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Ecosystem specific accumulation of organohalogenated compounds: A comparison between adjacent freshwater and terrestrial avian predators

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ABSTRACT

Insight into processes determining the exposure of organohalogenated contaminants (OHCs) in wildlife might be gained from comparing predators in different ecosystems. This study compared two avian predator species with similar food chain lengths: the goldeneye duck (*Bucephala clangula*) and the tawny owl (*Strix aluco*) breeding in adjacent freshwater- and terrestrial ecosystems in central Norway. We measured lipophilic organochlorines (OCs) and protein-bound perfluorinated substances (PFASs) in eggs of the two species over 21 years (1999–2019). Across years, the proportional distribution of OCs (~90% of the ΣOHC load) relative to PFASs (~10%) was similar in the two species. Moreover, ΣOC concentrations were similar between the species, but PFAS compounds were 2–12 times higher in the goldeneyes than in tawny owls. OC-pesticides dominated in tawny owls (~60% of ΣOC), whereas persistent polychlorinated biphenyl (PCBs) congeners were the main OC components in goldeneyes (~70% of ΣOC). The lipid-normalized concentrations of most OC-pesticides and the less persistent PCB101 declined significantly in both species. Hexachlorobenzene (HCB), *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE), and more persistent PCBs decreased in tawny owls, while they tended to increase in goldeneyes. The increase in HCB was particulary robust. Among the PFASs, contrasted temporal trends were found across the species for four out of 11 compounds: PFOS declined while most perfluorocarboxylic acids (PFCAs) increased in tawny owls. In contrast, most PFASs were stable in goldeneyes. Moreover, there was no annual covariance between the OHC exposure in the two species: *i.e.,* high concentrations in one species in a given year did not translate into high concentrations in the other. Hence, the two avian predators in adjacent ecosystems seem to be subject to different processes determining the OHC exposure, probably related to variation in diet and climate, long-range transport of different contaminants, and emissions of pollution locally.

1. Introduction

Many organohalogenated contaminants (OHCs) are persistent, bioaccumulative, and have spread worldwide through long-range transport. Among the most widely distributed OHCs are 1) lipophilic organochlorines (OCs), notably polychlorinated biphenyls (PCBs) and organochlorine pesticides; 2) protein-bound perfluoroalkyl substances (PFASs). The contamination of OCs in the environment declined strongly after bans in the 1970s and 1980s (Hebert et al., 1999; Bignert et al., 1998; Helander et al., 2002; Bustnes et al., 2007), and some PFASs, such as perfluorooctane sulfonate (PFOS), have also decreased due to restrictions in production and use. However, other less regulated PFAS substances, including several perfluoroalkyl carboxylates (PFCAs), are still increasing in many regions (Ahrens et al., 2011; Miller et al., 2015; Eriksson et al., 2016; Sun et al., 2019; Jouanneau et al., 2020).

Exposure to biomagnifying OHCs in wildlife populations may differ among ecosystems. For example, marine predators tend to be more exposed to OHCs than their terrestrial and freshwater counterparts,

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Table 1
Concentrations (ng/g) of organohalogenated compound, organochlorines (OCs: lipid-normalized), and perfluorinated substances (PFASs: wet weight) in eggs of goldeneye and tawny owl. Data from Trøndelag, central Norway, 1999 and 2019.

Compound	Goldeneye						Tawny Owl						
	n	Mean	median	Std.Err	Min	Max	n	Mean	median	Std.Err	Min	Max	P-value
HCB	92	13.0	12.5	0.5	6.5	34.9	125	47.0	30.5	4.2	5.0	281.9	<0.0001
α-HCH	92	0.3	0.2	0.0	0.1	1.0	125	1.2	0.7	0.1	0.1	8.1	<0.0001
β-HCH	92	10.6	6.3	1.3	0.4	79.0	125	2.9	1.5	0.3	0.4	23.5	<0.0001
γ-HCH	92	0.3	0.3	0.0	0.2	1.5	125	6.9	0.7	3.7	0.1	446.2	<0.0001
Oxychlordane	92	17.4	14.6	1.3	3.3	71.4	125	27.7	20.6	2.1	2.1	177.4	<0.0001
trans-Nonachlor	92	9.0	6.9	0.7	0.3	28.5	125	6.9	3.8	0.8	0.4	65.9	<0.0001
p,p'-DDE	92	335.3	259.5	31.1	0.6	2202.0	154	1243.0	771.3	123.0	0.7	10424.5	<0.0001
PCB101	92	2.6	1.6	0.4	0.7	24.6	154	5.3	2.8	0.5	0.7	64.5	<0.0001
PCB99	92	67.6	34.4	13.7	0.9	1076.1	154	23.2	12.7	2.5	0.5	201.4	<0.0001
PCB118	92	195.9	96.9	39.7	12.4	2772.7	154	28.5	16.9	2.6	1.4	194.4	<0.0001
PCB153	92	509.3	341.2	80.0	41.1	6791.1	154	291.9	181.9	24.8	10.0	1756.6	<0.0001
PCB138	92	307.9	188.2	55.6	22.0	4351.2	154	158.1	98.7	13.9	3.7	904.8	<0.0001
PCB187	92	90.0	61.6	11.9	8.7	981.8	154	117.8	80.8	9.9	6.7	625.7	0.07890
PCB180	92	159.6	92.2	28.5	14.7	2522.8	154	195.0	127.9	17.4	7.6	1357.5	0.11520
ΣPCB	92	1332.8	857.1	216.9	104.2	17471.2	154	819.7	565.4	68.4	31.0	4619.1	<0.0001
ΣOC	92	1718.7	1257.0	234.1	228.2	18646.0	125	2068.1	1407.0	191.1	75.1	11097.3	0.59540
PFOSA	100	0.03	0.02	0.00	0.02	0.27	121	0.02	0.02	0.00	0.02	0.23	0.00140
PFHxS	100	0.43	0.26	0.06	0.02	3.29	121	0.15	0.04	0.03	0.02	3.40	<0.0001
PFHpS	100	0.23	0.09	0.04	0.02	2.39	121	0.07	0.02	0.01	0.02	0.38	<0.0001
PFOS	100	19.62	11.68	3.08	1.82	282.05	121	8.19	4.95	0.84	0.34	63.41	<0.0001
PFOA	100	0.42	0.24	0.06	0.02	3.71	121	0.04	0.02	0.00	0.02	0.35	<0.0001
PFNA	100	2.11	1.32	0.23	0.13	11.71	121	0.20	0.14	0.02	0.02	1.48	<0.0001
PFDcA	100	1.06	0.72	0.10	0.02	4.91	121	0.43	0.32	0.04	0.02	3.21	<0.0001
PFUnA	100	3.03	2.54	0.20	0.67	13.16	121	0.66	0.55	0.05	0.02	3.55	<0.0001
PFDoA	100	1.50	1.23	0.10	0.02	5.32	121	0.57	0.45	0.09	0.02	8.35	<0.0001
PFTriA	100	3.37	2.91	0.22	0.12	13.35	121	1.07	0.90	0.08	0.02	6.26	<0.0001
PFTeA	100	0.56	0.43	0.05	0.02	3.41	121	0.42	0.28	0.06	0.02	5.24	<0.0001
ΣPFAS	100	32.34	22.01	3.47	3.64	296.45	121	11.79	8.31	1.02	1.13	69.77	<0.0001

often attributed to the longer marine food chains (Dietz et al., 2000; Elliott et al., 2009; Eulaers et al., 2011; Bustnes et al., 2013). In addition, OHC exposure may fluctuate within wildlife populations over the years (Bignert et al., 1995; Helander et al., 2002; Elliott et al., 2012; Bustnes et al., 2005, 2011, 2015; Eriksson et al., 2016). The drivers of such variation are often poorly understood. Still, they may depend on alterations in local emissions or long-range transport, changes in feeding habits, and species-specific- or individual ability to eliminate different compounds (Hebert et al., 2000, 2008; Elliott et al., 2009, 2012; Noyes et al., 2009; Custer et al., 2010; Bustnes et al., 2011, 2015; Jouanneau et al., 2020). Most natural- and urban landscapes are patchily divided into different habitat types or ecosystems, both terrestrial- (forest, grassland, parks, etc.) and aquatic systems (streams, rivers, ponds, and lakes). Hence, an important question is how different drivers may impact the OHC exposure in such adjacent ecosystems. For example, OHCs, both of local- and long-transported origin, may spread more easily to- and within freshwater systems than terrestrial environments due to precipitation and subsequent drainage toward water bodies. Little is known about the importance of such processes in determining OHC exposure in freshwater and terrestrial food webs. A natural step toward increasing our understanding of the ecosystem-specific accumulation of OHC is to compare adjacent freshwater and terrestrial ecosystems over time. However, there is a paucity of long-term data from adjacent freshwater and terrestrial ecosystems suitable for comparisons of accumulation patterns and trends of OHCs. A novel approach to examine such relationships is to study predators at higher trophic levels, and in this study OHCs were measured in the terrestrial tawny owl (*Strix aluco*) and the aquatic goldeneye (*Bucephala clangula*) in adjacent terrestrial- and freshwater ecosystems in central Norway, over two decades. The tawny owl is a raptor that feeds predominantly on voles (sometimes shrew) and passerine birds (Cramp, 1985; Yoccoz et al., 2009; G. Bangjord unpublished data). The goldeneye is a diving duck breeding near freshwater lakes and rivers, mainly feeding on molluscs, crustaceans, and small fish (Cramp and Simmons, 1977). The feeding ecology of the two species thus suggests that their food chains are of similar length, predicting that the concentrations of OHCs in their eggs would be within the same order of magnitude (Dietz et al., 2000). The tawny owl is a territorial resident species. In contrast, the goldeneye is a short-distant migrant that often spends the winters in coastal areas, both in freshwater- and marine environments, and arrives at the breeding lakes when the ice breaks up in spring (Cramp and Simmons, 1977; Bakken et al., 2003; Clark et al., 2014). Hence, goldeneyes potentially include nutrient resources from the wintering areas in their eggs (Hobson et al., 2005). However, several studies have suggested that exogenous nutrients from freshwater breeding locations form the bulk of egg nutrients in most migrating ducks (Hobson, 2006; Bond et al., 2007; DeVink et al., 2011), indicating that this is a minor problem when comparing such species.

