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The impact of climate sensitive factors on the exposure to organohalogenated contaminants in an aquatic bird exploiting both marine and freshwater habitats

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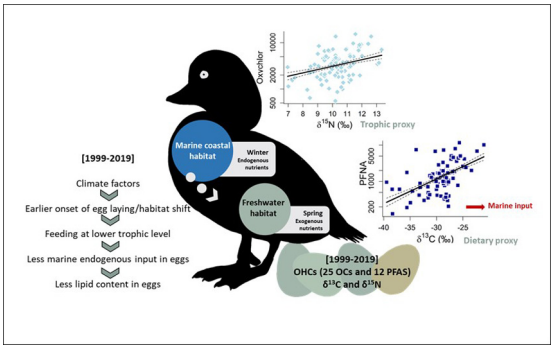
HIGHLIGHTS

- Exposure to OCs and perfluoroalkyl substances was assessed in common goldeneyes.
- Few compounds showed significant temporal trends over the last two decades.
- In general organochlorines were positively associated to trophic position ($\delta^{15}N$).
- Positive associations between PFASs and $\delta^{13}C$ isotopes may suggest some marine input.
- Exposure to OHCs in goldeneyes is significantly influenced by climate sensitive factors.

GRAPHICAL ABSTRACT



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ABSTRACT

To assess how climate-sensitive factors may affect the exposure to organochlorines (OCs) and perfluoroalkyl substances (PFASs), we monitored concentrations in eggs of the common goldeneye (*Bucephala clangula*) over two decades (1999–2019) in central Norway. The goldeneye alternates between marine and freshwater habitats and is sensitive to climate variation, especially due to alterations in ice conditions which may affect feeding conditions. We assessed how biological factors such as diet (stable isotopes $\delta^{13}C$ and $\delta^{15}N$), the onset of egg laying, and physical characteristics such as winter climate (North Atlantic Oscillation: NAO_w) influenced exposure. We predicted compounds to show different temporal trends depending on whether they were still in production (*i.e.* some PFASs) or have been banned (*i.e.* legacy OCs and some PFASs). Therefore, we controlled for potential temporal trends in all analyses. There were declining trends for α- and γ-hexachlorocyclohexane (HCH), oxychlordane, *cis*-chlordane, *cis*-nonachlor, *p,p′*-dichlorodiphenyltrichloroethane (*p,p′*-DDT) and less persistent polychlorinated biphenyl (PCB) congeners (*e.g.* PCB101). In contrast, the dominant compounds, such as *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE) and persistent PCB congeners, were stable, whereas hexachlorobenzene (HCB) increased over time. Most OCs were positively related to $\delta^{15}N$, suggesting higher exposure in birds feeding at upper trophic levels. Chlordanes and HCB were positively associated with $\delta^{13}C$,

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indicating traces of marine input for these compounds, whereas the relationships to most PCBs were negative. Among PFASs, perfluorooctanesulfonamide (PFOSA) and perfluorohexane sulfonic acid (PFHxS) declined. Most PFASs were positively associated with $\delta^{13}C$, whereas there were no associations with $\delta^{15}N$. Egg laying date was positively associated to perfluoroheptanesulfonic acid (PFHpS), perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), suggesting that some of the PFAS load originated from the wintering locations. Although NAO*w* had little impact on the exposure to organohalogenated contaminants, factors sensitive to climate change, especially diet, were associated with the exposure to OHCs in goldeneyes.

1. Introduction

An important question is how a rapidly changing climate will impact the distribution of organochlorinated contaminants (OHCs), such as lipophilic organochlorines (OCs) and perfluoroalkyl substances (PFASs), in the physical environment and biota. Although most OCs have declined strongly in wildlife since bans were introduced in the US and western Europe in 1970s and 1980s, environmental concentrations are still considerable due to their high persistence (Loganathan and Kannan, 1994; Hebert et al., 1999; Bignert et al., 1998; Helander et al., 2002; Bustnes et al., 2007). Perfluoroalkyl substances (PFASs) have mostly been used as processing additives in fluoropolymer production and many consumer products. They were, however, not recognized as an environmental problem before the 1990s (Giesy and Kannan, 2001). PFASs generally bind to proteins, and many compounds are environmentally persistent, bioaccumulative and may have adverse effects on biota (Lau et al., 2004; Hoff et al., 2005; Land et al., 2018). Recent wildlife studies have documented decreasing trends of some PFASs, notably perfluorooctanesulfonic acid (PFOS), due to restrictions in production and use. However, other less regulated substances, such as many perfluorocarboxylic acids (PFCAs), still show increasing trends in different wildlife species (Holmström et al., 2010; Ahrens et al., 2011; Braune and Letcher, 2013; Miller et al., 2015; Jouanneau et al., 2020; Pereira et al., 2015; Sun et al., 2019).

Concentrations of biomagnifying organohalogenated contaminants (OHCs) often show great spatiotemporal variability within wildlife species, and monitoring such compounds in biota may not be straight forward since various environmental and biological factors determine exposure, in addition to emission rate (Hebert et al., 1997; Hebert and Weseloh, 2006; Hebert, 1998; Noyes et al., 2009; Bustnes et al., 2011, 2015). Hence, pollution trends may go undetected or spurious trends could be established, for example due to dietary changes and climate variation (Hebert et al., 2000, 2008; Noyes et al., 2009).

Marine food chains are generally more exposed to OHCs than their terrestrial- and freshwater counterparts, reflected in concentrations in apex predators (Elliott et al., 2009; Eulaers et al., 2013), although freshwater systems may be highly polluted from point sources. However, some species may switch habitats over their annual cycle, for example sea ducks, a group of large-bodied, predominantly marine diving ducks, which mainly breed in freshwater systems. Females lay voluminous clutches relative to their body size and thus must build up large lipid reserves prior to egg laying. Nutrients of both marine- (often endogenous nutrients) and freshwater-origins (exogenous nutrients) may be exploited for egg formation in sea ducks (Hobson et al., 2005, 2015; DeVink et al., 2011). In Barrow's goldeneye (*Bucephala islandica*), for example, allocation among different nutrients varied by the tactics of individual females, laying order of the eggs, and across years (Hobson et al., 2005). Little is known about how different nutrient sources affect the build-up of OHC burdens in sea ducks. However, whether eggs are produced from endogenous or exogenous nutrients is expected to influence the magnitude of OHC sequestration into the eggs. Endogenous lipid reserves are a major storage compartment for lipophilic OHCs in sea ducks (Swift et al., 1993; Franson et al., 2004; Braune and Malone, 2006; Bustnes et al., 2012; Tomza-Marciniak et al., 2019).

Since maternal transfer is a major source of OHCs excretion, eggs of aquatic birds have frequently been used for monitoring purposes (Bignert et al., 1998; Hebert et al., 1999; Helander et al., 2002). In this study, we collected eggs of the common goldeneye (*Bucephala clangula*), a close relative of the Barrow's goldeneye, in Central Norway. Goldeneyes breed in freshwater habitats and winter in coastal areas, in both marine and freshwater habitats (Bakken et al., 2003; Clark et al., 2014). The birds usually arrive on the lakes in spring, as soon as the ice breaks up, and the egg laying may start shortly after as air temperatures rise (Pöysä, 1996; Clark et al., 2014; G. Bangjord, unpublished data).

The ongoing climatic changes are unprecedented and profoundly impact many bird species. The goldeneye may be greatly affected due to its sensitivity to ice conditions, both in the wintering- and breeding grounds (Clark et al., 2014; Adde et al., 2020). Hence, the objective of this study was to unravel how different biological and physical climate-sensitive factors may influence the accumulation of OHCs in goldeneyes. Changes in habitat use will affect their diet, both the composition and the food quality (Clark et al., 2014), which again is expected to influence the exposure to OHCs. Hence, if goldeneyes expand their period in marine habitats, the exposure to OHCs may increase because of the generally higher levels in marine food chains. Moreover, if climate changes result in goldeneyes changing the trophic level at which they feed, exposure to biomagnifying OHCs will also change.

We collected eggs from goldeneyes over two decades (1999–2019) and analysed concentrations of 25 OCs and 11 PFASs, and dietary tracers (stable isotopes: $\delta^{13}C$ and $\delta^{15}N$). In general, $\delta^{15}N$ has been interpreted as an indicator of trophic position, although it varies spatially due to different baselines in different areas. $\delta^{13}C$ is depleted in terrestrial and offshore habitats but enriched in coastal habitats (Hobson, 1987; Hobson, 2006; Elliott et al., 2009), in which goldeneyes in this study spend the winter. Hence $\delta^{13}C$ will be a good indicator of the type of habitat from which the nutrients used to produce eggs originates. Each year, we recorded the onset of egg laying in the population, a proxy for variation in breeding conditions among seasons, as it reflects temperatures and ice break-up (Clark et al., 2014). Finally, we included the winter North Atlantic Oscillation index (NAO*w*), a composite measurement of climate conditions (Stenseth et al., 2002; Hurrell et al., 2003), as a predictor. In central Norway, NAO*w* has proven a reliable proxy for temperature and precipitation and is an important predictor of OHC accumulation in a terrestrial raptor inhabiting areas close to the goldeneyes in the present study (Bustnes et al., 2011, 2015).

