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Exposure to persistent organic pollutants is linked to over-wintering latitude in a Pacific seabird, the rhinoceros auklet, *Cerorhinca monocerata*[☆]

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ABSTRACT

Seabirds are wide-ranging organisms often used to track marine pollution, yet the effect of migration on exposure over the annual cycle is often unclear. We used solar geolocation loggers and stable isotope analysis to study the effects of post breeding dispersal and diet on persistent organic pollutant (POP) and mercury (Hg) burdens in rhinoceros auklets, *Cerorhinca monocerata*, breeding on islands along the Pacific Coast of Canada. Hg and four classes of POPs were measured in auklet eggs: organochlorine insecticides (OCs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and perfluoralkyl substances (PFASs). Stable isotope values of adult breast feathers grown during winter were used in conjunction with geolocation to elucidate adult wintering latitude. Wintering latitude was the most consistent and significant predictor of some POP and of Hg concentrations in eggs. The magnitude and pattern of exposure varied by contaminant, with \sum PCBs, \sum PBDEs and DDE decreasing with wintering latitude, and mirex, perfluoro-n-tridecanoic acid, and Hg increasing with latitude. We suggest that concentrations of these contaminants in rhinoceros auklet eggs are influenced by variation in uptake at adult wintering locations related to anthropogenic inputs and oceanic and atmospheric transport.

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1. Introduction

Chemicals such as persistent organic pollutants (POPs) and mercury (Hg) can move long distances via atmospheric and oceanic processes, leading to contamination of ecosystems distant from their points of release (Selin, 2009; Lohmann et al., 2007). A high affinity for lipids and resistance to degradation leads to bioaccumulation of POPs and their metabolites in wildlife (Borgå et al., 2004), which can affect behaviour, development, reproduction, survival and ultimately, in some instances, cause population

declines (e.g. Anderson et al., 1975; Elliott et al., 1988; Blus, 2011; Harris and Elliott, 2011). Hg, meanwhile, has been released from a variety of industrial and commercial activities, particularly the burning of fossil fuels, and because it is a naturally occurring element, does not chemically degrade (Driscoll et al., 2013; Mason et al., 2012). Hg is converted by bacteria in sediments or the water column to methyl-mercury which is both toxic and bioaccumulative (Scheuhammer et al., 2008; Driscoll et al., 2013). In response to demonstrated impacts on ecosystems and human health, regulations and restrictions on many contaminants have been implemented internationally, e.g. the 1999 Canadian Environmental Protection Act, the 2008 Stockholm Convention on Persistent Organic Pollutants, the 2009 Long Range Transboundary Air Pollution Protocol on POPs (Miller et al., 2020), and the 2013 Minamata Convention on Mercury (Selin, 2014). Despite those efforts to limit or ban the production, use, and distribution of POPs

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and Hg, problems persist. Numerous contaminated sites remain, some compounds continue to be produced in countries outside of North America, legacy contaminants are transported through ecosystems, and new contaminants are developed and used; thus, ongoing monitoring to assess the fate of both legacy and newer POPs is necessary (Elliott and Elliott, 2013).

Seabirds are effective indicators of marine pollution as they integrate contaminant exposure over time and space, and different species enable sampling of a range of habitats, from coastal to offshore. Congregation at breeding colonies enables researchers to sample returning adults by collection of eggs, blood, feathers or other tissues (Elliott et al., 1989; Furness and Camphuysen, 1997; Holmström et al., 2005; Elliott and Elliott, 2013; Miller et al., 2014, 2015, 2020). The egg, for example, is a matrix in a convenient and robust package for sampling with a high and consistent lipid and protein content and represents an integrated measure of exposure.

Seabird populations can, however, make complex seasonal migrations, and unless at least the broad scope of such movements outside the breeding season is known, there is uncertainty about the source(s) of exposure (Fort et al., 2013; Leat et al., 2013; Ely and Franson, 2014; Miller et al., 2020). Where species lie on the capital to income breeding strategy is also an important factor in determining the effect of overwintering location on the degree of contamination measured at breeding sites (Stephens et al., 2009). Capital breeders mobilize lipid and other reserves accumulated over the winter or during migration to reproduce, and thus a strong signal is carried over from the overwinter grounds. Income breeders, in contrast, rely on energy and nutrient intake from the breeding area, and, therefore, would more closely represent breeding ground signals (Elliott et al., 2007). Recently, broad scale spatial data obtained by advanced telemetry devices, such as satellite, GPS and solar geolocator devices, has enabled elucidation of migration patterns, which are essential to determine if non-breeding season movements impact contaminant uptake and concentrations (e.g. Elliott et al., 2007; Yates et al., 2010; Leat et al., 2013; Miller et al., 2020; Albert et al., 2021). Interpretation of data on contaminant exposure in recent years also has improved with the measurement of stable isotopes in egg fractions, which function as biochemical tracers. Stable isotopes of nitrogen or the ratio of ^{15}N – ^{14}N , represented in relative terms as $\delta^{15}\text{N}$, provide information on the trophic enrichment of biomagnifying pollutants and is useful in distinguishing effects of diet vs environmental contaminant concentrations (Hebert et al., 2009; Miller et al., 2014; Braune et al., 2015). However, baseline $\delta^{15}\text{N}$ varies spatially and so variation in $\delta^{15}\text{N}$ may also capture variation within and across habitats (Elliott et al., 2021). The ratio of ^{13}C – ^{12}C varies depending on the carbon source for the local community, with terrestrial communities usually depleted in ^{13}C compared to marine communities and benthic/littoral communities being enriched compared to pelagic communities (Kainz et al., 2002; Ethier et al., 2008). However, $\delta^{13}\text{C}$ also increases with trophic level, albeit slower than $\delta^{15}\text{N}$.

