White-tailed eagle (*Haliaeetus albicilla*) feathers from Norway are suitable for monitoring of legacy, but not emerging contaminants

Mari E. Løseth[a,*], Nathalie Briels[a], Jørgen Flo[a], Govindan Malarvannan[b], Giulia Poma[b], Adrian Covaci[b], Dorte Herzke[c], Torgeir Nygård[d], Jan O. Bustnes[e], Bjørn M. Jenssen[a], Veerle L. B. Jaspers[a]

Affiliations:

[a]Department of Biology, Norwegian University of Science and Technology (NTNU), Høgskoleringen 5, 7491 Trondheim, Norway

[b]Toxicological Center, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium

[c]Norwegian Institute for Air Research (NILU), FRAM - High North Research Centre on Climate and the Environment, 9007 Tromsø, Norway

[d]Norwegian Institute for Nature Research (NINA), Høgskoleringen 9, 7034 Trondheim, Norway

[e]Norwegian Institute for Nature Research (NINA), FRAM - High North Research Centre on Climate and the Environment, 9007 Tromsø, Norway

***Corresponding author:**

Mari Engvig Løseth: mari.loseth@ntnu.no

Submission to:

Science of the Total Environment

Abstract

While feathers have been successfully validated for monitoring of internal concentrations of heavy metals and legacy persistent organic pollutants (POPs), less is known about their suitability for monitoring of emerging contaminants (ECs). Our study presents a broad investigation of both legacy POPs and ECs in non-destructive matrices from a bird of prey. Plasma and feathers were sampled in 2015 and 2016 from 70 white-tailed eagle (*Haliaeetus albicilla*) nestlings from two archipelagos in Norway. Preen oil was also sampled in 2016. Samples were analysed for POPs (polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and organochlorinated pesticides (OCPs)) and ECs (per- and polyfluoroalkyl substances (PFASs), dechlorane plus (DPs), phosphate and novel brominated flame retardants (PFRs and NBFRs)). A total of nine PCBs, three OCPs, one PBDE and one PFAS were detected in over 50 % of the plasma and feather samples within each sampling year and location. Significant and positive correlations were found between plasma, feathers and preen oil concentrations of legacy POPs and confirm the findings of previous research on the usefulness of these matrices for non-destructive monitoring. In contrast, the suitability of feathers for ECs seems to be limited. Detection frequencies (DF) of PFASs were higher in plasma (mean DF: 78 %) than in feathers (mean DF: 38 %). Only perfluoroundecanoic acid could be quantified in over 50 % of both plasma and feather samples, yet their correlation was poor and not significant. The detection frequencies of PFRs, NBFRs and DPs were very low in plasma (mean DF: 1 - 13 %), compared to feathers (mean DF: 10 - 57 %). This may suggest external atmospheric deposition, rapid internal biotransformation or excretion of these compounds. Accordingly, we suggest prioritising plasma for PFASs analyses, while the sources of PFRs, NBFRs and DPs in feathers and plasma need further investigation.

1. Introduction

Polychlorinated biphenyls (PCBs), organochlorinated pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) are compounds previously used in industrial applications, agriculture and consumer products (Mackay et al., 2006). Classified as persistent organic pollutants (POPs), these compounds are generally lipophilic, semi-volatile and resistant to chemical and

biological degradation (Buccini, 2003; Mackay et al., 2006). Consequently, POPs persist in the environment (Letcher et al., 2010; Mackay et al., 2006) and may result in high uptake in biota, followed by bioaccumulation and biomagnification, especially in long and lipid-rich food webs (Borgå et al., 2004; Jones and de Voogt, 1999). As replacements for the legacy POPs regulated by the Stockholm Convention (UNEP, 2009), new and (re-) emerging contaminants (ECs) have entered the market. Those include phosphorus flame retardants (PFRs; van der Veen and de Boer, 2012), "novel" brominated flame retardants (NBFRs; Covaci et al., 2011), dechlorane plus (DPs; Sverko et al., 2011) and certain per- and polyfluoroalkyl substances (PFASs; Lau et al., 2007). These ECs exhibit different physicochemical properties than the legacy POPs and may accumulate in other matrices, such as protein-rich tissues (Lau et al., 2007), or become rapidly metabolised and excreted (Briels et al., 2018; Covaci et al., 2011; van der Veen and de Boer, 2012).

Wild birds are important biomonitors for numerous environmental contaminants (Burger and Gochfeld, 2004; Furness, 1993). Due to ethical and species conservational aspects, non-destructive sampling methods such as the collection of blood or addled eggs are often applied in environmental monitoring programs of wild birds (Espín et al., 2016). The contaminant concentrations detected in blood plasma provide a snapshot of recent exposure through diet (Henriksen et al., 1998), but during periods of low food availability or starvation concentrations can also originate from internal fat reserves (re-exposure) (Fenstad et al., 2014). Egg concentrations on the other hand reflect maternal concentrations deposited during the egg formation (Becker and Sperveslage, 1989). Feathers, either plucked or moulted, present another non-destructive sampling matrix. Feathers are connected to the blood circulation during formation and growth, and during this period the internal contaminant concentrations may

thereby be transferred and deposited into the feather (Jaspers et al. 2006; García-Fernández et al., 2013).

The use of feathers as a non-destructive matrix for biomonitoring is increasing (García-Fernández et al., 2013; Gómez-Ramírez et al., 2014). While feathers have been used for decades as a matrix for monitoring environmental concentrations of metal (Burger, 1993), it was only in the early 2000s that feathers were proposed for legacy POP analyses (Dauwe et al., 2005; Jaspers et al., 2006). Recently, feathers have also been investigated as a matrix for analysing and monitoring PFASs (Gómez-Ramírez et al., 2017; Jaspers et al., 2013; Li et al., 2017; Meyer et al., 2009), and only a few studies published to date have investigated the suitability of NBFRs and PFRs monitoring in feathers (Eulaers et al., 2014; Svendsen et al., 2018). Consequently, little is known about the exposure to and deposition of these ECs into feathers. Preen oil has also been proposed as a non-destructive matrix for monitoring PCBs, PBDEs and OCPs (Eulaers et al., 2011b; Van den Brink, 1997), but few studies have collected preen oil for contaminant analyses (Eulaers et al., 2011a, 2011b; Van den Brink, 1997).

Studies investigating non-destructive sampling matrices in birds have been conducted on a wide variety of bird species (García-Fernández et al., 2013). However, there is a general lack of studies with larger sample sizes that have investigated both legacy POPs and ECs in several non-destructive matrices (Espín et al., 2016; García-Fernández et al., 2013). This may improve the evaluation of the suitability of these matrices for monitoring purposes. An overview of contaminant monitoring activities in Europe revealed that 100 monitoring programs from 28 countries have included feathers samples from birds of prey (Espín et al., 2016).

Due to their apex trophic position, large body size and long lifespan, birds of prey such as the white-tailed eagle (*Haliaeetus albicilla*), are good sentinel species for monitoring the presence of contaminants in the environment (Burger and Gochfeld, 2004). White-tailed eagle nestlings are stationary in their nests and therefore good indicators of local exposure to a wide range of environmental contaminants (Olsson et al., 2000). They are also relatively easy to sample while still in the nest (Espín et al., 2016; Eulaers et al., 2011b). The white-tailed eagle was listed as threatened by the International Union for Conservation of Nature in 1988, but today it is listed as of least concern (Birdlife Int., 2016).

In this study, we aimed to evaluate if body feathers and preen oil from white-tailed eagle nestlings present a good non-destructive matrix to monitor internal concentrations of both legacy POPs and ECs. Consequently, we investigated concentrations of legacy POPs and ECs in plasma, feathers and preen oil from 70 white-tailed eagle nestlings. Furthermore, we investigated correlations of POP and EC concentrations in these matrices and evaluated the consistency of these results by including samples from two field locations during two consecutive years. As the sampled feathers were still growing and connected to the blood circulation, we expected to find strong correlations between feathers and plasma concentrations of POPs and ECs. We also expected to find strong correlations between plasma and preen oil, as the oil is produced by an internal gland which is connected to the blood circulation.

2. Materials and methods

2.1. Field sampling

The study was conducted on 70 white-tailed eagle nestlings from two archipelagos in Norway, Smøla (63.35°N; 8.03°E) and Steigen (67.93°N; 14.98°E), during the breeding seasons of 2015 and 2016. We sampled 13 nestlings in Smøla in 2015 and 22 nestlings in 2016. In Steigen, 14

125 nestlings were sampled in 2015 and 21 nestlings in 2016. All nestlings, aged from 8-12 weeks
126 old, were caught at the nest site and handled for approximately 15 min. Body feathers were
127 gently pulled from the dorsal region, approximately 10 per individual, and stored in
128 polyethylene zipper bags (VWR, USA) at -20°C. A blood sample of 8 mL was collected in
129 heparinised vacutainers through brachial venepuncture. The blood samples were centrifuged
130 (860 g), after which plasma was transferred to cryogenic tubes (Nalgene®, USA) and stored at
131 -20 °C. Preen oil could only be collected in a sufficient amount in 2016. It was collected in a
132 1.5 mL Eppendorf tube (VWR, USA) by massaging the preen gland using disposable gloves
133 and avoiding traces of feathers in the sample. The sampling was approved by the Norwegian
134 Food Safety Authority (Mattilsynet; 2015/6432 and 2016/8709) and the handling of the birds
135 were in accordance with the regulations of the Norwegian Animal Welfare Act.
136
137 **2.2. Chemical analyses**
138 ***2.2.1. Feather pre-treatment***
139 Clean stainless steel and glass tools were used to wash and cut the feathers. Tools were
140 thoroughly rinsed between individual samples with acetone for POP and EC analyses and
141 methanol for PFASs analyses. The feather quills (calamus) were removed and remaining
142 feathers were washed in MilliQ-water to remove dust and particles from the feathers prior to
143 analysis (Jaspers et al., 2007a, 2007b, 2008). For a thorough wash, two pairs of tweezers were
144 used to separate the barbs by pulling the barbs downwards and away from each other. Feathers
145 were placed on clean lab paper, covered with tissue paper (Facial tissues, VWR) and dried
146 overnight at room temperature. Finally, the feathers were cut into approximately 1–2 mm
147 pieces and homogenates were accurately weighed prior to analyses (range: 0.10 – 0.40 grams).
148 The feather pre-treatment was conducted on the bench in a clean lab (not used for chemical
149 analyses).

2.2.2. Legacy POPs and ECs

Chemical analyses of legacy POPs and ECs in feathers, plasma and preen oil were performed at the Toxicological Centre of the University of Antwerp, Belgium. The targeted compounds for the analyses were 23 PCBs, 10 OCPs, seven PBDEs, eight PFRs, three NBFRs and two DPs. The full compound list can be found in the Supplementary information (SI), Tables S1 and S2. Contents of the internal standards (IS1 (POPs), IS2 (ECs) and IS3 (DPs)) can be found in Table S3.

Plasma extraction: One mL of plasma was spiked with 100 µL IS1 and with 40 µL IS2. To this, 1 mL of Milli-Q water, 200 µL of formic acid (98 %) and 4 mL of the extraction solvent *n*-hexane/dichloromethane (DCM) mixture (4:1, v/v) were added before 1 min of vortexing. This mixture was then centrifuged for 5 min (2200 g) before the organic layer was transferred to a clean glass tube. This extraction was repeated before the extracts were evaporated to near dryness and resolubilised in 0.50 mL *n*-hexane followed by 1 min vortexing.

Feather extraction: To approximately 200 mg feathers, 100 µL of IS1, 40 µL of IS2, 5 mL of hydrochloric acid (HCl, 4M) and 5 mL of *n*-hexane/DCM mixture (4:1, v/v) were added before the samples were incubated at 45 °C overnight. The incubated sample solutions were vortexed thoroughly for 1 min and the organic layer was retrieved. This liquid-liquid extraction was repeated with 5 mL of *n*-hexane/DCM mixture (4:1, v/v). Extracts were then evaporated to near dryness (~200 µL) by a gentle nitrogen stream and resolubilised in 0.50 mL *n*-hexane followed by 1 min vortexing.

Preen oil extraction: Between 13-40 mg of preen oil was transferred to clean glass tubes using a spatula and the accurate weight was recorded. The spatula was thoroughly cleaned with

175 acetone between samples. Prior to extraction, the samples were spiked with 100 µL IS1 and 20
176 µL of IS3. Subsequently, 2 mL of *n*-hexane was added to the spiked sample, which was then
177 vortexed for 1 min.
178
179 Further clean-up and fractionation of all sample extracts were performed according to Eulaers
180 et al. (2011b) and Poma et al. (2017), with slight modifications. Detailed descriptions of these
181 modifications are available in the SI. The preen oil samples could not be analysed for PFRs
182 due to the high lipid content of the oil which made it difficult to use the PFR clean-up procedure
183 to get a lipid-free extract. After clean-up, all the extracts were concentrated to near dryness
184 under a gentle nitrogen stream and resolubilised in 100 µL of iso-octane. For each batch of 24
185 samples, 100 µL of recovery standard (CB 207, 50 pg/µL in iso-octane/toluene 9:1, v/v) was
186 added to five of the samples and vortexed for 30 s. Extracts were transferred to injection vials
187 and analysed by gas chromatography with electron capture negative ionization and mass
188 spectrometry (GC-ECNI/MS) according to Eulaers et al. (2011b) for legacy POPs and Poma et
189 al. (2017) for ECs (details in SI).
190 ***2.2.3. Per- and polyfluoroalkyl substances***
191 The analysis of PFASs in feathers and plasma was performed at the Norwegian Institute of Air
192 Research in Tromsø, Norway. The targeted PFASs were one perfluorinated sulfonamide, seven
193 perfluorinated sulfonates and 11 perfluorinated acids. See Table S4 for the full list of targeted
194 compounds. The preen oil samples were not analysed for PFASs due to their high lipid content
195 and small sample volumes. The contents of the internal standard for PFASs (IS4) can be found
196 in Table S3.
197
198 **Plasma extraction:** Plasma samples were extracted and analysed according to Herzke et al.
199 (2009). Aliquots of 200 and 300 µL of plasma were thawed and homogenised, then spiked with

20 µL of IS4. One mL methanol (MeOH) was added to the samples and the solutions were mixed by shaking and vortexing for 1 min. The samples were ultrasonicated three times for 10 min, with intermittent vortexing. To enhance phase separation and sedimentation, the samples were centrifuged for 10 min (1500 g). The supernatant (methanol phase) was then purified in 1.70 mL Eppendorf tubes (VWR, USA) containing 25 mg Supelclean™ ENVI-Carb™ graphitised carbon absorbent (Sigma-Aldrich, USA) and 50 µL glacial acetic acid. After centrifuging for 10 min (1500 g), an exact volume of 0.50 mL supernatant was transferred to glass vials and added 20 µL of recovery standard solution (3,7-diMe-PFOA, 0.102 ng/µL).

