*Environmental Toxicology and Chemistry*—Volume 41, Number 6—pp. 1508–1519, 2022
Received: 10 December 2021 | Revised: 12 January 2022 | Accepted: 16 March 2022


Environmental Toxicology

# Temporal Trends of Organochlorine and Perfluorinated Contaminants in a Terrestrial Raptor in Northern Europe Over 34 years (1986–2019)

Jan Ove Bustnes,[a,*] Bård-Jørgen Bårdsen,[a] Dorte Herzke,[b] Georg Bangjord,[c] Sophie Bourgeon,[d] Clementine Fritsch,[e] and Igor Eulaers[f]

[a]Norwegian Institute for Nature Research (NINA), The Fram Centre, 9296, Tromsø, Norway
[b]Norwegian Institute for Air Research (NILU), The Fram Centre, 9296, Tromsø, Norway
[c]Oddatunet, 7057, Jonsvatnet, Norway
[d]Department of Arctic and Marine Biology, UiT The Arctic University of Norway, 9037, Tromsø, Norway
[e]Chrono-environnement UMR 6249 CNRS/University of Franche-Comté, Besançon, France
[f]Norwegian Polar Institute, The Fram Centre, 9296, Tromsø, Norway

Abstract: Fourteen legacy organochlorine (OC) contaminants and 12 perfluoroalkyl substances (PFASs) were measured in eggs of tawny owls (*Strix alueco*) in central Norway (1986–2019). We expected OCs to have reached stable equilibrium levels due to bans, and that recent phase-out of some PFASs would have slowed the increase of these compounds. ∑OC comprised on average approximately 92% of the measured compounds, whereas ∑PFAS accounted for approximately 8%. However, whereas the ∑OC to ∑PFAS ratio was approximately 60 in the first 5 years of the study, it was only approximately 11 in the last 5 years. Both OC pesticides and polychlorinated biphenyls (PCBs) showed substantial declines over the study period (~85%–98%): hexachlorocyclohexanes and chlordanes seemed to be levelling off, whereas *p,p*′-dichlorodiphenyldichloroethylene (*p,p*′-DDE) and hexachlororbenzene (HCB), and most PCB congeners still seemed to decline at a more or less constant rate. While the concentration of perfluorooctane sulfonic acid (PFOS), the dominating PFAS, was reduced by approximately 43%, other perfluorinated sulfonates (PFSAs) showed only minor changes. Moreover, the median concentrations of seven perfluorinated carboxylic acids (PFCAs) increased approximately five-fold over the study period. Perfluorononanoic acid and perfluoroundecanoate acid, however, seemed to be levelling off in recent years. In contrast, perfluorododecanoic acid, perfluorodecanoate acid, perfluorotridecanoic acid, and perfluorotetradecanoic acid seemed to increase more or less linearily. Finally, perfluorooctanoic acid (PFOA) was increasingly likely to be detected over the study period. Hence, most legacy OCs and PFOS have not reached a lower threshold with stable background levels, and voluntary elimination of perfluoroalkyl carboxylates still has not resulted in declining levels in tawny owls in central Norway. *Environ Toxicol Chem* 2022;41:1508–1519. © 2022 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: Organochlorine; Polychlorinated biphenyls; Perfluorinated substances; Perfluorinated carboxylic acid; Tawny owl; Raptor; Norway

## INTRODUCTION

Long-term monitoring has documented that the environmental concentrations of most oganohalogenated contaminants (OHCs), notably lipophilic organochlorines (OCs), such as polychlorinated biphenyls (PCBs) and OC pesticides, declined strongly in biota following bans imposed in the 1970s and early 1980s (Bignert et al., 1998; Bustnes et al., 2007, 2011; Helander et al., 2002; Herbert et al., 1999; Loganathan & Kannan, 1994). However, our knowledge about the temporal trends for other classes of OHCs, such as the perfluoroalkyl substances (PFAS), is insufficient. Perfluoroalkyl substances are hydrophobic, and unlike the OCs they bind primarily to proteins. However, they tend to have similar properties to OCs in terms of environmental persistency, ability to bioaccumulate, and potential for adverse effects in biota (Hoff et al., 2005; Land et al., 2018;

This article contains online-only Supporting Information.
This is an open access article under the terms of the Creative Commons Attribution-NonCommercial License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited and is not used for commercial purposes.
* Address correspondence to Jan.Bustnes@nina.no
Published online 21 March 2022 in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.5331

Lau et al., 2004). These compounds, commonly used as processing additives in fluoropolymer production, were not considered a general environmental problem before the 1990s (Giesy & Kannan, 2001). High concentrations of some PFAS, especially perfluorooctane sulfonate (PFOS) in wildlife and humans, led to the voluntary phase-out of production by the 3M company, the leading manufacturer. Moreover, the Stockholm Convetion listed PFOS in its Annex B in 2009. Subsequently, several bird studies have found decreasing temporal trends of PFOS (Ahrens et al., 2011; Holmström et al., 2010; Jouanneau et al., 2020; Sun et al., 2019). Still, PFOS is used in many products, and some studies have failed to document declining trends (e.g., Park et al., 2021). Moreover, a voluntary stewardship program aimed at eliminating long-chained perfluoroalkyl carboxylates (PFCAs) from production was established by the US Environmental protection agency (US EPA, 2006). Nevertheless, PFCAs have continued to increase in terrestrial and marine birds until recently (Braune & Letcher, 2013; Jouanneau et al., 2020; Miller et al., 2015; Route et al., 2014; Sun et al., 2019). Hence, it is important to assess further whether different OHCs continue to show increasing or declining trends or if they have reached equilibrium thresholds. Although many factors influence the temporal trends of OHCs, such as diet and climate, the lack of trends over long periods indicates stable chronic background pollution levels.

Organochlorines and PFAS are the dominating organic contaminants in Europe, but they have different physiochemical properties. Hence, comparing the developmental trends for these compounds in the same sampling matrix (egg homogenates) will provide important information about expected threats from these contaminants in a rapidly changing environment.

Avian top predators are suitable for monitoring biomagnifying OHCs and have been used extensively in studies for more than 50 years (Gómez-Ramírez et al., 2014). In the present study, we measured OHCs in eggs of tawny owls (*Strix alueco*) at the northern distribution range in central Norway. Tawny owls are a sentinel for monitoring organic pollutants in terrestrial ecosystems because they are resident top-predators accumulating pollutants locally in their territories (Bustnes et al., 2007). Previous analyses of tawny owls in the region, between 1986 and 2009 (Ahrens et al., 2011; Bustnes et al., 2007, 2011, 2015), reported that both PCBs and OC pesticides declined strongly, especially in the late 1980s and early 1990s (Bustnes et al., 2007, 2011). For PFAS, the picture was more complex, with a decline for PFOS, but overall increases of PFCAs (Ahrens et al., 2011; Bustnes et al., 2015). The present study examined 10 additional years (2010–2019) for 14 OCs and 12 PFASs, providing a data series spanning over 34 years (breeding seasons). Based on the previous studies and emission statistics, we hypothesized that legacy OCs would be stabilizing over the last decade and that PFOS would continue to decline. For PFCAs, we predicted that previously documented increases would be levelling off. The annual collection of eggs and the large sample size ($n = 234$ for OCs and $n = 172$ for PFAS) enabled us to assess temporal trends with relatively high precision.

# MATERIALS AND METHODS

## Study area

The study was carried out in the area surrounding the city of Trondheim (63.42°N, 10.23°E) in Trøndelag County, Central Norway (Bustnes, Bårdsen, Bangjord, et al., 2013). Tawny owl eggs were collected annually between 1986 and 2019.

## Sample collection

More than 100 tawny owl nest boxes were deployed, and annually each nest box was visited twice, first in early April, shortly after egg laying, and then in the first half of May, when all nonhatched eggs were collected. Eggs were frozen shortly after collection (within hours). In this analysis, 234 eggs were selected for contaminants analysis. The number of eggs sampled per year varied between one (1990) and 23 (2004), except for 1988 and 2016, when no eggs were found (for more details of egg collections see Bustnes et al., 2007, 2011, 2015). In each year, we included only one egg per nest box in the analyses. The number of eggs analysed per year for each compound group can be found in Supporting Information, Table S1.

## Chemical analysis

All eggs were homogenized before the OHC analyses. Bustnes et al. (2007) provide information regarding the analyses of OCs in the eggs collected between 1986 and 2004. The National Veterinary Institute (in Oslo) did all the chemical analyses for this period. For 2005–2009, the Norwegian Institute for Air Research (NILU, in Tromsø) did the chemical analyses (Bustnes et al., 2011). In these analyses, we used National Institute of Standards and Technology (NIST) Standard Reference Material 1588b. Recovery was between 77% and 84%. To assess the repeatability of the analyses from the two laboratories, NILU reanalyzed pooled samples from the years 1987, 1991, 1993–1994, 1997, 1999, 2001, and 2003–2004. The deviation of the mean concentrations reported in the earlier study (Bustnes et al., 2007) with the mean of the recent samples was low, 16% ($R^2 = 0.69$) and 22.5% ($R^2 = 0.70$), for the PCBs and *p,p*′-dichlorodiphenyldichloroethylene (*p,p*′-DDE), respectively. The NILU carried out the OC analyses for the period 2010–2019 (see Hitchcock et al., 2019 for details).

