



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

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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 ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) in general increased performance of the temporal trend models, but this was dependent on the subpopulation and study period. The downward trend of Hg coincided with the decreasing $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ 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 ($40\mu\text{g g}^{-1}$), while the maximum exposure in the Swedish subpopulation was distinctively elevated (median: $46.0\mu\text{g g}^{-1}$) and still remains well above natural background concentrations (maximum $5\mu\text{g g}^{-1}$).

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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; Willgoos, 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 concentra-

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

Table 1

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

tions (Braune et al., 2014). It is therefore important to study ecological tracers such as the stable nitrogen and carbon isotopes ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) 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 (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, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ 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

Table 2

Comparison of generalized additive models with different covariates (year; $\delta^{15}\text{N}$; $\delta^{13}\text{C}$) 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	ΔAICc	
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
				$\delta^{15}\text{N}$	1.67	0.47	0.08			
Greenland (1970–2013)	41	1.65	0.05	Year	0.45	0.09	0.19	0.03	4.51%	2.25
		1.74	0.10	Year	0.00	0.00	0.39	0.11	9.34%	0.00
				$\delta^{13}\text{C}$	1.05	0.18	0.20			
Norway (1866–2015)	101	1.75	0.10	Year	0.52	0.12	0.16	0.03	3.70%	2.24
		1.74	0.10	Year	0.75	0.32	0.06			
				$\delta^{13}\text{C}$	0.75	0.34	0.05			
Sweden (1967–2011)	86	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
				$\delta^{15}\text{N}$	1.21	0.50	0.03			
Sweden (1967–2011)	86	1.73	0.05	Year	1.31	0.57	0.02	0.08	9.27%	1.88
				$\delta^{13}\text{C}$	1.16	0.22	0.16			
		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
				$\delta^{15}\text{N}$	0.90	1.52	<0.01			
		3.23	0.05	Year	1.89	21.3	<0.01	0.66	74.0%	2.77
		$\delta^{13}\text{C}$	1.61	1.24	0.01					
		Year	1.90	17.1	<0.01	0.66	72.8%	0.00		
		$\delta^{15}\text{N}$	0.88	1.45	<0.01					
		$\delta^{13}\text{C}$	1.54	0.84	0.02					

Edf: estimated degree of freedom.

R^2 : adjusted R^2 .

D : deviance explained.

ΔAICc : the difference in AICc values for each candidate model compared to the candidate model with the lowest AICc value.

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 differing concentrations of Hg exposure and also potentially differing temporal trends among these subpopulations. In addition, we investigated the temporal trends of Hg in relation to feather $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. 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 comprises 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 plastic bags upon collection and transferred to the 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 from various body regions was taken from each nest and specific year. These subsamples then represent the status (Hg exposure and diet) of the breeding pairs in their specific terri-