Feather extraction: Feather samples were extracted and analysed according to Jaspers et al. (2013). Pre-cleaned and homogenised feathers were transferred to sterile polypropylene tubes (VWR, USA). For preen oil removal, feather homogenates were immersed in 20 mL of *n*-hexane and ultra-sonicated for 10 min. The *n*-hexane was decanted after centrifugation and the tubes with the homogenates were dried overnight. Previous tests have shown no removal of PFASs from feathers by *n*-hexane washes (pers. comm. Dorte Herzke). When dry, samples were spiked with 20 µL of IS4. To resolve the PFASs bound to proteins in the feather homogenate, we added 2 mL 200 mM NaOH in MeOH. The homogenate was then vortexed for 1 min and set to soak for 60 min. Then, we added 10 mL of MeOH and the homogenate was mixed, ultra-sonicated for 3 x 10 min and let to soak overnight. The next day, PFASs were further extracted from the samples by adding 200 µL of 2M HCl in MeOH. Extracts were then centrifuged for 5 min at 1500 g, transferred to new polypropylene tubes and evaporated to 1 mL with RapidVap (Labconco, USA). The 1 mL extracts were then cleaned up with carbon and recovery standard was added similar as to the plasma samples.

Prior to quantification analysis, extract aliquots of 100 µL were transferred to autosampler vials with insert and an equal amount of 2 mM aqueous ammonium acetate was added. The extracts were then refrigerated until quantification. Quantification was performed according to Hanssen et al. (2013), using ultrahigh pressure liquid chromatography and triple–quadrupole mass-spectrometry (UHPLC-MS/MS). All labelled and internal standards were provided by NILU (IRMM-427, ID 0119) and all solvents were purchased from Merck (Darmstadt, Germany).

2.2.4 Quality assurance and quality control

Quality assurance of the analytical method was carried out by measurements of procedural blanks and standard reference material (SRM). For POPs extractions from plasma, the SRM was human plasma from the AMAP (Arctic Monitoring and Assessment Program) interlaboratory exercise. For PFASs extractions from plasma, the SRM was a commercially available human plasma sample (NIST SRM 1957, USA). For POPs extraction from preen oil, the SRM was whale blubber (NIST, SRM 1945, USA). These SRMs were used to control the performance of the analytical method for every 10^{th} sample, together with a procedural blank. No SRM was available for feather samples. However, a procedural blank was analysed for every 10^{th} sample and recoveries of internal standards calculated for every sample as well as for the blanks. For legacy POPs, PFRs, NBFRs and DPs, the limits of quantification (LOQs) were calculated as three times the standard deviation of the procedural blanks for each compound and sample type. For PFAS, the LOQs were calculated as three times the limit of detection (LOD), which again was calculated as the sum of the average of the procedural blanks and three times the signal-to-noise ratio for each compound and sample type. The LOQs for all compounds are available in the SI (Table S1, S2 and S4). For analytes that were not detected in the blanks, LOQs were set to ten times the signal-to-noise ratio of the sample runs. Recoveries of internal standards can be found in Table S5 and S6. No contamination was

observed in the feather blanks. For plasma, only perfluorohexane sulfonate (PFHxS) was observed in 33 % of the blanks at average concentration of 0.15 ng/mL. No blank corrections were carried out for any of the investigated compounds. All PFAS samples fulfilled the requirements for QA/QC except for PFDoA were recoveries were less than 50 % in some occasions, and lower than 35 % for one sample. Even so, the low standard deviations give good confidence in the robustness of the applied method.

2.3. Statistical analyses

Statistical analyses were performed using R version 3.4.2. Descriptive statistics of all the investigated compounds are available in the SI (Table S7 – S13). Concentrations of the compounds are expressed in ng/mL wet weight (ww) for plasma, ng/g ww for feathers and ng/g ww for preen oil. Data were treated in the same way as in most previous studies on bird feathers and preen oil to allow for direct comparison (i.e. Eulaers et al., 2011a, 2011b, Gómez-Ramírez et al., 2017). Thus, compounds quantified in over 50 % of samples and detected in both feathers and plasma samples within each location and each year were included in the statistical analyses. Data below the LOQ were substituted with LOQ * detection frequency (DF) within the year and location of each matrix (Voorspoels et al., 2002). Results from Shapiro-Wilk's test for normality and visual inspection of normal quantile-quantile plots showed that the concentrations of the compounds were not normally distributed, some not even after $\log_e$ transformation. All statistical analyses were therefore performed using non-parametric tests on untransformed data. Significance levels were set to $\alpha = 0.05$.

Concentration differences between years and locations of Σ_9PCBs, Σ_3OCPs, 2,2',4,4'-tetrabromodiphenyl ether (BDE 47) and perfluoroundecanoic acid (PFUnA) were investigated by Kruskal-Wallis analyses for each matrix separately. Significant Kruskal-Wallis analyses

were further investigated by Dunn's test of multiple comparisons with Bonferroni correction. Correlations between concentrations in the three matrices of each selected compound were investigated by Spearman's rank correlation (r_S) for each year and location separately, as well as combined. Concentrations from both years (n = 70) were included for feather and plasma correlations. For preen oil, only data from 2016 (n = 43) could be included. PFUnA concentrations in feathers and plasma from 2015 and 2016 were not in a monotonic relationship due to the large differences between the years and thus could not be analysed with Spearman's rank correlation. To investigate if a larger sample size could create a monotonic relationship, we included PFUnA concentrations reported in feathers and plasma from white-tailed eagle nestlings sampled in 2014 in Steigen (data from Gómez-Ramírez et al. 2017). By adding the latter samples and pooling samples from 2014 – 2016, a monotonic relationship was established between feather and plasma concentrations, and the relationship was analysed by Spearman's rank correlation.

3. Results

3.1. Detection frequencies and concentrations of legacy POPs and ECs

The compounds that were quantified in over 50 % of both plasma and feather samples from all white-tailed eagle nestlings within each year and location (n = 70) included nine PCBs (Σ_9PCBs: CB 99, 101, 105, 118, 138, 153, 170, 180 and 187), three OCPs (Σ_3OCPs: *oxy*-chlordane (OxC), dichlorodiphenyldichloroethylene (*p,p'*-DDE) and dichlorodiphenyl-trichloroethane (*p,p'*-DDT)), BDE 47 and PFUnA. Table 1 presents the median (min-max) concentrations of these compounds, while the same info for all 54 targeted compounds is listed in the SI (Table S7 – S13). The concentrations of Σ_9PCBs, Σ_3OCPs, BDE 47 and PFUnA differed between the three matrices, and the general concentration pattern on a wet weight basis was preen oil > feathers > plasma. The most abundant compounds (highest concentrations)

among the legacy POPs were CB 153, *p,p'*-DDE and BDE 47 in all matrices (Table 1). For PFASs, linear PFOS was found in the highest concentrations in both plasma (2.3 – 31.9 ng/mL, mean DF: 100 %) and feathers (< 0.03 – 90.2 ng/g, mean DF: 48.9 %) (Table S12). However, the average detection frequencies of linear PFOS in feathers over the two years were strongly influenced by the low detection frequencies in 2015 of only 8 and 29 % in Smøla and Steigen, respectively. Detection frequencies of all targeted PFASs averaged at 78 % in plasma and 38 % in feathers, with higher concentrations detected in plasma than in feathers. On the contrary, perfluorooctanesulfonamide (PFOSA) was only detected in feathers, at detection frequencies between 46 – 100 % in both years and locations. Of the PFRs, the most abundant compounds were tris(1,3-dichloro-2-propyl) phosphate (TDCIPP) in plasma (< 0.2 – 1.4 ng/mL, mean DF: 17.8 %) and triphenyl phosphate (TPhP) in feathers (< 1.0 – 1229.3 ng/g, mean DF: 94 %) (Table S11), however at detection frequencies lower than 50 % in feathers and plasma, respectively. Contrary to PFASs, the concentrations of PFRs detected in feathers exceeded those in plasma and the average detection frequencies of the targeted PFRs were 13 % in plasma and 80 % in feathers. The most abundant NBFR was bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (TBPH) in both plasma (0.08 ng/mL) and feathers (< 0.40 – 1.03 ng/g) (Table S11), but both at low detection frequencies (< 5 % and < 36 %, respectively). No NBFRs were detected in the preen oil. Of the DPs, the most dominating isomer was *anti*-DP in both plasma (< 0.002 – 0.03 ng/mL, mean DF: 12 %) and feathers (< 0.10 – 1.14 ng/g, mean DF: 21 %) (Table S11). *Anti*-DP was also detected in one of the preen oil samples, at 0.45 ng/g.

3.1.1 Differences between locations and years

We detected significant concentration differences for Σ_9PCBs, Σ_3OCPs, BDE 47 and PFUnA in plasma ($\chi^2_{(70,3)}$ = 9.04 – 51.2, $p < 0.05$) and feathers ($\chi^2_{(70,3)}$ = 28.8 – 34.0, $p < 0.05$) between the two years and between locations (Table S7 – S12). The median concentrations of these

contaminant groups were generally higher in feathers and plasma samples from Steigen than Smøla. For preen oil, the median concentrations were also slightly higher in Steigen than in Smøla, although not significantly ($\chi^2_{(43,1)} = 0.3 – 0.7$, $p > 0.05$).

3.2. Correlations between matrices

3.2.1 Plasma and feather correlations

Strong and significant positive correlations between plasma and feather concentrations were found for all PCBs, OCPs (except for *p,p'*-DDT) and BDE 47 (r_S: 0.33 – 0.95, $p < 0.02$), when both years and locations were combined (Table 2, Figure 1). When years and locations were investigated separately, we detected significant positive correlations between plasma and feathers for all compounds ($r_S = 0.43 – 0.93$, $p < 0.05$), except for CB 101, 105 and 180. Contrary to the POPs, a correlation analysis of PFUnA concentrations in feathers and plasma on all samples combined was not possible in the current study, as the relationship was non-monotonic. However, when the years and locations were analysed separately, positive and significant correlations within Steigen were detected for 2015, as well as for 2016 ($r_S = 0.69$ and 0.56, respectively, $p < 0.01$). A study from Gómez-Ramírez et al. (2017) has investigated PFASs in plasma and feathers from white-tailed eagle nestlings from Steigen, sampled in 2014. Since data from Gómez-Ramírez et al. (2017) were produced in the same lab, using the same methodology, we combined the raw data from their study with our data and performed new statistical analysis (since a monotonic relationship was achieved). However, no correlation was detected between PFUnA concentrations in plasma and feathers on the combined data ($r_S = 0.001$, $p = 0.99$; Figure 1). Figure 1 illustrates a highly scattered distribution, indicating that the concentrations of PFUnA in these two matrices are highly variable and poorly correlated, both within locations and years.

Table 1: Summary statistics [median (min – max)] of contaminants quantified in over 50 % of plasma and body feathers samples within each year and location in white-tailed eagle nestlings from Smøla and Steigen (Norway). Concentrations of other PFASs, PFRs and NBFRs were below LOQ in > 50% of the samples and can be found in Supplementary information. The preen oil concentrations (ng/g ww) were only available from 2016. Units are ng/ml ww for plasma and ng/g ww for feathers. Samples not available for analyses are marked with "n.a".

