Science of the Total Environment 687 (2019) 77–86

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Temporal trends of mercury differ across three northern white-tailed eagle (*Haliaeetus albicilla*) subpopulations

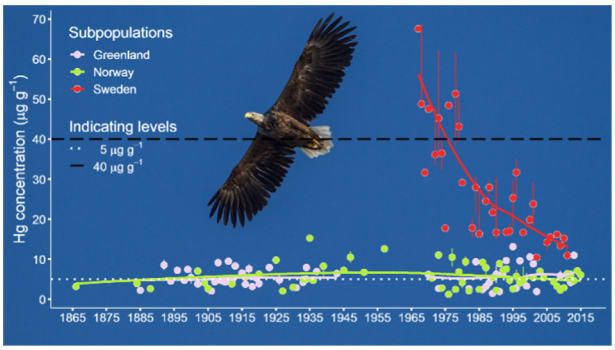
Jiachen Sun [a,*], Jan Ove Bustnes [b], Björn Helander [c], Bård-Jørgen Bårdsen [b], David Boertmann [d], Rune Dietz [d], Veerle L.B. Jaspers [e], Aili Lage Labansen [f], Gilles Lepoint [g], Ralf Schulz [h], Jens Søndergaard [d], Christian Sonne [d], Kasper Thorup [i], Anders P. Tøttrup [i], Jochen P. Zubrod [h], Marcel Eens [a], Igor Eulaers [d,**]

[a] Behavioural Ecology & Ecophysiology Group, Department of Biology, University of Antwerp, Universiteitsplein 1, BE-2610 Wilrijk, Belgium
[b] Arctic Ecology Department, Norwegian Institute for Nature Research (NINA), FRAM Centre, NO-9296 Tromsø, Norway
[c] Environmental Research & Monitoring, Swedish Museum of Natural History, Frescativägen 40, PO Box 50007, SE-104 05 Stockholm, Sweden
[d] Department of Bioscience, Arctic Research Centre, Aarhus University, Frederiksborgvej 399, PO Box 358, DK-4000 Roskilde, Denmark
[e] Environmental Toxicology Group, Department of Biology, Norwegian University of Science and Technology, Høgskoleringen 5, NO-7491 Trondheim, Norway
[f] Greenland Institute of Natural Resources, Kivioq 2, PO Box 570, GL-3900 Nuuk, Greenland
[g] MARE Centre, Oceanology, University of Liège, Allée de la Chimie 3, BE-4000 Liège, Belgium
[h] iES, Institute for Environmental Sciences, University of Koblenz-Landau, Fortstrasse 7, DE-76829 Landau in der Pfalz, Germany
[i] Center for Macroecology, Evolution and Climate, Natural History Museum of Denmark, University of Copenhagen, Universitetsparken 15, DK-2100 Copenhagen, Denmark

HIGHLIGHTS

- 150 y of Hg exposure using feathers were reconstructed for white-tailed eagle.
- Significant trends in feathers of Norwegian and Swedish eagles were observed.
- For Greenland individuals had higher concentrations recently.
- Distinctively higher concentrations were present in the Swedish subpopulation.
- Decreasing Hg, δ^{13}C, and δ^{15}N were found for the Norwegian subpopulation.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 7 March 2019
Received in revised form 2 June 2019
Accepted 2 June 2019
Available online 4 June 2019

Editor: Mae Sexauer Gustin

Keywords:
δ^{13}C
δ^{15}N

ABSTRACT

The spatiotemporal trends of mercury (Hg) are crucial for the understanding of this ubiquitous and toxic contaminant. However, uncertainties often arise from comparison among studies using different species, analytical and statistical methods. The long-term temporal trends of Hg exposure were reconstructed for a key sentinel species, the white-tailed eagle (*Haliaeetus albicilla*). Body feathers were sampled from museum collections covering 150 years in time (from 1866 to 2015) from West Greenland ($n = 124$), Norway ($n = 102$), and Sweden ($n = 87$). A significant non-linear trend was observed in the Norwegian subpopulation, with a 60% increase in exposure occurring from 1866 to 1957 followed by a 40% decline until 2015. In the Swedish subpopulation, studied at a later period, the Hg exposure showed a drastic decline of 70% from 1967 to 2011. In contrast, no significant trend could be observed in the Greenland subpopulation. The additional analysis of dietary proxies (δ^{13}C and δ^{15}N) in general increased performance of the temporal trend models, but this was dependent on the

* Correspondence to: J. Sun, Department of Biology, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium.
** Correspondence to: I. Eulaers, Department of Bioscience, Arctic Research Centre, Aarhus University, Frederiksborgvej 399, PO Box 358, 4000 Roskilde, Denmark.
E-mail addresses: Jiachen.Sun@uantwerpen.be (J. Sun), ie@bios.au.dk (I. Eulaers).

https://doi.org/10.1016/j.scitotenv.2019.06.027

Hg
Stable isotope
Feather
Museum collection

subpopulation and study period. The downward trend of Hg coincided with the decreasing δ^{13}C and δ^{15}N in the Norwegian subpopulation, suggesting a potential dietary mitigation of Hg contamination. Hg exposure in both the Greenland and Norwegian subpopulations was consistently below the suggested threshold for adverse health effects (40.0 μg g^{-1}), while the maximum exposure in the Swedish subpopulation was distinctively elevated (median: 46.0 μg g^{-1}) and still remains well above natural background concentrations (maximum 5.0 μg g^{-1}).

1. Introduction

Mercury (Hg) contamination has become a ubiquitous problem since the industrial revolution (Boening, 2000; Dietz et al., 2009). Historically, Europe and North America were the major Hg emitters, making, for instance, the northern marine environment of particular concern for Hg pollution (AMAP, 2011; Beattie et al., 2014; Dietz et al., 2018; Farmer and Leonard, 2011). There has been an increasing focus on temporal trends in Hg exposure (Berg et al., 1966; Dietz et al., 2006; Dittmann et al., 2012; Lindqvist et al., 1991; Rigét et al., 2007, 2011; Westermark et al., 1975). However, the use of different study species, analytical and statistical methods, and vastly differing observation periods, makes it difficult to compare temporal trends across regions and to draw conclusions on future scenarios under different political, industrial and climate regimes.

Predatory birds are excellent sentinels providing first-hand information on changing environmental conditions and pressures (Kovács et al., 2008). Top predators are prone to increasing concentrations of Hg in the environment by integrating Hg in their body through digesting contaminated prey (Dietz et al., 2000, 2018; Monteiro and Furness, 2001). The white-tailed eagle (*Haliaeetus albicilla*) is a Palearctic species occurring mainly in marine coast and freshwater habitats and feeding predominantly on fish and waterfowl (Helander, 1983; Sulkava et al., 1997; Wille and Kampp, 1983; Willgohs, 1961); however, its diet can vary over time and geographic region (Helander, 1983; Mlíkovský, 2009). It is also a key sentinel species in the environmental monitoring of various contaminants including Hg (Berg et al., 1966; Helander et al., 1982; Koivusaari et al., 1976). Since diet is a major pathway of Hg uptake, dietary shifts can affect Hg concentrations (Braune et al., 2014). It is therefore important to study ecological tracers such as the stable nitrogen and carbon isotopes (δ^{15}N and δ^{13}C) as proxies of diet, concurrently with Hg exposure, when addressing the spatiotemporal patterns of Hg contamination (Burgess et al., 2013; Thompson et al., 1998; Vo et al., 2011).

Feathers have long been used to monitor Hg in the environment (Appelquist et al., 1984; Berg et al., 1966; Goede and De Bruin, 1984; Johnels et al., 1979; Lindberg and Odsjö, 1983; Westermark et al., 1975). Hg is deposited into the feathers via blood circulation during feather growth and is bonded with the keratin molecules almost exclusively in its organic form (methyl-Hg - MeHg) (Johnels et al., 1979; Thompson and Furness, 1989; Westermark et al., 1975). MeHg is bioavailable and bioaccumulative in aquatic food chains, and it accounts for >90% of total feather Hg concentrations (Renedo et al., 2017; Thompson and Furness, 1989). Moulting is a major detoxification pathway as over 50% of a bird's total body burden of Hg can be excreted via the replacement of plumage, depending on the species and exposure levels (Braune and Gaskin, 1987; Honda et al., 1986). In addition, δ^{15}N and δ^{13}C of feather keratin reflect a bird's feeding behaviour during feather growth (Mizutani et al., 1990, 1992; Peterson and Fry, 1987). Since keratin is resistant to biochemical and physical degradation (Crewther et al., 1965), both the sequestered Hg burden (Appelquist et al., 1984) and the stable isotope compositions (Thompson et al., 1995) remain chemically stable in feathers throughout time. This allows employing archived feathers to retrospectively establish the Hg exposure and dietary habits of an individual bird, and to investigate the relationship between them.