Firstly, we hypothesised that the annual accumulation of OHCs in goldeneye eggs would show temporal trends depending on whether they are currently being used (increasing) or have been banned (declining). Secondly, we predicted that ducks feeding at higher trophic levels (increasing $\delta^{15}N$) would be more exposed to OHCs. Thirdly, based on the assumption that marine food chains generally are more exposed to OHC than freshwater ones, we predicted that if egg production included endogenous lipids acquired in winter areas, OHC concentrations would increase with increasing $\delta^{13}C$ values. Fourth, late onset of egg laying suggests late arrival in the breeding habitat, and hence the potential for more use of endogenous nutrients in egg production. However, this might differ among lipophilic OCs and protein-bound PFAS due to their different affinity, but we were unable to make predictions about these possible relationships. Finally, we predicted a negative effect of NAO*w* (mild winters) on OHC as mild winters result in less ice and better feeding conditions for the ducks. This might shorten migration distances, resulting in lower use of endogenous lipids and hence lowered OHC concentrations in the eggs.

2. Materials and method

2.1. Study area

The study area was at Jonsvatnet (63.22°N, 10.35°E) a lake (15 km^2) near the city of Trondheim in Trøndelag County, in central Norway. Eggs of goldeneyes were collected at Litlvantnet (1.6 km^2), a section of Jonsvatnet. 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 in the period from late March, and freshly laid eggs were collected and frozen. Five goldeneye eggs were analysed per year for all years (n = 100), except 2017 when a freezer breakdown destroyed the samples.

2.2. Climate variables

Climate variation may be measured by a multitude of factors/parameters (temperature, wind, precipitation, *etc.*). In the present study, we used the North Atlantic Oscillation (NAO) index as a proxy for large-scale climate variation (Hurrell et al., 2003; Stenseth et al., 2002). The NAO measures sea-level pressure variability between the Azores and Iceland and thus represents, on a large scale several climate variables in central Norway, including the atmospheric air transport from southern regions (Hurrell et al., 2003). The winter NAO index (NAO_w) in our study covers winter months (December–March) for all relevant years and where obtained from the website of Jim Hurrell of the National Centre for Atmospheric Research (http://www.cgd.ucar.edu/cas/jhurrell/indices.html). In central Norway, NAO*w* has proven a reliable proxy for temperature and precipitation and is a predictor of OHC accumulation in tawny owls (*Strix aluco*), a terrestrial raptor (Bustnes et al., 2011, 2015).

2.3. Stable isotopes

The ratio of ^{13}C to ^{12}C discriminates between carbon sources. Terrestrial ecosystems are depleted in ^{13}C compared to aquatic ones, and the ratio of ^{13}C to ^{12}C is in addition, distinctively different between freshwater, littoral, benthic and pelagic communities. The ratio of ^{15}N to ^{14}N typically increases by 2–5 ‰ per trophic level and thus represents a proxy for trophic feeding levels (Hobson, 2006). The analysis for stable carbon and nitrogen isotopes was carried out at the Stable Isotope Lab of the University of Koblenz-Landau (Germany). Ratios of carbon and nitrogen isotopes were determined in a homogenized and freeze-dried egg subsample (1.36 ± 0.32 mg) using a Flash 2000 HT elemental analyser coupled *via* a ConFlo IV interface to a Delta V Advantage isotope ratio mass spectrometer (all Thermo Fisher Scientific, Bremen, Germany). The stable isotope values for carbon and nitrogen are conventionally expressed as δ values (‰) relative to their respective international measurement standards Vienna Pee Dee Belemnite and atmospheric N_2, respectively. Internal reference material (casein) was measured concurrently in duplicate every ten samples, revealing a precision (±SD) ≤ 0.06 ‰ for $\delta^{13}C$ and $\delta^{15}N$, respectively.

2.4. Contaminant analysis

2.4.1. OC analysis

Approximately 1.5 g of pre-weighed egg homogenate was mixed with anhydrous sodium sulfate (approximately 1:5 weight ratio) burnt at 600 °C. The sample homogenate was allowed to freeze dry overnight to ensure the removal of moisture content within the sample. Samples were spiked with ^{13}C-labelled internal standards prior to being three times extracted with cyclohexane/acetone (3:1) under sonication. Supernatant from each step was combined (total volume 100 ml), where 10 % of the combined supernatant was aliquoted into a pre-weighed vial for gravimetric lipid determination. The remaining supernatant was evaporated to dryness and reconstituted in 0.5 ml of isooctane and transferred to EZ-POP NP cartridges (Supelco®) for clean-up purposes. The OCs were eluted from the cartridges with 3 × 5 ml of acetonitrile, and the collected acetonitrile eluent was evaporated and reconstituted in 0.5 ml of isooctane. An additional clean-up step was performed using automated solid phase extraction where the extract was eluted with 1 g of activated Florisil® (burnt at 450 °C) with 12 ml of 1:10 dichloromethane/hexane. The collected extract was evaporated to approximately 0.1 ml, quantitatively transferred to a GC vial and evaporated to 100 μl and spiked with ^{13}C-labelled PCB-159 recovery standard.

Analysis was performed by gas chromatography mass spectrometry (GC–MS) using a Waters Quattromicro mass analyser (Waters, US). Separation of analytes was performed on at TG-5SilMS GC (30 m × 0.25 mm ID × 250 μm) capillary column using helium as a carrier gas at a constant flow rate of 1 ml/min. The GC oven program utilized for separation was as follows: an initial temperature of 40 °C held for 1.5 min followed by a temperature ramp of 25 °C/min to 180 °C. Then, the temperature increased at 5 °C/min to 280 °C followed by a final temperature ramp of 40 °C/min to 320 °C and held for 5 min. Analysis was performed using electron impact ionization (70 eV) in targeted single ion monitoring (TSIM) mode.

Table 1
Summary statistics for lipids, stable isotopes ($\delta^{15}N$ and $\delta^{13}C$) and organohalogenated compounds (OHCs: pg/g wet weight) in goldeneye eggs from central Norway, 1999–2019.

Variable	N	Mean	Median	Std. err	Min	Max
%Lipids	92	23.57	22.42	0.54	16.05	39.99
$\delta^{13}C$	100	−29.23	−28.76	0.33	−39.45	−21.03
$\delta^{15}N$	100	10.19	10.22	0.13	6.96	13.34
HCB	100	2920.7	2941.5	66.1	1581	6283
α-HCH	100	58.2	26.0	5.9	26	287
β-HCH	100	2338.6	1442.5	256.8	33	15,716
γ-HCH	100	64.9	26.5	12.6	6	1071
Oxychlordane	100	4123.0	3216.0	296.2	555	15,280
trans-Chlordane	100	415.5	294.0	39.9	5	2642
cis-Chlordane	100	72.0	28.0	12.0	11	650
trans-Nonachlor	100	2222.1	1631.5	182.3	3	8264
cis-Nonachlor	100	714.2	492.5	68.7	66	3194
Mirex	100	601.0	392.0	68.4	107	4821
p,p′-DDT	100	24,319.3	8065.5	7224.8	420	661,521
p,p′-DDD	100	2462.3	841.0	652.7	36	59,549
p,p′-DDE	100	83,084.3	57,406.5	10,335.6	138	913,093
PCB28/31	100	3196.4	1475.5	555.0	299	35,197
PCB52	100	176.8	80.5	46.0	25	4339
PCB101	100	636.2	358.5	103.5	39	7173
PCB99	100	16,850.3	7461.5	3608.9	34	313,467
PCB105	100	14,445.3	6468.5	3503.9	908	311,892
PCB118	100	47,453.5	22,297.0	9902.5	2576	807,684
PCB138	100	71,569.0	39,712.5	11,596.5	4580	791,916
PCB153	100	115,648.6	70,849.5	15,022.5	8553	1,235,972
PCB180	100	35,932.4	22,414.0	5103.7	3051	459,157
PCB183	100	7292.1	4492.0	980.5	597	85,379
PCB187	100	20,504.5	12,850.5	2276.0	1811	178,685
PCB194	100	3350.7	2037.5	439.7	215	35,017
ΣPCB	100	337,055.6	190,945.0	48,652.3	23,552	3,413,009
ΣOC	100	460,451.6	297,716.5	56,869.8	50,970	3,716,804
PFOSA	100	34.4	15.0	4.8	15	268
PFHpS	100	234.3	94.0	38.5	18	2393
PFHxS	100	425.5	257.0	56.0	13	3289
PFOS	100	19,619.8	11,681.0	3082.5	1819	282,051
ΣPFSA	100	20,314.0	12,418.5	3148.3	1883	287,754
PFOA	100	421.9	240.5	58.4	10	3708
PFNA	100	2106.9	1319.0	231.4	129	11,713
PFDcA	100	1061.6	724.0	99.2	8	4911
PFUnA	100	3030.5	2539.5	203.6	667	13,163
PFDoA	100	1501.8	1234.5	103.6	8	5318
PFTriA	100	3371.0	2908.0	222.2	117	13,349
PFTeA	100	562.9	425.0	52.7	10	3405
ΣPFCA	100	12,056.4	9546.0	780.5	1765	41,327
ΣPFAS	100	32,370.4	22,045.0	3473.4	3648	296,475

Quantification was performed using internal standard calibration with isotopic dilution.