In the current study, we measured Hg, four classes of POPs and stable isotopes in eggs of an indicator species the rhinoceros auklet, *Cerorhinca monocerata*. Our objective was to use solar geolocation and stable isotope analysis to examine how variation in breeding colony, diet and particularly wintering location may influence concentrations of contaminants in eggs collected at breeding colonies.

2. Methods

2.1. Study species

The rhinoceros auklet, *Cerorhinca monocerata*, (hereafter auklet)

is a nocturnal, monogamous seabird with biparental care that spends the majority of its life at sea in temperate waters of the North Pacific Ocean (Bertram et al., 1991; Ydenberg, 1989; Kouwenberg et al., 2016). In winter, auklets breeding on colonies in British Columbia disperse widely after breeding, and in winter are distributed in continental shelf waters along the west coast of North America from Mexico to the Gulf of Alaska (Hipfner et al., 2020). By March they return to their breeding grounds and will lay a single egg in a colonial burrow-nesting environment in April or May (Wilbur, 1969; Wilson and Manuwal, 1986). Rhinoceros auklets are epipelagic predators that feed across a range of trophic levels, from zooplankton to forage fish (Burger et al., 1993; Hipfner et al., 2013). They are mainly zooplanktivorous through the pre- and early-breeding season, but become increasingly piscivorous after chicks hatch (Hipfner et al., 2013). At the breeding colony, it is possible to access a burrow during incubation, tag an adult with a geolocator to track movement throughout the year, and in the following year retrieve the tag and collect the egg for contaminant analysis (Elliott and Elliott, 2013). Collection of the egg early in incubation facilitates relaying of a replacement egg (Hipfner et al., 2008). Stable isotopes in breast feathers grown outside the breeding season provide a spatio-temporally integrated signal of wintering diet (Kainz et al., 2002). Across 9 auk species and 42 sites, rhinoceros auklets in the Gulf of Alaska had the highest head feather (breeding season signal) and fifth highest breast feather (non-breeding season signal) Hg concentrations, implying that auklets in our region of study may have exceptionally high exposure (Albert et al., 2021).

2.2. Sites and geolocator attachments

From 2013 to 2017, a total of 300 solar geolocation loggers (GLS) were deployed on adult auklets at four breeding colonies located off the Pacific coast of British Columbia, Canada (Table S1). Details of the geolocators used, deployment methods, and data retrieval can be found in Hipfner et al. (2020), including permit numbers and field methods.

2.3. Analysis of tracking data

Details of tracking data analysis have been previously reported (Miller et al., 2020; Hipfner et al., 2020). Two GLS tag types were used in the current study, Migrate Intigeo-C65 and Lotek LAT 2900 Series Avian tags. The Migrate tags store light level data, which were converted to latitude/longitude using the TwilightFree methods (Bindoff et al., 2018). Lotek tags have a built-in algorithm that automatically converts light level data into coordinates (Ekstrom 2007). Wintering centroid coordinates were calculated for Jan 1-Feb 28 to minimize potential overlap with movements during migration periods.

2.4. Egg and feather sampling

The year after GLS deployment, burrows were re-accessed within 48 h of laying. The adult bird with the leg-mounted GLS unit was removed from the burrow, weighed and measured, the unit retrieved and a few breast feathers taken. The relatively undeveloped egg was then collected. A total of 84 complete GLS datasets were retrieved, of which 37 eggs were analyzed for contaminants. Of these 37 egg-tracking data pairs, 28 were associated with tracking data from female auklets (Table S2). Eight eggs were collected in 2014 (Lucy Island, n = 3; Pine Island, n = 1; Triangle Island, n = 4), 22 in 2015 (Cleland Island, n = 2; Lucy Island, n = 6; Pine Island, n = 1; Triangle Island, n = 13), four in 2016 (Lucy Island, n = 1; Triangle Island, n = 3) and two in 2017 (Triangle Island,

$n = 2$). Eggs were kept in cool locations and where possible stored on ice in the field, then frozen at $-20\text{ }^{\circ}\text{C}$ until chemical analysis.

2.5. Stable isotope analysis

Adult breast feathers and the same egg homogenate as used for chemical analyses were analyzed for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ at the G.G. Hatch Stable Isotope Laboratory, Ottawa University following procedures reported earlier (Miller et al., 2015) (See Electronic Supplement, Section 1). Egg homogenate $\delta^{13}\text{C}$ values were lipid-corrected following previous methods (Elliott et al., 2014), as variation in lipid content can obscure variation in $\delta^{13}\text{C}$. Final delta values are presented in parts per thousand (‰) relative to international standards Vienna PeeDee Belemnite ($\delta^{13}\text{C}$) and air ($\delta^{15}\text{N}$).

2.6. Chemical analysis and quality assurance

Further details on analytical methods for chemicals and stable isotopes can be found in the electronic supplement to this paper.

2.6.1. Mercury

Total mercury (THg) was analyzed in 15 egg homogenates from burrows with a tagged female auklet in an accredited laboratory for metals analyses at the National Wildlife Research Centre (NWRC), Environment Canada (Electronic Supplement, 2.1). Established analytical methods were followed, as previously described (Weech et al., 2006; Scheuhammer et al., 2008). The method limit of quantification (MLOQ) was 0.09 ng/g dw.

2.6.2. Organochlorine pesticides (OCs) and polychlorinated biphenyls (PCBs)

Egg homogenates from 37 eggs (28 from tracked female auklets), were analyzed for OCs and PCBs at NWRC ($n = 8$) and the GLIER laboratory ($n = 29$) (Electronic Supplement, Section 2.2). Photomirex was only analyzed at NWRC ($n = 8$) and therefore removed prior to calculating the sum of all detected OCs and not further analyzed statistically. The MLOQ was <0.10 ng/g ww for all PCBs and OCs. PCB-17 and -18 were only analyzed at GLIER and therefore removed prior to calculating \sum PCBs and further statistical analyses.