The Baltic Sea is one of the most contaminated marine areas in the world (Bignert et al., 2012; Elmgren, 1989; Koivusaari et al., 1976; Lehtonen et al., 2017). Factors contributing to the accumulation of contaminants in the Baltic sea include the small water volume in relation to precipitation area, the limited water exchange with the North Sea and a comparatively low water temperature (Jensen et al., 1972). Accordingly, exceptionally high concentrations of Hg have been reported in tissues from white-tailed eagle and its prey (Helander et al., 1982; Koivusaari et al., 1976). Downward trends have been reported in various species in the Baltic countries following national regulations (Johnels et al., 1979). But in remote regions, such as the Arctic and West Greenland, increasing trends of Hg have been reported due to factors such as long-range transport (LRT; Dietz et al., 2018) and changing climate (Provencher et al., 2014). Given the inconsistent pattern of Hg contamination, studies that provide direct comparison across regions are crucial for a better understanding of this global contaminant.

In the present study, we reconstructed and compared the long-term temporal trends of Hg exposure of white-tailed eagles from three northern subpopulations (the West Greenland, Norwegian and Central Swedish Baltic Sea coasts). The three subpopulations thus represent potentially different degrees of contamination influx from high (Swedish Baltic Sea) to low (West Greenland). We therefore expected to find variable Hg exposure and also potentially different temporal trends among these subpopulations. In addition, we investigated the temporal trends of Hg in relation to feather δ^{13}C and δ^{15}N. Finally, we evaluated the spatiotemporal trends of Hg with regard to the effect of exposure on the species and the implications for future monitoring.

2. Materials and methods

2.1. Feather collection and preparation

For the purpose of the present study we obtained body feathers from white-tailed eagle specimens found dead in West Greenland ($n = 124$; 1884–2013) and along the Norwegian coast ($n = 102$; 1866–2015), as well as moulted body feathers collected at individual nests during the breeding season at the Central Swedish Baltic Sea coast ($n = 87$ pairs; 1967–2011), see Fig. 1. The sample collection in the present study (Table Supplemental Information S1) therefore compiles feathers provided by the Natural History Museum (NHM) of Denmark, and Greenland Institute of Natural Resources, several Norwegian NHMs including the Bergen University Museum, the NTNU University Museum, the Oslo University NHM, the Rana Museum, the Stavanger Museum and the Tromsø University Museum, and the Swedish NHM. Inorganic Hg contamination from Hg containing preservatives has not been found in more recent museum skins, e.g. after 1936 (Head et al., 2011) or after 1980 (Vo et al., 2011), but was apparent in skins from the 19th and early 20th century (Bond et al., 2015; Head et al., 2011; Vo et al., 2011). Other studies found no evidence of inorganic Hg contamination in samples as early as 1910 through 1970 (Frederick et al., 2009) or 1901 through 1940 (Movalli et al., 2017). In the present study, it is unlikely that there has been inorganic Hg contamination in the early museum feathers as museum correspondence affirmed the lack thereof. The moulted feathers collected at nests in Sweden were stored in polyethylene bags upon collection and transferred to the

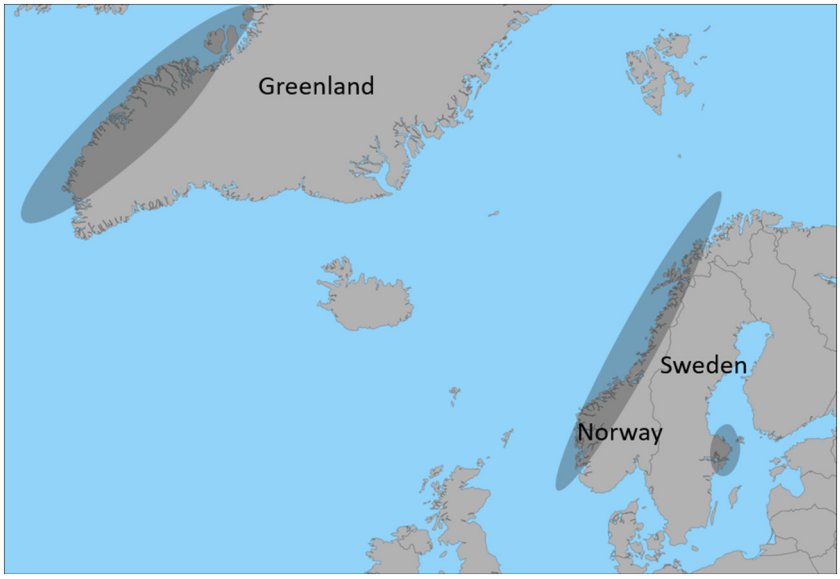


Fig. 1. Overview of original collecting locations (grey shades) of white-tailed eagle feathers sampled for the present study.

museum after 2000, and have not been subjected to any preservative treatment prior to or during storage at the museum.

From the West Greenland and Norwegian collections, only specimens with confirmed date of collection were used. Additional information on maturity, sex and more detailed location, were not always available, and such information was not included in further analysis (Table S1). The sampling and preparation followed the protocol from Espín et al. (2014). Briefly, ten body feathers (five breast and five back contour feathers) were plucked or cut at the base. From the Swedish collection of moulted feathers, a subsample of approximately ten body feathers originating from various body regions was collected at each nest for a specific year. These subsamples then represent the status (Hg exposure and diet) of the breeding pairs in their specific territory during the year before collection, i.e. the period when the feathers were grown assuming a one-year turnover rate of body feathers (Forsman, 1999). These nest sites (territories) were monitored annually during the period 1968–2012 with repetitive sampling from the same territories over the years. After sampling of the original collections for the present study all feathers were stored in polyethylene bags under dark, ambient temperature and humidity conditions until preparation for chemical analysis. During the preparation for chemical analysis for Hg and stable isotopes, the calamus of each feather was first removed using a pair of sterilized stainless-steel scissors (Bortolotti, 2010; Espín et al., 2014). Feathers were thoroughly cleaned using distilled water and stainless-steel forceps to remove possible external contamination with dirt particles. The cleaned feathers were dried overnight at ambient room temperature in a controlled laboratory environment, and subsequently cut into 1 mm pieces using stainless-steel scissors. Once homogenised, the feather material was stored in aluminium foil under dry, dark and ambient temperature conditions until chemical analysis.

2.2. Mercury analysis

Analysis for total Hg concentrations (referred to as Hg throughout this article) was performed at the Trace Element Lab of the Department of Bioscience, Aarhus University, Denmark. Body feather Hg concentrations were determined using a Milestone DMA-80 Direct Hg Analyser (Sorisole, Italy) following the U.S. EPA Method 7473 (EPA, 1998). Dry homogenised feather samples weighing on average 5.27 ± 0.42 mg (mean ± *SD*) were analysed. The instrumental analytical quality control was verified by analysing procedural blanks, duplicates, aqueous standards (10 ng and 100 ng Hg, prepared from 1000 ± 4 mg L^{-1} stock solution, Sigma-Aldrich, Switzerland), and Certified Reference Material (CRM; DORM-4, National Research Council, Ottawa, Canada). Procedural blanks and CRMs were analysed concurrently every 10 samples. All samples and CRMs were corrected for the average blank amount of Hg (0.07 ± 0.13 ng; $n = 131$) as well as for the recovery of aqueous standards (108.6 ± 1.3%; $n = 21$). The measured recovery percentage of the CRMs fell within the acceptable range (105.9 ± 2.3%; $n = 52$) of the certified value (0.410 ± 0.055 μg g^{-1} dry weight). Relative percent difference for duplicate samples ranged from 0.02% to 34.08% ($n = 13$). All body feather Hg concentrations are expressed in μg g^{-1}.

2.3. Stable isotope analysis

For the Greenland and Norwegian subpopulations, the analysis for stable carbon (^{13}C and ^{12}C) and nitrogen isotopes (^{15}N and ^{14}N) was carried out at the Stable Isotope Lab of the University of Koblenz-Landau, Germany. Stable isotope ratios of carbon (^{13}C:^{12}C) and nitrogen (^{15}N:^{14}N) in bulk homogenised feather material were determined using a Flash 2000 HT elemental analyser coupled via a ConFlo IV interface to a Delta V Advantage isotope ratio mass spectrometer (all Thermo Fisher Scientific, Bremen, Germany). For the Swedish subpopulation, the analysis was performed at the Laboratory of Oceanology of the University of Liège, Belgium, using an Isoprime 100 isotopic ratio mass spectrometry (Isoprime, UK) coupled in continuous flow to a Vario MICRO cube elemental analyser (Elementar, Germany). For all three subpopulations, 1.52 ± 0.10 mg of dry and homogenised feather material was crimped into a tin capsule and introduced to the elemental analysers. The stable isotope ratios for carbon and nitrogen are expressed as δ values (‰) relative to their respective international measurement standards Vienna Pee Dee Belemnite and atmospheric N_2. For the subpopulations in Greenland and Norway, 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 $\delta^{15}N$. For the subpopulation in Sweden, glycine was used as the internal reference material and was measured every 15 samples, showing an imprecision (± *SD*) ≤ 0.20‰ for both $\delta^{13}C$ and $\delta^{15}N$.

