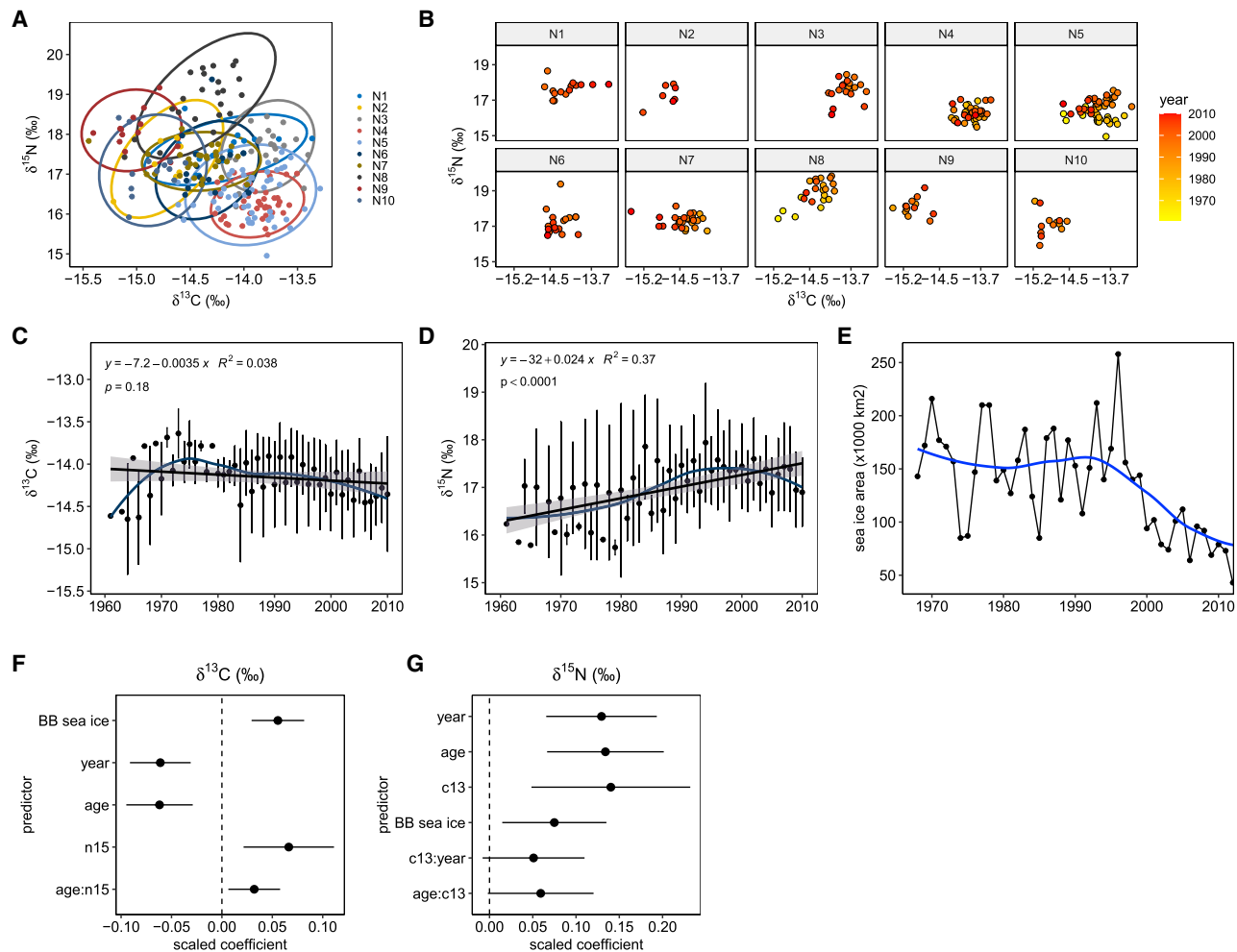


Figure 2. Profile of the feeding ecology of Northwest Greenland narwhals determined from tusk growth layer group (GLG) analysis of stable isotopes

(A) Isotopic niches of individual narwhals based on lifelong $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ concentrations. Isotope niches are visualized as statistical ellipses based on bivariate confidence intervals of stable isotope data.