		Smøla		**Steigen**	
		2015 (*n* = 13)	**2016** (*n* = 22)	**2015** (*n* = 14)	**2016** (*n* = 21)
	Matrix	median (min - max)	median (min – max)	median (min – max)	median (min – max)
CB 99	Plasma	0.16 (0.08 – 0.59)	0.18 (0.06 – 1.47)	0.50 (0.18 – 4.61)	0.23 (0.06 – 0.98)
	Feathers	1.10 (0.18 – 3.85)	0.92 (0.41 – 7.89)	7.78 (2.71 – 31.05)	1.08 (0.21 – 3.61)
	Preen oil	n.a	28.00 (12.34 – 198.68)	n.a	33.59 (0.95 – 161.58)
CB 101	Plasma	0.21 (0.09 – 0.31)	0.14 (0.01 – 0.56)	0.16 (0.07 – 0.56)	0.12 (0.02 – 0.25)
	Feathers	0.72 (0.18 – 1.82)	0.55 (0.31 – 1.57)	1.50 (0.80 – 1.81)	0.44 (0.19 – 0.99)
	Preen oil	n.a	16.68 (9.99 – 49.55)	n.a	19.86 (0.95 – 32.62)
CB 105	Plasma	0.08 (0.04 –0.30)	0.11 (0.04 – 0.79)	0.26 (0.10 – 2.59)	0.14 (0.04 – 0.66)
	Feathers	0.23 (0.11 – 0.57)	0.24 (0.11 – 0.94)	1.37 (0.52 – 3.85)	0.31 (0.12 – 1.05)
	Preen oil	n.a	17.94 (7.99 – 91.04)	n.a	22.33 (7.99 – 91.04)
CB 118	Plasma	0.23 (0.11 –0.81)	0.41 (0.17 – 2.92)	0.70 (0.28 – 7.30)	0.50 (0.14 – 2.30)
	Feathers	0.72 (0.23 – 2.1)	0.90 (0.45 – 4.12)	5.12 (2.18 – 15.03)	1.07 (0.4 – 3.37)
	Preen oil	n.a	51.84 (19.68 – 240.09)	n.a	66.46 (23.95 – 289.96)
CB 138	Plasma	0.27 (0.11 –1.25)	1.10 (0.40 – 10.55)	0.66 (0.29 – 5.63)	1.26 (0.28 – 8.88)
	Feathers	0.42 (0.17 – 1.51)	1.86 (0.75 – 11.78)	1.34 (0.60 – 5.89)	2.64 (0.62 – 6.26)
	Preen oil	n.a	129.63 (39.37 – 720.65)	n.a	168.07 (40.26 – 602.9)
CB 153	Plasma	0.74 (0.21 – 3.06)	1.44 (0.55 – 9.48)	2.05 (1.12 – 26.27)	1.75 (0.43 – 10.16)
	Feathers	1.77 (0.63 – 6.77)	3.13 (1.22 – 17.07)	12.86 (5.48 – 38.64)	4.15 (0.92 – 9.73)
	Preen oil	n.a	259.08 (81.81 – 1420.57)	n.a	327.62 (94.03 – 1164.43)
CB 170	Plasma	0.07 (0.02 – 0.36)	0.22 (0.07 – 1.30)	0.18 (0.06 – 2.16)	0.23 (0.07 – 1.98)
	Feathers	0.14 (0.07 – 0.48)	0.34 (0.13 – 1.45)	0.52 (0.27 – 1.73)	0.42 (0.11 – 1.23)
	Preen oil	n.a	37.74 (10.16 – 180.79)	n.a	50.51 (12.85 – 184.32)
CB 180	Plasma	0.17 (0.04 – 0.84)	0.70 (0.20 – 3.55)	0.45 (0.13 – 5.29)	0.65 (0.19 – 5.89)
	Feathers	0.23 (0.10 – 1.03)	0.74 (0.26 – 3.06)	0.96 (0.56 – 3.30)	0.92 (0.24 – 2.73)
	Preen oil	n.a	117.86 (29.89 – 489.81)	n.a.	139.58 (40.9 – 579.26)
CB 187	Plasma	0.11 (0.03 – 0.43)	0.32 (0.13 – 2.55)	0.22 (0.07 – 1.81)	0.36 (0.10 – 1.95)
	Feathers	0.17 (0.08 – 0.78)	0.48 (0.17 – 3.21)	0.76 (0.26 – 1.89)	0.45 (0.14 – 1.08)
	Preen oil	n.a	55.94 (16.74 – 434.19)	n.a	59.11 (20.75 – 186.85)
Σ_9 PCB	Plasma	1.90 (0.78 – 7.97)	4.44 (1.70 – 32.37)	4.87 (2.81 – 56.17)	5.34 (1.44 – 32.98)
	Feathers	5.56 (1.86 – 18.39)	9.20 (3.86 – 51.10)	34.10 (14.89 – 101.29)	11.94 (2.96 – 27.61)
	Preen oil	n.a	723.56 (231.26- 3804.89)	n.a	863.79 (269.37 – 3196.99)
OxC	Plasma	0.04 (0.02 – 0.14)	0.08 (0.02 – 0.53)	0.24 (0.05 – 2.16)	0.13 (0.04 – 0.6)
	Feathers	0.11 (0.07 – 0.32)	0.26 (0.13 – 1.28)	1.36 (0.36 – 6.89)	0.48 (0.08 – 1.47)
	Preen oil	n.a	9.52 (3.36 – 30.37)	n.a	13.54 (4.80 – 53.73)
***p,p'*-DDE**	Plasma	1.21 (0.56 – 5.23)	1.20 (0.56 – 9.47)	3.95 (2.18 – 47.61)	1.45 (0.48 – 8.64)
	Feathers	4.96 (2.35 – 22.1)	3.23 (1.48 – 17.03)	26.59 (12.38 – 94.38)	3.74 (1.14 – 8.81)
	Preen oil	n.a	298.56 (170.99 -1447.32)	n.a	364.88 (185.38 – 932.96)
***p,p'*-DDT**	Plasma	0.20 (0.08 – 0.30)	0.15 (0.06 – 0.63)	0.12 (0.02 – 0.31)	0.27 (0.02 – 0.38)
	Feathers	0.50 (0.14 – 1.14)	0.24 (0.15 – 1.03)	0.89 (0.17 – 1.92)	0.24 (0.14 – 0.47)
	Preen oil	n.a	7.07 (3.85 – 15.09)	n.a	7.65 (5.06 – 46.63)
Σ_3 OCP	Plasma	1.54 (0.67 – 5.62)	1.47 (0.64 – 10.64)	4.37 (2.40 – 49.83)	3.48 (1.31 – 12.96)
	Feathers	5.68 (2.95 – 23.56)	4.12 (2.03 – 18.63)	28.62 (13.98 – 103.19)	4.36 (1.37 – 10.08)
	Preen oil	n.a	319.63 (180.46 – 1491.16)	n.a	384.06 (198.87 – 1024.27)
BDE 47	Plasma	0.06 (0.03 – 0.28)	0.08 (0.01 – 0.81)	0.19 (0.06 – 1.82)	0.09 (0.01 – 0.36)
	Feathers	0.56 (0.28 – 1.58)	0.30 (0.16 – 2.43)	1.73 (0.63 – 3.59)	0.45 (0.14 – 1.06)
	Preen oil	n.a	9.19 (4.25 – 96.35)	n.a	10.47 (4.45 – 60.28)
PFUnA	Plasma	3.59 (2.43 – 4.36)	1.15 (0.68 – 2.05)	3.36 (2.3 – 5.08)	1.40 (0.94 – 2.15)
	Feathers	0.15 (0.05 – 0.6)	0.58 (0.07 – 0.95)	0.38 (0.13 – 0.81)	0.82 (0.26 – 1.07)*
	Preen oil	n.a	n.a	n.a	n.a

* Feathers for PFUnA Steigen 2016: *n* = 19

3.2.2 Plasma and preen oil correlations

Significant positive correlations were found for all compounds that were detected > 50 % in preen oil and plasma sampled in 2016, when both locations were combined ($r_S > 0.35$, $p < 0.02$, Table 2). When samples from Steigen and Smøla were analysed for correlations separately, significant positive correlations were found for all compounds except CB 101 from Smøla ($r_S = 0.21$, $p < 0.35$). Unfortunately, the preen oil samples could not be analysed for PFRs and PFAS due to their high lipid content and small sample volume.

3.2.3 Feathers and preen oil correlations

Similar to plasma and feathers, strong and significant correlations were found between feathers and preen oil concentrations from 2016 for all compounds detected > 50 % ($r_S > 0.46$, $p < 0.01$), except for *p,p'*-DDT (Table 2), when both locations were combined. When samples from Smøla and Steigen were analysed for correlations separately, the relationships between feathers and preen oil from Smøla were weak and not significant for CB 101, *p,p'*-DDT and BDE 47 ($r_S = 0.18 – 0.25$, $p > 0.3$).

Table 2: Spearman's correlation coefficients (r_S) and significance values (p) between contaminant concentrations in blood plasma, body feathers and preen oil from white-tailed eagle nestlings from Smøla and Steigen (Norway). Correlation coefficients are not available for PFUnA due to the non-monotonic relationship between feathers and plasma. Correlations for compounds not present in both matrices could not be calculated ("n.a."). Significant *p*-values are marked with *.

	Blood plasma ~ body feathers		**Blood plasma ~ preen oil**		**Body feathers ~ preen oil**	
	n = 70		*n* = 43		*n* = 43	
	r_S	*p*-value	r_S	*p*-value	r_S	*p*-value
CB 99	0.73	< 0.001*	0.78	< 0.001*	0.81	< 0.001*
CB 101	0.33	0.006*	0.35	0.02*	0.46	0.002*
CB 105	0.76	< 0.001*	0.86	< 0.001*	0.74	< 0.001*
CB 118	0.74	< 0.001*	0.86	< 0.001*	0.77	< 0.001*
CB 138	0.78	< 0.001*	0.83	< 0.001*	0.80	< 0.001*
CB 153	0.72	< 0.001*	0.83	< 0.001*	0.79	< 0.001*
CB 170	0.73	< 0.001*	0.77	< 0.001*	0.73	< 0.001*
CB 180	0.72	< 0.001*	0.76	< 0.001*	0.73	< 0.001*
CB 187	0.72	< 0.001*	0.78	< 0.001*	0.79	< 0.001*
OxC	0.83	< 0.001*	0.95	< 0.001*	0.75	< 0.001*
***p,p'*-DDE**	0.73	< 0.001*	0.84	< 0.001*	0.71	< 0.001*
***p,p'*-DDT**	0.12	0.32	0.52	< 0.001*	0.24	0.116
BDE 47	0.67	< 0.001*	0.86	< 0.001*	0.73	< 0.001*
PFUnA	-	-	n.a.	n.a.	n.a.	n.a.

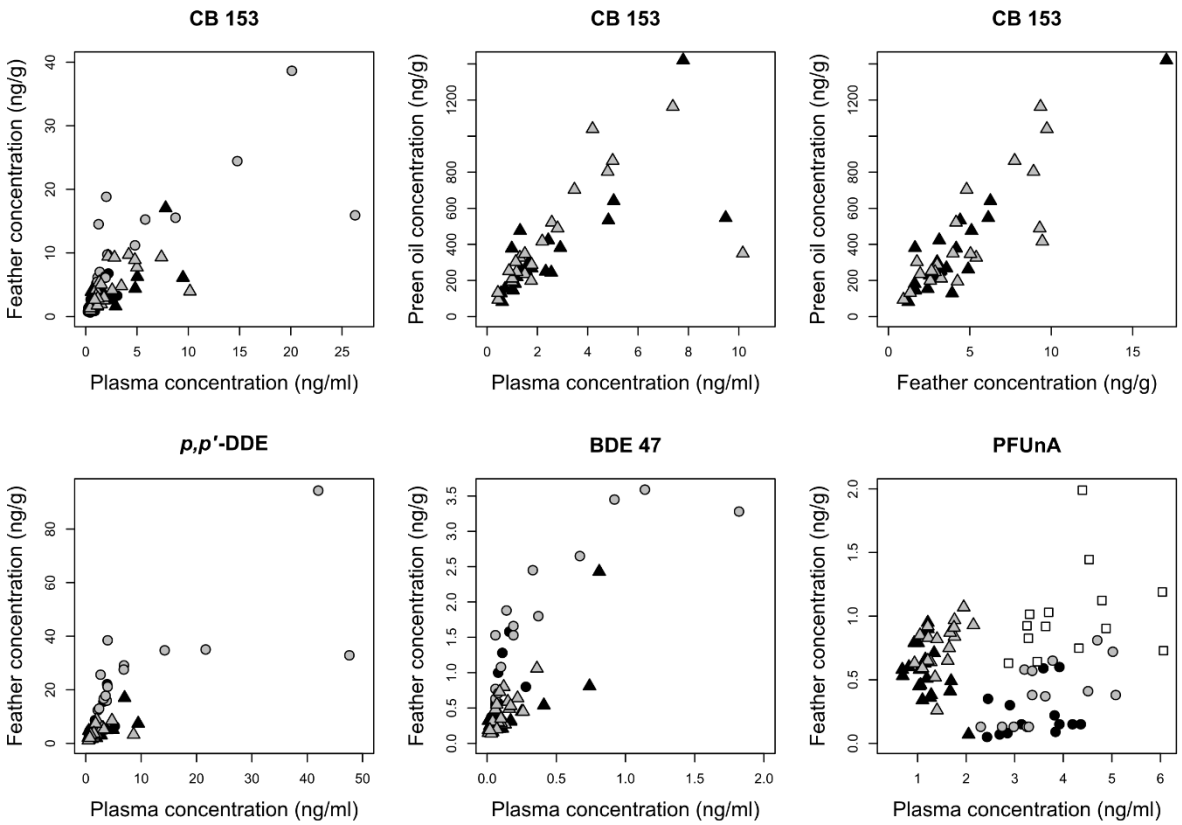

Figure 1: Correlation plots of concentrations in plasma (ng/ml ww), feathers (ng/g ww) and preen oil (ng/g ww) of CB 153, *p,p'*-DDE, BDE 47 and PFUnA from white-tailed eagle nestlings from Steigen in grey and from Smøla in black. Samples from 2015 are in circles (○), while samples from 2016 are in triangles (Δ). Preen oil was only sampled in 2016. Samples from Gómez-Ramírez et al. 2017 are in open squares (□).

4. Discussion

4.1 Detection frequencies and concentrations of legacy POPs and ECs

We expected to find that the concentrations and detection frequencies of legacy POPs and ECs in plasma would also be reflected in the feathers and preen oil. This was true for legacy POPs, as we found high detection frequencies for PCBs, OCPs and BDE 47 in plasma, feathers and preen oil. The concentration profile with high concentrations in the preen oil was expected due to the high lipid content of the oil and the lipophilic nature of these compounds (Eulaers et al., 2011b). The concentrations of the main contaminant contributors (CB 153, *p,p'*-DDE, BDE 47 and PFUnA, Table 1) in plasma, feathers and preen oil were slightly lower in the current study than previously reported in white-tailed eagle nestlings from Norway (Eulaers et al., 2011a, 2011b, 2013, 2014; Gómez-Ramírez et al., 2017). The samples from the previous studies were

collected in 2008, 2009, 2011 and 2014, and some also at other locations than Smøla and Steigen. Hence, some of this variation may be due to temporal, spatial, biological or dietary differences (Eulaers et al., 2013, Løseth et al., in preparation).

The detection frequencies of the analysed ECs differed between plasma and feathers and may suggest different exposure routes or different toxicokinetics in the two matrices. The higher detection frequencies of PFASs in plasma than in feathers, is contrary to a study were PFOS concentrations were compared between keratinous tissues (hair and nails) and serum in humans (Li et al., 2013). Although the detection frequencies of PFASs were low in feathers in the current study, the concentrations correspond to those reported in a previous study on feathers from white-tailed eagle nestlings (Gómez-Ramírez et al., 2017).