2.6.3. Polybrominated diphenyl ethers (PBDEs) and non-PBDE halogenated flame retardants

Egg homogenates from 37 eggs (28 from tagged female auklets), were analyzed for PBDEs at NWRC ($n = 8$) and the Great Lakes Institute for Environmental Research (GLIER, $n = 29$). NWRC Methods for flame retardant analyses have been described earlier (Chen et al., 2013; Miller et al., 2015) and are detailed in the Electronic Supplement (Section 2.3).

At GLIER, chemical extraction and clean up followed the procedures of Lazar et al. (1992). Some congeners (b-TBECH/BDE15, BDE 190, and BDE209) were only analyzed at NWRC ($n = 8$) and were thus excluded from the calculated \sum PBDE and statistical analyses. Non-PBDE halogenated flame retardants (BTBPE, HBB, syn-D, anti-DP, a-TBECH, HBCD, BB101) were only analyzed at NWRC ($n = 8$) and therefore excluded from statistical analysis due to limited sample size; furthermore, HBCD was the only non-brominated FR detected above MLOQ. The MLOQ was <0.5 ng/g ww for the majority of PBDE congeners.

2.6.4. Perfluoroalkyl substances (PFASs)

Egg homogenates from 15 burrows with tagged female auklets were analyzed for PFASs at NWRC, Environment Canada. The perfluoroalkyl substance (PFAS) extraction, cleanup, and analysis method has been described previously (Gebbinck and Letcher 2012;

See Electronic Supplement, 2.4). In brief, this method includes extraction of the analytes from the sample using acetonitrile, and clean-up with a weak anion exchange solid phase extraction (SPE) cartridge (WAX). After SPE separation and clean-up process the sample fraction containing carboxylics and sulfonates was analyzed by ultra-high performance liquid chromatography tandem mass spectrometry equipped with an electrospray ionization source. MLOQ values varied from 0.01 ng/g ww for PFHxDA to 9.03 ng/g ww for L-PFDS.

2.7. Statistical analyses

All contaminant concentrations were \log_{10} -transformed prior to statistical analyses, with the exception of Hg, which did not require \log_{10} -transformation to achieve normality. Values $<$ MLOQ were assigned half the MLOQ. Trace congeners, with $\geq 75\%$ reported below the MLOQ, were removed prior to calculating the \sum contaminant values (i.e., \sum PBDE, \sum PCB, \sum PFCA).

We selected a suite of 11 contaminant variables to represent the major contaminant groups (Fig. S1). For each contaminant, we used the *glm* function in R to fit a series of General Linear Models (GLMs; R Core Team, 2020) for each of four explanatory variables (breeding colony, dN15, dC13, and wintering latitude). Wintering latitude and longitude are very strongly correlated ($r = -0.93$, $P < 0.001$; Fig. 1), and therefore only latitude was considered. In each model, both response and explanatory variables were scaled by centering on the mean and dividing by the standard deviation. We then used an information-theoretic approach (Anderson and Burnham, 2002) to evaluate which of the four variables received the most support in explaining variation in the response variable. To broadly assess whether contaminant loads were mostly related to breeding colony, diet, or wintering location, we calculated the Akaike Information Criterion, corrected for small samples sizes (AIC_c) for each model, and used the Akaike weights (w) as the measure of model support. We also based our inferences on specific correlations between explanatory variables and each contaminant, and on parameter estimates and their confidence intervals. Given the small sample sizes, we considered both 95% and 80% confidence intervals to evaluate any potential correlations.

3. Results

Of the 37 adult birds with GLS tracks and the egg collected, nine were males. Since members of auklet breeding pairs may overwinter in different locations, we conducted a separate statistical analysis of the 37 birds of both sexes, and the 28 females. We focus on the data for the 28 female birds, but include the results for all 37 birds in an electronic supplement (Fig. S2).

Contaminant loads varied among rhinoceros auklets and were most strongly affected by wintering distribution. Across the four explanatory variables, variation in contaminant values was best explained by wintering latitude, and on average, models with this explanatory variable received more support from the data than other models (mean $w = 0.36$; Table 1). Models with $\delta^{15}\text{N}$, on average, received the second most support (mean $w = 0.29$), and models with breeding colony or $\delta^{13}\text{C}$ had reduced support (mean $w < 0.20$).

Correlations with measures of wintering distribution, diet, and breeding colony varied widely among the 11 focal contaminants (Fig. 2). Of the 11 contaminants considered, five of them (\sum PCBs, \sum PBDEs, PFTrDA, mirex, Hg, and DDE) were associated with wintering latitude, with parameter values having 80% or 95% confidence intervals that did not encompass 0 (Fig. 2). For mirex, PFTrDA, and Hg, this relationship was positive, indicating birds wintering further south had lower values (Fig. 3), in contrast to

