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An assessment of mercury and its dietary drivers in fur of Arctic wolves from Greenland and High Arctic Canada

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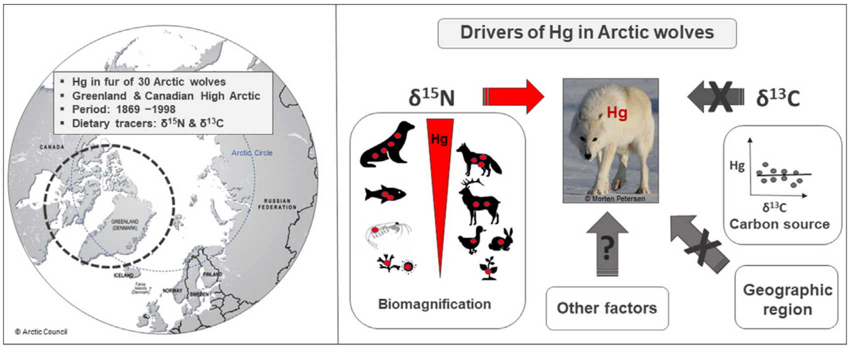
HIGHLIGHTS

- Total mercury (THg) measured in Arctic wolf fur ($n = 30$) from 1869 to 1998 in 3 regions.
- THg fur levels of 0.20–5.45 μg g^{-1} dw indicate no or low risk for health effects.
- Trophic position, not carbon source nor region, explains variation in THg exposure.
- Stable isotope values suggest high dietary plasticity.
- Historic fur samples are useful for retrospective THg monitoring in Arctic canids.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury has become a ubiquitous hazardous element even ending up in pristine areas such as the Arctic, where it biomagnifies and leaves especially top predators vulnerable to potential health effects. Here we investigate total mercury (THg) concentrations and dietary proxies for trophic position and habitat foraging (δ^{15}N and δ^{13}C, respectively) in fur of 30 Arctic wolves collected during 1869–1998 in the Canadian High Arctic and Greenland. Fur THg concentrations (mean ± *SD*) of 1.46 ± 1.39 μg g^{-1} dry weight are within the range of earlier reported values for other Arctic terrestrial species. Based on putative thresholds for Hg-mediated toxic health effects, the studied Arctic wolves have most likely not been at compromised health. Dietary proxies show high dietary plasticity among Arctic wolves deriving nutrition from both marine and terrestrial food sources at various trophic positions. Variability in THg concentrations seem to be related to the wolves' trophic position rather than to different carbon sources or regional differences (East

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Greenland, the Foxe Basin and Baffin Bay area, respectively). Although the present study remains limited due to the scarce, yet unique historic study material and small sample size, it provides novel information on temporal and spatial variation in Hg pollution of remote Arctic species.

1. Introduction

Due to increased deposition of atmospheric mercury (Hg) of anthropogenic origin over the past century, environmental Hg contamination has become an issue (AMAP/UN Environment, 2019; Dietz et al., 2019). Thousands of tons of Hg from industrial processes such as coal combustion, municipal waste incineration, and mercury production are transported and released into the oceans and to the atmosphere each year (Beckers and Rinklebe, 2017; Driscoll et al., 2013; Streets et al., 2017, 2019; AMAP/UN Environment, 2019) resulting in the atmospheric Hg pool having increased an estimated 3-fold relative to 1850 (Enrico et al., 2017; Fitzgerald et al., 1998). Long-range transport via atmospheric pathways, ocean currents and migrating animals distribute Hg to remote regions such as the Arctic (Fitzgerald et al., 1998; Obrist et al., 2018). On the basis of the high anthropogenic Hg concentrations worldwide, the anthropogenic releases of Hg are now globally regulated by Minamata Convention on Mercury that was ratified in 2017 (www.mercuryconvention.org) in order to take action to protect human health and the environment. Microbial activity facilitates methylation of monomethylmercury (MeHg), which particularly bioaccumulates in tissues of organisms (Hammerschmidt and Fitzgerald, 2006; Mason et al., 2012), and is biomagnified along the food chain (Jæger et al., 2009; Lavoie et al., 2013; Ruus et al., 2015). As a result, Hg concentrations have increased over time in multiple Arctic species (Braune et al., 2005, 2015; Dietz et al., 2006a, 2006b, 2009, 2011; Dietz et al., 2021; Rigét et al., 2011) reaching especially high levels in Arctic high-trophic species such as polar bears (*Ursus maritimus*; Dietz et al., 1998, 2006a, 2006b; Dietz et al., 2013a,b), seals and narwhals (*Monodon monoceros*; Dietz et al., 2021; Houde et al., 2020), birds of prey (e.g. Dietz et al., 2006b; Ekblad et al., 2021; Sun et al., 2019), and Arctic foxes (*Vulpes lagopus;* e.g. Bocharova et al., 2013). Hence, such high trophic species are potentially at increased risk for harmful effects associated to Hg exposure, comprising changes in immune functioning, hormone imbalances, oxidative stress, tissue pathology, behavioural, neurochemical and reproductive impairments (see reviews by: Desforges et al., 2016; Dietz et al., 2013a,b, 2019, 2022; Sonne, 2010).