The objective of this study was to compare the long-term exposure to OHCs in avian predators in adjacent terrestrial- and freshwater ecosystems in the same landscape. The hypothesis that the same environmental processes play a similar role in explaining OHC exposure in both ecosystems provided a few testable predictions. Firstly, the concentrations and profiles of OHCs and their temporal trends should be similar for the predators in the freshwater and terrestrial ecosystem. Secondly, we predicted a high annual covariance in concentrations between the two species: *i.e.,* high levels in one species would mirror the levels in the other species in any given year. Eggs of tawny owls (n = 154) and goldeneyes (n = 100) were collected annually over two decades (1999–2019) and measured for 14 OCs and 11 PFASs.

2. Materials and methods

2.1. Study area and sampling

The study was carried out in the area surrounding the city of Trondheim (63.42° N, 10.23°E) in Trøndelag County, central Norway. Eggs of tawny owl and goldeneye were collected opportunistically between 1999 and 2019, a total of 21 seasons (available for both species for 19 of the 21 breeding seasons).

Table 2
Estimates from the selected models assessing temporal trends of OC-pesticides and PCB-congeners in tawny owl and goldeneye eggs from central Norway.

Compound	OC-pesticides						PCBs						
	Model Type	Parameter	Estimate	Std. Error	t/z	p	Compound	Model Type	Parameter	Estimate	Std. Error	t/z	p
HCB	LM	Intercept	7.865	0.048	162.803	<0.00001	PCB101	VGLM	Intercept 1	5.899	0.100	58.708	<0.00001
	LM	Year	0.005	0.007	0.732	0.465		VGLM	Intercept 2	-0.049	0.058	-0.847	0.397
	LM	Species (TO)	0.060	0.054	1.110	0.268		VGLM	Year	-0.085	0.017	-4.944	<0.00001
	LM	$Year^2$	0.003	0.001	3.193	0.00161		VGLM	Species (TO)	-0.830	0.144	-5.782	<0.00001
	LM	Year: Species	-0.056	0.008	-6.692	<0.00001		VGLM	Year: Species	-0.042	0.025	-1.728	0.084
	LM	R^2	0.317					VGLM	R^2	0.101			
α-HCH	GLM	Intercept	-2.852	0.734	-3.889	0.00010	PCB99	LM	Intercept	9.071	0.152	59.611	<0.00001
	GLM	Year	-0.730	0.164	-4.460	0.00001		LM	Year	-0.029	0.018	-1.608	0.109
	GLM	Species (TO)	3.408	0.779	4.378	0.00001		LM	Species (TO)	-2.373	0.202	-11.750	<0.00001
	GLM	Year: Species	0.454	0.169	2.677	0.0074		LM	$Year^2$	-0.003	0.003	-1.031	0.304
								LM	Year: Species	-0.072	0.022	-3.218	0.0015
								LM	$Year^2$: Species	0.009	0.004	2.318	0.021
								LM	R^2	0.530			
β-HCH	LM	Intercept	7.160	0.095	75.555	<0.00001	PCB118	LM	Intercept	9.965	0.098	101.813	<0.00001
	LM	Year	0.034	0.016	2.139	0.034		LM	Year	-0.008	0.014	-0.612	0.541
	LM	Species (TO)	-1.972	0.127	-15.513	<0.00001		LM	Species (TO)	-2.803	0.105	-26.668	<0.00001
	LM	Year: Species	-0.074	0.020	-3.674	0.0003		LM	$Year^2$	0.004	0.002	2.852	0.0047
	LM	R^2	0.541					LM	Year: Species	-0.058	0.017	-3.374	0.00086
								LM	R^2	0.748			
γ-HCH	GLM	Intercept	-1.711	0.421	-4.066	0.00005	PCB138	LM	Intercept	10.482	0.100	104.424	<0.00001
	GLM	Year	-0.419	0.086	-4.895	<0.00001		LM	Year	-0.005	0.014	-0.328	0.743
	GLM	Species (TO)	0.546	0.474	1.150	0.250		LM	Species (TO)	-1.718	0.108	-15.938	<0.00001
	GLM	Year: Species	0.340	0.092	3.709	0.0002		LM	$Year^2$	0.005	0.002	3.223	0.0014
								LM	Year: Species	-0.079	0.018	-4.454	0.00001
								LM	R^2	0.551			
Oxychlordan	LM	Intercept	8.092	0.066	123.030	<0.00001	PCB153	LM	Intercept	11.046	0.095	115.756	<0.00001
	LM	Year	-0.035	0.011	-3.197	0.0016		LM	Year	-0.001	0.013	-0.067	0.946
	LM	Species (TO)	-0.540	0.088	-6.120	<0.00001		LM	Species (TO)	-1.584	0.102	-15.454	<0.00001
	LM	Year: Species	0.005	0.014	0.380	0.705		LM	$Year^2$	0.005	0.002	3.231	0.0014
	LM	R^2	0.215					LM	Year: Species	-0.074	0.017	-4.412	0.00002
								LM	R^2	0.534			
trans-Nonachlor	LM	Intercept	7.330	0.106	69.373	<0.00001	PCB180	LM	Intercept	10.082	0.081	124.712	<0.00001
	LM	Year	-0.049	0.009	-5.390	<0.00001		LM	Year	-0.012	0.014	-0.864	0.389
	LM	Species (TO)	-1.489	0.118	-12.654	<0.00001		LM	Species (TO)	-0.780	0.104	-7.517	<0.00001
	LM	Year: Species	0.002	0.002	0.961	0.338		LM	Year: Species	-0.033	0.017	-1.958	0.051
	LM	R^2	0.464					LM	R^2	0.232			
p,p'-DDE	LM	Intercept	10.766	0.140	76.986	<0.00001	PCB187	LM	Intercept	9.567	0.079	121.255	<0.00001
	LM	Year	0.007	0.020	0.346	0.729		LM	Year	-0.011	0.013	-0.824	0.411
	LM	Species*	-0.080	0.150	-0.534	0.594		LM	Species (TO)	-0.748	0.101	-7.385	<0.00001
	LM	$Year^2$	0.005	0.002	2.343	0.020		LM	Year: Species	-0.031	0.017	-1.871	0.063
	LM	Year: Species	-0.092	0.025	-3.756	0.0002		LM	R^2	0.223			
	LM	R^2	0.119										

NOTE: A set of seven *a priori* based different candidate models were fitted with varying combinations of the following preditors: 1) Year (a continuous centred variable); 2) its second-order polynomial ($Year^2$); and 3) Species [factor variable with 'Goldeneye' (baseline) and 'Tawny owl' (TO) as levels]. In analyzing each contaminant, we selected a single model based on differences in the second-order Akaike's Information Criteria (AICc; see main text for details). The output differs slightly depending on whether our rule of thumb dictated us to fit simple linear models (i.e., an LM), a logistic model (i.e., a GLM with a binomial family and logit link to binary response), or a Tobit regression (i.e., a VGLM with a tobit-family and the contaminant-specific level of detection [LOD] as the lower-limit; see main text for details).

Table 3
Estimates from the selected models assessing temporal trends of different PFASs in tawny owl and goldeneye eggs from central Norway (see Table 2 for technical details).

