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Temporal trends of legacy organochlorines in different white-tailed eagle (*Haliaeetus albicilla*) subpopulations: A retrospective investigation using archived feathers

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ABSTRACT

Understanding the spatiotemporal patterns of legacy organochlorines (OCs) is often difficult because monitoring practices differ among studies, fragmented study periods, and unaccounted confounding by ecological variables. We therefore reconstructed long-term (1939–2015) and large-scale (West Greenland, Norway, and central Sweden) trends of major legacy OCs using white-tailed eagle (*Haliaeetus albicilla*) body feathers, to understand the exposure dynamics in regions with different contamination sources and concentrations, as well as the effectiveness of legislations. We included dietary proxies ($\delta^{13}C$ and $\delta^{15}N$) in temporal trend models to control for potential dietary plasticity. Consistent with the hypothesised high local pollution sources, levels of polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs) in the Swedish subpopulation exceeded those in the other subpopulations. In contrast, chlordanes (CHLs) and hexachlorobenzene (HCB) showed higher concentrations in Greenland, suggesting the importance of long-range transport. The models showed significantly decreasing trends for all OCs in Sweden in 1968–2011 except for CHLs, which only decreased since the 1980s. Nevertheless, median concentrations of DDTs and PCBs remained elevated in the Swedish subpopulation throughout the 1970s, suggesting that the decreases only commenced after the implementation of regulations during the 1970s. We observed significant trends of increasing concentrations of PCBs, CHLs and HCB in Norway from the 1930s to the 1970s/1980s and decreasing concentrations thereafter. All OC concentrations, except those of PCBs were generally significantly decreasing in the Greenland subpopulation in 1985-2013. All three subpopulations showed generally increasing proportions of the more persistent compounds (CB 153, *p.p′*-DDE and *β*-HCH) and decreasing proportions of the less persistent ones (CB 52, *p.p′*-DDT, *α*- and γ-HCH). Declining trends of OC concentrations may imply the decreasing influence of legacy OCs in these subpopulations. Finally, our results demonstrate the usefulness of archived museum feathers in retrospective monitoring of spatiotemporal trends of legacy OCs using birds of prey as sentinels.

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1. Introduction

Organochlorine (OC) contamination has been and still is a worldwide concern ever since the ubiquitous presence of polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) was reported in the environment in the 1960s (Jensen, 1966; Holden and Marsden, 1967). Their toxicity, bioaccumulative and long-range transport (LRT) potential were consequently investigated and have led to national regulations of their production and use in developed countries since the 1970s (Goldberg, 1975). Internationally, the Stockholm Convention on Persistent Organic Pollutants (POPs) was implemented in 2004 and took legislative restrictions on the production and use of these compounds a step further towards transboundary and global management of the major OCs (UNEP, 2008).

Decreasing temporal trends of many OCs in the environment have been reported since the 1970s following national regulations, especially in highly polluted regions such as the Baltic Sea (Westöö, 1974; Kveseth et al., 1979). However, OCs such as DDTs are still exempted for use in some countries where alternative vector control methods are lacking (van den Berg et al., 2017). Moreover, PCBs, lindane (γ-hexachlorocyclohexane; HCH) and hexachlorobenzene (HCB) have been released into the environment from former production sites and stockpiles (Weber et al., 2015). As a result, OC levels in remote regions, such as the Arctic, have not decreased consistently across biota due to its function as a global sink for contamination (Hung et al., 2016; Rigét et al., 2019). In addition, OCs are partitioned in the environment according to their temperature mediated physical-chemical properties. Thus, previously immobilised stocks can be remobilised due to climate change (Ma et al., 2011).

Comparing temporal trends across administrative boundaries is crucial for a better understanding of the environmental behaviour and distribution of OCs as well as the efficiency and geographical results of legislative decisions. It is however often difficult to reliably compare trend studies conducted using different matrices, species, and analytical or statistical methods. Moreover, many studies are not adequately long or statistically powerful enough to detect clear trends due to typically observed short-term fluctuations (Bignert et al., 1993; Rigét et al., 2019) or are confounded by the lack of information on sources or pathways. Since diet is a major pathway for contaminant exposure, controlling for variability in dietary habits using proxies such as stable carbon (indicating food source) and nitrogen (indicating trophic level) isotopes is desired in the reconstruction of long-term temporal trends (Hebert and Weseloh, 2006; Braune, 2007; Gauthier et al., 2008; Braune et al., 2019).

Feathers are increasingly being regarded as a useful nondestructive biomonitoring tool for OC concentrations (Dauwe et al., 2005; Jaspers et al., 2006, 2019). Feathers are connected to the blood circulation during the period of formation and they have been shown to be correlated with concentrations measured in internal tissues (Jaspers et al., 2019). Therefore they represent a unique integrated documentation of information on the circulating blood chemicals (Goede and De Bruin, 1984). Although feathers have thus far not been used in long-term and large-scale spatiotemporal trend studies of OCs, archived feathers from museums or environmental specimen banks may be particularly valuable in environmental monitoring as their collections allow spatial and temporal comparisons of contaminant exposure (Rocque and Winker, 2005; Campbell and Drevnick, 2015).

In the present study, we aimed to reconstruct long-term temporal trends of major OCs over a large geographical area encompassing a hypothesised contamination gradient. To this end, we sampled feather collections of an established sentinel species, the white-tailed eagle (*Haliaeetus albicilla*), from three subpopulations inhabiting the coasts of West Greenland (low local contamination but LRT), Norway (limited local contamination) and the central Swedish Baltic Sea (high local contamination). White-tailed eagles are suitable for studying large-scale spatial trends as they are largely sedentary. Their lack of migratory movements has been supported by phylogeographical studies (e.g. Hailer et al., 2007) and field observations from colour ringing programmes (Helander, 2003; Lyngs, 2003). The objectives of the present study were to (1) reconstruct temporal trends of the intensity and composition of OC exposure, while accounting for stable carbon and nitrogen isotopes as dietary proxies; and (2) to investigate these exposure dynamics with respect to the hypothesised contamination gradient and known regulation history of OCs. Additionally, we looked into the suitability of archived feathers in reconstructing spatiotemporal trends of OCs. Finally, we discussed the potential effect of OC exposure on the species and implications for future monitoring.