From a risk assessment perspective apex predators are valuable as sentinel species because of integration of chemical exposure across their food chain over larger space and time spans (Burger and Gochfeld, 2001; Harley et al., 2016). Fur samples have been shown promising for minimally-invasive biomonitoring of Arctic pollution using sentinel species (Bechshoft et al., 2016, 2019; Dietz et al., 2006a, 2009). Specifically for Hg issues, MeHg is sequestered from the blood circulation into this keratinized tissue due to its strong binding affinity to sulfur groups (George et al., 2010) and reflects previous exposure levels now archived upon growth of the fur (Wang et al., 2014). Hair has been widely studied as an indicator of blood Hg levels and dietary exposure in humans (FAO/WHO, 2003). In addition, fur Hg has proven to be significantly correlated with concentrations in animal soft tissues, including kidney, liver, muscle, cerebrum and cerebellum, and is hence considered a valuable biomonitoring matrix (for review see Dietz et al., 2009), especially when studying Arctic canids (Dainowski et al., 2015; Treu et al., 2018). Moreover, the complementary analysis for stable carbon and nitrogen isotope composition reflects the diet composition during the period of tissue synthesis (Bearhop et al., 2002; Hobson and Clark, 1992). As such, joint analysis for contaminants and stable isotope values allows for an integrated evaluation of contaminant exposure and its potential dietary ecological drivers such as trophic position (proxied by δ^{15}N) and carbon source (proxied by δ^{13}C). The physicochemical and biochemical stability of hair (Crewther et al., 1965) warrant both sequestered contaminants, such as Hg (Appelquist et al., 1984), as well as biomolecules to remain stable throughout time. This is the tenet on which the analysis of archived museum or private collection samples rest in order to allow for retrospective assessment of Hg exposure and its potential dietary pathways.

Stable isotope (SI) analysis is an integrated approach to improve the overall understanding of feeding behaviour and food web ecology and to unravelling animal diets, as stable isotopes in animal tissues and excreta reflect dietary preferences, and yield insight into the environmental conditions experienced by the animal (e.g. West et al., 2006, review by Kelly, 2011). Nitrogen stable isotopes are commonly used as proxies for estimating the trophic position of animals since consumers get enriched with ^{15}N in relation to ^{14}N (ratio expressed as δ^{15}N) by ~2 to 3.4‰ compared to their prey (e.g. Vanderklift and Ponsard, 2003), whereas measuring the enrichment of ^{13}C in relation to ^{12}C (ratio expressed as δ^{13}C) can be used to distinguish different carbon sources of terrestrial (depleted δ^{13}C) and aquatic (enriched δ^{13}C) environments (Kelly, 2011; Peterson and Fry, 1987). The SI signature of a tissue generally reflects the diet composition during the period of tissue synthesis (Bearhop et al., 2002; Hobson and Clark, 1992), this is why SI in hair, in particular, offer insights of an animal's dietary history over extended periods (Dalerum and Angerbjörn, 2000; Roth and Hobson, 2000).

Many ecotoxicological studies have focused on marine Arctic species, while insight into contaminant exposure and its sources in terrestrial wildlife remains a latent knowledge gap (AMAP, 2018; Dietz et al., 2019, 2022), with only few efforts on Arctic foxes (Bocharova et al., 2013; Hallanger et al., 2019; Treu et al., 2018). Therefore, the overall aim of the present study was to study Hg concentrations and potential dietary ecological drivers in Arctic wolves, another valuable sentinel species in the poorly documented terrestrial environment. We used fur sampled over a long time period (from 1869 to 1998) across the Greenlandic and Canadian High Arctic (Foxe Basin/Baffin Island, Baffin Bay area/Ellesmere Island). Specifically, we aimed i) to assess the magnitude of THg exposure using fur levels and compare these to putative risk thresholds for potential exposure-mediated individual health effects; ii) to identify potential drivers of variation in exposure, with geographical region, trophic position, and carbon source as potential candidates. Based on previous studies on Hg contamination in marine and terrestrial Arctic predators (Bocharova et al., 2013; Burnham et al., 2018; Dehn et al., 2006; Desforges et al., 2022; Dietz et al., 2022; Dietz et al., 2019; Gamberg and Braune, 1999; McGrew et al., 2014; Treu et al., 2018), we hypothesize that Arctic wolves are likely not at risk of Hg-associated compromised health and that individuals feeding on a higher trophic diet may ingest marine prey and therefore exhibit higher THg fur levels.

2. Materials and methods

2.1. Study species and sample collection

The Arctic is roughly divided into two regions: The High Arctic, which appears as polar desert, and the Low Arctic, which corresponds to the tundra (AMAP, 1997). Among grey wolves (*Canis lupus ssp.*) from the high Arctic region, three distinct wolf populations were recently identified genetically comprising a novel and highly distinct polar wolf population endemic to Ellesmere Island and Greenland (Sinding et al., 2018). The three populations occur in the West Arctic (representing the Banks and Victoria Islands), East Arctic (representing the area near the Baffin Island), and in the High Arctic polar region (representing Ellesmere Island and Greenland), respectively (Sinding et al., 2018). Several subspecies of the grey wolves have been reported by Nowak (1995) in the Canadian Arctic, North America and Greenland but systematic information on the numbers and distribution in particular on the Canadian Arctic Archipelago are lacking (Miller, 1995). In North and East Greenland the number of Arctic wolves between 1978 and 1998 was estimated up to 55 individuals based on sightings (Marquard-Petersen, 2009). The two wolf subspecies, *Canis lupus arctos* and *Canis lupus nubilus,* are reported to occur in the three study areas (Nowak, 1995) but wolf taxonomy and distribution ranges are still a matter of research due to lack of systematic data. Since no genetic analysis was applied in the current study, the distinction of wolf subspecies was not possible and thus we hereinafter refer to the study species as *Canis lupus* ssp. Arctic wolves are top predators and scavengers both in the terrestrial and marine food web (Dalerum et al., 2018; Marquard-Petersen, 1998). As opportunistic scavenging carnivores, wolves from Greenland and Arctic Canada have been documented to mainly feed on muskoxen (*Ovibos moschatus*), caribou (*Rangifer tarandus*), lemmings (*Lemmus* and *Dicrostonyx* spp), Arctic hare (*Lepus arcticus*), geese and other seabirds, insects, marine and terrestrial mammal carcasses as well as refuse (Dalerum et al., 2018; Marquard-Petersen, 1998).

