Spatiotemporal analysis of perfluoroalkyl substances in White-tailed eagle (*Haliaeetus albicilla)* nestlings from northern Norway – a ten-year study

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ABSTRACT

The white-tailed eagle (*Haliaeetus albicilla*) in Scandinavia has suffered from impaired reproduction due to high exposure to industrial pollution between the 1960s and 1980s. While population numbers are rising again, new contaminants, such as per- and polyfluoroalkyl substances (PFAS), are increasingly found in high trophic avifauna and are of concern to potentially impact once again population health. In the present study, we examined PFAS levels in plasma of white-tailed eagle nestlings from northern Norway over the last decade (2008-2017). While PFOA and PFNA exposure did not follow a significant time trend, PFOS and PFHxS concentrations decreased over time, and $\geq C_{11}$ perfluorinated carboxylic acids only seem to level-off during the last four years. This may in fact be the first evidence for a change in the trend for some of these compounds. Furthermore, since several PFAS are expected to be highly present in aqueous film forming foams used at airports, we also investigate the potential of the two main airports in the region to act as hotspots for PFAS. Our results indeed show decreasing exposure to PFOA with distance to the airports. Altogether, our results seem to show that legislation actions are effective, continued concern for PFAS exposure of high trophic wildlife is still warranted, even in the northern environment.

INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are environmentally stable man-made chemicals emitted into the environment as a product or by-product of various industries. These substances are classified in different groups[1], in which the two most common are perfluoroalkane sulfonates (PFSAs) and perfluorinated carboxylic acids (PFCAs). PFAS are essential for numerous industrial applications due to their surfactant properties, oil and water repellence as well as for their stability at high temperature.[2] Despite the fact that they have been used since the 1950s, they have only been studied in relation to wildlife since the early 2000s when some of them were found to be ubiquitous, persistent and to biomagnify along food chains.[3-12] Some PFAS were in addition found to have adverse effects on human and animal health in several *in vitro* and *in vivo* studies.[2,3,6,13-17] Such findings led the 3M Company, a main producer, to phase-out its production of C_8-PFAS (perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) related products) during 2000-2002. Subsequently, PFOS and related products are regulated in the European Union (EU) since 2006,[18] and were added in 2009 to Annex B of the Stockholm Convention on Persistent Organic Pollutants (POPs), which restrict, but do not completely ban their use.[19] PFOA and higher homologues, have been phased-out as well by other main producers in the United States (US) during the 2010 decade,[20] but are still produced in some Asian countries[21].

Airports have been important local emission sources as their use of Aqueous Film Forming Foams (AFFFs), containing some fluorosurfactants, have led to uncontrolled emissions into the surrounding environment,[22-24] causing significant persistent ground contamination[25-27]. While PFAS have not been produced in Norway, AFFFs have been found to be responsible for high contamination levels (of PFOS in particular) close to

airports across the country.[28,29] In northern Norway, activities at the Harstad/Narvik airport (Supporting Information (SI), Figure S1) have caused a particularly elevated PFOS contamination in surrounding lakes, freshwater fish and groundwater.[28-30] In addition, a recent study found a positive relationship between elevated levels of PFOS, perfluorohexane sulfonic acid (PFHxS) and perfluorononanoic acid (PFNA) in human serum and the consumption of freshwater fish from lakes around this specific airport.[29] AFFFs containing PFAS have not been used at public airports in Norway since 2012,[31] but no public data are available for Harstad/Narvik military airbase.[29] Moreover, some of these compounds are extremely persistent and might still be abundant in the environment. Such findings raise concern regarding how such contamination spreads locally away from the sources, and support the need for an assessment of PFAS' scattering impact on the contamination level of local wildlife.

In Norway, recent studies on terrestrial and marine fauna showed some decreasing time trends in the exposure to PFOS over the last 30 years.[32-35] On the other hand, and despite legal restrictions, C_{10} to C_{13} PFCAs were found to be increasing over the same time period.[32-35] However, time trends for a variety of PFAS in Norway are still scarce and were mainly conducted in the remote Svalbard. It is thus difficult to assess the impact of restrictions on production, use and emissions on the Norwegian coastal environment. Moreover, close to eighty per cent of the Norwegian population live less than 10 kilometers from the coast, consequently, having sources of concern for both legacy and potentially emerging PFAS, such as aforementioned airports in coastal northern Norway, makes it crucial to have clear understanding and follow up of these compounds local time trends.

The White-tailed eagle (*Haliaeetus albicilla*; WTE) is a coastal raptor that breeds in Northern Europe, Russia and West Greenland. Its diet is largely composed of seabirds and fish.[36,37] Because of its residency and apex food chain position, the WTE is particularly at exposure risk for bioaccumulative and biomagnifying contaminants, and, therefore, at the same time a highly valued and established bioindicator of local contamination with PFAS.[38] PFAS are now found in higher concentrations than legacy POPs in WTE nestlings of northern Norway.[39,40] In fact, while information of physiological impacts of PFAS on raptors remain scarce, PFAS have been found to be in sufficiently high concentrations in the Norwegian environment to potentially cause adverse effects on development in a terrestrial lower trophic level bird of prey, the common kestrel (*Falco tinnunculus*).[41]

In the present study we aimed at investigating the spatiotemporal trends of plasma PFAS concentrations in WTE nestlings from northern Norway from 2008 to 2017. We hypothesized that: 1) time trends for phased-out long-chain PFAS (i.e. $\geq C_8$ PFSAs, $\geq C_7$ PFCAs), in particular PFOS and PFOA, are decreasing; and 2) spatial trends would show a hotspot pattern only for some PFAS locally released from AFFFs (PFOS, PFHxS and PFNA) with distance to the County airports.

MATERIALS AND METHODS

Study regions

The study was conducted, in Troms County (68-70°N, 15-22°E), in northern Norway (SI Figure S1). The area consists of a long coast line defined by numerous fjords and islands, and contains two major towns, Tromsø (75,000 inhabitants) and Harstad (21,000 inhabitants), with civilian airports, the Tromsø airport (2,000,000 passengers per

year) and Harstad/Narvik airport (700,000 passengers per year; these numbers do not include military activity, which is only relevant for the latter).[42,43] Besides the airports, other local industries may be potential sources of PFAS due to their use of AFFF. Close to Harstad, a firemen training school (Norges brannskole) and a fuel depot (Statoil), whereas in Tromsø PFAS are spread by diffuse sources in wastewater treatment plants of the area treating both households and hospital waste water.[44-47]

Field methodology

WTE nestlings were sampled for blood during the late-spring of 2008 to 2017 (n=164). Nests were initially checked for breeding activity in April using binoculars and telescopes, while keeping a distance in order to minimize disturbance of birds. The presence of a bird incubating on the nest was used as a confirmation of breeding activity (see Sletten *et al*.[39] for more details).

When the birds were approximately eight weeks old, just before fledging, the nest were visited. Blood samples (5-10 mL) were taken from the brachial vein using heparin-coated syringes which were stored in a cooler for transport to the lab. Plasma was obtained through centrifugation of the blood at 8,000 rpm for 10 min and was subsequently stored at -20°C until chemical analysis. The field protocol was approved by the Norwegian National Animal Research Authority.

PFAS analysis

The chemical analysis of PFAS in plasma was carried out at the Norwegian Institute for Air Research (NILU) in Tromsø, Norway. We used the method previously described by

Sletten *et al.*[39] In brief, the plasma samples (0.20 mL) were spiked with internal standards (0.1 ng μL^{-1} of a ^{13}C-labeled PFAS mix; Wellington Laboratories Inc., Guelph, Canada), then extracted in methanol using a consecutive series of vortexing followed by sonication baths. After centrifugation (10,000 rpm) for sedimentation, the supernatant was cleaned using glacial acetic acid and ENVI-Carb 120/400 (Supelco 57.210-U). After final centrifugation, the supernatant was spiked with recovery standards (0.1 ng μL^{-1} of 3.7-diMe-PFDcA in methanol; ABCR, Karlsruhe, Germany) and then stored at +4°C. Prior to quantification, an aliquot (50 µL) of the prepared mix was transferred into an auto-sampler vial with the same amount of 2mM NH_4O Ac (>99%, Sigma-Aldrich, St-Louis, MO, USA) and vortexed.

Quantification was conducted by ultrahigh performance liquid chromatography triple-quadrupole mass spectrometry (UHPLC-MS/MS), as previously described by Hanssen *et al*.[48] The chromatograms were quantified with LCQuan software (version 2.6, Thermo Fisher Scientific Inc., Waltham, MA, USA). Quantification was done using the internal standard method and isotopically labelled compounds and an eight-point calibration curve with a concentration range from 0.02 pg μL^{-1} to 10 pg μL^{-1} was used. We quantified six PFCAs (PFOA, PFNA, perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA)) and two PFSAs (PFHxS and PFOS); SI Table S1.