2.4. Statistical analysis

All statistical analyses, and plotting of results, were performed using R 3.5.1 (R Core Team, 2018). Data of the Greenland subpopulation were divided into two periods (1884–1943 and 1970–2013) due to the lack of

samples between 1943 and 1970. For each dataset the response variable (Hg concentration) was tested for various distribution assumptions (i.e. normal, gamma, log-normal and Weibull, package 'goft', Table S2), and the predictors (year of exposure, $\delta^{13}C$ and $\delta^{15}N$) were tested for correlation (Pearson product-moment correlation test, Fig. S1). We did this to assess potential effects of confounding and collinearity. Hg concentrations in all three populations followed a gamma or log-normal distribution. Therefore, we used the gamma family with a log-link in the statistical models (Fu and Moncher, 2004). $\delta^{13}C$ and $\delta^{15}N$ were strongly correlated in both the Greenland and Norwegian subpopulations, but not in the one from Sweden, thus, we avoided including both $\delta^{13}C$ and $\delta^{15}N$ in one model when studying the Greenland and Norwegian subpopulations. There were weaker correlations between year and $\delta^{15}N$ in the Greenland (1970–2013) and Norwegian subpopulations, thus when both covariates were present in models we performed extra validation on the collinearity. Two samples with unusual high Hg concentrations (>mean + 5*SD*) from the Greenland (1884–1948; 24.88 μg g^{-1}) and Norwegian subpopulations (26.10 μg g^{-1}) and one sample with low $\delta^{13}C$ (<mean − 5*SD*) from the Sweden subpopulation (−25.93) were regarded as outliers and removed from further analysis.

$\delta^{13}C$ values were corrected for the oceanic Suess effect, the temporal decline of atmospheric $\delta^{13}C$ values and consequently oceanic $\delta^{13}C$ values due to the large quantity of CO_2 released from fossil fuel burning in recent industrial times (Gruber et al., 1999). We used the mathematical correction established for the North Atlantic Ocean by Farmer and Leonard (2011), a modelled global annual decline of −0.007‰ was used for samples before 1950 (Tagliabue and Bopp, 2008) and −0.026‰ was used for samples after 1950 (Körtzinger et al., 2003). The magnitude of the correction for the Suess effect was 0.00–2.10‰, 0.00–2.28‰ and 0.32–1.47‰ for the Greenland, Norwegian and Swedish data, respectively.

The temporal trends of Hg for each subpopulations were evaluated using Generalized Additive Models (GAMs, package 'mgcv'; Wood, 2017). We used the restricted maximum likelihood (REML) method instead of generalized cross-validation (GCV) as REML avoids overfitting (Marra and Wood, 2011; Wood, 2011). We used cubic regression splines as part of the univariate smoothing modelling. For variable selection, we chose a double penalty approach that is computationally stable and has good predictive ability (Marra and Wood, 2011). Because of repetitive sampling of some territories in the Swedish collection, we included territory as a random effect variable in the models for the Swedish subpopulation. The assumptions underlying GAM (i.e. residual normality and homoscedasticity) and knot selections were visually validated using the gam.check function. In addition, we checked the observed concurvity (a generalization of collinearity) to validate model fit for year and $\delta^{15}N$ in the Greenland (1970–2013) and Norwegian subpopulations. Concurvity values of the models presented in the results were within the acceptable range.

3. Results

3.1. Spatial trends of Hg

Across all three subpopulations, Hg concentrations (median and mean; Table 1) were highest in the Swedish subpopulation, while concentrations in the Greenland and Norwegian subpopulations were similar to each other and substantially lower than in Sweden. Body feather concentrations of Hg in the Greenland and Norwegian subpopulations were consistently well below 20 μg g^{-1}. In contrast, values at the start of the study period in Sweden (median = 46.0 μg g^{-1}; Table 1) were more than five-fold higher than peak concentrations in the Norwegian subpopulation during the mid-1940s to mid-1960s (Table 1).

3.2. Temporal trends of Hg

In the Greenland subpopulation, year explained a limited part of the observed variation (adjusted R^2) in Hg concentrations: 1% and 3% of the variation in the period 1884–1943 and 1970–2013, respectively (Table 2). After integrating information on dietary plasticity ($\delta^{15}N$) into the model, there were still no significant temporal trends ($P = 0.11$; 1884–1943 and $P = 0.39$; 1970–2013) (Table 2; Fig. 2*A* and *B*). The concentrations of Hg in the Greenland subpopulation remained constant during both periods. The overall Hg concentrations were similar between the two periods (Fig. 2*A* and *B*).

In the Norwegian subpopulation, year explained more of the observed Hg variation (6%) and showed a significant effect also after controlling for $\delta^{15}N$ ($P = 0.01$; Table 2). Hg concentrations at the beginning of the study period (the 1860s–1880s) were similar to those in the Greenland subpopulation, but increased considerably (0.5% $year^{-1}$; Fig. 2*C*) and peaked in the mid-1950s. After this, a steadily decreasing trend (0.8% $year^{-1}$) is observed leading to a value at recent times (median=4.51 μg g^{-1}; 2013–2015) similar to that at the start of the study period (median=4.39 μg g^{-1}; 1866–1884; Fig. 2*C*).

In the Swedish subpopulation, the observed variation in Hg concentrations was mostly explained by year (59%) and its effect remained highly significant when integrating both $\delta^{15}N$ and $\delta^{13}C$ ($P < 0.01$; Table 2). With a shorter study period (starting in 1967) we observed a continuous and significant decreasing trend (Fig. 2*D*). Compared to the observed decrease in Hg in the Norwegian subpopulation from the mid-1950s to present, the decrease in the Swedish subpopulation after 1967 was much more pronounced: a 72.4% decrease over 44 years (2.9% $year^{-1}$; Fig. 2*D*). The decreasing rate was not uniform

Table 1
Body feather Hg concentrations (μg g^{-1}), summarized as median (with sample size *n*), mean and standard deviation (*SD*), at 10-year intervals for three white-tailed eagle subpopulations. "–" indicates periods with no available sample. The sample size *n* represents individual birds in Greenland and Norway, and breeding pairs in Sweden.

Period	Greenland		Norway		Sweden	
	Median (*n*)	Mean ± *SD*	Median (*n*)	Mean ± *SD*	Median (*n*)	Mean ± *SD*
1866–1875	–	–	3.17 (1)	3.17	–	–
1876–1885	3.75 (7)	3.68 ± 1.47	3.93 (2)	3.93 ± 0.74	–	–
1886–1895	4.87 (6)	5.62 ± 2.92	2.65 (1)	2.65	–	–
1896–1905	5.55 (9)	5.43 ± 2.38	6.53 (5)	4.99 ± 2.72	–	–
1906–1915	4.66 (20)	5.12 ± 2.55	3.84 (1)	3.84	–	–
1916–1925	5.37 (32)	5.52 ± 2.59	6.33 (5)	7.38 ± 2.33	–	–
1926–1935	4.24 (5)	4.95 ± 2.60	4.63 (9)	5.41 ± 3.89	–	–
1936–1945	6.44 (2)	6.09 ± 1.05	7.24 (5)	6.50 ± 2.57	–	–
1946–1955	–	–	8.10 (4)	8.62 ± 3.47	–	–
1956–1965	–	–	12.6 (1)	12.6	–	–
1966–1975	4.44 (2)	4.44 ± 2.50	5.33 (3)	6.38 ± 4.22	46.0 (16)	47.5 ± 21.5
1976–1985	5.98 (2)	5.98 ± 4.78	5.52 (14)	6.05 ± 4.56	31.4 (24)	33.4 ± 15.9
1986–1995	4.04 (14)	5.57 ± 4.00	7.08 (15)	6.12 ± 3.17	22.2 (16)	22.5 ± 8.04
1996–2005	3.32 (15)	5.66 ± 4.26	4.60 (14)	5.53 ± 2.50	17.2 (16)	19.4 ± 8.54
2006–2015	6.53 (8)	6.78 ± 3.26	4.62 (21)	4.69 ± 2.52	14.4 (10)	14.1 ± 2.75

Table 2
Comparison of generalized additive models with different covariates (year; $\delta^{15}N$; $\delta^{13}C$) and their capacity in predicting Hg concentrations in body feathers of white-tailed eagles from the West Greenland, the Norwegian and the Central Swedish Baltic Sea coasts. Final models selected for each subpopulation are shown in bold. The sample size *n* represents individual birds in Greenland and Norway, and breeding pairs in Sweden.