Hair samples of 30 Arctic wolves from West and East Greenland (n = 13) and the Canadian High Arctic (n = 17) had been collected between 1869 and 1998 and stored at the Natural History Museum of Denmark, University of Copenhagen, Danish Museum of Hunting and Forestry, Natural History Museums in London and Oslo, and within the Ulf Marquard-Petersen Greenland Wolf Research Program, USA. Nine wolves were collected from East Greenland between 1901 and 1998, 11 wolves from North-eastern Canada/North-West Greenland between 1897 and 1940, and 10 wolves from Central Eastern Canada from 1869 to 1928. We had to rely on unique specimens opportunistically available at different museums and scientific collections, which determined the final sample collection the present study is based upon. Biological metadata for each Arctic wolf is given in Table S1 of the Supplement. For the majority of the 30 Arctic wolf fur samples no information was available neither on sample treatment nor on biological metadata (Table S1).

Hair degrades slowly relative to other tissues (Pereira et al., 2004) and therefore, samples from museum collections containing pelts of different species can be valuable resources to assess mercury trends over time and space, retrospectively. Still, it should be noted that mercuric chloride ($HgCl_2$ or sublimate) was sometimes used in the late 1800s and early 1900s during sampling, museum cataloguing or subsequent storage (e.g. Horton et al., 2009; Dietz et al., 2011). In the current study, the treatment and storage processes of the fur samples at the different museums had not been reported. We tried to reduce external Hg contamination following recommendations for sample preparation (Morton et al., 2002). Since, the observed THg levels in fur of the Arctic wolves lie within or below the biological plausible range of those reported in canids, i.e. grey and Arctic wolves (Gamberg and Braune, 1999; McGrew et al., 2015) and Arctic foxes (Hallanger et al., 2019; Treu et al., 2018), it is unlikely that external mercury contamination has occurred but still cannot be fully excluded.

2.2. Mercury analysis

The analysis for THg was performed on homogenized fur samples using a Direct Mercury Analyzer (DMA-80, Milestone, Italy) at the accredited Trace Element Laboratory at the Department of Ecoscience, Aarhus University, Denmark following the US-EPA Method 7473 (US-EPA, 1998). To minimize potential external Hg contamination from exogenous dust or soil particles each fur sample was thoroughly washed with ultra-clean de-ionized water and dried overnight at room temperature (Morton et al., 2002). Dried hair material was minced using stainless steel scissors into <1 mm pieces, wrapped in aluminium foil and stored at room temperature until Hg analysis. The analytical quality was controlled by concurrently analysing aqueous standard solutions (10 ng and 100 ng Hg, prepared from 1000 ± 4 mg L^{-1} stock solution, Sigma-Aldrich, Switzerland), procedural blanks, duplicates and the Certified Reference Material DORM-4 (fish protein from the National Research Council, Canada; 0.412 ± 0.036 μg Hg g^{-1} dry weight). Measurements were subsequently corrected for instrumental drift using results from the aqueous 10 and 100 ng standards applied to the low- and high-concentration cell, respectively (drift was always <10%). All samples were corrected for concurrent blanks (0.09 ± 0.09 ng; n = 9), and duplicates showed $RSD \leq$ 3,98%. The measured recovery percentage of the DORM-4 was 101.4 ± 5.0% (mean ± SD; n = 444 during 2017–2021). The laboratory is accredited by the Danish Accreditation Fond DANAK following ISO 17025 to DMA-80 analyses of Hg in biota with a detection limit of 0.001 μg g^{-1} dry weight and an extended measurement uncertainty (2*SD) of 20%. As part of the laboratory QA/QC, the lab participates twice a year in the international laboratory performance study programme QUASIMEME (www.quasimeme.org). The participation has proved excellent long-term measurement accuracy and precision (n = 15 during 2017–2021; assigned concentration from 0.009 to 0.931 μg g^{-1} dry weight; Z-scores from −1.0 to 0.7 with a mean of −0.1). All hair THg concentrations are expressed as μg g^{-1} dry weight (dw).

2.3. Stable isotope analysis

The analysis for stable carbon and nitrogen isotopes was carried out at the Stable Isotope Lab of the University of Koblenz-Landau, Germany. The analysis was carried out on homogenized fur material using a Flash 2000 HT elemental analyzer coupled via a ConFlo IV interface to a Delta V Advantage isotope ratio mass spectrometer (all Thermo Fisher Scientific, Bremen, Germany). Stable carbon and nitrogen isotope ratios were expressed using the standard δ notation (‰) against the standard international reference material Vienna Peedee Belemnite and atmospheric nitrogen, respectively. An internal reference material (i.e., casein) was measured in duplicate every ten samples revealing an imprecision (± SD) ≤ 0.06‰ for both $\delta^{13}C$ and $\delta^{15}N$.

2.4. Data analysis

Raw data for stable isotopes and THg for each Arctic wolf is given in Table S1. We conducted all statistical analyses and visualization using R version 4.0.2 (R Core Team, 2020) and SPSS version 27 (IBM, 2020), setting the level of significance at $P < 0.05$, while $P < 0.1$ was considered a trend given the limited sample size.

The established putative thresholds for assessing the health effects of Hg in terrestrial mammals are yet based on Hg liver levels as liver is usually the target organ in ecotoxicological studies. No transformation factor is, however, available for Artic or grey wolves to convert THg fur levels into liver concentrations. In order to assess the risk for Hg-mediated health effects, we therefore estimated THg liver levels in μg g^{-1} wet weight (ww) from fur concentrations (in μg g^{-1} dw) of the Arctic wolves using the formula from another Arctic canid species, the Arctic fox (Treu et al., 2018). This conversion is based on significant linear regression analyses ($P < 0.01$; n = 35) between Arctic fox liver and fur (Treu et al., 2018) with the formula: Log (THg Liver in μg g^{-1} ww) = 0.852 x log (THg Hair in μg g^{-1} dw) + 0.078. We then compared the estimated hepatic THg concentrations in Arctic wolves with the putative risk categories proposed by Dietz et al., 2019, 2022 which are based on risk quotient assessment for terrestrial mammals across the Arctic. Dietz et al., 2019, 2022 used hepatic Hg threshold values from studies in mink (*Mustela vison*, Wobeser et al., 1976; Wren et al., 1987) yielding five risk categories for Hg-mediated health effect: no-risk at THg liver burden (μg g^{-1} ww) below 4.2, low risk at 4.2–7.3, moderate risk at 7.3–22.7, high risk 22.7–30.5 and severe risk ≥30.5. These risk thresholds reflect effects on reproduction and adverse effects on body condition and behaviour (Dietz et al., 2019, 2022).