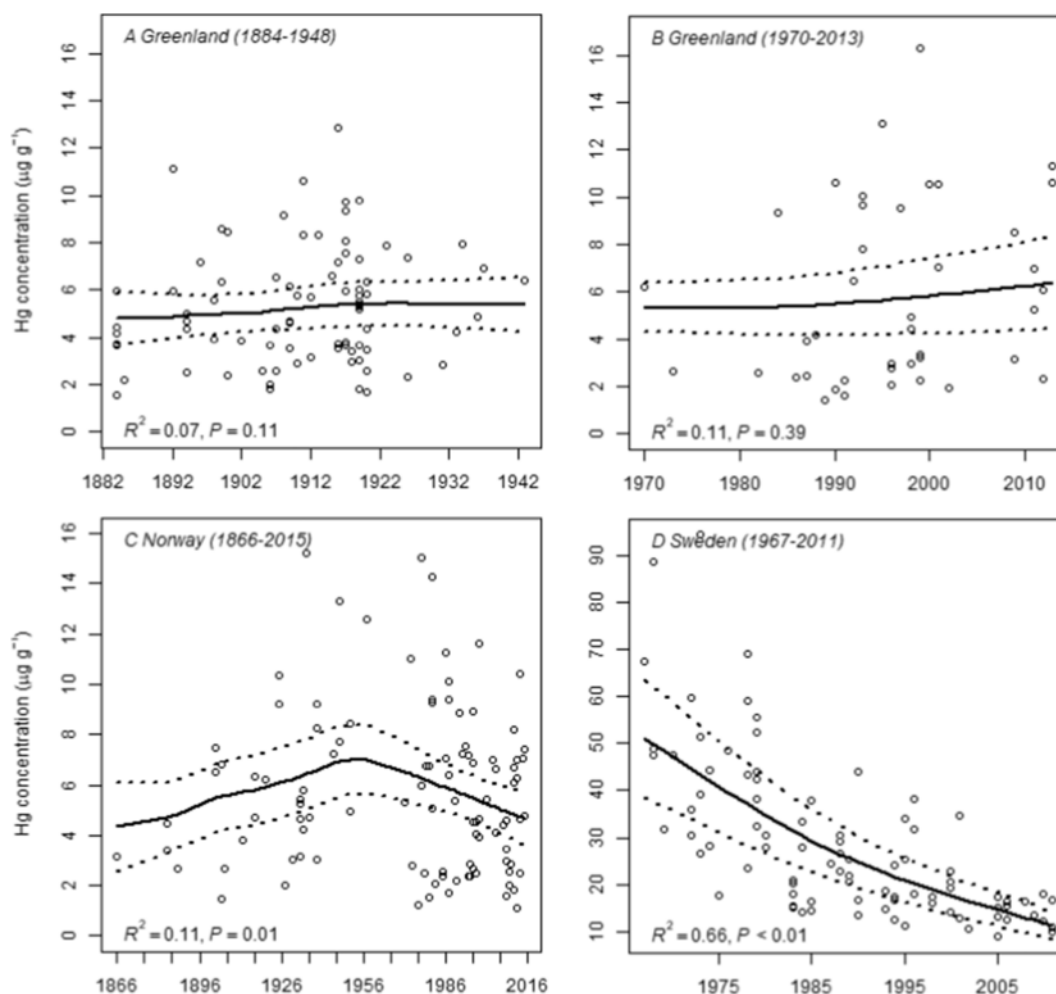


Fig. 2. Temporal trends of Hg in body feathers of white-tailed eagles from the West Greenland coast (A and B), the Norwegian coast (C) and the Central Swedish Baltic Sea coast (D). Open dots are original Hg concentrations. Trend lines (with a 95% prediction interval) were fitted using GAMs with year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

tory 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; Espin 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 (Soriso, Italy) following the U.S. EPA Method 7473 (EPA, 1998). Dry homogenised feather samples weighing on average 5.27 ± 0.42 (mean \pm SD) mg were analysed. The instrumental analytical quality control was verified by analysing procedural blanks, duplicates, aqueous standards (10 ng and 100 ng Hg, prepared from $1000 \pm 4 \text{ mg L}^{-1}$ 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 ($0.07 \pm 0.13 \text{ ng}$; $n=131$) as well as for the recovery of aqueous standards ($108.6 \pm 1.3\%$; $n=21$). The measured recovery percentage of the CRMs fell within the acceptable range ($105.9 \pm 2.3\%$; $n=52$) of the certified value ($0.410 \pm 0.055 \mu\text{g g}^{-1}$ 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 $\mu\text{g g}^{-1}$.