Subpopulation (period)	*n*	Coefficients		Smooth terms				Model performance		
		Estimate	*SD*		Edf	*F*	*P*	R^2	*D*	ΔAIC_c
Greenland (1884–1943)	82	1.66	0.05	Year	0.40	0.07	0.21	0.01	1.19%	1.46
		1.65	**0.05**	**Year**	**0.62**	**0.18**	**0.11**	**0.07**	**8.67%**	**0.00**
				$\delta^{15}N$	**1.67**	**0.47**	**0.08**			
		1.65	0.05	Year	0.45	0.09	0.19	0.03	4.51%	2.25
				$\delta^{13}C$	1.05	0.18	0.20			
Greenland (1970–2013)	41	1.75	0.10	Year	0.52	0.12	0.16	0.03	3.70%	2.24
		1.74	**0.10**	**Year**	**0.00**	**0.00**	**0.39**	**0.11**	**9.34%**	**0.00**
				$\delta^{15}N$	**0.75**	**0.32**	**0.06**			
		1.74	0.10	Year	0.29	0.04	0.26	0.11	10.7%	0.54
				$\delta^{13}C$	0.75	0.34	0.05			
Norway (1866–2015)	101	1.73	0.05	Year	1.46	0.74	0.01	0.06	7.06%	1.01
		1.73	**0.05**	**Year**	**1.82**	**0.79**	**0.01**	**0.11**	**12.2%**	**0.00**
				$\delta^{15}N$	**1.21**	**0.50**	**0.03**			
		1.73	0.05	Year	1.31	0.57	0.02	0.08	9.27%	1.88
				$\delta^{13}C$	1.16	0.22	0.16			
Sweden (1967–2011)	86	3.24	0.05	Year	1.80	18.4	<0.01	0.59	70.4%	5.12
		3.25	0.04	Year	1.72	14.3	<0.01	0.60	68.6%	1.19
				$\delta^{15}N$	0.90	1.52	<0.01			
		3.23	0.05	Year	1.89	21.3	<0.01	0.66	74.0%	2.77
				$\delta^{13}C$	1.61	1.24	0.01			
		3.24	**0.04**	**Year**	**1.90**	**17.1**	**<0.01**	**0.66**	**72.8%**	**0.00**
				$\delta^{15}N$	**0.88**	**1.45**	**<0.01**			
				$\delta^{13}C$	**1.54**	**0.84**	**0.02**			

Edf: estimated degree of freedom.
R^2: adjusted R^2.
D: deviance explained.
ΔAIC_c: the difference in AIC*c* values for each candidate model compared to the candidate model with the lowest AIC*c* value.

across years, as we observed a clearly steeper decline until about 1985 (3.8% year^{-1} from 1967 to 1985), afterwards the decrease has slowed down (2.2% year^{-1} from 1985 to 2011).

3.3. Spatiotemporal variation in Hg, $\delta^{13}C$ and $\delta^{15}N$

Including $\delta^{13}C$ or $\delta^{15}N$ resulted in the lowest AIC*c* values of fitted Hg temporal trend models, compared to when only year was used as a predictor (Table 2). However, ΔAIC*c* was less than two in the Greenland (1884–1943) and Norwegian subpopulations, which indicated essentially similar models. In contrast, in the Greenland (1970–2013) and Swedish subpopulations, ΔAIC*c* was considerably lower when including these proxies (Table 2). Moreover, there were no substantial differences in the model AIC*c* including either $\delta^{13}C$ or $\delta^{15}N$ in Greenland (1970–2013) and Norway, whereas including $\delta^{15}N$ resulted in better models in Greenland (1884–1943) and Sweden.

We observed significant temporal trends of $\delta^{15}N$ in Greenland (1970–2013), Norway and Sweden, and $\delta^{13}C$ in Norway and Sweden (all $P < 0.05$; Fig. 3). Trends lines of $\delta^{15}N$ and $\delta^{13}C$ appeared to be mostly constant in Sweden throughout the study period (Fig. 3*D*) whereas $\delta^{15}N$ appeared to be decreasing in Greenland (1970–2013; Fig. 3*B*). In comparison, the variation in both $\delta^{15}N$ and $\delta^{13}C$ values were much more pronounced in the Norwegian subpopulation (Fig. 3*C*), noticeably the clear downward trends since the 1960s. The decrease coincided with decreasing Hg concentrations in the Norwegian subpopulation (Fig. 2*C*). Median and mean $\delta^{13}C$ and $\delta^{15}N$ values per ten-year period are given in Table S3.

4. Discussion

4.1. Spatiotemporal trends of Hg

Despite the geographical proximity between Norway and Sweden, we observed remarkably different Hg concentrations in feathers of these two white-tailed eagle subpopulations. Since the Swedish feather collection has not been subjected to any preservative treatment, the feathers that contain the highest Hg values reflect high environmental concentrations rather than external contamination. Moreover, there is a lack of migratory movement of white-tailed eagles between Norway and Sweden, confirmed by an extensive international colour ringing program implemented since 1976 (Helander et al., 2003), as well as clear differences in genetic composition between the Norwegian and Swedish subpopulations (Hailer et al., 2007). Therefore, the considerably different Hg exposure in white-tailed eagles could be attributed to different anthropogenic Hg influxes in these two countries. Large amounts of Hg were emitted to the atmosphere in Sweden during the 1950s and 1960s, mainly from chlor-alkali plants and from metal production (Lindqvist et al., 1991). In accordance, we observed elevated Hg concentrations from 1967 and throughout the 1970s in the Swedish subpopulation. Although Hg use in agriculture and industry in Norway has been documented (Skurdal et al., 1985), it was likely much more restricted than that in Sweden. For instance, the agriculture and pasture lands in 1989 were approximately 10,000 km^2 and 34,000 km^2 in Norway and Sweden, respectively (Pettersson and Lehman, 1994). Relatively low Hg concentrations were previously reported in white-tailed eagles (2.4 μg g^{-1} in liver; median) from Norway during 1965–1983 (Frøslie et al., 1986), well below the concentrations reported in white-tailed eagles from the Stockholm Archipelago (60–235 μg g^{-1} in liver) during 1964–1966 (Falandysz et al., 1988). Hg concentrations observed here in body feathers from Swedish eagles during the 1960s to 1970s are comparable with the concentrations reported previously in tail feathers (Berg et al., 1966): 38.4 μg g^{-1} ($n = 1$; 1952) and 47.0–64.0 μg g^{-1} (mean = 51.6 μg g^{-1}; $n = 4$; 1964–1965). Hg exposure in eagles from prior to 1950, however, showed lower concentrations ranged 2.7–15.5 μg g^{-1} (mean = 6.6 μg g^{-1}; $n = 8$; 1832–1940) and 3.7–7.8 μg g^{-1} (mean = 5.8 μg g^{-1}; $n = 4$; 1942–1949). These concentrations are comparable to MeHg concentrations in primary feathers of adult white-tailed eagles from West Greenland that averaged 5.8 μg g^{-1} during 1875–1944 (Dietz et al., 2006). Although analysed with different methods and based on tail/primary feathers, results for early years in Sweden (before 1949) and Greenland (before 1944) correspond well with the Hg concentrations presented here (Table 1 and Fig. S2) in the

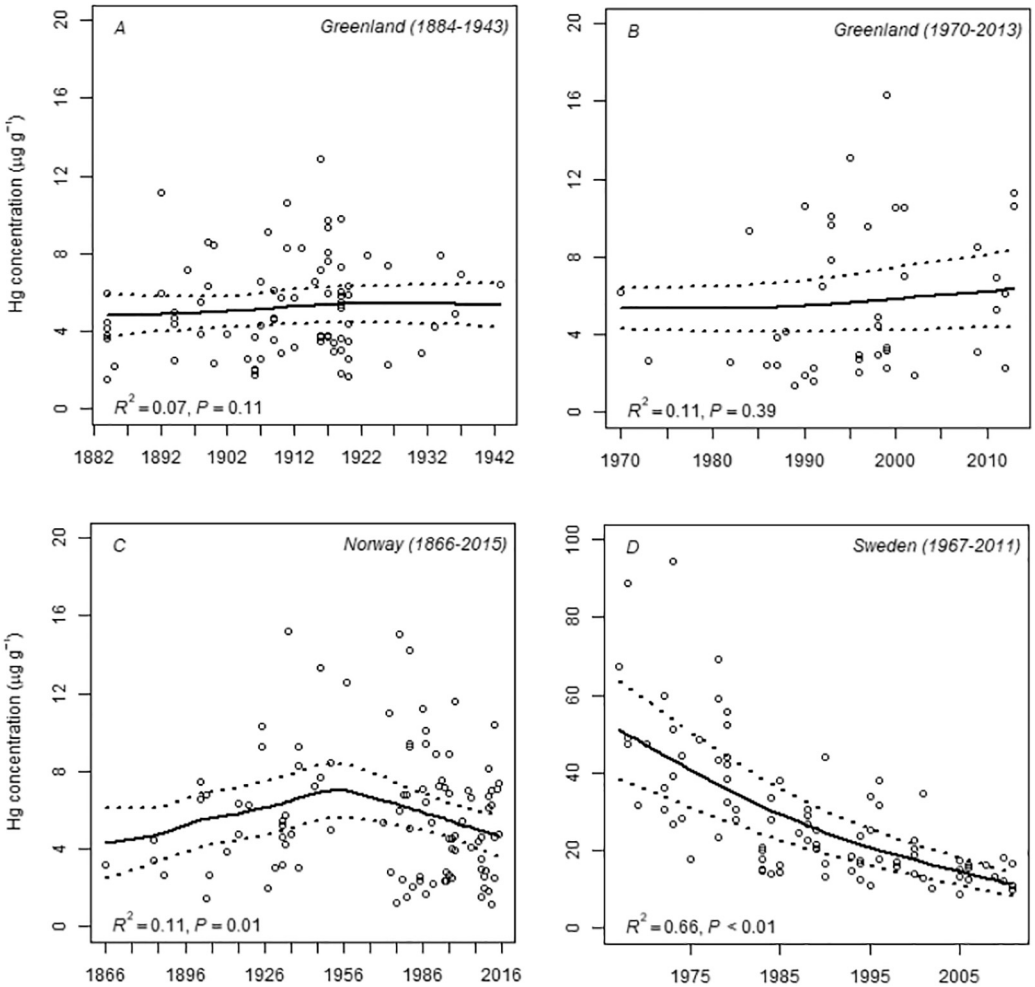


Fig. 2. Temporal trends of Hg in body feathers of white-tailed eagles from the West Greenland coast (*A* and *B*), the Norwegian coast (*C*) and the Central Swedish Baltic Sea coast (*D*). Open dots are original Hg concentrations. Trend lines (with a 95% prediction interval) were fitted using GAMs with year and δ^{15}N; δ^{15}N and δ^{13}C as covariates (per the outcome of the model selection; see Table 1).

body feathers of eagles from Greenland and Norway up to the 1940s and indicate no significant Hg influx even throughout the 1940s.