(B) Isotopic bi-plot for each individual narwhal. Color of points indicate the reconstructed year of deposition based on GLGs.

(C) Time trend of mean annual $\delta^{13}\text{C}$ values ($\pm\text{SD}$). A smoothed conditional mean curve is fitted using loess functions (blue line), and the linear regression line (black) and statistics are included.

(D) Time trend of mean annual $\delta^{15}\text{N}$ values ($\pm\text{SD}$). Smoothed conditional means are fitted using loess functions (blue line) and linear regression line (black) and statistics are included.

(E) Temporal profile of summer sea-ice coverage (in square kilometers) in Baffin Bay. Summer period is defined as June 25 to October 15. Data are from Environment and Climate Change Canada.²⁰

(F and G) Summary results of the linear mixed-effect models for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ indicating the effect sizes of predictor variables. Shown are the conditional model averaged scaled coefficient estimates with 85% confidence intervals from top selected models.

See also [Figure S1](#) and [Tables S1–S4](#).

saida) before the 1990 sea-ice decline to more pelagic prey like capelin (*Mallotus villosus*) and boreoatlantic armhook squid (*Gonatus fabricii*) following the increased open-water season in the area from 1990 to 2010.²⁶ Similar reports from soft tissue analysis of beluga whales, narwhals, and ringed seals in the region^{26–28} provide a consistent pattern of increased intake of sub-Arctic and pelagic prey in Arctic top predators with climate warming and sea-ice loss.

Studies using stable isotopes in narwhal soft tissue (muscle/skin) have shown that this species has higher variability in foraging ecology than previously thought, suggesting some

capacity to adapt to rapid climate-induced environmental changes in the Arctic.^{26,29} Our study confirms these findings and adds insights at both the individual and group (population) levels. Consistent with previous studies, we find variability in foraging patterns between individual males at a snapshot in time; however, we have also shown that foraging patterns of individuals fluctuate over time to a similar degree ([Figure S1](#)). Our analysis thus provides rare insight into the dietary plasticity of the male narwhal over time, revealing variability of foraging ecology at the individual and group levels in this strongly ice-associated species with presumed high dietary specialization.^{10,29}

