



Temporal trends (1968–2019) of legacy persistent organic pollutants (POPs) in seabird eggs from the northeast Pacific: Is it finally twilight for old POPs?

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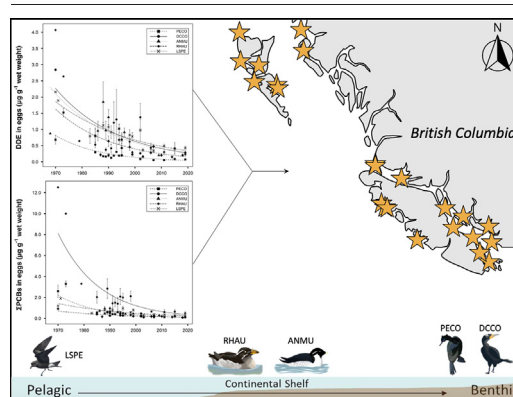
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HIGHLIGHTS

- Seabird eggs were collected from the Pacific coast (1968–2019) and analyzed for POPs.
- Σ PCBs, *p,p'*-DDE, β -HCH, HCB, and oxychlorane dominated the contaminant profile.
- Most POPs significantly declined or showed no directional change over time.
- In some years, slight increases in photomirex and β -HCH were observed in the eggs of some species.
- SIAs suggested no major changes in trophic position or baseline food web signature.

GRAPHICAL ABSTRACT



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ABSTRACT

Legacy persistent organic pollutants (POPs), such as organochlorine pesticides (OCs) and polychlorinated biphenyls (PCBs), are known to persist in the marine environment; however, whether concentrations of these POPs have decreased or stabilized from Canada's Pacific coast in recent years is unclear. Here, we examined temporal trends of various legacy POPs in the eggs of five seabird species; two cormorants (*Nannopterum auritum* and *Urile pelagicus*), an auklet (*Cerorhinca monocerata*), a murrelet (*Synthliboramphus antiquus*), and a storm-petrel (*Hydrobates leucorhous*), sampled 1968 to 2019 from 23 colonies along the Pacific coast of British Columbia, Canada. The contaminant profile in the eggs of all species and sampling years was dominated by Σ PCBs, followed by Σ DDT (mostly *p,p'*-DDE), Σ HCH (β -HCH), Σ CHLOR (oxychlorane), and Σ CBz (HCB). Σ OC and Σ PCB concentrations were generally higher in double-crested cormorant eggs than in the other four species. The majority of legacy POPs are either significantly declining (e.g. *p,p'*-DDE, HCB, HE, oxychlorane, Σ PCBs) or showing no directional change over time (Σ Mirex) in the eggs of our monitoring species. Contaminants such as α -HCH, *cis*- and *trans*-chlordanes, *p,p'*-DDT, dieldrin, and octachlorostyrene also showed evidence of downward trends, largely influenced by non-detect values during more recent sampling periods. Increasing trends were observed for β -HCH in the eggs of some species; however, mean concentrations eventually returned to early 2000 levels by the end of the study period. Although bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ egg values varied interannually, compound-specific amino acid analyses suggested no major changes in trophic position or baseline food web signature. Temporal trends observed here were comparable to those found in other seabird species and pelagic food webs. As most legacy POPs in our data set were at very low levels in recent years, we support the general consensus that it is indeed the twilight years for old POPs, and we attribute these declines largely to voluntary regulations and international restrictions on the production and use of these compounds, and thus their release into the marine environment.

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1. Introduction

POPs, or Persistent Organic Pollutants, are defined under the United Nations Stockholm Convention effectively as persistent, bioaccumulative and toxic (PBT) organic compounds produced by human activities, and which can be moved around by natural environmental processes (UNEP, 2019). The original group of 12 POPs, the 'dirty dozen' are now commonly regarded as 'legacy' compounds so as to separate them from more recently released compounds, which are often regarded as 'emergent'. The legacy group includes a number of organochlorine insecticides (OCs), e.g. DDT, six cyclodienes, and toxaphene. Some compounds, such as mirex and hexachlorobenzene (HCB), had both insecticidal and industrial uses (Blus et al., 1996; Elliott et al., 1989). Similarly, polychlorinated biphenyls (PCBs) were widely used commercial chemicals, and finally the polychlorinated dibenzo-*p*-dioxins (PCDDs) and furans (PCDFs) were unintentionally produced and released from a range of human activities (Elliott et al., 1996).

Widespread use of the legacy pesticides and industrial compounds began post WWII and continued until varying dates in the 1970s (Elliott et al., 1988). PCB production continued in Europe until the late 1980s and in Russia until the 1990s, and DDT is still manufactured in India for disease vector control (van den Berg et al., 2017). Most economically and developed jurisdictions eventually introduced severe use restrictions on POPs due to the accumulating evidence that they had become global pollutants. Bird species were at the core of this story, initially because of acute mortality of songbirds and waterfowl (Carson, 1962; Flickinger and King, 1972; Stanley and Bunyan, 1979) and eventually higher-trophic level predatory birds in both highly industrialized (Bagge, 1975; Gilman et al., 1977) and remote locations (Risebrough et al., 1968). The earliest evidence of the ecotoxicological effects of that contamination were also largely identified and developed in marine and piscivorous birds, both on survival and reproduction (Ratcliffe, 1967; Hickey and Anderson, 1968; Anderson et al., 1975; Newton and Bogan, 1978; Spitzer et al., 1978; Grier, 1982). Those research studies ultimately led to the effective use of birds as sentinel species for various long-term contaminant monitoring programs.

Seabirds are particularly useful indicators for monitoring levels of POPs and other contaminants in marine systems because they are long-lived and feed at relatively high trophic levels in marine food webs (Borgå et al., 2004), suitably integrating contaminant exposure across space, time, and communities (Elliott and Elliott, 2013). Generally, seabird eggs can be used as a relatively non-invasive sampling matrix (that is also easily homogenized) to represent the contaminant burden in the female adult prior to and during the egg-laying period. For example, the hydrophobic and lipophilic nature of recalcitrant POPs, such as DDT and its metabolites, allows them to freely diffuse across the oocyte plasma membrane and be deposited with yolk lipids directly into eggs (Kestic et al., 2021). Such contaminants can also bind to endogenous yolk proteins synthesized in the liver, which then move by active transport into the egg yolk (Eng et al., 2012). In addition, the congregation of seabirds at breeding colonies allows for the collection of eggs from multiple individuals, facilitating a large sample size while providing adequate statistical power to detect an effect (Braune et al., 2019; Elliott and Elliott, 2013).

New technologies have improved the resolution of factors influencing spatial and temporal variation of legacy POPs. Stable isotope ratios have been widely used as biogeochemical tracers to distinguish the effects of diet and environmental contaminant concentrations (Burgess et al., 2013; Elliott et al., 2021a, 2021b; Kelly et al., 2007; Kelly et al., 2008; van den Brink et al., 2015; Braune et al., 2019). Stable isotopes of carbon ($\delta^{13}\text{C}$) can be used to characterize habitat use and foraging behaviour in nearshore versus offshore waters since benthic and littoral marine communities are often enriched in $\delta^{13}\text{C}$, compared to terrestrial (and some pelagic) communities that are depleted in $\delta^{13}\text{C}$ (de la Vega et al., 2019). Conversely, stable isotopes of nitrogen ($\delta^{15}\text{N}$) can provide information on trophic position because $\delta^{15}\text{N}$ values in body tissues increase systematically by ~ 3 to 5‰ per each trophic level (Kelly, 2000); however, variation in baseline or source $\delta^{15}\text{N}$ can influence $\delta^{15}\text{N}$ analyses (Elliott et al., 2021b). At the same time, compound-specific amino acids have enabled further insights into

ecological factors influencing trophic dynamics and baseline food web signatures of biomagnifying contaminants in birds (Hebert and Popp, 2018; Grenier et al., 2020; Elliott et al., 2021b; Bode et al., 2021; Jones et al., 2022) and changes in food chain structure and climate oscillations and their relationships with POPs (Hebert et al., 2000a; Kalia et al., 2021; Braune et al., 2014; Houde et al., 2019).

Long-term monitoring studies using avian indicators of marine and large aquatic systems have showed significant declines for most of the POPs by the early 1980s (Elliott et al., 1988, Elliott et al., 1989; Pekarik and Weseloh, 1998; Bignert et al., 1995; Braune et al., 2001; Harris et al., 2005). With some variation led spatially and by compound, such declines have continued to the present day at highly contaminated sites, such as the Laurentian Great Lakes (de Solla et al., 2016) and near remote locations in the Arctic (Braune et al., 2019). Here, we assessed the state and toxicological significance of legacy POP exposure at varying trophic levels by: 1) examining temporal trends of POPs in the eggs of double-crested cormorants (*Nannopterum auritum*; formerly classified within the genus *Phalacrocorax*), pelagic cormorants (*Urile pelagicus*), Leach's storm-petrels (*Hydrobatas leucorhous*), rhinoceros auklets (*Cerorhinca monocerata*), and ancient murrelets (*Synthliboramphus antiquus*) breeding off the northeast Pacific coast of British Columbia, Canada from 1968 to 2019; 2) comparing egg contaminant concentrations within and among nearshore, continental shelf, and offshore species; and 3) evaluating differences in trophic relationships, foraging habitat, and dietary uptake of contaminants using stable isotope analyses of bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, and multiple compound-specific amino acids.

2. Materials and methods

2.1. Study species

Seabird eggs were routinely collected as part of the long-term contaminants monitoring program initiated in 1968 by the Canadian Wildlife Service (Gilbertson et al., 1987). This marine ecosystem monitoring program is predicated on niche partitioning of seabird species by habitat and diet. For the purposes of our study, we have divided the system into three zones; nearshore, continental shelf, and offshore and selected appropriate seabird indicator species for each zone. Monitoring sites were selected based on colony size, past monitoring history, and accessibility. The nearshore zone is covered by two cormorants; the double-crested cormorant (*N. auritum*; DCCO) and pelagic cormorant (*U. pelagicus*; PECO). Cormorants are opportunistic feeders, typically preying on slow-moving and schooling fish species (e.g., herring), as well as benthic prey (Harris et al., 2005; Elliott and Elliott, 2013). Both cormorants will lay, on average, 3–4 eggs (range 1–11) per clutch and will often re-lay if an egg is lost (Van Tets, 1959).

The continental shelf zone is covered by two auklets; rhinoceros auklets (*C. monocerata*; hereafter auklet; RHAU) and ancient murrelets (*S. antiquus*; hereafter murrelet; ANMU). Auklets generally consume zooplankton before the egg-laying period and switch to a diet of fish (sand lance, herring, rockfish) throughout the breeding season (Hipfner et al., 2013). Auklets nest in burrows under dense canopy and vegetation, laying a single egg per clutch (Elliott et al., 2021a). Murrelets forage mostly on zooplankton (e.g. euphausiids) and small fishes, including juvenile sand lance, perch, and rockfishes (Vermeer et al., 1985). Murrelets nest in mature forests, often in a burrow or under natural cavities and lay 1–2 eggs per clutch (Miller et al., 2020).