The higher concentrations of PFOSA, PFRs, NBFRs and DPs in feathers than in plasma, suggest that the feathers may not only reflect the internal contamination burden. Some of the concentrations may potentially originate from external contamination (Eulaers et al., 2014; Jaspers et al., 2008). Possible sources of external contamination can come from outdoor environments, field accommodations or other indoor environments (Cequier et al., 2014; Green et al., 2008, Möller et al., 2011, Tollbäck et al. 2006). The field accommodation at Smøla 2015 was a newly built house and even though the feathers were thoroughly rinsed before contaminant extraction, their PFR profile of TPhP > tris(2-chloroisopropyl) phosphate (TCPP) > tris(chloroethyl) phosphate (TCEP) show similarities to profiles reported in indoor air and dust (Cequier et al., 2014; Green et al., 2008, Tollbäck et al. 2006). The general PFR profile detected in feathers from the other location and years was TCPP > TCEP > TPhP. This same profile has been reported in a study of atmospheric air from the North Sea (Möller et al., 2011) and at a remote Arctic location (Green et al., 2008). *Anti*-DP and TBPH were also detected in

higher concentrations in feathers than in plasma. These compounds have also been detected in indoor air and dust from Norway (Cequier et al., 2014). The similarity of PFR profiles and the occurrence of *anti*-DP and TBPH in feathers and in air further suggests that feathers may act as air- and dust samplers. Some of the detected concentrations may therefore originate from external contamination, which may not have been removed by the washing procedure.

The lower detection frequencies and concentrations of PFRs and TBPH in plasma, compared to feathers, may also result from rapid metabolism and excretion of these compounds from internal tissues as reported in other studies (Bearr et al., 2012; Briels et al., 2018; Covaci et al., 2011; Hou et al., 2016). The PFRs and NBFRs detected in the current study have previously been detected in white-tailed eagle samples, primarily in feathers, from Trøndelag and Troms, Norway (Eulaers et al., 2014). As in Eulaers et al. (2014), our study found low concentrations and detection frequencies of PFRs and NBFRs in plasma, further suggesting high excretion rates, low absorption or low exposure of these compounds. Dechloranes, on the contrary, may not biotransform (Briels et al., 2018) and can accumulate in biota (Feo et al., 2012). To our knowledge, this is the first study detecting DPs in feathers and preen oil, and further studies are therefore needed to investigate if the concentrations of PFRs, NBFRs and DPs in feathers are of external and/or internal origin. Although the present study documents that white-tailed eagle nestlings are exposed to PFRs, NBFRs and DPs, we did not investigate the possible correlations between these compounds in plasma, feathers and preen oil due to the < 50 % detection frequencies in some of these matrices.

4.2. Correlation between matrices

In general, the relatively low concentrations of PCBs, OCPs, BDE 47 and PFUnA quantified in plasma reflected the nestlings' recent exposure through diet (Henriksen et al., 1998) and

445 remains from maternal transfer to the eggs (Bourgeon et al., 2013). The concentrations of these
446 compounds quantified in the feathers were incorporated into the feathers some weeks prior to
447 the sampling, and the concentrations therefore reflected blood concentrations at that time
448 (García-Fernández et al., 2013; Jaspers et al., 2006). Feather concentrations of POPs and ECs
449 can also be affected by preening activity and external contamination from air and dust. The
450 preen oil is lipid rich and may function as a passive excretion route for lipophilic compounds
451 onto the feathers (Eulaers et al., 2011b; Jaspers et al., 2008). However, in nestlings this activity
452 is considered to be of minor influence on feather concentrations (Eulaers et al., 2011b; Jaspers
453 et al., 2011). Although the concentrations of the quantified POPs and ECs seem to be higher in
454 feathers than in blood (on a ww basis), the pattern may vary depending on the structure and
455 toxicokinetics of the compound, as we generally found higher concentrations of PFASs in
456 plasma than in feathers (Table 1).
457
458 The high detection frequencies of legacy POPs in plasma and feathers also resulted in
459 significant correlations between these matrices. The strong correlations of PCBs between
460 plasma and feathers are in accordance with previous studies on white-tailed eagle nestlings
461 (Eulaers et al., 2011a, 2011b). The correlations also correspond with results from earlier studies
462 on PCBs correlations in internal tissues and feathers (Dauwe et al., 2005; Van den Steen et al.,
463 2007). Nevertheless, when the two locations were analysed separately, no significant
464 correlations were detected for CB 101, 105 and 180 between plasma and feathers. This lack of
465 correlation corresponds to previous studies with small sample sizes (Eulaers et al., 2011a,
466 2011b), and may reflect temporal, spatial or biological variation. The differences between the
467 two locations, regarding the mentioned variables, will be further investigated in another study
468 (Løseth et al., in preparation).
469

Our significant correlations for POP concentrations in plasma and feathers are contrary to a study on adult black-legged kittiwakes (*Rissa tridactyla*) from Svalbard, where the authors investigated several PCBs, OCPs and PFRs in plasma and feathers (Svendsen et al., 2018). In that study, a significant positive correlation was only identified for CB 153 (Svendesen et al., 2018). The authors argued that the absence of correlations between plasma and feathers concentration may be linked to the migratory behaviour of the adult kittiwakes. The sampled primary feathers were grown when the birds were at their wintering areas, and plasma and feathers were collected during summer (Svendsen et al., 2018). In our study, the nestlings were sampled when they were stationary in their nests, and concentrations detected in their growing feathers are therefore more likely to correlate with plasma concentrations.

The high detection frequencies and concentrations of legacy POPs in the preen oil also resulted in significant correlations between plasma, feathers and preen oil. These significant correlations were in accordance with a previous study on white-tailed eagle nestlings (Eulaers et al., 2011b). This is the fourth study, to our knowledge, where plasma and preen oil concentrations of legacy POPs have been compared (Eulaers et al., 2011b; Van den Brink, 1997; Yamashita et al., 2007). Therefore, our study further adds to the evidence of preen oil as a suitable matrix for biomonitoring of legacy POPs as it strongly reflects internal concentrations.

Of the analysed ECs, PFUnA was the only compound which could be investigated for correlations between plasma and feathers. The two significant correlations detected between plasma and feathers for PFUnA in Steigen 2015 and 2016 contrasts with reports from a study on white-tailed eagle nestlings at the same location in 2014 (Gomez-Ramirez et al., 2017). Their study detected significant correlations between plasma and feathers for other PFASs, but

not for PFUnA (Gomez-Ramirez et al., 2017). It should, however, be noted that in the present study, no correlations were detected between plasma and feather concentrations of PFUnA in the Smøla population, in either 2015 or 2016. Also, no significant correlation was detected when our data were combined with data from Gomez-Ramirez et al. (2017). The variability of the plasma and feather correlation suggests that feathers may not be a suitable matrix for investigating internal concentrations of PFUnA (Gómez-Ramírez et al., 2017). The large variation observed between years and locations in detection frequencies of several PFASs in feathers also leads us to question the general suitability of feathers for monitoring internal PFASs concentrations. As there is little knowledge on the deposition of PFASs into feathers, we suggest prioritising the use of plasma samples to investigate internal PFASs concentrations in birds.

5. Conclusions

This is the first study to present a wide investigation of feathers and preen oil, in relation to plasma, for monitoring of both legacy and emerging compounds in white-tailed eagle nestlings from Norway. Our results propose both feathers and preen oil as suitable matrices for legacy POP analyses as the concentrations were significantly and positively correlated with plasma concentrations. This was also the first study to investigate non-destructive sampling methods from one species at different locations and years. Despite inter-annual and spatial variation of POPs, our large sample size allowed strong and robust statistical analyses providing further support for the strong and significant correlations between the three matrices for legacy POPs found in previous studies. For PFASs on the other hand, the inter-annual and spatial variation as well as the low detection frequencies in feathers compared to plasma resulted in poor and non-significant correlations between feathers and plasma. Because of the generally high detection frequencies of PFASs in plasma despite inter-annual and spatial variation, we suggest

prioritising the use of plasma for PFAS analyses. Correlations could not be investigated for PFRs, NBFRs and DPs due to low detection frequencies in plasma. The higher detection frequencies and concentrations of these emerging contaminants in feathers compared to plasma may suggest that feathers are prone to external contamination and/or that these compounds are rapidly metabolised and excreted. Further studies are needed to investigate if PFRs, NBFRs and DPs detected in feathers are from external or internal origin.

6. Acknowledgements

The authors acknowledge the Norwegian Research Council and NTNU for funding the project NewRaptor (project number 230465), Mari E. Løseth, Nathalie Briels and Veerle L.B. Jaspers. Giulia Poma and Govindan Malarvannan acknowledge the University of Antwerp for their post-doctoral fellowships. Trond V. Johnsen, Igor Eulaers, Paula Marcinekova, Courtney Waugh, Espen L. Dahl, Johannes Schrøder and Aasmund Gylseth are acknowledged for their assistance during sample collection. We thank Grethe S. Eggen and Lene N. Torgersen for assisting the chemical analyses. We also thank Pilar M. Gómez-Ramírez for providing additional data on PFUnA concentrations in white-tailed eagle nestlings from Steigen (2014). Additional funding for fieldwork in Steigen was provided by the Hazardous Substances Flagship (the Raptor project) at the Fram Centre in Tromsø.

Supplementary information:

Table S1: List of targeted organochlorinated compounds for analyses

Table S2: List of targeted flame retardant compounds for analyses

Table S3: Contents of internal standards

Table S4: List of targeted per- and polyfluoroalkyl substances for analyses

Table S5: Recoveries of internal standards in plasma

545 Table S6: Recoveries of internal standards in feathers and preen oil

546 Table S7-S13: Detection frequencies and descriptive statistics for all analysed compounds in

547 plasma, feathers and preen oil

548 Additional analytical details of POPs and PFR analyses

549

7. References

Bearr, J.S., Mitchelmore, C.L., Roberts, S.C., Stapleton, H.M., 2012. Species specific differences in the in vitro metabolism of the flame retardant mixture, Firemaster®BZ-54. Aquat. Toxicol. 124–125, 41–47. doi:10.1016/j.aquatox.2012.06.006

Becker, P.H., Sperveslage, H., 1989. Organochlorines and heavy metals in Herring Gull (*Larus argentatus*) eggs and chicks from the same clutch. Bull. Environ. Contam. Toxicol. 42, 721–727. doi:10.1007/BF01700394

BirdLife International. 2016. *Haliaeetus albicilla*. The IUCN Red List of Threatened Species 2016: e.T22695137A93491570. http://dx.doi.org/10.2305/IUCN.UK.2016-3.RLTS.T22695137A93491570.en. Downloaded on 01 June 2018.

Borgå, K., Fisk, A.T., Hoekstra, P.E., Muir, D.C.G., 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in Arctic marine food webs. Environ. Toxicol. Chem. 23, 2367–85. doi: 10.1897/03-518

Bourgeon, S., Leat, E.K.H, Furness, R.W., Borgå, K., Hanssen, S.A., Bustnes, J.O., 2013. Dietary versus maternal sources of organochlorines in top predator seabird chicks: An experimental approach. Environ. Sci. Technol. 47, 5963–5970. doi:10.1021/es400442q

Briels, N., Løseth, M.E., Ciesielski, T.M., Malarvannan, G., Poma, G., Kjærvik, S.A., Léon, A., Cariou, R., Covaci, A., Jaspers, V.L.B., 2018. In ovo transformation of two emerging flame retardants in Japanese quail (*Coturnix japonica*). Ecotoxicol. Environ. Saf. 149. doi:10.1016/j.ecoenv.2017.10.069

Buccini, J., 2003. The development of a global treaty on persistent organic pollutants (POPs), Persistent Organic Pollutants, in: Fiedler H. (Ed.) Persistent Organic Pollutants. The Handbook of Environmental Chemistry (Vol. 3 Series: Anthropogenic Compounds), vol 3O. Springer, Berlin, Heidelberg, pp.13–30.

575 Burger, J., 1993. Metals in Avian Feathers: Bioindicators of Environmental Pollution, in:
576 Hodgson, E. (Ed.), Reviews in Environmental Toxicology, Vol. 5. Toxicology
577 Communications, INC. Raleigh, North Carolina, USA, pp. 203–311.
578 Burger, J., Gochfeld, M., 2004. Marine Birds as Sentinels of Environmental Pollution.
579 Ecohealth 1, 263–274. doi:10.1007/s10393-004-0096-4Bustnes, J.O., Bårdsen, B.J.,
580 Herzke, D., Johnsen, T. V, Eulaers, I., Ballesteros, M., Hanssen, S. a, Covaci, A.,
581 Jaspers, V.L.B., Eens, M., Sonne, C., Halley, D., Moum, T., Nøst, T.H., Erikstad, K.E.,
582 Ims, R.A., 2013. Plasma concentrations of organohalogenated pollutants in predatory
583 bird nestlings: associations to growth rate and dietary tracers. Environ. Toxicol. Chem.
584 32, 2520–7. doi:10.1002/etc.2329
585 Cequier, E., Ionas, A.C., Covaci, A., Marcé, R.M., Becher, G., Thomsen, C., 2014.
586 Occurrence of a broad range of legacy and emerging flame retardants in indoor
587 environments in Norway. Environ. Sci. Technol. 48, 6827–6835.
588 doi:10.1021/es500516u
589 Covaci, A., Harrad, S., Abdallah, M.A.E., Ali, N., Law, R.J., Herzke, D., de Wit, C.A., 2011.
590 Novel brominated flame retardants: A review of their analysis, environmental fate and
591 behaviour. Environ. Int. 37, 532–556. doi:10.1016/j.envint.2010.11.007
592 Dauwe, T., Jaspers, V.L.B., Covaci, A., Schepens, P., Eens, M., Auwe, T.O.M.D., Aspers,
593 V.E.J., Ovaci, A.D.C., Chepens, P.A.U.L.S., Ens, M.A.E., 2005. Feathers as a
594 nondestructive biomonitor for persistent organic pollutants. Environ. Toxicol. Chem. 24,
595 442–9. doi:10.1897/03-596.1
596 Espín, S., García-Fernández, A.J., Herzke, D., Shore, R.F., van Hattum, B., Martínez-López,
597 E., Coeurdassier, M., Eulaers, I., Fritsch, C., Gómez-Ramírez, P., Jaspers, V.L.B.,
598 Krone, O., Duke, G., Helander, B., Mateo, R., Movalli, P., Sonne, C., van den Brink,
599 N.W., 2016. Tracking pan-continental trends in environmental contamination using

sentinel raptors—what types of samples should we use? Ecotoxicology 25. doi:10.1007/s10646-016-1636-8