2. Materials and methods

2.1. Sampling and preparation

Body (chest and back contour) feathers of white-tailed eagles from the West Greenland (n = 38; 1984–2013), the Norwegian (n = 76; 1939–2015) and central Swedish Baltic coasts (n = 150; 1967–2011) were obtained from various natural history museum collections as well as specimen banks and prepared as reported in detail elsewhere (Sun et al., 2019b). Briefly, approximately 10 body feathers per breeding pair were sampled from a moulted feather collection of the Swedish subpopulation (collected at individual territories and stored at ambient conditions in polyethylene bags at the Swedish Museum for Natural History, Stockholm, Sweden). From the Greenland and Norwegian collections, the same amount of body feathers was sampled per individual from immature and adult specimens of both sexes. Since these two collections were based on dead eagles found by the public and given to the museums (or management authorities), we expect the numbers of both sexes and age groups to have been represented randomly across the study periods. These specimens were preserved as skins or mounted pieces (stored in cabinets at ambient conditions) or were frozen (stored in polyethylene bags at −20 °C). Museums are known to have treated their collections with organic biocides, and these would then potentially interfere with some of the targeted OCs such as DDT and γ-HCH (Schmidt, 2000; Marcotte et al., 2014). In the present study, all analysed Greenland and Swedish feathers can however be deemed free from external biocide contamination, since they were sampled from frozen carcasses or archived moulted feathers preserved in polyethylene bags upon collection in the field. The Norwegian feathers, however, include in addition to frozen samples also mounted and skin specimens acquired during years when preservative treatment was common. Based on the knowledge of the participating Norwegian museum collection curators, we do not suspect external contamination of the feathers with the targeted OCs except for DDTs and γ-HCH.

The body feathers collected for the present study were placed and stored in polyethylene bags at ambient conditions until chemical analysis. All feathers were prepared according to Espín et al. (2014) in a controlled lab environment: the calamus was removed from each feather and the rest was subsequently thoroughly cleaned using distilled water and stainless steel tweezers. By washing with distilled water only we were able to retain preen oil on the feather surface, as preen oil originates from internal sources of the bird, and has been found to be significantly correlated with internal muscle concentrations (Jaspers et al., 2011, 2019). The feathers were then dried overnight at ambient temperature, and finally cut into < 1 mm pieces using stainless steel scissors. The homogenised feather material was stored in aluminium foil at ambient conditions until chemical analysis.

2.2. Organochlorine analysis

The analysis was performed at the Toxicological Centre (University of Antwerp, Belgium). All feathers were analysed for 20 PCB congeners (CB 49, 52, 74, 95, 99, 101, 105, 110, 118, 138, 149, 153, 156, 170, 171, 177, 180, 183, 187, and 194), *p,p′*-

dichlorodiphenyltrichloroethane (*p,p′*-DDT) and its metabolites, *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE) and *p,p′*-dichlorodiphenyldichloroethane (*p,p′*-DDD), three chlordane-related compounds (CHLs): *cis*-nonachlor (CN), *trans*-nonachlor (TN) and oxychlordane (OxC), hexachlorobenzene (HCB) and hexachlorocyclohexanes (HCHs; *α*-, *β*- and γ-HCH). The extraction and analytical methods have been described elsewhere (e.g. Jaspers et al., 2007, 2011; Eulaers et al., 2011). Briefly, on average 204 ± 5 mg of feather material was spiked with 50 μL of internal standards (CB 143, *ε*-HCH and ^{13}C-HCB), and incubated overnight at 45 °C in a mixture of hexane:dichloromethane (4:1; *v:v*) and HCl (1 M). After liquid–liquid extraction, the extracts were cleaned up on acid silica (44%) and eluted with 10 mL of hexane:dichloromethane (1:1; *v:v*). All extracts were then concentrated until dryness under a gentle nitrogen flow, reconstituted in 100 μL of *iso*-octane and transferred to injection vials. Targeted compounds were quantified using an Agilent 6890 gas chromatograph coupled to an Agilent 5973 mass spectrometer, operated in electron capture negative ionization (GC/ECNI-MS) or in electron impact (GC/EI-MS) mode. The analytical quality assurance and control were positively evaluated based upon the analysis of procedural blanks (concurrently run every 11 samples) and internal standards. Since internal standards were added to each sample for quantification purposes, only the recovery of these internal standards could be computed. The values (mean ± *SD*) are as follows: CB 143: 82 ± 8% and *ε*-HCH: 69 ± 9%. All reported concentrations are corrected for average procedural blank values and recoveries. The limit of quantification (LOQ) was set at 3 × *SD* of the procedural blank values, or calculated from a 10:1 signal to noise ratio for compounds not detected in blanks. LOQs and detection frequencies (DFs) are listed in Supporting Information: Table S1.

2.3. Stable isotope analysis

The analysis for stable carbon (^{13}C and ^{12}C) and nitrogen (^{15}N and ^{14}N) isotopes was performed at the Stable Isotope Lab of the University of Koblenz-Landau (Greenland and Norwegian collections) and the Laboratory of Oceanology of the University of Liège, Belgium (Swedish collection), both using isotope ratio mass spectrometry. The SI ratios for carbon and nitrogen are conventionally expressed as δ values (‰). The imprecision (± *SD*) for both stable isotope values, as revealed by internal reference materials, was ≤0.06‰ for the Greenland and Norwegian collections and ≤0.20‰ for the Swedish collection. Further details on the instrumentation and quantitation are reported in Sun et al. (2019b).

2.4. Statistical analyses

2.4.1. Data pre-processing

Due to the potential external contamination of DDTs and γ-HCH in the Norwegian samples from museum preservative measures, we carefully examined the compound profiles of DDTs and HCHs, and decided that only data from samples stored frozen should be used for these compounds in the Norwegian data set (detailed information is given in Fig. S1). The resulted sample sizes and time periods are therefore: n = 38 (all OCs; 1984–2013) for the West Greenland coast, n = 76 (PCBs, CHLs and HCB; 1939–2015) and n = 23 (DDTs and HCHs; 1992–2015) for the Norwegian coast, and n = 150 (all OCs; 1967–2011) for the central Swedish Baltic coast.

All statistical analyses were performed using R 3.5.1 (R Core Team, 2018). Only compounds with a DF above 50% in each subpopulation were statistically analysed. Non-detects were excluded for temporal trend analysis and set to zero for calculation of summary statistics and proportions. Given that most compounds have a DF of over 95% and only *α*- and γ-HCH were detected in less than 50% of the samples from Norway, we expect very limited bias caused by this censoring approach (Table S1).