The OC pesticides were α-, β-, γ-hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), chlordanes (oxychlordane and *trans*-nonachlor), and *p,p*′-DDE. However, OC pesticides were not measured for 2005–2009, except *p,p*′-DDE, which was measured for all years. The following PCB congeners were determined from both laboratories: CB-101, -99, -118, -138, -153, -180, and -187.

All analyses of PFAS were conducted by the NILU as described by Ahrens et al. (2011). Out of the 234 eggs, PFAS were measured in 172. The following PFAS were measured: perfluorooctane sulfonamide (PFOSA), perfluorooctane sulfonic acid (PFOS), perfluoroheptanesulfonic acid (PFHpS), perfluorohexanesulfonic acid (PFHxS), perfluorodecane sulfonate

 © 2022 The Authors

(PFDcS), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoate acid (PFDcA), perfluorododecanoic acid (PFDoA), perfluorotetradecanoic acid (PFTeA), perfluorotridecanoic acid (PFTriA), and perfluoroundecanoate acid (PFUnA). All 26 OHCs were measured in 145 eggs.

Nondetects were replaced by multiplying their respective limit of detection (LOD) by 0.5, and when the LOD differed between different laboratory analyses, the highest LOD was used in the statistical analyses. The frequency of detection (%) and LODs for each compound can be found in Table 1.

In our previous temporal trend analyses of OCs, we used lipid-normalized concentrations (Bustnes et al., 2007). In this analysis we used wet weight concentrations because we compared concentrations and trends between OCs and PFASs, and the latter are predominantly bound to proteins.

## Statistical analyses

We did the statistical analyses and plotting in R (R Core Team, 2020). All tests were two-tailed, and we rejected the null hypothesis at an $\alpha$ level of 0.05. We used the treatment contrast and Wald statistics to test if the estimated parameters differed significantly from 0. Our predictions could be operationalized into models based on the following predictors: (1) Time (with Year as a continuous variable; in the model output, this variable was centred [i.e., subtracting the average value]) and (2) its second-order polynomial (Year$^2$). The biological rationale behind this is that we have a priori expectations to believe that temporal trends (i.e., the effect of Year) take several forms (Bustnes, Bårdsen, Bangjord, et al., 2013; Sun et al., 2019). Consequently, we used models that included both simple linear effects of Year and nonlinear temporal trends, operationalized as Year$^2$, were included as predictors.

For each response, we rescaled and ranked each model ($i$) relative to the model with the lowest second-order Akaike's Information Criterion (AICc) value ($\Delta_i$ denotes this for model $i$). From this, we selected the simplest model (as judged by the model's estimated number of parameters) with a $\Delta_i \leq 1.5$ value (e.g., Anderson, 2008; Burnham & Anderson, 2002) using the AICcmodavg-package in R (Mazerolle, 2019). We speak of temporal trends if either, or both, the main effect of Year or Year$^2$ was statistically significant. We mainly refer to the figures

**TABLE 1:** Frequency of detection (%), limit of detection (LOD), and destcriptive statistics for concentrations (pg/g, wet wt) of organochlorine, pesticides, polychlorinated biphenyl, congeners, and perfluoroalkyl substances in tawny owl eggs[a]

| Compound | Frequency of detection (%) | LOD | 1986–2019 | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | | *N* | Mean | Median | SE | Min | Max |
| α-HCH | 74.63 | 80 | 205 | 421 | 150 | 53 | 13 | 6110 |
| β-HCH | 61.95 | 210 | 205 | 506 | 240 | 57 | 69 | 8160 |
| γ-HCH | 56.1 | 110 | 205 | 842 | 55 | 249 | 17 | 31 990 |
| HCB | 100 | 237 | 205 | 6276 | 3890 | 619 | 200 | 95 410 |
| Oxychlordane | 99.51 | 200 | 205 | 4476 | 2440 | 681 | 100 | 124 250 |
| *trans*-nonachlor | 95.12 | 150 | 205 | 1124 | 540 | 150 | 70 | 22 450 |
| *p,p'*-DDE | 98.29 | 200 | 234 | 143 508 | 88 707 | 16 746 | 100 | 3 268 210 |
| CB101 | 53.42 | 420 | 234 | 787 | 313 | 91 | 210 | 10 220 |
| CB99 | 98.72 | 420 | 234 | 4608 | 1646 | 799 | 103 | 113 090 |
| CB118 | 99.57 | 420 | 234 | 4764 | 2075 | 858 | 210 | 173 050 |
| CB138 | 100 | 300 | 234 | 24 579 | 12 713 | 2978 | 778 | 518 240 |
| CB153 | 100 | 420 | 234 | 49 358 | 25 220 | 6592 | 1754 | 1 159 180 |
| CB180 | 100 | 250 | 234 | 33 288 | 16 190 | 4717 | 1217 | 864 230 |
| CB187 | 100 | 400 | 234 | 13 648 | 9445 | 1157 | 780 | 199 280 |
| ∑PCB | | | 234 | 131 031 | 69 730 | 16 561 | 5310 | 2.935 560 |
| ∑OC | | | 205 | 304 159 | 185 687 | 37 477 | 9635 | 6 456 920 |
| PFOSA | 13.95 | 30 | 172 | 27 | 15 | 4 | 15 | 449 |
| PFOS | 100 | 70 | 172 | 10 509 | 6680 | 964 | 344 | 86 943 |
| PFHpS | 57.56 | 35 | 172 | 98 | 40 | 9 | 18 | 597 |
| PFHxS | 59.88 | 30 | 172 | 134 | 44 | 25 | 15 | 3400 |
| PFDcS | 41.86 | 104 | 172 | 151 | 52 | 22 | 28 | 2500 |
| PFOA | 20.93 | 30 | 172 | 31 | 15 | 3 | 15 | 351 |
| PFNA | 60.47 | 30 | 172 | 168 | 98 | 17 | 15 | 1481 |
| PFDcA | 86.63 | 30 | 172 | 396 | 315 | 31 | 15 | 3212 |
| PFDoA | 79.65 | 36 | 172 | 467 | 333 | 65 | 18 | 8350 |
| PFTeA | 59.3 | 41 | 172 | 330 | 143 | 48 | 19 | 5240 |
| PFTriA | 88.37 | 30 | 172 | 984 | 790 | 77 | 15 | 7078 |
| PFUnA | 82.56 | 30 | 172 | 543 | 460 | 38 | 15 | 3551 |
| ∑PFCA | | | 172 | 2918 | 2506 | 217 | 114 | 24 072 |
| ∑PFAS | | | 172 | 13 837 | 9406 | 1073 | 1195 | 90 388 |

[a]Data from central Norway, 1986–2019.

α-, β-, γ-HCH = α-, β-, γ-hexachlorocyclohexanes; CB 101-187 = polychlorinated biphenyl congeners 101–187; HCB = hexachlorobenzene; OC = organochlorine; *p,p'*-DDE = *p,p'*-dichlorodiphenyldichloroethylene; PCB = polychlorinated biphenyl; PFAS = perfluoroalkyl substance; PFCA = perfluorinated carboxylic acid; PFDcA = perfluorodecanoate acid; PFDcS = perfluorodecane sulfonate; PFDoA = perfluorododecanoic acid; PFHpS = perfluoroheptanesulfonic acid; PFHxS = perfluorohexanesulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFOSA = perfluorooctane sulfonamide; PFTeA = perfluorotetradecanoic acid; PFTriA = perfluorotridecanoic acid; PFUnA = perfluoroundecanoate.

© 2022 The Authors 

when we speak of the resulting trends, because models including polynomials may take various forms based on the sign and strengths of the estimates.

Finally, the different responses were treated differently depending on the number of samples in each analysis at or below the LOD by creating our own rule of thumb based on defining the LOD for each contaminant as its minimum value. First, we fitted linear models using the lm-function if >15% of samples were ≤LOD. Second, we used logistic models with a binary response, classifying each observation ($X_i$) as 0 if $X_i \leq$ LOD and 1 if $X_i >$ LOD, using the glm-function with a logistic-link assuming a binomial distribution (both the lm- and glm-functions are a part of the stats-package: see, e.g., Fox 2002; Zuur et al. 2009 for how these models are fitted to data). Third, we fitted a tobit-regression model using the vglm-function in the vgam-package (using the tobit-family and the contaminant-specific LOD as the lower limit: see Yee & Moler 2020 for how these models were specified) if between 15% and 50% of the samples were ≤LOD. The tobit-regression is a left-censored model meaning that whenever $X_i =$ LOD, the observation is, in reality, ≤LOD (even though $X_i \geq 0$) and were fitted similar as in Thomas et al. (2016). We fitted both the tobit- and lm-models to $\log_e$-transformed responses.