The raw $\delta^{13}C$ values of the analysed fur were corrected for the Suess effect, the temporal decline of atmospheric $\delta^{13}C$ values and consequently oceanic $\delta^{13}C$ values due to increasing anthropogenic CO_2 emissions in recent industrial times (Gruber et al., 1999). We used a mathematical correction established for the North Atlantic Ocean (Farmer and Leonard, 2011), suggesting a modelled global annual decline of – 0.007‰ for samples collected before 1950 (Tagliabue and Bopp, 2008) and – 0.026‰ for samples collected after 1950 (Körtzinger et al., 2003). We hereafter report the corrected $\delta^{13}C$ values as $\delta^{13}C_c$.

Given the temporal and spatial heterogeneity of the current data set attributable to sample availability, temporal trend analyses were beyond the

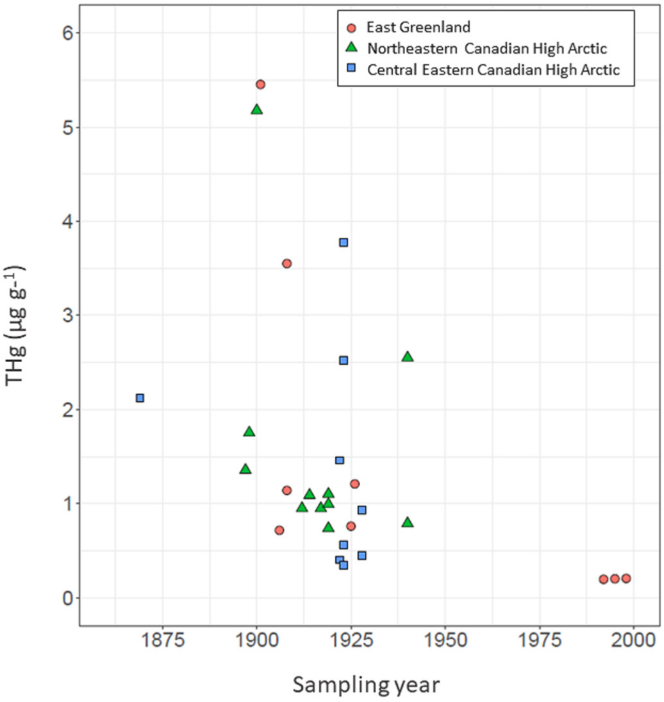

Fig. 1. Total mercury levels (µg g^{-1} dw) in Arctic wolves ($n = 30$) sampled during 1898–1998 in three study sites across Greenland and the Canadian High Arctic plotted by sampling year and study site.

scope of the present study. We divided the Arctic wolves according to their geographic sampling location, i.e. "Northeastern Canadian High Arctic" including North-West Greenland, Baffin Bay area, and the Canadian Arctic Islands Ellesmere and Axel Heiberg ($n = 11$), "Central Eastern Canadian High Arctic" ($n = 10$) including the Foxe Basin (North-West Nunavut, Baffin, Baffin Island, North-Western-Passage) as well as one individual (MS01) from West Greenland, and "East Greenland" ($n = 9$) .

We explored data for outliers (defined as data points exceeding 4-fold *SD*), heterogeneity of variance and normality following procedures described by Zuur et al. (2010). We then used linear models and included $\delta^{13}C_c$ as proxy for their feeding strategy and $\delta^{15}N$ as proxy for their trophic position. We further checked best distribution assumptions of THg concentrations as response variable (considering normal, gamma, ln, log_{10}, and Weibull with the package "goft" (González-Estrada and Villaseñor, 2018). No outlier was detected and THg was log_{10} transformed to meet model assumptions of normal distributed and homoscedastic residuals. Multicollinearity of predictor variables can be problematic for significance testing and should be avoided (e.g. Fox et al., 2010). Thus, all predictors (study sites, $\delta^{13}C_c$ and $\delta^{15}N$) were tested for possible intercorrelation (Pearson product-moment correlation test) (Zuur et al., 2010). Since $\delta^{13}C_c$ and $\delta^{15}N$ values were strongly correlated (Pearson's $r = 0.68$; $P < 0.01$; Table S2), we avoided including both dietary proxies as covariates in one model. The linear models included log_{10} THg as response variable and either $\delta^{13}C_c$ or $\delta^{15}N$ as covariate, respectively, and the three study regions as fixed factors. We then ran a set of candidate models with univariate, additive and multiplicative effects of fixed factors (regions) and dietary covariates ($\delta^{13}C_c$ or $\delta^{15}N$), and chose the model with the smallest Akaike's Information Criterion corrected for small sample sizes as the most parsimonious one (AICc; Burnham and Anderson, 2002) and largest Akaike weights (wAIC), specifying the amount of statistical confidence for the model with the lowest AICc value (e.g. Wagenmakers and Farrell, 2004). No model had a ΔAICc value (difference between AICc of the respective model and the smallest AICc of all models applied) smaller than 2 and thus no model averaging was performed. The overall significance of all created models was checked using analysis of variance by comparing the fitted model versus the null model (i.e. the intercept only model, $P < 0.05$). We did not compute more complex models to keep the number of models and predictors small relative to our sample size (Burnham and Anderson, 2002). Our sample size was sufficient though to fulfil the recommended minimal amount of data points for our model to avoid overfitting (e.g. Harrell, 2015). Model assumptions were verified by plotting residuals versus fitted values and using P—P and Q-Q plots (Zuur et al., 2010; Zuur and Ieno, 2016, Fig. S2A—C).