We corrected δ^{13}C values for the oceanic Suess effect (Sun et al., 2019b). There was one sample with low δ^{13}C from the Swedish subpopulation (−25.9, while the population mean ± 5*SD* = −17.4 ± 6.6) and therefore it was regarded as an outlier and removed from further analyses. Information on age and sex were only sporadically available and could therefore not be included in the statistical analyses. There could be a potential age effect (immature vs. adult; Jaspers et al., 2013) in our comparison among subpopulations, because we sampled all adults in Sweden compared to both immature and adult eagles in Greenland and Norway. However, we consider the impact of age effect on our results to be likely limited with regard to the temporal trends reconstructed specific to each subpopulation.

2.4.2. Spatiotemporal trend modelling of exposure concentrations

We used year of exposure as a primary predictor for modelling the temporal trends, while including the dietary proxies δ^{13}C (foraging origin) and δ^{15}N (trophic position) to control for potential confounding by dietary plasticity. We modelled temporal trends of δ^{13}C and δ^{15}N for each subpopulation and fitted trend lines to show potential dietary plasticity across time (Fig. S2). Additionally, we checked for multicollinearity among these predictors. Predictors with a variance inflation factor above the suggested threshold of three were not included in the same model (i.e. δ^{13}C and δ^{15}N for the Norwegian subpopulation; Table S2) to avoid inflated standard errors and overfitting (Zuur et al., 2010). In addition, territory was included as a random effect in the Swedish subpopulation due to repetitive sampling of the same territories over years. Likely due to random seasonal variations between years (Fryer and Nicholson, 1993; Rigét et al., 2019), we observed strong year-to-year variations in OC concentrations at the beginning of the Greenland (1984–1986) and Swedish (1967–1968) time series. Data from these years were pooled when reconstructing temporal trends, and the beginning year was presented as 1985 in Greenland and 1968 in Sweden to assist calculation. Given that the variations happened at the very beginning of the study periods, and since both periods are relatively long, we expect very limited influence of such data treatment on the systematic temporal trends (Fryer and Nicholson, 1993).

We fitted Generalised Additive Models (GAMs, package 'mgcv'; Wood, 2017) with penalised thin plate splines for each OC group. Smooth terms were shrunk to zero in the absence of a relationship between the predictor and response variable and therefore we did not perform additional variable selection. We used the Gamma family with a log link considering the positive skewness of concentration data. For the Norwegian subpopulation we fitted models with δ^{13}C and δ^{15}N separately and compared the Akaike Information Criteria corrected for small sample sizes (AIC*c*). The model with the lowest AIC*c* was selected for temporal trend investigation of each OC. Model adequacy was visually validated using residual plots.

We evaluated the spatiotemproal trends of the five OC groups according to the summaries of the final models (P values, deviance explained and adjusted R^2). Trend lines were fitted for each group in each subpopulation to help visualise the temporal trends using model predictions. We also used these predictions and their median values to calculate the total percentage of change (decrease or increase) and annual percentage of change (decrease or increase) in exposure during the study period. Additionally, we compared OC concentrations among subpopulations. Average concentrations of ΣPCBs, ΣDDTs, ΣCHLs, ΣHCHs and HCB per ten-year period were calculated to account for temporal variation, ANOVA models were fitted for each period and differences among subpopulations were compared using Tukey pairwise comparison.

2.4.3. Temporal trend modelling of exposure composition for individual OC groups

In addition to the temporal trends of the exposure to the five OC groups, we investigated the potential change in the composition of each OC group (PCBs, DDTs, CHLs and HCHs) over time. We chose three indicator PCB congeners that are relatively persistent and

bioaccumulative (CB 138 and 153) or relatively volatile and less persistent (CB 52) and calculated their respective proportion to Σ_{20}PCBs. The proportions of DDT and metabolites (*p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD), HCH isomers (*α*-, *β*- and γ-HCH) and chlordane-related compounds (OxC, TN and CN) were also calculated, respectively. We used year as the predictor, and territory was again included as a random effect variable for the Swedish subpopulation.

We fitted Generalised Additive Model with Location, Scale and Shape (GAMLSS) for the proportion data (package 'gamlss'; Stasinopoulos et al., 2018). The current version of P splines (pb) was used for smoothing. Distribution families were selected for each individual compound using chooseDist() function and validated using dignostic plots, i.e. normalized quantile residual plots and multiple worm plots. We evaluated model performance for the intepretation of potential temporal change in composition and used model predictions to fit trend lines.

2.4.4. Spatiotemporal differences in the exposure profile for total OCs

To investigate the potential differences in the composition of total OCs among subpopulations, we calculated the average proportion for each sum group (i.e. Σ_{20}PCBs, ΣDDTs, ΣCHLs, ΣHCHs and HCB) to the total OC exposure. We used average proportion data in the four decades prior to 2015 during which time we have data of the five OCs in at least two subpopulations within each ten-year period (i.e. 1976–1985, 1986–1995, 1996–2005 and 2006–2015). We fitted beta regression (package 'betareg'; Cribari-Neto and Zeileis, 2009) with interaction between the factors subpopulation and compound class. We then looked at Tukey pairwise comparisons and documented differences in OC composition among the subpopulations within each period.

3. Results

3.1. Spatiotemporal trends of legacy OC exposure concentrations and composition

3.1.1. PCBs

Concentrations of Σ_{20}PCBs in the Greenland subpopulation were at a lower level (mean: 239 ng g^{-1}; median: 110 ng g^{-1}; 1984–2013) compared to the other two subpopulations and did not show a significant temporal trend from 1985 to 2013 ($P = 0.15$, Tables S3 and 1; Fig. 1). We observed a significant non-linear trend in the Norwegian subpopulation ($P < 0.01$), showing a 563% increase from 1939 to 1976 followed by a 72% decrease up to 2015. Σ_{20}PCB concentrations in Norway in the recent decade were still higher than those observed in the 1940s or those in Greenland. Σ_{20}PCB exposure was highest in the Swedish subpopulation, for many years the concentrations (2,273 ng g^{-1}; 1967–1999) have exceeded almost twice the peak concentration in Norway (1,228 ng g^{-1}; 1976), and were significantly higher than in Norway and Greenland across the entire study period (all $P < 0.05$; Fig. S3). Σ_{20}PCBs in Sweden showed a significantly decreasing temporal trend ($P < 0.01$) from 1968 to 2011 (79%). Although it is apparent that the median concentrations decreased after the implementation of regulations during the 1970s (Fig. S4). Despite this declining trend, PCB exposure in Sweden over the last decade was still the highest among all three subpopulations (Table S4).