To assure the quality and control for reproducibility and precision of the method during the different sample preparation periods, one blank and a standard reference material (2008-2012: human serum NIST 1957; 2013-2016: human serum AM-S–Y1607 INSPQ within the Arctic Monitoring and Assessment Programme ring test) were

concurrently analysed every 15 samples to verify quality of the prepared samples, test reproducibility and precision of the method. Inter-day and intra-day relative variations of PFAS with standardized concentrations in the reference material are provided in SI Table S2 for the period 2013-2016. They ranged between 4-21% and 9-27%, respectively, with the exception of the inter-day variation of PFNA (25.6%) and the intra-day variation of PFHxS and PFDcA (31% and 27%). All blanks concentrations were below the instrument detection limits. Limit of detection (LOD) was defined as three times the signal to noise ratio for the specific matrix. The mass spectrometers have been replaced in 2012, thus, LODs were different throughout the years and can be found in SI Table S3. All concentrations are presented on a wet weight basis (ww).

Data processing and analyses

All data processing and analyses were done using R, version 3.4.3.[49] In addition to the sampling year and the PFAS concentration, models included two spatial variables: 1) a quantitative variable for the distance from each nest to the nearest airport; and 2) a qualitative variable dividing the study area halfway between Harstad and Tromsø (North: 69.3-70.2N 17.7-20.5°E; n = 107; South: 68.4-69.3°N 15.5-17.7°E; n = 57; SI Figure S1) in order to study possible spatiotemporal trend differences linked to both towns, each considered a source of contamination. A sample size of 164 nestlings was available for statistical analyses. Only compounds detected in more than 70% of the samples were analysed. By using a unique data set of a relatively high number of samples collected continually during ten years in the Troms County, this study provides a strong and valuable investigation of PFAS spatiotemporal trends at a local scale. The unique characteristic of

this dataset created some analytic challenges since the contaminant measurements were done in the lab at different period regarding the year of the samples. The sensitivity varied through years due to modernization of the equipment, leading to variations in the compound-specific LODs (SI Table S3). For some years when the LOD was high, most of the samples were lower than LOD. To prevent false high estimations on these years, for each contaminant independently of each other, years with more than 50% of points measured lower than LOD were removed from analyses. Years 2009-2010 were removed for PFOA and PFTrDA, years 2010 and 2012 were removed for PFHxS, and year 2010 only was removed for PFUnDA; SI Table S4. A difference between LOD per year leads to potential bias regarding the study of temporal trends, especially due to LODs being higher during the study's first years. This problem was addressed by setting, for each contaminant, a common LOD for all years. This assigned value was chosen as the maximum LOD among all years combined, and every sample value smaller or equal to this new LOD was set to half of its value. While risking to lose some information towards the more recent years in the study, for which LODs were the lowest, this conservative method removes the potential bias of using different LODs to study true temporal trends. PFDA showed different spatial trends whether original LODs were included in the analyses or not and was also removed from the dataset to avoid misinterpretation of the results.

Descriptive statistics were computed for both the northern and southern region (Table 1). Before modelling, all PFAS concentrations were first log-transformed to meet the assumption of homoscedasticity and normality of residuals, which we also confirmed through inspection of plots of residuals against fitted values and normal Q-Q plots.[50] Mixed models were used to investigate spatial and time trends of each homologue. As the majority

of the nests included only one (44.6% of the nests) or two (48.2%) nestlings, and some contained three (7.2%), a number was attributed to each brood (i.e. brood ID) and was used as a random variable. To do so, we used the R package nlme (version 3.1-137).[51]

We performed two sets of analyses – one to assess time trends and one to assess spatial trends. In the temporal analysis, the main effect of year (a numeric variable), its second-order polynomial ($year^2$) and geographical region (a factor variable with the division of the study area as levels) were used as potential predictors, while plasma concentration of each homologue was used as the response variable; SI Table S5. In the spatial analysis, the same set-up for the candidate models was used, but we replaced year with distance to the nearest airport (a numeric variable measured in km; SI Table S6). In both analyses, we rescaled and ranked models relative to the value of the model with the lowest second-order Akaike's Information Criterion value for small sample sizes (AICc; Δ_i denotes this difference for model i).[52-54] Among the models with a $\Delta_i \leq 1.5$, we selected the most parsimonious model, i.e. the one with the fewest degrees of freedom, and used this model for inference (an approach commonly applied; see e.g. Bårdsen & Fox[55], Næss et al.[56]). We used the R package AICcmodavg (version 2.1-1)[57] to rank the models based on Δ_i, and provide the statistics for our two sets of candidate models as judged from Δ_i values. Statistical significance was interpreted at an α-level of 0.05. Changes over time were obtained for PFAS explained by linear models. Log-transformed PFAS yearly changes (%) were derived from $100 \times (\exp(estimate\ for\ year) - 1)$.

RESULTS AND DISCUSSION

Descriptive statistics: PFAS exposure

Of the 7 targeted PFAS, six (PFOA, PFNA, PFUnDA, PFTrDA, PFHxS and PFOS) were detected above a 70% threshold (SI Table S3). PFDoDA was overall detected in 67.3% of the samples, but in only 30% of the samples before 2013 and in 97% afterwards, consequently, it was not included in the analyses. Results on the last 5 years of the study (2013-2017) can however be found for PFDoDA in SI Table S7, S8 and Figure S4 for further information.

Mean, median and range descriptors of concentrations are given for each PFAS in Table 1. Among the PFAS, PFOS had the highest concentrations over the decade in both regions and represents 61-75% of ∑PFAS. PFUnDA and PFNA were, respectively, the second and third major PFAS detected.

Table 1. Descriptive statistics (mean ± standard error (*SE*), median and range (min-max)) for PFAS concentrations (ng g^{-1} ww) in plasma of white-tailed eagle nestlings from northern Norway in both northern and southern regions (see SI Figure S1).

	Mean ± *SE*	**Median**	**Min-max**
PFOA (C_8)			
North	1.10 ± 0.12	0.78	0.15-7.79
South	0.91 ± 0.11	0.79	0.15-2.91
PFNA (C_9)			
North	3.70 ± 0.23	3.28	0.56-13.7
South	3.90 ± 0.50	2.87	0.55-22.8
PFUnDA (C_{11})			
North	4.57 ± 0.30	3.97	1.00-20.4
South	4.07 ± 0.26	3.77	1.14-7.79
PFTrDA (C_{13})			
North	2.15 ± 0.23	1.52	0.30-11.2

South	1.67 ± 0.20	1.35	0.30-6.25
PFHxS (C_6)			
North	1.50 ± 0.12	1.27	0.25-6.80
South	0.70 ± 0.11	0.53	0.25-3.10
PFOS (C_8)			
North	33.1 ± 3.06	27.10	7.32-247
South	36.0 ± 5.43	23.20	1.00-249
∑PFCAs			
North	11.5 ± 0.60	10.3	2.08-27.1
South	10.8 ± 0.91	9.95	2.74-34.7
∑PFSAs			
North	33.9 ± 3.29	26.9	7.46-254
South	28.7 ± 4.41	19.8	1.25-134
∑ PFAS			
North	41.1 ± 2.30	37.3	12.2-107
South	39.6 ± 5.68	30.7	8.84-156

The general exposure profile, with PFOS dominating followed by PFUnDA and PFNA was similar to what has been documented for eggs and blood of seabird species in Norway.[58-60] This may be explained by the historical predominance of highly persistent and bioaccumulative PFOS and its precursors in consumer products (e.g. in textiles or carpets) and industrial emissions before being regulated.[9] PFOA was the main PFCA produced together with PFNA (respectively estimated at 85% and 9% of the global historical PFCA release in 2006),[61] but was found in lower concentrations in the present study in comparison to the longer-chain PFCAs (i.e. ≥C_{10}). The longer chain PFCAs are however by-products of PFOA and PFNA production, they are more bioaccumulative than PFOA and PFNA, and therefore more likely to be found in wildlife.[9,61] Indeed, as it has been shown in

mammals, PFAS have different bioaccumulative characteristics according to the chain-length (balance between uptake, storage and excretion) and their functional groups.[62,63] For these reasons, the distribution profiles found in WTE nestlings might be different from PFAS concentrations in the abiotic media.