3. Results

Measured Hg fur levels are plotted by sampling year and study site of the Arctic wolves in Fig. 1 showing the temporal and spatial distribution of the samples. The concentrations, variance and ranges of THg, $\delta^{13}C_c$ and $\delta^{15}N$ are summarized in Table 1 and shown as heatmap (Fig. 2) and violin plots (Fig. S1A—C), indicating large variation. Among all candidate linear models, the most parsimonious one included only $\delta^{15}N$ as predictor ($n = 30$; estimate: 0.09; 95% CI: 0.03–0.15) and significantly outperformed the null model ($R^2 = 0.26$; $F_{1,28} = 9.60$; $P < 0.01$). In contrast, the fitted models including the predictors $\delta^{13}C_c$ and/or study region did not significantly vary from the null model as shown by ANOVA (all $P > 0.1$ Table 2). This goes against our hypothesis, that $\delta^{13}C_c$, $\delta^{15}N$ and geographical region explain the variability of THg exposure in fur. Trophic position ($\delta^{15}N$) explained 23% of the observed THg variance (*adjusted* $R^2 = 0.23$) with higher trophic position associated with a significant linear increase in THg levels (Table S3; Fig. 3). The THg liver levels calculated from THg fur concentrations using the conversion formula based on THg in Arctic fox tissues (Treu et al., 2018) are reported in Table S1 and ranged from 0.30 to 5.00 µg g^{-1} ww with a mean of 1.55 ± 1.25 µg g^{-1} ww. Based on the estimated liver THg levels the majority of Arctic wolves analysed in the current study fall into the no-risk category at levels below 4.2 µg g^{-1} (93%, $n = 28$, range: 0.20–3.78 µg g^{-1}) reported by Dietz et al., 2019, 2022, while two individuals fall into the low risk category at levels between 4.2 and 7.3 µg g^{-1} (7%, $n = 2$, levels: 5.18, 5.45 µg g^{-1}).

Table 1

Summary statistics for stable carbon ($\delta^{13}C_c$ corrected for the Suess effect and raw $\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope values of and THg concentrations in fur of Arctic wolves ($n = 30$) sampled during 1898–1998 in three study sites across Greenland and the Canadian High Arctic. NECHA: Northeastern Canadian High Arctic; CECHA: Central Eastern Canadian High Arctic; EGL: East Greenland.

	Mean	*SD*	Median	Range	Site	*n*	Mean	*SD*	Median	Range
$\delta^{13}C_c$ (‰)	−19.53	1.11	−19.63	−21.25–17.46	EGL	9	−19.72	1.04	−19.93	20.80 – −17.46
					NECHA	11	−20.08	1.18	−20.51	−20.08 to −20.51
					CECHA	10	−18.78	0.66	−18.85	−19.58 to −17.01
$\delta^{13}C$ (‰)	−19.98	1.24	−20.08	−21.75–17.73	EGL	9	−20.50	1.37	21.00	−21.75 to −17.73
					NECHA	11	−20.41	1.18	−20.86	−21.55–17.88
					CECHA	10	−19.05	0.58	−19.12	−19.83–18.29
$\delta^{15}N$ (‰)	+7.69	2.17	+7.24	+5.29 – +12.92	EGL	9	+7.03	2.71	+5.69	+5.29 – +12.92
					NECHA	11	+7.77	1.66	+7.50	+5.99 – +12.02
					CECHA	10	+8.18	2.20	+7.54	+5.49 – +12.21
THg (µg g^{-1} dw)	1.46	1.34	0.97	0.20–5.45	EGL	9	1.49	1.81	0.76	0.20–5.45
					NECHA	11	1.59	1.30	1.09	0.34–5.18
					CECHA	10	1.29	1.17	0.74	0.24–3.78