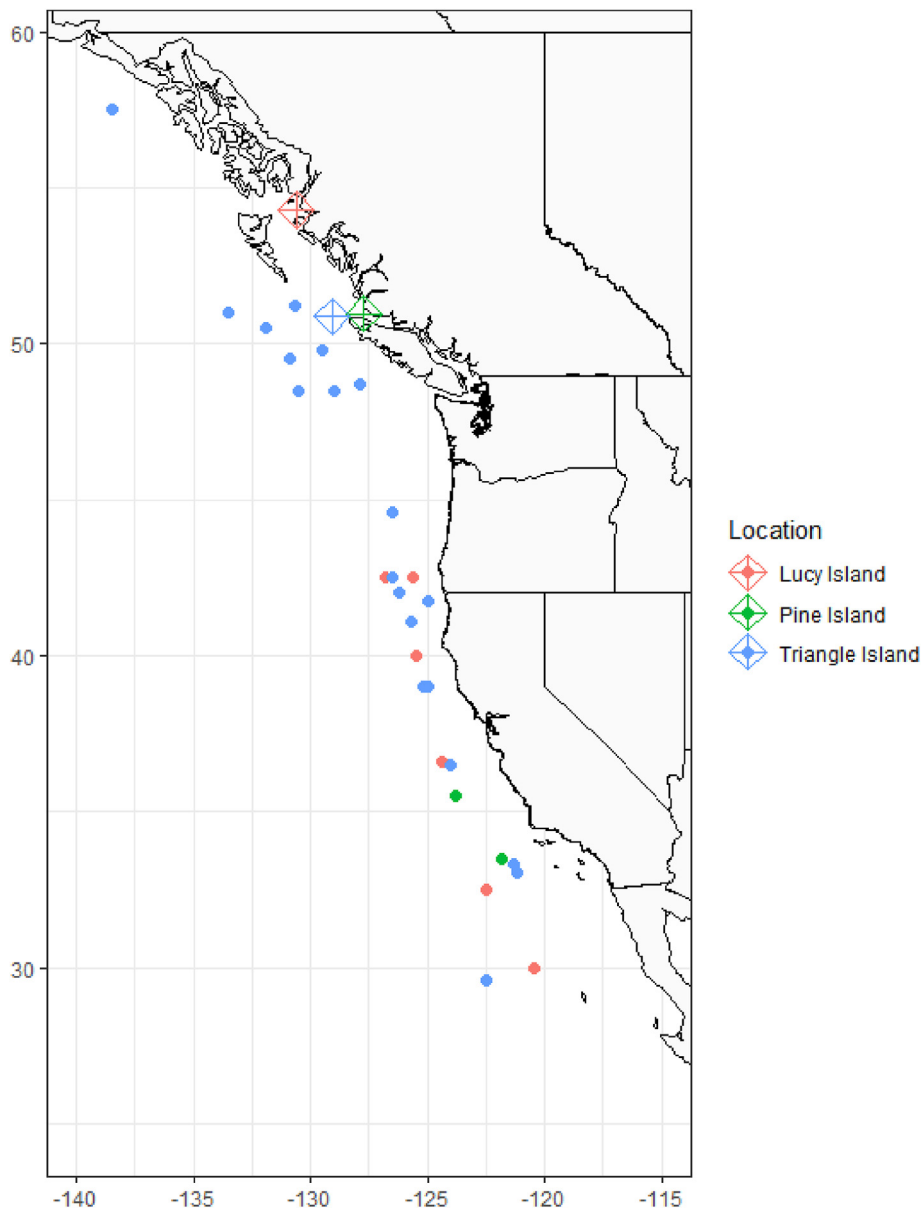


Fig. 1. Wintering (Jan 1 – Feb 28) range of female rhinoceros auklets GLS tracked from breeding colonies (diamonds) with individual wintering area centroids (points) in the eastern North Pacific Ocean. Wintering area colours correspond to breeding locations: Lucy Island 54°18'N; 130°37'W, Pine Island, 50°98'N; 127°72'W, Triangle Island, 50°86'N, 129°08'W. These locations are within the following traditional territories: Metlakatla and Lax Kw'alaams (Lucy Is.), Tlatasikwala and Gwa'sala-Nakwaxda'xw (Pine Is.), Tlatasikwala and Quatsino (Triangle Is). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1

Relative support for four models explaining variation in 11 selected contaminants measured in 28 eggs of rhinoceros auklets, collected from female birds which had been fitted with a geolocator to track movement throughout an annual cycle. Collection colonies were located on the Pacific coast of Canada, 2014 to 2017.

Model	Variable	Akaike Weight		
		Mean	Min	Max
1	Breeding Colony	0.16	0.00	0.61
2	Wintering latitude	0.36	0.01	0.94
3	Egg $\delta^{15}\text{N}$	0.29	0.01	0.98
4	Egg $\delta^{13}\text{C}$	0.19	0.01	0.59

DDE, PBDEs, and PCBs where lower values were found in birds wintering further north (Fig. 3).

Models with $\delta^{15}\text{N}$ had negative parameter values across all 11

focal contaminants (Fig. 2), with two of them, HCB, and β -HCH with 95% confidence intervals that did not overlap zero (Fig. 2), and two, Σ chlordanes and Hg at 80%. Relationships with $\delta^{13}\text{C}$ were both positive and negative across the 11 contaminants (Fig. 2), and none of them had non-zero 95% confidence intervals. Considering 80% confidence intervals, five contaminants (Σ PBDEs, PFUDA, PFTTrDA, HCB, and β -HCH) showed a negative association with $\delta^{13}\text{C}$ values.

Analysis of the results for the larger data set of 37 eggs from burrows with both males and females did not change the relationships between contaminants and wintering latitude or isotopes at the 95% confidence level, with one exception, β -HCH was negatively related to $\delta^{13}\text{C}$ (Fig. S2).

Mean values of contaminants were generally similar among the three breeding colonies (Fig. 4), with the exception of Σ chlordanes and Σ PCBs, which were higher from birds breeding on Pine Island. Mean values for concentrations of the various contaminants are