Compound	Modell Type	Parameter	Estimate	Std. Error	t/z	p	Compound	Modell Type	Parameter	Estimate	Std. Error	t/z	p
PFOSA	GLM	Intercept	−1.955	0.467	−4.183	0.00003	PFOA	LM	Intercept	5.406	0.101	53.749	<0.00001
	GLM	Year	−0.519	0.173	−3.000	0.0027		LM	Species (TO)	−2.253	0.136	−16.572	<0.00001
	GLM	Species (TO)	−1.430	0.471	−3.035	0.0024		LM	R^2	0.556			
	GLM	$Year^2$	−0.025	0.019	−1.308	0.191	PFNA	LM	Intercept	7.312	0.138	53.130	<0.00001
PFHpS	LM	Intercept 1	4.474	0.157	28.577	<0.00001		LM	Year	0.047	0.013	3.583	0.0004
	LM	Intercept 2	0.217	0.056	3.862	0.0001		LM	Species (TO)	−2.578	0.154	−16.790	<0.00001
	LM	Year	−0.026	0.021	−1.225	0.221		LM	$Year^2$	−0.004	0.002	−1.720	0.087
	LM	Species (TO)	−1.502	0.178	−8.418	<0.00001		LM	R^2	0.573			
	LM	$Year^2$	0.007	0.003	2.583	0.0098							
	LM	Year: Species	−0.141	0.030	−4.759	<0.00001	PFDcA	LM	Intercept	6.742	0.130	51.851	<0.00001
	LM	R^2	0.101					LM	Year	0.032	0.012	2.562	0.011
								LM	Species (TO)	−1.009	0.145	−6.954	<0.00001
PFHxS	LM	Intercept	5.572	0.117	47.531	<0.00001		LM	$Year^2$	−0.005	0.002	−2.178	0.030
	LM	Species (TO)	−1.598	0.158	−10.087	<0.00001		LM	R^2	0.207			
		R^2	0.317				PFUnA	LM	Intercept	7.834	0.084	93.480	<0.00001
PFOS	LM	Intercept	9.270	0.110	84.659	<0.00001		LM	Species (TO)	−1.658	0.113	−14.640	<0.00001
	LM	Year	0.001	0.015	0.082	0.934		LM	R^2	0.495			
	LM	Species (TO)	−0.848	0.122	−6.926	<0.00001							
	LM	$Year^2$	0.005	0.002	2.676	0.008	PFDoA	LM	Intercept	7.083	0.094	75.166	<0.00001
	LM	Year: Species	−0.039	0.020	−1.944	0.053		LM	Year	0.017	0.016	1.080	0.281
	LM	R^2	0.228					LM	Species (TO)	−1.401	0.129	−10.873	<0.00001
								LM	Year: Species	0.066	0.021	3.125	0.002
								LM	R^2	0.391			
							PFTriA	LM	Intercept	7.939	0.096	82.630	<0.00001
								LM	Year	−0.004	0.016	−0.226	0.822
								LM	Species (TO)	−1.458	0.131	−11.100	<0.00001
								LM	Year: Species	0.052	0.021	2.407	0.017
								LM	R^2	0.368			
							PFTeA	LM	Intercept	6.033	0.109	55.374	<0.00001
								LM	Year	0.017	0.018	0.912	0.363
								LM	Species (TO)	−1.021	0.149	−6.851	<0.00001
								LM	Year: Species	0.095	0.024	3.894	0.0001
									R^2	0.280			

Bustnes et al. (2007) provide information about the collection of tawny owl eggs. In short, more than 100 tawny owl nest boxes were deployed, and annually each nest box was visited twice: 1) in early April, shortly after egg laying; 2) in the first half of May when non-hatched eggs were collected and frozen shortly after ($n = 154$). Eggs of goldeneyes were collected at Litlvantnet (1.6 km^2), a section of lake Jonsvatnet (63.22° N, 10.35°E:15 Km^2). The breeding population of goldeneyes at Jonsvatnet has varied over the study period and was about 55 pairs at the start of the study, whereas at present, it is about 35 pairs. Over the years, the number of nest boxes deployed at the lakeshore of Litlvantnet has varied, the maximum number being 37, whereas the present number is 17. Goldeneye usually arrives at Litlvatnet in March, and egg-laying starts when night temperatures exceed 0 °C (Clark et al., 2014; G. Bangjord, unpublished data). Nest boxes were checked daily from late March, and freshly laid eggs were collected and frozen. Five goldeneye eggs were analyzed annually except in 2017 when a freezer breakdown destroyed the samples. The 1999–2009 tawny owl data have been used in previous publications (Bustnes et al., 2007, 2011, 2015), whereas our data from tawny owl eggs between 2010 and 2019, and the data on goldeneye are novel.

2.2. Chemical analysis

Bustnes et al. (2007) describe details regarding the analyses of OCs in tawny owl eggs collected between 1999 and 2004, when the National Veterinary Institute (NVI) in Oslo did the analyses. For 2005–2019, the tawny owl eggs, and the complete sample of goldeneye eggs, were analyzed at the Norwegian Institute for Air Research (NILU) in Tromsø; methods as described in Hitchcock et al. (2019). For these latter analyses, the reference fish tissue EDF-2524 by Cambridge isotope laboratories was used. Blank samples were prepared for every batch, showing no contamination for most OCs, except for HCB, oxychlordane, and PCB153, showing blank contamination of 45, 13, and 46 pg/g. We thus did not perform blank correction due to the very low nature of blank contributions. The Limit of Detection (LOD) was determined by either applying three times the signal (noise) ratio or, in the case of blank

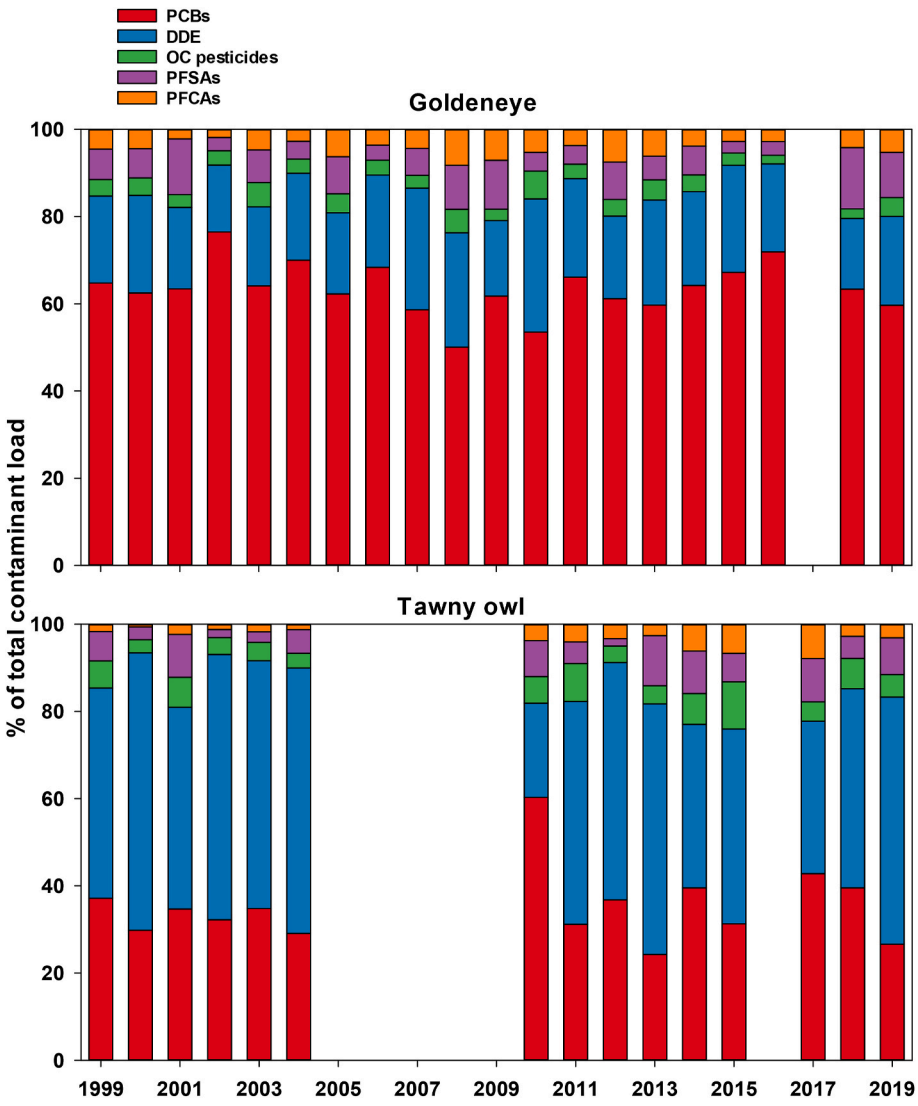

Fig. 1. Proportion (based on wet weight concentrations) of the organohalogenated compound (OHC) load comprised of different organochlorines (PCBs, *p,p'*-DDE and OC-pesticides) and perfluorinated substances (PFSAs and PFCAs) in different years in goldeneye (n = 100; 5 eggs per year) and tawny owl (n = 94; 2–10 eggs per year). Years where columns are missing indicate a complete lack of data, or that not all compounds were analyzed in those years. Data from central Norway, 1999, and 2019.

contribution, using the addition of the average concentration to three times the standard deviation. To ensure that the analyses from the two laboratories were comparable, the NILU reanalyzed pooled samples from the years 1987, 1991, 1993, 1994, 1997, 1999, 2001, 2003, and 2004 and compared the results with the mean concentrations reported in the earlier study (Bustnes et al., 2007). The concentrations were highly correlated (r = 0.986), when comparing the mean concentrations from NVI for these years with the mean concentrations from the NILU pools (Bustnes et al., 2011). This suggests a high degree of repeatability for these measures between the two laboratories. The following PCB-congeners were determined from both laboratories; PCB-101, -99, −118, −153, −138, −187, −180. The OC pesticides were hexachlorobenzene (HCB), α-, β-, γ-hexachlorocyclohexanes (HCHs), chlordanes (oxychlordane and *trans*-Nonachlor), and *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE).