Across years and subpopulations, CB 153 and CB 138 dominated the PCB profiles (Table S5). Although proportions of CB 153 were similar among the three subpopulations, noticeably higher ratios of CB 138 and CB 52 were observed in Sweden and Greenland, respectively (Fig. S5). We observed significant increases in the proportions of CB 153 in all three subpopulations over the study period (all $P < 0.01$). There were also significant overall increases in the proportions of CB 138 in the Norwegian and Swedish subpopulations (both $P < 0.01$), but less pronounced than those of CB 153. In contrast, significant decreases in the proportions of CB 52 were found in all subpopulations over the study period (all $P < 0.01$; Tables S7 and S8), with the most pronounced decrease observed for the Greenland subpopulation (from 4.5% to 1.6%; 1984–2013).

3.1.2. DDTs

White-tailed eagles from all locations were exposed to levels of ΣDDTs higher than all other pesticides and close to the levels of Σ_{20}PCBs (Table S4). The spatial variation in ΣDDT concentrations was smaller than observed for Σ_{20}PCBs, especially in the most recent decade when concentrations were not significantly different among subpopulations (all $P > 0.2$; Fig. S3). The overall gradient appears to be Sweden > Norway > = Greenland across the study period (Table S4; Fig. 1). Significant decreasing trends of ΣDDTs were observed for the Greenland ($P = 0.02$; 1985–2013) and Swedish subpopulations ($P < 0.01$; 1968–2011) with a total decrease of 65% and 95%, respectively. Similar to the ΣPCBs, the median ΣDDT concentrations in Sweden decreased after the implementation of regulations during the 1970s (Fig. S4). Contrasting the Greenland and Swedish trends, ΣDDT concentrations remained constant in the Norwegian subpopulation ($P = 0.70$; 1992–2015; Tables 1 and S3).

P,p′-DDE dominated the DDT profile in all three subpopulations (Table S6). Both *p,p′*-DDD and *p,p′*-DDT showed noticeably lower proportions in Sweden compared to the other two subpopulations. Overall increasing proportions of *p,p′*-DDE and decreasing proportions of *p,p′*-DDD were observed at the Swedish coast, whereas for *p,p′*-DDT the proportions showed a non-linear trend that first decreased from 4.5% in 1967 to 0.4% in 1996 and then increased again to 1.0% towards 2011 (all $P < 0.01$; Fig. S5). Proportions of *p,p′*-DDE and *p,p′*-DDT significantly increased and decreased (both $P < 0.01$) across the study period, respectively, in the Greenland subpopulation, while *p,p′*-DDD proportions remained constant ($P = 0.99$). We observed significantly increasing *p,p′*-DDT proportions ($P < 0.01$; 1992–2015) in the Norwegian subpopulation, but no significant temporal change for its metabolites (both $P > 0.10$; Tables S7 and S8).

3.1.3. CHLs

CHL exposure was significantly higher in the Greenland and Norwegian subpopulations compared to the Swedish one (Table S4; Fig. 1; all $P < 0.05$, 1976–2015; Fig. S3). Overall ΣCHL concentrations declined significantly (48%; 1985–2013) in the Greenland subpopulation ($P = 0.05$). Significant non-linear trend was observed in the Norwegian subpopulation ($P < 0.01$), with a 1183% increase from 1939 to 1989, followed by 32% decrease to 2015. Likewise, ΣCHL concentrations increased 91% from 1968 to 1981 at the Swedish coast, after which they declined 68% until 2011 ($P < 0.01$; Tables S3 and 1).

The CHL profile was dominated by OxC and TN across all subpopulations (Table S6). We observed noticeably higher proportions of OxC and lower proportions of CN in Greenland, while the proportions of TN were similar between all three subpopulations, especially in more recent years (Fig. S5). We found significantly decreasing proportions of OxC in Greenland across the study period ($P < 0.01$), but no apparent temporal trends were observed for CN or TN ($P = 0.37$ and 0.18, respectively). OxC proportions increased significantly at the Norwegian coast across the study period ($P < 0.01$), while proportions of CN and TN decreased significantly over time ($P = 0.02$ and $P < 0.01$, respectively). At the Swedish coast the proportions of CN increased significantly over the study period ($P < 0.01$), while no significant temporal trends were obvious for the proportions of OxC or TN ($P = 0.35$ and $P = 0.67$, respectively; Tables S7 and S8).

3.1.4. HCHs

The overall ΣHCH concentrations were highest in the Swedish subpopulation, followed by Greenland, and were significantly higher than at the Norwegian coast (Table S4; Fig. 1; $P = 0.02$, 1996–2005 and $P < 0.001$, 2006–2015; Fig. S3). ΣHCH concentrations decreased significantly in both the Greenland (60%; 1985–2013; $P < 0.01$) and Swedish subpopulations (96%; 1968–2011; $P < 0.01$). In contrast,

Table 1
Temporal trend model predictions of five legacy OC classes in body feathers from three white-tailed eagle subpopulations. GAM models were fitted using year, $\delta^{13}C$ and/or $\delta^{15}N$ as predictors. Model predicted concentrations (median; ng g^{-1}) of the starting and ending years, as well as the peak year (if applicable) are presented, with 95% prediction intervals (PI). Annual increasing (↑) and decreasing (↓) rates were computed using the predicted medians. Asterisk indicates significant temporal trends. "-" means not applicable. Σ_{20}PCBs: sum of CB 49, 52, 74, 95, 99, 101, 105, 110, 118, 138, 149, 153, 156, 170, 171, 177, 180, 183, 187, and 194, ΣDDTs: sum of *p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD, ΣHCHs: sum of *α*-, *β*- and γ-HCH, ΣCHLs: sum of CN, TN and OxC.