Eulaers, I., Covaci, A., Herzke, D., Eens, M., Sonne, C., Moum, T., Schnug, L., Hanssen, S.A., Johnsen, T.V., Bustnes, J.O., Jaspers, V.L.B., 2011a. A first evaluation of the usefulness of feathers of nestling predatory birds for non-destructive biomonitoring of persistent organic pollutants. Environ. Int. 37, 622–30. doi:10.1016/j.envint.2010.12.007

Eulaers, I., Covaci, A., Hofman, J., Nygård, T., Halley, D.J., Pinxten, R., Eens, M., Jaspers, V.L.B., 2011b. A comparison of non-destructive sampling strategies to assess the exposure of white-tailed eagle nestlings (*Haliaeetus albicilla*) to persistent organic pollutants. Sci. Total Environ. 410–411, 258–265. doi:10.1016/j.scitotenv.2011.09.070

Eulaers, I., Jaspers, V.L.B., Bustnes, J.O., Covaci, A., Johnsen, T. V., Halley, D.J., Moum, T., Ims, R.A., Hanssen, S.A., Erikstad, K.E., Herzke, D., Sonne, C., Ballesteros, M., Pinxten, R., Eens, M., 2013. Ecological and spatial factors drive intra- and interspecific variation in exposure of subarctic predatory bird nestlings to persistent organic pollutants. Environ. Int. 57–58, 25–33. doi:10.1016/j.envint.2013.03.009

Eulaers, I., Jaspers, V.L.B., Halley, D.J., Lepoint, G., Nygård, T., Pinxten, R., Covaci, A., Eens, M., 2014. Brominated and phosphorus flame retardants in White-tailed Eagle *Haliaeetus albicilla* nestlings: bioaccumulation and associations with dietary proxies ($\delta^{13}C$, $\delta^{1}5N$ and $\delta^{34}S$). Sci. Total Environ. 478, 48–57. doi:10.1016/j.scitotenv.2014.01.051

Fenstad, A.A., Jenssen, B.M., Moe, B., Hanssen, S.A., Bingham, C., Herzke, D., Bustnes, J.O., Krøkje, Å., 2014. DNA double-strand breaks in relation to persistent organic pollutants in a fasting seabird. Ecotoxicol. Environ. Saf. 106, 68–75. doi:10.1016/j.ecoenv.2014.04.020

Feo, M.L., Barón, E., Eljarrat, E., Barceló, D., 2012. Dechlorane Plus and related compounds

in aquatic and terrestrial biota: a review. Anal. Bioanal. Chem. 404, 2625–37. doi:10.1007/s00216-012-6161-x

Furness, R.W., 1993. Birds as monitors of pollution, in: Furness, R.W., Greenwood, J.J.D. (Eds.), Birds as Monitors of Environmental Change. Springer, Dordrecht, pp. 86–143.

García-Fernández, A.J., Espín, S., Martínez-López., E., 2013. Feathers as a biomonitoring tool of polyhalogenated compounds: A review. Environ. Sci. Technol. 47, 3028–3043. doi:10.1021/es302758x

Gómez-Ramírez, P., Bustnes, J.O., Eulaers, I., Herzke, D., Johnsen, T.V., Lepoint, G., Pérez-García, J.M., García-Fernández, A.J., Jaspers, V.L.B., 2017. Per- and polyfluoroalkyl substances in plasma and feathers of nestling birds of prey from northern Norway. Environ. Res. 158, 277–285. doi:10.1016/j.envres.2017.06.019

Gómez-Ramírez, P., Shore, R.F., van den Brink, N.W., van Hattum, B., Bustnes, J.O., Duke, G., Fritsch, C., García-Fernández, A. J., Helander, B.O., Jaspers, V., Krone, O., Martínez-López, E., Mateo, R., Movalli, P., Sonne, C., 2014. An overview of existing raptor contaminant monitoring activities in Europe. Environ. Int. 67, 12–21. doi:10.1016/j.envint.2014.02.004

Green, N., Schlabach, M., Bakke, T., Brevik, E.M., Dye, C., Herzke, D., Huber, S., Plosz, B., Remberger, Mikael Schøyen, M., Uggerud, H.T., Vogelsang, C., 2008. Phosphorus flame retardents, polyfluorinated organic compounds, nitro-PAHs, silver, platinum and sucralose in air, wastewater treatment facilities, and recipients, Screening of selected metals and new organic contaminants 2007 NIVA report no. 5569–2008.

Hanssen, L., Dudarev, A. a, Huber, S., Odland, J.Ø., Nieboer, E., Sandanger, T.M., 2013. Partition of perfluoroalkyl substances (PFASs) in whole blood and plasma, assessed in maternal and umbilical cord samples from inhabitants of arctic Russia and Uzbekistan. Sci. Total Environ. 447, 430–7. doi:10.1016/j.scitotenv.2013.01.029

650 Henriksen, E.O., Gabrielsen, G.W., Skaare, J.U., 1998. Validation of the use of blood
651 samples to assess tissue concentrations of organochlorines in glaucous gulls, *Larus*
652 *hyperboreus*. Chemosphere 37, 2627–2643. doi:10.1016/S0045-6535(98)00162-3
653 Herzke, D., Nygård, T., Berger, U., Huber, S., Røv, N., 2009. Perfluorinated and other
654 persistent halogenated organic compounds in European shag (*Phalacrocorax aristotelis*)
655 and common eider (*Somateria mollissima*) from Norway: a suburban to remote pollutant
656 gradient. Sci. Total Environ. 408, 340–8. doi:10.1016/j.scitotenv.2009.08.048
657 Hou, R., Xu, Y., Wang, Z., 2016. Review of OPFRs in animals and humans: Absorption,
658 bioaccumulation, metabolism, and internal exposure research. Chemosphere 153, 78–90.
659 doi:10.1016/j.chemosphere.2016.03.003
660 Jaspers, V.L.B., Voorspoels, S., Covaci, A., Eens, M., 2006. Can predatory bird feathers be
661 used as a non-destructive biomonitoring tool of organic pollutants? Biol. Lett. 2, 283–
662 285. doi:10.1098/rsbl.2006.0450
663 Jaspers, V.L.B., Covaci, A., Van den Steen, E., Eens, M., 2007a. Is external contamination
664 with organic pollutants important for concentrations measured in bird feathers? Environ.
665 Int. 33, 766–772. doi:10.1016/j.envint.2007.02.013
666 Jaspers, V.L.B., Voorspoels, S., Covaci, A., Lepoint, G., Eens, M., 2007b. Evaluation of the
667 usefulness of bird feathers as a non-destructive biomonitoring tool for organic
668 pollutants: A comparative and meta-analytical approach. Environ. Int. 33, 328–337.
669 doi:10.1016/j.envint.2006.11.011
670 Jaspers, V.L.B., Covaci, A., Deleu, P., Neels, H., Eens, M., 2008. Preen oil as the main
671 source of external contamination with organic pollutants onto feathers of the common
672 magpie (*Pica pica*). Environ. Int. 34, 741–748. doi:10.1016/j.envint.2007.12.002
673 Jaspers, V.L.B., Rodriguez, F.S., Boertmann, D., Sonne, C., Dietz, R., Rasmussen, L.M.,
674 Eens, M., Covaci, A., 2011. Body feathers as a potential new biomonitoring tool in

675 raptors: a study on organohalogenated contaminants in different feather types and preen
676 oil of West Greenland white-tailed eagles (Haliaeetus albicilla). Environ. Int. 37, 1349–
677 56. doi:10.1016/j.envint.2011.06.004
678 Jaspers, V.L.B., Herzke, D., Eulaers, I., Gillespie, B.W., Eens, M., 2013. Perfluoroalkyl
679 substances in soft tissues and tail feathers of Belgian barn owls (*Tyto alba*) using
680 statistical methods for left-censored data to handle non-detects. Environ. Int. 52, 9–16.
681 doi:10.1016/j.envint.2012.11.002
682 Jones, K.C.K.C., de Voogt, P., 1999. Persistent organic pollutants (POPs): state of the
683 science. Environ. Pollut. 100, 209–221. doi:10.1016/S0269-7491(99)00098-6
684 Lau, C., Anitole, K., Hodes, C., Lai, D., Pfahles-Hutchens, A., Seed, J., 2007. Perfluoroalkyl
685 acids: a review of monitoring and toxicological findings. Toxicol. Sci. 99, 366–94.
686 doi:10.1093/toxsci/kfm128
687 Letcher, R.J., Bustnes, J.O., Dietz, R., Jenssen, B.M., Jørgensen, E.H., Sonne, C., Verreault,
688 J., Vijayan, M.M., Gabrielsen, G.W., 2010. Exposure and effects assessment of
689 persistent organohalogen contaminants in arctic wildlife and fish. Sci. Total Environ.
690 408, 2995–3043. doi:10.1016/j.scitotenv.2009.10.038
691 Li, J., Guo, F., Wang, Y., Zhang, J., Zhong, Y., Zhao, Y., Wu, Y., 2013. Can nail, hair and
692 urine be used for biomonitoring of human exposure to perfluorooctane sulfonate and
693 perfluorooctanoic acid? Environ. Int. 53, 47–52. doi:10.1016/j.envint.2012.12.002
694 Li, Y., Gao, K., Duo, B., Zhang, G., Cong, Z., Gao, Y., Fu, J., Zhang, A., Jiang, G., 2017.
695 Analysis of a broad range of perfluoroalkyl acids in accipiter feathers: method
696 optimization and their occurrence in Nam Co Basin, Tibetan Plateau. Environ.
697 Geochem. Health 1–10. doi:10.1007/s10653-017-9948-z
698 Mackay, D., Shiu, W.Y., Ma, K., Lee, S.C., 2006. Halogenated Hydrocarbons, in: Handbook
699 of Physical-Chemical Properties and Environmental Fate for Organic Chemicals:

Second Edition. CRC Press, Taylor & Francis Group, III, 921–2258. ISBN: 9781566706872

Meyer, J., Jaspers, V.L.B., Eens, M., de Coen, W., 2009. The relationship between perfluorinated chemical levels in the feathers and livers of birds from different trophic levels. Sci. Total Environ. 407, 5894–5900. doi:10.1016/j.scitotenv.2009.07.032

Möller, A., Xie, Z., Caba, A., Sturm, R., Ebinghaus, R., 2011. Organophosphorus flame retardants and plasticizers in the atmosphere of the North Sea. Environ. Pollut. 159, 3660–3665. doi:10.1016/j.envpol.2011.07.022

Olsson, A., Ceder, K., Bergman, Å., Helander, B., 2000. Nestling blood of the white-tailed sea eagle (*Haliaeetus albicilla*) as an indicator of territorial exposure to organohalogen compounds - An evaluation. Environ. Sci. Technol. 34, 2733–2740. doi:10.1021/es991426k

Poma, G., Cuykx, M., Amato, E., Calaprice, C., Focant, J.F., Covaci, A., 2017. Evaluation of hazardous chemicals in edible insects and insect-based food intended for human consumption. Food Chem. Toxicol. 100, 70–79. doi:10.1016/j.fct.2016.12.006

Svendsen, N.B., Herzke, D., Harju, M., Bech, C., Gabrielsen, G.W., Jaspers, V.L.B., 2018. Persistent organic pollutants and organophosphate esters in feathers and blood plasma of adult kittiwakes (*Rissa tridactyla*) from Svalbard – associations with body condition and thyroid hormones. Environ. Res. 164, 158–164. doi:10.1016/j.envres.2018.02.012

Sverko, E., Tomy, G.T., Reiner, E.J., Li, Y.-F., McCarry, B.E., Arnot, J. a, Law, R.J., Hites, R. a, 2011. Dechlorane plus and related compounds in the environment: a review. Environ. Sci. Technol. 45, 5088–98. doi:10.1021/es2003028

Tollbäck, J., Tamburro, D., Crescenzi, C., Carlsson, H., 2006. Air sampling with Empore solid phase extraction membranes and online single-channel desorption/liquid chromathography/mass spectrometry analysis: Determination of volatile and semi-

volatile organophosphate esters. Journ. Chroma. A. 1129, 1-8. doi:10.1016/j.chroma.2006.05.086

UNEP (United Nations Environmental Programme), 2009. The Stockholm Convention on Persistent Organic Pollutants. United Nations, adoption of amendments to Annexes A, B and C. http://chm.pops.int/Convention/ConventionText/tabid/2232/Default.aspx

Van den Brink, N.W., 1997. Preen gland oil and blood samples: Non- destructive methods for monitoring organochlorine levels in Antartic top predators, in: Battaglia, B., Valencia, J., Walton, D.W.H. (Eds.), Antarctic Communities, Species Structure and Survival. Cambridge University, Cambridge, p. 413−416.