Ahrens et al. (2011) describe the PFAS analyses of in tawny owl eggs collected between 1986 and 2009. PFAS analyses of goldeneye- and tawny owls eggs between 2010 and 2019, are as described in Hitchcock et al. (2019). The following PFASs were measured: perfluorooctane sulfonic acid (PFOS), perfluorooctane sulfonamide (PFOSA), perfluoroheptanesulfonic acid (PFHpS), perfluorohexanesulfonic acid (PFHxS), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluoroundecanoate acid (PFUnA), perfluorododecanoic acid (PFDoA), perfluorodecanoate acid (PFDcA), perfluorotridecanoic acid (PFTriA), and perfluorotetradecanoic acid (PFTeA).

We substituted all values below LOD with LOD/2. When LOD differed (*i.e.,* due to analyses being carried out at different times or laboratories), we used the highest LOD for all the samples.

Sample sizes varied in different analyses. For goldeneyes, we used five eggs per year (total of 100) for OHCs, but due to lack of lipid measurements in eight eggs, the sample size for temporal trends of lipophilic OCs was only 92. Similarly, 154 tawny owl eggs were processed, but for 2005–2009, OC-pesticides, other than *p,p'*-DDE, were not analyzed. Hence, the sample size was 125 eggs for these compounds. Moreover, we measured 121 tawny owl eggs for PFAS, and 94 of these eggs were analyzed for both OCs and PFASs, which we used when comparing the profiles of the OHC loads in the two species.

2.3. Statistical analyses

We performed statistical analyses and plotting of results in R (R Core Team, 2020). All tests were two-tailed, and we rejected the null hypothesis at an α-level of 0.05. To compare the temporal trends of OHCs between the species, we used the treatment contrast (where all other levels measure change compared to the baseline: i.e., the factor's first level) and Wald statistics to test if estimated parameters were significantly different from zero. Our predictions were operationalized by defining models that contained the following predictors: 1) Time [with

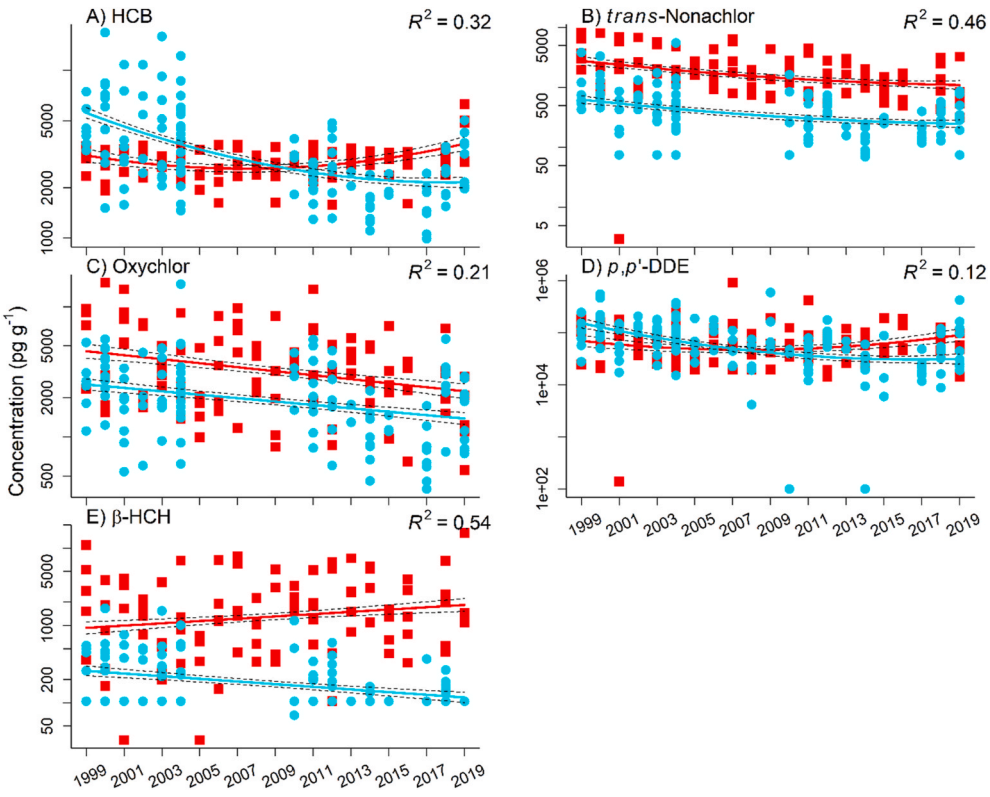

Fig. 2. Temporal trends of different organochlorine (OC) pesticides in goldeneyes (red) and tawny owls (blue). The figures show the predictions (thick coloured lines), including their precision (dotted thin lines; ± 1 Standard Error [SE]) and the coefficient of determination (R^2), from statistical models fitted to data from central Norway (1999–2019): A) hexachlorobenzene (HCB); B) *trans*-Nonachlor*; C) Oxychlordane*; D) *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE); E) β-hexachlorocyclohexanes (β-HCH). The X-axis shows time in years. Note: *Represents two different types of chlordanes.

Year as a continuous variable; in the model output, we centerd this variable (*i.e.*, subtracting the average value)]; 2) its second-order polynomial (Year^2); 3) Species [factor variable with 'Goldeneye' (baseline) and 'Tawny owl' as levels]. Additionally, our biological hypothesis also required us to assess the following interactions into our set of candidate models: 1) Species × Year; and 2) Species × Year^2. The biological rationale for this is that we hypothesized that the temporal trends, which could be non-linear due to the possible effect of Year^2, could be different for the two species. In sum, we ended up with seven *a priori* defined candidate models: including all combinations of the predictors and the two-ways interactions involving Species. For each response or analysis, each model (*i*) was rescaled and ranked relative to the model with the lowest second-order Akaike's Information Criterion (AICc) value (Δ_i denotes this for model *i*). From this set of candidate models we selected the simplest model (as judged by the models' estimated number of parameters) with a $\Delta_i \leq 1.5$ (e.g. Burnham and Anderson, 2002; Anderson, 2008) value using the AICcmodavg-package (Mazerolle, 2019).

Finally, the responses were treated differently depending on the number of samples in each analysis ≤ LOD. We created our own rule of thumb for this purpose. First, we defined LODs in the statistical analyses as the minimum value observed for each contaminant (*i.e.*, this may differ from how LOD was determined in the chemical analyses above). Second, based on the percentages of values ≤ LOD, we fitted different statical models to the data. If >15% of samples were ≤LOD, we fitted linear models using the lm-function. If, however, >50% of the samples were ≤LOD, we fitted logistic models to a binary response: classifying observations as 0 for ≤ LOD; 1 for > LOD. We fitted the latter model 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]. Finally, if between 15 and 50% of the samples were ≤LOD, a tobit regression model was fitted using the vglm-function in the vgam-package (using the tobit-family and the contaminant-specific LOD as the lower-limit: see Yee and Moler (2020) for how these models were specified). The tobit regression is a left-censored model where values at the LOD may either be equal to the lower threshold or smaller (even though values below zero are not defined) and we fitted these models similar to Thomas et al. (2016).

Our set of candidate models includes either Year, Year^2 (not included if Year was not included in any model), Species, and the Species-Year or -Year^2 interactions. Consequently, we speak of statistically significant temporal trends for goldeneye if either, or both, the main effect of Year or Year^2 was statistically significant. If neither the Species-Year nor -Year^2 interactions were statistically significant, both species' temporal trends were similar (or absent if the main effects were insignificant). A statistically significant Species-Year or -Year^2 interaction implies that the linear trend or the second-order polynomial differed for tawny owls compared to goldeneye (as we used the treatment contrast). Moreover, a statistically significant main effect of Species means that the average concentration for tawny owl differed from goldeneye (evaluated at the average value for Year if it was included in the model). We assessed statistical significance based on Tables 2 and 3. In contrast, we mainly refer to the figures when we speak of the resulting trends and differences between the species as models, including polynomials, may take various forms based on the sign and strengths of the estimates.