## RESULTS AND DISCUSSION

### OHC Profiles

Of the OHCs measured, the loads were dominated by OCs over PFASs ($\bar{x} = 91.6\% \pm 0.65$ SE vs. $8.4\% \pm 0.65$ SE, $n = 145$, respectively; Table 1 and Figure 1). *p,p*′-DDE was the dominating OC (i.e., in eggs where all 14 OCs were measured, $n = 205$), followed by PCBs ($\bar{x} = 50.3\% \pm 1.1$ SE vs. $43.9\% \pm 1.0$ SE of the ∑OC load, respectively; Figure 1A). The proportional relationship between *p,p*′-DDE and PCBs was similar to previous analyses (Bustnes et al., 2007), and the *p,p*′-DDE:PCB ratio ($\bar{x} = 1.47$, SD = 1.1) did not change significantly over the 34 years ($p = 0.43$). This suggests that the two compounds have declined relatively synchronously, although *p,p*′-DDE seemed to decline more rapidly in the early years (Bustnes et al., 2007). Similarly, there were no substantial changes in the proportion of other OC pesticides such as HCB, HCHs, and chlordanes, which on average made up 5.8% (SE = 0.26, $n = 205$) of the ∑OC across all years, varying from 4.7% (SE = 0.78, $n = 13$) to 6.8% (SE = 0.89, $n = 33$) in the first 5 years (1986–1990) and the last 5 years (2015–2019) of the present study, respectively (Figure 1B and Table 2).

Perfluorooctane sulfonic acid (PFOS) was the dominating PFAS, making up on average 68.3% (SE = 1.4, $n = 172$) of the ∑PFAS load across all years. However, this proportion changed throughout our study from a mean of 89.3% (SE = 2.3, $n = 10$) in the first 5 years of the study to 58.7% (SE = 2.5, $n = 32$) in the last 5 years (Figure 1). Moreover, there were significant changes in the proportional relationship between OCs and PFAS ($F_{1,143} = 49.6$, $p < 0.01$; Figure 1). In the first 5 years of the study, the ∑OC:∑PFAS ratio was 59.4 (SE = 10.2, $n = 10$) compared with only 11.4 (SE = 1.4, $n = 32$) in the last 5 years. Hence, ∑OC concentrations went from being approximately 60 to approximately 11 times higher than ∑PFAS in the earliest to the latest years (Table 2 and Figure 1) because OCs were declining faster than PFOS and the concentrations of long-chained PFCAs were increasing. Few temporal trend studies have compared the relative proportions of OCs and PFAS in the same samples. However, in marine birds such as white-tailed eagles (*Haliaeetus albicilla*) and lesser black-backed gulls (*Larus fuscus*), on the Norwegian Coast, PFOS concentrations were equally high or higher than PCBs (Bustnes et al., 2008; Sletten et al., 2016). Moreover, in chicks of terrestrial goshawks (*Accipiter nisus*), concentrations of PFOS were approximately 3.5 times higher than *p,p*′-DDE and PCB153 (Bustnes, Bårdsen, Herzke, et al., 2013). However, these studies measured OHCs in plasma, and how comparable these results are to the egg concentrations in the present study is unknown. A recent study from the Baltic Proper found that in eggs of marine birds PCBs and DDTs concentrations were orders of magnitude higher than those of PFASs (De Wit et al., 2020), whereas in eggs of terrestrial arctic barnacle goose (*Branta leucopsis*) mean ∑PFAS equalled ∑PCBs (Hitchcock et al., 2019). Thus it remains difficult to draw firm conclusions about the expected proportional relationship between OCs and PFASs in bird eggs because so few studies have compared these in the same samples and none, to our knowledge, on a long-term temporal scale.

### Temporal trends

Because OCs and PFASs are the dominating environmental contaminants in northern Europe and thus most likely to cause different negative impacts, it is essential to document the relative strength of these compound groups and how they change over time. All compounds subject to analysis in our study have been analyzed previously (Ahrens et al., 2011; Bustnes et al., 2007, 2011, 2015). However, whereas PCBs, *p,p*′-DDE and PFASs were analyzed between 1986 and 2009, other OC pesticides have only been analyzed until 2004. Hence, we added 15 additional years to the time series for the latter compounds.

The seven OC pesticides investigated declined strongly over time. For α- and β-HCH, oxychlordane and *trans*-nonachlor, the best models included the second-order polynomial, and we documented that the declines were levelling off. For γ-HCH, *p,p*′-DDE, and HCB the selected model only included the linear effect, suggesting steady declines of 7.3%–8.1% per year for these compounds (Figure 2, Table 2, Supporting information, Table S2 and S4). For α-HCH, the best model explained 84% of the variation in concentrations, and the median concentration in the last 5 years of our study was only 2% that of the first 5 years (Table 2). For the six other OC pesticides, the best models explained between 29% (γ-HCH) and 59% (β-HCH) of the variation (Figure 2 and Supporting Information Table S2). The median concentrations of these compounds declined between approximately 84% (*p,p*′-DDE) and approximately 98% (α-HCH) between the first ($n = 13$) and last ($n = 33$) 5 years of our study (Table 2).

The trends for the seven PCB congeners were similar to the OC pesticides, because they all declined strongly (Figure 3),

 © 2022 The Authors

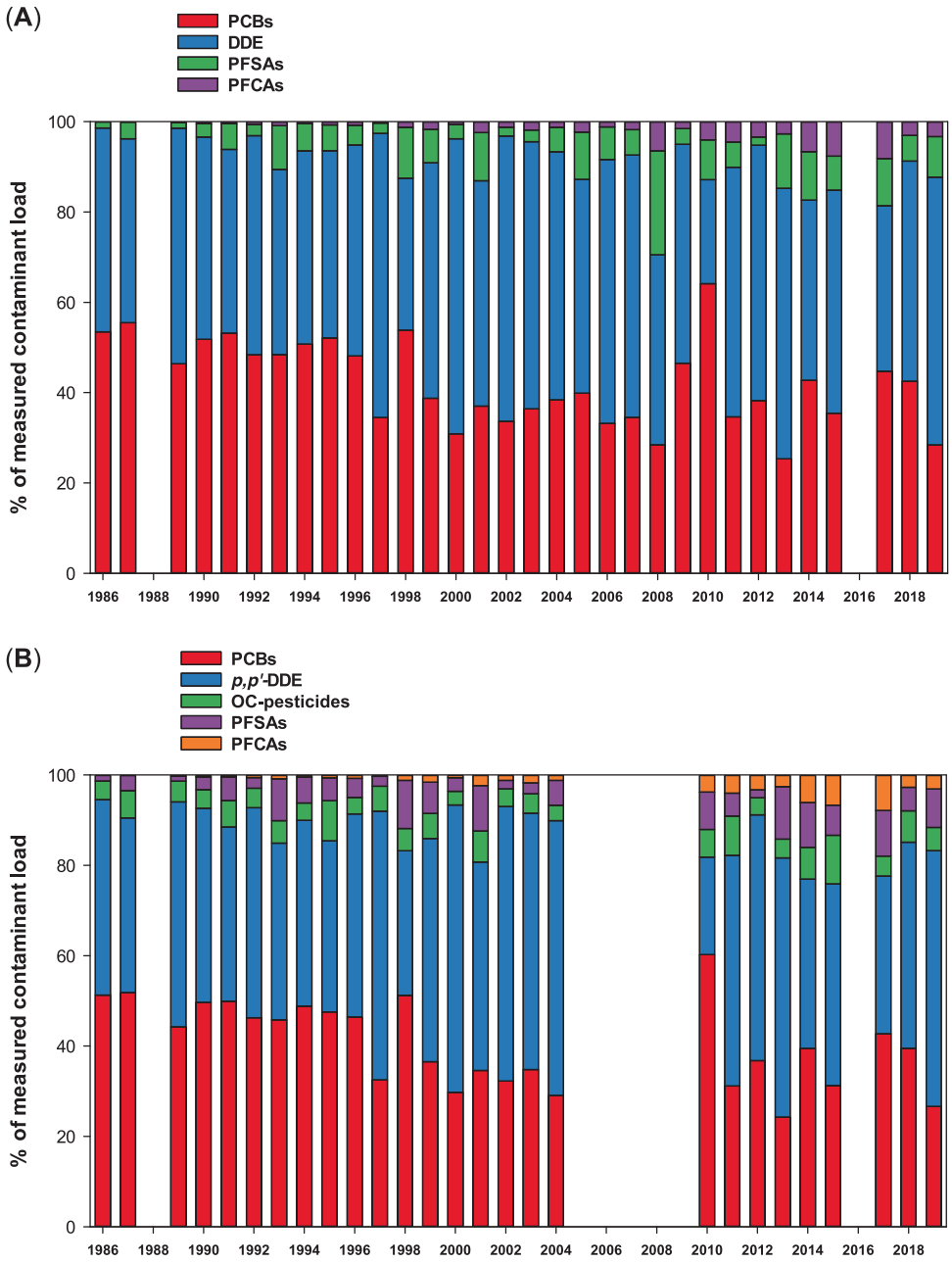


**FIGURE 1:** Proportional distribution of organochlorine contaminants (OCs) and perfluoroalkyl substances (PFAS) in tawny owl eggs. (**A**) All years included, showing the proportional relationships between *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE) and polychlorinated biphenyl (PCB) versus PFASs ($n = 172$). No eggs were collected in years with missing values. (**B**) All 26 compounds included ($n = 145$). Organochlorine pesticides, other than *p,p'*-DDE, were not measured in years with missing values. Data from central Norway, 1986–2019. PFSAs = perfluorinated sulfonates; PFCAs = perfluoroalkyl carboxylates.