The OC-pesticides were α-, β-, γ-hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), chlordanes (oxychlordane and *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), Mirex, *p,p′*-diklordifenyltrikloretan (*p,p′*-DDT), *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE), *p,p′*-dichlorodiphenyldichloroethane (*p,p′*-DDD). The following PCB-congeners were determined: PCB-28/31, -52, -101, -99, -105, -118, -138, -153, -180, -183, -187 and 194.

2.4.2. PFAS analysis

1–1.5 g of pre-weighed homogenized egg material was extracted using 8 ml acetonitrile, egg extracts were evaporated to 2 ml, respectively. Before extraction, all samples were spiked with 0.5 ng/μl of ^{13}C-labelled internal standards. Prior to quantification, each 0.5 ml of solution was spiked with 2 ng of 3,7-brPFDcA recovery standard and 0.1 ml was transferred to an autoinjector vial containing 0.1 ml of 2 mM NH_4OAc in HLB-water. Full details of the instrumental analysis are described in Hanssen et al. (2013). Ten μl of the extract was used to separate and analyse PFASs by ultrahigh pressure liquid chromatography triple-quadrupole mass-spectrometry (UHPLC-MS/MS). Data quantification was conducted with LCQuan software (Thermo Scientific). Unless specified, all PFASs refer to their linear isomers.

The following PFASs were measured: perfluorooctane sulfonamide (PFOSA), perfluorooctane sulfonic acid (PFOS), perfluoroheptanesulfonic acid (PFHpS), perfluorohexanesulfonic acid (*PFHxS*), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoate acid (PFDcA), perfluorododecanoic acid (PFDoA), perfluorotetradecanoic acid (PFTeA), perfluorotridecanoic acid (PFTriA) and perfluoroundecanoate (PFUnA).

2.5. Statistical analyses

We carried out the statistical analyses and plotting results in R (R Core Team, 2021). All tests were two-tailed, and we rejected the null hypothesis at an *α*-level of 0.05. We used Wald statistics to test if estimated parameters were significantly different from zero. Similar to Bustnes et al. (2017), we assessed multiple co-linearity among our *a priori* defined predictors of interest before any model fitting (Zuur et al., 2010, see also Næss et al., 2011 for an example of how to deal with this is practice). These predictors included: NAO*w*; our two dietary indices (*i.e.*, the index of trophic-level and marine input measured as δ^{15}N and δ^{13}C, respectively); laying date (the date when the egg-laying started: Laying date); and time (Year). Among the remaining predictors, the most problematic relationships were between δ^{15}N and Year ($r = -0.29$) and Year and Laying date ($r = 0.22$). Still, we considered neither correlation unsuitable, and did not exclude them from the candidate models set, which ended up consisting of 16 different models (SI Table S1). We rescaled and ranked each model in each analysis relative to the model with the lowest second-order Akaike's Information Criterion (AICc) value (Δ_i denotes this difference for model *i*). We selected the simplest model (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).

Common to ecotoxicological studies in general, our dataset also consists of many correlated contaminant measures (*e.g.*, Bustnes, 2006). In order to establish how correlated the different OHCs were, a Principal Component Analysis (PCA) using the contaminants as the input variables was applied using the *princomp*, based on the correlation matrix, function in the MASS-library (Venables and Ripley, 2002; Everitt, 2004). In this assessment, we excluded PFOSA (77 %), α-HCB (65 %), PCB52 (37 %) and *cis*-chlordane (34 %) due to their many observations ≤ the Limit of Detection (LOD; percentages refer to the number of observations). 93.94 % ($n = 33$) of the compounds showed statistically significant correlations with either the 1st or the 2nd Principal Component (PC). Moreover, the cumulative variance in the original variables explained by PC1-2 was 44.94 % (66.21 % for PC1-4). Yet of more importance, the PCA separated the contaminates from each other based on their chemical properties.

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

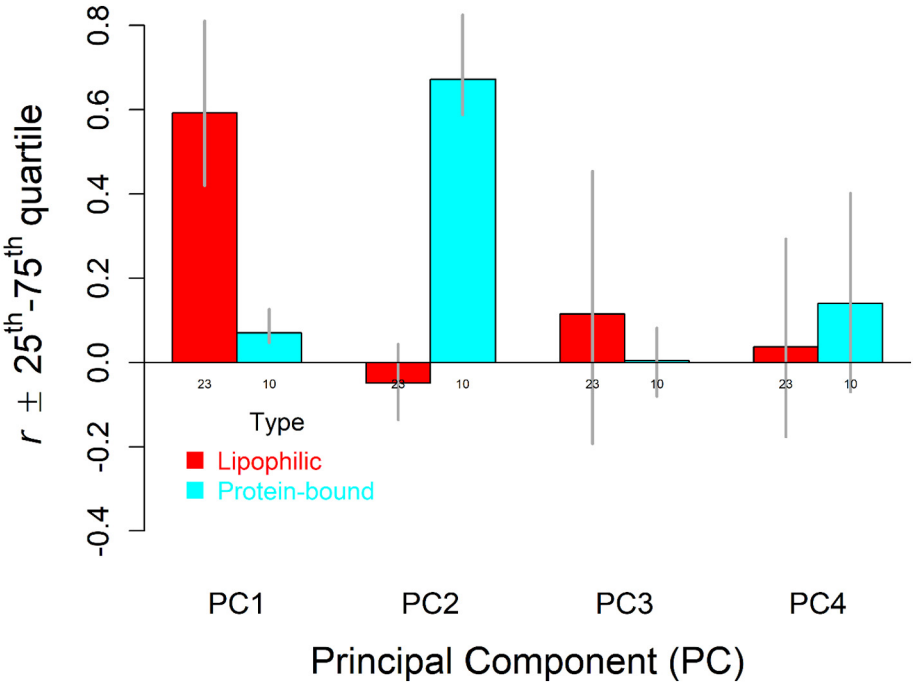

Fig. 1. The average Pearson's product-moment correlation (*r*) between contaminant concentrations and the scores for the Principal Component (PCs; in a Principle Component Analyses), grouped on the substances chemical properties (Lipophilic = OCs and Protein-bound = PFASs). The bars show the average for each group, whereas the lines show the variability of the correlations (showing the 25th and 75th percentiles). The 1st PC, which explained 28.67 % of the original variance in the actual concentrations, clearly separated the lipophilic from the protein-bound substances. The same pattern was also apparent using the 2nd PC, which explained 16.27 % of the original variance.

as its minimum value (using the same method as in Bustnes et al., 2022a). If <15 % of samples were ≤LOD we fitted linear models using the *lm* function, while if >50 % of the samples were ≤LOD we fitted logistic models to a binary response (observations were then classified as 0 = ≤LOD; 1 ≥ 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 we fitted these models to data). However, if 15 and 50 % of the samples were ≤LOD, 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 and Moler, 2020 for how these models were specified). Thomas et al. (2016) provide more details on the tobit-models we applied.

3. Results and discussion

PCBs ($\bar{x}$ = 62.3 %) and DDTs ($\bar{x}$ = 23.3 %), made up the bulk of the contaminant load in the goldeneye eggs across all years (Table 1). PFASs made up 10.5 % on average, PFOS being the dominating compound ($\bar{x}$ = 6.2 %). Moreover, there were no correlations between OC and PFAS concentrations; *i.e.* the eggs with high OC loads did not show high PFAS loads (Fig. 1). This is similar to findings in both marine- (Bustnes et al., 2008) and terrestrial bird species (Bustnes et al., 2015). Hence, these two OHC classes seem to have different exposure pathways into the food chain of goldeneyes and subsequently their eggs; *e.g.* through different dietary sources and different bioaccumulation characteristics during the various stages of egg formation (Hitchcock et al., 2019).