Finally, to further assess whether the concentrations for the different contaminants differed across species (i.e., covaried), we performed standard linear models (fitted using the lm-function) where median levels for each contaminant for tawny owls were predicted based on median levels of the same contaminant for goldeneye.

3. Results and discussion

3.1. OHC concentrations and profiles

Food is the primary exposure source of OHCs in birds, and based on the diets of the two species (Cramp and Simmons, 1977; Cramp, 1985; Yoccoz et al., 2009), we assumed that tawny owls and goldeneyes had about equal food chain length. As primary consumers, tawny owls depend on different species of voles, whereas goldeneyes depend on

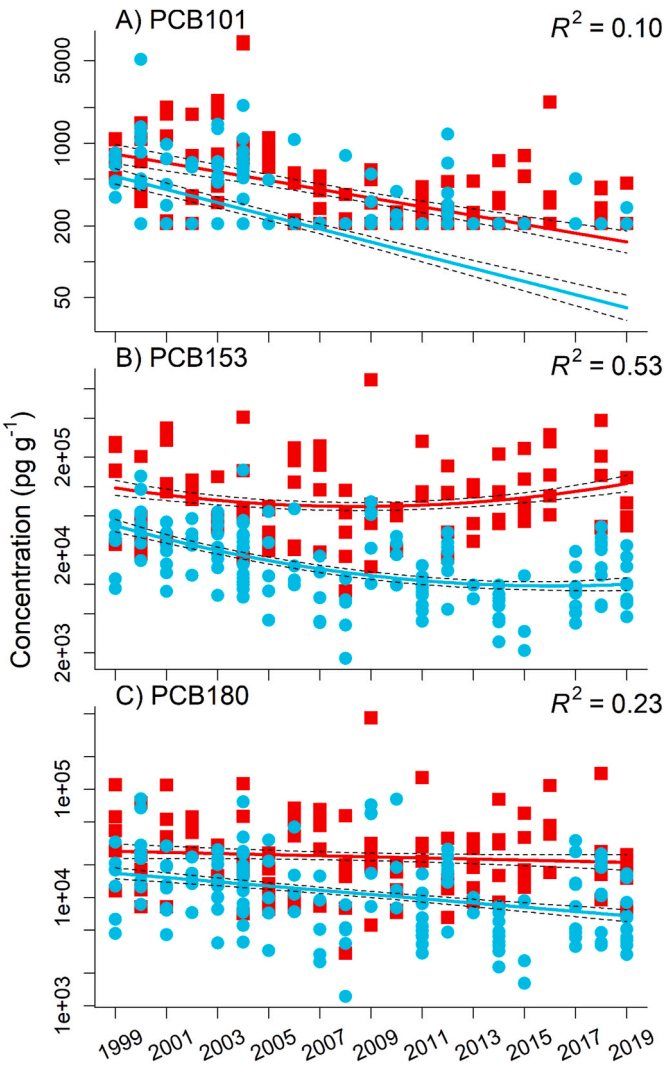

Fig. 3. Temporal trends of different selected persistent polychlorinated biphenyl (PCB) congeners in goldeneyes (red) and tawny owls (blue): A) PCB101; B) PCB153; C) PCB180. See legends for Fig. 2 for technical details.

molluscs and crustaceans. As secondary consumers, tawny owls and goldeneye consume passerine birds (sometimes shrews) and small fish, respectively. Hence, we predicted that the lipid normalized concentrations of OHCs in the two species' eggs would be similar. The median lipid-normalized concentrations of ΣOC (14 compounds) across all years was only ~11% lower in goldeneyes compared to tawny owls ($p > 0.50$; Table 1). The median concentrations (wet weight) of ΣPFAS (11 compounds) was, however, ~62% lower in tawny owls compared to goldeneyes ($p < 0.01$; Table 1).

The proportions of ΣOC and ΣPFAS were similar in the two species: *i.e.,* ~90% of the OHC load was made up by OCs ($\bar{x} = 88.4\% \pm 1.06$ SE *vs* 90.2% ± 0.82SE) and ~10% by PFASs ($\bar{x} = 11.6\% \pm 1.06$ SE *vs* 9.8% ± 0.82 SE) in goldeneye (n = 100) and tawny owl eggs (n = 94), respectively (Fig. 1). Among the OCs, PCBs strongly dominated in goldeneyes, comprising ~70% of the OC load ($\bar{x} = 71.3\% \pm 1.25$ SE, n = 100), whereas it only made up ~40% in tawny owl eggs ($\bar{x} = 40.6\% \pm 1.31$ SE, $n = 125$). Median concentrations of *p,p'*-DDE, HCB, oxychlordane, α-HCH, and γ-HCH were ~3, 2.4, 1.4, 3.5 and 3.5 times higher in tawny owls than in goldeneyes, respectively. On the contrary, median concentrations of *trans*-Nonachlor and β-HCH were ~2.4 and 4.2 times higher in goldeneyes (Table 1). The *p,p'*-DDE:PCB-ratio was very different between the species ($\bar{x} = 0.39 \pm 0.032$ SE *vs* 1.67 ± 0.01, $F_{1, 251} = 106.1$, $p < 0.0001$, in goldeneyes and tawny owls, respectively), as ~53% of the OC load was made up by *p,p'*-DDE in tawny owls, compared to only ~24% in goldeneyes. There were, however, no temporal trends in the *p,p'*-DDE:PCB-ratio in neither species ($p > 0.70$). Among the PCBs, median concentration of the less persistent PCB101 was 1.75 times higher in tawny owl eggs, whereas most persistent PCBs were higher in goldeneyes (~2–6 times), except PCB180 and PCB187 (Table 1). The consistent dominance of OC-pesticides in tawny owls, notably the DDT metabolite *p,p'*-DDE, seems somewhat unusual. Previous raptor studies in northern Europe have found *p,p'*-DDE to decline faster than PCBs, also in terrestrial environments (Newton et al., 1993, 1999; Helander et al., 2002; Wegner et al., 2005), probably because DDT was banned earlier. High levels of pesticides in the tawny owls in our study region might also reflect previous local use of pesticides such as DDT, chlordane, and HCHs in the terrestrial environment. PCBs, on the contrary, might be more influenced by long-range transport and may thus be more evenly distributed in the environment: the proportional relationship between *p,p'*-DDE and PCB in goldeneyes resembles most other aquatic bird species investigated in northern Europe (Bignert et al., 1995; Helander et al., 2002; Bustnes et al., 2005, 2006, 2008, 2013; Sun et al., 2020).

In the PFAS group, there were some differences in profiles in which sulfonates (PFSAs), on average, made up ~56% in goldeneyes and ~65% in tawny owls. The ΣPFCA:ΣPFSA-ratio, however, increased significantly over time in tawny owls ($F_{1, 119} = 22.1$, $p < 0.01$), but not in goldeneyes ($p = 0.34$; Fig. 1). Since PFASs tend to accumulate similarly in freshwater and terrestrial environments (Mueller et al., 2011; Xu et al., 2014; Eriksson et al., 2016), we predicted similar concentrations in the two species. However, the median concentration of PFOS was ~2.4 times higher in goldeneyes than tawny owls, whereas the ΣPFCAs were ~2–12 times higher, respectively (Table 1). The higher exposure of PFASs in goldeneyes could result from a more complex freshwater food chain (Chase, 2000). For instance, goldeneyes may feed at a higher trophic position than the tawny owl, or the freshwater food web may be more contaminated by PFASs than the adjacent terrestrial web (Eriksson et al., 2016). As PFASs are more protein-bound, they might have different environmental pathways than the lipophilic OCs, resulting in a low correlation between concentrations of OCs and PFASs found for some bird species (Bustnes et al., 2008, 2015). However, the sources of PFAS in Norway are poorly known, except that PFOS has been used in fire foam at airports, but long-range transport might also be of importance (Jouanneau et al., 2020).

Although the loads of most OHC in the predators from the two adjacent ecosystems were on the same order of magnitude, the great differences among individual compounds might be influenced by interspecific variation in the ability to bioaccumulate and biotransform different OHCs (e.g., Volta et al., 2009; Custer et al., 2010). One possible way around these potential confounding factors would be to study the same species in both ecosystems, or study closely related species. There are, however, no suitable species-pair of birds available for such comparison in the study region. Another issue we need to consider is that the study area for the goldeneyes is limited to one lake. In contrast, the eggs from tawny owls were collected over a larger area, consisting of different habitats such as forest, farmland, and more urbanized areas. This could lead to a more significant variation in the intake of OHCs for tawny owls than in goldeneyes. However, voles, the predominant prey of the tawny owl, usually fluctuate in cycles. When abundant, they are reasonably evenly distributed, as confirmed by regular checks of nest boxes' content, suggesting that feeding conditions are relatively similar over the different tawny owl territories in our study area (Yoccoz et al., 2009).