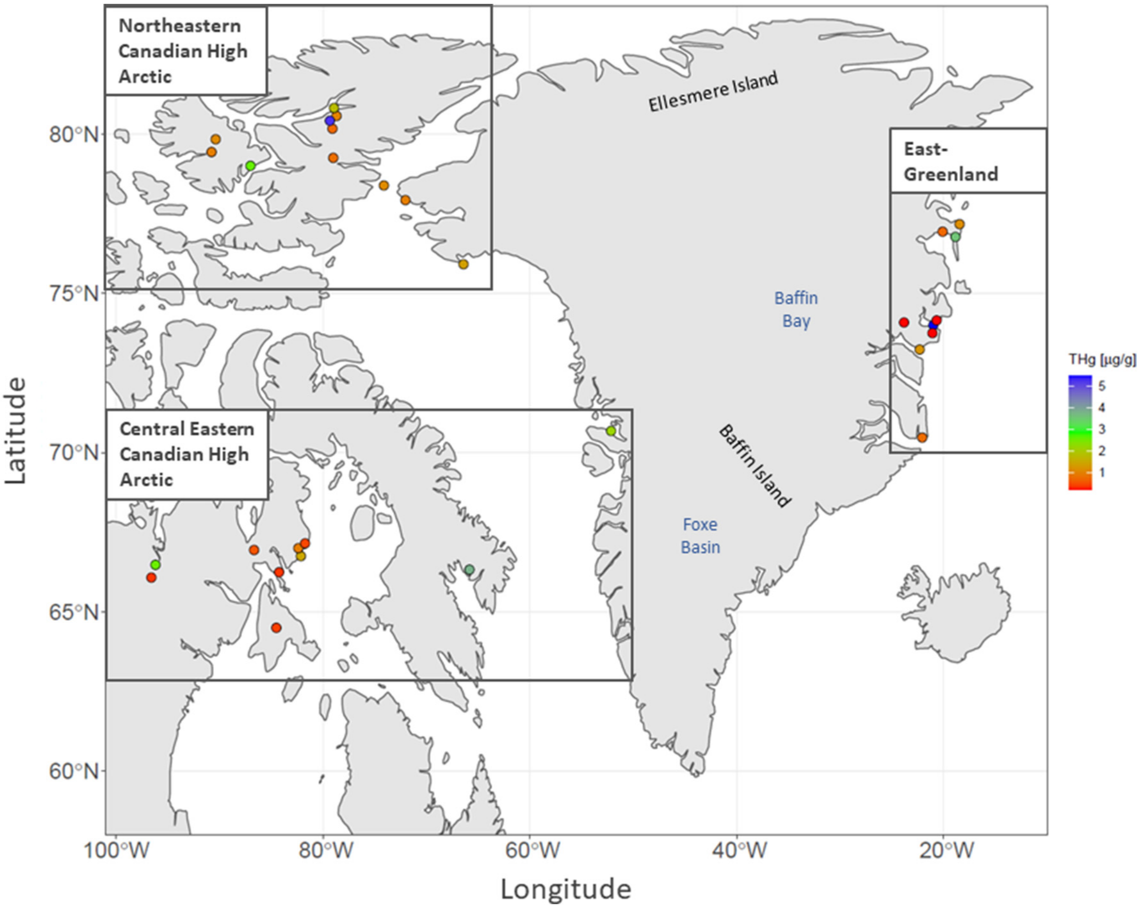

Fig. 2. Map showing origins of the Arctic wolf fur samples ($n = 30$) collected during 1869–1998 in Greenland and the Canadian High Arctic, composing three study areas (squares) and THg levels (µg g^{-1} dw) as heatmap.

4. Discussion

4.1. Mercury concentrations in Arctic wolves

The study relies on unique specimens opportunistically available at different museums and scientific collections which explains the spatial and temporal heterogeneity of the data. Furthermore, due to current lack of a systematic monitoring on Arctic wolves and strict conservation and hunting restrictions only three recent samples (post 1992) could be obtained while the remaining the wolves ($n = 27$) were collected before 1941.

There is a general paucity of information on Hg exposure levels in terrestrial Arctic predators, and no data is available on Hg contamination in Arctic wolves from the High Arctic. The only available study on hepatic THg exposure in Low Arctic wolves ($n = 19$) collected samples during

Table 2
Model selection of candidate models identifying predictors of $\log_{10}$ THg concentrations in Arctic wolves from Greenland and Canadian High Arctic. Among the potential candidate variables are stable carbon values corrected for Suess effect ($\delta^{13}C_c$) and stable nitrogen isotope values ($\delta^{15}N$), study site (Northeastern Canadian High Arctic, Central Eastern Canadian High Arctic, East Greenland) of Arctic wolf fur collected during 1869–1998. We report number of estimated coefficients (k), *ΔAICc* (difference between Akaike Information Criterion corrected for small sample size (AIC_c) of the respective model and the smallest AICc of all models applied), Akaike weight (wAIC) specifying the amount of statistical confidence for the model with the lowest AICc value, log Likelihood (*LogLik*), adjusted variance explained (R^2_{adj}) and *P* values of analysis of variance comparing variance of the fitted versus the null model. Candidate models are ranked by smallest ΔAIC_c.

Candidate model	k	*ΔAICc*	*wAIC*	*LogLik*	R^2_{adj}	*P*
$\sim \delta^{15}N$	1	0.00	0.704	−9.68	0.23	< 0.01
$\sim \delta^{15}N$ + site	2	4.28	0.083	−8.68	0.21	0.02
$\sim \delta^{15}N$: site	1	4.45	0.076	−8.68	0.2	0.03
$\sim \delta^{13}C_c$	1	8.98	0.008	−14.13	0.01	> 0.1
~ site	1	10.12	0.004	−13.23	0.01	> 0.1
$\sim \delta^{13}C_c$: site	1	11.35	0.002	−13.72	0.1	> 0.1
$\sim \delta^{13}C_c$ + site	2	13.78	0.001	−13.84	0.01	> 0.1

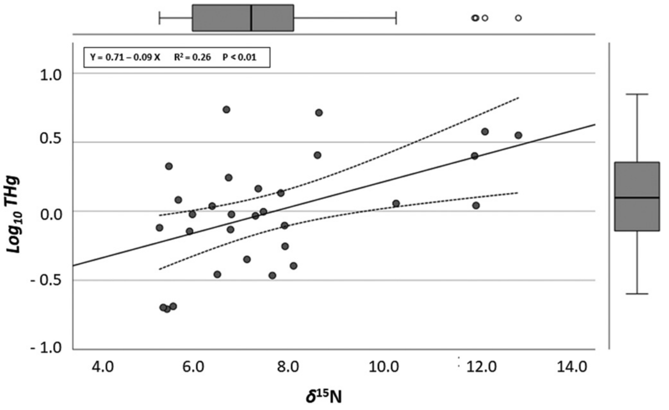

Fig. 3. Linear relationship between *log₁₀* THg concentrations (µg g^{-1} dw) and $\delta^{15}N$ values (‰) in Artic wolf fur ($n = 30$) collected during 1869–1998 in the Greenlandic and Canadian High Arctic. The plot shows the regression line (black), 95% confidence interval (dashed line) and statistical parameters. Boxplots are shown for *log₁₀* THg and $\delta^{15}N$, respectively. For each boxplot boxes represent 50% of data points, black lines: median; whiskers: 1,5 x inter-quartile range, white circles: data points > 3 x inter-quartile range.