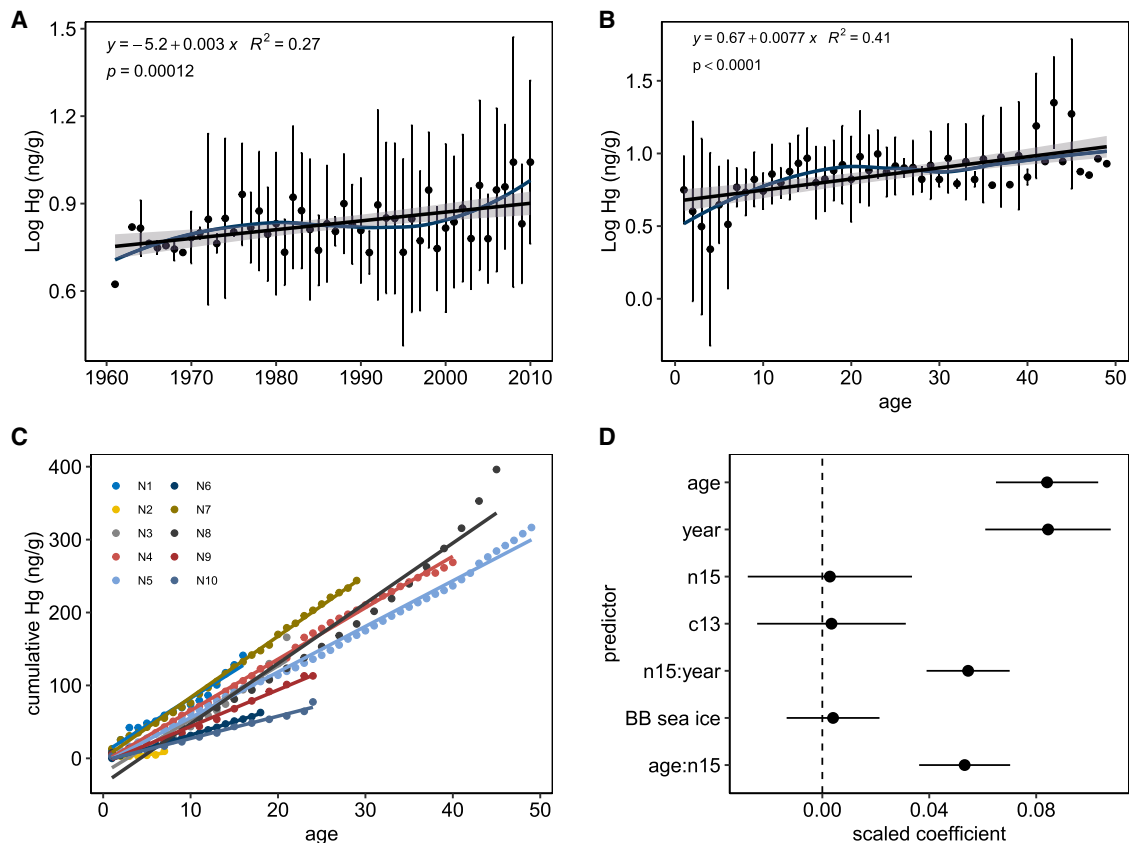


Figure 3. Profile of mercury accumulation of Northwest Greenland narwhal determined from tusk growth layer group (GLG) analysis of total mercury

(A) Time trend of total mercury concentrations (nanograms per gram of dry weight) reconstructed from GLGs. Indicated are the mean annual mercury concentrations (\pm SD). A smoothed conditional mean curve is fitted using loess functions (blue line), and the linear regression line (black) and statistics are included. (B) Age trend of total mercury concentrations. Indicated are the mean age mercury concentrations (\pm SD). A smoothed conditional mean curve is fitted using loess functions (blue line), and the linear regression line (black) and statistics are included.

(C) Cumulative lifetime mercury accumulation curves for individual narwhals. Cumulative lifetime mercury was calculated as the cumulative sum of annual concentrations for each individual.

(D) Summary results of the linear mixed-effect model indicating the effect sizes of predictor variables on concentrations of total mercury in annual GLGs of individual narwhals. Shown are the conditional model averaged scaled coefficient estimates, with 85% confidence intervals from top selected models.

See also [Figure S1](#) and [Tables S1](#), [S2](#), and [S5](#).

One of the major strengths of analyzing biogeochemical markers in growing and metabolically inert tissues such as teeth is that they provide rare insight into the foraging ecology of individual animals over time, data otherwise near impossible to acquire with standard monitoring approaches (e.g., soft tissue sampling). While we show how the narwhal tusk can be used to explore temporal feeding patterns, similar temporal analyses in other species have been carried out using regular teeth growth layers^{30–32} or gradients along hair/fur,^{33,34} feathers,³⁵ whiskers,^{36,37} baleen plates,³⁸ or layers of cerumen in earplugs.³⁹ Different tissues provide information at various temporal scales (weeks to years), providing researchers with a plethora of information with which to study animal ecology and physiology.¹⁸