between 7% and 11% per year (Supporting Information, Table S4). For CB101 and CB180, the best models included the second-order polynomial, suggesting a levelling off. For the others, the selected models only included a simple linear effect of year (Figure 3 and Supporting Information, Table S2). For all congeners, the median concentrations in the last 5 years of the present study were only between approximately 6% (CB153) and approximately 14% (CB101) of those in the first 5 years (Table 2). Hence, our analysis demonstrated that most OCs have continued to decline at a constant rate over the last 10 years. However, a closer examination of the graphs suggested an increasing trend over the last 3 years of our study (2017–2019). In fact, in 2019, most OC pesticides and PCBs concentrations were as high as the models predicted approximately for year 2000 (Figures 2 and 3). The importance of this is currently uncertain because these concentrations show high interannual variability. This makes it challenging for us to discriminate between noise (random fluctuations) and actual signals (i.e., changing trends) over just 3 years at the end of the time series. Nonetheless, this highlights the importance of

© 2022 The Authors 

**TABLE 2:** Mean and median concentrations (pg/g, wet wt) of organochlorine pesticides, polychlorinated biphenyl congeners, and perfluoroalkyl substances in tawny owl eggs during the first and last 5 years of a 34-year study[a]

| | 1986–1990 | | | | | | 2015–2019 | | | | | | | % change | | Fold change | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Compound | *N* | Mean | Median | SE | Min | Max | *N* | Mean | Median | SE | Min | Max | Trend | Mean | Median | Mean | Median |
| HCB | 13 | 18 909 | 14 760 | 6756 | 2050 | 95 410 | 33 | 2369 | 2051 | 162 | 991 | 5052 | – | 87.5 | 86.1 | 8.0 | 7.2 |
| α-HCH | 13 | 2415 | 2040 | 472 | 550 | 6110 | 33 | 51 | 40 | 9 | 34 | 309 | – | 97.9 | 98.0 | 47.4 | 51.0 |
| β-HCH | 13 | 2288 | 2310 | 577 | 240 | 8160 | 33 | 126 | 105 | 10 | 105 | 369 | – | 94.5 | 95.5 | 18.2 | 22.0 |
| γ-HCH | 13 | 1124 | 560 | 603 | 120 | 8160 | 33 | 373 | 55 | 293 | 17 | 9711 | – | 66.8 | 90.2 | 3.0 | 10.2 |
| Oxychlordan | 13 | 17 797 | 8390 | 9047 | 1240 | 124 250 | 33 | 1737 | 1212 | 206 | 397 | 5652 | – | 90.2 | 85.6 | 10.2 | 6.9 |
| *trans*-nonachlor | 13 | 4501 | 2640 | 1684 | 75 | 22 450 | 33 | 311 | 265 | 31 | 75 | 863 | – | 93.1 | 90.0 | 14.5 | 10.0 |
| *p,p′*-DDE | 13 | 57 3826 | 19 8290 | 24 0095 | 51 190 | 3 268 210 | 33 | 62 124 | 32 477 | 13 454 | 5949 | 42 3348 | – | 89.2 | 83.6 | 9.2 | 6.1 |
| CB101 | 13 | 3338 | 1560 | 997 | 210 | 10 220 | 33 | 221 | 210 | 9 | 210 | 503 | – | 93.3 | 86.5 | 15.1 | 7.4 |
| CB99 | 13 | 21 000 | 10 350 | 8454 | 1440 | 113 090 | 33 | 654 | 557 | 68 | 103 | 1611 | – | 96.9 | 94.6 | 32.1 | 18.6 |
| CB118 | 13 | 16 300 | 10 460 | 5336 | 1530 | 71 450 | 33 | 1242 | 1032 | 127 | 276 | 3287 | – | 92.4 | 90.1 | 13.1 | 5.2 |
| CB153 | 13 | 21 3448 | 15 2250 | 83 779 | 20 080 | 1 159 180 | 33 | 12 674 | 9551 | 1546 | 2107 | 38 925 | – | 94.1 | 93.7 | 16.8 | 15.9 |
| CB138 | 13 | 10 4375 | 64 220 | 37 292 | 11 380 | 518 240 | 33 | 6497 | 4903 | 857 | 778 | 20 349 | – | 93.8 | 92.4 | 16.1 | 13.1 |
| CB187 | 13 | 43 244 | 28 010 | 14 845 | 7990 | 199 280 | 33 | 6148 | 4522 | 700 | 1417 | 13 979 | – | 85.8 | 83.9 | 7.0 | 6.2 |
| CB180 | 13 | 15 5850 | 80 010 | 63 109 | 16 030 | 864 230 | 33 | 10 599 | 7693 | 1443 | 1611 | 33 601 | – | 93.2 | 90.4 | 14.7 | 10.4 |
| ∑PCB | 13 | 55 7555 | 41 6400 | 21 2008 | 59 190 | 2 935 560 | 33 | 38 036 | 29 646 | 4430 | 6531 | 97 456 | – | 93.2 | 92.9 | 14.7 | 14.0 |
| ∑OC | 13 | 11 78415 | 71 4660 | 46 7953 | 11 5280 | 6 456 920 | 33 | 10 5126 | 80 531 | 16 023 | 15 838 | 48 8456 | – | 91.1 | 88.7 | 11.2 | 8.9 |
| PFOSA | 10 | 41 | 15 | 26 | 15 | 273 | 32 | 15 | 15 | 0 | 15 | 15 | – | 63.4 | - | 2.7 | - |
| PFHxS | 10 | 59 | 15 | 21 | 15 | 196 | 32 | 144 | 33 | 49 | 15 | 1288 | + | 144.1 | 120.0 | 2.4 | 2.2 |
| PFHpS | 10 | 151 | 90 | 54 | 18 | 543 | 32 | 37 | 18 | 8 | 18 | 205 | – | 75.5 | 80.0 | 4.1 | 5.0 |
| PFOS | 10 | 16 651 | 8145 | 5757 | 2421 | 52 872 | 32 | 9086 | 4617 | 2377 | 794 | 63 408 | – | 45.4 | 43.3 | 1.8 | 1.8 |
| PFDcS | 10 | 72 | 52 | 12 | 52 | 157 | 32 | 126 | 52 | 49 | 52 | 1474 | | 75.0 | - | 1.8 | - |
| PFOA | 10 | 15 | 15 | 0 | 15 | 15 | 32 | 38 | 15 | 6 | 15 | 143 | + | 153.3 | - | 2.5 | - |
| PFNA | 10 | 15 | 15 | 0 | 15 | 15 | 32 | 159 | 147 | 18 | 15 | 476 | + | 960.0 | 880.0 | 10.5 | 9.8 |
| PFDcA | 10 | 195 | 200 | 55 | 15 | 427 | 32 | 416 | 318 | 63 | 15 | 1707 | + | 113.3 | 59.0 | 2.1 | 1.6 |
| PFUnA | 10 | 144 | 15 | 71 | 15 | 637 | 32 | 641 | 609 | 72 | 160 | 2122 | + | 345.1 | 3960.0 | 4.5 | 40.6 |
| PFDoA | 10 | 92 | 18 | 50 | 18 | 453 | 32 | 1042 | 548 | 316 | 124 | 8350 | + | 1032.6 | 2944.4 | 11.3 | 30.4 |
| PFTriA | 10 | 423 | 124 | 170 | 15 | 1548 | 32 | 1285 | 983 | 220 | 175 | 6257 | + | 203.8 | 692.7 | 3.0 | 7.9 |
| PFTeA | 10 | 70 | 21 | 49 | 21 | 515 | 32 | 798 | 523 | 212 | 19 | 5240 | + | 1040.0 | 2390.5 | 11.4 | 24.9 |
| ∑PFCA | 10 | 954 | 624 | 293 | 114 | 3197 | 32 | 4379 | 3445 | 866 | 79 | 24 072 | + | 359.0 | 452.1 | 4.6 | 5.5 |
| ∑PFAS | 10 | 17 886 | 9218 | 5989 | 2619 | 54 451 | 32 | 13 773 | 9021 | 3072 | 2045 | 70 103 | – | 23.0 | 2.1 | 1.3 | 1.0 |

[a]Data from central Norway, 1986–2019.