Concerning exposure and toxicological implications, PFAS have been reported to have adverse effects such as immunotoxicity, hepatotoxicity, hormonal disruption and impairment of reproductive success on different marine species.[14,15] In particular, in some avian top-predators, such as in chicks of black-legged kittiwakes (*Rissa tridactyla*), PFAS have been found to disrupt thyroid hormone homeostasis at lower exposure than for the WTE nestlings in the present study.[64] In great tits (*Parus major*), high concentrations of PFAS interfered with reproduction success, by failure in hatching in particular.[65,66] Conversely, some studies found no consequences of PFAS concentrations on wild birds. For instance, no effect of PFAS on humoral immunity has been measured in WTE chicks in the studied population.[39] Also, no consequences on demographic parameters have been observed in a population of adult lesser black-backed gull (*Larus fuscus fuscus*) carrying a similar contamination burden in northern Norway.[59]

As a consequence, it is difficult to apprehend the impact of PFAS on wild birds. Too few studies have been conducted on raptors, especially on WTEs, to draw consistent conclusions. This is especially also due to the environmental behavior and fate of PFAS to vary considerably among different species.

Time trends

The year of sampling was the only variable in the trend models significantly explaining the temporal variation in PFOA and PFNA exposure. However, for the longer-chain PFUnDA and PFTrDA the quadratic time component was also retained (SI Table S5). PFOA showed a non-significant decreasing tendency along years (t=-0.05, p=0.20; Table 2 and Figure 1). PFNA exhibited no statistically significant time trend in neither region (t= 0.03, p=0.24), in contrast to PFUnDA and PFTrDA which both showed a significant time trend with increasing concentrations up to 2014 (after which the concentrations declined). No other variable than the sampling year was retained in the model explaining temporal PFOS variation, but for PFHxS the sampling region was also retained in the selected model (SI Table S5). PFHxS also showed significant decreasing time trends in both regions (t=-0.07, p=0.02), but the contamination level was significantly lower in the southern region (t=-0.72, p<0.001; Table 2 and Figure 2). PFOS concentrations decreased over the entire decade (t=-0.07, p<0.01). Summary statistics of the targeted PFAS concentrations per year can be found in SI Table S4.

277 **Table 2.** Factors affecting PFAS concentrations (ng g^{-1} ww) time trends in white-tailed
278 eagle chicks from northern Norway, estimated by mixed linear regression models.
279 Significant *p*-values are bolded.

280

	Time trend[a]			
Parameter	**Estimate**	***SE***	***t*-value**	***p*-value**
PFOA (R^2m[b]: 0.02, R^2c[c]: 0.62)				
Year	-0.05	0.04	-1.29	0.20
PFNA (R^2m: 0.01, R^2c: 0.75)				
Year	0.03	0.02	1.18	0.24
PFUnDA (R^2m: 0.11, R^2c: 0.85)				
Year	133	36.1	3.69	**<0.001**
Year²	-0.03	0.01	-3.69	**<0.001**
PFTrDA (R^2m: 0.10, R^2c: 0.76)				
Year	169	54.89	3.07	**<0.01**
Year²	-0.04	0.01	-3.07	**<0.01**
PFHxS (R^2m: 0.22, R^2c: 0.74)				
Year	-0.07	0.03	-2.47	**0.02**
Region (South)	-0.72	0.18	-4.11	**<0.001**
PFOS (R^2m: 0.07, R^2c: 0.71)				
Year	-0.07	0.02	-2.99	**<0.01**

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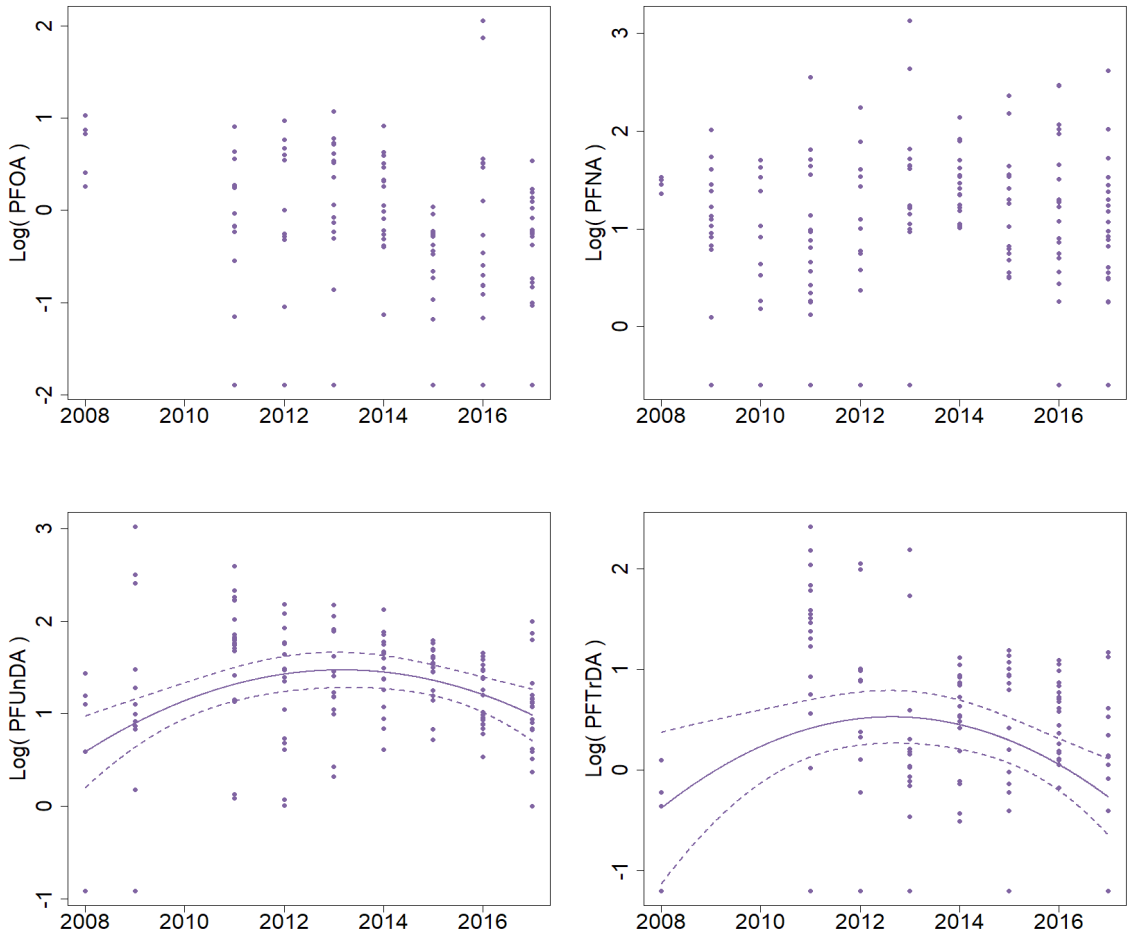
[a] The predictor variables in the statistical models were year, geographical Region (Region), their interaction, as well as the second-order polynomial (year²); these variables were selected based on the most parsimonious model with the lowest second-order Akaike's Criterion (AICc).
[b] R^2m: Marginal coefficient of determination, i.e. variance explained by the fixed effects.
[c] R^2c: Conditional coefficient of determination, i.e. variance explained by the entire model, including both fixed and random effects.

Among the PFCAs targeted in the present study, PFOA showed no significant temporal trends on the decade, despite legal regulation and voluntary phase-out that greatly reduced its production and related >C_8 PFCA compounds in Western Europe and in the US between 2006 and 2015.[20,67,68] Similarly, in previous studies, no significant trends have generally been observed in biota for PFOA, indicating that the phase-outs and regulations did not affect the global level of this compound, yet.[69] Some long-chain PFCAs such as PFNA, PFDA, PFUnDA, PFDoDA and PFTrDA are by-products of PFOA and are therefore impacted by PFOA restrictions, nevertheless, these compounds are generally found increasing with time in biota.[69] In the present study, as for PFOA, PFNA showed no change over time despite the regulations, in contrast to PFUnDa And PFTrDA which showed decreasing or levelling off trends after 2013 (Figure 1). For comparison, increasing temporal tendencies were generally found in Norwegian wildlife.[32,33] However, these studies are based on data collected mostly before 2010 and were collected on terrestrial biota, which might present different time responses as PFAS exposure main pathway is distinct from marine environments.[69] In Greenland, Norway, and Sweden, another study on PFAS concentrations in feathers of dead WTE was conducted on a larger time period (1968-2015).[35] Contrary to what we measured in northern Norway nestlings, the sum of 6 ≥C_8 PFCAs was found to be increasing on the whole time period in Norwegian dead specimens. We believe that such a difference might be due to the low number of dead WTEs sampled each years, as well as the fact that the levelling off tendency in the present study happened at the end of Sun et al. study time-period. Consequently, a late decrease might have been concealed on such a long time study. In the US, Bald eagles (*Haliaeetus leucocephalus*) nestlings' plasma was found with strong declining concentrations of PFOA