Unexpectedly, the lipid contents in the eggs declined linearly, from ~30 % to ~20 %, over the study period ($F_{1,\ 91}$ = 60.28, P < 0.01, Fig. 2A). This seems to be caused by dietary changes as there was a corresponding decline in $\delta^{15}N$ over time ($F_{1,99}$ = 9.31, P < 0.01, Fig. 2B), as well as a positive relationship between $\delta^{15}N$ and lipids ($F_{1,91}$ = 6.06, P = 0.02, Fig. 2C). In addition, the lipid content in 2018 was as high as in the early years of the study (Fig. 2A). This suggests that water evaporation (leading to less egg mass and thus higher lipid proportion) was not the cause of the eggs' declining lipid content during storage. Moreover, the goldeneyes seemed to have increased their feeding at lower trophic levels. Possibly, climate change has reduced the period of ice cover, allowing goldeneyes to reduce the input of nutrients from winter locations in egg production over time. There are no data on the diet of goldeneyes from this region. Still, in other areas, the species is known to feed on molluscs, crustaceans, and small fish (Cramp and Simmons, 1977), and changes in the proportions of different prey may have reduced the level of lipids in the eggs. For example, an earlier ice break-up will lead to a longer growing season, which may impact the food web structure. Since the lipid content changed, we also ran the models for the lipophilic OCs on lipid-normalized concentrations to check if the trends changed or if different variables were associated with the lipid-normalized- compared to the wet weight concentrations (see below).

Most of the OCs measured in this study were banned in many countries 35–50 years ago, and environmental concentrations declined strongly worldwide in the 1980s and 1990s (Loganathan and Kannan, 1994; Hebert et al., 1999; Bignert et al., 1998; Helander et al., 2002; Bustnes et al., 2007). Hence, we predicted declining temporal trends over the two decades covered by this study, and when estimating the impact of the selected climate sensitive factors, we always kept time (year) in the statistical models (plots of the selected models for the compounds not presented in the main text are found in SI, Figs. S1–28). Of the 13 banned OC-pesticides analysed, the wet-weight concentrations of seven showed significant negative trends in the selected models (Table 2); *i.e.* the trends for α- and γ-HCH, *cis*-chlordan and *cis*-nonachlor being strongly negative. Four compounds showed no trends, and two showed positive trends (Table 2, Figs. 3–7); *i.e.* the trend for β-HCH was marginally significant, whereas HCB showed a strong positive trend. Four of the 12 PCB congeners showed significant negative trends (PCB28/31, −52, −101, and 194), whereas the other congeners were stable (Table 2). Thus, the wet weight concentrations of the dominant OHCs, such as *p,p′*-DDE and persistent PCB congeners, were

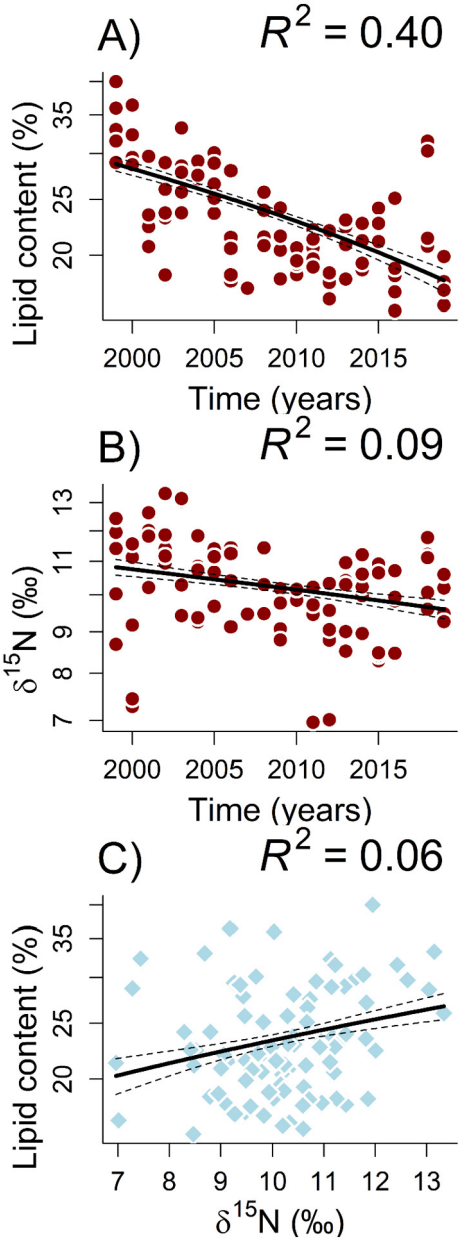

Fig. 2. Relationships between: A) Lipids and year; B) $\delta^{15}N$ and year; C) $\delta^{15}N$ and lipids in goldeneye eggs from central Norway, 1999–2019.

stable, while less persistent OCs, with relatively low concentrations declined. It is possible that a major decline of OCs in goldeneyes took place in the 1980s and 1990s, soon after the bans, as seen in some other species (Bignert et al., 1998; Helander et al., 2002; Bustnes et al., 2007), and that the levels have reached equilibrium states. The increasing concentrations of HCB may not be surprising since studies of both 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). Unlike most other OCs, emissions and releases of HCB continue as it is a byproduct of the manufacturing of chlorinated compounds and its industrial- and combustion-processes (Barber et al., 2005). Moreover, the consequences of decreasing lipid contents over the study period were increasing concentrations of some OCs on a lipid weight basis, even if the wet weight concentrations were stable. This was especially strong for HCB, but also the case for *p,p′*-DDE and some persistent PCB congeners (SI Table S2). Hence, the developing goldeneye embryos may have been exposed to higher OC concentrations in lipids over time. Indeed, *p,p′*-DDE and HCB are known to

Table 2
Variables in selected models (AICc) for different OC-pesticides and PCB-congeners in goldeneye eggs from central Norway. z/t: z GLM models; t in LM and VGLM models (for details see Materials and method).