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 nitro-

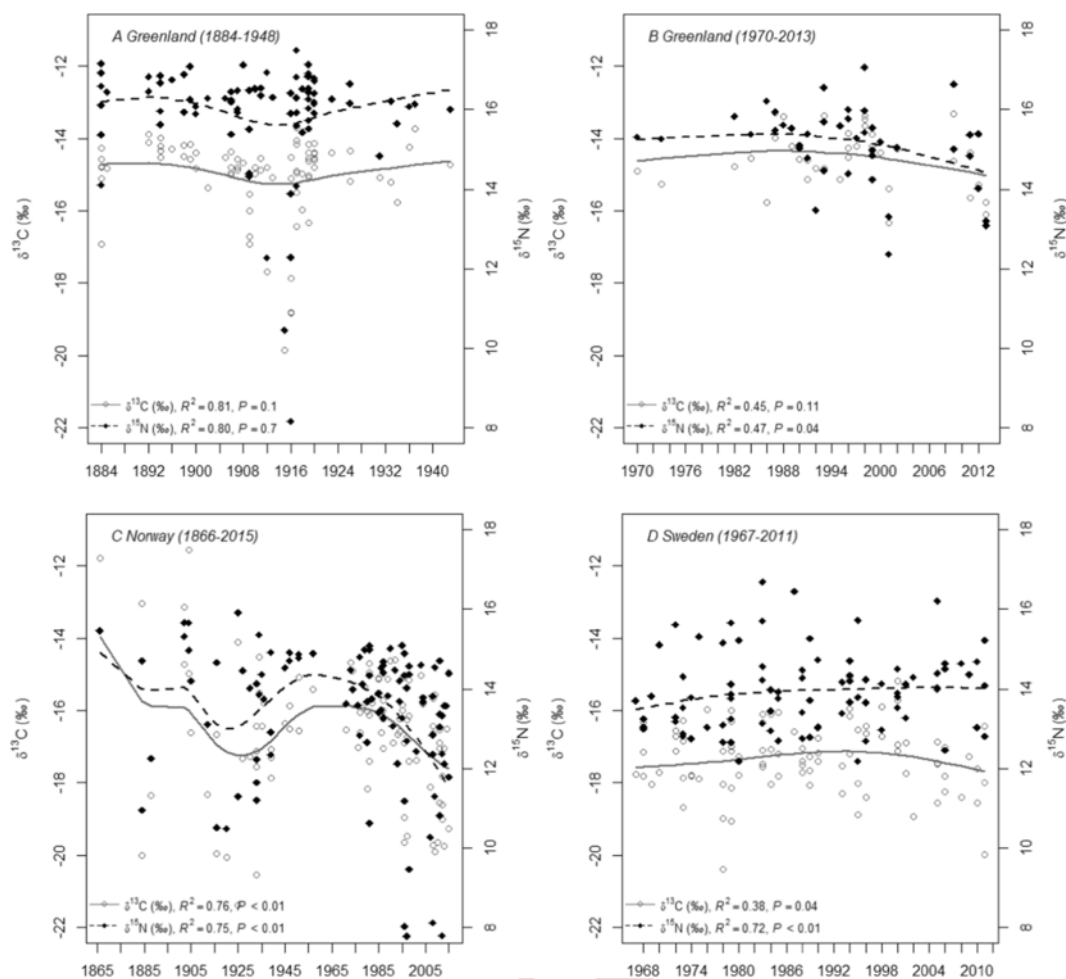


Fig. 3. Temporal trends of $\delta^{13}\text{C}$ (open dots) and $\delta^{15}\text{N}$ (closed dots) in body feathers of white-tailed eagles (*Haliaeetus albicilla*) collected in West Greenland (A and B), the Norwegian coast (C), and the Central Swedish Baltic Sea coast (D). $\delta^{13}\text{C}$ values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

gen ($^{15}\text{N}:^{14}\text{N}$) 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 , respectively. For the subpopulations in Greenland and Norway, an internal reference material (i.e., casein) was measured in duplicate every ten samples revealing an imprecision ($\pm SD$) $\leq 0.06\text{‰}$ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. For the subpopulation in Sweden, glycine was used as the internal reference material and was measured every 15 samples, showing an imprecision ($\pm SD$) $\leq 0.20\text{‰}$ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$.

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, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) 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). $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ were strongly correlated in both the Greenland and Norwegian subpopulations, but not in the one from Sweden, thus, we avoided including both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in one model when studying the Greenland and Norwegian subpopulations. There were weaker correlations between year and $\delta^{15}\text{N}$ 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 ($>\text{mean} + 5SD$) from the Greenland (1884–1948; $24.88 \mu\text{g g}^{-1}$) and Norwegian subpopulations ($26.10 \mu\text{g g}^{-1}$) and one sample with low