and PFUnDA from 2006 to 2011.[70] As in Norwegian WTE, PFNA showed no clear trend in Bald eagles. Despite these variations were not uniform across the study area in Bald eagles, these findings in both sea eagles have some similarities and might be evidences of the phase-outs consequences. The present study might thus be one of the first showing the effect of regulatory actions on $\geq C_{11}$ PFCAs concentrations in biota from Northern Norway. As an additional factor for consideration, the waste stocks of long-chain fluorotelomer-based precursors phased-out in 2010 are becoming a source of increasing importance as their final degradation products are long-chained PFCAs, forming a new source of release for these compounds in the environment.[71] The intermediate degradation products are highly volatile and travel long distances, in particular in polar and sub-polar regions in the northern hemisphere.[21,72] Despite these potential secondary sources, we observed that concentrations of long-chain PFCAs levelled-off. However, in marine environments, atmospheric transport of $\geq C_{10}$ PFCAs precursors is considered to be much lower than long-range oceanic transportation of the long-chain final products in the Arctic.[73] Thus, oceanic long-chain final products as well as potential local fluorotelomer-based precursors emissions might represent the main sources for these contaminants found in WTEs. For the above reasons, long-chain PFCAs should remain to receive attention in order to confirm temporal changes, preferably in reaction to legal or voluntary phase-out.

Corroborating our expectations, PFOS showed a decreasing time trend (~9.5% per year) during these last ten years (Figure 2). Such a decline has been documented on the previous decade as well in other studies on Norwegian birds and their eggs.[32,33] It also follows the continuous decline measured in dead WTE specimens collected in Norway,[35] as well as the decline evidence found in Bald eagles of the US.[70] Nevertheless, in a

systematic review, Land *et al.*[69] compared all temporal studies and found no overall trend; i.e. patterns for PFOS varied in Europe and the Arctic where the effect of phase-outs has been unclear. Concentrations of PFHxS also decreased over time in the entire study area (9% per year), though the contamination level was found to be higher in the northern subregion. Such a decline along time has also been observed in Bald eagles from the upper Midwestern US.[70] PFHxS has been used in aqueous firefighting foams for years, but regulations might explain the observed decreasing time trend.[68,74] The intensity difference in PFHxS exposure between the two regions might be due to the dissimilarities in local coastal characteristics. In the south, the nests are situated in large fjords exposed to the open sea, whereas in the north a long and narrow fjord system could act as a trap for contaminants. Hence, such spatial dissimilarities could cause differences in the time trends in each of the regions. However, as PFOS is relatively close to PFHxS in physicochemical characteristics, we would expect a similar regional difference, but this was not the case. Contrasted emissions between both regions may also offer an explanation. In conclusion, not only production and release may explain the levels of PFAS found in biota, but also the behaviour and fate of these compounds in the environment. As an additional factor of consideration, their absorption, tissue distribution and excretion driven by differences in intrinsic characteristics of each congener, are also major dynamic mechanisms which contribute to explain PFAS concentrations.[75,76]



348

349 **Figure 1.** Time trends for the concentrations of PFOA, PFNA, PFUnDA and PFTrDA in
350 white-tailed eagle nestlings from northern Norway. The solid line refers to a statistically
351 significant time trend (and are based on the selected models presented in SI Table S5), lined
352 by the dotted lines representing 95% confident intervals (CIs); no lines indicate the absence
353 of a significant trend.

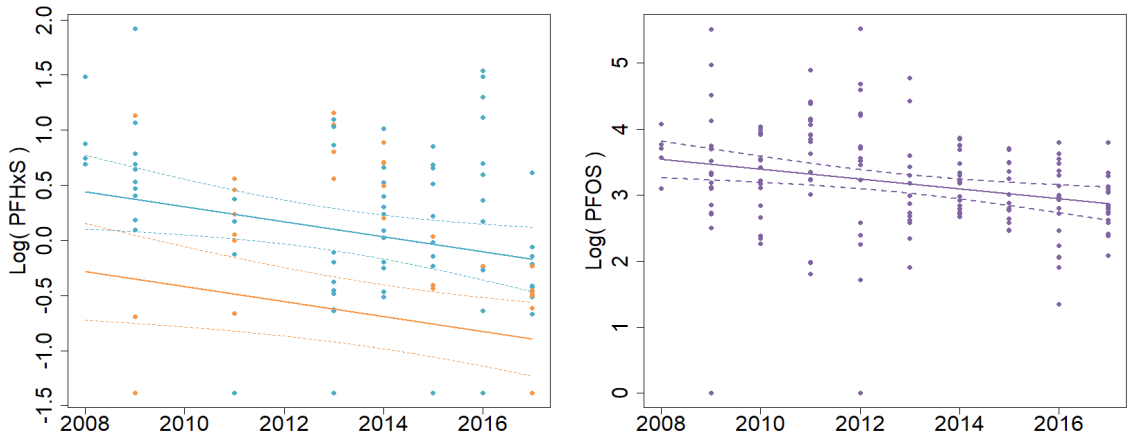


Figure 2. Time trends for the concentrations of PFHxS and PFOS in white-tailed eagle nestlings in both study regions of the Troms County. The solid line refers to a statistically significant linear regression (based on the selected models presented in SI Table S5), lined by the dotted lines representing 95% CIs (orange for the southern region and blue for the northern one, purple is used if the trend is similar in both regions).

Spatial trends

The distance to the nearest airport and the region were selected as explanatory variables in the model explaining PFHxS variations, while the models for the remaining PFAS only retained the distance to the nearest airport (SI Table S6). However, PFOA only decreased significantly with distance of the airport (t=-0.01, p=0.03). PFHxS concentrations in both regions were not significantly linked to the distance to the airports (t=-0.01, p=0.32), but as found in the temporal trends, the southern region was significantly less contaminated than the northern one (t=-0.67, p<0.01; Table 3 and Figure 3).

Table 3. Factors affecting PFAS concentrations (ng g^{-1} ww) spatial trends in white-tailed eagle chicks from northern Norway, estimated by mixed linear regression models. Significant *p*-values are bolded.

	Spatial trend[a]			
Parameter	**Estimate**	***SE***	***t*-value**	***p*-value**
PFOA (R^2m: 0.05, R^2c: 0.61)				
Distance	-0.01	0.01	-2.16	**0.03**
PFNA (R^2m: 0.01, R^2c: 0.75)				
Distance	0.00	0.00	-0.82	0.42
PFUnDA (R^2m: 0.01, R^2c: 0.85)				
Distance	0.00	0.00	0.76	0.45
PFTrDA (R^2m: 0.00, R^2c: 0.76)				
Distance	0.00	0.01	-0.32	0.75
PFHxS (R^2m: 0.18, R^2c: 0.73)				
Distance	-0.01	0.01	-1.00	0.32
Region (South)	-0.67	0.21	-3.12	**<0.01**
PFOS (R^2m: 0.00, R^2c: 0.71)				
Distance	0.00	0.00	0.60	0.55

[a] The predictor variables were distance to the nearest airport (Distance), geographical region (Region) as well as their interaction.