Compound	Model type	Parameter	Estimate	Std. error	t/z	P
OC-pesticides						
HCB	LM	Intercept	7.959	0.021	379.770	<0.00001
	LM	Year	0.011	0.004	3.082	0.0027
	LM	$\delta^{13}C$	0.013	0.006	2.023	0.046
	LM	$\delta^{15}N$	0.049	0.017	2.828	0.0058
	LM	R^2	0.161			
α-HCH	GLM	Intercept	−2.520	0.758	−3.322	0.00089
	GLM	Year	−0.734	0.174	−4.221	0.00002
β-HCH	LM	Intercept	7.173	0.128	55.866	<0.00001
	LM	Year	0.038	0.021	1.790	0.077
	LM	R^2	0.034			
γ-HCH	VGLM	Intercept 1	2.858	0.164	17.401	<0.00001
	VGLM	Intercept 2	−0.037	0.087	−0.426	0.670
	VGLM	Year	−0.239	0.022	−10.795	<0.00001
	VGLM	Year^2	−0.002	0.004	−0.602	0.547
	VGLM	$\delta^{15}N$	0.283	0.091	3.123	0.002
	VGLM	R^2	0.504			
Oxychlordane	LM	Intercept	8.076	0.062	130.045	<0.00001
	LM	Year	−0.021	0.011	−2.015	0.047
	LM	$\delta^{13}C$	0.078	0.019	4.061	0.00011
	LM	$\delta^{15}N$	0.163	0.051	3.205	0.0019
	LM	R^2	0.294			
trans-Chlordane	LM	Intercept	5.664	0.085	66.287	<0.00001
	LM	Year	−0.019	0.014	−1.369	0.175
	LM	$\delta^{13}C$	0.075	0.026	2.850	0.0054
	LM	R^2	0.104			
cis-Chlordane	VGLM	Intercept 1	4.309	0.398	10.815	<0.00001
	VGLM	Intercept 2	0.069	0.088	0.785	0.43249
	VGLM	Year	−0.069	0.022	−3.213	0.0013
	VGLM	Year^2	0.007	0.004	1.649	0.099
	VGLM	Laying date	−0.077	0.024	−3.148	0.002
	VGLM	$\delta^{13}C$	0.164	0.041	4.014	0.00006
	VGLM	$\delta^{15}N$	0.479	0.105	4.571	<0.00001
	VGLM	R^2	0.299			
trans-Nonachlor	LM	Intercept	7.936	0.311	25.495	<0.00001
	LM	Year	−0.020	0.017	−1.229	0.222
	LM	Laying date	−0.033	0.017	−1.966	0.052
	LM	$\delta^{13}C$	0.087	0.029	3.017	0.0033
	LM	$\delta^{15}N$	0.168	0.079	2.132	0.036
	LM	R^2	0.197			
cis-Nonachlor	LM	Intercept	6.153	0.067	91.644	<0.00001
	LM	Year	−0.044	0.011	−3.855	0.00022
	LM	$\delta^{13}C$	0.099	0.021	4.758	0.00001
	LM	$\delta^{15}N$	0.319	0.055	5.786	<0.00001
	LM	R^2	0.508			
Mirex	LM	Intercept	6.097	0.073	83.891	<0.00001
	LM	Year	−0.006	0.012	−0.492	0.624
	LM	R^2	0.003			
p,p′-DDT	LM	Intercept	8.868	0.142	62.573	<0.00001
	LM	Year	−0.057	0.023	−2.449	0.016
	LM	R^2	0.062			
p,p′-DDD	LM	Intercept	7.600	0.446	17.058	<0.00001
	LM	Year	−0.016	0.024	−0.678	0.500
	LM	Laying date	−0.049	0.024	−2.031	0.045
	LM	$\delta^{13}C$	0.093	0.041	2.249	0.027
	LM	$\delta^{15}N$	0.317	0.113	2.808	0.0061
	LM	R^2	0.175			
p,p′-DDE	LM	Intercept	10.931	0.098	111.592	<0.00001
	LM	Year	0.015	0.016	0.911	0.365
	LM	R^2	0.009			
PCB-congeners						
PCB28/31	LM	Intercept	7.441	0.089	83.905	<0.00001
	LM	Year	−0.031	0.015	−2.162	0.033
	LM	R^2	0.049			
PCB52	VGLM	Intercept 1	5.346	0.488	10.949	<0.00001
	VGLM	Intercept 2	0.278	0.090	3.078	0.0021
	VGLM	Year	−0.024	0.025	−0.946	0.34433
	VGLM	Year^2	0.023	0.005	4.651	<0.00001
	VGLM	Laying date	−0.126	0.030	−4.176	0.00003
	VGLM	$\delta^{15}N$	0.293	0.124	2.360	0.018
	VGLM	R.sq	0.043			
PCB101	LM	Intercept	6.855	0.329	20.807	<0.00001
	LM	Year	−0.063	0.018	−3.596	0.00053
	LM	Laying date	−0.058	0.018	−3.261	0.0016
	LM	$\delta^{15}N$	0.193	0.084	2.312	0.023
	LM	R^2	0.325			
PCB99	LM	Intercept	8.926	0.142	63.013	<0.00001
	LM	Year	−0.025	0.023	−1.075	0.285
	LM	R^2	0.013			
PCB105	LM	Intercept	8.866	0.095	93.067	<0.00001
	LM	Year	−0.006	0.016	−0.402	0.689
	LM	$\delta^{13}C$	−0.069	0.029	−2.357	0.021
	LM	R^2	0.060			
PCB118	LM	Intercept	10.108	0.094	107.268	<0.00001
	LM	Year	0.010	0.016	0.611	0.543
	LM	$\delta^{13}C$	−0.067	0.029	−2.292	0.024
	LM	$\delta^{15}N$	0.148	0.077	1.918	0.058
	LM	R^2	0.095			
PCB138	LM	Intercept	10.655	0.089	119.672	<0.00001
	LM	Year	0.014	0.015	0.945	0.347
	LM	$\delta^{13}C$	−0.060	0.028	−2.192	0.031
	LM	$\delta^{15}N$	0.169	0.073	2.311	0.023
	LM	R^2	0.108			
PCB153	LM	Intercept	11.214	0.091	123.230	<0.00001
	LM	Year	0.018	0.016	1.155	0.251
	LM	$\delta^{15}N$	0.164	0.075	2.197	0.031
	LM	R^2	0.054			
PCB180	LM	Intercept	10.071	0.086	117.746	<0.00001
	LM	Year	−0.009	0.014	−0.658	0.512
	LM	$\delta^{13}C$	−0.057	0.026	−2.167	0.033
	LM	R^2	0.053			
PCB183	LM	Intercept	8.471	0.088	96.446	<0.00001
	LM	Year	0.007	0.015	0.493	0.623
	LM	$\delta^{15}N$	0.154	0.072	2.132	0.036
	LM	R^2	0.049			
PCB187	LM	Intercept	9.568	0.083	115.197	<0.00001
	LM	Year	0.003	0.014	0.181	0.857
	LM	$\delta^{15}N$	0.170	0.068	2.490	0.015
	LM	R^2	0.068			
PCB194	LM	Intercept	7.671	0.088	87.506	<0.00001
	LM	Year	−0.029	0.014	−1.995	0.049
	LM	$\delta^{13}C$	−0.084	0.027	−3.103	0.0026
	LM	R^2	0.128			

be embryotoxic compounds (Boersma et al., 1986; Helander et al., 2002), though overall potential adverse effects are impossible to conclude on.

For PFASs, there were significant negative temporal trends for PFOSA and PFHxS, and marginal significant for PFHpS in the selected models (Table 3, Figs. 8–10). None of the PFCAs showed significant temporal trends, except for a positive trend for PFNA (Fig. 10). This was unexpected since most wildlife studies have found that major compounds such as PFOS to be declining, and many PFCAs to be increasing (Holmström et al., 2010, Ahrens et al., 2011, Land et al., 2018; Miller et al., 2015, Jouanneau et al., 2020, Sun et al., 2019, Pereira et al., 2015), also in the terrestrial ecosystem adjacent to our study area (Ahrens et al., 2011; Bustnes et al., 2015; Bustnes et al., 2022b).

The lack of measurable temporal trends for most OHCs might be partly caused by the fact that goldeneyes alternate between different habitats, and there are several examples of temporal trends of OHCs being modified by other, often climate sensitive factors, such as diet (Hebert et al., 2000, 2008; Hebert and Weseloh, 2006; Bustnes et al., 2010, 2011, 2015; Elliott et al., 2012). The diet of goldeneyes is strongly dependent on climate since several climate-related factors (temperature, ice, wind, precipitation *etc.*) will influence their habitat use (Clark et al., 2014). Dietary tracers, in particular stable nitrogen and carbon isotopes, have been used extensively in ecotoxicological studies, and proven useful in explaining variation in

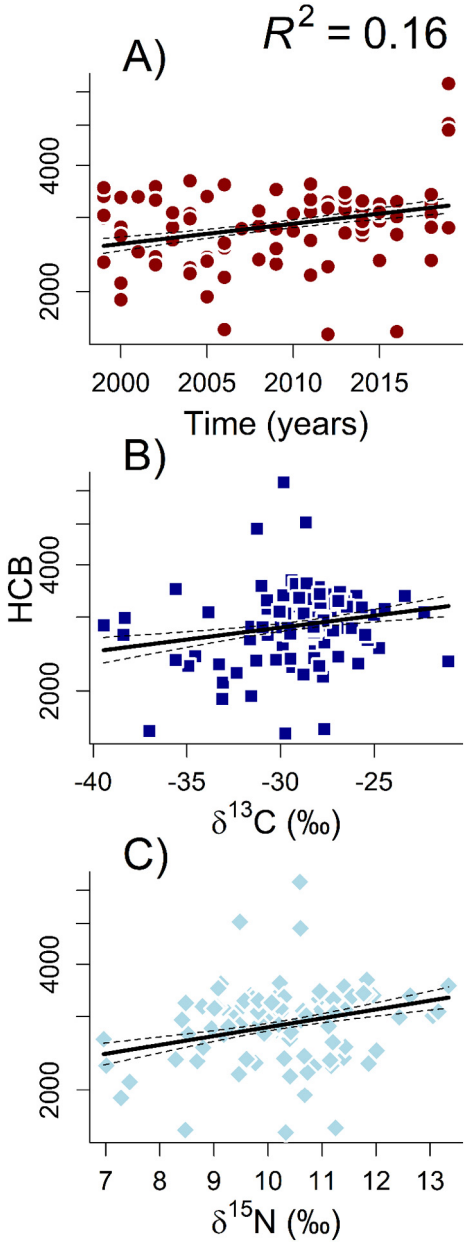

Fig. 3. Relationships between HCB concentrations (pg/g, wet weight) and year (A), and stable isotopes (B: δ^{13}C and C: δ^{15}N) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