The offshore zone is predominantly covered by the Leach's storm-petrel (*H. leucorhous*; hereafter storm-petrel; LSPE). The LSPE is a trans-equatorial migrant ranging widely throughout the Pacific and Atlantic Oceans (Pollet et al., 2019). Storm-petrels are partial planktivores, feeding on crustaceans, squid, myctophids, and other fish picked from the surface (Elliott et al., 1989). Storm-petrels breed in large colonies in the Haida Gwaii archipelago and along the northwest coast of Vancouver Island and nest in burrows/crevices, laying a single egg (Halpin et al., 2018).

2.2. Sampling sites and egg collections

Eggs of double-crested cormorants, pelagic cormorants, rhinoceros auklets, ancient murrelets, and Leach's storm-petrels were sampled from various colonies off the Pacific coast of British Columbia (BC), Canada (Fig. 1). A freshly laid egg (i.e. unincubated) was collected by hand from haphazardly selected nests early in the breeding season. Eggs were kept in cool locations and stored in a cooler or refrigerator. Internal egg contents were transferred to chemically rinsed (acetone/hexane) glass jars and frozen (−20 to −40 °C) until chemical analysis. In some years, eggs were analyzed as large composite pools (i.e., up to 15 eggs) or individual eggs (in 1973, 1985, 1987 to 1990, 1992, and 1994). In all other years, eggs were analyzed as five pools of 2–3 eggs each per species. Generally, egg samples were analyzed for contaminants within the year of collection. Handling and collection of seabird eggs

were conducted under research and collection permits authorized by Environment and Climate Change Canada (ECCC). Details about egg collection locations and dates are in the Supplementary Information.

2.3. Organochlorine insecticide and polychlorinated biphenyl (PCB) chemical analyses

From 1971 to 1983, egg homogenates were analyzed for OCs and PCBs at the Ontario Research Foundation, as described by Reynolds and Cooper (1975). In 1988, double-crested and pelagic cormorant egg homogenates were analyzed for OCs and PCBs at the Great Lakes Institute for Environmental Research (GLIER) laboratory at the University of Windsor. In all other years, egg homogenates were analyzed for POPs at the National Wildlife Research Centre (NWRC) in Ottawa, ON. The full list of OCs (Σ OCs) analyzed included:

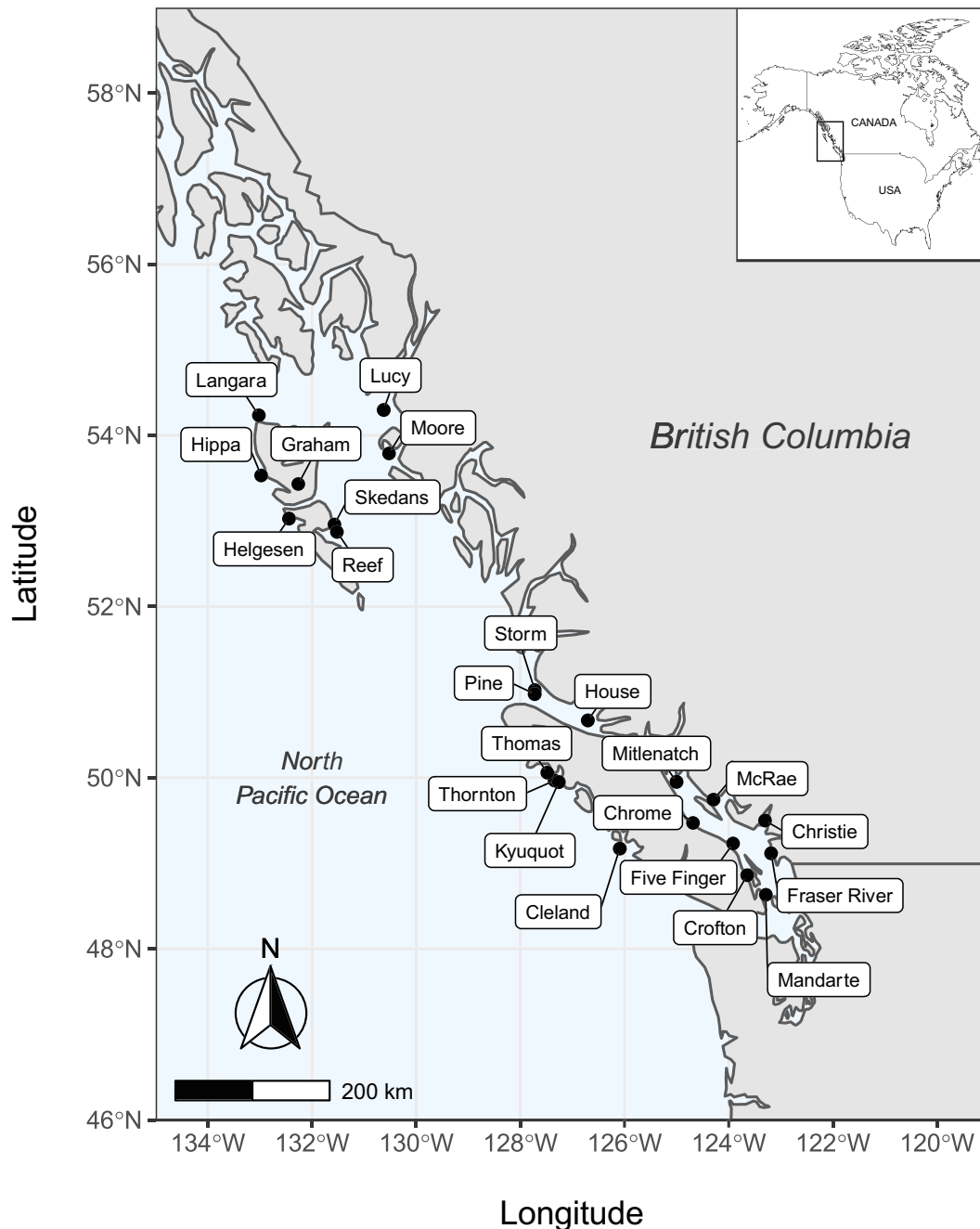


Fig. 1. Location of seabird colonies and egg collection sites ($n = 23$) sampled between 1968 and 2019 from the Pacific coast of British Columbia (BC), Canada. The spatial extent of sampling sites is indicated by the rectangle in the inset (top right corner). Map made programmatically in R (RStudio, Ver 4.1.2).

chlorobenzenes (Σ Cbz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene), hexachlorocyclohexanes (Σ HCH = α -, β - and γ -hexachlorocyclohexane), chlordan-related compounds (Σ CHL = oxychlordan, *trans*-chlordan, *cis*-chlordan, *trans*-nonachlor, *cis*-nonachlor), heptachlor epoxide (HE), dichlorodiphenyldichlorethane (1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane) (DDT and its metabolites; Σ DDT = *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD), octachlorostyrene (OCTS), mirex (Σ Mirex = photomirex and mirex), dieldrin, and PCB congeners. For a small number of egg samples collected between the 1970s and 1990s, PCBs were estimated using 1:1 mixtures of Aroclor 1254:1260 or Aroclor 1260 (Elliott et al., 1989). For these samples, an equivalent to later sums of Σ PCBs was estimated using significant linear regressions derived for each species (see Harris et al., 2005; Turle et al., 1991). In all other years, Σ PCBs were standardized to congeners according to their respective IUPAC numbers. Thus, to facilitate direct comparisons across species and sampling years, we focussed on the overall sum of PCBs (Σ PCBs) when analyzing temporal trends; however, in the recent year of sampling (2019), we also compared concentrations of several penta-, hexa-, and hepta-chlorinated PCB congeners due to their relatively high bioaccumulation potential (Borgå et al., 2005; Braune et al., 2019). Details about chemical analyses, instrumentation, quality control and assurance are provided in the Supplementary Information.

2.4. Stable isotope analyses

Stable isotope analyses of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) were generally carried out using the same pooled egg homogenates as used for chemical analyses and were performed at the G. G. Hatch Stable Isotope Laboratory in Ottawa, ON. Compound-specific amino acid analyses were conducted on a subset of seabird egg samples at the University of California Davis Stable Isotope Facility. Amino acid analyses were conducted using the same egg homogenates as used for bulk tissue $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ analyses and have been described previously (Elliott et al., 2021b). Trophic amino acids analyzed in seabird eggs included: alanine (Ala), aspartate (Asp), glutamate (Glu), glycine (Gly), isoleucine (Ile), leucine (Leu), proline (Pro), and valine (Val). Source amino acids included phenylalanine (Phe) and lysine (Lys). We used the term " $\delta^{15}\text{N}_{\text{glu-phe}}$ " to denote the difference in $\delta^{15}\text{N}$ between glutamate (i.e., trophic amino acid) and phenylalanine (i.e., source amino acid), and " $\delta^{15}\text{N}_{\text{trophic-source}}$ " as the difference in $\delta^{15}\text{N}$ averaged across multiple trophic (Ala, Asp, Glu, Gly, Ile, Leu, Pro, Val) and source (Lys, Phe) amino acids. We used the term 'bulk' to distinguish $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analyses in whole body samples from compound-specific analyses. Details about stable isotope analyses are in the Supplementary Information.

2.5. Statistical analyses

Statistical analyses were carried out in R (RStudio, Ver 9.2) and were performed on contaminants that were detected in at least 60 % of all egg samples. For contaminant concentrations that were below the Method Detection Limit (MDL), average concentrations, standard deviation (SD), and standard error (SE) were calculated using a Kaplan-Meier (KM) statistical model in NADA, as recommended for left-censored data (Helsel, 2012). We also used the KM method to summarize non-detects and compute means and confidence intervals for major contaminant groups (Helsel, 2009) and to calculate the percent contribution of individual OCs to the total POP burden. To be consistent with previous data and current literature, all chemical data is presented on a wet weight basis.

Legacy POP concentrations typically exhibit a right-skewed distribution with time (Elliott et al., 1988); therefore, to determine whether POP concentrations decreased over time, we fitted a set of generalized linear models for each species with a gamma distribution and log link function in R. Although significant relationships were found between % egg lipid content and some POP concentrations (e.g. DDT) for the two cormorants, we did not find significant relationships between % lipid and all of the other POPs for these species, nor did we find significant relationships between these variables for the other offshore species. Given these inconsistencies, we included % lipid as a covariate rather than use lipid normalized

concentrations to control for the variation in % lipid over time (Hebert and Keenleyside, 1995). Accordingly, each POP was analyzed separately as the dependent variable with year, % lipid, and breeding site as independent variables. For contaminants with non-detects, we fitted a set of models for each species by first using the lowest AIC before analyzing model coefficients with a censored multiple regression using the NADA2 (Julian and Helsel, 2021) package in R. For the latter, we included a quadratic effect (year^2) to account for possible first-order nonlinearity. Trend analyses were plotted with a Kendall's Tau (τ) test of change and an Akritas-Theil-Sen regression line for censored data. A principal component analysis (PCA) was used to examine patterns of PCB congeners for the most recent year of sampling (2019) and to determine whether species may have been exposed to PCBs from either long-range/atmospheric or local sources.