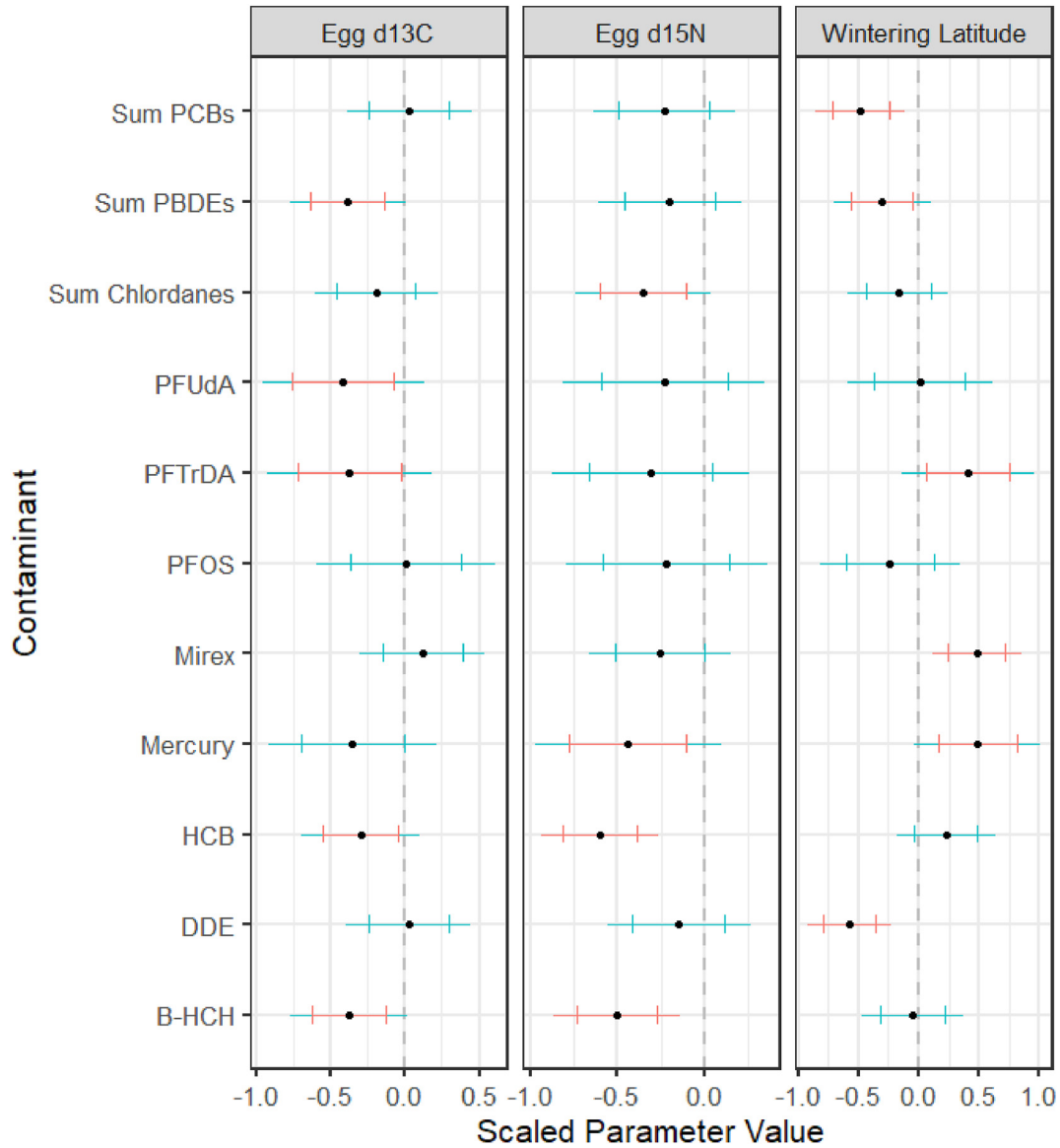


Fig. 2. Parameter estimates of General Linear Models explaining variation in 11 contaminants measured in 28 eggs of rhinoceros auklets, collected from female birds which had been fitted with a geolocator to track movement throughout an annual cycle. Collection colonies were located on the Pacific coast of Carravieri et al. (2014) to 2017. Circles depict the parameter value and horizontal bars depict 80% and 95% confidence intervals. Vertical dashed line indicates 0 value. Parameters with non-zero overlapping confidence intervals are shown in red, and zero-overlapping intervals are shown in blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

available in the electronic supplement (Tables S3 to S10).

Across all breeding colonies, adult feathers were enriched in ¹⁵N and ¹³C compared to eggs (Fig. S4). Overlap of standard error bars for egg isotope values between breeding locations suggests a similar diet during egg production among breeding colonies, although the small sample of two eggs from Cleland Island was ¹⁵N-enriched compared to other locations. There was a significant negative correlation between $\delta^{13}\text{C}$ in adult feathers and wintering latitude ($r = -0.41$, 95% CI = -0.65 to -0.09 , $p = 0.014$), but feather $\delta^{15}\text{N}$ and wintering latitude were not correlated (Fig. S5). Stable isotopes in eggs were not significantly correlated with adult wintering latitude, but there was a trend of lower $\delta^{15}\text{N}$ in eggs laid by adults overwintering at higher latitudes (Fig. S3).

4. Discussion

4.1. Influence of wintering latitude

Wintering latitude was the most consistent predictor of POP and Hg concentrations in rhinoceros auklet eggs, with the magnitude and pattern of exposure varying by contaminant. At the 95% confidence level, DDE and $\sum\text{PCBs}$ were significantly greater and mirex lower in eggs laid by auklets wintering at lower latitudes. Similarly, $\sum\text{PBDE}$ concentrations were higher in eggs of females that wintered at lower latitudes and Hg and PFtriDA lower, but only with an 80% confidence level. It appears, therefore, that some of the more recalcitrant contaminants can be acquired over the winter,