1993–1994 in Yukon (Low Arctic Canada), reported mean THg liver levels in adult male wolves of 0.23 ± 0.28 μg g $^{-1}$ dw and 0.15 ± 0.03 μg g $^{-1}$ dw in adult females (Gamberg and Braune, 1999). Given that the current existing data are on different tissues, meaningful comparisons cannot be made, and we can only cautiously estimate how these data compare to the ones reported here. Our reported average fur THg concentration of 1.46 ± 1.39 μg g $^{-1}$ dw resulted in estimated THg liver levels of 1.55 ± 1.25 μg g $^{-1}$ ww or 0.44 ± 0.36 μg g $^{-1}$ dw (assuming a water content of 75% (Dainowski et al., 2015)), following a conversion from reported fur to hepatic concentrations for Arctic foxes (Treu et al., 2018). Again, while caution should be taken with such extrapolation, Hg levels in the wolves in the current study were approximately two to threefold higher than those from Yukon. The wolves in the study by Gamberg and Braune (1999) in Yukon (Canada) were all sampled in inland, while in our study all individuals were assumed to have access to the coast based on the sampling locations and reported home ranges of Arctic wolves (Mech and Cluff, 2011), and thus to marine dietary resources, which generally tend to accumulate higher Hg levels (Jæger et al., 2009; Rigét et al., 2004, 2005). Moreover, Yukon is located in the far West of Canada, where several wildlife species show generally lower Hg body burden than in East Canada (Brown et al., 2018; Evans et al., 2005), which might further explain why the Yukon wolves had lower putative Hg residues than the studied Arctic wolves. Altogether, the observed THg fur levels in Arctic wolves lie within the ranges reported in various tissues of terrestrial Arctic wildlife, typically below 5.00 μg g $^{-1}$ dw (reviewed by Braune et al., 2015, Rigét et al., 2011).

A technical issue potentially influencing the Hg hair levels may be external contamination of the archived hair samples. Some authors have demonstrated that washing procedures may significantly reduce external Hg contamination but cannot totally remove all Hg even if reagents containing sulfhydryl groups that strongly bind to Hg (e.g. cysteine or 2-mercaptoethanol (Li et al., 2008; Morton et al., 2002), are used. This is because Cysteine residues in hair which make up approximately 14% of human hair, may bind Hg (Li et al., 2008). However, as all samples underwent the same procedure there would be no individual sample bias affecting the results. Still, we cannot exclude that a small portion of the measured THg derives from external contamination.

We did not distinguish the most toxic form, MeHg, analytically in the current study, though earlier works reported the proportion of MeHg in hair of polar bears to be as high as 97.1% (Dietz et al., 2011) and 75% (Bechshoft et al., 2019) of the total Hg. As such, hair is a promising tissue to assess potential exposure-associated health effects. Subclinical neurological effects have been measured in polar bear brains at THg concentrations of 6.00 μg g $^{-1}$ in hair (Dietz et al., 2013b), while clinical neurological effects in mink occurred at THg hair concentrations above 30.00 μg g $^{-1}$ (Basu et al., 2007). The latter are all well above the THg concentrations we found in the Arctic wolves' fur. Moreover, the comparison of the estimated Arctic wolves' liver levels with putative risk thresholds for Hg-mediated health effects proposed by Dietz et al., 2019, 2022 indicates that the majority (93%) of Arctic wolves analysed in the current study were not at risk (THg < 4.2 μg g $^{-1}$ ww), and only 7% at low risk (at 4.2–7.3 μg g $^{-1}$ ww). However, again due to potentially different toxicokinetic behaviour of Hg and its forms in Arctic foxes and as compared to Arctic wolves, limitations remain on the use of Hg tissue to fur transformation formulas derived from predator species other than wolves.

4.2. Drivers of mercury levels

Previous studies report that environmental and biotic THg concentrations are highest in the Western Arctic including High Arctic Canada (review by AMAP, 2018; Dietz et al., 2000, 2013b; Rigét et al., 2005). Particularly polar bears and ringed seals (*Pusa hispida*) from the Northern Canadian Archipelago around the Jones and Lancaster Sound and in Baffin Bay area (covering the study area Northeastern Canadian High Arctic in the present study) and Foxe Basin/Baffin Island (covering the Central Eastern Canadian High Arctic study area) have higher THg liver and blood levels as compared to East Greenland (AMAP, 2018; Dietz et al., 2019, 2022). Our study, however, indicates THg concentrations in Arctic wolves' fur to not vary geographically. This finding should however be nuanced by the spatiotemporal heterogeneity of the data, the lack of sex and age information and the relatively small sample size ($n = 30$).

The observed ranges and variability of $\delta^{13}C_c$ in Arctic wolf fur indicate that the wolves fed across ecosystem on prey with both marine and terrestrial carbon sources. This also resulted in variability in $\delta^{15}N$ values, supporting the general observation that as opportunistic scavenging carnivores Arctic wolves show considerable dietary plasticity among and within populations. Similar observations were reported for grey wolves from Alaska most likely in response to variation in the distribution and density of local food resources in the study areas (Dalerum et al., 2018; Dalerum and Angerbjörn, 2000; Watts et al., 2010; Watts and Newsome, 2017). Carnivores consuming a larger proportion of their diet from the marine ecosystem are expected to have higher Hg body burdens, since marine biota also tend to have higher levels of Hg (Lehnherr, 2014; Brown et al., 2018). Against our hypothesis that THg would increase with marine dietary carbon sources (i.e., increasing $\delta^{13}C_c$ values), as shown for grey wolves from Alaska (McGrew et al., 2014), $\delta^{13}C_c$ did not explain the variation of THg exposure in the studied Arctic wolves. Our results, again which are potentially biased due to a small sample size and large spatial and temporal heterogeneity contrast findings in Arctic foxes from Svalbard, where $\delta^{13}C$ value was shown to be the most important predictor explaining variation in hepatic THg levels and terrestrial food consumption reduced THg loads (Hallanger et al., 2019). Accordingly, THg fur concentrations in Arctic foxes from Iceland and the Commander Island in the Bering Sea were also related to their feeding habits (Bocharova et al., 2013). The relatively low $\delta^{13}C_c$ values observed in the Arctic wolves in the present study as compared to the aforementioned works indicate, that the contribution of marine food items in the overall wolf diet seemed to be minor, which in turn might explain why $\delta^{13}C_c$ had no significant effect on the Hg exposure. This assumption concords with dietary studies on wolves from Greenland, Western Nunavut and Canadian High Arctic showing that wolves preferentially fed on terrestrial prey, i.e. caribou and muskoxen, and to a minor part on Arctic hares and lemmings, while marine food items like carcasses of marine mammals or water birds were generally used to a lesser extent (Dalerum et al., 2018; Marquard-Petersen, 1998; Samelius and Alisauskas, 2000). Still, the larger temporal and spatial scale of our study impedes direct comparisons with the aforementioned studies and caution needs to be taken when interpreting the SI data.