Egg $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values were compared among species and sampling years using a linear mixed-effects model with a Tukey's multiple comparisons test in R. Each stable isotope was analyzed separately as the dependent variable with year as a fixed effect and breeding site as a random effect. Relationships between bulk $\delta^{15}\text{N}$ and compound specific stable isotopes were examined using simple linear regression. Isotopic niche parameters were generated with SIBER (Jackson et al., 2011) in R. Isotopic niche overlap, defined as the area in units of per mil squared, was calculated using the standard ellipse area for each species corrected according to the sample size (SEA_C). Two outliers were removed (LSPE, Hippa Island 1995, $\delta^{15}\text{N} = 0.32\text{‰}$, $\delta^{13}\text{C} = -29\text{‰}$ and DCCO, Mandarte Island 1994, $\delta^{15}\text{N} = 11.2\text{‰}$, $\delta^{13}\text{C} = -25\text{‰}$) from the stable isotope analyses. The statistical significance of p values for all models was assessed at $\alpha = 0.05$.

3. Results

3.1. Dominant chemical congeners and profiles

With the exception of Σ PCBs, the primary OCs detected in the eggs of all five species and sampling years were Σ DDT (mostly *p,p'*-DDE), Σ HCH (mostly β -HCH), Σ CHLOR (mostly oxychlordan), and Σ Cbz (mostly HCB) (Fig. 2). Σ DDT was comprised almost entirely of *p,p'*-DDE, which averaged 90 % of Σ DDT in LSPE eggs, 95 % in PECO eggs, and 99 % in the eggs of double-

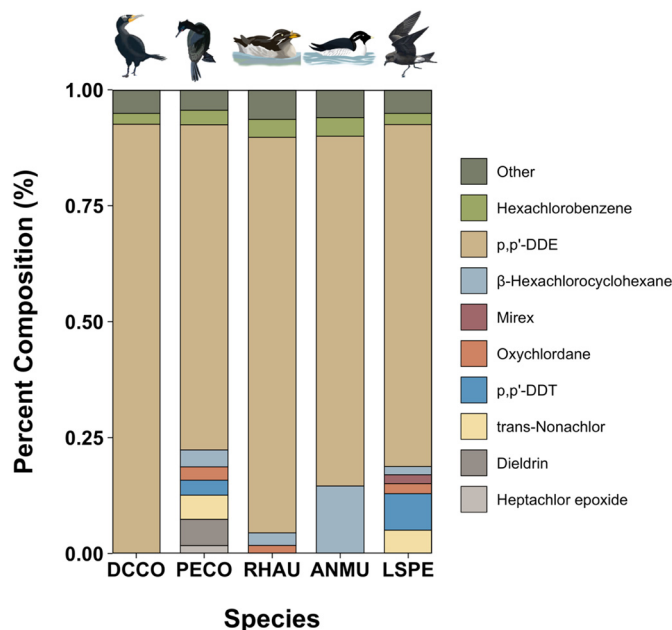


Fig. 2. Percent contribution (%) of major organochlorine insecticides averaged across all sampling years (1968–2019) in the eggs of nearshore, continental shelf, and offshore seabird monitoring species located at the breeding colonies in Fig. 1. Nearshore species: double-crested cormorant (DCCO), pelagic cormorant (PECO). Continental shelf species: rhinoceros auklet (RHAU), ancient murrelet (ANMU). Offshore species: Leach's storm-petrel (LSPE).

crested cormorants, murrelets, and auklets. Proportions of the *p,p'*-DDD metabolite and parent compound, *p,p'*-DDT, were generally at levels below detection limits, averaging <1 % of the total ΣOCs across all species and sampling years. DDE:DDT ratios, typically used as a proxy for the age and rate of transformation of ΣDDT, were highly variable and ranged over several orders of magnitude, averaging 9.4 in LSPE eggs, 21.7 in PECO eggs, 264 in RHAU eggs, 358 in ANMU eggs, and 374 in DCCO eggs across all years.

ΣHCH was predominantly made up of β-HCH, averaging 70 % in DCCO eggs, 79 % in PECO eggs, 83 % in LSPE eggs, 93 % in RHAU eggs, and 99 % in ANMU eggs. The α-HCH isomer was detected less frequently, averaging 1 % in ANMU eggs, 6 % in RHAU eggs, and 7 % in LSPE eggs, of ΣHCH. For the cormorants, α-HCH proportions were higher, comprising an average of 18 % in PECO eggs and 29 % in DCCO eggs, of ΣHCH over the entire study period.

The relative compositions of ΣCHLs differed among species. In eggs of auklets and double-crested cormorants, oxychlordane was the predominant isomer, comprising an average of 46 % and 81 %, respectively, of ΣCHL, while *cis*-nonachlor averaged 14 % and 13 %, respectively, of ΣCHL. Meanwhile, *trans*-nonachlor was the predominant isomer in the eggs of murrelets, pelagic cormorants, and storm-petrels, averaging 34 %, 52 %, and 62 %, respectively, of ΣCHL, while *cis*-nonachlor in these latter species averaged 36 %, 13 %, and 6 %, respectively, of ΣCHL. Proportions of *cis*- and *trans*-chlordane isomers were generally at levels below detection limits.

ΣCBz was predominately made up of HCB for all species, averaging 88 % in RHAU eggs, 94 % in PECO and LSPE eggs, 95 % in ANMU eggs, and 97 % in DCCO eggs. ΣMirex was generally comprised of mirex, which averaged 58 % in LSPE eggs, 60 % in RHAU eggs, 62 % in ANMU eggs, 73 % in PECO eggs, and 91 % in DCCO eggs. The photodegradation product, photomirex, was detected in greater proportions in the eggs of storm-petrels (42 %) and auklets (40 %), and 1–39 % in the eggs of all other species over the entire study period. Other OCs, such as pentachlorobenzene, octachlorostyrene, heptachlor epoxide, and dieldrin were either detected in small proportions or at levels below detection limits in the eggs of most species and sampling years.

3.2. Temporal trends

3.2.1. Nearshore environment (cormorants; double-crested cormorant and pelagic cormorant)

From 1970 to 2019, significant decreases occurred for *p,p'*-DDE ($t_{68} = -2.58$; $p < 0.05$) and HCB ($t_{67} = -5.99$; $p < 0.001$) concentrations in

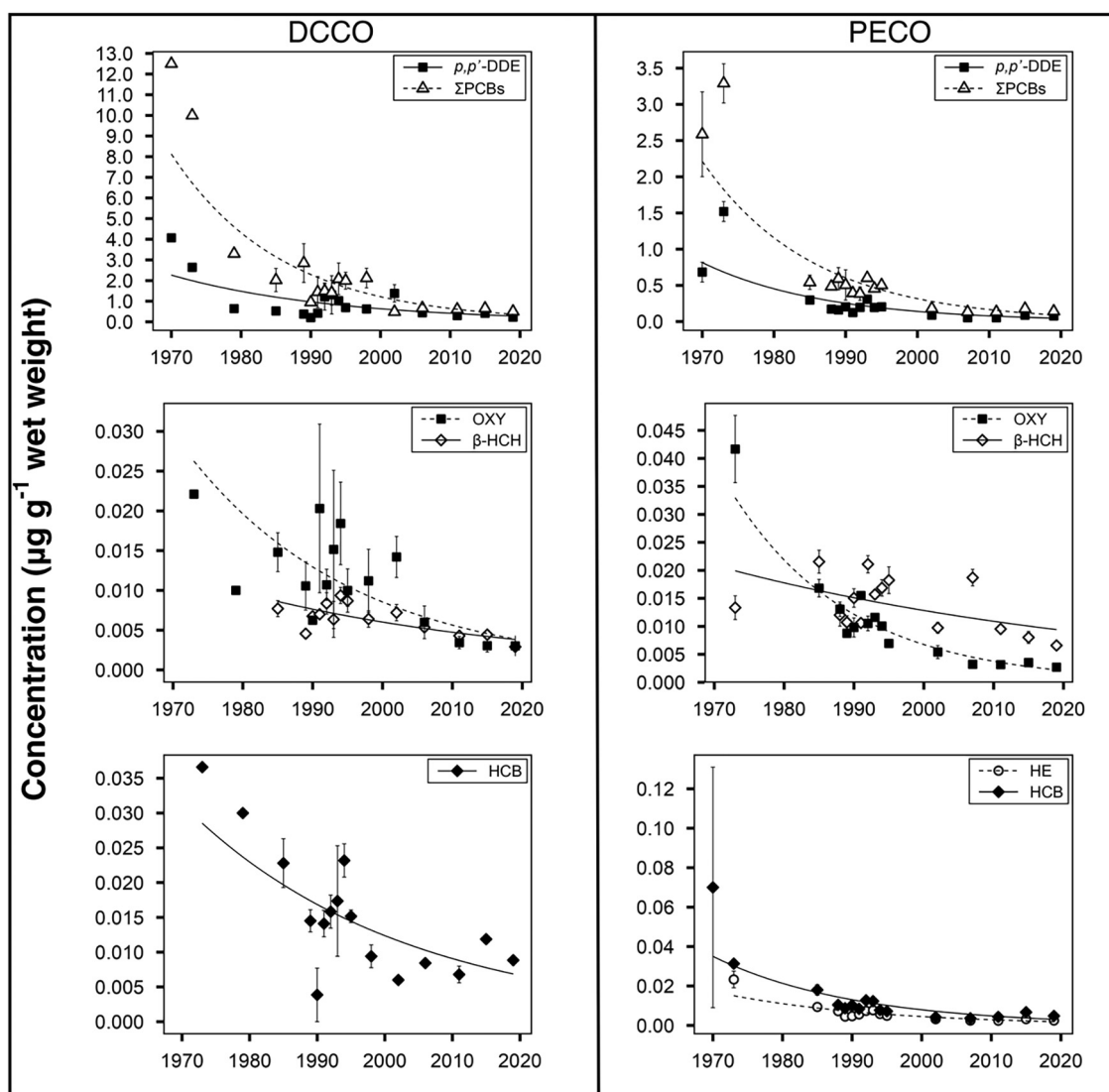


Fig. 3. Temporal trends of persistent organic pollutants (POPs) in the eggs of two nearshore indicator seabird species, sampled 1970 to 2019 from the Pacific coast of British Columbia, Canada. Left: double-crested cormorant (DCCO). Right: pelagic cormorant (PECO). Trends are plotted for mean contaminant concentrations that were fully detected (i.e., non-censored) in egg samples for that species. “*p,p'*-DDE” = para,para-dichlorodiphenyldichloroethylene (1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene); “ΣPCBs” = sum of polychlorinated biphenyls; “OXY” = oxychlordane; “β-HCH” = beta-hexachlorocyclohexane; “HE” = heptachlor epoxide; “HCB” = hexachlorobenzene. Error bars represent the standard error.

DCCO eggs (Fig. 3). By 1998, mean p,p' -DDE concentrations dropped by ~85 % before reaching a steady state from 2006 to 2019. Although decreases in Σ PCBs were slightly offset by elevated mean concentrations in 1989 ($2.85 \pm 0.94 \mu\text{g/g}$), mean Σ PCB concentrations still decreased exponentially by ~96 % from 1970 to 2019 ($t_{68} = -6.84$; $p < 0.001$) and remained at uniformly low concentrations ($<1 \mu\text{g/g}$) for the last 17 years of the study period. Mean concentrations of oxychlordanone peaked in the early period and then decreased ($t_{67} = -6.65$; $p < 0.001$), while those for β -HCH remained relatively constant during the 1990s before levelling off to $<0.01 \mu\text{g/g}$ in the late period ($t_{65} = -5.90$; $p < 0.001$). Concentrations of other contaminants in DCCO eggs also showed evidence of downward trends, largely influenced by non-detect values during recent sampling periods (STable 1; SFigure 1).