Lifetime mercury profiles and predictors of annual exposure

Lifetime profiles of mercury revealed large inter-individual variability in concentrations over time ([Figures 1C](#), [3A](#), and [3B](#))

through consistent cumulative lifelong accumulation ([Figure 3C](#)). Combining all animals, mercury increased log-linearly by 0.3%/year ($R^2 = 0.27$, $p < 0.001$; $n = 48$ years) and by 1.9%/year post-2000 where there was a distinct deviation from the historical trend ([Figure 3A](#)). Similarly, average mercury increased log-linearly with age by 0.7%/year ($R^2 = 0.44$, $p < 0.001$; $n = 48$ years) ([Figure 3B](#)). The cumulative annual deposition of mercury in tusks across the full lifetime was linear in all individuals, and maximum values reached up to ~ 400 ng/g in the oldest animals ([Figure 3C](#)). The slope of the cumulative mercury deposition, representing the annual accumulation rate, was similar across individuals and ranged from 0.79 to 8.40, with a mean of 6.70 ng/g per year. At the individual level, we found significant log-linear increases in annual tusk mercury in five of the 10 narwhals over time (0.097% to 12.10%/year) ([Table S1](#)).

Model-averaged regression estimates from top selected linear mixed-effect models indicated that year, age, and $\delta^{15}\text{N}$ were the strongest predictors of annual mercury concentrations

(Figure 3D; Tables S2 and S5). Concentrations of mercury increased over time and increased with animal age, and the interaction with $\delta^{15}\text{N}$ indicated that mercury accumulated at a greater rate over time in animals with greater values of $\delta^{15}\text{N}$. These results suggest that feeding ecology—namely, trophic position of prey species—is an important determinant of mercury in narwhal-tusk GLGs. Consistent with our proposed isotopic temporal profile, mercury levels increased during the sympagic period concurrently with $\delta^{15}\text{N}$ (1962–1990), likely due to intake of the high-trophic and high-mercury-laden Greenland halibut.⁴⁰ Levels of mercury during the 1990s leveled off as sea-ice cover declined and pelagic prey, relatively lower in mercury,⁴⁰ began to dominate the narwhal dietary signal. This pattern of stable or slightly declining mercury concentrations continued until 2000, after which concentrations increased rapidly (Figure 3A), uncoupling from the sea-ice-associated dietary signal.

The rapid rise of mercury concentrations in recent years (post-2000) above the historical trend (Figure 3A), violating the linear trend in cumulative lifetime accumulation (Figure 3C), suggests a higher input of mercury to the environment and/or a change in diet or food web structure. A change in diet is consistent with the increased pelagic signal from dietary tracers in recent decades, though not likely related to trophic magnification of mercury, as $\delta^{15}\text{N}$ declined during this period (2000–2010) (Figure 1C). Evidence from other Arctic top predators support our hypothesis in showing that tissue mercury and other pollutant levels tend to increase in years with less sea ice.⁸ Studies of the Arctic fox (*Vulpes lagopus*) and polar bears have similarly shown greater than expected rises in tissue mercury levels in the past 2 decades.^{41–43} Climate-change-induced sea-ice loss can influence mercury bioaccumulation in Arctic food webs via changes in mercury distribution, transport, oxidation, and methylation rates; changes in primary productivity; and shifts in food web structure.^{3,44} We posit that the unexpected rapid rise in recent mercury concentrations as revealed through narwhal tusk GLGs likely reflects increased global anthropogenic emissions,³ increased secondary emissions,⁴² and/or increased bioavailability in pelagic compared to sympagic food webs,⁴⁴ resulting from the effects of climate warming in the Arctic.

Overall, the rate of mercury increase over time in our study is similar to that reported in the same region for tooth GLGs of beluga whales (1885–1997),⁴⁵ hair of polar bears (1898–2008),⁴⁶ and feathers of three species of birds of prey (1850–2004),³⁵ suggesting a consistent pattern across species in this Arctic region. In the narwhals of this study, though, age likely contributes to the temporal increase of mercury concentrations, suggesting that the temporal profile of mercury is the sum of lifetime bioaccumulation and increased environmental inputs. Mercury also increased with $\delta^{15}\text{N}$, suggesting increased intake of mercury as the whales feed on higher trophic-level prey. Organic mercury (methyl mercury) is the major component of dietary mercury in fish-eating wildlife and is highly bioaccumulative.¹² Indeed, $\delta^{15}\text{N}$ is typically used as a marker of trophic position and strongly predicts the food web magnification of mercury concentrations in Arctic marine food webs.³⁵