These results did not confirm our initial hypothesis pointing out airports as important local sources of PFAS. Indeed, among the different PFAS known to have been used in large amount in AFFFs or found close to Harstad/Narvik airport (e.g. PFOS, PFHxS and PFNA),[25,29,77] none was decreasing with distance to the airports (see graphs in SI Figure S2, S3). Hansen *et al*.[29] found elevated levels of PFOS, PFHxS and PFNA in serum of humans consuming fish from lakes around Harstad/Narvik airport, compared to low-consumption groups and non-consumers. In the same study, similar differences were observed with higher levels of these three PFAS measured in the fish from the vicinity of Harstad/Narvik airport compared to fish from a control lake sampled 15km away. Hence, we would expect a similar decreasing spatial trend with distance to the airports for these three compounds in WTE nestlings, contrarily to what we measured. A simple explanation for most compounds lack of decreasing spatial trends could be that local contamination decreases to baseline levels by fast environmental dilution after being released in the fjords. Similarly, in water and sediment of Resolute Bay (Nunavut, Canada), elevated levels of PFAS (PFHxS, PFOS, Perfluoroheptanoate (PFHpA) and PFOA) were measured, and was attributed to emissions into the environment by a local airport and a treatment plant.[78] However, Butt *et al.* [79] did not find these contaminants in ringed seals (*Phoca hispida*) from the vicinities. Consequently, it is difficult to make assumptions about how the historical use of AFFFs containing PFAS in the Troms County may influence the concentrations in the surrounding marine fauna. Another reason why we found no effect of distance for the homologues historically emitted by AFFFs could be that other local sources may be spreading PFAS, blurring the effect of the airports. For example, wastewater treatment plant effluents, which have been found to be potential important sources of PFAS for the

ecosystems.[80] For PFOS, however, the high concentrations, and the fact that Harstad airport is a regional source,[29] did not result in significant variations with distance to the airport. Thus, the local sources seem to have minimal impact on the environmental levels measured outside of their close vicinities. The physicochemical characteristics of PFAS might also explain their absence of spatial trends with distance to the airports. Indeed, the carbon chain length has been described as a discriminant factor for PFAS partitioning between water and sediments, longer carbon chains being more adsorbed to sediment than shorter chains, prevailing in the water.[81] Consequently, the three PFAS known to have been mostly found in in AFFFs used in the airports might not be bioavailable for aquatic WTE preys as they are long-chain PFASs, and thus, potentially directly adsorbed to local sediment for the most part.

Despite this absence of variation for these three targeted PFAS, PFOA was found decreasing in a significant manner with distance to the airports. PFOA has not been found to be associated with higher fish consumption in human living close to Harstad/Narvik airport,[29] therefore, this result is surprising. Some of the new generations PFOS-free AFFFs used in Norway contain traces of PFOA, however, the concentration measured in these products are much lower than those of PFOS, PFHxS and PFNA in old-generation AFFFs.[44] Since PFAS adsorption to particles is increasing with their chain-length, PFSAs are also more bound to sediment than PFCAs with a similar chain-length.[82,83] Consequently, despite PFOA being released in far lower amount by AFFFs in airports, it might be available in higher concentrations than PFOS, PFHxS and PFNA in aquatic ecosystems adjoining the airports. Such a difference may be amplified enough in WTE's diet to detect a decreasing trend with distance to the airports.

In the present study, only 12 nests were sampled between 0 and 7 kilometres from the two airports and a gap exists from 7 to 15 kilometres with no nests sampled. In that context, a decrease with distance in a close range from the airport would be difficult to bring to light. Additionally, graphically for PFOA, only the few nests situated between 0 and 7 kilometres from the airports seem to have a leverage effect on this decreasing tendency. To confirm our results and for a better resolution on spatial PFAS variability, data would be valuable to acquire on water, sediments and low trophic biota closer to potential sources. For future studies, investigating stable isotopes in WTE chicks of the Troms County could give a general understanding of how plasticity in feeding behaviour may affect local exposure, and the biomagnification potential of the studied PFAS in general.

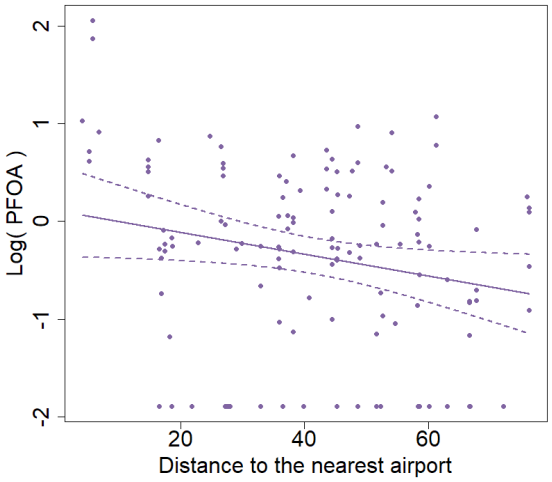


Figure 3. Spatial trends of the concentrations of PFOA in white-tailed eagle nestlings from northern Norway with distance to the nearest airport (km). The solid line refers to a statistically significant linear time trend, lined by the dotted lines representing 95% confident intervals.

ASSOCIATED CONTENT

Supporting Information

The following files are available free of charge.

Map of the study regions; targeted PFAS, classification, abbreviations, names, chemical structures, molecular weight and CAS-numbers; list of standard reference material inter-day and intra-day variations; targeted PFAS, level of detection and limit of detection by sampling year; descriptive statistics of each PFAS for each year; model selection for each selected PFAS; Graphs of spatial trends for each PFAS; PFDoDA temporal variations (PDF).

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Notes

The authors declare no competing financial interest.

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463 REFERENCES

464 (1) Buck, R. C.; Franklin, J.; Berger, U.; Conder, J. M.; Cousins, I. T.; de Voogt, P.;
465 Jensen, A. A.; Kannan, K.; Mabury, S. A.; van Leeuwen, S. P., J. Perfluoroalkyl and
466 Polyfluoroalkyl Substances in the Environment: Terminology, Classification, and
467 Origins. *Integr. Environ. Assess. and Manag*. **2011**, 7 (4), 513–541. DOI
468 10.1002/ieam.258.

469 (2) Lindstrom, A. B.; Strynar, M. J.; Libelo, E. L. Polyfluorinated Compounds: Past,
470 Present, and Future. *Environ. Sci. Technol*. **2011**, 45 (19), 7954–7961. DOI
471 10.1021/es2011622.

472 (3) Giesy, J. P.; Kannan, K. Global Distribution of Perfluorooctane Sulfonate in
473 Wildlife. *Environ. Sci. Technol*. **2001**, 35 (7), 1339–1342. DOI 10.1021/es001834k.

474 (4) Kannan, K.; Franson, J. C.; Bowerman, W. W.; Hansen, K. J.; Jones, P. D.; Giesy,
475 J. P. Perfluorooctane Sulfonate in Fish-Eating Water Birds Including Bald Eagles
476 and Albatrosses. *Environ. Sci. Technol*. **2001**, 35 (15), 3065–3070. DOI
477 10.1021/es001935i.

478 (5) Kannan, K.; Koistinen, J.; Beckmen, K.; Evans, T.; Gorzelany, J. F.; Hansen, K. J.;
479 Jones, P. D.; Helle, E.; Nyman, M.; Giesy, J. P. Accumulation of Perfluorooctane
480 Sulfonate in Marine Mammals. *Environ. Sci. Technol*. **2001**, 35 (8), 1593–1598.
481 DOI 10.1021/es001873w.

482 (6) Kannan, K.; Corsolini, S.; Falandysz, J.; Fillmann, G.; Kumar, K. S.; Loganathan,
483 B.G.; Mohd, M. A.; Olivero, J.; Van Wouve, N.; Yang, J. H.; Aldoust K. M.
484 Perfluorooctanesulfonate and Related Fluorochemicals in Human Blood from
485 Several Countries. *Environ. Sc. Technol*. **2004**, 38 (17), 4489–4495. DOI

10.1021/es0493446.

(7) Tomy, G. T.; Budakowski, W.; Halldorson, T.; Helm, P. A.; Stern, G. A.; Friesen, K.; Pepper, K.; Tittlemier, S. A.; Fisk, A. T. Fluorinated Organic Compounds in an Eastern Arctic Marine Food Web. *Environ. Sci. Technol*. **2004**, 38 (24), 6475–6481. DOI 10.1021/es049620g.

(8) Haukås, M.; Berger, U.; Hop, H.; Gulliksen, B.; Gabrielsen, G. W. Bioaccumulation of Per- and Polyfluorinated Alkyl Substances (PFAS) in Selected Species from the Barents Sea Food Web. *Environ. Pollut*. **2007**, 148 (1), 360–371. DOI 10.1016/j.envpol.2006.09.021.

(9) Conder, J. M.; Hoke, R. A.; De Wolf, W.; Russell, M. H.; Buck, R. C. Are PFCAs Bioaccumulative? A Critical Review and Comparison with Regulatory Criteria and Persistent Lipophilic Compounds. *Environ. Sci. Technol*. **2008**, 42 (4), 995–1003. DOI 10.1021/es070895g.

(10) Kelly, B. C.; Ikonomou, M. G.; Blair, J. D.; Surridge, B.; Hoover, D.; Grace, R.; Gobas. F. A. Perfluoroalkyl Contaminants in an Arctic Marine Food Web: Trophic Magnification and Wildlife Exposure. *Environ. Sci. Technol*. **2009**, 43 (11), 4037–4043. DOI 10.1021/es9003894.