α-, β-, γ-HCH = α-, β-, γ-hexachlorocyclohexanes; HCB = hexachlorobenzene; OC = organochlorine; *p,p′*-DDE = *p,p′*-dichlorodiphenyldichloroethylene; PCB = polychlorinated biphenyl; PFAS = perfluoroalkyl substance, PFCA = perfluorinated carboxylic acid; PFDcA = perfluorodecanoate acid; PFDcS = perfluorodecane sulfonate; PFDoA = perfluorododecanoic acid; PFHpS = perfluoroheptanesulfonic acid; PFHxS = perfluorohexanesulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFOSA = perfluorooctane sulfonamide; PFTeA = perfluorotetradecanoic acid; PFTriA = perfluorotridecanoic acid; PFUnA = perfluoroundecanoate.

© 2022 The Authors

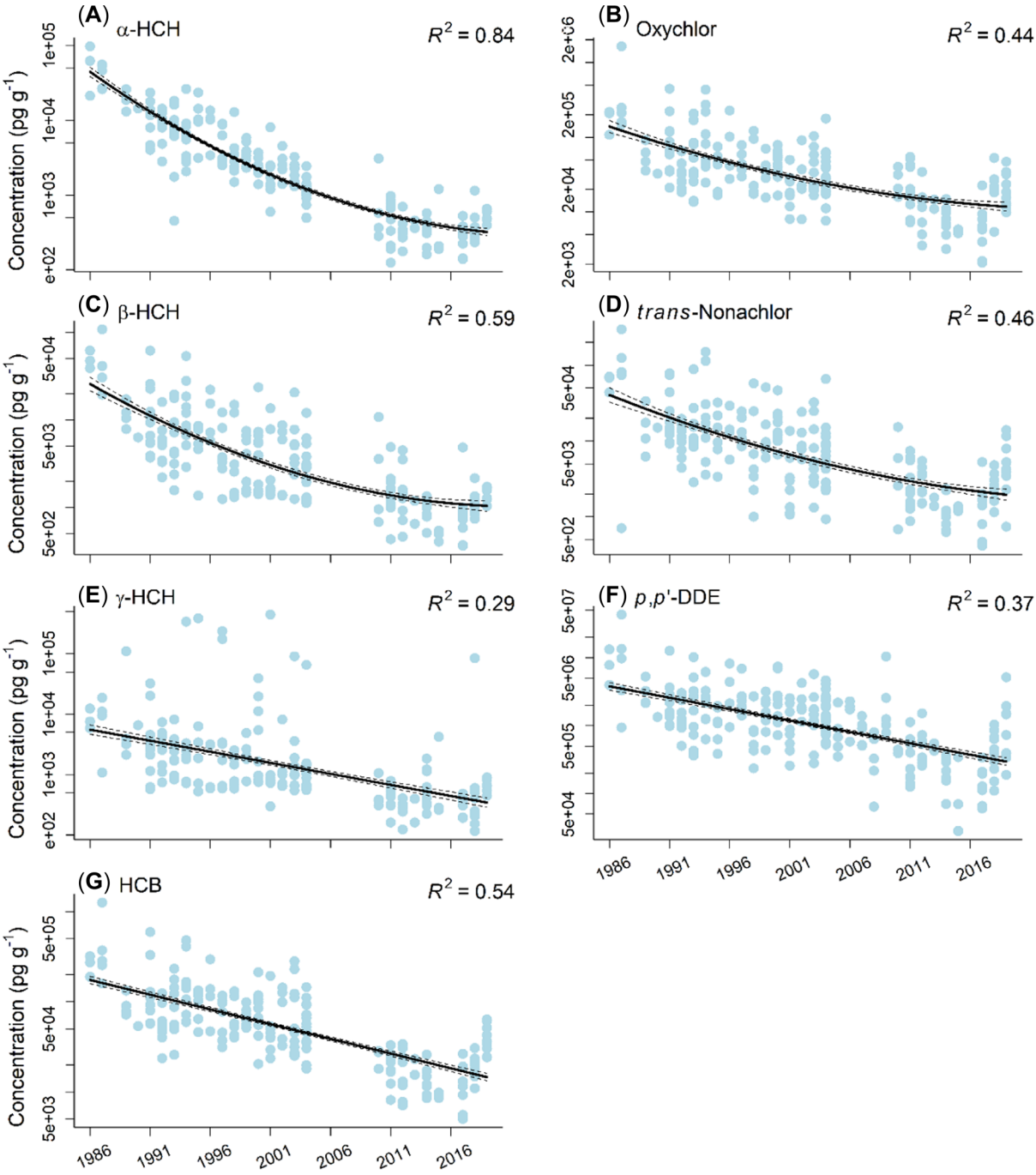


**FIGURE 2:** Temporal trends for concentrations (pg/g, wet weight) of different organochlorine contaminant (OC) pesticides, 1986–2019. Data from central Norway. HCH = hexachlorocyclohexane; *p*,*p*′-DDE = *p*,*p*′-dichlorodiphenyldichloroethylene; HCB = hexachlorobenzene.

continuous monitoring of this population because such questions are easily answered when we add a few more years to the data.

The temporal variations for the PFAS compounds differed from those of OCs. Ahrens et al. (2011) showed an annual decline of PFOS of 1.6% between 1986 and 2009, whereas the long-chained PFCAs increased by approximately 4% to approximately 12%. Our updated analysis still reports declines for PFOS and PFHpS, whereas there are no overall trends for the other perfluorinated sulfonates (PFSAs; Figure 4 and Supporting Information, Table S3). The median concentrations of PFOS in the last 5 years ($n = 32$) of our study were approximately 43% of the concentrations in the first 5 years ($n = 10$; Table 2). For PFOS, this analysis shows a decline of approximately 3.2% per year (Figure 4 and Supporting Information, Table S4), suggesting that the phase-out of PFOS production has had a similar impact on PFOS as the banning had on legacy OCs. However, PFOS is still used in some products, which may account for the relatively slower decline. The reported decline is in line with other studies from Scandinavia investigating both terrestrial and marine species (Holmström et al., 2010; Jouanneau et al., 2020; Sun et al., 2019). For both PFHxS and

© 2022 The Authors 

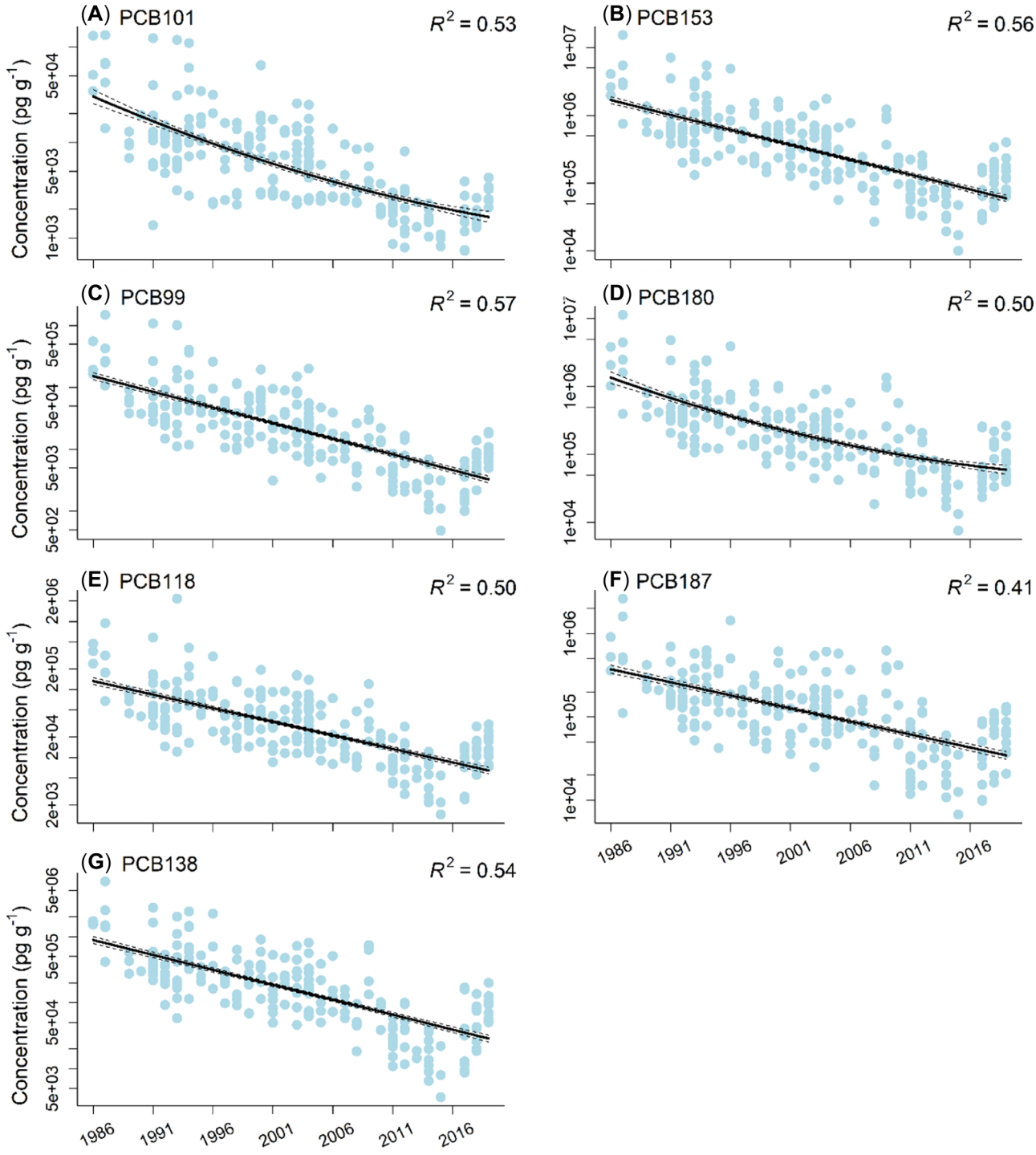


**FIGURE 3:** Temporal trends for concentrations (pg/g, wet weight) of different polychlorinated biphenyl (PCB) congeners, 1986–2019. Data from central Norway.