Our study has revealed unprecedented insight into individual lifelong feeding ecology and contaminant accumulation in a long-lived Arctic top predator. Although our analyses preclude

a causal link to climate change, they are consistent with an emerging pattern across Arctic marine mammals: warmer temperatures are causing steady declines in sea-ice coverage, leading to increased borealization of Arctic marine ecosystems⁷ and changes in the habitat use and diet of top predators.^{26,27} As demonstrated here, narwhal plasticity in feeding ecology suggests that narwhals are able to change their foraging ecology in response to changing environments. Follow-up dietary studies (stomach content, fatty acid analysis, etc.) are needed to further explain the variability in feeding ecology with time demonstrated in this study and to investigate whether narwhals are able not only to change the composition of their historic diet over time but also to shift to entirely new prey items. Changing environmental conditions and biological communities in the Arctic have profound implications for the fate of environmental contaminants like highly neurotoxic mercury,^{8,47} which, when combined with increased anthropogenic emissions,³ can lead to unexpected increases in tissue concentrations of endemic wildlife. The use of the male narwhal tusk as a chronological record of biogeochemical data, as shown in this study, can provide insight into these important questions, especially considering archived collections in museums.

STAR★METHODS

Detailed methods are provided in the online version of this paper and include the following:

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SUPPLEMENTAL INFORMATION

Supplemental Information can be found online at <https://doi.org/10.1016/j.cub.2021.02.018>.

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AUTHOR CONTRIBUTIONS

R. Dietz designed the study. E.G., C.S., M.P.H.-J., A.A., P.A., R. Drimmie, and R. Dietz provided samples or laboratory analysis; J.-P.D. performed data analysis, with support from R. Dietz and F.F.R.; and J.-P.D. and R. Dietz generated

figures and tables. J.-P.D. and R. Dietz drafted the manuscript. All authors were involved in the interpretation of results and writing of the final manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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STAR★METHODS

KEY RESOURCES TABLE

REAGENT or RESOURCE	SOURCE	IDENTIFIER
Biological samples		
Adult male narwhal tusk	Greenland Inuit subsistence hunt	Table S1
Chemicals, peptides, and recombinant proteins		
SRM 1400	National Institute of Standards and Technology, USA	SRM 1400
SRM DOLT-5	National Research Council of Canada	DOLT-R
Deposited data		
Raw and analyzed data	This paper	
Software and algorithms		
R v4.0.1	⁴⁸	https://cran.r-project.org/

RESOURCE AVAILABILITY

Lead contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the Lead Contact, Rune Dietz (rди@bios.au.dk).

Materials availability

This study did not generate new unique reagents.

Data and code availability

Original data for all figures, tables and statistical analyses in the paper are available at Mendeley Data: <https://doi.org/10.17632/y3tf8mw2dv.1>.

EXPERIMENTAL MODEL AND SUBJECT DETAILS

Ten male narwhal tusks were collected from northwest Greenland. Nine animals were from Avanersuaq, Inglefield Bredning summering stock (Thule) from 2007–2010 and the last animal was from Niaqornat, Uummanaq wintering stock from 2007 (Table 1). All tusks were purchased from Inuit subsistence hunters (following hunting quotas) as part of scientific research granted by the Danish Environmental Protection Agency (Danish Cooperation for Research in the Arctic: DANCEA). Narwhal are listed under CITES-List II and were therefore legally exported from Greenland with a CITES export permit (#10GL0806157) and imported to Denmark with a CITES import permit (#IM 0611-297/10) for wild animals (W) and shipped for scientific research (S). Tusks are stored at the Danish Natural History Museum in Copenhagen and Aarhus University, Denmark. For information and access to samples, contact Lead Author Prof. Rune Dietz.