	Subpopulation	*n*	Period (peak year)	Median $_{start}$ (95% PI)	Median $_{peak}$ (95% PI)	Median $_{end}$ (95% PI)	Annual (↑)	Annual (↓)
Σ_{20}PCBs	Greenland	38	1985–2013 (–)	273 (56.5–490)	–	187 (12.6–361)	–	1.4%
	Norway	76	1939–2015 (1976)	185 (14.5–356)	1228 (650–1807)	342 (140–544)	5.2%*	3.2%*
	Sweden	150	1968–2011 (–)	4165 (2979–5352)	–	897 (619–1161)	–	3.5%*
ΣDDTs	Greenland	38	1985–2013 (–)	407 (105–708)	–	143 (29.6–256)	–	3.7%*
	Norway	23	1992–2015 (–)	174 (105–243)	–	174 (105–243)	–	0.0%
	Sweden	150	1968–2011 (–)	3348 (2138–4559)	–	187 (120–251)	–	6.5%*
ΣCHLs	Greenland	38	1985–2013 (–)	83.1 (32.5–134)	–	42.9 (14.4–71.4)	–	2.3%*
	Norway	76	1939–2015 (1989)	3.0 (0.1–6.0)	38.7 (24.0–53.3)	26.4 (10.2–42.7)	5.2%*	1.5%*
	Sweden	150	1968–2011 (1981)	7.6 (5.0–10.2)	14.6 (10.7–18.4)	4.6 (3.1–6.1)	5.1%*	3.8%*
ΣHCHs	Greenland	38	1985–2013 (–)	25.4 (11.1–39.7)	–	10.3 (4.3–16.3)	–	3.2%*
	Norway	22	1992–2015 (–)	5.0 (1.9–8.0)	–	3.2 (1.7–4.6)	–	1.9%
	Sweden	150	1968–2011 (–)	168 (104–232)	–	7.2 (4.7–9.9)	–	7.1%*
HCB	Greenland	38	1985–2013 (–)	18.3 (8.3–28.2)	–	11.5 (4.7–18.3)	–	1.7%
	Norway	75	1939–2015 (1974)	1.2 (0.5–2.0)	4.5 (2.9–6.1)	1.1 (0.7–1.6)	3.8%*	3.3%*
	Sweden	148	1968–2011 (–)	6.8 (4.4–9.1)	–	0.8 (0.5–1.1)	–	4.8%*

ΣHCH concentrations did not show a significant decrease at the Norwegian coast ($P = 0.16$; 1992–2015; Tables 1 and S3).

The HCH profiles were dominated by *β*-HCH in all three subpopulations (Table S6). Given that both *α*- and γ-HCH had DFs < 50% in the Norwegian samples, we compared the proportions between the Greenland and Swedish subpopulations only. Distinctively higher proportions of *α*-HCH were observed at the Greenland coast, especially in early years (Fig. S5). Proportions of all three isomers showed significant temporal trends at both the Greenland and Swedish coasts (all $P < 0.01$; Table S7): *β*-HCH increased considerably in proportion in both subpopulations while *α*- and γ-HCH both decreased over the study period. Interestingly, proportions of *α*-HCH at the Swedish coast, although decreased overall, first showed an increasing trend from 1.2% in 1967 to 4.8% in 1979, followed by a decreasing trend to 0.8% in 2000, after which *α*-HCH was not detected anymore in this subpopulation. Proportions of γ-HCH in the Swedish subpopulation decreased considerably from 80% in 1967 to 10% in 1995, yet increased again to 18% in 2011. In contrast, proportions of *β*-HCH increased from 28% in 1967 to 89% in 1996, and then decreased again to 85% in 2011 (Table S8).

3.1.5. HCB

Concentrations of HCB were lowest among all targeted OCs, and were significantly higher in the Greenland subpopulation than those in Norway and Sweden (Table S4; all $P < 0.001$, 1986–2015; Fig. S3). A marginally significantly ($P = 0.07$; 1985–2013) decreasing trend was found in the Greenland subpopulation. In the Norwegian subpopulation, we observed a 264% increase in HCB concentrations from 1939 to 1974, followed by a 75% decrease until 2015 ($P < 0.01$). At the Swedish coast, the overall HCB concentrations decreased significantly (88%; 1968–2011; $P < 0.01$; Tables 1 and S3). The HCB exposure was similar in the Norwegian and Swedish subpopulations during the recent decade, and both were considerably lower than at the Greenland coast (Fig. 1; Table S4).

3.2. Differences in the OC exposure profile among subpopulations

Within each subpopulation, we observed a consistent pattern of dominating Σ_{20}PCB and ΣDDT proportions, and relatively lower proportions of the other OC classes (Fig. S6). Noticeable differences in OC exposure profile can be observed across the subpopulations. Σ_{20}PCBs were consistently significantly presented at higher proportions in Sweden than in Greenland (all $P < 0.01$), and in Norway in 1996–2015 ($P < 0.01$, Fig. S7). Also striking were the higher proportions of organochlorine pesticides (OCPs) at the Greenland coast: unlike at the Norwegian and Swedish coasts, proportions of the four pesticides (i.e. ΣDDTs, ΣCHLs, ΣHCHs and HCB) combined were consistently above 50% at the Greenland coast during the entire study period. In addition, proportions of each OCP were significantly higher in the Greenland subpopulation than the Swedish one from 1985 onwards (all $P < 0.05$, Fig. S7) and the Norwegian one in 1986–1995 (all $P < 0.01$). From 1995 onwards, there were no significant differences in the proportions of ΣDDTs between the Greenland and Norwegian subpopulations ($P = 0.07$ and 0.18 in 1996–2005 and 2006–2015, respectively). However, ΣCHLs, ΣHCHs and HCB were still observed at significantly higher proportions in the Greenland subpopulation than in the Norwegian one (all $P < 0.01$, Fig. S7).