3.2. Temporal trends in concentrations

Since most OC compounds in this study were banned 35–50 years ago, we predicted an overall decline over the course of the study. In tawny owls, previous studies have documented declining trends between 1986 and 2004 (2009) (Bustnes et al., 2007, 2011), but there have been no studies of goldeneyes in this region. Similarly, among PFASs,

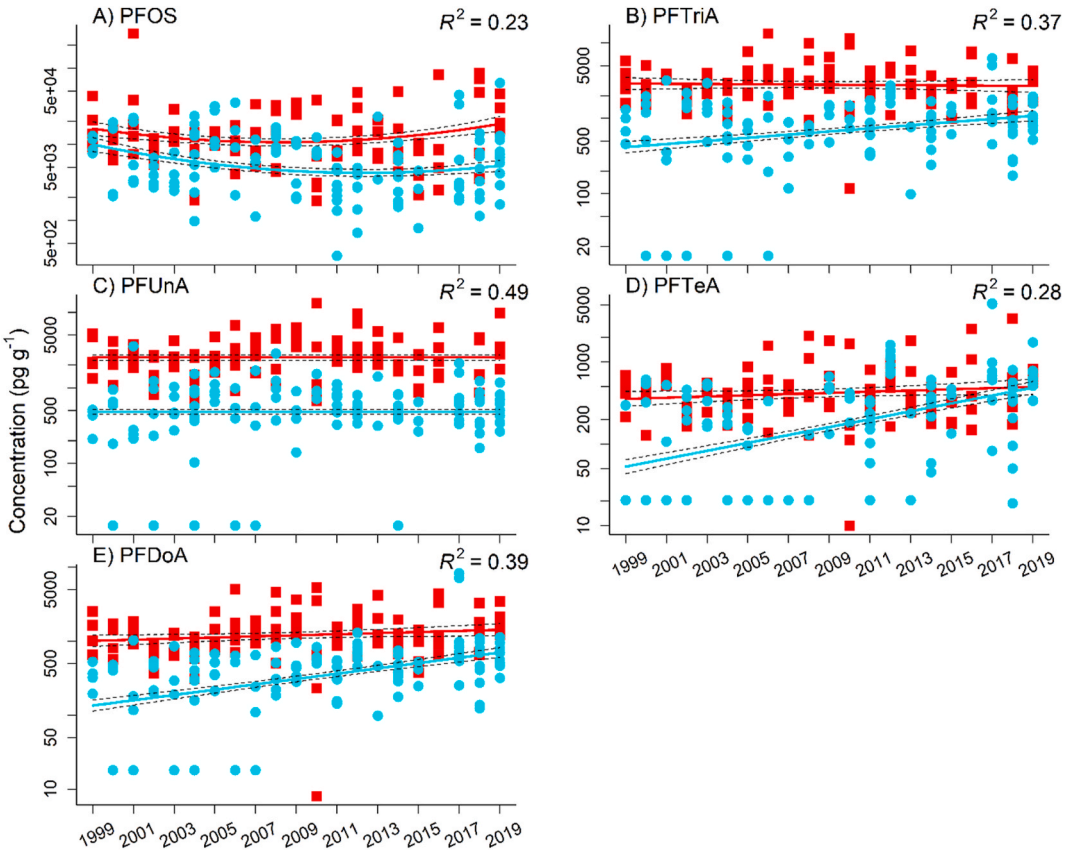

Fig. 4. Temporal trends of selected and protein-bound perfluorinated substances (PFASs) compounds in goldeneyes (red) and tawny owls (blue): A) perfluorooctane sulfonate (PFOS); B) perfluorotridecanoic acid (PFTriA); C) perfluoroundecanoate acid (PFUnA); D) perfluorotetradecanoic acid (PFTeA); E) perfluorododecanoic acid (PFDoA). See legends for Fig. 2 for technical details.

sulfonates (PFSAs) such as PFOS have been shown to decline in tawny owls while PFCAs have been increasing (Ahrens et al., 2011; Bustnes et al., 2015). We ran statistical models on lipid-normalized concentrations to compare the temporal trends for OCs between the two species. For the 14 OCs measured, either one or both of the year-species interactions were statistically significant, or marginally so. For 12 compounds, the trends were stronger for tawny owls than for goldeneyes (Table 2). However, for the following OC-pesticides; α-, β-, and γ-HCH, oxychlordane and *trans*-Nonachlor, and for PCB101, there were also significant declines in goldeneyes, but still stronger in tawny owls (Table 2, Figs. 2–3). In comparison, *p,p'*-DDE, and heavily chlorinated PCBs were all declining, significantly in tawny owls but not in goldeneyes (Table 2, Figs. 2–3). Moreover, whereas HCB showed significant declining trends in tawny owls, it increased strongly in goldeneyes, especially in recent years (Fig. 2, Table 2). The reason for this substantial ecosystem difference is unknown, but studies both in air, wildlife, and humans have found increasing HCB levels, especially in the Arctic (Hung et al., 2016; Ma et al., 2011; Rigét et al., 2020; Abass et al., 2018). Further, the physical-chemical properties of HCB drive its distribution towards the aquatic phase rather than the particulate-bound terrestrial sphere, causing higher background concentrations. Thirdly, unlike most other OCs, emissions of HCB continue as it is a byproduct of the manufacturing of chlorinated compounds and its industrial or combustion processes (Barber et al., 2005). It might also be that HCB in the goldeneye eggs has different sources outside the lake, brought in from the wintering grounds in the coastal areas by the birds. However, exogenous nutrients from freshwater breeding locations usually form the bulk of egg nutrients in migrating sea ducks (Hobson, 2006; Bond et al., 2007; DeVink et al., 2011).

Among PFASs (wet weight concentrations), the sulfonates (PFSAs), PFHpS, and PFOS (to a lesser extent) showed evidence for decreasing trends in tawny owls but not in goldeneyes (Fig. 4). PFOSA, however, showed strong declining trends in both species (Table 3). Among the PFCAs, the PFDoA, PFTriA and PFTeA increased in tawny owls but were stable in goldeneyes (Table 3, Fig. 4). However, PFNA and PFDcA showed similar trajectories increasing and levelling-off in both species (Table 3). For other PFAS compounds, we documented no significant trends for either species.

Hence, although some compounds, notably OC-pesticides, showed similar trends, the dominating OCs differed significantly among the species. In tawny owls, the declining trends were in line with the negative trends previously reported between 1986 and 2004 (2009) (Bustnes et al., 2007, 2011) and overall complied with other studies of terrestrial raptors (Newton et al., 1993; Wegner et al., 2005; Bustnes et al., 2007; Gómez-Ramírez et al., 2014). We know little about trends of OHCs in freshwater predators at relatively low trophic levels, such as ducks. However, in Scandinavia, OCs such as PCBs, have declined in freshwater fish since the 1980s (Nyberg et al., 2014). The osprey, a high trophic freshwater predator, shows decreasing OHC levels and improved reproductive performance worldwide (Henny et al., 2010). Hence, we expected declining OC trends in the goldeneyes over the two last decades. For PFAS, the trends for tawny owls corresponded to prior observed trends (Ahrens et al., 2011; Bustnes et al., 2015; Miller et al., 2015; Eriksson et al., 2016; Sun et al., 2019). However, the lack of trends in goldeneyes was somewhat unexpected since a study on osprey from Sweden, feeding on freshwater fish, reported declining PFAS trends, similar to tawny owls in this study (Eriksson et al., 2016).