Significant decreases occurred for p,p' -DDE ($t_{81} = -7.69$; $p < 0.001$), HCB ($t_{81} = -7.41$; $p < 0.001$), HE ($t_{79} = -8.29$; $p < 0.001$), oxychlordanone ($t_{79} = -12.6$; $p < 0.001$), and *cis*-nonachlor ($t_{73} = -11.6$; $p < 0.001$) concentrations in PECO eggs (Fig. 3). Most of the declines for Σ PCBs in PECO eggs occurred through the late 1990s ($t_{81} = -12.0$; $p < 0.001$) and were followed by little change ($<0.1 \mu\text{g/g}$) during the 2000s (Fig. 3). Σ PCBs and p,p' -DDE in PECO eggs generally followed an exponential decline

pattern, with mean concentrations decreasing by ~95 % and 89 %, respectively, by the end of the study period. By contrast, mean concentrations of β -HCH peaked in 1985, 1992, and 2007 at roughly the same concentration (mean $0.02 \pm 0.002 \mu\text{g/g}$) before declining in the late period but without statistical significance ($t_{79} = -1.94$; $p < 0.1$). Concentrations of other contaminants in PECO eggs all declined over time and in the case of photomirex, showed no major trend (STable 2; SFigure 2).

3.2.2. Continental shelf environment (alacids; ancient murrelet, rhinoceros auklet)

From 1970 to 2019, significant decreases occurred for p,p' -DDE ($t_{133} = -3.01$; $p < 0.01$), HCB ($t_{133} = -3.59$; $p < 0.001$), HE ($t_{132} = -12.2$; $p < 0.001$), oxychlordanone ($t_{132} = -14.0$; $p < 0.001$), *trans*-nonachlor ($t_{126} = -11.0$; $p < 0.001$), and Σ PCB ($t_{133} = -12.7$; $p < 0.001$) concentrations in RHAU eggs (Fig. 4). Mean concentrations of p,p' -DDE and Σ PCBs followed an exponential decline pattern, decreasing by ~90 % and 88 %, respectively, by the end of the study period. Although a sharp peak in β -HCH was observed at Lucy Island in 2003 (mean $0.084 \pm 0.07 \mu\text{g/g}$), β -HCH concentrations declined shortly thereafter, though not statistically significant ($t_{133} = -1.05$; $p > 0.1$). Concentrations of other contaminants in

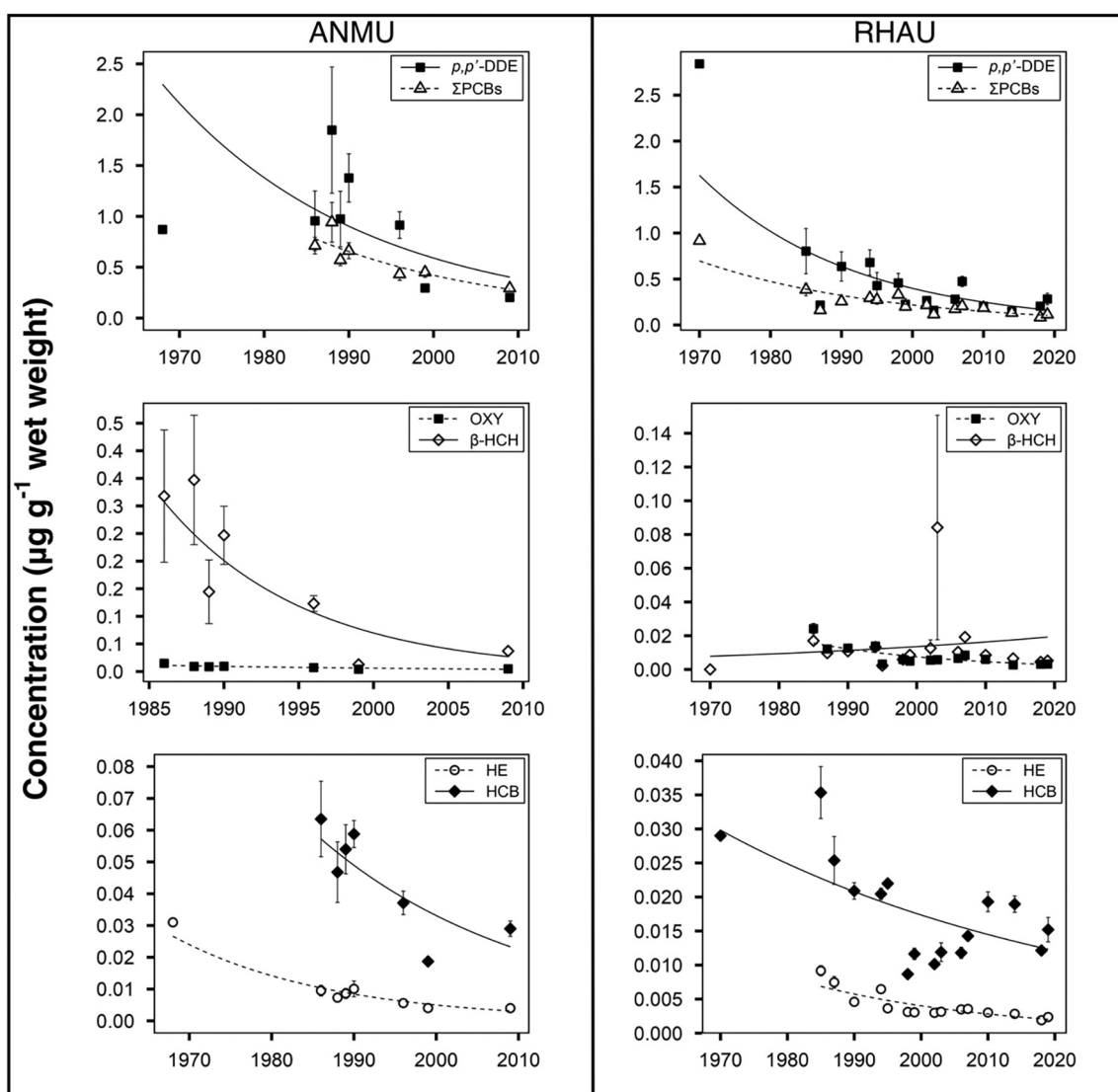


Fig. 4. Temporal trends of persistent organic pollutants (POPs) in the eggs of two continental shelf indicator seabird species, sampled 1968 to 2019 from the Pacific coast of British Columbia, Canada. Left: ancient murrelet (ANMU). Right: rhinoceros auklet (RHAU). Trends are plotted for mean contaminant concentrations that were fully detected (i.e., non-censored) in egg samples for that species. “ p,p' -DDE” = para,para-dichlorodiphenyldichloroethylene (1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene); “ Σ PCBs” = sum of polychlorinated biphenyls; “OXY” = oxychlordanone; “ β -HCH” = beta-hexachlorocyclohexane; “HE” = heptachlor epoxide; “HCB” = hexachlorobenzene. Error bars represent the standard error.

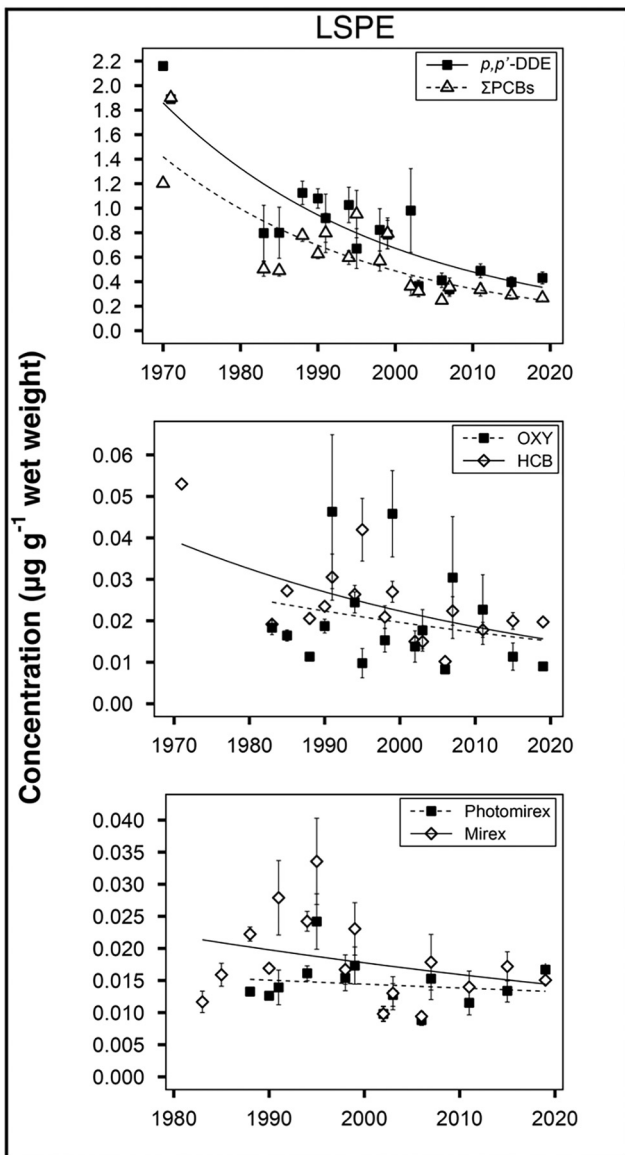


Fig. 5. Temporal trends of persistent organic pollutants (POPs) in the eggs of an offshore indicator seabird species, the Leach's storm-petrel (LSPE), sampled 1970 to 2019 from the Pacific coast of British Columbia, Canada. Trends are plotted for mean contaminant concentrations that were fully detected (i.e., non-censored) in egg samples. “*p,p'*-DDE” = para,para-dichlorodiphenyldichloroethylene (1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene); “ΣPCBs” = sum of polychlorinated biphenyls; “OXY” = oxychlordan; “HCB” = hexachlorobenzene. Error bars represent the standard error.

Table 1

Amino acid-specific stable isotope analyses. Values represent arithmetic means (range in parentheses) for multiple trophic (Ala, Asp, Glu, Gly, Ile, Leu, Pro, Val) and source (Phe, Lys) amino acids, as well as for Glu, Phe, bulk $\delta^{15}\text{N}$, and $\delta^{15}\text{N}_{\text{trophic-source}}$ in Pacific seabird eggs collected from the breeding colonies in Fig. 1.

Species	Trophic (%)	Source (%)	Glu (%)	Phe (%)	Bulk $\delta^{15}\text{N}$	$\delta^{15}\text{N}_{\text{trophic-source}}$ (%)
Double-crested cormorant (DCCO)	22.92 (20.54–27.55)	7.23 (5.39–9.04)	25.16 (19.77–30.46)	7.34 (4.99–9.34)	15.01 (11.22–17.26)	15.69 (14.07–18.52)
Pelagic cormorant (PECO)	21.91 (19.89–23.60)	6.88 (5.44–8.04)	23.57 (21.33–25.71)	7.03 (5.73–8.38)	13.95 (12.83–15.01)	15.02 (13.50–17.81)
Rhinoceros auklet (RHAU)	23.22 (21.48–24.90)	6.89 (3.74–8.28)	26.14 (21.75–30.60)	6.46 (4.45–8.71)	13.42 (11.92–14.64)	16.33 (14.47–18.07)
Ancient murrelet (ANMU)	19.51 (17.53–21.89)	5.92 (4.84–7.52)	22.18 (18.80–26.40)	5.59 (4.18–7.11)	12.27 (11.43–13.09)	13.60 (12.45–16.71)
Leach's storm-petrel (LSPE)	24.23 (20.92–25.79)	6.49 (4.71–8.27)	26.32 (20.70–29.70)	6.12 (3.88–7.81)	14.11 (12.01–15.57)	17.74 (15.00–19.46)

RHAU eggs all declined over time, while those for PCB and OCTS showed slight increasing trends in the late study period (STable 3, SFigure 3).