The 2007–2008 tusks ($n = 3$) were sampled from every second growth layer group (GLG), while all others ($n = 7$) were sampled from every annual GLG. Age was estimated by counting the dentine layers in the tusk¹⁷ and used for historical reconstruction of Hg and stable isotope profiles dating back up to 1962 for the oldest sample. Available animal details are provided in Table 1.

METHOD DETAILS

Sample preparation

Tusks were cut into two longitudinal sections with a jigsaw and then ground with fine sandpaper (Figure 1B). Next, the surface of the tusk was decalcified by acetic acid (12%) after which they were rinsed with water. This procedure improved the readability of the GLG or annuli of the tusks. A series of double analyses were conducted with and without acetic acid treatment, which revealed that the Hg concentration in the dentinal layers of the tusk was not affected by the treatment ($p = 0.157$). Dentine samples of ca. 0.10 g for Hg analysis and ca. 0.05 g for stable isotope analysis were cut from GLGs using a hand-held Dremel grinding tool with a diamond cutting plate.

Mercury analysis

Analysis of mercury was performed at Aarhus University (Roskilde), Denmark, using a solid sample atomic absorption spectrometer AMA-254 (Advanced Mercury Analyzer-254 from LECO, Sweden). Details on the analytical methods and quality assurance were

based on previously published work.^{45,49} Briefly, the analytical process consists of a drying period at 120°C for 50 s, prior to a combustion phase at 750°C for 250 s, which leads to the desorption of Hg from the samples. Subsequently, the Hg vapor produced is carried by an oxygen flow to a gold amalgamator and trapped on its surface. Thereafter, the collected Hg is released from the amalgamator by a short heat-up to 900°C, and carried in a pulse through a spectrophotometer, where it is measured by UV absorption. Standard Reference Materials (SRMs) included NIST 1400 Bone ash (National Institute of Standards and Technology, USA) and the DOLT-5 (Dogfish Liver Tissue from the National Research Council of Canada). These were run at the beginning and end of the analytical cycle as well as every ten samples. Our laboratory also participates in the international inter-laboratory comparison exercises conducted by the EEC (QUASIMEME), and the latest 2007 analyses by AMA-254 showed satisfactory results ($0 < z < 0.5$). All data are presented on a dry weight basis (dw) and the detection limit (d.l.) was 0.01 ng.

Dietary tracers - stable isotope analysis

Stable isotopes were analyzed in GLGs as general dietary tracers of trophic position and feeding ecology (pelagic versus benthic/ice-associated). The 2007 and 2008 ($n = 3$) tusks were analyzed at the Environmental Isotope Laboratory (EIL), University of Waterloo, Canada, while remaining samples were run at Department of Chemical and Biochemical Engineering, DTU Risø Campus, Denmark. Sample preparation and analysis followed methods previously described.^{45,49} Briefly, GLG samples were crushed into small pieces and ground into homogeneous powder using a ball mill (Retsch MM2000). The powdered dentine was stored in small glass flasks where carbonates were removed by digestion with ~ 1 mL of a 4M hydrochloric acid solution at 45°C for 48 h. The digesta was diluted with in milli-Q ultrapure water and freeze-dried. The freeze-dried samples were homogenized and kept at -20°C until analysis. Approximately 1.45 mg of sample was loaded into tin capsules and analyzed for the relative abundance of stable isotopes of nitrogen ($^{15}\text{N}/^{14}\text{N}$) and carbon ($^{13}\text{C}/^{12}\text{C}$) using an Elemental Analyzer connected on-line to an Isotope-Ratio Mass Spectrometer (Iso-prime, Micromass, UK). International standards included atmospheric nitrogen for $\delta^{15}\text{N}$ and the Pee Dee Belemnite (PDB) marine fossil limestone formation from South Carolina for $\delta^{13}\text{C}$. We also included acetanilide as an internal laboratory reference material.