Van den Steen, E., Covaci, A., Jaspers, V.L.B., Dauwe, T., Voorspoels, S., Eens, M., Pinxten, R., 2007. Experimental evaluation of the usefulness of feathers as a non-destructive biomonitor for polychlorinated biphenyls (PCBs) using silastic implants as a novel method of exposure. Environ. Int. 33, 257–264. doi:10.1016/j.envint.2006.09.018

van der Veen, I., de Boer, J., 2012. Phosphorus flame retardants: properties, production, environmental occurrence, toxicity and analysis. Chemosphere 88, 1119–53. doi:10.1016/j.chemosphere.2012.03.067

Voorspoels, S., Covaci, A., Maervoet, J., Schepens, P., 2002. Relationship between age and levels of organochlorine contaminants in human serum of a Belgian population. Bull. Environ. Contam. Toxicol. 69, 22–29. doi:10.1007/s00128-002-0004-y

Yamashita, R., Takada, H., Murakami, M., Fukuwaka, M.A., Watanuki, Y., 2007. Evaluation of noninvasive approach for monitoring PCB pollution of seabirds using preen gland oil. Environ. Sci. Technol. 41, 4901–4906. doi:10.1021/es0701863

White-tailed eagle (*Haliaeetus albicilla*) feathers from Norway are suitable for monitoring of legacy, but not emerging contaminants

Mari E. Løseth[a*], Nathalie Briels[a], Jørgen Flo[a], Govindan Malarvannan[b], Giulia Poma[b], Adrian Covaci[b], Dorte Herzke[c], Torgeir Nygård[d], Jan O. Bustnes[e], Bjørn M. Jenssen[a] and Veerle L. B. Jaspers[a]

Affiliations:

[a]Department of Biology, Norwegian University of Science and Technology (NTNU), Høgskoleringen 5, 7491 Trondheim, Norway

[b]Toxicological Center, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium

[c]Norwegian Institute for Air Research (NILU), FRAM- High North Research Centre on Climate and the Environment, 9007 Tromsø, Norway

[d]Norwegian Institute for Nature Research (NINA), Høgskoleringen 9, 7034 Trondheim, Norway

[e]Norwegian Institute for Nature Research (NINA), FRAM- High North Research Centre on Climate and the Environment, 9007 Tromsø, Norway

***Corresponding author:**

Mari Engvig Løseth: mari.loseth@ntnu.no

Number of pages: 16

Number of tables: 11

Contents:

Table S1: List of targeted organochlorinated compounds for analyses
Table S2: List of targeted flame retardant compounds for analyses
Table S3: Contents of internal standards
Table S4: List of targeted per- and polyfluoroalkyl substances for analyses
Table S5: Recoveries of internal standards in plasma
Table S6: Recoveries of internal standards in feathers and preen oil
Table S7-S13: Detection frequencies and descriptive statistics for all analysed compounds in plasma, feathers and preen oil

Additional analytical details of POPs and PFR analyses

Table S3: Targeted chlorinated compounds analysed in plasma, feather and preen oil samples from white-tailed eagle nestlings sampled at Steigen and Smøla (Norway) in 2015 and 2016. Only samples from 2016 were analyzed for *p,p'*-DDD. PCB congeners are numbered by the IUPAC system (International Union of Pure and Applied Chemistry). Limit of quantification (LOQ) for the compounds are the same for 2015 and 2016 and are presented as ng/mL for plasma and ng/g for feathers and preen oil.

Organochlorinated compounds

Group	**Abbreviations**	**Compounds**	LOQ plasma	LOQ feathers	LOQ preen oil
Organo-	OxC	*oxy*-chlordane	0.01	0.10	1.0
Chlorinated	TN	*trans*-nonachlor	0.01	0.10	1.0
Pesticides	CN	*cis*-nonachlor	0.01	0.10	1.0
(OCPs)	HCB	hexachlorobenzene	0.01	0.10	1.0
	a-HCH	1α,2α,3β,4α,5β,6β-hexachlorocyclohexane	0.01	0.10	1.0
	b-HCH	1α,2β,3α,4β,5α,6β-hexachlorocyclohexane	0.01	0.10	1.0
	g-HCH	1α,2α,3β,4α,5α,6β-hexachlorocyclohexane	0.02	0.10	1.0
	p,p',-DDT	*p,p'*,-dichloro-α,α-diphenyl-β,β,β-trichloroethane	0.02	0.20	2.0
	p,p',-DDE	*p,p'*,-dichloro-diphenyl-dichloroethylene	0.02	0.20	2.0
	p,p',-DDD	*p,p'*,-dichloro-diphenyl-dichloroethane	0.02	0.20	2.0
Polychlorinated	CB 28	2,4,4'-trichlorobiphenyl	0.05	0.30	2.0
Biphenyls	CB 49	2,2',4,5'-tetrachlorobiphenyl	0.05	0.30	2.0
(PCBs)	CB 52	2,2',5,5'-tetrachlorobiphenyl	0.05	0.30	2.0
	CB 74	2,4,4',5-tetrachlorobiphenyl	0.05	0.30	2.0
	CB 95	2,2',3,5',6-pentachlorobiphenyl	0.02	0.20	1.0
	CB 99	2,2',4,4',5-pentachlorobiphenyl	0.02	0.20	1.0
	CB 101	2,2',4,5,5'-pentachlorobiphenyl	0.02	0.20	1.0
	CB 105	2,3,3',4,4'-pentachlorobiphenyl	0.01	0.10	1.0
	CB 110	2,3,3',4',6-pentachlorobiphenyl	0.01	0.10	1.0
	CB 118	2,3',4,4',5-pentachlorobiphenyl	0.01	0.10	1.0
	CB 138	2,2',3,4,4',5'-hexachlorobiphenyl	0.01	0.10	1.0
	CB 149	2,2',3,4',5',6-hexachlorobiphenyl	0.01	0.10	1.0
	CB 153	2,2',4,4',5,5'-hexachlorobiphenyl	0.01	0.10	1.0
	CB 156	2,3,3',4,4',5-hexachlorobiphenyl	0.01	0.10	1.0
	CB 170	2,2',3,3',4,4',5-heptachlorobiphenyl	0.01	0.10	1.0
	CB 171	2,2',3,3',4,4',6-heptachlorobiphenyl	0.01	0.10	1.0
	CB 177	2,2',3,3',4,5',6'-heptachlorobiphenyl	0.01	0.10	1.0
	CB 180	2,2',3,4,4',5,5'-heptachlorobiphenyl	0.01	0.10	1.0
	CB 183	2,2',3,4,4',5',6-heptachlorobiphenyl	0.01	0.10	1.0
	CB 187	2,2',3,4',5,5',6-heptachlorobiphenyl	0.01	0.10	1.0
	CB 194	2,2',3,3',4,4',5,5'-octachlorobiphenyl	0.01	0.10	1.0
	CB 206	2,2',3,3',4,4',5,5',6-nonachlorobiphenyl	0.01	0.10	1.0
	CB 209	Decachlorobiphenyl	0.01	0.10	1.0

Table S4: Targeted flame retardants analysed in plasma, feather and preen oil samples from white-tailed eagle nestlings, sampled at Steigen and Smøla (Norway) in 2015 and 2016. PBDE congeners are numbered by the IUPAC system (International Union of Pure and Applied Chemistry). Only samples from 2016 were analysed for 2'-MeO-BDE 68 and 6'-MeO-BDE 47. Limit of quantification (LOQ) for the compounds are the same for 2015 and 2016 and are presented as ng/mL for plasma and ng/g for feathers and preen oil. Compounds not targeted (analysed) are marked with "n.a".

Flame retardants					
Group	**Abbreviations**	**Compounds**	LOQ plasma	LOQ feathers	LOQ preen oil
Polybrominated diphenyl ethers (PBDEs)	BDE 28	2',4,4'-tribromodiphenyl ether	0.002	0.10	0.4
	BDE 47	2,2',4,4'-tetrabromodiphenyl ether	0.002	0.10	0.4
	BDE 99	2,2',4,4',5'-pentabromodiphenyl ether	0.002	0.10	0.4
	BDE 100	2,2',4,4',6'-pentabromodiphenyl ether	0.002	0.10	0.4
	BDE 153	2,2',4,4',5,5'-hexabromobiphenyl ether	0.002	0.10	0.8
	BDE 154	2,2',4,4',5,6'-hexabromobiphenyl ether	0.004	0.10	0.8
	BDE 183	2,2',3',4,4',5',6'-heptabromodiphenyl ether	0.004	0.20	0.8
2016	2'-MeO-BDE 68	1,5-Dibromo-3-(2,4-dibromophenoxy)-2-methoxybenzene			n.a
2016	6'-MeO-BDE 47	1,5-Dibromo-2-(2,4-dibromophenoxy)-3-methoxybenzene			n.a
Dechlorane plus isomers (DPs)	Syn-DP	Syn-Dechlorane plus	0.002	0.10	0.4
	Anti-DP	Anti-Dechlorane plus	0.002	0.10	0.4
Novel brominated flame retardants (NBFRs)	TBB	2-ethylhexyl-2,3,4,5-tetrabromobenzoate	0.010	0.20	1.6
	TBPH	bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate	0.020	0.40	2.0
	BTBPE	1,2 Bis(2,4,6-tribromophenoxy)ethane	0.005	0.10	0.4
Phosphate flame retardants (PFRs)	TCEP	tris(chloroethyl) phosphate	0.100	1.00	n.a
	TBOEP	tris(2-butoxyethyl) phosphate	0.400	4.00	n.a
	EHDPHP	2-ethylhexyl diphenyl phosphate	0.100	1.00	n.a
	TPhP	triphenyl phosphate	0.100	1.00	n.a
	TCPP	tris(2-chloroisopropyl) phosphate	0.100	1.00	n.a
	TCIPP	tris(1-chloro-2-propyl) phosphate	0.100	1.00	n.a
	TDCIPP	tris(1,3-dichloro-2-propyl)phosphate	0.200	1.00	n.a

798 **Table S5:** Compounds and their concentrations in internal standards used for extraction of targeted PCBs, PBDEs,
799 OCPs, NBFRs, PFRs, DPs and PFASs.

Internal standard	Concentrations and compounds
IS1 (POPs)	200 pg/µL PCB 143
	25 pg/µL BDE 77
	25 pg/µL ε-hexachlorocyclohexane (ε-HCH)
IS2 (ECs)	200 pg/µL ^{13}C-bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (TBPH)
	50 pg/µL ^{13}C-syn-dechlorane plus (DP)
	50 pg/µL ^{13}C-anti-DP
	1 ng/µL triphenyl phosphate (TPHP-d15)
	1 ng/µL tris(chloroethyl) phosphate (TCEP-d12)
	1 ng/µL tris-(1,3-dichloro-2-propyl) phosphate (TDCIPP-d15)
	1 ng/µL triamyl phosphate (TAP)
	2 ng/µL tri-(2-butoxyethyl) phosphate (TBOEP-d6)
IS3 (DPs)	200 pg/µL ^{13}C-DPs
IS4 (PFASs)	0.1 ng/µL ^{13}C-PFAS mix

800

801

802 **Table S6:** Targeted compounds for per-and polyfluorinated substance analysed in plasma and feather samples
803 from white-tailed eagle nestlings, sampled at Steigen and Smøla (Norway) in 2015 and 2016. Limit of
804 quantification (LOQ) is given as ng/mL for plasma and ng/g for feathers.

Per- and polyfluorinated substances (PFASs)						
	Abbreviations	**Compounds**	LOQ plasma 2015	LOQ feathers 2015	LOQ plasma 2016	LOQ feathers 2016
Carboxylic acids	PFBA	Perfluorobutanoic acid	169.11	0.498	0.05	
	PFPeA	Perfluoropentanoic acid	1.31	0.200	0.05	
	PFHxA	Perfluorohexanoic acid	1.31	0.002	0.10	0.193
	PFHpA	Perfluoroheptanoic acid	1.31	0.002	0.05	0.267
	PFOA	Perfluorooctanoic acid	0.10	0.029	0.05	0.210
	PFNA	Perfluorononanoic acid	0.10	0.029	0.08	0.259
	PFDcA	Perfluorodecanoanoic acid	0.10	0.029	0.05	0.262
	PFUnA	Perfluoroundecanoic acid	0.20	0.029	0.08	0.181
	PFDoA	Perfluorododecanoic acid	0.20	0.029	0.08	0.145
	PFTrA	Perfluorotridecanoic acid	0.20	0.029	0.10	0.186
	PFTeA	Perfluorotetradecanoic acid	0.20	0.029	0.10	0.200
Sulfonamides	PFOSA	Perfluorooctanesulfonamide	23.35	0.029	0.10	0.200
Sulfonic acids	PFBS	Perfluorobutane sulfonate	169.11	0.498	0.05	0.100
	PFPS	Perfluoropentane sulfonate		0.002		0.100
	PFHxS	Perfluorohexane sulfonate	0.10	0.002	0.05	0.393
	PFHpS	Perfluoroheptane sulfonate	0.01	0.002	0.08	0.127
	Lin-PFOS	Linear perfluorooctane sulfonate	0.20	0.029	0.10	0.050
	Br-PFOS	Branched perfluorooctane sulfonate	0.20	0.029	0.10	0.050
	PFNS	Perfluorononane sulfonate		0.030	0.10	0.050

805

806

Additional analytical details

Clean-up of sample extracts for PCB, PBDE, OCP, NBFR, DP and PFR analyses

Further procedures for clean-up, fractionation of the concentrated extracts and quantification were the same for plasma and feathers. Fractionation was performed on Supelclean™ ENVI™18 Florisil cartridges (500 mg, 3mL, Supelco® Analytical). Anhydrous sodium sulfate (Na_2SO_4) was added to the cartridge before cleaning with 6 mL ethyl acetate, followed by 6 mL *n*-hexane. The sample extract was transferred to the cartridge. The sample tube was washed twice with 0.5 mL of *n*-hexane and vortexed. The extracts were eluted in two fractions: the first fraction (F1), containing PCBs, PBDEs, DPs, NBFRs, and OCPs was eluted with 10 mL *n*-hexane:DCM (1:1, v/v). A second fraction (F2) was collected, only for plasma and feather extracts, in new tubes, containing the PFRs. F2 was eluted from the same columns as F1 by 10 mL ethyl acetate. Both fractions were evaporated to near dryness by a gentle nitrogen steam. F1 was re-solubilised in 0.5 mL of *n*-hexane, followed by a second clean-up of F1 on acidified silica (5 %) in a 3 mL cartridge, pre-cleaned with 6 mL *n*-hexane. The tube was washed twice with 0.5 mL of *n*-hexane and vortexed.