From 1968 to 2009, significant decreases occurred for HCB ($t_{42} = -7.43$; $p < 0.001$), β -HCH ($t_{42} = -7.05$; $p < 0.001$), oxychlordan ($t_{42} = -5.07$; $p < 0.001$), *trans*-nonachlor ($t_{42} = -6.74$; $p < 0.001$), *cis*-nonachlor ($t_{42} = -5.17$; $p < 0.001$), HE ($t_{43} = -5.48$; $p < 0.001$), and OCTS ($t_{36} = -14.4$; $p < 0.001$) concentrations in ANMU eggs (Fig. 4). Mean concentrations of *p,p'*-DDE peaked in 1988 ($1.85 \pm 0.6 \mu\text{g/g}$) before declining ($t_{43} = -8.58$; $p < 0.001$) with an overall reduction of 77 % by 2009. Mean concentrations of ΣPCBs declined at a steadier rate ($t_{42} = -7.66$; $p < 0.001$), decreasing by ~59 % by 2009 (Fig. 4). Concentrations of other contaminants in ANMU eggs either declined over time or showed no major trends (STable 4; SFigure 4).

3.2.3. Offshore environment (storm-petrels; Leach's storm-petrel)

From 1970 to 2019, mean concentrations of *p,p'*-DDE decreased ($t_{141} = -7.59$; $p < 0.001$) in tandem with ΣPCBs ($t_{141} = -8.61$; $p < 0.001$; Fig. 5), dropping by >80 % and 78 %, respectively, by the end of the study period. Similarly, mean concentrations of HCB ($t_{140} = -3.01$; $p < 0.01$), *p,p'*-DDT ($t_{141} = -6.01$; $p < 0.001$), *trans*-nonachlor ($t_{130} = -2.48$; $p < 0.05$), and oxychlordan ($t_{139} = -4.12$; $p < 0.001$) in LSPE eggs peaked in the 1990s and/or early 2000s before continuously declining in the late period. Opposing trends were observed for mirex ($t_{139} = -2.37$; $p < 0.05$) and photomirex ($t_{124} = -0.19$; $p > 0.1$). Specifically, there was a rapid annual increase in mean concentrations of mirex in the early period (1983 to 1995), while those for photomirex stabilized and showed slight increasing trends in the late period (2011 to 2019), consequently resulting in no net change for ΣMirex (Fig. 5). Concentrations of other contaminants in LSPE eggs generally declined over time (STable 5, SFigure 5).

3.2.4. Comparisons of contaminant concentrations within and across species

Legacy POP concentrations in seabird eggs declined over time at all breeding colonies (Table 2). For species with the longest time trends, mean concentrations of ΣPCBs were highest in the eggs of DCCO ($1.75 \pm 0.25 \mu\text{g/g ww}$) > PECO ($0.643 \pm 0.1 \mu\text{g/g}$) > LSPE ($0.484 \pm 0.03 \mu\text{g/g}$) > RHAU ($0.212 \pm 0.01 \mu\text{g/g}$) across all years. For *p,p'*-DDE, concentrations followed a slightly different pattern than those for ΣPCBs, averaging highest in the eggs of DCCO ($0.723 \pm 0.09 \mu\text{g/g}$) > LSPE ($0.670 \pm 0.04 \mu\text{g/g}$) > RHAU ($0.379 \pm 0.04 \mu\text{g/g}$) > PECO ($0.276 \pm 0.04 \mu\text{g/g}$) across all years. Comparatively, mean concentrations of *p,p'*-DDE dropped by 77 % in ANMU eggs, 80 % in LSPE eggs, 89 % in PECO eggs, 90 % in RHAU eggs, and 94 % in DCCO eggs, during the study period. Similarly, mean concentrations of ΣPCBs dropped by 59 % in ANMU eggs, 78 % in LSPE eggs, 88 % in RHAU eggs, 95 % in PECO eggs, and 96 % in DCCO eggs during the study period. In the recent year of sampling (2019), the top four PCB congeners detected in the eggs of all species, in descending order, were: PCB-153, -138, -180, and/or -118, with concentrations averaging highest in the eggs of DCCO, relative to those from other species (STable 6).

>77 % of the total variance was explained by the first axis of principal component analysis examining several dominant PCB congeners in the recent year of sampling (2019; Fig. 7). The PCAs suggest that rhinoceros

Table 2
 Concentrations of major organochlorines (µg/g wet weight) in Pacific seabird eggs collected from the breeding colonies located in Fig. 1. Concentrations shown are representative of the earliest (1968/70), intermediate (1990/4), and recent years (2009 or 2018/9) of reporting for that species. Concentrations shown are for individual eggs or as a pooled sample of eggs (arithmetic mean ± standard error). “HE” = Heptachlor epoxide. “OCTS” = Octachlorostyrene. “β-HCH” = β-Hexachlorocyclohexane. “HCB” = Hexachlorobenzene. “ΣPCBs” = Polychlorinated biphenyls^a.

Species	Year	Sample size	% moisture	% lipid	p,p'-DDE	HE	OCTS	Oxychlorthane	β-HCH	HCB	Dieldrin	Mirex	ΣPCBs ^b
Nearshore environment													
Double-crested cormorant (DCCO)	1970	1 pool, n = 3	81.4	6.90	4.07	–	–	–	–	–	0.04	–	12.5
	1994	5 pools, n = 3	83.7 ± 0.1	5.30 ± 0.08	1.02 ± 0.13	0.006 ± 0.002	ND	0.018 ± 0.005	0.009 ± 0.001	0.023 ± 0.002	0.014 ± 0.01	0.005	2.08 ± 0.76
	2019	5 pools, n = 3	84.0 ± 0.1	3.39 ± 0.27	0.23 ± 0.05	0.002	ND	0.003 ± 0.001	0.003	0.009 ± 0.001	0.0017	ND	0.50 ± 0.17
Pelagic cormorant (PECO)	1970	1 pool, n = 10	83.7 ± 0.05	4.85 ± 0.5	0.68 ± 0.14	–	–	–	–	0.07 ± 0.06	0.072 ± 0.01	–	2.59 ± 0.58
	1994	6 pools, n = 3–10	83.6 ± 0.16	5.25 ± 0.14	0.19 ± 0.02	0.006	0.001	0.01 ± 0.001	0.017 ± 0.002	0.008	0.012 ± 0.001	0.002	0.46 ± 0.04
	2019	5 pools, n = 2–3	83.6 ± 0.13	3.20 ± 0.51	0.076 ± 0.01	0.003	ND	0.003	0.007 ± 0.001	0.005	0.007	0.001	0.14 ± 0.01
Continental shelf environment													
Rhinceros auklet (RHAU)	1970	1 pool, n = 10	71.7	15.0	2.84	–	–	–	–	0.029	–	–	0.917
	1994	10 pools, n = 2–3	69.4 ± 1.04	14.0 ± 0.21	0.68 ± 0.14	0.007 ± 0.003	ND	0.014 ± 0.001	0.013 ± 0.001	0.020 ± 0.001	0.007	0.003	0.294 ± 0.03
	2018/9	15 pools, n = 3	69.2 ± 0.35	12.5 ± 0.51	0.22 ± 0.03	0.002	0.001	0.003	0.005	0.013 ± 0.001	0.004	0.001	0.089 ± 0.01
Ancient murrelet (ANMU)	1968	1 pool, n = 3	65.2	15.3	0.87	0.031	–	–	–	–	0.008	–	–
	1990	6 pools, n = 2	64.6 ± 0.2	16.8 ± 0.4	1.38 ± 0.2	0.01 ± 0.002	0.03 ± 0.001	0.0094 ± 0.001	0.247 ± 0.05	0.059 ± 0.004	0.017 ± 0.003	0.003	0.662 ± 0.08
	2009	14 pools, n = 2–3	60.9 ± 0.7	19.0 ± 1.0	0.20 ± 0.02	0.004	0.002	0.005	0.037 ± 0.003	0.029 ± 0.002	0.0034	0.0029	0.294 ± 0.02
Offshore environment													
Leach's storm-petrel (LSPE)	1970	n = 1	73.4	12.7	2.16	–	–	–	–	–	0.045	–	1.20
	1994	10 pools, n = 3	72.9 ± 0.56	11.7 ± 0.55	1.03 ± 0.14	0.004	0.005	0.024 ± 0.003	0.017 ± 0.002	0.026 ± 0.002	0.006 ± 0.001	0.024	0.60 ± 0.06
	2019	15 pools, n = 3	69.4 ± 0.43	11.5 ± 0.45	0.43 ± 0.05	0.003	0.001	0.009 ± 0.001	0.011 ± 0.001	0.020 ± 0.001	0.005	0.015	0.27 ± 0.02

^a “ND” = Not detected based on the Method Detection Limit (MDL). “–” = Not analyzed.

^b ΣPCBs is the total sum of PCB congeners. In a small number of early samples (1970–1990), ΣPCBs were measured as 1:1 mixtures of either Aroclor 1260 or Aroclor 1254:1260. For these samples, an equivalent to later sums of ΣPCBs was estimated using the following species-specific significant regression relationships: ANMU: Log(SumPCBs) = 0.8652(Log(Aroclor1260)) + 0.3398 (n = 23; p < 0.001; R² = 0.90). DCCO: Log(SumPCBs) = 0.90971(Log(Aroclor1260)) + 0.05639 (n = 48; p < 0.001; R² = 0.96); or Log(SumPCBs) = 0.90(Log(Aroclor1254:1260)) - 0.2551 (n_{total} = 36; p < 0.001; R² = 0.99). PECO: Log(SumPCBs) = 0.81872(Log(Aroclor1260)) + 0.01463 (n = 37; p < 0.001; R² = 0.97); or Log(SumPCBs) = 0.9508(Log(Aroclor1254:1260)) - 0.3413 (n_{total} = 34; p < 0.001; R² = 0.99). LSPE: Log(SumPCBs) = 1.01821(Log(Aroclor1260)) + 0.04184 (n = 90; p < 0.001; R² = 0.93); or Log(SumPCBs) = 0.9590(Log(Aroclor1254:1260)) - 0.2173 (n_{total} = 86; p < 0.001; R² = 0.97). RHAU: Log(SumPCBs) = 0.5685(Log(Aroclor1260)) - 0.2102 (n = 75; p < 0.001; R² = 0.60); or Log(SumPCBs) = 0.9530(Log(Aroclor1254:1260)) - 0.2428 (n_{total} = 74; p < 0.001; R² = 0.90).