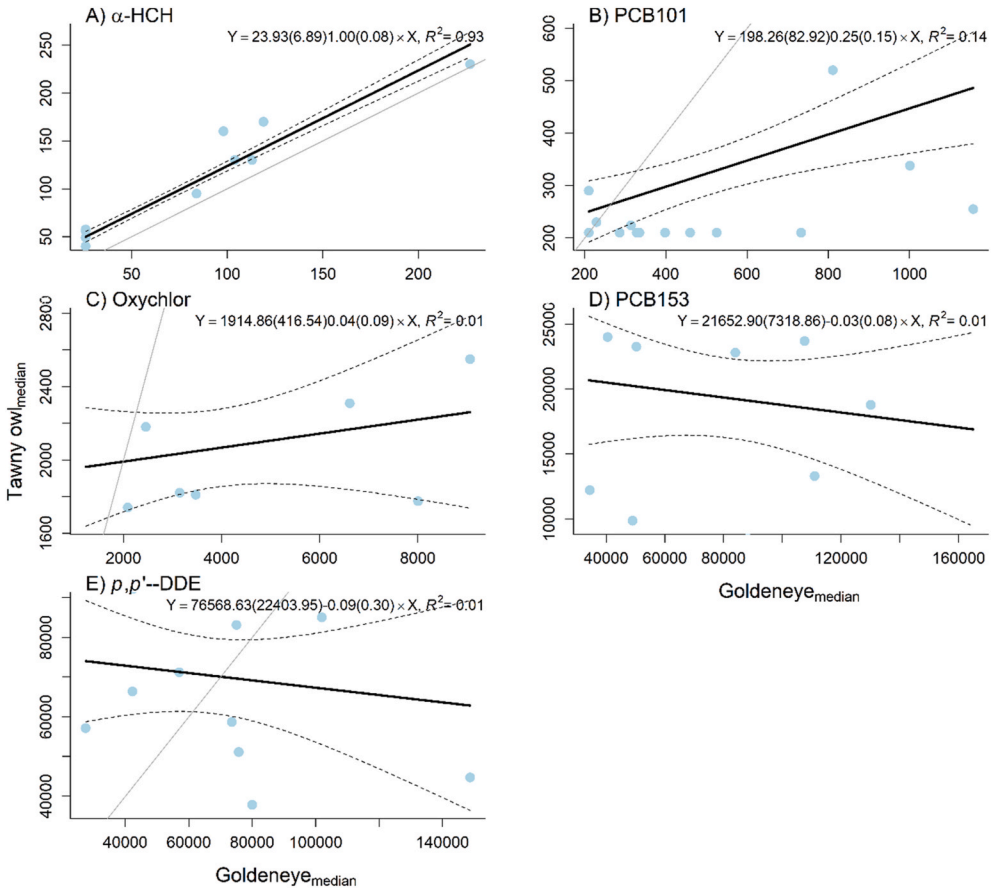

Fig. 5. Relationships between annual medians of selected organohalogenated compounds in tawny owls and goldeneyes. See legends for Figs. 2–4 for technical details.

3.3. Annual covariation in OHC concentrations between the species

Variation in concentrations among years is a prominent feature of OHC exposure in wildlife populations (Bignert et al., 1995; Helander et al., 2002; Elliott et al., 2012; Bustnes et al., 2005, 2011, 2015; Eriksson et al., 2016). Although the underlying causes have been under-studied, dietary changes and climate variation, influencing transport and biomagnifying processes, have been suggested to be major components (Noyes et al., 2009; Bustnes et al., 2011, 2015; Elliott et al., 2012). A solid annual covariance between adjacent freshwater- and terrestrial ecosystems would suggest that the processes determining exposure and accumulation of OHCs function evenly over landscapes and within different ecosystems. To test the annual between-species covariance of different compounds, we predicted the yearly median concentration for each compound in tawny owls based on the median for goldeneye using linear regression models. Among the 14 OCs, there was very little covariance in the concentrations, supported by R^2 values close to zero for most compounds (see Fig. 5 for examples of relationships). We documented a notable exception for α-HCH, which showed a high covariance (R^2 = 0.93). However, this result should be viewed with caution as it was one of the analyses where a high proportion of the observations (55.1%) was below the LOD (Fig. 5). Another exception was PCB101, where we documented a significant covariance despite a low coefficient of determination (R^2 = 0.14). Similar to OCs, there was very little covariation in concentrations between the two species for PFASs (R^2< 0.1; Fig. 5).

The main aim of this long-term study was to compare the OHC exposure to avian predators in adjacent terrestrial- and freshwater ecosystems in landscapes consisting of different ecosystems. We hypothesized that if the same environmental processes were equally important drivers for OHC exposure in our study region's freshwater and terrestrial ecosystems, we predicted similar concentrations and profiles of OHCs and temporal trends. Moreover, we expected this to result in a positive covariance in concentrations between the two species. Although the ∑OC were similar among the two species, PFAS concentrations were considerably higher in the goldeneyes. In addition, the profiles of the OHC loads were different between the species, which might be due to several reasons. First, greater input of some OHCs in the freshwater ecosystem might occur because of possible drainage to and easier spread of OHCs in the aquatic environment (higher baseline concentrations in the lowest levels of the aquatic food chain). Second, the added complexity (e.g., more trophic levels) in the freshwater ecosystem (Chase, 2000; Eriksson et al., 2016) may lead to higher biomagnification. Third, the physical-chemical properties of individual OHCs results in a partition into either the aquatic or terrestrial ecosystem: more polarized compounds will distribute more quickly in an aquatic environment. In contrast, highly hydrophobic compounds spread more efficiently in a terrestrial environment. Establishing the importance of each of these factors is difficult without detailed knowledge about the diet, *i.e.* the OHC content in the dietary component and the abiotic surroundings.

Moreover, the temporal trends of OHCs were different between the species, and the major components in the OHC loads such as persistent PCBs, *p,p'*-DDE, PFOS showed no signs of declining in goldeneyes whereas they did in the tawny owls. The latter might be due to the higher input of OHCs to the freshwater systems resulting in a slower response to diminishing environmental OHC concentrations than in the terrestrial systems. Finally, there were no indications that high levels of OHCs in one species were concomitant to high concentrations in the other species. We thus conclude that the processes at play in shaping the

contaminant loads of predators in adjacent terrestrial and aquatic ecosystems within the same landscapes differ. Therefore, it is arduous to predict OHC exposure in different components of landscapes by studying one ecosystem. The reasons for the differences observed in this study may be multifaceted. Firstly, we cannot exclude that it has to do with species differences, and ideally, it would be better to compare species that were closer taxonomically. Hence, some of the differences we found between the goldeneye and the tawny owl in terms of the composition of OHC loads may be due to different inter-specific physiological traits: ability to eliminate or bio-transform different compounds, amount of lipids in the eggs, etc.

In conclusion, the two avian predators in adjacent ecosystems seem to be subject to different processes determining the OHC exposure, probably related to diet variation and climate, long-range transport of various contaminants in addition to variation in local pollution. These results may be a step towards a better understanding of the vulnerability of different ecosystems to the accumulation of legacy and emerging pollutants with a broad range of chemical-physical properties. Future studies measuring OHCs in the lower food chains, *i.e.* the prey items of the different species, might help us gain more insights into the mechanisms determining OHC exposure in different ecosystems.

CRediT authorship contribution statement

Jan Ove Bustnes: Conceptualization, Writing – review & editing, analyses, writing. **Bård-Jørgen Bårdsen:** Conceptualization, statistical analyses, Writing – review & editing. **Dorte Herzke:** Methodology, chemical analyses. **Georg Bangjord:** Methodology, Data collection. **Sophie Bourgeon:** Conceptualization, Writing – review & editing. **Clementine Fritsch:** Conceptualization, Writing – review & editing. **Igor Eulaers:** Conceptualization, Writing – review & editing.

Declaration of competing interest

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

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References

Abass, K., Emelyanova, A., Rautio, A., 2018. Temporal trends of contaminants in Arctic human populations. Environ. Sci. Pollut. Res. 25, 28834–28850.

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. Environ. Sci. Technol. 45, 8090–8097.

Anderson, D.R., 2008. Model Based Inference in the Life Sciences: a Primer on Evidence. Springer Science, New York, United States of America.

Bakken, V., Runde, O., Tjørve, E., 2003. Norsk Ringermingsatlas, vol. 1. Stavanger Museum, Stavanger).

Barber, J.L., Sweetman, A.J., van Wijk, D., Jones, K.C., 2005. Hexachlorobenzene in the global environment: emissions, levels, distribution, trends and processes. Sci. Total Environ. 349, 1–44.

Bignert, A., Litzén, K., Odsjö, T., Olsson, M., Persson, W., Reutergårdh, L., 1995. Time-related factors influence the concentrations of DDT, PCBs and shell parameters in eggs of Baltic guillemot (Uria aalge), 1861-1989. Environ. Pollut. 89, 27–36.

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. Environ. Pollut. 99, 177–198.

Bond, J.C., Esler, D., Hobson, K.A., 2007. Isotopic evidence for sources of nutrients allocated to clutch formation by harlequin ducks. Condor 109, 698–704. https://doi.org/10.1650/8241.1.

Burnham, K.P., Anderson, D.R., 2002. Model Selection and Multimodel Inference: a Practical Information-Theoretic Approach, second ed. Springer, Inc., New York, USA.

Bustnes, J.O., Skaare, J.U., Berg, V., Tveraa, T., 2005. Interseasonal variation in blood concentrations of organochlorines in great black-backed gulls (Larus marinus). Environ. Toxicol. Chem. 24, 1801–1806.

Bustnes, J.O., Helberg, M., Strann, K.B., Skaare, J.U., 2006. Environmental pollutants in endangered vs. increasing subspecies of lesser black-backed gulls along the Norwegian Coast. Environ. Pollut. 144, 893–901.

Bustnes, J.O., Yoccoz, N., Bangjord, G., Polder, A., Skaare, J., 2007. Temporal trends (1986-2004) of organochlorines and brominated flame retardants in tawny owl eggs from northern Europe. Environ. Sci. Technol. 41, 8491–8497.

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). Environ. Toxicol. Chem. 27, 1383–1392.

Bustnes, J.O., Yoccoz, N., 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. Environ. Sci. Technol. 45, 7542–7547.

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. Environ. Toxicol. Chem. 32, 2520–2527.