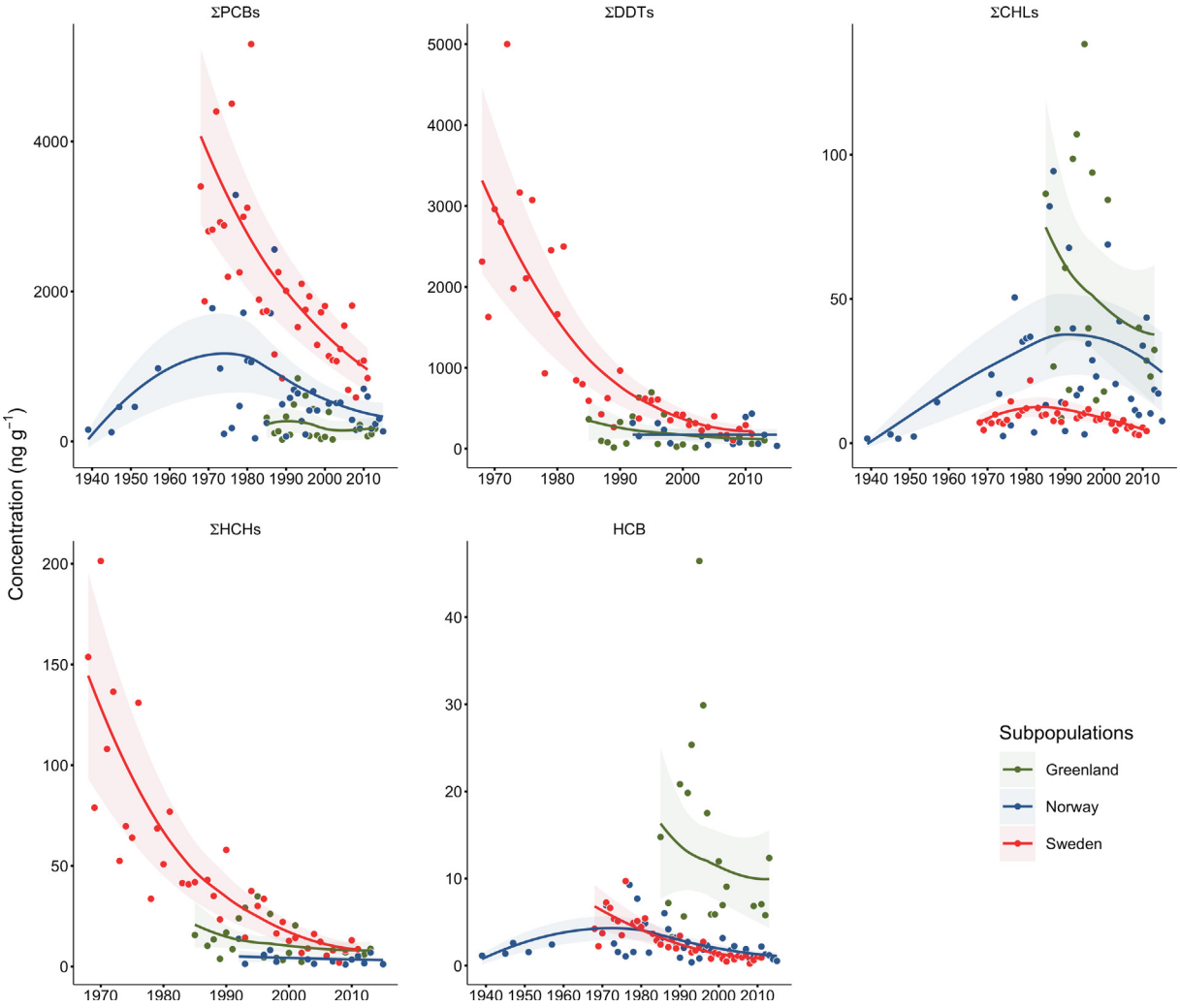

Fig. 1. Temporal trends of major legacy OC classes in body feathers of white-tailed eagles from the West Greenland, Norwegian and central Swedish Baltic coasts. Temporal trend lines and 95% prediction intervals (shaded) are fitted using GAM models with year, δ^{13}C and/or δ^{15}N as predictors (see Table S3 for further details). ΣPCBs: sum of CB 49, 52, 74, 95, 99, 101, 105, 110, 118, 138, 149, 153, 156, 170, 171, 177, 180, 183, 187, and 194, ΣDDTs: sum of *p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD, ΣHCHs: sum of *α*-, *β*- and γ-HCH, ΣCHLs: sum of CN, TN and OxC.

4. Discussion

4.1. Exposure concentrations and profiles in relation to local contamination and LRT

To our knowledge, this is the first long-term and large-scale spatiotemporal trend study of legacy OCs using archived museum feathers. The spatial differences in OC concentrations and proportions that we observed in general reflected the hypothesised spatial gradient from high local contamination at the central Swedish Baltic coast over lower local influx at the Norwegian coast to virtually no local sources, but influx through LRT, at the Greenland coast. Prior to their ban at the beginning of the 1970s, PCBs were imported in large quantities and widely used in Sweden at volumes of up to ten times higher than in Norway (De Voogt and Brinkman, 1989), consistent with the different exposure levels of the white-tailed eagles in the present study. DDTs were widely applied as an insecticide in Sweden (Jensen et al., 1969) and Norway (Kveseth et al., 1979). We could not compare the exposure levels between Sweden and Norway until the 1990s due to external contamination of early Norwegian samples. Nevertheless, comparable with PCBs, DDT concentrations were reported to be considerably lower in the Norwegian environment compared to central-south Sweden based on various species analysed in the late 1960s to early 1970s (Bagge, 1975). The same spatial pattern was found in harbour seals (*Phoca vitulina*) during the 1980s (Luckas et al., 1990). This is consistent with the history of intensive agricultural and industrial activities within the Baltic Sea region (Bagge, 1975).

The use of HCHs was also documented in both Sweden and Norway, but likely to have occurred in lower quantities than DDTs and was more restricted than in other European countries (Breivik et al., 1999; Li et al., 2003; Vijgen, 2006). HCH exposure in the present study was lower than the exposure to PCBs and DDTs by one to two orders of magnitude. Comparable differences between HCHs and PCBs and DDTs have been reported in fish from the Baltic Sea (Falandysz, 1985; Haahti and Perttila, 1988) and in Norwegian seabirds (Barrett et al., 1985). The exposure to HCB seems to have been comparable at the Norwegian and central Swedish Baltic coasts, whereas the exposure to CHLs was higher at the Norwegian coast than at the Swedish one. This could have been expected as CHLs were not registered for use in Sweden (Falandysz et al., 1994) but were so in Norway until 1967 (Skåre et al., 1985). HCB has not been used in Norway (Brevik et al., 1978), but has been used as a fungicide in Sweden (UNEP, 2017a), however likely in smaller quantities than HCHs. Previous studies also reported lower HCB concentrations than HCHs in fish from the Baltic Sea (Falandysz, 1984; Roots, 1995; Strandberg et al., 1998). Levels of HCB were also lower than HCHs in different fish species from Norwegian fjords and both

were lower than CHLs (Skåre et al., 1985).

OC exposure concentrations and proportions in the Greenland subpopulation appear to indicate a combination of low local input and high influence of LRT compared to the other subpopulations. In contrast to the high concentrations of PCBs (Sweden and Norway), DDTs (Sweden) and HCHs (Sweden), exposure to these compounds in the Greenland subpopulation was less pronounced, indicating a relative lack of high local contamination sources of these compounds in Greenland. CHLs and volatile HCB however, were detected in considerably higher concentrations at the Greenland coast, in comparison to the limited contamination at the Swedish and Norwegian ones. In addition, levels of DDTs and HCHs were comparable or higher in the Greenland subpopulation compared to the Norwegian one. Our findings therefore appear to support LRT from North America to West Greenland for these compounds (Muir and Norstrom, 2000), especially CHLs as they were never used in Sweden (Falandysz et al., 1994), whereas their production in the U.S. continued until 1997 (EFSA, 2007). Furthermore, the higher proportions of the volatile congener CB 52 and isomer α-HCH observed in the Greenland subpopulation are strongly indicative of LRT. Our results are in general agreement with previous studies comparing OCs in biota among the Baltic Sea region and other regions, such as Norway, the north Atlantic and the Arctic. In general, higher levels of PCBs, DDTs and HCHs, but lower levels of HCB and CHLs were found in the Baltic compared to remote areas (Luckas and Harms, 1987; Andersson et al., 1988; Luckas et al., 1990; Granby and Kinze, 1991; Berggren et al., 1999; Bruhn et al., 1999; Jorundsdottir et al., 2009; Fenstad et al., 2016).