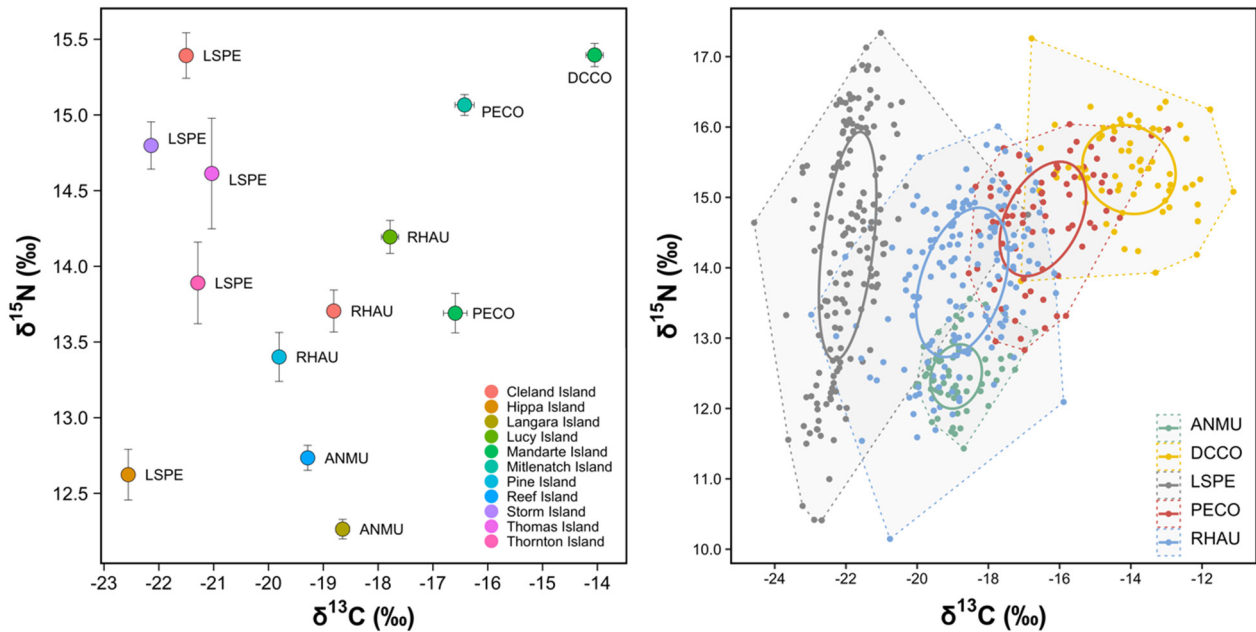


Fig. 6. Left plot: Mean bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ isotope biplot for Pacific seabird eggs collected from 1970 to 2019 at the breeding colonies located in Fig. 1. DCCO, double-crested cormorant; PECO, pelagic cormorant; RHAU, rhinoceros auklet; ANMU, ancient murrelet; LSPE, Leach’s storm-petrel. Error bars represent the standard error. Right plot: Population niche width and dietary overlap of bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ isotope signatures of Pacific seabird eggs collected from the breeding colonies located in Fig. 1 from 1970 to 2019. Circles represent maximum likelihood standard ellipses (sample size corrected area). Dashed lines and shaded regions represent convex hull total areas.

auklet and pelagic cormorant eggs did not overlap with respect to certain PCB congeners, nor did they appear to be influenced by any one congener or groups of PCB congeners. By contrast, one double-crested cormorant egg sample had higher levels of more chlorinated congeners (PCB – 158, – 195), possibly indicating a local source signal. Several storm-petrel egg samples had higher levels of lower chlorinated congeners (PCB – 101, – 110, – 151), possibly indicating a long-range atmospheric source further offshore.

3.3. Stable isotope analyses

3.3.1. Spatial variation in bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values in seabird eggs

$\delta^{13}\text{C}$ varied among species ($F_{4,19} = 126; p < 0.001$) with average $\delta^{13}\text{C}$ values ranging from -21.95‰ ($\pm 0.06\text{‰}$) in LSPE, -18.91‰ ($\pm 0.09\text{‰}$) in ANMU, -18.73‰ ($\pm 0.1\text{‰}$) in RHAU, -16.47‰ ($\pm 0.1\text{‰}$) in PECO, and -14.17‰ ($\pm 0.2\text{‰}$) in DCCO eggs across all locations and years (Fig. 6). Egg $\delta^{13}\text{C}$ values were lowest in storm-petrels breeding on Hippa Island (mean $\delta^{13}\text{C} = -22.56 \pm 0.1\text{‰}$) and Storm Island ($-22.14 \pm 0.1\text{‰}$), indicating a preference for offshore pelagic feeding. The intermediate egg $\delta^{13}\text{C}$ values for auklets and murrelets are consistent with these alcids foraging mostly on the continental shelf and/or near slope waters. Eggs of both cormorant species showed the greatest $\delta^{13}\text{C}$ enrichment relative to the other species ($p < 0.001$), a pattern consistent with their nearshore and benthic feeding habits. Interspecific differences for egg $\delta^{13}\text{C}$ values were also found between the two cormorant species ($p < 0.001$).

Similarly, bulk $\delta^{15}\text{N}$ varied among species ($F_{4,19} = 40.4; p < 0.001$) with average bulk $\delta^{15}\text{N}$ values ranging from 12.45‰ ($\pm 0.06\text{‰}$) in ANMU, 13.79‰ ($\pm 0.08\text{‰}$) in RHAU, 14.31‰ ($\pm 0.13\text{‰}$) in LSPE, 14.69‰ ($\pm 0.09\text{‰}$) in PECO, and 15.33‰ ($\pm 0.1\text{‰}$) in DCCO eggs across all locations and years. Mean bulk $\delta^{15}\text{N}$ values were lowest in ANMU eggs collected from Langara ($12.26 \pm 0.06\text{‰}$) and Reef Islands ($12.73 \pm 0.08\text{‰}$), and in LSPE eggs from Hippa Island ($12.62 \pm 0.17\text{‰}$). Mean bulk $\delta^{15}\text{N}$ values were highest in DCCO eggs from Mandarte Island ($15.33 \pm 0.1\text{‰}$) and LSPE eggs from Cleland Island ($15.39 \pm 0.15\text{‰}$). Pelagic cormorants breeding on Mitlenatch Island had more enriched

values of bulk $\delta^{15}\text{N}$ ($15.07 \pm 0.07\text{‰}$) than those breeding on Mandarte Island ($13.69 \pm 0.13\text{‰}$; Fig. 6).

For the subsamples of individuals with amino acid-specific data, bulk $\delta^{15}\text{N}$ correlated poorly with trophic position from $\delta^{15}\text{N}_{\text{glu-phe}}$ ($F_{1,92} = 0.22; p = 0.64; R^2 = 0$) while bulk $\delta^{15}\text{N}$ increased with $\delta^{15}\text{N}_{\text{trophic-source}}$ ($F_{1,92} = 7.7; p < 0.01; R^2 = 0.10$), providing evidence that the trophic relationships in our species were better predicted using several baseline and trophic amino acids, rather than just phenylalanine and glutamate. Respectfully, egg $\delta^{15}\text{N}$ data adjusted for isotopic baselines (i.e., $\delta^{15}\text{N}_{\text{trophic-source}}$) revealed minor differences in trophic position and baseline food web

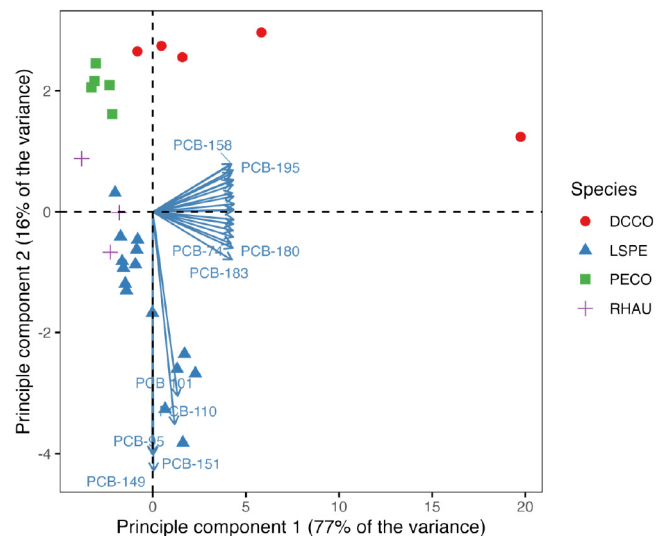


Fig. 7. Principal component analysis of several dominant PCB congeners for double crested cormorant eggs (DCCO; red circles, $n = 5$ pools of 3 eggs), pelagic cormorant eggs (PECO; green squares, $n = 5$ pools of 3 eggs), rhinoceros auklet eggs (RHAU; purple plus signs, $n = 3$ pools of 3 eggs), and Leach’s storm-petrel eggs (LSPE; blue triangles, $n = 15$ pools 3 eggs) collected in 2019 along the Pacific coast of British Columbia, Canada (Breeding colonies located in Fig. 1).

Table 3
Temporal trends of major legacy persistent organic pollutants (POPs) based on other published studies.

Area	Time period	Species	Contaminants	Sampling matrix	Trend	Reference
Eastern Canada	1974–2013	Herring gull	<i>p,p'</i> -DDE, Dieldrin, Mirex, Σ CHL, HCB, HE, Σ PCBs	Egg	↓	de Solla et al., 2016
Eastern Canada	1987–2007	Beluga whale	Σ DDT, Σ CHL, Σ HCH, TCPs, HCB, Mirex, Σ PCBs	Blubber	↓	Lebeuf et al., 2014
Canadian Arctic	1975–2003	Thick-billed murre Northern fulmar Black-legged kittiwake	Σ DDT, Σ CBz, Σ HCH, Σ CHL, OCS, Σ Mirex, Dieldrin, Σ PCBs	Egg	↓, ↔	Braune, 2007
Canadian Arctic	1975–2015	Thick-billed murre Northern fulmar Black-legged kittiwake Black guillemot Glaucous gull	Σ DDT, Σ CBz, Σ HCH, Σ CHL, OCS, Σ Mirex, Dieldrin, Σ PCBs	Egg	↓, ↔	Braune et al., 2019
Canadian Arctic	1972–2016	Ringed seal	Σ DDT, HCB, Σ HCH, Σ PCBs	Blubber	↓	Houde et al., 2019
Antarctic	1998–2016	Wilson's storm-petrel	Σ DDT and Σ PCBs	Egg	↓	Kuepper et al., 2022
United Kingdom	1977–1998	Northern gannet	Σ PCBs	Egg	↓	Alcock et al., 2002
Eastern Canada	1968–1984	Northern gannet	Σ DDT, Σ CHL, Mirex, Dieldrin, Σ HCH, HCB, Σ PCBs	Egg	↓	Elliott et al., 1988
Northern Norway	1983–2003	Herring gull Black-legged kittiwake Common murre Atlantic puffin	Σ DDT, HCB, Σ HCH, Σ CHL, Mirex, Σ PCBs	Egg	↓	Helgason et al., 2008
Central Norway	1986–2004	Tawny owl	Σ DDT, HCB, Σ HCH, Σ PCBs	Egg	↓	Bustnes et al., 2007
Baltic Sea	1969–1989	Common murre	Σ DDT, Σ PCBs	Egg	↓	Bignert et al., 1995
Baltic Sea, Sweden	1969–2012	Herring European perch Baltic cod Eelpout Blue mussel Common murre	Σ DDT, HCB, Σ HCH, Σ PCBs	Whole tissue, Egg	↓	Nyberg et al., 2015
Mediterranean	2000–2013	Mediterranean mussel	Σ DDT, α -HCH, γ -HCH, HCB, Dieldrin, Σ PCBs	Whole tissue	↓, ↔	Campillo et al., 2017
Western Mediterranean	1996–2005	Mediterranean mussel Saltwater clam Date mussel Sea urchin	Σ DDT, γ -HCH, HCB, Σ PCBs	Whole tissue	↓, ↔	Deudero et al., 2007
Korea	2015–2019	Black-tailed gull	Σ DDT, HCB, Σ HCH, Σ CHL, Σ PCBs	Egg	↓	Jang et al., 2022
Western Canada	1971–1986	Leach's storm-petrel Fork-tailed storm-petrel Rhinoceros auklet Double-crested cormorant Pelagic cormorant Glaucous-winged gull Ancient murrelet	<i>p,p'</i> -DDE, Dieldrin, HE, oxychlorthane, Mirex, HCB, α -HCH, β -HCH, Σ PCBs	Egg	↓	Elliott et al., 1989
Western Canada	1970–2002	Double-crested cormorant Pelagic cormorant	Σ DDT, Σ CBz, OCS, Σ CHL, HE, Σ Mirex, Σ HCH, Dieldrin, Σ PCBs	Egg	↓	Harris et al., 2005
Western Canada and USA	1991–1997	Osprey	Σ DDT, Dieldrin, HE, Σ CHL, Σ Mirex, Σ HCH, Σ CBz, Σ PCBs	Egg	↔	Elliott et al., 2000