The observed mean 2.9% annual decrease in the Swedish subpopulation is in agreement with an overall significant decrease of Hg in herring (*Clupea harengus*) muscle from Ängskärsklubb within our study area (2.3% year^{-1}; 1971–2012; Bignert et al., 2014) and in guillemot (*Uria aalge*) eggs from Gotland just south of our study area (1.8% year^{-1}; 1969–2014; Bignert et al., 2016). On the other hand, we did not find significantly elevated Hg concentrations in Greenland as reported in a previous study: Dietz et al. (2006) reported significantly elevated MeHg concentrations in juvenile white-tailed eagles during 1980–2000 compared to 1880–1920, but not in immature or adult birds. Even though we sampled many of the same individuals as presented in Dietz et al. (2006) and extended the study period to 2013, we could not detect a clear difference between the two sampling periods 1884–1943 and 1970–2013 or any significant temporal variation within each period. This could be due to combining juveniles and adults in our analysis. No significant trends of Hg ($r = -0.05$, $n = 72$) were observed in livers of white-tailed eagles from Norway during 1973–1983 (Frøslie et al., 1986). In the present study, covering a longer period, a significantly decreasing trend in the Norwegian subpopulation was observed from the mid-1950s to recent times, albeit a very mild decline (0.8% year^{-1}) compared to the pronounced drop in Sweden. Nevertheless, the decreasing trends observed in both Sweden and Norway reflect that legal actions taken to control Hg emission, and improved technologies introduced in Europe (e.g. at the end of the 1960s in Sweden; Lindqvist et al., 1991) have been effective. A large contaminant-free feeding program was implemented in Sweden starting from 1970. This program aimed to reduce the contamination level in eagles and to increase the survival rate of juveniles by placing tons of uncontaminated carrion along the coast (Helander et al., 2003; Koivusaari et al., 1980). This conservation effort might have contributed additionally to the sharp decreasing Hg concentrations in the local subpopulation. However, in Sweden, despite the sharp decline, the legacy from the industrial past is carried on to the present day as clearly indicated by the still elevated concentrations in the Swedish subpopulation contrasting to those in the Greenland and Norwegian subpopulations.

4.2. Spatiotemporal dynamics of Hg in relation to δ^{13}C and δ^{15}N

Incorporating information on dietary proxies, especially the trophic level proxy δ^{15}N, in environmental contaminants studies has become increasingly popular (Braune et al., 2002, 2019; Rigét et al., 2007). In the present study, including δ^{13}C or δ^{15}N substantially improved model outcome for Greenland (1970–2013) and Swedish data, but not for Greenland (1884–1943) and Norway that suggests that the effect of dietary tracers is dependent on region and study period, even for the same species. In addition, the similar effect of δ^{13}C and δ^{15}N in predicting Hg variation in Greenland and Norway was expected due to their high inter-correlation, while when not correlated (e.g. the Swedish subpopulation), δ^{15}N resulted in better model performance than δ^{13}C. These findings indicate a likely more important role of trophic level in

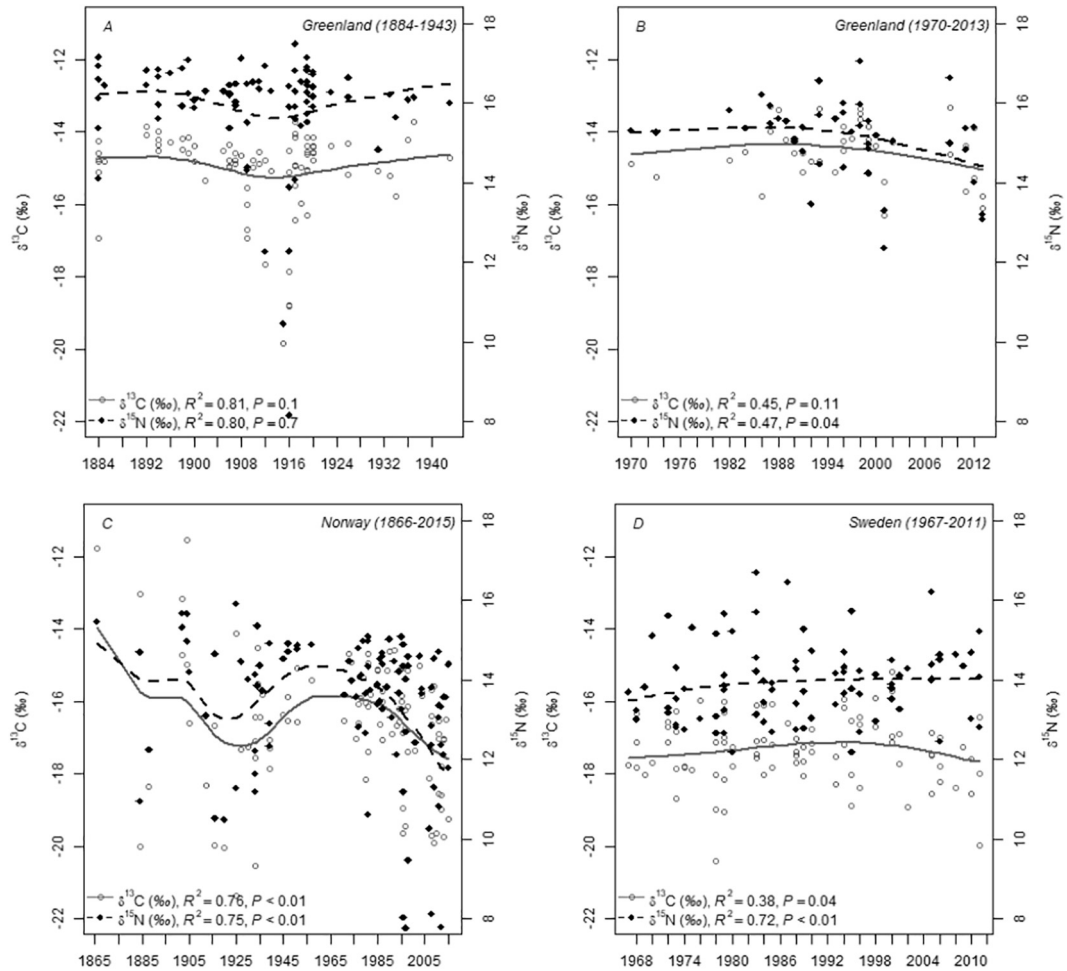


Fig. 3. Temporal trends of δ^{13}C (open dots) and δ^{15}N (closed dots) values of body feathers of white-tailed eagles (*Haliaeetus albicilla*) collected in West Greenland (*A* and *B*), the Norwegian coast (*C*), and the Central Swedish Baltic Sea coast (*D*). δ^{13}C values were detrended for the oceanic Suess effect. Trend lines were fitted using GAMs with year and δ^{13}C or year and δ^{15}N as covariates for the modelling of δ^{15}N and δ^{13}C, respectively.

explaining Hg exposure relatively to δ^{13}C in white-tailed eagles. All three studied subpopulations are from marine coastal areas, and all feed predominantly on fish and waterfowl as previously reported for Greenland (Wille and Kampp, 1983), Norway (Willgohs, 1961), and Sweden (Helander, 1983). Consequently, there appeared to be no marked variation in δ^{13}C due to major difference in terms of terrestrial versus aquatic diet predominance in the studied white-tailed eagles. Therefore, considerable variation in Hg concentrations driven by fundamentally different foraging habitat is unlikely.

In contrast to the decline of δ^{13}C and δ^{15}N reported in the German Baltic Sea during 1988–2016 (Corman et al., 2018), we observed relatively stable δ^{13}C and δ^{15}N values in white-tailed eagles from the Swedish Baltic Sea during 1967–2011. The pronounced Hg decline in the Swedish subpopulation therefore could have been majorly driven by the changing anthropogenic input as discussed above. On the other hand, the decreasing δ^{15}N could be linked to the stable Hg concentrations in Greenland observed during 1970–2013. In addition, δ^{15}N levels in the Greenland subpopulation during 1970–2013 were relatively lower than during 1884–1943 that could also be a reason for the fact that significantly elevated Hg concentrations were not observed in the later period. Furthermore, the decreasing Hg in the Norwegian subpopulation after mid-1950s occurred concurrently with the clear downward trends of δ^{13}C and δ^{15}N. These results indicate that potential dietary shifts or baseline foodchain shifts in the Greenland and Norwegian subpopulations have likely played a major role in mitigating Hg exposure. Since we integrated dietary proxies in the Hg trend models rather than correcting Hg concentrations, i.e. the trend lines were already accounted for the dietary factor, we did not observe reverse trends as reported in previous studies (Braune et al., 2014; Burgess et al., 2013). Nevertheless, it is crucial to take into account the trends of δ^{13}C and δ^{15}N. It is possible that anthropogenic Hg influx is still relatively high in both Norway and Greenland, although the effect was masked by potential dietary shifts.