PFHpS, there were indications of a decline until 2009 (Ahrens et al., 2011), but only for PFHpS did this continue when including data for the last decade (Figure 4 and Supporting Information, Table S3). Moreover, PFOSA was very low in concentration and was less likely to be detected after 2005 (Figure 5 and Supporting Information, Table S3).

Most PFCAs increased, and the median concentrations of seven PFCAs ($\sum PFCA_7$) increased approximately five-fold over the study period (Table 2). However, for PFNA and PFUnA the second-order term (Year$^2$) was selected, suggesting that the increasing temporal trends seen in analyses for the first 25 years (Ahrens et al., 2011) levelled off in the last decade (Figure 6 and Supporting Information, Table S3). For PFDoA, Year$^2$ was also in the best model, but the trend seemed to be still increasing (Figure 6 and Supporting Information, Table S3). For the other three PFCAs measured, linear temporal trends suggested an increase between approximately 3% and 10% per year (Figure 6 and Supporting Information, Table S4) as

 © 2022 The Authors

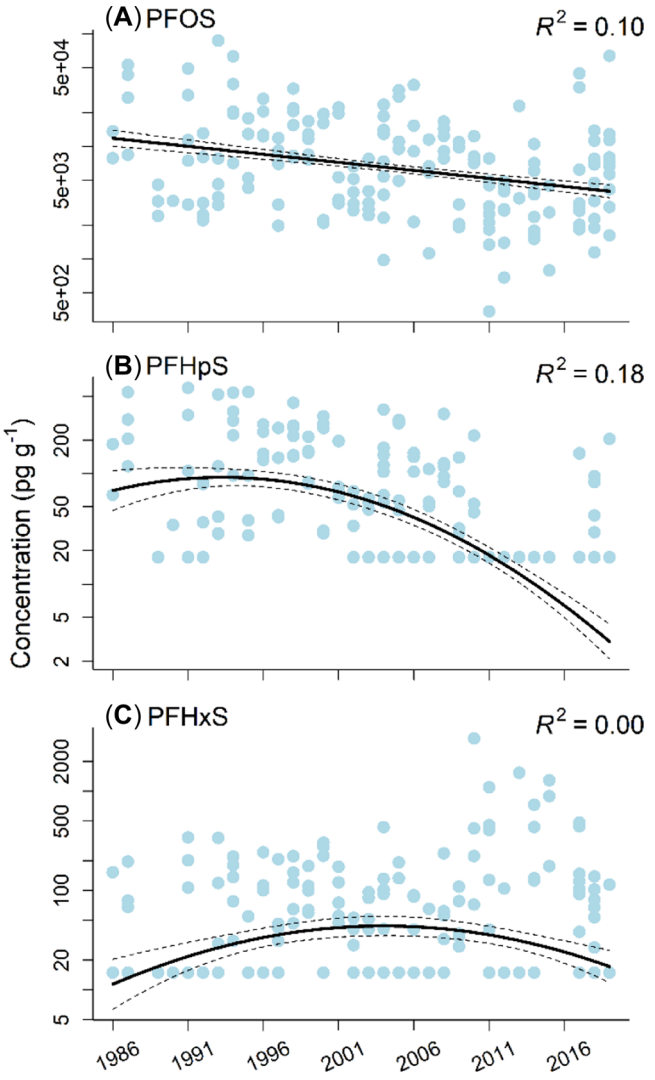


**FIGURE 4:** Temporal trends for concentrations (pg/g, wet weight) of different perfluorinated sulfonates (PFSA), 1986–2019. Data from central Norway. PFOS = perfluorooctane sulfonic acid; PFHpS = perfluoroheptanesulfonic acid; PFHxS = perfluorohexanesulfonic acid.

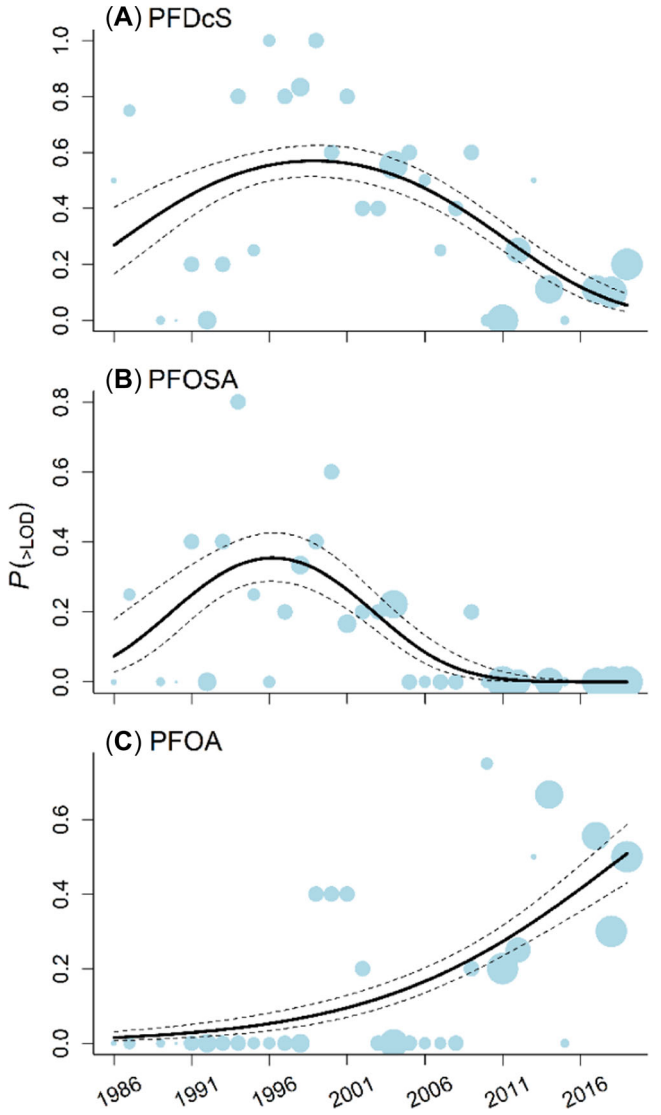


**FIGURE 5:** Temporal trends for concentrations (pg/g, wet weight) of perfluorodecane sulfonate (PFDcS), perfluorooctane sulfonamide (PFOSA), and perfluorooctanoic acid (PFOA), 1986–2019. Data from central Norway. Note that the analyses were done fitting logistic models to binary data predicting the probability of observations being ≥ limit of detection (LOD; 1) or not (0).

before (Ahrens et al., 2011). The median levels of PFDcA, PFDoA, PFTriA, and PFTeA were 1.6, 30, 8, and 25 times higher in the last 5 years compared to the first 5 years, respectively (Table 2). Ahrens et al. (2011) noted that PFTriA showed a tendency to level off between 2000 and 2009, but the additional data presented in our study did not support this (Figure 6). Finally, PFOA concentrations were low, but showed an increased probability of being detected in recent decades, that is, PFOA was not being detected in tawny owl eggs from our study area before the late 1990s (Figure 5 and Supporting Information, Table S3).

Ahrens et al. (2011) argued that the increasing PFCA concentrations could be caused by a production shift from perfluorooctane sulfonyl fluoride (POSF)-based products to fluorotelomer-based products, potential precursors for PFCAs whose production increased significantly after 2002. The global stewardship program, launched by the US Environmental Protection Agency (US EPA, 2006) and eight major fluoropolymer and telomer manufacturers in 2006, had a goal of reducing PFOA and related chemicals from facility emissions and product content by 95% until 2010, and to work toward the elimination of emissions and content by 2015 (Ahrens et al., 2011). However, our study has not shown evidence of any impact on PFCAs in tawny owls yet.