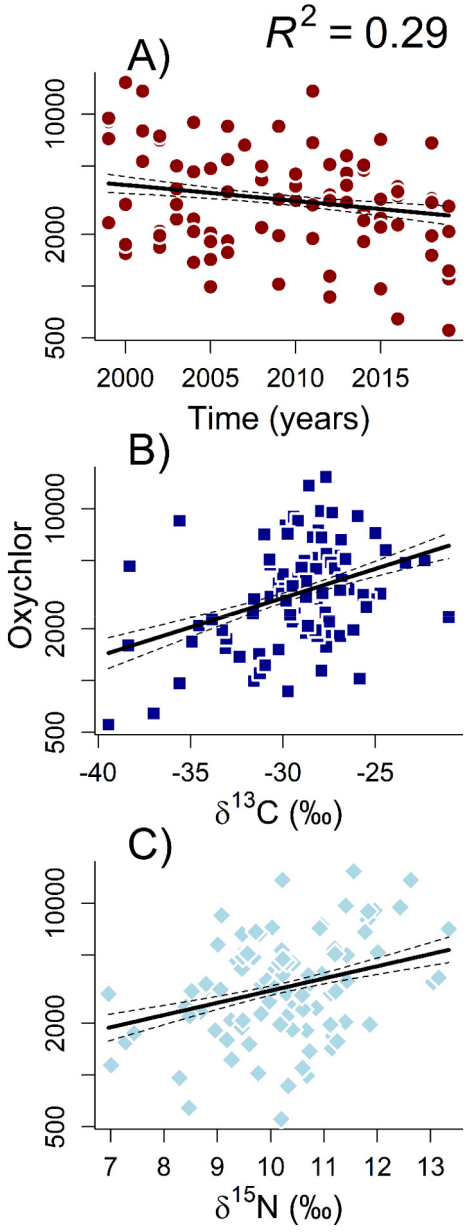

Fig. 4. Relationships between Oxychlordane concentrations (pg/g, wet weight) and year (A), and stable isotopes (B: δ^{13}C and C: δ^{15}N) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

OHCs between populations and individuals (Elliott et al., 2009; Gebbink et al., 2011; Eulaers et al., 2013; Bustnes et al., 2013; Lopez-Antia et al., 2021). The trophic proxy δ^{15}N has been used as an indicator of biomagnifying OCs in feathers, eggs and blood of various bird species (Elliott et al., 2009; Eulaers et al., 2013; Bustnes et al., 2013). In the present study, δ^{15}N showed strong significant positive relationships to seven out of 13 OC-pesticides, particularly for some chlordanes (Table 2, Fig. 4). However, the relationships were insignificant for α- and β-HCH, *trans*-chlordane, Mirex, *p,p′*-DDT and notably *p,p′*-DDE (Table 2). Similarly, δ^{15}N was positively related to the concentrations of PCBs, significantly so for six out of the 12 PCB-congeners, but the associations were relatively weak (Table 2, Figs. 5–7). Hence, for OCs the relationship with trophic position reveals a mixed picture, in which some compounds showed solid relationships, whereas other relatively similar compounds showed no association. The reason for this is not known, but recent studies have shown that different groups of lipophilic contaminants may accumulate differently in the freshwater food web depending on transfer pathways and biological traits (Windsor et al., 2019, 2020). That is, ducks feeding at the same trophic level but on different prey species may be exposed to different contaminants. Another complicating factor is that δ^{15}N has declined over the years, suggesting a dietary change (see above), which may confound some of the expected relationships; *i.e.* declining δ^{15}N values predict declining OHC concentrations. Finally, since we do not know exactly where the goldeneyes in this study winters, we are unable to adjust for potential differences in δ^{15}N baselines, which might influence the relationships we are measuring. However, SIAs in our study were exclusively used to estimate relative dietary contributions, so we are confident that this should not affect our conclusions.

Although PFASs have lower biomagnifying potential than many OCs, a natural prediction would be a significant association to δ^{15}N. With few exceptions (Gebbink et al., 2011; Lopez-Antia et al., 2021), however, bird studies have failed to find such relationships (Bustnes et al., 2013;

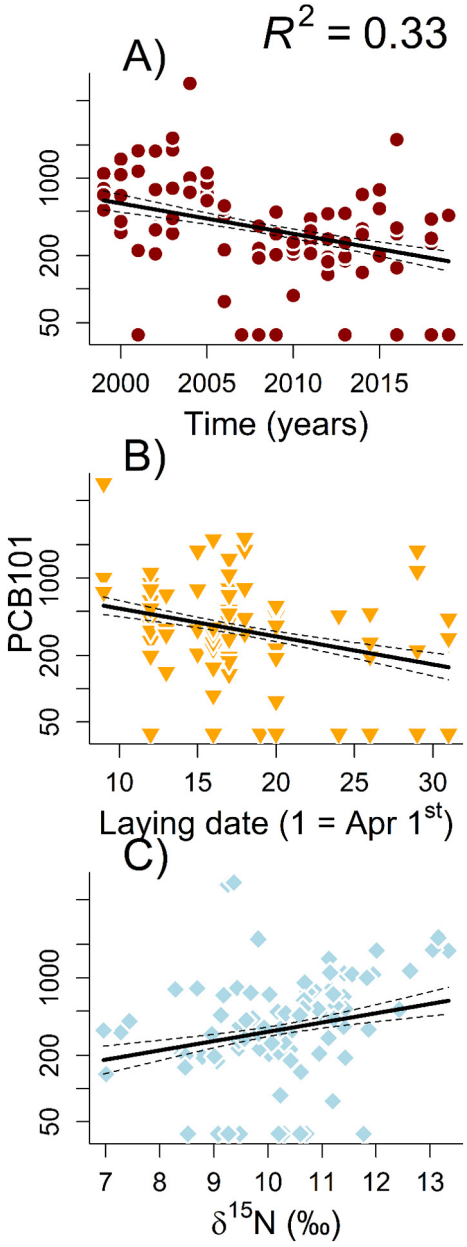

Fig. 5. Relationships between PCB101 concentrations (pg/g, wet weight) and year (A), egg laying date (B) and $\delta^{15}N$ (C), and in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

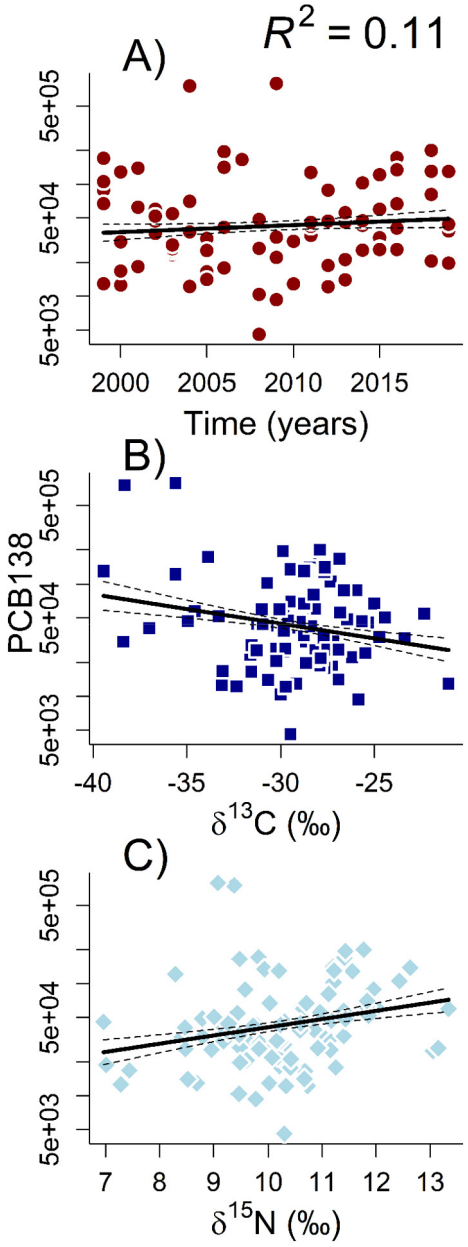

Fig. 6. Relationships between PCB138 concentrations (pg/g, wet weight) and year (A), and stable isotopes (B: $\delta^{13}C$ and C: $\delta^{15}N$) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

Gómez-Ramírez et al., 2017; Haukås et al., 2007; Leat et al., 2013; Løseth et al., 2019, Carravieri et al., 2020; Vicente et al., 2015), and the present study also showed no association between $\delta^{15}N$ and any of the 12 PFAS measured. The general lack of associations between PFASs and $\delta^{15}N$ may be multifaceted and could depend on a wide range of factors, including study designs, sample sizes, compound-dependent trophic magnification capabilities food web structure *etc.*, and discussing such potential causes is beyond the scope of the present study.