The preen oil samples were only cleaned up on 6 mL columns, containing about 2.5 g of 44 % acidified silica. The cartridges were pre-cleaned with 6 mL *n*-hexane. Samples were added, and the tubes were washed twice with 1 mL of *n*-hexane and transferred to the cartridge. Preen oil samples were eluted with 10 mL of hexane:DCM (1:1, v/v).

All extracts were finally concentrated to near dryness under a gentle nitrogen stream and re-solubilized in 100 µL of iso octane. For each batch of 24 samples, 100 µL recovery standard (RS, CB 207, 50 pg/µL in iso-octane toluene 9:1, v/v) was added to five samples and vortexed for 30 s. Extracts were transferred to injection vials for gas chromatography with electron capture negative ionization and mass spectrometry (GC-ECNI/MS) analysis.

Instrumental analysis of PCBs, PBDEs, OCPs, NBFRs, DPs and PFRs

The analysis was performed with an Agilent 6890 GC (Palo Alto, CA, USA) coupled to an Agilent 5973 MS operated in electron capture negative ionization (ECNI) mode and equipped with a DB 5ms capillary column (30 m x 0.25 mm x 0.25 mm). The GC system was equipped with electronic pressure control and a programmable temperature vaporizer (PTV) inlet. The injection temperature was set at 92 °C, held 0.03 min, ramped at 700 °C/min to 300 °C, held 30 min. Injection (1 µL) was performed under a pressure of 10.06 psi until 1.25 min and purge flow to split vent of 50 mL/min after 1.25 min. The GC temperature ramp started from 92 °C,

held 1.25 min, ramped at 10 °C/min to 300 °C, held 1 min, ramped at 40 °C/min to 310 °C and held 9.5 min. Helium was used as a carrier gas with a flow rate of 1.0 mL/min until 25 min, then increased to 1.5 mL/min. The ion source and quadrupole temperatures were set at 170 °C and 150 °C, respectively. The mass spectrometer was operated in selected ion monitoring (SIM) for the quantification of BDE 28, 47, 100, 99, 154, 153, 183, BTBPE, *s*-DP, *a*-DP, CB 101, 99, 118, 153, 138, 187, 183, 180, 170, oxychlordane (OxC), trans nonachlor (TN), HCB, *p,p'*-DDE, *p,p'*-DDT, α-HCH, β-HCH, γ-HCH. BDE 103 and BDE 128 were used as IS for all PBDE congeners and BTBPE; 13C *s*-DP and 13C *a*-DP, were used as IS for *s*-DP, and *a*-DP, respectively; CB 143 was used as IS for the targeted PCBs and OCPs.

PFRs were analysed using an Agilent 6890 GC coupled to an Agilent 5973 MS operated in EI mode. The GC system was equipped with an HT 8 column (25 m x 0.22 mm x 0.25 mm), electronic pressure control and a PTV inlet. The injection temperature was set at 80 °C, held 0.03 min, ramped at 700 °C/min to 300 °C, held 40 min. Injection (1 uL) was performed under a pressure of 13.65 psi until 1.25 min and purge flow to split vent of 50 mL/min after 1.25 min. The GC temperature ramp started from 80 °C, held 1.25 min, ramped at 15 °C/min to 200 °C, held 3 min, ramped at 5 °C/min to 270 °C, ramped at 20 °C/min to 310 °C and held 12 min. Helium was used as a carrier gas with a flow rate of 1.0 mL/min until 28 min, then increased to 1.5 mL/min. The mass spectrometer was run in SIM mode and TEHP, TCEP, TCIPP (2 isomers), EHDPHP, TPHP, TDCIPP, TNBP and TBOEP were analysed. TAP was used as IS for TEHP, TNBP; TCEP-d12 was used for TCEP and TCIPP (2 isomers); TBOEP-d6 was used for TBOEP; TPHP-d15 was used for TPHP and EHDPHP; TDCIPP-d15 was used for TDCIPP.

Table S7: Recoveries of internal standards in plasma samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na".

	2015		2016	
Plasma	Mean	sd	Mean	sd
CB 143	69	39	108	8
e-HCH	53	12	79	11
BDE 77	83	9	87	10
^{13}C-HCB	na	na	102	17
^{13}C-TBPH	18	14	25	12
^{13}C-s-DP	47	13	55	10
^{13}C-a-DP	50	10	52	11
TAP	93	13	79	14
TCEP-d12	89	33	72	22
TBEP-d6	78	35	68	28
TPhP-d15	86	15	80	12
TDCPP-d15	92	15	80	11
^{13}C-PFPA	101	16	87	12
^{13}C-PFHxA	93	22	86	10
^{13}C-PFHpA	90	16	93	12
^{13}C-PFOA	88	15	87	12
^{13}C-PFDcA	81	32	89	11
^{13}C-PFUnA	77	13	84	12
^{13}C-PFDoA	59	15	83	19
^{13}C-PFHxS	90	17	87	12
^{13}C-PFOS	79	17	84	12
^{13}C-PFOSA	83	17	75	9

868 **Table S8:** Recoveries of internal standards in feathers and preen oil samples from white-tailed eagles from Smøla
869 and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na".

	2015		2016	
Feathers	Mean	sd	Mean	sd
CB 143	79	10	92	5
e-HCH	75	10	82	4
BDE 77	91	9	86	10
^{13}C-HCB	na	na	106	6
^{13}C-TBPH	45	13	40	14
^{13}C-s-DP	69	9	75	8
^{13}C-a-DP	68	9	72	9
TAP	87	11	92	10
TCEP-d12	105	19	93	15
TBEP-d6	97	15	88	18
TPhP-d15	74	12	81	10
TDCPP-d15	82	13	85	11
^{13}C-PFPA	43	5	101	22
^{13}C-PFHxA	42	5	101	20
^{13}C-PFHpA	40	4	118	29
^{13}C-PFOA	37	15	114	22
^{13}C-PFDcA	44	3	121	27
^{13}C-PFUnA	43	7	121	29
^{13}C-PFDoA	46	12	129	36
^{13}C-PFHxS	39	3	150	55
^{13}C-PFOS	38	6	89	16
^{13}C-PFOSA	43	4	90	21
Preen oil	Mean	sd	Mean	sd
CB 143	93	9	84	13
e-HCH	88	14	76	10
BDE 77	120	9	104	11
^{13}C-HCB	na	na	109	13

870

Table S9: Detection frequency, median, min and max concentrations of PCBs quantified in plasma samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Plasma concentrations are in ng/ml ww. Congeners not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
Plasma	2015 *n* = 13				2016 *n* = 22				2015 *n* = 14				2016 *n* = 21			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
CB 52	na				54	0.11	0.07	0.23	na				57	0.1	0.06	0.12
CB 74	na				9	0.18	0.12	0.25	na				24	0.15	0.06	0.25
CB 99	100	0.16	0.08	0.59	100	0.18	0.06	1.47	100	0.5	0.18	4.61	100	0.23	0.06	0.98
CB 101	100	0.21	0.09	0.31	100	0.14	0.01	0.56	100	0.16	0.07	0.56	86	0.14	0.02	0.25
CB 105	100	0.08	0.04	0.3	100	0.11	0.04	0.79	100	0.26	0.1	2.59	100	0.14	0.04	0.66
CB 118	100	0.23	0.11	0.81	100	0.41	0.17	2.92	100	0.7	0.28	7.3	100	0.50	0.14	2.30
CB 138	100	0.27	0.11	1.25	100	1.1	0.4	10.55	100	0.66	0.29	5.63	100	1.26	0.28	8.88
CB 149	na				14	0.38	0.09	0.47	na				na			
CB 153	100	0.74	0.21	3.06	100	1.44	0.55	9.48	100	2.05	1.12	26.27	100	1.75	0.43	10.16
CB 156	85	0.02	0.01	0.11	100	0.06	0.02	0.43	100	0.07	0.03	0.8	100	0.07	0.02	0.54
CB 170	100	0.07	0.02	0.36	100	0.22	0.07	1.3	100	0.18	0.06	2.16	100	0.23	0.07	1.98
CB 171	69	0.02	0.01	0.07	100	0.04	0.01	0.23	100	0.03	0.02	0.37	100	0.04	0.01	0.26
CB 177	77	0.02	0.01	0.07	100	0.04	0.02	0.43	100	0.03	0.01	0.18	100	0.05	0.01	0.17
CB 180	100	0.17	0.04	0.84	100	0.7	0.2	3.55	100	0.45	0.13	5.29	100	0.65	0.19	5.89
CB 183	85	0.04	0.01	0.19	100	0.12	0.04	0.76	100	0.1	0.04	1.15	100	0.13	0.03	1.03
CB 187	100	0.11	0.03	0.43	100	0.32	0.13	2.55	100	0.22	0.07	1.81	100	0.36	0.1	1.95
CB 194	69	0.03	0.02	0.08	100	0.08	0.02	0.3	93	0.05	0.02	0.38	100	0.07	0.02	0.79
CB 199	69	0.02	0.01	0.08	na				79	0.05	0.01	0.18	na			
CB 196/203	69	0.02	0.01	0.09	na				93	0.04	0.01	0.43	na			
CB 206	10	0.01	0.01	0.01	59	0.02	0.01	0.05	62	0.02	0.01	0.07	62	0.03	0.01	0.2
CB 209	nd				55	0.03	0.01	0.1	15	0.02	0.02	0.3	48	0.03	0.01	0.12

Table S10: Detection frequency, median, min and max concentrations of OCPs and PBDEs quantified in plasma samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Plasma concentrations are in ng/ml ww. OCPs and PBDEs not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
Plasma	2015 *n* = 13				2016 *n* = 22				2015 *n* = 14				2016 *n* = 21			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
OxC	100	0.04	0.02	0.14	100	0.08	0.02	0.53	100	0.24	0.05	2.16	100	0.13	0.04	0.6
TN	100	0.19	0.06	0.36	100	0.26	0.08	0.98	100	0.22	0.1	1.22	100	0.29	0.14	0.59
CN	100	0.07	0.04	0.12	100	0.13	0.05	0.39	100	0.07	0.04	0.44	100	0.14	0.08	0.25
p,p'-DDE	100	1.21	0.56	5.23	100	1.2	0.56	9.47	100	3.95	2.18	47.61	100	1.45	0.48	8.64
p,p'-DDD	na				77	0.09	0.03	0.32	na				67	0.08	0.05	0.19
p,p'-DDT	100	0.2	0.08	0.3	100	0.15	0.06	0.63	86	0.13	0.06	0.31	95	0.27	0.09	0.38
HCB	92	0.09	0.04	0.21	100	0.76	0.26	2.96	100	0.15	0.05	0.8	100	1.02	0.32	2.46
α-HCH	nd				nd				7	0.04	0.04	0.04	nd			
β-HCH	100	0.04	0.03	0.06	55	0.02	0.01	0.08	100	0.06	0.03	0.32	86	0.02	0.01	0.07
γ-HCH	nd				5	0.04	0.04	0.04	nd				nd			
BDE 28	85	0.003	0.002	0.005	23	0.003	0.002	0.005	86	0.01	0.004	0.02	nd			
BDE 47	100	0.06	0.03	0.28	100	0.08	0.01	0.81	100	0.19	0.06	1.82	100	0.09	0.01	0.36
BDE 99	85	0.01	0.003	0.03	86	0.02	0.003	0.16	100	0.04	0.01	0.23	95	0.03	0.002	0.08
BDE 100	100	0.03	0.01	0.11	96	0.03	0.004	0.35	100	0.08	0.02	0.5	100	0.03	0.002	0.14
BDE 153	62	0.01	0.003	0.02	96	0.01	0.004	0.08	100	0.02	0.003	0.07	95	0.01	0.002	0.08
BDE 154	92	0.01	0.004	0.03	100	0.03	0.01	0.17	93	0.01	0.01	0.04	100	0.02	0.003	0.99
BDE 183	nd				36	0.01	0.01	0.03	nd				33	0.01	0	0.01
2'-MeO-BDE68	na				45	0.02	0.01	0.08	na				81	0.01	0.01	0.04
6-MeO-BDE47	na				50	0.04	0.01	0.16	na				86	0.05	0.01	0.12

Table S11: Detection frequency, median, min and max concentrations of PCBs quantified in feather samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Feather concentrations are in ng/g ww. Congeners not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
Feathers	2015 *n* = 13				2016 *n* = 22				2015 *n* = 14				2016 *n* = 21			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
CB 49	na				9	0.37	0.35	0.39	na				5	0.71	0.71	0.71
CB 52	na				59	0.70	0.31	1.14	na				81	0.79	0.37	1.33
CB 74	na				9	0.43	0.39	0.47	na				24	0.56	0.50	0.96
CB 95	na				9	0.22	0.21	0.23	na				na			
CB 99	92	1.18	0.40	3.85	100	0.92	0.41	7.89	100	7.78	2.71	31.05	100	1.08	0.21	3.61
CB 101	92	0.75	0.56	1.82	100	0.55	0.31	1.57	100	1.50	0.80	1.81	95	0.44	0.20	0.99
CB 105	85	0.30	0.11	0.57	100	0.24	0.11	0.94	100	1.37	0.52	3.85	100	0.31	0.12	1.05
CB 118	100	0.72	0.23	2.10	100	0.90	0.45	4.12	100	5.12	2.18	15.03	100	1.07	0.40	3.37
CB 138	100	0.42	0.17	1.51	100	1.86	0.75	11.78	100	1.34	0.60	5.89	100	2.64	0.62	6.26
CB 153	100	1.77	0.63	6.77	100	3.13	1.22	17.07	100	12.86	5.48	38.64	100	4.15	0.92	9.73
CB 156	8	0.10	0.10	0.10	36	0.12	0.10	0.20	71	0.18	0.10	0.45	52	0.16	0.11	0.26
CB 170	69	0.19	0.11	0.48	100	0.34	0.13	1.45	100	0.52	0.27	1.73	100	0.42	0.11	1.23
CB 171	15	0.12	0.10	0.14	14	0.12	0.11	0.34	57	0.20	0.16	0.47	48	0.15	0.10	0.19
CB 177	23	0.12	0.10	0.18	14	0.11	0.11	0.72	57	0.17	0.11	0.31	33	0.14	0.11	0.17
CB 180	100	0.23	0.10	1.03	100	0.74	0.26	3.06	100	0.96	0.56	3.30	100	0.92	0.24	2.73
CB 183	38	0.19	0.10	0.30	82	0.18	0.13	0.84	100	0.38	0.17	1.37	86	0.26	0.10	0.58
CB 187	85	0.19	0.10	0.78	100	0.48	0.17	3.21	100	0.76	0.26	1.89	100	0.45	0.14	1.08
CB 194	23	0.13	0.12	0.14	45	0.15	0.11	0.40	64	0.18	0.10	0.36	62	0.20	0.10	0.36
CB 199	8	0.10	0.10	0.10	na				14	0.15	0.12	0.18	na			
CB 196/203	8	0.10	0.10	0.10	na				57	0.17	0.11	0.47	na			
CB 206	nd				nd				7	0.11	0.11	0.11	nd			
CB 209	nd				23	0.14	0.12	0.22	7	0.11	0.11	0.11	24	0.14	0.1	0.16