(11) Tomy, G. T.; Pleskach, K.; Ferguson, S. H.; Hare, J.; Stern, G.; MacInnis, G.; Marvin, C. H.; Loseto, L. Trophodynamics of Some PFCs and BFRs in a Western Canadian Arctic Marine Food Web. *Environ. Sci. Technol*. **2009**, 43 (11), 4076–4081. DOI 10.1021/es900162n.

(12) *AMAP Assessment 2015: Temporal Trends in Persistent Organic Pollutants in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP): Oslo,

Norway, 2016; https://www.amap.no/documents/download/2866/inline.

(13) Olsen, G. W.; Burris, J. M.; Mandel, J. H.; Zobel, L. R. Serum Perfluorooctane Sulfonate and Hepatic and Lipid Clinical Chemistry Tests in Fluorochemical Production Employees. *J. Occup. Environ. Med*. **1999**, 41 (9), 799–806. DOI 10.1097/00043764-199909000-00012.

(14) Lau, C.; Anitole, K.; Hodes, C.; Lai, D.; Pfahles-Hutchens, A.; Seed, J. Perfluoroalkyl Acids: A Review of Monitoring and Toxicological Findings. *Toxicol. Sci*. **2007**, 99 (2), 366–394. DOI 10.1093/toxsci/kfm128.

(15) Letcher, R. J.; Bustnes, J. O.; Dietz, R.; Jenssen, B. M.; Jørgensen, E. H.; Sonne, C.; Verreault, J.; Vijayan, M. M.; Gabrielsen, G. W. Exposure and Effects Assessment of Persistent Organohalogen Contaminants in Arctic Wildlife and Fish. *Sci. Total Environ*. **2010**, 408 (15), 2995–3043. DOI 10.1016/j.scitotenv.2009.10.038.

(16) Sonne, C.; Bustnes, J. O.; Herzke, D.; Jaspers, V. L. B.; Covaci, A.; Halley, D. J.; Moum, T.; Eulaers, I.; Eens, M.; Ims, R. A.; Hanssen, S. A.; Erikstad, K. E.; Johnsen, T.; Schnug, L.; Rigét, F. F.; Jensen, A. L. Relationships between Organohalogen Contaminants and Blood Plasma Clinical–Chemical Parameters in Chicks of Three Raptor Species from Northern Norway. *Ecotoxicology and Environmental Safety*. **2010**, 73 (1), 7–17. DOI 10.1016/j.ecoenv.2009.08.017.

(17) Sonne, C.; Bustnes, J. O.; Herzke, D.; Jaspers, V. L.; Covaci, A.; Eulaers, I.; Halley, D. J.; Moum, T.; Ballesteros, M.; Eens, M.; Ims, R. A.; Hanssen, S. A.; Erikstad, K. E.; Johnsen, T. V.; Rigét, F. F.; Jensen, A. L.; Kjelgaard-Hansen, M. Blood Plasma Clinical–Chemical Parameters as Biomarker Endpoints for

532 Organohalogen Contaminant Exposure in Norwegian Raptor Nestlings. *Ecotoxicol.*
533 *Environ. Saf.* **2012**, 80, 76–83. DOI 10.1016/j.ecoenv.2012.02.012.
534 (18) Directive 2006/122/EC of the European Parliament and of the Council,
535 2006; http://eur-
536 lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2006:372:0032:0034:en:PDF.
537 (19) Stockholm Convention on Persistent Organic Pollutants (POPs*),* 2009;
538 https://www.wipo.int/edocs/lexdocs/treaties/en/unep-pop/trt_unep_pop_2.pdf.
539 (20) PFOA Stewardship Program, United States Environmental Protection
540 Agency (EPA), 2006;
541 https://www.regulations.gov/contentStreamer?documentId=EPA-HQ-OPPT-2006-
542 0621-0001&contentType=pdf.
543 (21) Wang, Z.; Cousins, I. T.; Scheringer, M.; Buck, R. C.; Hungerbühler, K.
544 Global Emission Inventories for C_4–C_{14} Perfluoroalkyl Carboxylic Acid (PFCA)
545 Homologues from 1951 to 2030, Part I: Production and Emissions from
546 Quantifiable Sources. *Environ. Int*. **2014**, 70, 62–75. DOI
547 10.1016/j.envint.2014.04.013.
548 (22) Place, B. J.; Field, J. A. Identification of Novel Fluorochemicals in Aqueous
549 Film-Forming Foams Used by the US Military. *Environ. Sci. Technol*. **2012**, 46
550 (13), 7120–7127. DOI 10.1021/es301465n.
551 (23) Weiner, B.; Yeung, L. W.; Marchington, E. B.; D'Agostino, L. A.; Mabury,
552 S. A. Organic Fluorine Content in Aqueous Film Forming Foams (AFFFs) and
553 Biodegradation of the Foam Component 6:2 Fluorotelomermercaptoalkylamido
554 Sulfonate (6:2 FTSAS)." *Environ. Chem*. **2013**, 10 (6), 486–493. DOI

10.1071/EN13128.

(24) Favreau, P.; Poncioni-Rothlisberger, C.; Place, B. J.; Bouchex-Bellomie, H.; Weber, A.; Tremp, J.; Field, J. A.; Kohler, M. Multianalyte Profiling of Per- and Polyfluoroalkyl Substances (PFASs) in Liquid Commercial Products. *Chemosphere*. **2017**, 171, 491–501. DOI 10.1016/j.chemosphere.2016.11.127.

(25) Moody, C. A.; Field, J. A.; Perfluorinated Surfactants and the Environmental Implications of Their Use in Fire-Fighting Foams. *Environ. Sci. Technol*. **2000**, 34 (18), 3864–3870. DOI 10.1021/es991359u.

(26) Kärrman, A.; Elgh-Dalgren, K.; Lafossas, C.; Møskeland, T. Environmental Levels and Distribution of Structural Isomers of Perfluoroalkyl Acids after Aqueous Fire-Fighting Foam (AFFF) Contamination. *Environ. Chem*. **2011**, 8 (4), 372-380. DOI 10.1071/EN10145.

(27) Baduel, C.; Paxman, C. J.; Mueller, J. F. Perfluoroalkyl Substances in a Firefighting Training Ground (FTG), Distribution and Potential Future Release. *J. Hazard. Mat*. **2015**, 296, 46–53. DOI 10.1016/j.jhazmat.2015.03.007.¨

(28) *Harstad/Narvik lufthavn, Evenes Undersøkelser av PFAS i jord, vann of biota med risikovurdering*; Sweco; Norconsult, 2015; https://avinor.no/globalassets/_konsern/miljo-lokal/miljorapporter/pfos-rapport-harstad-narvik-lufthavn-april-2015.pdf.

(29) Hansen, S.; Vestergren, R.; Herzke, D.; Melhus, M.; Evenset, A.; Hanssen, L.; Brustad, M.; Sandanger, T. M. Exposure to Per- and Polyfluoroalkyl Substances through the Consumption of Fish from Lakes Affected by Aqueous Film-Forming Foam Emissions — A Combined Epidemiological and Exposure Modeling

578 Approach. The SAMINOR 2 Clinical Study. *Environ. Int*. **2016**, 94, 272–282. DOI
579 10.1016/j.envint.2016.05.030.
580 (30) *Avinor, Miljøprosjektet DP 2, Miljøtekniske grunnundersøkelser*
581 *Harstad/Narvik Lufthavn Evenes*; Sweco; Cowi, 2013;
582 https://avinor.no/globalassets/_konsern/miljo-og-samfunn/pfos/2013-
583 harstad_narvik---dp2-miljotekniske-grunnundersokelser.pdf.
584 (31) Forskrift om begrensning i bruk av helse- og miljøfarlige kjemikalier og
585 andre produkter (produktforskriften); FOR-2004-06-01-922; Norwegian Ministry
586 of Climate and Environment: Oslo, 2004;
587 https://lovdata.no/dokument/SF/forskrift/2004-06-01-922.
588 (32) Ahrens, L.; Herzke, D.; Huber, S.; Bustnes, J. O.; Bangjord, G.; Ebinghaus,
589 R. Temporal Trends and Pattern of Polyfluoroalkyl Compounds in Tawny Owl
590 (*Strix Aluco*) Eggs from Norway, 1986−2009. *Environ. Sci. Technol*. **2011**, 45 (19),
591 8090–8097. DOI 10.1021/es103473v.
592 (33) Bustnes, J. O.; Bangjord, G.; Ahrens, L.; Herzke, D.; Yoccoz. N. G.
593 Perfluoroalkyl Substance Concentrations in a Terrestrial Raptor: Relationships to
594 Environmental Conditions and Individual Traits. *Environ. Toxicol. Chem*. **2015**, 34
595 (1), 184–191. DOI 10.1002/etc.2782.
596 (34) Routti, H.; Aars, J.; Fuglei, E.; Hanssen, L.; Lone, K.; Polder, A.; Pedersen,
597 Å. Ø.; Tartu, S.; Welker, J. M.; Yoccoz, N. G. Emission Changes Dwarf the
598 Influence of Feeding Habits on Temporal Trends of Per- and Polyfluoroalkyl
599 Substances in Two Arctic Top Predators. *Environ. Sci. Technol*. **2017**, 51 (20),
600 11996–12006. DOI 10.1021/acs.est.7b03585.