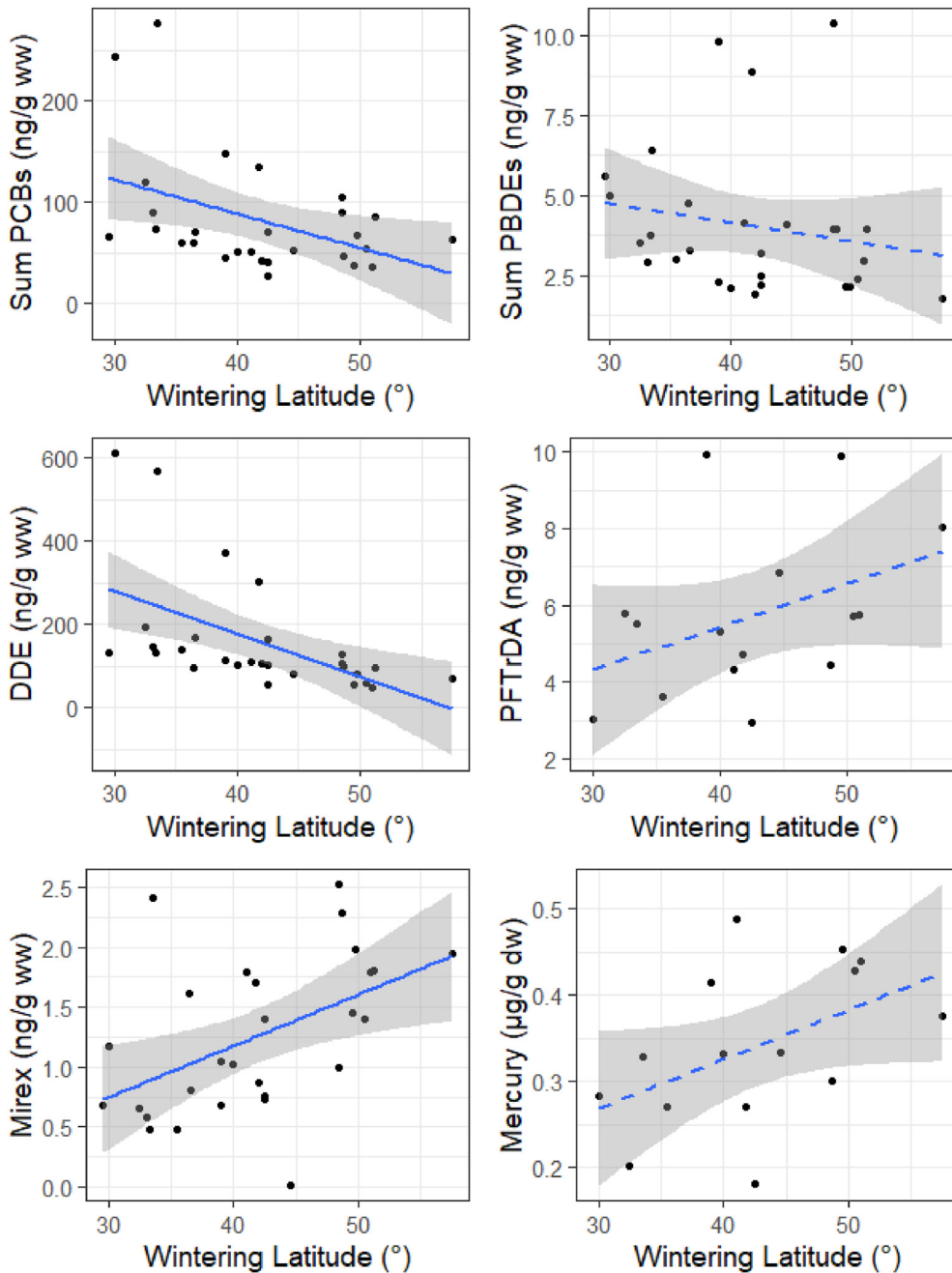


Fig. 3. Representative plots of contaminants in eggs of rhinoceros auklets, collected from female birds which had been fitted with a geolocator to track movement throughout an annual cycle. Contaminants concentrations are compared to wintering latitudinal gradients determined as the centroid of the wintering distribution. All regressions have coefficient values with 80% confidence intervals that do not overlap 0. Solid lines are regressions that have coefficients with 95% confidence intervals that do not overlap 0.

retained in the body lipid pool until returning to the breeding colonies, and deposited into eggs of rhinoceros auklets. The spatial relationships found here also are reasonably consistent with latitudinal differences in onshore sources, and atmospheric and oceanic transport of POPs and Hg. Contaminants measured in rhinoceros auklet eggs from colonies on the Pacific coast of Canada provide an indication of contamination of the continental shelf region of the west coast of North America. That finding is reasonably consistent with the original objective of this long term monitoring program to monitor trends in contamination in this key indicator species representative of the Pacific continental shelf (Elliott et al., 1992).

Most of the auklets that went south wintered on the American coastline from southern Oregon, along the length of California to the Baja. That region has an extensive agricultural sector with widespread and intensive DDT use for several decades, particularly in the Central Valley region of California. For example, runoff contaminated the San Joaquin River, which in turn transported DDT compounds to the coast (Pereira et al., 1996). Dicofol, an insecticide that includes several DDT-related compounds, was used as a replacement for DDT, with as much as 250,000 kg applied in the San Joaquin Valley in 1991 alone (Pereira et al., 1996). Similarly, a major DDT manufacturing site located near Los Angeles was the source of extensive contamination of the offshore region known as