Since generally, the carbon and nitrogen cycles in aquatic and terrestrial ecosystems are empirically linked in many ways as integral elements of a broader set of nutrient dynamics, (Guiry, 2019) one would expect an effect of the interaction between the two SI on Hg levels in the studied wolves, i.e. an increase of Hg levels with increasing carbon and nitrogen SI values in individuals feeding on higher-trophic marine prey. However, due to the small sample size and large spatial temporal heterogeneity of the samples in the present study shown to bias δ^{13} C and δ^{15} N values and statistical power (Guiry, 2019; Kelly, 2011; Krzywinski and Altman, 2013; Newsome et al., 2010) and due to the strong correlation of both variables reducing the reliability of models applied (Zuur et al., 2007), study site was prioritized rather than pursuing the interaction between both SI to assess the effect on Hg exposure.

Previous research indicates that Hg biomagnification occurs in Arctic food webs via uptake by primary producers and subsequent consumption by invertebrates, fish, birds and marine mammals and terrestrial carnivores (e.g. Atwell et al., 1998). This is because particularly MeHg is lipophilic and therefore bioaccumulates in tissues of organisms due to the high affinity to protein thiol groups, following absorption in the gut, but is slowly eliminated from the body (Hammerschmidt and Fitzgerald, 2006). Our study demonstrates that $\delta^{15}N$ was a strong, positive driver of THg concentrations in Artic wolves' fur, supporting the expected bioaccumulation and biomagnification behaviour of Hg (Jæger et al., 2009; Lavoie et al., 2013; Ruus et al., 2015; Atwell et al., 1998; Burnham et al., 2018; Ruus et al., 2015). THg and MeHg were also found to biomagnify through the food web on Ellesmere and in Baffin Bay area (covering the North-West study area of the current study) sampled in 1998, with positive relationships between log THg and log MeHg concentrations versus δ^{15}N (Campbell et al., 2005). Our results are well consistent with observations made in grey wolves from Alaska showing a significant increase in hepatic THg levels with increasing δ^{15} N values which was assumed to be linked to a cross-ecosystem utilization of food resources, particularly when feeding at higher trophic marine prey (McGrew et al., 2014). Other works on marine Arctic carnivores also demonstrated an increase of THg with higher trophic level prey, e.g. in polar bears (Lippold et al., 2020; McKinney et al., 2017; Yurkowski et al., 2020) and in narwhals (Dietz et al., 2021).

The large variance of the THg fur concentrations among individual wolves ranging from 0.10 to 5.45 μg g^{-1} dw is assumed to be related to the interaction of several biotic and abiotic factors, since concentrations of THg and MeHg in Arctic biota are generally controlled by complex interacting processes which act on one or both of the primary regulating factors including the size of the abiotic pool of THg/MeHg available for bioaccumulation, and the transfer of MeHg through aquatic food webs (St. Louis et al., 2011; Kirk et al., 2012; Lehnherr, 2014; Loseto et al., 2008a, 2008b). The size of the abiotic MeHg pool, representing a bottom-up control on MeHg concentrations in biota (Kirk et al., 2012; Sluis et al., 2013), is determined by the atmospheric delivery of Hg(II) to Arctic aquatic ecosystems, riverine, and oceanic sources, sea ice cover, as well as the methylation efficiency within Arctic ecosystems (Lehnherr, 2014). Thus, despite the observed biomagnification as major driver of Arctic wolves Hg fur levels, other determinants explain the remaining 76% of the observed variance in fur THg levels, are most likely associated with differences in (local) geographical, ecologic, biological and climatic conditions, global emission patterns and dietary differences of Arctic wolves among the three study areas, owing to distribution and density of local food resources and their THg loads. We assume, that also foraging behaviour and life history of the wolves as well as their age affected their THg exposure levels, since these are usually important in explaining Hg and MeHg concentrations in biota (Loseto et al., 2008a, 2008b; St. Louis et al., 2011; Swanson and Kidd, 2010).

5. Conclusions

To our knowledge, this is the first data reported on Arctic wolves from the High Arctic investigating Hg exposure and its potential dietary pathways, covering a large spatial-temporal scale (1869–1998). The present study remains limited, though, due to both the small sample size and its temporal and spatial heterogeneity in Hg data. Nonetheless, the unique historic material has shown useful to fill an important gap in the understanding of concentrations and potential drivers of Hg in Arctic terrestrial wildlife. Our data, however, allow us to interpret relationships of Hg biomagnification within this Arctic terrestrial, sentinel species, providing yet another clear perspective of global Hg pollution affecting remote and pristine species. Moreover, our findings further support the promising use of archived fur samples in retrospective risk assessment, and we encourage further research in this direction especially as novel analytical methods become accessible.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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CRediT authorship contribution statement

Gabriele Treu: Conceptualization, Writing - Original Draft; Review & Editing; Formal analysis (statistical analysis, modelling), Data Curation, Visualization, Funding acquisition; **Thomas Gräff**: Writing - review & editing, Formal analysis (statistical modelling), Visualization, Data Curation; **Igor Eulaers:** Conceptualization, Investigation, Writing - Original Draft, Review & Editing, Formal analysis (Chemical analysis), Validation; Supervision, Funding acquisition, **Jochen P Zubrod, Ralf Schulz:** Performance and supervision of stable isotope analysis, Review & Editing**. The remaining authors**: Review & Editing.

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