4.2. Spatiotemporal trends in relation to regulatory actions

In the present study, we included dietary proxies $\delta^{13}C$ (foraging origin) and/or $\delta^{15}N$ (trophic position) in all temporal trend models. This allowed us to control for any potential confounding by individual dietary plasticity over time. The overall stable $\delta^{13}C$ and $\delta^{15}N$ values in Greenland are in contrast to the significant temporal trends in Norway and Sweden (Fig. S2), highlighting potential changes in feeding behaviour and/or food web structure (Braune et al., 2014) in the latter two subpopulations. Nevertheless, since all populations in the present study are from marine coastal regions, and all feed predominantly on fish and waterfowl (Willgohs, 1961; Helander, 1983; Wille and Kampp, 1983), it is unlikely that there have been major variations due to fundamental differences in terms of terrestrial versus aquatic diet predominance, as also evidenced by the $\delta^{13}C$ values (Fig. S2). In addition, although dietary tracers were not consistently significant in trend models (Table S3), keeping them as predictors has likely resulted in the better comparability of the OC temporal trends among subpopulations.

The temporal trend reconstructed for PCBs in the Norwegian subpopulation coincides well with the global production history as the commercial production of PCBs started in 1929 and global consumption peaked in the 1960s (Breivik et al., 2002), as well as with the Norwegian regulation on PCBs in 1971 (Skåre et al., 1985). We observed increasing PCB concentrations in white-tailed eagles since 1939 while they peaked in 1974, a few years after the regulation (such time lag in biota is not uncommon due to the mixed effects from other factors like climate; Rigét et al., 2019), then decreased continuously until the most recent sample. We did not observe a significant temporal change in DDT exposure from 1992 to 2015 in the Norwegian subpopulation, likely due to the limited sample size ($n = 23$). As a matter of fact, previous studies have reported considerable declines in DDT concentrations in both biotic and abiotic samples shortly after the DDT ban in 1970 (Brevik et al., 1978; Kveseth et al., 1979). In Sweden, the regulations on DDTs and PCBs were implemented in 1970 and 1971, respectively (Weissglas, 1973; Westöö, 1974), but was enforced stepwise: DDT was still in use in forestry until 1975, and a total ban of the use of PCBs in new products was enforced in 1978. Our temporal trend models suggest a slightly early onset of decreasing PCBs and DDTs in the Swedish subpopulation. Similar temporal trends have been previously reported in other monitoring programmes using Swedish Baltic fish (Sapota, 2006) and sediment records (Olsson et al., 2000). However, the appearance of continuous decreasing trends of DDTs and PCBs from the late 1960s is likely a result of our set of Swedish samples starting when concentrations of DDTs and PCBs in the environment were peaking. If the Swedish data set had started earlier, as is the case for Norway, we would most likely had seen a similar rise to the peak, and a subsequent decline. It is obvious from the data points in Figs. 1 and S3, as well as from data in white-tailed eagle eggs from the same area (Bignert and Helander, 2015), that the declines are not continuous from the late 1960s but commence from the late 1970s (DDTs) or around 1980 (PCBs). In fact, the highest concentrations of PCBs and DDTs in the Swedish subpopulation were detected in 1972 (10,924 ng g^{-1} and 6985 ng g^{-1}, respectively) and 1979 (13,134 ng g^{-1} and 8,867 ng g^{-1}, respectively) in feathers from the same territory, indicating continued accumulation throughout the 1970s. In addition, PCBs and DDTs were decreasing at different rates at the Swedish coast, with ΣDDTs decreasing at an annual rate twice as high as the rate for ΣPCBs, suggesting that PCBs are more persistent. This is also consistent with previous findings in temporal trend studies in the Baltic (Bignert et al., 1998; Helander et al., 2002). The decreasing proportions of *p,p′*-DDT from 1967 (4.5%) to 1996 (0.4%) in the Swedish subpopulation coincided with the diminishing DDT concentrations due to regulations. However, the proportions of *p,p′*-DDT again increased slowly but continuously to 1.0% in 2011. Likewise, *p,p′*-DDT proportions in the Norwegian subpopulation showed an increase from 1992 (3.2%) to 2015 (4.9%). Such increases seem to indicate recent DDT input or remobilisation. As a matter of fact, the use of postemergence herbicides such as glyphosate has been shown to cause remobilisation of sediment DDT due to their effect on soil erosion (Sabatier et al., 2014). Besides, both glyphosate and its metabolite have been detected in estuary water of the Baltic Sea (Skeff et al., 2015).

Unlike other OCs, CHLs in both the Norwegian and Swedish subpopulations peaked during the 1980s, which seems to indicate continued contamination after national regulations. CHL was introduced as an insecticide during the 1950s, though it was never used in Sweden (Falandysz et al., 1994) and banned in 1971 (UNEP, 2017a). CHL has been regulated in Norway since 1967 (Barrett et al., 1985) and banned since 2002 (UNEP, 2006). The prolonged CHL exposure in these eagles thus was likely due to emissions from existing or legacy sources, and/or LRT from the European continent (banned in the EU since 1981) and North America (produced until 1997; EFSA, 2007). The increasing temporal trends throughout the 1970s are in agreement with increasing CHL concentrations observed in northern pike (*Esox lucius*) and Atlantic herring (*Clupea harengus*) from northern Finnish waters of the Baltic Sea in 1971–1982 (Moilanen et al., 1982).