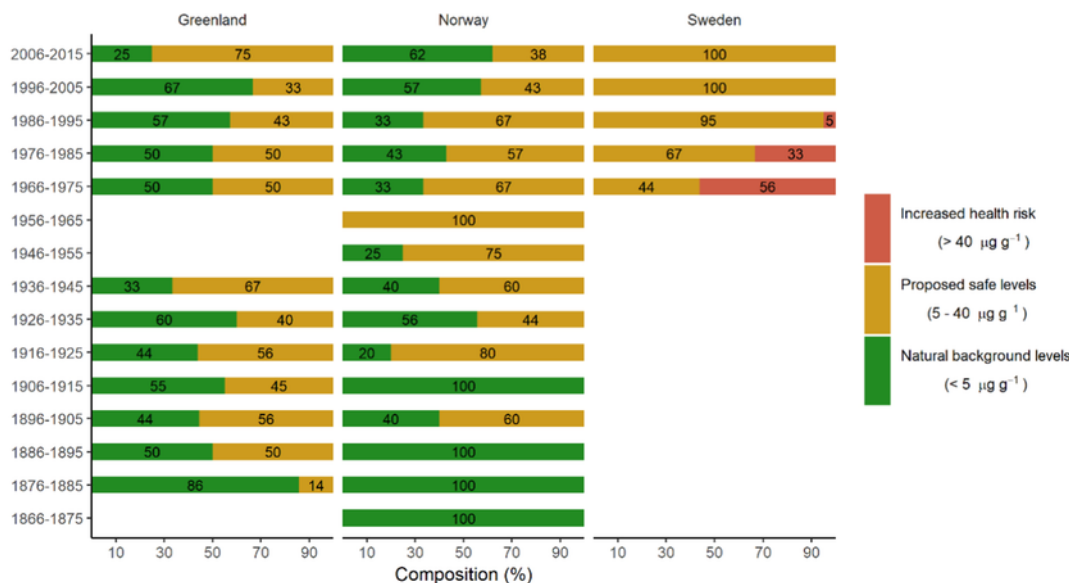


Fig. 4. Proportions of individual white-tailed eagle from the West Greenland, the Norwegian and Central Swedish Baltic Sea coasts at risk for Hg exposure impacted health at 10-year intervals. Empty fields are periods without available samples.

$\delta^{13}\text{C}$ ($< \text{mean} - 5\text{SD}$) from the Sweden subpopulation (-25.93) were regarded as outliers and removed from further analysis.

$\delta^{13}\text{C}$ values were corrected for the oceanic Suess effect, the temporal decline of atmospheric $\delta^{13}\text{C}$ values and consequently oceanic $\delta^{13}\text{C}$ 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 $0.00\text{--}2.10\text{‰}$, $0.00\text{--}2.28\text{‰}$ and $0.32\text{--}1.47\text{‰}$ 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 $\delta^{15}\text{N}$ 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 $20\ \mu\text{g g}^{-1}$. In contrast, values at the start of the study period in Sweden (median = $46.0\ \mu\text{g g}^{-1}$; 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 ($\delta^{15}\text{N}$) 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 (Table 2).

In the Norwegian subpopulation, year explained more of the observed Hg variation (6%) and showed a significant effect also after controlling for $\delta^{15}\text{N}$ ($P=0.01$; 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 = $4.51\ \mu\text{g g}^{-1}$; 2013–2015) similar to that at the start of the study period (median = $4.39\ \mu\text{g g}^{-1}$; 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 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ ($P<0.01$; Table 2). With a shorter sampling 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 across years, as we observed a clearly steeper decline until about 1985 (3.8% year $^{-1}$ from 1967 to 1985), af-

terwards the decrease has slowed down (2.2% year⁻¹ from 1985 to 2011).

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

Including $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ resulted in the lowest AICc values of fitted Hg temporal trend models, comparing to when only year was used as a predictor (Table 2). However, ΔAICc 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, ΔAICc was considerably lower when including these proxies (Table 2). Moreover, there were no substantial differences in the model AICc including either $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ in Greenland (1970–2013) and Norway, whereas including $\delta^{15}\text{N}$ resulted in better models in Greenland (1884–1943) and Sweden.