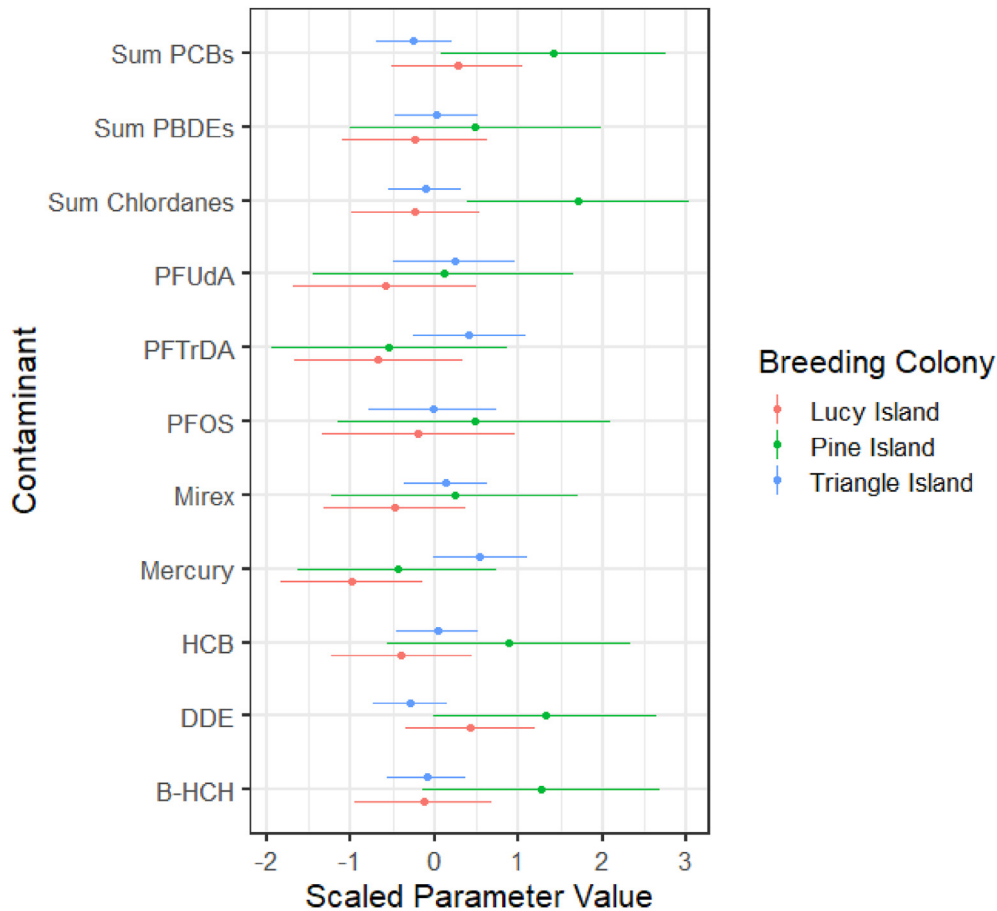


Fig. 4. Parameter estimates of General Linear Models explaining variation in 11 contaminants measured in 28 eggs of rhinoceros auklets, collected at three colonies on the coast of British Columbia, Canada. Circles depict parameter value (the mean relative contaminant value for each colony), and horizontal bars depict 95% confidence intervals.

the Palos Verdes Shelf, including marine birds and raptors. That contamination caused significant negative effects on eggshell quality and reproduction, causing population declines of brown pelicans (*Pelecanus occidentalis*) (Albert et al., 2021), cormorants (Gress et al., 1973), bald eagles (*Haliaeetus leucocephalus*) (Sharpe and Garcelon, 2005), and peregrine falcons (*Falco peregrinus*) (Peakall and Kiff, 1979). Blood samples of bald eagle nestlings sampled in the early 2000s at Catalina Island off the coast of Southern California had two to three-fold greater concentrations of DDE than eaglets sampled on the British Columbia coast (Elliott et al., 2009). The Columbia River, which has the largest discharge volume on the Pacific coast of North America, is also likely to be a major source of DDT input to the marine environment of the Pacific coastline. DDT use was intensive for decades in the fruit growing regions of the Columbia basin extending up to the Okanagan valley of southern Canada (Blus et al., 1987; Harris et al., 2000). Species such as osprey and bald eagle nesting along the Lower Columbia River still had elevated residues of DDE in eggs into at least the late 1990s (Elliott et al., 2000; Buck et al., 2005), as did aquatic mammals (Elliott et al., 1999). The effect of agricultural runoff along the U.S. coast on auklet DDE levels is evident where the nine birds that wintered along the Canadian/Alaskan coastline (above 45°N) averaged lower levels of DDE than those that wintered along the contiguous U.S. coastline (below 45°N; Fig. 3).

Elevated exposure to commercial chemicals such as PCBs and PBDEs in eggs of birds that wintered along the U.S. coastline reflect the greater overall human population (~51 million) and the larger urban areas compared to the coast of British Columbia (~5 million).

Similar to the spatial trends in DDT-related compounds, PCB exposure was greater in eagles from the California coast compared to British Columbia (Elliott et al., 2009), and in eagles, osprey and aquatic mammals from the Lower Columbia compared to the Fraser River (Elliott et al., 1999; Elliott et al., 2007; Buck et al., 2005). The greater contamination of the Columbia River is due to the extensive development of hydroelectric generation facilities along its entire length, and past use of PCBs in transformers. In contrast, the other major Pacific rivers, such as the Fraser and the Skeena, have relatively minor or no hydroelectric development. Air samples taken during a cruise across the Pacific found that greatest PCB concentrations occurred off the southern coast of California; air masses originated from the west coast of North America (Zhang and Lohmann, 2010). For PBDEs and likely other flame retardants, degree of contamination of ambient environments is related to size of urban populations. A study of peregrine falcon eggs from the California coast revealed a significant positive relationship between egg concentrations of \sum PBDEs and human population size of the city where the nest was located (Newsome et al., 2010; Park et al., 2009). Other studies have shown increased PBDE concentrations in various avian indicator species in more urban situations (Henny et al., 2011; Chen et al., 2013; Elliott et al., 2015; Brogan et al., 2017; Currier et al., 2020). Greater concentrations of PBDEs in air over urban than rural areas has also been reported (Harner et al., 2006). The effect of human population on auklet PCB/PBDE levels is evident in Fig. 3 where the nine birds with winter home range centroids along the Canadian/Alaskan coastline (above 45°N) averaged lower levels than those with winter home range centroids

along the contiguous U.S. coastline (below 45°N). The one exception was three birds whose centroids were off of Vancouver Island, at the northern edge of the Davidson Current that brings waters north from California. This bird had the highest levels of PBDEs, perhaps due to partial foraging in the California Current. Interestingly, the two birds wintering off of Mexico, along relatively remote coastlines, averaged higher levels of PBDEs and PCBs (and DDE) than those wintering farther north. Possibly, they obtained those contaminants during their long migration along the U.S. coast.