4.3. Effect of sampling on the spatiotemporal trends

Our models accounted for 6.5%, 10.5%, 11.2% and a very high 65.7% of the observed variation in Hg concentrations in body feathers of white-tailed eagle from West Greenland (1884–1943 and 1970–2013), Norway and Sweden, respectively. Compared to those in Greenland and Norway, sampling of the Swedish subpopulation was less subjected to confounding biological and ecological variables (e.g. age or location) by using only moulted adult feathers within a smaller geographical area. This likely contributed to the higher model performance in the Swedish subpopulation. These results are strong evidence for popularising the establishment of archives of moulted feathers as was done for the Swedish subpopulation in the framework of environmental monitoring (Espín et al., 2016). However, due to the age effect, it should be kept in mind that the difference in Hg concentrations between Sweden (only adult pairs) and the other two subpopulations (both adults and juveniles) could be smaller than we observed, as Hg concentrations in juveniles have been found to be lower than in adults (Dietz et al., 2006; Lindberg and Odsjö, 1983).

4.4. Magnitudes, thresholds and implications

Adverse health effects of Hg exposure range from behavioural abnormalities, impaired reproduction to sterility or even mortality depending on the exposure magnitude (Burger and Gochfeld, 1997). Five μg g^{-1} of Hg in feathers has been used as a threshold for adverse effect in various seabird studies (Blévin et al., 2013; Bond et al., 2015). Scheuhammer (1991) suggested that a Hg concentration of 20 μg g^{-1} in feathers of piscivorous birds would indicate significant threat to reproductive success, while in loons ≥40 μg g^{-1} of feather Hg concentration was associated with a substantial reduction in individual fitness (Evers et al., 2008). In contrast, three adult bald eagles (*Haliaeetus leucocephalus*) sampled at Pinchi lake with a median feather concentration of 40 μg g^{-1} did not show reproductive impairment or other signs of toxicity (Weech et al., 2009). Neither was 66 μg g^{-1} feather Hg associated with five-year productivity in adult bald eagles from the Great lakes regions (Bowerman et al., 1994). Furthermore, no significant correlation was found between productivity and Hg in eggs in the highly contaminated Swedish white-tailed eagle population from 1965 to 1978 (Helander et al., 1982). The study was largely done on eggs from the same nests sampled for feathers as represented in the present study, with concentrations in feathers in the range of 28–68 μg g^{-1} during the same time period (Table 1 and Fig. 2*D*). Based on these observations, and because of the high trophic position of the white-tailed eagle, we suggest that concentrations below 40 μg g^{-1} in feathers might be regarded as "safe", while concentrations above this threshold should still be considered as a range with increased risk for negative health effects in white-tailed eagles. This conservative approach extends the commonly accepted range of 1 to 5 μg g^{-1}, a concentration frequently found in raptors inhabiting habitats with natural background concentrations of Hg (Scheuhammer, 1991).

Classifying individuals of the assessed subpopulations into the just-mentioned categories, it is reasonable to conclude that the Greenland and Norwegian subpopulations have not been at risk for Hg-mediated toxic health effects (Fig. 4). In contrast, the contamination in Sweden has been severe, especially during the time when Hg was widely applied in industry and agriculture during the 1960s. At that time body feather Hg concentrations in many individuals exceeded the threshold for concern. Concentrations in the Swedish subpopulation only fell below the threshold after around 1980, and they remain twice as high compared to the Greenland and Norwegian subpopulations (Fig. S2 and Table 1). In addition, it is important to note that the percentage of individuals associated with increased anthropogenic Hg input (>5 μg g^{-1}; Scheuhammer, 1991) in Greenland increased considerably in the most recent decade.

5. Conclusions

In the present study, we reconstructed long-term temporal trends of Hg and stable carbon and nitrogen isotopes in three white-tailed eagle subpopulations from the West Greenland, Norwegian and Central Swedish Baltic Sea coasts. We found distinctly different concentrations in feathers and trends of Hg exposure between the Swedish subpopulation and the other two subpopulations. The considerably higher concentrations and sharp declining Hg concentrations in the Swedish subpopulation was in accordance to the intensive agricultural and industrial use during the 1960s and the national regulation thereafter. Even though the eagles are unlikely to remain under severe threat posed by current Hg exposure in Sweden, Hg concentrations in all pairs still showed elevated anthropogenic input as opposed to natural background concentrations (>5 μg g^{-1}). In contrast, consistent with lower anthropogenic input, Hg concentrations were much lower in the Norwegian subpopulation. Our trends showed increasing Hg values in white-tailed eagle feathers in Norway until the mid-1950s, the concentrations decreased afterwards that coincided with decreases in $\delta^{13}C$ and $\delta^{15}N$ values. Our finding indicate that anthropogenic influence may still be of importance in Norway, even though more than half of the individuals in the most recent decades showed a Hg exposure associated with natural background contamination (< 5 μg g^{-1}). We observed low Hg concentrations and the lack of significant temporal trends in the Greenland subpopulation. This confirms Greenland being less contaminated, although there was an increasing number of individuals showing concentrations associated with increased anthropogenic Hg input (>5 μg g^{-1}) in the recent decade. Finally, our findings demonstrated the feasibility of using archived feathers for Hg and stable isotope trends, and we encourage to continue and expand monitoring in these subpopulations. This is important because the concentrations in freshwater fish are still high enough to be of concern for human consumption in some regions of Sweden (Åkerblom et al., 2014), and an increasing influx of Hg originating from Asia is to be expected during the coming years (Chen et al., 2018).

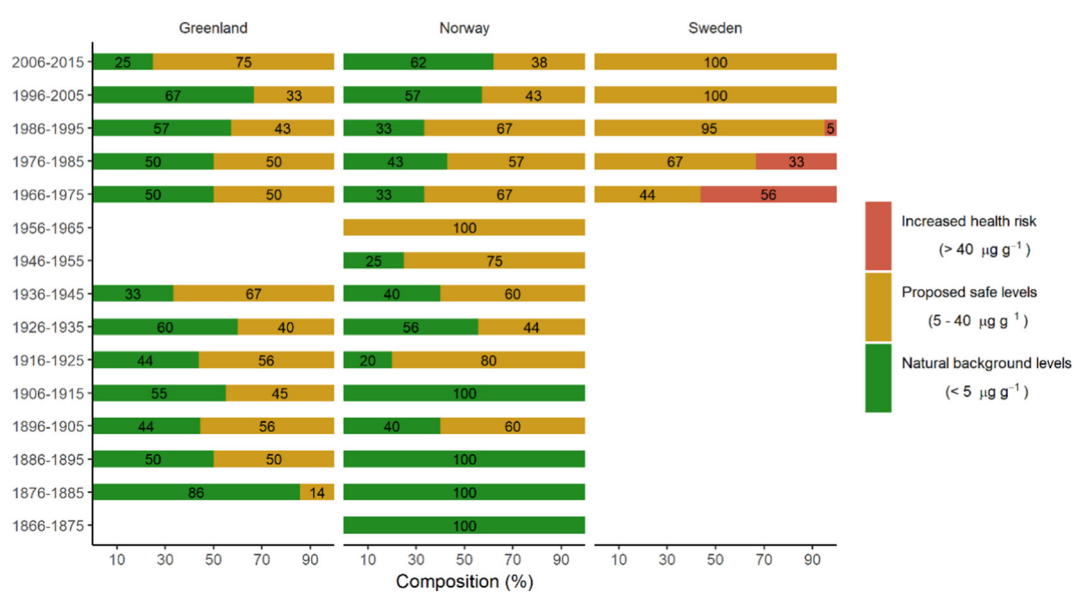


Fig. 4. Proportions of individual white-tailed eagles from the West Greenland, the Norwegian and Central Swedish Baltic Sea coasts at risk for Hg exposure impacted health at 10-year intervals. Empty fields are periods without available samples.

Conflict of interest statement

The authors declare that there are no intellectual or financial conflicts of interest.

Acknowledgements

The present study was made possible by the ECOSTRESS project, funded by the Norwegian Research Council (project number 255681) and the FRAM Centre Flagship Program "Hazardous substances – effects on ecosystems and human health" (project: Impacts of environmental contaminants and natural stressors on northern raptors: RAPTOR), as well as the BONUS BALTHEALTH project, which received funding from BONUS (Art. 185), funded jointly by the EU, Innovation Fund Denmark (grants 6180-00001B and 6180-00002B), Forschungszentrum Jülich GmbH, German Federal Ministry of Education and Research (grant FKZ 03F0767A), Academy of Finland (grant 311966) and Swedish Foundation for Strategic Environmental Research (MISTRA). We especially thank Jan Bolding Kristensen and Jon Fjeldså (Natural History Museum of Denmark), Trond Johnsen (Norwegian Institute for Nature Research), Ingvar Byrkjedal (Bergen University Museum), Jan T. Lifjeld (Oslo University Natural History Museum), Alf Tore Mjøs (Stavanger Museum), Per Gustav Thingstad (NTNU University Museum), Stig Lundmo (Helgeland - Rana Museum) and Swedish Museum of Natural History. We thank the Universiteit Antwerpen and FWO-Flanders for support.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.06.027.