Previous studies have assumed that owls in general (Blus, 1996) and tawny owls in particular (Yoccoz et al., 2009) have a low risk for adverse effects of biomagnifying OCs, primarily because their mammalian prey contain low residues. Similarly, a Swedish study (Eriksson et al., 2016) showed that the tawny owl only had approximately 10% of the PFAS levels found in the aquatic fish-feeding osprey (*Pandion haliaetus*). Hence, the tawny owl may not be a relevant species to document the potential adverse effects of OHCs. Moreover, one potential

© 2022 The Authors 

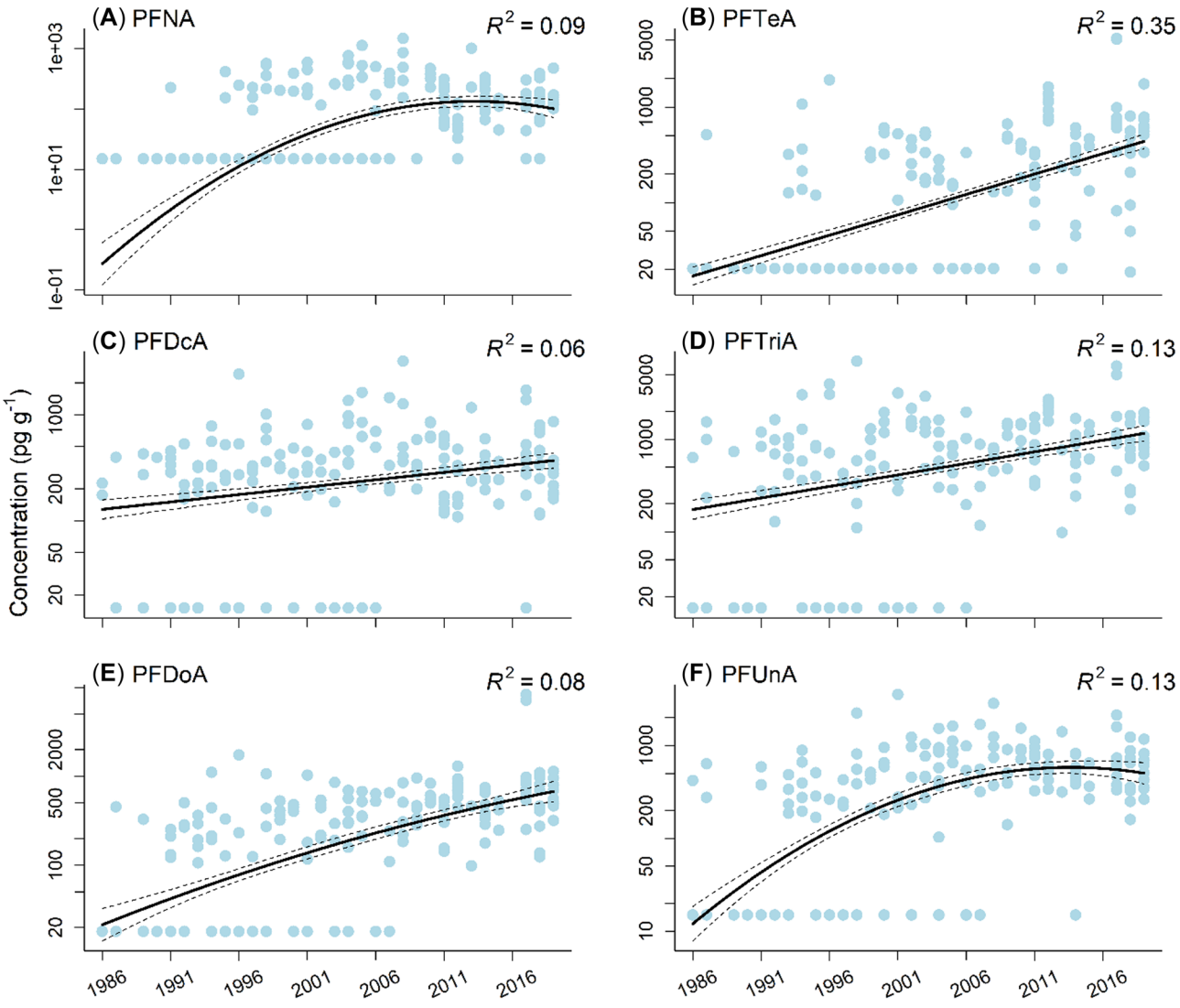


**FIGURE 6:** Temporal trends for concentrations (pg/g, wet weight) of different perfluoroalkyl carboxylates (PFCAs), 1986–2019. Data from central Norway. PFNA = perfluorononanoic acid; PFTeA = perfluorotetradecanoic acid; PFDcA = perfluorodecanoate acid; PFTriA = perfluorotridecanoic acid; PFDoA = perfluorododecanoic acid; PFUnA = perfluoroundecanoate acid.

weakness of the present study is that only abandoned eggs were collected. Our sample thus contained unfertilized eggs, addled eggs, and eggs in various stages of incubation. However, the state of embryo development does not seem to add much variation to persistent organic polluntant concentrations measured in eggs (Drouillard et al., 2003). In addition, features of the egg-laying female, such as possible diet specialization, may be important. The only way to reduce this effect would have been to sample eggs at the same incubation stage, for example freshly laid eggs. However, this may not be feasible in rare and threatened species such as raptors. Finally, to improve the understanding of how different abiotic and biotic environmental factors influence the accumulation of OHCs in terrestrial raptors, it will be a great advantage to measure OHCs in separate compartments of the food chain.

Because the tawny owl is a resident top predator, it is a precious sentinel species for documenting spatiotemporal distributions of contaminants and the processes that may influence the variation in OHC exposure (Ahrens et al., 2011; Bustnes et al., 2007, 2011, 2015; Eriksson et al., 2016). Hence, the present study provided important information about the recent development in OHC exposure in Scandinavia. The major strength of our data series is its high number of samples (234 eggs for OCs and 172 for PFAS) continuously collected over 34 years in the same area, allowing us to estimate the temporal changes in OHCs with relatively high precision. An important finding was that most legacy OCs are still declining 40–50 years after being banned, and a threshold where a stable chronic background level has not been reached for most of these compounds. Furthermore, we demonstrated that the previously observed decline in PFOS continued, suggesting that banning the use of these compounds efficiently reduced their environmental concentrations. However, this appears to have had little impact on the PFCAs so far, a global phenomenon reported for

 © 2022 The Authors

marine, freshwater, and terrestrial environments (Braune & Letcher, 2013; Holmström et al., 2010; Jouanneau et al., 2020; Route et al., 2014; Sun et al., 2019).

Because several of the compounds analyzed in our study are of major environmental concern and their temporal trends often are influenced by several climate-related factors (Bustnes et al., 2011, 2015), long-term monitoring is necessary to assess the threat of such compounds in a changing environment. In this respect, it is highly relevant to monitor the tawny owls in our study area because the long unbroken time series and high sample size allow better assessment of temporal trends.

*Supporting Information*—The Supporting Information are available on the Wiley Online Library at https://doi.org/10.1002/etc.5331.

*Acknowledgments*—The project has been funded by the Norwegian Research Council, Projects: EnviStress (No. 268482) and by the FRAM Centre Flagship Program "Hazardous substances–effects on ecosystems and human health" (project: Impacts of environmental contaminants and natural stressors on northern raptors: RAPTOR). We are very grateful to L. Ahrens for conducting some of the chemical analyses and to M. Gamain for valuable help with laboratory analyses. Finally, we thank two anonymous reviewers for comments that greatly improved the manuscript.

*Author Contribution Statement*—**Jan Ove Bustnes:** Conceptualization; Analyses; Writing—original draft. **Bård-Jørgen Bårdsen**: Conceptualization; Statistical analyses; Writing—review & editing. **Dorte Herzke**: Methodology; Chemical analyses; Writing—review & editing. **Georg Bangjord**: Methodology; Data collection. **Sophie Bourgeon**: Conceptualization; Writing—review & editing. **Clementine Fritsch**: Conceptualization; Writing—review & editing. **Igor Eulaers**: Conceptualization; Writing—review & editing.

*Data Availability Statement*—Data, associated metadata, and calculation tools are available from the corresponding author (Jan.Bustnes@nina.no).

## REFERENCES

Ahrens, L., Huber, S., Herzke, D., Bustnes, J. O., Bangjord, G., & Ebinghaus, R. (2011). Temporal trends of polyfluoroalkyl compounds in tawny owl (*Strix aluco*) eggs from Norway, 1986–2009. *Environmental Science and Technology*, *45*, 8090–8097.

Anderson, D. R. (2008). Model based inference in the life sciences: A primer on evidence. Springer Science.

Bignert, A., Olsson, M., Persson, W., Jensen, S., Zakrisson, S., Litzén, K., Erikson, U., Häggberg, L., & Alsberg, T. (1998). Temporal trends of organochlorines in northern Europe, 1967–1995. Relation to global fractionation, leakage from sediments and international measures. *Environmental Pollution*, *99*, 177–198.

Blus, L. J. (1996). Effects of pesticides on owls in North America. *Journal of Raptor Research*, *30*, 198–206.

Braune, B. M., & Letcher, R. J. (2013). Perfluorinated sulfonate and carboxylate compounds in eggs of seabirds breeding in the Canadian Arctic: Temporal trends (1975–2011) and interspecies comparison. *Environmental Science & Technology*, *47*, 616–624.