(35) Sun, J.; Bossi, R.; Bustnes, J. O.; Helander, B.; Boertmann, D.; Dietz, R.; Herzke, D.; Jaspers, V. L. B.; Labansen, A. L.; Lepoint, G.; Schultz, R.. Sonne, C.; Thorup, K.; Tøttrup, A. P.; Zubrod, J. P.; Eens, M.; Eulaers, I. White-Tailed Eagle (*Haliaeetus albicilla*) Body Feathers Document Spatiotemporal Trends of Perfluoroalkyl Substances in the Northern Environment. *Environ. Sci. Technol.* **2019**, 53 (21), 12744-12753 DOI 10.1021/acs.est.9b03514

(36) Hailer, F.; Helander, B.; Folkestad, A. O.; Ganusevich, S. A.; Garstad, S.; Hauff, P.; Koren, C.; Nygård, T.; Volke, V.; Vilà, V.; Ellegren, H.; Bottlenecked but Long-Lived: High Genetic Diversity Retained in White-Tailed Eagles upon Recovery from Population Decline. *Biol. Lett*. **2006**, 2 (2), 316–319. DOI 10.1098/rsbl.2006.0453.

(37) Halley, D. J. Golden and White-Tailed Eagles in Scotland and Norway: Coexistence, Competition and Environmental Degradation. *Br. Birds*. **1998**, 91 (5), 171-179.

(38) Helander, B.; Bignert, A.; & Asplund, L. Using Raptors as Environmental Sentinels: Monitoring the White-tailed Sea Eagle *Haliaeetus albicilla* in Sweden. Ambio. **2008**, 37(6), 425–431. DOI 10.1579/0044-7447(2008)37[425:uraesm]2.0.co;2

(39) Sletten, S.; Bourgeon, S.; Bårdsen, B. J.; Herzke, D.; Criscuolo, F.; Massemin, S.; Zahn, S.; Johnsen, T. V.; Bustnes, J. O. Organohalogenated Contaminants in White-Tailed Eagle (*Haliaeetus Albicilla*) Nestlings: An Assessment of Relationships to Immunoglobulin Levels, Telomeres and Oxidative Stress. *Sci. Total Environ*. **2016**, 539, 337–349. DOI

10.1016/j.scitotenv.2015.08.123.

(40) Løseth, M. E.; Briels, N.; Eulaers, I.; Nygard, T.; Malarvannan, G.; Poma, G.; Covaci, A.; Herzke, D.; Bustnes, J. O.; Lepoint, G.; Jenssen, B. M. and Jaspers, V. L. B. Plasma concentrations of organohalogenated contaminants in white-tailed eagle nestlings - The role of age and diet. *Environ. Pollut.* **2019**, 246, 527-534. DOI 10.1016/j.envpol.2018.12.028.

(41) Eriksson, U.; Roos, A.; Lind, Y.; Hope, K.; Ekblad, A.; & Kärrman, A. Comparison of PFASs contamination in the freshwater and terrestrial environments by analysis of eggs from osprey (*Pandion haliaetus*), tawny owl (*Strix aluco*), and common kestrel (*Falco tinnunculus*). *Environ. Res*. **2016**, 149, 40–47 DOI 10.1016/j.envres.2016.04.038

(42) Statistics – Norway: Norway Urban Population and Region 2017; https://www.ssb.no/befolkning/statistikker/beftett/aar/2017-12-19.

(43) Statistics – Avinor: Traffic Statistics; https://avinor.no/en/corporate/about-us/statistics/archive.

(44) Herzke, D.; Olsson, E.; Posner, S. Perfluoroalkyl and Polyfluoroalkyl Substances (PFASs) in Consumer Products in Norway – A Pilot Study. *Chemosphere*. **2012**, 88 (8), 980–987. DOI 10.1016/j.chemosphere.2012.03.035.

(45) Sinclair, E.; Kannan, K. Mass Loading and Fate of Perfluoroalkyl Surfactants in Wastewater Treatment Plants. *Environ. Sci. Technol*. **2006**, 40 (5), 1408–1414. DOI 10.1021/es051798v.

(46) McLachlan, M. S.; Holmström, K. E.; Reth, M.; Berger, U. Riverine Discharge of Perfluorinated Carboxylates from the European Continent. *Environ.*

Sci. Technol. **2007**, 41 (21), 7260–7265. DOI 10.1021/es071471p.

(47) Becker, A. M.; Gerstmann, S.; Frank, H. Perfluorooctane Surfactants in Waste Waters, the Major Source of River Pollution. *Chemosphere*. **2008**, 72 (1), 115–121. DOI 10.1016/j.chemosphere.2008.01.009.

(48) Hanssen, L.; Dudarev, A. A.; Huber, S.; Odland, J. Ø.; Nieboer, E.; Sandanger, T. M. 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.* **2013**, 447 (March), 430–437. DOI 10.1016/j.scitotenv.2013.01.029.

(49) R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. URL https://www.R-Project.Org/. (version 3.4.3). 2017. Eng. R Core Team (2017).

(50) Zuur, A.; Ieno, E. N.; Walker, N.; Saveliev, A. A.; Smith, G. M. *Mixed Effects Models and Extensions in Ecology with R;* Statistics for Biology and Health. Springer-Verlag: New York, 2009. www.springer.com/in/book/9780387874579.

(51) Pinheiro J, Bates D, DebRoy S, Sarkar D and R Core Team (2018). nlme: Linear and Nonlinear Mixed Effects Models. R package version 3.1-137, https://CRAN.R-project.org/package=nlme

(52) Burnham, K. P.; Anderson, D. R. *Model Selection and Multimodel Inference: A Practical Information-Theoretic Approach*. 2nd ed. Springer-Verlag: New York, 2002. www.springer.com/us/book/9780387953649.

(53) Buckland, S. T.; Burnham, K. P.; Augustin, N. H. Model Selection: An Integral Part of Inference. *Biometrics*. 1997, 53 (2), 603-618. DOI

10.2307/2533961.

(54) Anderson, D. R.; Burnham, K. P.; Thompson, W. L. Null Hypothesis Testing: Problems, Prevalence, and an Alternative. *J. Wild. Manage*. **2000**, 64 (4), 912–923. DOI: 10.2307/3803199.

(55) Bårdsen, B. J.; Fox, J. L. Evaluation of Line Transect Sampling for Density Estimates of Chiru Pantholops Hodgsoni in the Aru Basin, Tibet. *Wildlife Biol*. **2006**, 12 (1), 89–100. DOI 10.2981/0909-6396

(56) Næss, M. W.; Bårdsen, B. J.; Fauchald, P.; Tveraa, T. Cooperative Pastoral Production — the Importance of Kinship. *Evol. Hum. Behav.* **2010**, 31 (4), 246–258. DOI 10.1016/j.evolhumbehav.2009.12.004.

(57) Mazerolle, M. J. AICcmodavg: Model Selection and Multimodel Inference Based on (Q)AIC(c). R Package Version 2.1-1, 2017. https://cran.r-project.org/web/packages/AICcmodavg/index.html

(58) Bustnes, J. O.; Borgå K.; Erikstad, K. E.; Lorentsen, S. H.; Herzke, D. Perfluorinated, Brominated, and Chlorinated Contaminants in a Population of Lesser Black-Backed Gulls (*Larus Fuscus*). *Environ. Toxicol. Chem*. **2008**, 27 (6), 1383–1392. DOI 10.1897/07-473.

(59) Bustnes, J. O.; Erikstad, K. E.; Lorentsen, S. H.; Herzke, D. Perfluorinated and Chlorinated Pollutants as Predictors of Demographic Parameters in an Endangered Seabird. *Environ. Pollut.* **2008**, 156 (2), 417–424. DOI 10.1016/j.envpol.2008.01.028.