The dietary proxy $\delta^{13}C$ is commonly used to assess the proportion of marine input in the diet of many coastal birds, since coastal diets are more ^{13}C-enriched than terrestrial and freshwater ones (Kelly, 2000; Bond et al., 2007; Eriksson et al., 2016; Kloskowski et al., 2019). In sea ducks which stay in coastal habitats during winter and breed in freshwater, both endogenous and exogenous nutrients sources may be exploited for egg formation. Although marine input in eggs of such species seems low, early laid eggs have a higher probability of containing endogenously derived nutrients (Hobson, 2006; DeVink et al., 2011; Kloskowski et al., 2019). In general, a $\delta^{13}C$ value lower than −20 ‰ suggests a diet with low marine influence (Hobson, 1987; Eriksson et al., 2016), but it is recommended to lipid normalize $\delta^{13}C$ in lipid-rich tissues since lipids are depleted in ^{13}C (Hobson, 2006). One such technique applied for homogenized eggs of aquatic birds is arithmetic corrections equations using C:N-ratios (Elliott et al., 2014). However, this has proven complicated in sea ducks, especially if they incorporate nutrients from distinct environments (Oppel et al., 2010). In the present study, the uncorrected $\delta^{13}C$ values varied between −21 and − 39.5 ‰ (mean = 29.2 ‰), and using the non-linear equation proposed by Elliott et al. (2014) altered the mean by −1.16 ‰, but had negligible effects on the predictive power of $\delta^{13}C$ as the correlation between the lipid-corrected and un-corrected $\delta^{13}C$ was very high (r = 0.999, df = 90, P < 0.001), and consequently we chose to run the statistical models without corrections.

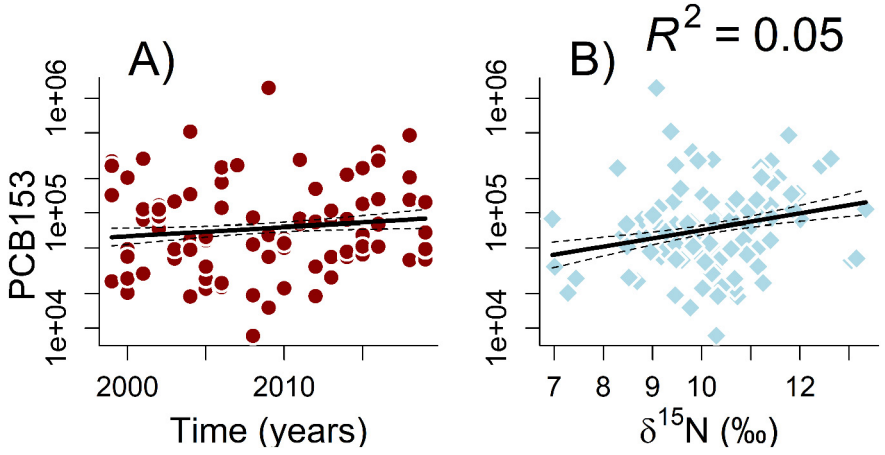

Fig. 7. Relationships between PCB153 concentrations (pg/g, wet weight) and year (A), and δ^{15}N (B) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

Although we could not firmly establish the source of nutrients in the present study, the low δ^{13}C values suggested that the eggs were produced predominantly by exogenous food resources from the freshwater environment. However, since the eggs were homogenized the dominating freshwater nutrients may have "overshadowed" a potentially low coastal/marine input in some early laid eggs (Hobson, 2006; DeVink et al., 2011; Kloskowski et al., 2019), since different compartments of the eggs are formed at different times (Nager, 2006). If there were marine nutrients in the eggs, the amount of contaminants sequestered into the eggs is expected to increase since endogenous lipid reserves are major storage compartments for lipophilic OHCs (Swift et al., 1993; Franson et al., 2004; Braune and Malone, 2006; Bustnes et al., 2012; Tomza-Marciniak et al., 2019). Among the OCs, δ^{13}C was significantly and positively associated with HCB, and all five chlordane compounds, the δ^{13}C effect being much stronger than the temporal changes for chlordanes (Table 2, Figs. 3, 4). There was also a slight positive effect of δ^{13}C on *p,p′*-DDD (Table 2). δ^{13}C, however, was negatively associated to concentrations of the PCBs, significantly for the following congeners: PCB52, -105, -118, -138, -180, and 194, although the effects were relatively weak (Table 2, Fig. 6). The reason for the variation between different OC groups is unclear but suggests that PCBs, to some extent, have different dietary sources than HCB and chlordanes (possibly some marine lipids). PCBs may originate more from the exogenous nutrient ingested in freshwater than the other compounds. Alternatively, it might depend on contaminants accumulating differently in the freshwater food web depending on transfer pathways and biological traits (Windsor et al., 2019, 2020).

δ^{13}C showed strong and positive associations with both PFSAs and PFCAs with the exception of PFOSA, PFDoA and PFTeA (Table 3, Figs. 8–10). This suggest that there may be traces of marine nutrients from the coast in the eggs with high PFAS levels since PFAS usually are higher in marine- than freshwater ecosystems (Bustnes et al., 2008, 2013; Eriksson et al., 2016; Lopez-Antia et al., 2021). The fact that the onset of egg laying (the date when the first egg was laid) was, in general, a positive predictor of the concentrations of PFAS (particularly for PFHpS and PFOS; Table 3, Fig. 8) further supported the hypothesis that some of the PFAS loads have a coastal marine origin. The mechanism may be that the egg laying starts late when the ducks arrive late on the lake, and they consequently have less time to feed locally before egg laying. In these situations, they may use more endogenous nutrient reserves acquired in winter for egg formation, either marine or freshwater nutrients. For PFCAs, however, egg laying date had only a significant positive effect on PFTeA, and negative effect on PFOA (Table 3). Similarly, the onset of egg laying had only minor effects in the models for OCs, being significantly negative for four low concentration compounds: *cis*-chlordane, *trans*-nonachlor, *p,p′*-DDD, PCB101 (Table 2). The fact that most OCs, except HCB and some chlordanes, showed no

Table 3
Variables in selected models (AICc) for different PFASs in goldeneye eggs from central Norway. z/t: z GLM models; t in LM and VGLM models (for details see Materials and method).

Compound	Model type	Parameter	Estimate	Std. error	t/z	P
PFSAs						
PFOSA	GLM	Intercept	−2.567	−4.084	−2.738	0.00004
	GLM	Year	−0.376	0.100	−3.742	0.0002
	GLM	δ^{15}N	0.485	0.253	1.919	0.055
PFHpS	LM	Intercept	3.549	0.368	9.646	<0.00001
	LM	Year	−0.032	0.019	−1.683	0.096
	LM	Laying date	0.067	0.020	3.383	0.00107
	LM	δ^{13}C	0.096	0.035	2.738	0.0075
	LM	R^2	0.200			
PFHxS	LM	Intercept	5.612	0.090	62.121	<0.00001
	LM	Year	−0.037	0.015	−2.527	0.013
	LM	δ^{13}C	0.093	0.028	3.317	0.0013
	LM	R^2	0.170			
PFOS	LM	Intercept	8.638	0.262	32.986	<0.00001
	LM	Year	−0.003	0.014	−0.247	0.806
	LM	Laying date	0.047	0.014	3.341	0.0012
	LM	δ^{13}C	0.079	0.025	3.154	0.0022
	LM	R^2	0.210			
PFCAs						
PFOA	LM	Intercept	6.193	0.329	18.849	<0.00001
	LM	Year	−0.001	0.017	−0.045	0.964
	LM	Laying date	−0.044	0.018	−2.508	0.014
	LM	δ^{13}C	0.193	0.032	6.111	<0.00001
	LM	NAO*w*	0.258	0.074	3.500	0.00074
	LM	R^2	0.362			
PFNA	LM	Intercept	7.191	0.079	91.067	<0.00001
	LM	Year	0.028	0.014	2.088	0.040
	LM	δ^{13}C	0.157	0.024	6.410	<0.00001
	LM	δ^{15}N	−0.142	0.065	−2.198	0.031
	LM	R^2	0.378			
PFDcA	LM	Intercept	6.598	0.094	70.001	<0.00001
	LM	Year	0.013	0.015	0.868	0.388
	LM	δ^{13}C	0.144	0.029	4.931	<0.00001
	LM	R^2	0.217			
PFUnA	LM	Intercept	7.848	0.057	136.760	<0.00001
	LM	Year	0.013	0.009	1.350	0.180
	LM	δ^{13}C	0.074	0.018	4.147	0.00008
	LM	R^2	0.172			
PFDoA	LM	Intercept	7.099	0.083	85.322	<0.00001
	LM	Year	0.018	0.014	1.307	0.195
	LM	R^2	0.019			
PFTriA	LM	Intercept	7.965	0.059	135.048	<0.00001
	LM	Year	−0.001	0.010	−0.073	0.942
	LM	δ^{13}C	0.042	0.018	2.285	0.025
	LM	R^2	0.056			
PFTeA	LM	Intercept	5.575	0.235	23.708	<0.00001
	LM	Year	0.013	0.012	1.063	0.291
	LM	Laying date	0.029	0.013	2.249	0.027
	LM	R^2	0.079			