QUANTIFICATION AND STATISTICAL ANALYSIS

Several approaches were taken to evaluate temporal patterns and overall predictors of Hg, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$. Linear regression analyses of GLG concentrations of each variable against time (calendar year or animal age) were applied using log-transformed concentrations of Hg and untransformed $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values as the dependent variable and year or age as the explanatory variable. This was carried out for each individual separately and using the mean annual values of all individuals. The isotopic feeding niche for each individual was assessed graphically by fitting bivariate confidence intervals of stable isotope data for each individual narwhal assuming a Student-t distribution using the `stat_ellipse` function of the 'ggplot2' package⁵⁰ in the software R.⁴⁸

We used multi-variate linear mixed-effect models (LMMs) to investigate the influence of biological and ecological drivers on variables of interest in narwhal tusk GLGs, including Hg, $\delta^{15}\text{N}$, and $\delta^{13}\text{C}$. Fixed effects included biological and ecological variables (see below) and individual ID was used as the random effect to account for pseudo-replication. We followed established protocols for conducting and presenting regression analyses.⁵¹ For each response variable, we generated *a priori* a list of candidate models containing all appropriate (see below) iterations of predictor variables (including a null model with intercept only) and used an information-theoretic approach based on Akaike's information criterion corrected for small sample size (AIC_c) for the selection of best models.⁵² Strongly correlated predictor variables (year and age) were never included within the same model in order to avoid collinearity issues, which was tested using variance inflation factors ($\text{VIF} < 3$) in all models.⁵³ All predictor variables were centered and scaled to further reduce collinearity (i.e., VIFs), especially among interaction terms, and facilitate comparison of effect sizes.⁵⁴ The final selected model set was chosen conservatively by including all models within $\Delta\text{AIC}_c < 10$.⁵⁵ Conditional model averaging using the final top-selected models was conducted to generate averaged regression coefficient estimates for all predictors, weighted by AIC weight.⁵² Calculated 85% confidence intervals (CI) for averaged parameter estimates were used to determine meaningful predictor variables, assuming that variables with 85% CIs that do not overlap zero significantly affect the response variable.⁵⁵ Model assumptions were verified by plotting residuals versus fitted values, residuals versus each covariate in the model, and normal quantile-quantile plots for residual normality. LMMs were run with the 'nlme' package,⁵⁶ model selection and condition averaging were performed using the 'MuMIn' package,⁵⁷ in the software R.

The model of GLG concentrations of $\delta^{15}\text{N}$ included the following predictor variables (fixed effects): year (continuous), age (continuous), $\delta^{13}\text{C}$ (continuous), and Baffin Bay sea ice cover (km^2) (continuous). Interaction terms included age \times $\delta^{13}\text{C}$, year \times $\delta^{13}\text{C}$, and sea ice cover \times $\delta^{13}\text{C}$. The model of GLG concentrations of $\delta^{13}\text{C}$ was similar and included year, age, $\delta^{15}\text{N}$ (continuous), and Baffin Bay sea ice cover as predictor variables, with age \times $\delta^{15}\text{N}$, year \times $\delta^{15}\text{N}$, and sea ice cover \times $\delta^{15}\text{N}$ as interaction terms. Year and age variables were never included in the same model as these were significantly correlated. For both stable isotopes, untransformed concentrations were used. To model the GLG concentrations of Hg as a function of available covariates, LMM with \log_{10} transformed Hg concentrations were used. Predictor variables included year, age, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, and Baffin Bay sea ice cover with age \times $\delta^{15}\text{N}$, year \times $\delta^{15}\text{N}$, age \times $\delta^{13}\text{C}$, year \times $\delta^{13}\text{C}$, sea ice cover \times $\delta^{13}\text{C}$, and sea ice cover \times $\delta^{15}\text{N}$ as interaction terms. Baffin Bay sea ice cover was taken from Environment and Climate Change Canada.²⁰ Sea ice data represent the average summer sea ice coverage in Baffin Bay, defined as the period from June 25 to October 15th.