References

AMAP, 2011. AMAP assessment 2011: mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), p. 193.
Appelquist, H., Asbirk, S., Drabæk, I., 1984. Mercury monitoring: mercury stability in bird feathers. Marine. Poll. Bull. 15, 22–24.
Beattie, S.A., Armstrong, D., Chaulk, A., Comte, J., Gosselin, M., Wang, F., 2014. Total and methylated mercury in Arctic multiyear sea ice. Environ. Sci. Technol. 48, 5575–5582.
Berg, W., Johnels, A., Sjöstrand, B., Westermark, T., 1966. Mercury content in feathers of Swedish birds from the past 100 years. Oikos 17, 71–83.
Bignert, A., Bäcklin, B.-M., Helander, B., Roos, A., 2012. Contaminants and health of aquatic wildlife. In: Norrgren, L., Levengood, J.M. (Eds.), Ecosystem Health and Sustainable Agriculture. Ecology and Animal Health. Uppsala University, Uppsala, pp. 73–85.
Bignert, A., Dahlgren, H., Danielsson, S., Faxneld, S., Kylberg, E., Nyberg, E., Vasileiou, M., Staveley Öhlund, J., Berger, U., Borg, H., 2014. Comments concerning the national Swedish contaminant monitoring programme in marine biota, 2014. Report 1. Swedish Museum of Natural History 62–75.
Bignert, A., Danielsson, S., Faxneld, S., Nyberg, E., 2016. Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota, 2016. Report 5. Swedish Museum of Natural History, pp. 73–85.
Blévin, P., Carravieri, A., Jaeger, A., Chastel, O., Bustamante, P., Cherel, Y., 2013. Wide range of mercury contamination in chicks of southern ocean seabirds. PLoS One 8, e54508.
Boening, D.W., 2000. Ecological effects, transport, and fate of mercury: a general review. Chemosphere 40, 1335–1351.
Bond, A.L., Hobson, K.A., Branfireun, B.A., 2015. Rapidly increasing methyl mercury in endangered ivory gull (*Pagophila eburnea*) feathers over a 130 year record. Proc. R. Soc. B 282, 20150032.
Bortolotti, G.R., 2010. Flaws and pitfalls in the chemical analysis of feathers: bad news–good news for avian chemoecology and toxicology. Ecol. Appl. 20, 1766–1774.
Bowerman, W.W., Evans, E.D., Giesy, J.P., Postupalsky, S., 1994. Using feathers to assess risk of mercury and selenium to bald eagle reproduction in the Great Lakes region. Arch. Environ. Contam. Toxicol. 27, 294–298.
Braune, B.M., Gaskin, D.E., 1987. Mercury levels in Bonaparte's gulls (*Larus philadelphia*) during autumn molt in the Quoddy region, New Brunswick, Canada. Arch. Environ. Contam. Toxicol. 16, 539–549.
Braune, B.M., Donaldson, G.M., Hobson, K.A., 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. Environ. Pollut. 117, 133–145.
Braune, B.M., Gaston, A.J., Hobson, K.A., Gilchrist, H.G., Mallory, M.L., 2014. Changes in food web structure alter trends of mercury uptake at two seabird colonies in the Canadian Arctic. Environ. Sci. Technol. 48, 13246–13252.
Braune, B.M., Gaston, A.J., Mallory, M.L., 2019. Temporal trends of legacy organochlorines in eggs of Canadian Arctic seabirds monitored over four decades. Sci. Total Environ. 646, 551–563.
Burger, J., Gochfeld, M., 1997. Risk, mercury levels, and birds: relating adverse laboratory effects to field biomonitoring. Environ. Res. 75, 160–172.
Burgess, N.M., Bond, A.L., Hebert, C.E., Neugebauer, E., Champoux, L., 2013. Mercury trends in herring gull (*Larus argentatus*) eggs from Atlantic Canada, 1972–2008: temporal change or dietary shift? Environ. Pollut. 172, 216–222.
Chen, L., Zhang, W., Zhang, Y., Tong, Y., Liu, M., Wang, H., Xie, H., Wang, X., 2018. Historical and future trends in global source-receptor relationships of mercury. Sci. Total Environ. 610-611, 24–31.
Corman, A.-M., Schwemmer, P., Mercker, M., Asmus, H., Rüdel, H., Klein, R., Boner, M., Hofem, S., Koschorreck, J., Garthe, S., 2018. Decreasing $\delta^{13}C$ and $\delta^{15}N$ values in four coastal species at different trophic levels indicate a fundamental food-web shift in the southern North and Baltic Seas between 1988 and 2016. Environ. Monit. Assess. 190, 461.
Crewther, W.G., Fraser, R.D., Lennox, F.G., Lindley, H., 1965. The chemistry of keratins. Adv. Protein Chem. 20, 191–346.
Dietz, R., Riget, F., Cleemann, M., Aarkrog, A., Johansen, P., Hansen, J.C., 2000. Comparison of contaminants from different trophic levels and ecosystems. Sci. Total Environ. 245, 221–231.
Dietz, R., Riget, F., Boertmann, D., Sonne, C., Olsen, M.T., Fjeldså, J., Falk, K., Kirkegaard, M., Egevang, C., Asmund, G., Wille, F., Moller, S., 2006. Time trends of mercury in feathers of West Greenland birds of prey during 1851—2003. Environ. Sci. Technol. 40, 5911–5916.
Dietz, R., Outridge, P.M., Hobson, K.A., 2009. Anthropogenic contributions to mercury levels in present-day Arctic animals—a review. Sci. Total Environ. 407, 6120–6131.
Dietz, R., Mosbech, A., Flora, J., Eulaers, I., 2018. Interactions of climate, socio-economics, and global mercury pollution in the North Water. Ambio 47, 281–295.
Dittmann, T., Becker, P.H., Bakker, J., Bignert, A., Nyberg, E., Pereira, M.G., Pijanowska, U., Shore, R.F., Stienen, E., Toft, G.O., Marencic, H., 2012. Large-scale spatial pollution patterns around the North Sea indicated by coastal bird eggs within an EcoQO programme. Environ. Sci. Pollut. R. 19, 4060–4072.
Elmgren, R., 1989. Man's impact on the ecosystem of the Baltic sea: energy flows today and at the turn of the century. Ambio 18, 326–332.
EPA, U.S., 1998. Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry. Environmental Protection Agency, Washington, DC.
Espín, S., García-Fernández, A., Herzke, D., Shore, R., van Hattum, B., Martínez-López, E., Coeurdassier, M., Eulaers, I., Fritsch, C., Gómez-Ramírez, P., 2014. Sampling and contaminant monitoring protocol for raptors. Research Networking Programme-EURAPMON, Research and Monitoring for and With Raptors in Europe.
Espín, S., García-Fernández, A.J., Herzke, D., Shore, R.F., van Hattum, B., Martínez-López, E., Coeurdassier, M., Eulaers, I., Fritsch, C., Gómez-Ramírez, P., 2016. Tracking pan-continental trends in environmental contamination using sentinel raptors—what types of samples should we use? Ecotoxicology 25, 777–801.
Evers, D.C., Savoy, L.J., DeSorbo, C.R., Yates, D.E., Hanson, W., Taylor, K.M., Siegel, L.S., Cooley, J.H., Bank, M.S., Major, A., Munney, K., Mower, B.F., Vogel, H.S., Schoch, N., Pokras, M., Goodale, M.W., Fair, J., 2008. Adverse effects from environmental mercury loads on breeding common loons. Ecotoxicology 17, 69–81.
Falandysz, J., Jakuczun, B., Mizera, T., 1988. Metals and organochlorines in four female white-tailed eagles. Marine. Poll. Bull. 19, 521–526.
Farmer, R.G., Leonard, M.L., 2011. Long-term feeding ecology of great black-backed gulls (*Larus marinus*) in the northwest Atlantic: 110 years of feather isotope data. Can. J. Zool. 89, 123–133.
Forsman, D., 1999. The Raptors of Europe and the Middle East: A Handbook of Field Identification (London).
Frederick, P.C., Hylton, B., Heath, J.A., Spalding, M.G., 2009. A historical record of mercury contamination in southern Florida (USA) as inferred from avian feather tissue: contribution R-09888 of the. Journal Series, Florida Agricultural Experiment Station. Environ. Toxicol. Chem. 23, 1474–1478.
Frøslie, A., Holt, G., Norheim, G., 1986. Mercury and persistent chlorinated hydrocarbons in owls Strigiformes and birds of prey Falconiformes collected in Norway during the period 1965–1983. Environ. Pollut. B. 11, 91–108.
Fu, L., Moncher, R.B., 2004. Severity distributions for GLMs: gamma or lognormal? Evidence from Monte Carlo simulations. Casualty Actuarial Society Discussion Paper Program, pp. 149–230.
Goede, A.A., De Bruin, M., 1984. The use of bird feather parts as a monitor for metal pollution. Environ. Pollut. B. 8, 281–298.
Gruber, N., Keeling, C.D., Bacastow, R.B., Guenther, P.R., Lueker, T.J., Wahlen, M., Meijer, H.A., Mook, W.G., Stocker, T.F., 1999. Spatiotemporal patterns of carbon-13 in the global surface oceans and the oceanic Suess effect. Glob. Biogeochem. Cycles 13, 307–335.
Hailer, F., Helander, B., Folkestad, A., Ganusevich, S., Garstad, S., Hauff, P., Koren, C., Masterov, V., Nygård, T., Rudnick, J., 2007. Phylogeography of the white-tailed eagle, a generalist with large dispersal capacity. J. Biogeogr. 34, 1193–1206.
Head, J.A., DeBofsky, A., Hinshaw, J., Basu, N., 2011. Retrospective analysis of mercury content in feathers of birds collected from the state of Michigan (1895–2007). Ecotoxicology 20, 1636–1643.
Helander, B., 1983. Food Ecology of the White-tailed Sea Eagle Haliaeetus albicilla in Sweden, in: Reproduction of the White-tailed Sea Eagle *Haliaeetus albicilla*, in Relation to Food and Residue Levels of Organochlorine and Mercury Compounds in the Eggs (Stockholm).
Helander, B., Olsson, M., Reutergårdh, L., 1982. Residue levels of organochlorine and mercury compounds in unhatched eggs and the relationships to breeding success in white-tailed sea eagles *Haliaeetus albicilla* in Sweden. Ecography 5, 349–366.
Helander, B., Marquiss, M., Bowerman, B., 2003. Sea Eagle 2000: Proceedings from the International Sea Eagle Conference in Björkö, Sweden, 13–17 September 2000 (Stockholm).