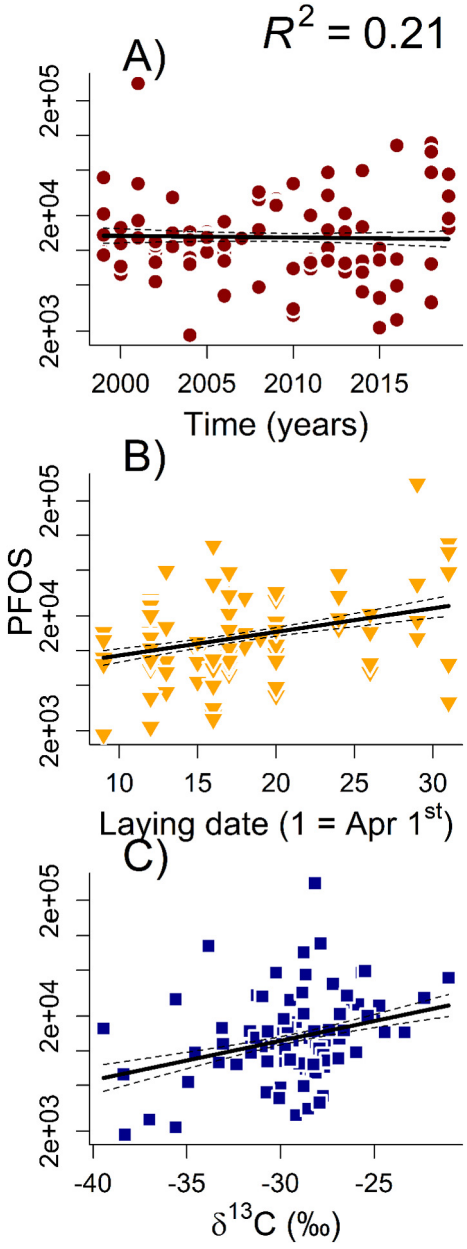

Fig. 8. Relationships between PFOS concentrations (pg/g, wet weight) and year (A), egg laying date (B) and $\delta^{13}C$ (C), and in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

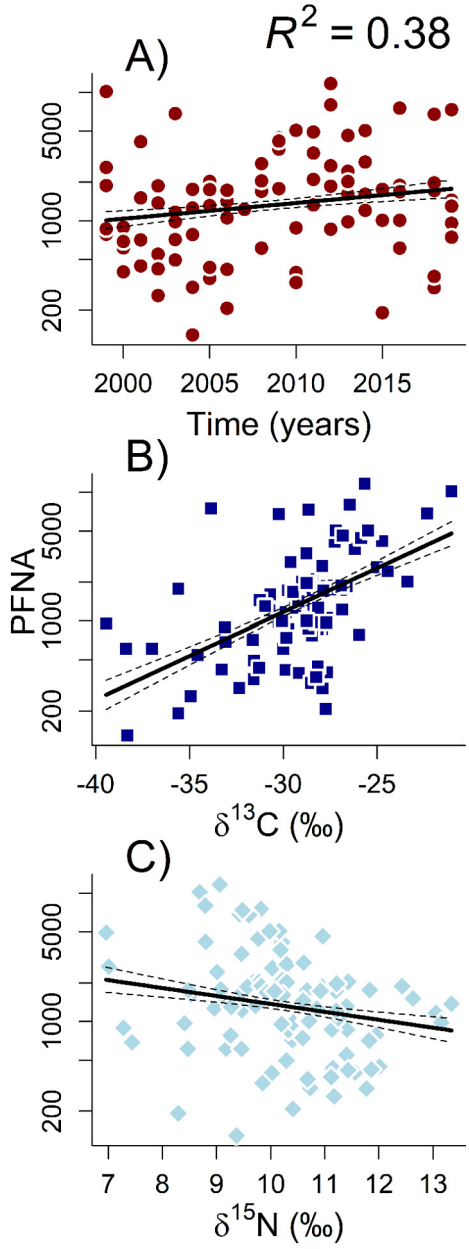

Fig. 9. Relationships between PFNA concentrations (pg/g, wet weight) and year (A), stable isotopes (B: $\delta^{13}C$ and C: $\delta^{15}N$) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

relationship to $\delta^{13}C$ and laying date suggests that they originate predominantly from exogenous lake nutrients.

The physical climate may have a potential impact on the transport and accumulation pathways of POPs (Noyes et al., 2009), and several studies have found such associations in species from different ecosystems (Hebert et al., 2000; Bustnes et al., 2010, 2011, 2015). In the present study, we used the NAO_w (see Materials and method) as an index of winter climate, predicting that mild winters would allow the ducks to arrive early and produce eggs predominantly from lake nutrients. However, the only significant effect of NAO*w* was a positive effect on PFOA (Table 3). Hence, our study did not suggest that winter climate plays a vital role in explaining variability in the accumulation of OHCs in goldeneye eggs. The significant relationship uncovered for PFOA, however, is of relevance, as this is one of the most ubiquitous PFASs. PFOA is highly water-soluble and can bind to particles, all aspects of a compound prone to multi-hopper transport behavior, which is highly affected by changing weather patterns such as increased precipitations during winter in the form of snow.

Because the lipid contents of the eggs changed over the years, we tested if lipid normalization altered the effect of the different predictors on the concentrations of lipophilic OCs. When running the same models on lipid weights of the 25 OCs as we did for wet weight, the temporal trends changed significantly for seven compounds; *i.e.* for *p,p′*-DDE, PCB138 and -153, the results changed from no trends to weak positive relationships with year, whereas for oxychlordane, PCB28, -52 and -194 the trends changed from negative to no trends (SI Table S2). However, only for three compounds did lipid-normalization result in different covariates included in the selected models. For HCB and *p,p′*-DDD, all other variables than year were excluded (SI Table S2). In addition, for PCB52 laying date was included in the best model for lipid weight, and for PCB118, $\delta^{15}N$ was excluded in the selected model (SI Table S2). Hence, the changing

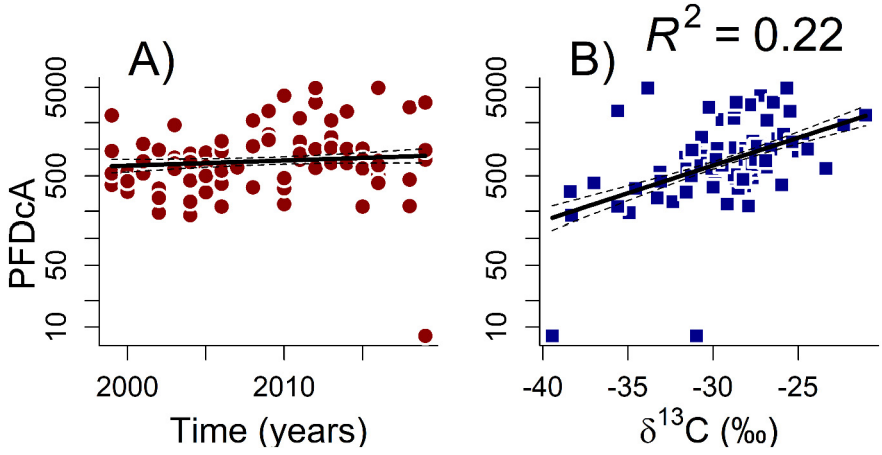

Fig. 10. Relationships between PFDcA concentrations (pg/g, wet weight) and year (A), and δ^{13}C (B) in eggs of goldeneyes. Plots based on the selected model. Data from central Norway, 1999–2019.

lipid concentrations over time had little impact on the effects of climate-sensitive factors on the accumulation of OCs in goldeneye eggs.

The goldeneye is a good model species for gaining insight into how future climate change may affect the exposure of OHCs in aquatic predators, since it alternates between different habitats and is sensitive to factors bound to change in a warming climate. Firstly, the present study demonstrated the complications of monitoring temporal trends of OHCs: *i.e.* although most of the studied compounds have been banned for several decades the concentrations of the major compounds in this OHC mixture (>90 %) showed no temporal trends. Rather, we found that other variables than time in general were more important in explaining variations in OHCs concentrations. Hence, exposure to OHCs in goldeneyes in our study area is significantly determined by climate-sensitive factors. The reduction of emission and environmental decline also plays a minor role suggesting that future climate change will have repercussions in terms of changing OHC loads. A notable development in the goldeneye is the changing diet (δ^{15}N) over the years, which may or may not be an indirect effect of climate change, and the impact it seemed to have on the OCs in the lipids and possible increased exposure to the embryos. However, how the predicted climate changes will affect the exposure in the long run is still unclear. A warmer climate will lead to less ice, and there may be less need for the ducks to stay in more polluted coastal environments, suggesting that PFAS exposure and possibly some OCs may decline. Increased strengths of precipitation and air currents, on the other hand, might cause the more volatile and water-soluble OHCs to increasingly travel over long distances from emission sources to northern regions as central Norway. A changing OHC composition towards smaller molecules and lower halogenation degree in bird eggs might be a result.

CRediT authorship contribution statement

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

Data availability

Data will be made available on request.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157667.

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