signatures among the five seabird species (Table 1). Isotopic niche width assessed using SEA_C indicated some overlap between the two alcids (auklets and murrelets; 1.76 %) and between pelagic cormorants and auklets (1.45 %; Fig. 6).

3.3.2. Temporal variation in bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values and compound-specific amino acids

Apart from ANMU eggs, which showed no significant temporal variation in bulk $\delta^{15}\text{N}$ values ($t_{57} = -0.75$; $p = 0.46$), bulk $\delta^{15}\text{N}$ values statistically increased over time in DCCO ($t_{65} = 2.56$; $p < 0.05$), PECO ($t_{72} = 5.05$; $p < 0.001$), LSPE ($t_{163} = 3.80$; $p < 0.001$), and RHAU ($t_{166} = 6.45$; $p < 0.001$) eggs. By contrast, $\delta^{13}\text{C}$ values statistically decreased over time in ANMU ($t_{52} = -4.20$; $p < 0.001$), LSPE ($t_{161} = -2.48$; $p < 0.05$), and RHAU ($t_{165} = -2.07$; $p < 0.05$) eggs, but increased in PECO ($t_{24} = 3.11$; $p < 0.01$) eggs. Source amino acids (Phe, Lys) for all species but ANMU (Phe: $t_8 = -2.83$; $p < 0.05$; Lys: $t_8 = -3.67$; $p < 0.01$) showed no significant temporal change in $\delta^{15}\text{N}$ values ($p > 0.1$). Likewise, $\delta^{15}\text{N}$ values in most trophic amino acids (i.e., Ala, Asp, Glu, Ile, Leu, Val) remained relatively constant over time in the eggs of all species, ($p > 0.1$), suggesting minor changes in trophic position. $\delta^{15}\text{N}$ values in non-essential amino acids (Gly) statistically decreased over time in DCCO ($t_{23} = -2.21$; $p <$

0.05) and RHAU ($t_{19} = -2.37$; $p < 0.05$) eggs, while $\delta^{15}\text{N}$ values in proline decreased over time in ANMU ($t_8 = -3.13$; $p < 0.05$) eggs.

4. Discussion

Most legacy POPs have either declined significantly (e.g. *p,p'*-DDE, β -HCH, HCB, HE, oxychlorthane, Σ PCBs) or showed no directional change (e.g. Σ Mirex) over time in the eggs of our monitoring species. Temporal trends for major contaminants (e.g. *p,p'*-DDE and Σ PCBs) generally followed an exponential decline pattern, while those for other contaminants (e.g. oxychlorthane and β -HCH) followed a combination of linear and/or second-order declines. This may reflect large environmental variability of legacy POP concentrations in the northeast Pacific Ocean; however, continued monitoring and larger sample sizes would be needed to accurately determine the statistical nature of such declines. While there was considerable interannual variation in bulk $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values in the eggs of our monitoring species, compound-specific amino acid analyses (after accounting for baseline $\delta^{15}\text{N}$) suggested no major changes in trophic position or baseline food web signature. Thus, temporal trends observed in our Pacific seabirds appear to be attributable to restrictions on usage and

not shifts in trophic position, as reported previously for other contaminants in the same species and region (Miller et al., 2014).

Temporal trends in our study were comparable to those found in other species of seabirds (Table 3). Decreasing concentrations of Σ PCBs, Σ DDT, Σ CBz, and Σ CHL were observed in the eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*), black-legged kittiwakes (*Rissa tridactyla*), black guillemots (*Cepphus grylle*), and glaucous gulls (*Larus hyperboreus*) sampled from the Canadian Arctic from the 1970s through the 1990s (Braune et al., 2001; Braune, 2007) and early 2000s (Braune et al., 2019). Temporal trends of POPs have also been monitored pre- and post- production in the Canadian Great Lakes for over 40 years using herring gull (*Larus argentatus*) eggs (Pekarik and Weseloh, 1998; Hebert et al., 2000b; de Solla et al., 2016). Those studies revealed that concentrations of most POPs followed an exponential decline from 1970 to 2013 at all breeding colonies, with the largest overall percent decline (96.4 %) reported at Lake Michigan. However, some gulls may have supplemented their diets with a combination of aquatic and/or terrestrial food items with low PCB concentrations, subsequently resulting in subtle annual fluctuations in egg PCB levels (Hebert et al., 1997, 2000a). Temporal trends in our species were also comparable to seabird egg data collected from the Baltic Sea (Bignert et al., 1995; Nyberg et al., 2015), Norway (Helgason et al., 2008), United Kingdom (Alcock et al., 2002), Asia (Jang et al., 2022), and the Antarctic (Kuepper et al., 2022), further illustrating the decline of these contaminants in seabird food webs globally.

Temporal trends in our Pacific seabirds were also similar to those in other top predators and pelagic food webs (Table 3). Between 1989 and 2008, mean concentrations of Σ CHL and Σ DDT declined in multiple sub-populations of polar bears (*Ursus maritimus*), but not for dieldrin or Σ PCBs, which was likely in part due to a greater reliance on contaminated ringed seals (*Phoca hispida*) from the Western Canadian Arctic, continued emissions, and/or reservoir redistribution (Brown et al., 2018). Long-term monitoring of blubber samples of ringed seals from the Beaufort Sea, Arctic Archipelago, Hudson Bay, and coastal Labrador showed significant declines for Σ DDT, Σ CHL, α -HCH, dieldrin, and most Σ PCBs between 1972 and 2016 with the strongest declines (9.1 % per year) occurring in the southeastern Arctic, possibly due to shorter chemical residence times (Houde et al., 2019). Similarly, mean concentrations of Σ PCBs, Σ DDTs, and Σ HCHs measured in blubber samples of beluga whales (*Delphinapterus leucas*) from the St. Lawrence Estuary between 1987 and 2007 showed weak but statistically significant declines of ≤ 11 % per year (Lebeuf et al., 2014). Concentrations of β -HCH and photomirex appear to have increased in some species (Braune, 2007; Braune et al., 2019; Houde et al., 2019), including those from our study, while showing little discernable trends in others (Bustnes et al., 2007; Rig  t et al., 2008). Variation in the proportions of these (and potentially other POPs) is likely influenced by dietary exposure, biotransformation rates, detection limits, environmental partitioning behaviour, and potentially greater usage in Asia (Elliott et al., 1989), consequently leading to atmospheric transport and deposition to the Pacific Ocean (Brown et al., 2018).

From 1970 to 2019, DCCO eggs collected from Mandarte Island had the highest mean concentrations of Σ OCs and Σ PCBs, relative to eggs from the other four species. Unlike other monitoring colonies, which are all relatively distant from human population centres, Mandarte Island is located 8–10 km from urbanized areas and landfills (Davis et al., 2015) and is listed as a hotspot for contaminants in our study area (Elliott et al., 1989). Cormorants within the Strait of Georgia and nearby Puget Sound likely fed on more contaminated prey (Harris et al., 2005) or resident populations of fish with high PCB levels (West et al., 2008). That may have decreased metabolism and excretion efficiencies due to a thermodynamic gradient established in the gastrointestinal tract (Gobas et al., 1993, 1999), consequently increasing chemical fugacities and concentrations in eggs. These findings are corroborated by other studies showing high POP concentrations in wildlife foraging in contaminated environments near the south coast of Vancouver Island (Ross et al., 2004; Cesh et al., 2008; Huang et al., 2018) and more broadly along the Pacific coast of North America (Pereira et al., 1996; Elliott et al., 1996, 2007, 2021a; Mora et al., 2016). Conversely,

concentrations of most POPs were lower in eggs of murrelets and auklets, relative to eggs from the other species. These alcids feed at low trophic levels, often consuming small fish and other juvenile prey (Elliott et al., 1989). Furthermore, most of these alcids nest in remote colonies away from densely populated areas and industrial sources with lower land-based POP inputs. Oceanic currents and large-scale mixing events may have further reduced bioavailability (Brown et al., 2018; Elliott et al., 2021a) or enhanced the dilution of contaminants relative to more enclosed inland waters in the Strait of Georgia (West et al., 2008).

Temporal trends may also be influenced by local environmental conditions and toxicokinetic parameters. For instance, proportions of sand lance decreased in the diets of rhinoceros auklets during warm years at the southern edge of their range and increased during warm years at the northern edge of their range (Hedd et al., 2006; Cunningham et al., 2018); foraging behaviour and prey selection also varied among and within years due to individual nutritional demands and breeding stage (Hipfner et al., 2013). Accordingly, Thayer et al. (2008) demonstrated that forage fish communities can vary across multiple continental shelf regions with different sea surface temperatures, with juvenile rockfish frequently observed in the California Current, sand lance in the Eastern Coastal Transition Zone, and capelin in the Gulf of Alaska. We can expect that the POP concentrations in our study would be impacted by diet changes such as those. In addition, biomagnification of POPs resulting from trophic transfer via prey may be influenced by differences in body temperature and lipid composition between different organisms. Recently, Fremlin et al. (2021) argued that high body temperatures maintained by birds and endotherms could contribute to a temperature induced decrease in the lipid solubility of hydrophobic substances, compared to fish and other poikilothermic organisms with lower body temperatures. These associations suggest that specific warming patterns could alter trophic structure and dynamics in marine systems; however, whether this directly influences the biomagnification potential of legacy POPs in all seabirds is still unclear (Kalia et al., 2021).