We observed significant temporal trends of $\delta^{15}\text{N}$ in Greenland (1970–2013), Norway and Sweden, and $\delta^{13}\text{C}$ in Norway and Sweden (all $P < 0.05$; Fig. 3). Trends lines of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ appeared to be mostly constant in Sweden throughout the study period (Fig. 3, D) whereas $\delta^{15}\text{N}$ appeared to be decreasing in Greenland (1970–2013; Fig. 3, B). In comparison, the variation in both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ 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 movements 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 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 km² and 34,000 km² in Norway and Sweden, respectively (Pettersson and Lehman, 1994). Relatively low Hg concentrations were previously reported in white-tailed eagles (2.4 $\mu\text{g g}^{-1}$ 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 $\mu\text{g g}^{-1}$ liver) during 1964–1966 (Falandysz et al., 1988). Hg concentrations observed here in body feathers from Swedish eagles during the 1960s–1970s are comparable with the concentrations reported previously in tail feathers (Berg et al., 1966): 38.4 $\mu\text{g g}^{-1}$ ($n=1$; 1952) and 47.0–64.0 $\mu\text{g g}^{-1}$ (mean=51.6; $n=4$; 1964–1965). Hg exposure in eagles from prior to 1950, however, showed lower concentrations ranged 2.7–15.5 $\mu\text{g g}^{-1}$

(mean=6.6; $n=8$; 1832–1940) and 3.7–7.8 $\mu\text{g g}^{-1}$ (mean=5.8; $n=4$; 1942–1949). These concentrations are comparable to MeHg concentrations in primary feathers of adult white-tailed eagles from West Greenland that averaged 5.8 $\mu\text{g g}^{-1}$ 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 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 Ångskärsklubb within our study area (2.3% year⁻¹; 1971–2012; Bignert et al., 2014) and in guillemot (*Uria aalge*) eggs from Gotland 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$

Incorporating information on dietary proxies, especially the trophic level proxy $\delta^{15}\text{N}$, in environmental contaminants studies has become increasingly popular (Braune et al., 2002, 2019; Rigét et al., 2007). In the present study, including $\delta^{13}\text{C}$ or $\delta^{15}\text{N}$ 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ 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), $\delta^{15}\text{N}$ gave better model performance than $\delta^{13}\text{C}$. These findings indicate a likely more important role of trophic level in explaining Hg exposure relatively to $\delta^{13}\text{C}$ in white-tailed eagles. All three studied subpopulations are from marine coastal areas, and all feed predominantly on fish and waterfowl as previously re-

ported in Greenland (Wille and Kampp, 1983), Norway (Willgohs, 1961), and Sweden (Helander, 1983). Consequently, there appeared to be no marked variation in $\delta^{13}\text{C}$ 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 food-web decline of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ reported in the German Baltic Sea during 1988–2016 (Corman et al., 2018), we observed relatively stable $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ 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 $\delta^{15}\text{N}$ could be linked to the stable Hg concentrations in Greenland observed during 1970–2013. In addition, $\delta^{15}\text{N}$ levels in the Greenland subpopulation during 1970–2013 were relatively lower than 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. These results indicate that potential dietary shifts or baseline food-web 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. 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 and location) by using only moulted adult feathers within a smaller geographical area. This 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 $\mu\text{g g}^{-1}$ 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 $20\mu\text{g g}^{-1}$ in feathers of piscivorous birds would indicate significant threat to reproductive success, while in loons $\geq 40\mu\text{g g}^{-1}$ 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 $40\mu\text{g g}^{-1}$ did not show reproductive impair-

ment or other signs of toxicity (Weech et al., 2009). Neither was $66\mu\text{g g}^{-1}$ 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 $28\text{--}68\mu\text{g g}^{-1}$ 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 $40\mu\text{g g}^{-1}$ 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 $5\mu\text{g g}^{-1}$, 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 ($>5\mu\text{g g}^{-1}$; 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 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 ($>5\mu\text{g g}^{-1}$). 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 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. 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 ($<5\mu\text{g g}^{-1}$). 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 ($>5\mu\text{g g}^{-1}$) 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 fresh-

water 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).

Conflict of interest statement

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

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Appendix A. Supplementary data

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