Honda, K., Min, B.Y., Tatsukawa, R., 1986. Distribution of heavy metals and their age-related changes in the Eastern great white egret, *Egretta alba modesta*, in Korea. Arch. Environ. Contam. Toxicol. 15, 185–197.
Jensen, S., Johnels, A.G., Olsson, M., Otterlind, G., 1972. DDT and PCB in herring and cod from the Baltic, the Kattegat and the Skagerrak. Ambio Special Report 71–85.
Johnels, A., Germund, T., Westermark, T., 1979. A history of mercury levels in Swedish fauna. Ambio 8, 160–168.
Koivusaari, J., Nuuja, I., Palokangas, R., Hattula, M.-L., 1976. Chlorinated hydrocarbons and total mercury in the prey of the white-tailed eagle (*Haliaeetus albicilla L.*) in the Quarken Straits of the Gulf of Bothnia, Finland. Bull. Environ. Contam. Toxicol. 15, 235–241.
Koivusaari, J., Nuuja, I., Palokangas, R., Finnlund, M., 1980. Relationships between productivity, eggshell thickness and pollutant contents of addled eggs in the population of white-tailed eagles *Haliaëtus albicilla L.* in Finland during 1969–1978. Environ. Pollut. 23, 41–52.
Körtzinger, A., Quay, P.D., Sonnerup, R.E., 2003. Relationship between anthropogenic CO_2 and the ^{13}C Suess effect in the North Atlantic Ocean. Glob. Biogeochem. Cycles 17, 1–20.
Kovács, A., Mammen, U.C.C., Wernham, C.V., 2008. European monitoring for raptors and owls: state of the art and future needs. Ambio 37, 408–412.
Lehtonen, K.K., Bignert, A., Bradshaw, C., Broeg, K., Schiedek, D., 2017. Chemical pollution and ecotoxicology. In: Snoeijs-Leijonmalm, P., Schubert, H., Radziejewska, T. (Eds.), Biological Oceanography of the Baltic Sea. Springer Netherlands, Dordrecht, pp. 547–587.
Lindberg, P., Odsjö, T., 1983. Mercury levels in feathers of peregrine falcon *Falco peregrinus* compared with total mercury content in some of its prey species in Sweden. Environ. Pollut. B. 5, 297–318.
Lindqvist, O., Johansson, K., Bringmark, L., Timm, B., Aastrup, M., Andersson, A., Hovsenius, G., Håkanson, L., Iverfeldt, Å., Meili, M., 1991. Mercury in the Swedish environment — recent research on causes, consequences and corrective methods. Water Air Soil Pollut. 55, 1–261.
Marra, G., Wood, S.N., 2011. Practical variable selection for generalized additive models. Comput. Stat. Data Anal. 55, 2372–2387.
Mizutani, H., Fukuda, M., Kabaya, Y., Wada, E., 1990. Carbon isotope ratio of feathers reveals feeding behavior of cormorants. Auk 107, 400–403.
Mizutani, H., Fukuda, M., Kabaya, Y., 1992. C13 and N15 enrichment factors of feathers of 11 species of adult birds. Ecology 73, 1391–1395.
Mlíkovský, J., 2009. The food of the white-tailed sea eagle (*Haliaeetus albicilla*) at Lake Baikal, East Siberia. Slovak Raptor J 3, 35–39.
Monteiro, L.R., Furness, R.W., 2001. Kinetics, dose—response, and excretion of methylmercury in free-living adult Cory's shearwaters. Environ. Sci. Technol. 35, 739–746.
Movalli, P., Bode, P., Dekker, R., Fornasari, L., van der Mije, S., Yosef, R., 2017. Retrospective biomonitoring of mercury and other elements in museum feathers of common kestrel *Falco tinnunculus* using instrumental neutron activation analysis (INAA). Environ. Sci. Pollut. R. 24, 25986–26005.
Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. Annu. Rev. Ecol. Evol. Syst. 18, 293–320.
Pettersson, O., Lehman, H., 1994. Reduced pesticide use in Scandinavian agriculture. Crit. Rev. Plant Sci. 13, 43–55.
Provencher, J.F., Mallory, M.L., Braune, B.M., Forbes, M.R., Gilchrist, H.G., 2014. Mercury and marine birds in Arctic Canada: effects, current trends, and why we should be paying closer attention. Environ. Rev. 22, 244–255.
R Core Team, 2018. R: A Language and Environment for Statistical Computing, Vienna. https://www.R-project.org/.
Renedo, M., Bustamante, P., Tessier, E., Pedrero, Z., Cherel, Y., Amouroux, D., 2017. Assessment of mercury speciation in feathers using species-specific isotope dilution analysis. Talanta 174, 100–110.
Rigét, F., Dietz, R., Born, E.W., Sonne, C., Hobson, K.A., 2007. Temporal trends of mercury in marine biota of west and northwest Greenland. Marine. Poll. Bull. 54, 72–80.
Rigét, F., Braune, B., Bignert, A., Wilson, S., Aars, J., Born, E., Dam, M., Dietz, R., Evans, M., Evans, T., 2011. Temporal trends of Hg in Arctic biota, an update. Sci. Total Environ. 409, 3520–3526.
Scheuhammer, A.M., 1991. Effects of acidification on the availability of toxic metals and calcium to wild birds and mammals. Environ. Pollut. 71, 329–375.
Skurdal, J., Qvenild, T., Skogheim, O.K., 1985. Mercury accumulation in five species of freshwater fish in Lake Tyrifjorden, south-east Norway, with emphasis on their suitability as test organisms. Environ. Biol. Fish 14, 233–237.
Sulkava, S., Tornberg, R., Koivusaari, J., 1997. Diet of the white-tailed eagle *Haliaeetus albicilla* in Finland. Ornis Fennica 74, 65–78.
Tagliabue, A., Bopp, L., 2008. Towards understanding global variability in ocean carbon-13. Glob. Biogeochem. Cycles 22, 1–13.
Thompson, D.R., Furness, R.W., 1989. Comparison of the levels of total and organic mercury in seabird feathers. Marine. Poll. Bull. 20, 577–579.
Thompson, D.R., Furness, R.W., Lewis, S.A., 1995. Diets and long-term changes in $\delta^{15}N$ and $\delta^{13}C$ values in northern fulmars *Fulmarus glacialis* from two northeast Atlantic colonies. Mar. Ecol. Prog. Ser. 125, 3–11.
Thompson, D.R., Bearhop, S., Speakman, J.R., Furness, R.W., 1998. Feathers as a means of monitoring mercury in seabirds: insights from stable isotope analysis. Environ. Pollut. 101, 193–200.
Vo, A.T.E., Bank, M.S., Shine, J.P., Edwards, S.V., 2011. Temporal increase in organic mercury in an endangered pelagic seabird assessed by century-old museum specimens. Proc. Natl. Acad. Sci. 108, 7466–7471.
Weech, S.A., Scheuhammer, A.M., Elliott, J.E., 2009. Mercury exposure and reproduction in fish-eating birds breeding in the Pinchi Lake region, British Columbia. Canada. Environ. Toxicol. Chem 25, 1433–1440.
Westermark, T., Odsjö, T., Johnels, A., 1975. Mercury content of bird feathers before and after Swedish ban on alkyl mercury in agriculture. Ambio 4, 87–92.
Wille, F., Kampp, K., 1983. Food of the white-tailed eagle *Haliaeetus albicilla* in Greenland. Ecography 6, 81–88.
Willgohs, J.F., 1961. The White-tailed Eagle *Haliaëtus albicilla albicilla* (Linné) in Norway (Trondheim).
Wood, S.N., 2011. Fast stable restricted maximum likelihood and marginal likelihood estimation of semiparametric generalized linear models. J. Royal Stat. Soc. B. 73, 3–36.
Wood, S.N., 2017. P-splines with derivative based penalties and tensor product smoothing of unevenly distributed data. Stat. Comput. 27, 985–989.
Åkerblom, S., Bignert, A., Meili, M., Sonesten, L., Sundbom, M., 2014. Half a century of changing mercury levels in Swedish freshwater fish. Ambio 43, 91–103.