Spatial and temporal variation in POPs can also be a function of migratory status and reproductive investment. Banding recoveries of double-crested cormorants from Mandarte Island confirm that most spend time near Padilla Bay, Skagit River, and Puget Sound (Moul, 1996), but radio telemetry data also indicate concentrated movements south towards California, Oregon, and the Columbia River (Harris et al., 2005). DCCO eggs laid on Mandarte Island could reflect contaminant exposure acquired on both breeding and wintering grounds. Geolocator loggers deployed on rhinoceros auklets breeding at the same colonies sampled in the present study indicated that concentrations of *p,p'*-DDE and Σ PCBs were up to two times higher in eggs laid by females wintering at lower latitudes (Elliott et al., 2021a). Similarly, geolocators on ancient murrelets breeding off Haida Gwaii in 2014 revealed that most birds migrated across the north Pacific Ocean to the Sea of Okhotsk and south towards China, Korea, and Japan before returning to their breeding colonies in BC (Gaston et al., 2017; Miller et al., 2020). That is consistent with the temporal decline in bulk $\delta^{13}\text{C}$ values in our ANMU eggs and suggests more shelf habitat feeding offshore north and west of Haida Gwaii. Moreover, murrelets from Hippa Island may have started migrating east ahead of those from other colonies (Gaston et al., 2017) and been exposed to recent or ongoing sources of POPs in Asia (van den Berg et al., 2017), which may have aligned with the relatively high Σ OCs in our Hippa Island eggs. As capital breeders, those murrelets may have additionally accrued and stored endogenous reserves for reproduction (Miller et al., 2014), resulting in carryover from wintering exposure to deposition in eggs (Yates et al., 2010). In some species, egg contaminant concentrations are also thought to be influenced by clutch size and egg-laying order (Braune et al., 2018), maternal reserves (Drouillard and Norstrom, 2001), and other physio-chemical factors (Verreault et al., 2006). Yet, Custer et al. (1997) found no significant differences in mean concentrations of *p,p'*-DDE, Σ PCBs, and other OCs between fresh DCCO eggs and sibling embryos from Green Bay, Wisconsin, suggesting that intra-clutch variation in POP concentrations played a negligible role in the contamination profile of our cormorant eggs. Nonetheless, our data is consistent with a latitudinal gradient and may explain why some birds

that nest or overwinter at lower latitudes are exposed to local sources of POPs (e.g. Σ PCBs), in contrast to other birds, notably the storm-petrels, that forage further offshore and are exposed to potentially more long-range atmospheric sources of PCBs.

In a global context, use of DDT and other OCs was relatively lower in British Columbia than in other North American regions (Elliott et al., 1989), albeit up to three times higher than most Canadian provinces based on recommended application rates (Harris et al., 2000). There was no broad scale spraying of forests due to the nature of pests in BC, in contrast to Oregon, Washington, and Idaho where emergency DDT spray programs were initiated to control outbreaks of the Douglas-fir tussock moth (*Orgyia pseudotsugata*) (Henny, 1977). However, there was intensive use of DDT to mitigate codling moth (*Cydia pomonella* L.) outbreaks and other pest infestations in fruit orchards in the BC southern interior, with *p,p'*-DDE persisting and biomagnifying at relatively high concentrations (mean 37 $\mu\text{g/g}$ ww, range 3.28–107 $\mu\text{g/g}$) in local avian food chains to the present day (Kesic et al., 2021). Some BC areas may have received >90,000 kg of technical DDT over the total period of use (Elliott et al., 1989), undoubtedly resulting in short to medium-range transport and deposition via drift, thereby contaminating nearby aquatic systems and alpine environments (Grenier et al., 2020). The low *p,p'*-DDT concentrations coupled with the relatively high DDE:DDT ratios in our seabird eggs confirms these findings and suggests either older applications, shorter half-lives, rapid biotransformation of the parent compound, and/or greater bio-availability and preferential uptake of *p,p'*-DDE in the marine environment due to its higher solubility and lower K_{OC} values compared with *p,p'*-DDT.

Our approach improves on previous studies that have simply used bulk $\delta^{15}\text{N}$ to assess trophic position and biomagnification potential. Specifically, egg $\delta^{15}\text{N}$ data adjusted for isotopic baselines ($\delta^{15}\text{N}_{\text{trophic-source}}$) indicated that over time, murrelets in our study occupied among the lowest trophic positions, consistent with this species' zooplanktivorous diet. Cormorants fed at intermediate trophic levels, reflecting mixed diets of fish and invertebrates and these groupings are similar to those reported by Elliott et al. (1989) and Hobson et al. (1994) for the same species and region. Storm-petrels and auklets occupied among the highest trophic positions relative to the other species, suggesting an exclusive diet of fish or higher trophic level zooplankton among laying females. For storm-petrels breeding on Cleland Island, the high trophic position would be synonymous with more myctophid fish in their diets (Vermeer and Devito, 1988; Hebert et al., 2022) but also high mercury concentrations since myctophids perform diel vertical migrations and can reach the oxygen minimum zone where methylmercury is produced by sulfate-reducing bacteria (Elliott and Elliott, 2016). Conversely, lower POP concentrations in our auklet eggs, despite their apparent high trophic position, may have been due to egg lipids obtained during the overwintering period compared with egg protein obtained during the breeding season (Elliott et al., 2014). Braune et al. (2002) reported low Σ OC concentrations and high bulk $\delta^{15}\text{N}$ values in thick-billed murre eggs collected from Prince Leopold Island (Canadian Arctic) in 1998 and argued that some seabirds could be acquiring lipids from invertebrates and protein from fish, resulting in a mismatch between stable isotope signatures and contaminant data.

Although bulk $\delta^{15}\text{N}$ values increased over time in the eggs of our species, bulk $\delta^{15}\text{N}$ (and $\delta^{13}\text{C}$) did not correlate with Σ DDT or Σ PCBs in our species, as established by Elliott et al., 2021b. Instead, egg $\delta^{15}\text{N}$ values in most trophic and source amino acids remained relatively constant over time. Our results are consistent with Hebert et al. (2022) who found no significant change in fatty acid concentrations (EPA and DHA) in Pacific food webs as measured in LSPE eggs collected from Hippa, Storm, and Cleland Islands from 1994 to 2015. Recent increases in bulk $\delta^{15}\text{N}$ values in higher consumers may have been influenced by eutrophication (Elliott et al., 2021a), N_2 fixation (Montoya et al., 2002), or other allochthonous inputs. Alternatively, the increasing baseline $\delta^{15}\text{N}$ values for double-crested cormorant eggs in our study suggests that these birds shifted to a diet of benthic prey and away from herring and schooling fish possibly due to overfishing, as previously documented by the temporal decline in egg $\delta^{34}\text{S}$ values for cormorants (Elliott and Elliott, 2016). Thus, although bulk

$\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ egg values varied interannually, such trends may have overestimated the extent to which seabirds biomagnified certain POPs (Hebert and Popp, 2018). We recommend that future studies incorporate compound-specific amino acids and other dietary tracers when analyzing contaminant temporal trends.

4.1. Toxicological implications

The time frame of the present study encompasses the era when ambient legacy POPs were causing toxicological effects, particularly in top predators until the present time when those effects largely disappeared, and previously affected species of aquatic and predatory birds recovered (Anderson et al., 1975; Grier, 1982; Best et al., 2010; Blus, 2011). Concentrations of POPs in our species are now below any thresholds associated with overt effects on reproduction (STable 7), as they have been for some time (Elliott et al., 1989; Harris et al., 2005). Available laboratory and field data on, for example, PCBs, dioxins, and polybrominated diphenyl ethers (PBDEs) in birds, were reviewed and concluded that, except at some extreme contaminated site hotspots, overt effects were not occurring at least since the early 2000s (Harris and Elliott, 2011). Recently, more work has been done on POPs in Arctic locations, despite generally lower exposures, but related to long range transport issues and other climate indices (Vorkamp et al., 2022; Bianchini et al., 2022; de Wit et al., 2022). Balanced reviews concluded that there was minimal evidence that organohalogenes were having significant impacts on the overall health of most Arctic species, including seabirds (Letcher et al., 2010). The only convincing evidence for ongoing effects of POPs in recent years has been from areas impacted by past major point sources, many of which are now designated superfund sites in the USA and subject to Natural Resource Damage Assessments (Custer, 2011; Edson et al., 2011; Fredricks et al., 2011; Bursian et al., 2013). Some lingering effects have also been reported from agricultural and urban areas in Canada heavily treated with DDT (Iwaniuk et al., 2006; Kesic et al., 2021; Elliott et al., 2005; Read et al., 2021) and cyclodienes (Okoniewski et al., 2006; Elliott and Bishop, 2011; Brogan et al., 2017).

Thus, the evidence from many quarters is that most legacy POPs have significantly declined in wildlife, with limited evidence of overt effects outside of highly contaminated sites. Regardless, some authors contend and have used individual-based models to support arguments that PCBs will continue to exert such drastic toxicological effects in marine ecosystems that they will result in the extirpation of, for example, many populations of killer whales (*Orcinus orca*; Desforges et al., 2018). Not all agreed with those methods or the rationale (Elliott and Trites, 2018; Witting, 2019). As we have shown here, top predators were historically exposed to much higher concentrations of POPs in the 1970s. Levels of Σ PCBs in the eggs of our seabird species have declined precipitously, including in areas designated as critical habitat (Kim et al., 2022) and, in some cases, stabilized and/or remained uniformly low in recent years. In stark contrast to the model-based forecasts of Desforges et al. (2018), some northern and transient killer whales along the Pacific coast of BC continue to thrive and populations to grow (Ross et al., 2000), despite being potentially more contaminated than resident orcas due to a more generalist diet (COSEWIC, 2008).

Within the area of our monitoring species, Victoria Harbor is a federally contaminated site due to past industrial activity. Subtidal surface (5–10 cm) sediments recently collected from Selkirk Waterway and Victoria/Esquimalt Harbors had some of the highest PCB concentrations, relative to samples collected from 96 other sites along the south coast of BC (Kim et al., 2022). Accordingly, Huang et al. (2018) found that ~18 % of river otters (*Lontra canadensis*) inhabiting Victoria had PCB concentrations in their scat and plasma that exceeded the critical effect level (> 16 mg/kg ww) with some evidence of endocrine disruption; however, reproduction and survival did not appear to be impacted at the population level since otters exhibited high levels of self-recruitment and emigration (Elliott et al., 2008; Guertin et al., 2010, 2012). We are in the process of developing molecular tools to explore whether such subtle perturbations can be detected

in our long-term monitoring seabird species at current POP exposure levels (King et al., in preparation).

5. Summary and conclusions

From the temporal data presented in this study (spanning a period of 51 years), we find that many legacy POPs are either significantly declining (e.g., *p,p'*-DDE, β -HCH, HCB, HE, oxychlorane, Σ PCBs) or showing no directional change over time (Σ Mirex) in the eggs of our long-term monitoring species sampled from the Pacific coast of BC. Contaminants such as α -HCH, *cis*- and *trans*-chlorane, OCTS, *p,p'*-DDT, *p,p'*-DDD, and dieldrin in our seabird eggs also showed evidence of downward trends, largely influenced by non-detect values during more recent sampling periods. Bulk and compound-specific amino acid analyses suggested no major changes in trophic position or baseline food web signatures during the study period. Temporal trends observed here were comparable to other seabird species and pelagic food webs, as well as other top predators from different geographic locations. Collectively, our findings suggest that international controls on POPs releases, principally the Stockholm Convention on Persistent Organic Pollutants, have been effective in greatly reducing the contamination of marine and other ecosystems by these persistent (P), bioaccumulative (B), and toxic (T) compounds.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.160084>.

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