(60) Huber, S.; Warner, N. A.; Nygård, T.; Remberger, M.; Harju, M.; Uggerud, H. T.; Kaj, L.; Hanssen, L. A Broad Cocktail of Environmental Pollutants Found in

693 Eggs of Three Seabird Species from Remote Colonies in Norway: A Broad Cocktail
694 of Environmental Pollutants in Seabirds. *Environ. Toxicol. Chem*. **2015**, 34 (6),
695 1296–1308. DOI 10.1002/etc.2956.
696 (61) Prevedouros, K.; Cousins, I. T.; Buck, R. C.; Korzeniowski. S. H. Sources,
697 Fate and Transport of Perfluorocarboxylates. *Environ. Sci. Technol*. **2006**, 40 (1),
698 32–44. DOI 10.1021/es0512475.
699 (62) Ng, C. A.; Hungerbühler, K. Bioaccumulation of Perfluorinated Alkyl
700 Acids: Observations and Models. *Environ. Sci. Technol*. **2014**, 48 (9), 4637–4648.
701 DOI 10.1021/es404008g
702 (63) Shi, Y.; Vestergren, R.; Haugdahl Nost, T.; Zhou, Z.; Cai, Y. Probing the
703 Differential Tissue Distribution and Bioaccumulation Behavior of Per- and
704 Polyfluoroalkyl Substances of Varying Chain-Lengths? Isomeric Structures and
705 Functional Groups in Crucian Carp. *Environ. Sci. Technol*. **2018**, 52, 4592-4600.
706 DOI 10.1021/acs.est.7b06128
707 (64) Nøst, T. H.; Helgason, L. B.; Harju, M.; Heimstad, E. S.; Gabrielsen, G. W.;
708 Jenssen, B. M. Halogenated organic contaminants and their correlations with
709 circulating thyroid hormones in developing Arctic seabirds. *Sci. Total Environ*.
710 **2012**, 414, 248–256. DOI 10.1016/j.scitotenv.2011.11.051
711 (65) Custer, C. M.; Custer, T.W.; Delaney, R.;Dummer, P. M.; Schultz, S.;
712 Karouna-Renier, N. Perfluoroalkyl Contaminant Exposure and Effects in Tree
713 Swallows Nesting at Clarks Marsh, Oscoda, Michigan, USA. *Arch. Environ.*
714 *Contam. Toxicol*. **2019**, 77(1), 1-13. DOI 10.1007/s00244-019-00620-1

(66) Groffen, T.; Lasters, R.; Lopez-Antia, A.; Prinsen, E.; Bervoets, L.; Eens, M. Limited Reproductive Impairment in a Passerine Bird Species Exposed Along a Perfluoroalkyl Acid (PFAA) Pollution Gradient, *Sci. Total. Environ.* **2019**, 652, 718-728. DOI 10.1016/j.scitotenv.2018.10.273.

(67) Commission Regulation (EC) No 552/2009 of the European Parliament and of the Council, 2009; https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2009:164:0007:0031:EN:PDF.

(68) Wang, Z.; Cousins, I. T.; Scheringer, M.; Hungerbühler, K. Fluorinated Alternatives to Long-Chain Perfluoroalkyl Carboxylic Acids (PFCAs), Perfluoroalkane Sulfonic Acids (PFSAs) and Their Potential Precursors. *Environ. Int*. **2013**, 60, 242–248. DOI 10.1016/j.envint.2013.08.021.

(69) Land, M.; de Wit, C. A.; Bignert, A.; Cousins, I. T.; Herzke, D.; Johansson, J. H.; Martin, J. W. What Is the Effect of Phasing out Long-Chain per- and Polyfluoroalkyl Substances on the Concentrations of Perfluoroalkyl Acids and Their Precursors in the Environment? A Systematic Review. *Environ. Evid*. **2018,** 4 (3). DOI 10.1186/s13750-017-0114-y

(70) Route, W. T.; Russel, R. E.; Lindstrom, A. B., Strynar, M. J.; Key, R. L. Spatial and Temporal Patterns in Concentrations of Perfluorinated Compounds in Bald Eagle Nestlings in the Upper Midwestern United States. *Environ. Sci. Technol.* **2014**, 48, 6653-6660. DOI 10.1021/es501055d

(71) Li, L.; Liu, J.; Hu, J.; Wania, F. Degradation of Fluorotelomer-Based Polymers Contributes to the Global Occurrence of Fluorotelomer Alcohol and

738 Perfluoroalkyl Carboxylates: A Combined Dynamic Substance Flow and
739 Environmental Fate Modeling Analysis. *Environ. Sci. Technol.* **2017**, 51, 4461–
740 4470. DOI 10.1021/acs.est.6b04021
741 (72) Schenker, U.; Scheringer, M.; MacLeod, M.; Martin, J. W.; Cousins, I. T.;
742 Hungerbühler, K. Contribution of Volatile Precursor Substances to the Flux of
743 Perfluorooctanoate to the Arctic. *Environ. Sci. Technol*. **2008**, 42, 3710-3716. DOI
744 10.1021/es703165m
745 (73) Wania, F. A Global Mass Balance Analysis of the Source of
746 Perfluorocarboxylic Acids in the Arctic Ocean. *Environ. Sci. Technol.* **2007**, 41,
747 4529–4535. DOI 10.1021/es070124c
748 (74) Commission Regulation (EU) No 757/2010 of the European Parliament and
749 of the Council, 2010; https://eur-
750 lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2010:223:0029:0036:en:PDF.
751 (75) Ahrens, L.; Bundschuh, M. Fate and effects of poly- and perfluoroalkyl
752 substances in the aquatic environment: A review. *Environ. Toxicol. Chem*. **2014**, 33
753 (9), 1921–1929. DOI 10.1002/etc.2663
754 (76) Numata, J.; Kowalczyk, J.; Adolphs, J.; Ehlers, S.; Schafft, H.; Fuerst, P.;
755 Müller-Graf, C.; Lahrssen-Wiederholt, M.; Greiner, M. Toxicokinetics of Seven
756 Perfluoroalkyl Sulfonic and Carboxylic Acids in Pigs Fed a Contaminated Diet. *J.*
757 *Agric. Food Chem*. **2014**, 62(28), 6861–6870. DOI 10.1021/jf405827u
758 (77) Rotander, A.; Toms, L. M.; Aylward, L.; Kay, M.; Mueller, J. F. Elevated
759 levels of PFOS and PFHxS in firefighters exposed to aqueous film forming foam
760 (AFFF). *Environ. Int.* 2015, 82, 28-34. DOI 10.1016/j.envint.2015.05.005.

(78) Stock, N. L.; Furdui, V. I.; Muir, D. C.; Mabury, S. A. Perfluoroalkyl Contaminants in the Canadian Arctic: Evidence of Atmospheric Transport and Local Contamination. *Environ. Sci. Technol*. **2007**, 41 (10), 3529–3536. DOI 10.1021/es062709x.

(79) Butt, C. M.; Mabury, S. A.; Kwan, M.; Wang, X.; Muir, D. C. Spatial Trends of Perfluoroalkyl Compounds in Ringed Seals (*Phoca Hispida*) from the Canadian Arctic. *Environ. Toxicol. Chem*. **2008**, 27 (3), 542–553. DOI 10.1897/07-428.

(80) Pistocchi, A.; Loos, R. A Map of European Emissions and Concentrations of PFOS and PFOA. *Environ. Sci. Technol*. 2009, 43 (24), 9237–9244. DOI 10.1021/es901246d.

(81) Zhao, P.; Xia, X.; Dong, J.; Xia, N.; Jiang, X.; Li, Y.; Zhu, Y. Short- and long-chain perfluoroalkyl substances in the water, suspended particulate matter, and surface sediment of a turbid river. *Sci. Total Environ.* **2016**, 568, 57-65 DOI 10.1016/j.scitotenv.2016.05.221

(82) Ahrens, L.; Yamashita, N;. Yeung, L. W. Y.; Taniyasu, S.; Horii, Y.; Lam, P. K. S.; Ebinghaus, R. Partitioning Behavior of Per- and Polyfluoroalkyl Compounds Between Pore Water and Sediment in Two Sediment Cores from Tokyo Bay, Japan. *Environ. Sci. Toxicol.* **2009**, 43 (18) 6969-6975. DOI 10.1021/es901213s

(83) Ahrens, L.; Taniyasu, S.; Yeung, L. W. Y.; Yamashita. N.; Lam, P. K. S.; Ebinghaus, R. Distribution of polyfluoroalkyl compounds in water, suspended particulate matter and sediment from Tokyo Bay, Japan. *Chemosphere*. **2010**, 79, 266-272. DOI 10.1016/j.chemosphere.2010.01.045

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