Burnham, K. P., & Anderson, D. R. (2002). *Model selection and multimodel inference: A practical information-theoretic approach* (2nd ed.). Springer, Inc.

Bustnes, J. O., Bangjord, G., Herzke, D., Ahrens, L., & Yoccoz, N. G. (2015). Perfluoroinated alkyl substance (PFAS) concentrations in a terrestrial raptor: Relationships to environmental conditions and individual traits. *Environmental Toxicology and Chemistry*, *34*, 184–191.

Bustnes, J. O., Bårdsen, B. J., Bangjord, G., Lierhagen, S., & Yoccoz, N. (2013). Temporal trends (1986–2005) of essential and non-essential elements in a terrestrial raptor in northern Europe. *Science of the Total Environment*, *458–460*, 101–106.

Bustnes, J. O., Bårdsen, B.-J., Herzke, D., Johnsen, T. V., Hanssen, S. A., Eulaers, I., Ballesteros, M., Covaci, A., Jaspers, V. L. B., Eens, M., Sonne, C., Halley, D. J., Moum, T., Erikstad, K. E., & Ims, R. A. (2013). Plasma concentrations of organohalogenated pollutants in predatory bird nestlings: Associations to growth rate and dietary tracers. *Environmental Toxicology and Chemistry*, *32*, 2520–2527.

Bustnes, J. O., Borgå, K., Erikstad, K. E., Lorentsen, S. H., & Herzke, D. (2008). Perfluorinated, brominated and chlorinated compounds in a population of lesser black-backed gulls (*Larus fuscus*). *Environmental Toxicology and Chemistry*, *27*, 1383–1392.

Bustnes, J. O., Yoccoz, N. G., Bangjord, G., Herzke, D., Ahrens, L., & Skaare, J. U. (2011). Impacts of climate and feeding condition on the annual accumulation (1986–2009) of persistent organic pollutants in a terrestrial raptor. *Environmental Science and Technology*, *45*, 7542–7547.

Bustnes, J. O., Yoccoz, N. G., Bangjord, G., Polder, A., & Skaare, J. U. (2007). Temporal trends (1986–2004) of organochlorines and brominated flame retardants in tawny owl eggs from northern Europe. *Environmental Science and Technology*, *41*, 8491–8497.

De Wit, C. A., Bossi, R., Dietz, R., Dreyer, A., Faxneld, S., Garbus, S. E., Hellstrom, P., Koschorreck, J., Lohmann, N., Roos, A., Sellstrom, U., Sonne, C., Treu, G., Vorkamp, K., Yuan, B., & Eulaers, I. (2020). Organohalogen compounds of emerging concern in Baltic Sea biota: Levels, biomagnification potential and comparisons with legacy contaminants. *Environment International*, *144*, 106037.

Drouillard, K. G., Nostrom, R. J., Fox, G. A., Gilman, A., & Peakall, D. B. (2003). Development and validation of a herring gull emryo toxicokinetic model for PCBs. *Ecotoxicology*, *12*, 55–68.

Eriksson, U., Roos, A., Lind, Y., Hope, K., & Ekblad, A. (2016). 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*). *Environmental Research*, *149*, 40–47.

Fox, J. (2002). *An R and S-PLUS companion to applied regression*. Sage Publications.

Giesy, J. P., & Kannan, K. (2001). Global distribution of perfluorooctane sulfonate in wildlife. *Environmental Science and Technology*, *35*, 1339–1342.

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. *Environment International*, *67*, 12–21.

Herbert, C. E., Norstrom, R. J., & Weseloh, D. V. C. (1999). A quarter century of environmental surveillance: The Canadian Wildlife Service's Great Lakes herring gull monitoring program. *Environmental Reviews*, *7*, 147–166.

Helander, B., Olsson, A., Bignert, A., Asplund, L., & Litzén, K. (2002). The role of DDE, PCB, coplanar PCB and eggshell parameters for reproduction in the white-tailed sea eagle (*Haliaeetus albicilla*) in Sweden. *Ambio*, *31*, 386–403.

Hitchcock, D. J., Andersen, T., Varpe, Ø., Loonen, M. J. J. E., Warner, N. A., Herzke, D., Tombre, I. M., Griffin, L. R., & Shimmings, P. (2019). Potential effect of migration strategy on pollutant occurrence in eggs of Arctic breeding barnacle geese (*Branta leucopsis*). *Environmental Science and Technology*, *53*, 5427–5435.

Hoff, P. T., Van de Vijver, K., Dauwe, T., Covaci, A., Maervoet, J., Eens, M., Blust, R., & De Coen, W. (2005). Evaluation of biochemical effects related to perfluorooctane sulfonic acid exposure in organohalogen-contaminated great tit (*Parus major*) and blue tit (*Parus caeruleus*) nestlings. *Chemosphere*, *61*, 1558–1569.

© 2022 The Authors 

Holmström, K., Johansson, A.-K., Bignert, A., Lindberg, P., & Berger, U. (2010). Temporal trends of perfluorinated surfactants in Swedish peregrine falcon eggs (*Falco peregrinus*), 1974–2007. *Environmental Science and Technology*, *44*, 4083–4088.

Jouanneau, W., Bårdsen, B.-J., Herzke, D., Johnsen, T. V., Eulaers, I., & Bustnes, J. O. (2020). Spatial and temporal analysis of PFASs in White-tailed eagle (*Haliaeetus albicilla*) nestlings of Northern Norway –a ten-year study. *Environmental Science and Technology*, *54*, 5011–5020.

Land, M., de Wit, C. A., Bignert, A., Cousins, I. T., Herzke, D., Johansson, J. H., & Martin, J. W. (2018). 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. *Environmental Evidence*, *7*, 4.

Lau, C., Butenhoff, J. L., & Rogers, J. M. (2004). The developmental toxicity of perfluoroalkyl acids and their derivatives. *Toxicology and Applied Pharmacolgy*, *198*, 231–241.

Loganathan, B. G., & Kannan, K. (1994). Global organochlorine contamination trends: An overview. *Ambio*, *23*, 187–191.

Mazerolle, M. J. (2019). *AICcmodavg: Model selection and multimodel inference based on (Q)AIC(c)*.

Miller, A., Elliott, J. E., Elliott, K. H., Lee, S., & Cyr, F. (2015). Temporal trends of perfluoroalkyl substances (PFAS) in eggs of coastal and offshore birds: Increasing PFAS levels associated with offshore bird species breeding on the Pacific coast of Canada and wintering near Asis. *Environmental Toxicology and Chemistry*, *34*, 1799–1808.

Park, K., Barghi, M., Lim, J. E., Ko, H. M., Nam, H. Y., Lee, S. I., & Moon, H. B. (2021). Assessment of regional and temporal trends in per- and polyfluoroalkyl substances using the Oriental Magpie (Pica serica) in Korea. *Science of the Total Environment*, *793*, 148513.

R Core Team. (2020). *R: A language and environment for statistical computing*. R Foundation for Statistical Computing, Vienna, Austria.

Route, W. T., Key, R. L., Russell, R. E., Lindstrom, A. B., & Strynar, M. J. (2014). Spatial and temporal patterns in concentrations of perfluorinated compounds in bald eagle nestlings in the upper Midwestern United States. *Environmental Science and Technology*, *48*, 6653–6660.

Sletten, S., Bourgeon, S., Bårdsen, B.-J., Criscuolo, F., Massemin, S., Johnsen, T. V., & Bustnes, J. O. (2016). Organohalogenated contaminants in white-tailed eagle (*Haliaeetus albicilla*) nestlings: An assessment of relationships to immunoglobulin levels, telomeres and oxidative stress. *Science of the Total Environment*, *539*, 337–349.

Sun, J., Bossi, R., Bustnes, J. O., Helander, B., Boertmann, D., Dietz, R., Herzke, D., Jaspers, V. L. B., Labansen, A. L., Lepoint, G., Schulz, R., Sonne, C., Thorup, K., Tøttrup, A., Zubrod, J., Eens, M., & Eulaer, I. (2019). White-tailed eagle (*Haliaeetus albicilla*) body feathers document spatiotemporal trends of poly- and perfluoroalkyl substances in the northern environment. *Environmental Science and Technology*, *53*, 12744–12753.

Thomas, M. G., Næss, M. W., Bårdsen, B.-J., & Mace, R. (2016). Smaller Saami Herding groups cooperate more in a public goods experiment. *Human Ecology*, *44*, 633–642.

U.S. Environmental Protection Agency. (2006). PFOA Stewardship Program, 2006, Docket EPA-HQ-OPPT-2006-0621.

Yee, T., & Moler, C. (2020). VGAM: Vector Generalized Linear and Additive Models.

Yoccoz, N. G., Bustnes, J. O., Bangjord, G., & Skaare, J. U. (2009). Reproduction and survival of tawny owls in relation to persistent organic pollutants. *Environment International*, *35*, 107–112.

Zuur, A. F., Ieno, E. N., Walker, N. J., Saveliev, A., & Smith, G. M. (2009). *Mixed effects models and extensions in ecology with R*. Springer.

 © 2022 The Authors