Table S12: Detection frequency, median, min and max concentrations of OCPs and PBDEs quantified in feather samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Feather concentrations are in ng/g ww. OCPs and PBDEs not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
Feathers	2015 *n* = 13				2016 *n* = 22				2015 *n* = 14				2016 *n* = 21			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
OxC	69	0.13	0.10	0.32	100	0.26	0.13	1.28	100	1.36	0.36	6.89	100	0.48	0.08	1.47
TN	46	0.15	0.10	0.32	95	0.16	0.11	0.29	100	0.24	0.13	0.41	100	0.16	0.10	0.33
CN	15	0.11	0.10	0.12	59	0.11	0.10	0.16	64	0.12	0.07	0.16	67	0.15	0.10	0.17
p,p'-DDE	100	4.96	2.35	22.10	100	3.23	1.48	17.03	100	26.59	12.38	94.38	100	3.74	1.14	8.81
p,p'-DDD	na				50	0.66	0.28	0.97	na				57	0.55	0.37	1.30
p,p'-DDT	69	0.71	0.31	1.14	73	0.28	0.21	1.03	86	0.91	0.55	1.92	71	0.27	0.20	0.47
α-HCH	nd				5	0.13	0.13	0.13	nd				10	0.12	0.10	0.13
β-HCH	77	0.30	0.17	0.44	36	0.12	0.10	0.29	100	0.46	0.18	0.71	48	0.12	0.10	0.18
γ-HCH	92	0.15	0.10	0.24	14	0.11	0.11	0.11	100	0.14	0.05	0.18	5	0.17	0.17	0.17
HCB	23	0.14	0.11	0.17	100	0.32	0.21	0.67	100	0.18	0.09	0.33	100	0.40	0.25	0.99
BDE 28	nd				nd				7	0.10	0.10	0.10	nd			
BDE 47	100	0.56	0.28	1.58	100	0.30	0.16	2.43	100	1.73	0.63	3.59	100	0.45	0.14	1.06
BDE 99	69	0.16	0.1	0.37	23	0.11	0.10	0.49	100	0.32	0.14	0.69	48	0.15	0.10	0.2
BDE 100	92	0.40	0.11	3.76	27	0.15	0.12	0.18	93	0.16	0.12	5.08	14	0.11	0.10	0.27
BDE 153	31	0.16	0.15	0.21	nd				nd				nd			
BDE 154	nd				5	0.12	0.12	0.12	nd				nd			
6-MeO-BDE47	na				73	0.12	0.10	0.18	na				71	0.12	0.10	0.19

Table S13: Detection frequency, median, min and max concentrations of DPs, PFRs and NBFRs quantified in plasma and feather samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Plasma concentrations are in ng/ml ww, while feather concentrations are in ng/g ww. PFRs and NBFRs not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
	2015 *n* = 13				2016 *n* = 22				2015 *n* = 14				2016 *n* = 21			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
Plasma																
TPhP	15	0.14	0.11	0.17	nd				36	0.19	0.11	0.55	nd			
EHDPHP	15	0.22	0.16	0.27	nd				29	0.12	0.10	0.28	nd			
TDCIPP	nd				nd				71	0.55	0.14	1.41	nd			
TCPP	8	0.14	0.14	0.14	nd				29	0.18	0.10	0.34	nd			
BTBPE	nd				nd				nd				5	0.01	0.01	0.01
TBPH	nd				nd				nd				5	0.08	0.08	0.08
s-DP	31	0.01	0.0002	0.02	5	0.002	0.002	0.002	7	0.06	0.06	0.06	nd			
a-DP	38	0.01	0.003	0.03	5	0.01	0.01	0.01	7	0.01	0.01	0.01	nd			
Feathers																
TPhP	100	21.65	2.54	1229.25	95	3.59	1.29	10.89	100	7.98	3.26	64.34	81	3.08	1.29	10.75
EHDPHP	85	2.67	1.43	11.49	73	3.11	1.28	8.74	100	2.48	1.07	4.42	76	2.19	1.06	3.91
TDCIPP	62	2.61	1.07	8.32	41	2.30	1.30	8.20	71	1.87	1.27	4.28	19	2.70	1.20	4.00
TCPP1	100	18.43	9.22	52.34	100	24.40	6.08	139.41	100	13.27	7.84	28.18	100	20.89	13.13	55.98
TCPP2	54	4.56	1.65	7.28	68	4.94	1.97	29.19	57	2.44	1.38	4.88	33	4.82	2.88	12.38
ΣTCPP		21.21	10.30	59.62		26.95	6.10	168.60		13.96	7.84	32.64		20.90	13.10	68.40
TCEP	92	9.47	3.72	14.79	100	9.28	4.24	23.42	100	4.83	2.43	9.01	100	7.57	4.56	14.65
TBB	nd				nd				7	0.64	0.64	0.64	nd			
TBPH	31	0.62	0.22	0.70	8	0.44	0.44	0.44	36	0.51	0.40	1.03	nd			
s-DP	8	0.10	0.10	0.10	14	0.10	0.10	0.17	nd				33	0.17	0.11	0.43
a-DP	23	0.13	0.11	0.47	32	0.17	0.10	0.62	21	0.11	0.0001	0.76	33	0.43	0.14	1.14

Table S14: Detection frequency, median, min and max concentrations of PFASs quantified in plasma and feather samples from white-tailed eagles from Smøla and Steigen (Norway), from 2015 and 2016. Compounds not analysed are marked with "na", while not detected are marked with "nd". Plasma concentrations are in ng/ml ww, while feather concentrations are in ng/g ww. PFASs not listed in this table were not detected in any of the samples.

	Smøla								Steigen							
	2015				2016				2015				2016			
	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max	df (%)	median	min	max
Plasma	*n* = 13				*n* = 22				*n* = 14				*n* = 21			
PFOSA	nd				9	0.25	0.21	0.28	nd				nd			
PFHxS	15	1.64	1.49	1.79	86	0.09	0.04	0.48	21	0.91	0.90	1.25	81	0.28	0.06	0.57
PFhpS	54	0.11	0.02	0.26	nd				57	0.15	0.08	0.31	24	0.21	0.08	0.29
branched PFOS	100	2.23	0.55	4.20	100	0.65	0.29	1.49	100	5.38	1.85	11.70	100	2.12	0.72	6.83
linear PFOS	100	14.12	6.04	31.85	100	5.25	2.34	8.47	100	16.55	9.55	27.07	100	7.01	3.86	17.5
PFOA	92	0.35	0.12	0.57	86	0.12	0.03	0.29	93	0.53	0.14	1.27	100	0.40	0.11	0.95
PFNA	100	1.82	0.57	4.86	100	0.56	0.35	1.77	100	3.58	1.56	6.48	100	1.69	0.63	6.60
PFDcA	100	1.22	0.66	2.3	100	0.39	0.25	0.82	100	1.44	0.91	2.52	95	0.69	0.36	1.82
PFUnA	100	3.59	2.43	4.36	100	1.15	0.68	2.05	100	3.36	2.30	5.08	100	1.40	0.94	2.15
PFDoA	85	0.57	0.32	0.94	100	0.27	0.09	0.46	100	0.38	0.20	0.86	100	0.22	0.15	0.51
PFTriA	62	0.94	0.31	1.66	100	0.31	0.14	0.65	71	0.63	0.20	1.02	100	0.29	0.07	0.62
Feathers	*n* = 13				*n* = 22				*n* = 14				*n* = 19			
PFOSA	46	0.56	0.23	0.86	95	1.43	0.47	6.52	50	0.53	0.34	1.11	100	1.39	0.45	3.17
PFHxS	nd				59	2.7	1.16	5.50	29	2.92	0.62	5.17	68	3.25	1.03	7.70
PFHpS	nd				14	0.37	0.28	3.27	nd				58	0.35	0.24	0.92
linear PFOS	8	0.22	0.22	0.22	68	1.33	0.47	90.2	29	0.92	0.58	1.82	89	1.69	0.88	4.49
PFHxA	nd				5	0.27	0.27	0.27	21	0.04	0.02	0.07	32	0.28	0.20	0.33
PFOA	nd				36	0.34	0.24	0.91	7	0.11	0.11	0.11	5	0.23	0.23	0.23
PFNA	nd				45	0.4	0.27	0.45	21	0.18	0.05	0.28	79	0.37	0.29	0.45
PFDcA	8	0.07	0.07	0.07	14	0.29	0.23	0.30	nd				58	0.28	0.23	0.46
PFUnA	77	0.18	0.05	0.60	95	0.58	0.34	0.95	64	0.57	0.37	0.81	100	0.82	0.26	1.07
PFDoA	23	0.18	0.07	0.20	32	0.24	0.2	0.50	21	0.14	0.11	0.15	47	0.29	0.20	0.78
PFTriA	31	0.28	0.11	0.51	91	0.66	0.25	1.59	50	0.30	0.11	0.46	100	1.14	0.50	2.02
PFTeA	nd				9	0.23	0.21	0.32	nd				53	0.30	0.19	0.91

Table S15: Detection frequency, median, min and max concentrations of PCBs, OCPs, PBDEs, NBFRs and DPs quantified in preen oil samples from white-tailed eagles from Smøla and Steigen (Norway), from 2016. Compounds not detected are marked with nd. Preen oil was not sampled in 2015. Preen oil concentrations are in ng/g ww. PCBs, OPCs, PBDEs and NBFRs not listed here were not detected in any of the samples.

		Smøla				Steigen			
Preen oil		2016			*n* = 22	2016			*n* = 21
		df (%)	median	min	max	df (%)	median	min	max
Polychlorinated biphenyls (PCBs)	CB 28	18	8.32	5.28	10.27	18	10.15	8.16	12.43
	CB 49	18	3.33	3.13	3.52	38	4.84	2.88	7.22
	CB 52	73	8.81	4.94	12.89	95	9.54	7.07	15.08
	CB 74	14	22.01	20.20	26.79	43	11.25	5.36	30.78
	CB 95	91	8.85	2.75	16.15	95	9.98	5.89	14.65
	CB 99	100	28.00	12.33	198.68	95	35.10	14.22	161.58
	CB 101	100	16.68	9.99	49.56	95	19.95	12.03	32.62
	CB 105	100	14.94	6.70	70.56	100	22.33	7.99	91.04
	CB 118	100	51.84	19.68	240.09	100	66.46	23.94	289.96
	CB 138	100	129.63	39.37	720.65	100	168.08	40.26	602.90
	CB 149	100	16.39	9.04	58.56	95	15.30	11.71	26.81
	CB 153	100	259.08	81.81	1420.57	100	327.32	94.03	1163.43
	CB 156	100	10.20	3.32	37.61	100	14.21	4.17	54.16
	CB 170	100	37.74	10.16	180.79	100	44.12	12.85	184.32
	CB 171	100	5.69	1.86	35.80	100	7.10	2.12	25.53
	CB 177	100	7.01	2.44	78.40	100	6.04	2.74	16.39
	CB 180	100	117.86	29.89	489.81	100	139.58	40.90	579.26
	CB 183	100	21.96	6.03	102.53	100	25.66	7.31	99.74
	CB 187	100	55.94	16.74	434.19	100	59.11	20.74	186.85
	CB 194	100	9.44	1.56	33.76	100	9.11	1.63	69.10
	CB 206	100	3.87	1.31	17.45	90	3.57	1.15	19.34
	CB 209	100	4.50	1.01	25.02	76	4.56	1.16	14.56
Organochlorinated pesticides (OCPs)	OxC	100	9.52	3.36	30.37	100	13.55	4.80	53.73
	TN	100	34.92	14.32	63.44	100	39.04	23.64	75.39
	CN	100	13.22	6.47	22.28	100	15.75	10.03	26.21
	p.p'-DDE	100	289.56	170.99	1447.32	100	364.88	185.38	932.96
	p.p'-DDD	91	13.39	7.89	29.52	95	13.40	9.59	23.43
	p.p'-DDT	100	7.07	3.85	15.09	100	7.65	5.06	46.63
	β-HCH	82	1.67	1.13	7.07	100	2.32	1.05	4.41
Polybrominated diphenyl ethers (PBDEs)	BDE 28	18	0.60	0.36	1.66	33	0.43	0.37	1.51
	BDE 47	100	9.19	4.25	96.35	100	10.47	4.45	60.28
	BDE 99	100	1.68	0.81	10.34	100	1.67	0.83	6.70
	BDE 100	100	2.89	0.99	22.25	100	3.36	1.25	17.20
	BDE 153	82	1.26	0.76	5.77	67	1.44	0.83	3.83
	BDE 154	100	1.68	0.81	10.34	100	1.67	0.83	6.70
	BDE 183	nd				5	0.75	0.75	0.75
	6-MeO-BDE47	95	8.17	2.43	21.57	100	12.44	7.33	74.81
	2-MeO-BDE68	95	1.84	0.70	3.66	48	2.52	1.21	8.42
Dechlorane plus (DPs)	*s*-DP	nd				nd			
	a-DP	5	0.45	-	-	nd			

901