The decrease of ΣHCHs in Sweden occurred slightly earlier than the regulation of γ-HCH (1970; Weissglas, 1973). The temporal trend is consistent with global consumption history as the global peak consumption of γ-HCH was estimated to have occurred during the 1960s and early 1970s (Vijgen et al., 2010). These findings are comparable with the decreasing trends found in fish from Swedish waters, the annual decreasing rate (7.1%) is also consistent with the reported decreasing γ-HCH (8% $year^{-1}$) in common guillemot (*Uria aalge*) eggs (Nyberg et al., 2015). Possibly, due to the limited sample size, we could not detect a clear trend of HCH exposure in the Norwegian subpopulation from 1992 to 2015. Nonetheless, the HCH profile (low DFs of the α- and γ-isomers) reflects the likely effective emission control in Norway, considering that γ-HCH has been regulated in Norway since 1992 (UNEP, 2017b). Similar results were observed in the Swedish subpopulation, as α-HCH was no longer detected since 2000 in spite of its increased proportions up to 1979. Proportions of γ-HCH decreased sharply from 1967 to 1995, but afterwards proportions increased 8% until 2011. Such an increase may also suggest potential remobilisation of lindane in Sweden, and may potentially be related to the influence of

anthropogenic activities such as the use of postemergence herbicides as discussed above or climate change.

HCB was introduced as a fungicide in 1933, however, it was never authorized to use in Norway and a total Norwegian ban was implemented in 2002 (UNEP, 2006). The increasing and then decreasing temporal trend observed in the Norwegian subpopulation is thus more likely a reflection of background contamination due to LRT. The peak time of 1974 is in general agreement with the suggested global consumption trend in the 1960s (Bailey, 2001; Barber et al., 2005) and the estimated decrease of total HCB emissions between 1970 and 1995 (Eurochlor, 2015). On the other hand, the use of HCB in Sweden was likely phased-out voluntarily much earlier than its official national ban in 1980 (UNEP, 2017a), as we observed a continuous decline since 1968. Continuous declines of HCB concentrations in 1969–2012 were also observed in various fish species across Sweden, as well as in guillemot eggs (Nyberg et al., 2015).

Temporal trends of legacy OCs in Greenland suggest the effectiveness of international and/or regional regulations on their use during the 1970s and the 1980s. We found significant or marginally significant declines in all OCs, except for PCBs. The annual decreasing rates ranked DDTs > HCHs > CHLs > HCB. Although ΣPCBs did not show a significantly decreasing trend in the Greenland samples, we observed pronounced decreasing proportions for lower chlorinated PCB congeners (e.g. CB 52), which seems to indicate a diminishing input of the background contamination from less persistent PCBs. In fact, 50% of the long-term trends in the Arctic in general reported decreasing concentrations of PCBs, in addition to a general decreasing pattern in other legacy OCs (Rigét et al., 2019). The annual decreasing rates of DDTs (3.7%), HCHs (3.2%), CHLs (2.3%) and HCB (1.7%) in the present study are somewhat lower than the annual rates reported for the general Arctic biota (4.2%, 8.9%, 4.6% and 2.6%, respectively; Rigét et al. 2019). In addition, we observed decreasing trends in the proportions of α- and γ-HCH as well as *p,p′*-DDT in the Greenland subpopulation, which may suggest the effective control on new emissions of these compounds.

4.3. Implications and suggestions for future monitoring

OC concentrations in the Swedish white-tailed eagle feathers have decreased approximately seven-fold (DDTs) and four-fold (PCBs), in rough agreement with the decreases of these compounds in eggs from the same population during a time period when reproduction returned to normal (Bignert and Helander, 2015). This appears to indicate that PCB and DDT contamination may now not be of concern at the population-level for the studied white-tailed eagles. Nevertheless, eagles in all subpopulations have been exposed to a mixture of contaminants including mercury (Sun et al., 2019b) and perfluoroalkyl substances (Sun et al., 2019a) as well, and many are endocrine-disrupting chemicals. This may potentially result in synergistically concerted effects on the health of these individuals and ultimately the populations. Further studies are thus warranted to assess the health risks of contaminant mixtures in these and other birds of prey.

Our findings also underscore the need for continued monitoring in these subpopulations, especially considering the still elevated PCB levels at the central Swedish Baltic coast, the relatively high HCB levels at the Greenland coast, and the relatively high CHL levels at the Norwegian and Greenland coasts. The recent increases in the proportions of γ-HCH in Swedish feathers and *p,p′*-DDT in Norwegian and Swedish feathers also warrant further investigation to track potential new input or remobilisation.

Finally, our results highlight the usefulness of archived feathers for retrospective spatiotemporal trend monitoring of legacy OCs. However, there are several important issues to be considered with regard to using feathers in general and archived feathers in particular. External contamination from wet/dry deposition onto the feather surface is possible but likely of minor importance for OCs (Jaspers et al., 2019). Furthermore, the application of biocides in natural history museums needs to be considered, especially when analysing OCPs, such as DDTs and HCHs. If possible, systematic sampling as was done for the Swedish subpopulation in the present study is highly recommended, because the feather collection comprised moulted feathers from adult breeding pairs in known territories, and feathers had never been subjected to any preservative treatment.

CRediT authorship contribution statement

Jiachen Sun: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing - original draft, Writing - review & editing, Data curation, Visualization. **Adrian Covaci:** Conceptualization, Methodology, Validation, Investigation, Resources, Writing - review & editing, Project administration, Supervision. **Jan Ove Bustnes:** Conceptualization, Methodology, Resources, Writing - review & editing, Project administration, Funding acquisition. **Veerle L.B. Jaspers:** Conceptualization, Methodology, Investigation, Writing - review & editing, Funding acquisition. **Björn Helander:** Conceptualization, Methodology, Resources, Writing - review & editing. **Bård-Jørgen Bårdsen:** Formal analysis, Writing - review & editing. **David Boertmann:** Resources, Writing - review & editing. **Rune Dietz:** Resources, Writing - review & editing. **Aili Lage Labansen:** Resources, Writing - review & editing. **Gilles Lepoint:** Investigation, Resources, Writing - review & editing. **Ralf Schulz:** Investigation, Resources, Writing - review & editing. **Govindan Malarvannan:** Investigation, Writing - review & editing. **Christian Sonne:** Resources, Writing - review & editing. **Kasper Thorup:** Resources, Writing - review & editing. **Anders P. Tøttrup:** Resources, Writing - review & editing. **Jochen P. Zubrod:** Investigation, Resources, Validation, Writing - review & editing. **Marcel Eens:** Conceptualization, Methodology, Writing - review & editing, Funding acquisition, Supervision. **Igor Eulaers:** Conceptualization, Methodology, Validation, Investigation, Resources, Writing - review & editing, Project administration, Funding acquisition, Data curation, Supervision.

Declaration of Competing Interest

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

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Appendix A. Supplementary material

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