Bustnes, J.O., Bangjord, G., Herzke, D., Ahrens, L., Yoccoz, N., 2015. Perfluoroinated alkyl substance (PFASs) concentrations in a terrestrial raptor: relationships to environmental conditions and individual traits. Environ. Toxicol. Chem. 34, 184–191.

Chase, J.M., 2000. Are there real differences among aquatic and terrestrial food webs? Trends Ecol. Evol. 15 (10), 408–412.

Clark, R.G., Poysa, H., Poysa, H., Runko, P., Paasivaara, A., 2014. Spring phenology and timing of breeding in short-distance migrant birds: phenotypic responses and offspring recruitment patterns in common goldeneyes. J. Avian Biol. 45, 457–465.

Cramp, S., 1985. Handbook of the Birds of Europe the Middle East and North Africa. The Birds of the Western Palearctic, vol. IV. Oxford University Press.

Cramp, S., Simmons, K.E.L., 1977. Handbook of the Birds of Europe the Middle East and North Africa. The Birds of the Western Palearctic, vol. IV. Oxford University Press.

Custer, C.M., Custer, T.W., Dummer, P.M., 2010. Patterns of organic contaminants in eggs of an insectivorous, and omnivorous, and a piscivorous bird nesting on the Hudson River, New York, USA. Environ. Toxicol. Chem. 29 (10), 2286–2296.

DeVink, E.M., Slattery, S.M., Clark, R.G., Alisauskas, R.T., Hobson, K.A., 2011. Combining stable-isotope and body-composition analyses to assess nutrient-allocation strategies in breeding white-winged scoters (Melanitta fusca). Auk 128, 166–174. https://doi.org/10.1525/auk.2010.10069.

Dietz, R., Riget, F., Cleemann, M., Aarkrog, A., Johansen, P., Hansen, J.C., 2000. Comparison of contaminants from different trophic levels and ecosystems. Sci. To. Environ. 245, 221–231.

Elliott, K.H., Cesh, L.S., Dooley, J.A., Letcher, R.J., Elliott, J.E., 2009. PCBs and DDE, but not PBDEs, increase with trophic level and marine input in nestling bald eagles. Sci. Total Environ. 407, 3867–3875.

Elliott, J.E., Levac, J., Guigueno, M.F., Shaw, D.P., Wayland, M., Morrissey, C.A., Muir, D.C.G., Elliott, K.H., 2012. Factors influencing legacy pollutant accumulation in alpine osprey: biology, topography, or melting glaciers? Environ. Sci. Technol. 46 (17), 9681–9689.

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). Environ. Res. 149, 40–47.

Eulaers, I., Covaci, A., Herzke, D., Eens, M., Sonne, C., Moum, T., Schnug, L., Hanssen, S. A., Johnsen, T.V., Bustnes, J.O., Jaspers, V.L.B., 2011. A first evaluation of the usefulness of feathers of nestling predatory birds for non-destructive biomonitoring of persistent organic pollutants. Environ. Int. 37, 622–630.

Fox, J., 2002. An R and S-PLUS Companion to Applied Regression. Sage Publications, London, UK and Thousand Oak, USA.

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

Hebert, 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. Environ. Rev. 7, 147–166.

Hebert, C.E., Hobson, K.A., Shutt, J.L., 2000. Changes in food web structure affect rate of PCB decline in herring gull (Larus argentatus) eggs. Environ. Sci. Technol. 34, 1609–1614.

Hebert, C.E., Weseloh, D.V.C., Idrissi, A., Arts, M.T., O’Gorman, R., Gorman, O.T., Locke, B., Madenjian, C.P., Roseman, E.F., 2008. Restoring piscivorous fish populations in the Laurentian Great Lakes causes seabird dietary change. Ecology 89 (4), 891–897.

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 whithe-tailed sea eagle (Haliaeetus albicilla) in Sweden. Ambio 31, 386–403.

Henny, C.J., Grove, R.A., Kaiser, J.L., Johnson, B.L., 2010. North American osprey populations and contaminants: historic and contemporary perspectives. J. Toxicol. Environ. Health B Crit. Rev. 13 (7–8), 579–603.

Hitchcock, D.J., Andersen, T., Varpe, O., Loonen, M.J.J.E., Warner, N.A., Herzke, D., Tombre, I.M., Griffin, L.R., Shimmings, P., Borgå, K., 2019. Potential effect of migration strategy on pollutant occurrence in eggs of arctic breeding barnacle geese (Branta leucopsis). Environ. Sci. Technol. 53 (9), 5427–5435.
Hobson, K.A., 2006. Using stable isotopes to quantitatively track endogenous and exogenous nutrients allocations to eggs of birds that travel to breed. Ardea 94, 359–369.
Hobson, K.A., Thompson, J.E., Evans, M.R., Boyd, S., 2005. Tracing nutrient allocation to reproduction in Barrow's goldeneye. J. Wildl. Manag. 69 (3), 1221–1228.
Hung, H., Katsoyiannis, A.A., Brorstrom-Lunden, E., Olafsdottir, K., Aas, W., Breivik, K., Bohlin-Nizzetto, P., Sigurdsson, A., Hakola, H., Bossi, R., Skov, H., Sverko, E., Barresi, E., Fellin, P., Wilson, S., 2016. Temporal trends of persistent organic pollutants (POPs) in arctic air: 20 years of monitoring under the arctic monitoring and assessment programme (AMAP). Environ. Pollut. 217, 52–61.
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. Environ. Sci. Technol. 54, 5011–5020.
Ma, J., Hung, H.L., Tian, C., Kallenborn, R., 2011. Revolatilization of persistent organic pollutants in the Arctic induced by climate change. Nat. Clim. Change 1, 255–260.
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 Asia. Environ. Toxicol. Chem. 34, 1799–1808.
Mueller, C.E., De Silva, A.O., Small, J., Morris, A., Katz, S., Gamberg, M., Muir, D.C.G., 2011. Biomagnification of perfluorinated compounds in a remote terrestrial food chain: lichen-caribou-wolf. Environ. Sci. Technol. 45 (20), 8665–8673.
Newton, I., Wyllie, I., Asher, A., 1993. Long-term trends in organochlorine and mercury residues in some predatory birds in Britain. Environ. Pollut. 79, 143–151.
Newton, I., Dale, L., Little, B., 1999. Trends in organochlorine and mercurial compounds in the eggs of British Merlins Falco columbarius. Hous. Theor. Soc. 46, 356–362.
Noyes, P.D., McElwee, M.K., Miller, H.D., Clark, B.W., Van Tiem, L.A., Lindsey, A., Walcott, K.C., Erwin, K.N., Levin, E.D., 2009. The toxicology of climate change: environmental contaminants in a warming world. Environ. Int. 35, 971–986.
Nyberg, E., Danielsson, S., Eriksson, U., Faxneld, S., Miller, A., Bignert, A., 2014. Spatio-temporal trends of PCBs in the Swedish freshwater environment 1981-2012. Ambio 43, 45–57.
R Core Team, 2020. R: a Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria.
Rigét, F., Vorkamp, K., Eulaers, I., Dietz, R., 2020. Influence of climate and biological variables on temporal trends of persistent organic pollutants in Arctic char and ringed seals from Greenland. Environ. Sci. Process. Impact. 22, 993–1005.
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., Eulaers, I., 2019. White-tailed eagle (Haliaeetus albicilla) body feathers document spatiotemporal trends of poly- and perfluoroalkyl substances in the northern environment. Environ. Sci. Technol. 53, 12744–12753.
Sun, J., Bustnes, J.O., Bårdsen, B.J., Dietz, R., Helander, B., Jaspers, V.L.B., Lepoint, G., Søndergaard, J., Sonne, C., Thorup, K., Tøttrup, A.P., Zubrod, J., Eens, M., Eulaers, I., 2020. Temporal trends of legacy organochlorines in different white-tailed eagle (Haliaeetus albicilla) subpopulations. Environ. Int. 138, 105618.
Thomas, M.G., Næss, M.W., Bårdsen, B.-J., Mace, R., 2016. Smaller Saami herding groups cooperate more in a public goods experiment. Hum. Ecol. 1–10.
Volta, P., Tremolada, P., Neri, M.C., Giussani, G., Galassi, S., 2009. Age-dependent dioaccumulation of organochlorine compounds in fish and their selective biotransformation in top predators from Lake Maggiore (Italy). Water. Soil. Pollut. 197, 193–209.
Wegner, P., Kleinstauber, G., Baum, F., Schilling, F., 2005. Long-term investigation of the degree of exposure of German peregrine falcons (Falco peregrinus) to damaging chemicals from the environment. J. Ornithol. 146, 34–54.
Xu, J., Guo, C.S., Zhang, Y., Meng, W., 2014. Bioaccumulation and trophic transfer of perfluorinated compounds in a eutrophic freshwater food web. Environ. Pollut. 184, 254–261.
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. Environ. Int. 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, New York, USA.