With one exception, PFTrDA (Perfluorotridecanoic acid), which exhibited a weak increase with wintering latitude, there were no significant spatial patterns in PFAS contamination. These compounds are also generally associated with greater urbanization (Gewurtz et al., 2016, 2018), and similar to the other POPs, we might expect greater relative contamination of the California versus British Columbia coastal environment. Wildlife near some large urban areas of California have elevated PFAS contamination (Sedlak et al., 2017). Perfluorinated contaminants can be transported long distances and are contaminants of seabirds in the arctic and pelagic species in the north Pacific (Braune and Letcher, 2013; Miller et al., 2015). PFTrDA and PFUdA (perfluoroundecanoic acid) increased over the past three decades in seabird species monitored in various north temperate and arctic regions (Miller et al., 2015). Production sources of the precursors for these compounds were identified as mainly in Asia, particularly Japan. However, Miller et al. (2020) did not find greater concentrations of these longer chained PFCAs in ancient murrelets, *Synthliboramphus antiquus*, which wintered on the north Asian coast versus those wintering on the Canadian coast.

Other telemetry studies of the effects of migration on contaminant exposure have produced varying results. Leat et al. (2011) used feather stable isotopes and geolocators to study the influence of migration on exposure of great skuas, *Stercorarius skua*, breeding at three colonies in the NE Atlantic and which wintered at three different locations. The influence of wintering location on POPs concentrations in plasma samples of adults captured on the breeding colony was small relative to differences in exposure at breeding colonies, although accumulation of POPs during the winter may be important for specific populations of seabirds. Miller et al. (2020) measured concentrations of POPs and Hg in blood samples of another species of seabird, the ancient murrelet (*Synthliboramphus antiquus*), at four colonies on the Pacific Coast of Canada. No significant differences in exposure were found between murrelets that wintered off the Canadian Pacific coastline versus those that migrated to and wintered along the north Asian coast (Miller et al., 2020). The eggs of thick-billed murrelets (*Uria lomvia*) wintering farther north in the High Arctic had lower levels of Σ PCB, Σ DDT, Σ CHLOR, Σ CBZ, and dieldrin than conspecifics wintering farther south, due to different atmospheric deposition patterns in the High and Low Arctic (Braune et al., 2002). Annual migration of Arctic seabirds to more temperate latitudes generally leads to higher OC concentrations than species that remain in northern latitudes year-round (Braune et al., 2005). However, in the Southern Ocean, latitudinal variation, inferred from $\delta^{13}\text{C}$, and measured in blood of wandering albatross (*Diomedea exulans*) sampled at a subantarctic colony was the reverse of what we observed here (Carravieri et al., 2014). PCBs increased with latitude, thought to support the global distillation hypothesis of transfer of POPs to polar regions, and Hg decreased, thought to relate to more complex food chains in subtropical waters. Meanwhile, spatial trends in most POPs in giant petrel (*Macronectes giganteus*), and consistent with our northern hemisphere study, decreased with increasing latitude of colonies sampled in the Southern Ocean (Roscales et al., 2016). Examination of congener patterns did find that, for example, lighter PCB compounds, increased with latitude, which they considered to be consistent with predictions of global transport

models.

Telemetry has been used to investigate sources of DDE contamination in non-marine long-distance migrants in Western North America. Results depended on factors such as relative contamination of breeding grounds in the US Pacific Northwest and British Columbia versus wintering grounds further south in the US and in Mexico and Central America, and income versus capital life history strategy. (Henny and Blus, 1986) used radio telemetry on white-faced ibis (*Plegadis chihi*) to identify sources of DDE at wintering sites in the US Southwest. A satellite telemetry study of osprey (*Pandion haliaetus*) reported greater DDE exposures on the breeding grounds, thought to be caused by overall greater use of DDT in the USA and Canada, along with the slower rates of degradation and volatilization from colder climate soils (Elliott et al., 2007). A later study of ibis using satellite telemetry located elevated DDE exposure on the wintering grounds in Mexico (Yates et al., 2010). The capital breeding strategy of the white-faced ibis, which lays eggs within a few days of returning to nesting sites, is likely an important factor in the evident carry-over of contamination in that instance (Yates et al., 2010).

Although sample size is low and the data variable, egg Hg concentration increased with winter latitude in rhinoceros auklets. There are broad hemispheric patterns of greater Hg concentrations in air masses over the northern than southern half of the Pacific consistent with greater past and present releases in the Northern Hemisphere (UNEP, 2018). We did not find any data on spatial Hg trends over the approximately 30 degrees of latitude of our study, 28° to 58°. Higher average Hg concentrations were found in North Pacific Ocean water above 40° and reflect circulation of the three main water masses in the region (Sunderland et al., 2009). Differences in Hg burdens among seabird species suggest exposure at higher latitudes in the arctic (Braune et al., 2006).

Using museum stored feathers, increasing temporal trends have been reported in seabirds from the tropical Pacific and the Canadian Arctic (Braune et al., 2006; Vo et al., 2011; Braune et al., 2016; Bond et al., 2015). Those increases took place from the late 1800s during the industrial revolution until the 1970s and 1980s and the regulatory restrictions on Hg release in many jurisdictions, and are consistent with trends and deposition from air (UNEP, 2018). However, over the most recent five decades at temperate latitudes, temporal trends of Hg in eggs of rhinoceros auklets and other seabird species in both the North Pacific and North Atlantic have remained flat (Burgess et al., 2013; Elliott and Elliott, 2016). More recently, there are disconnects between Hg trends in biota versus air, apparently because levels of methylmercury in biota are no longer limited by availability of Hg, but by rates of methylation and demethylation and other variables affecting bioaccumulation (Elliott and Elliott 2016; UNEP, 2018).

4.2. Influence of breeding and winter diet

The significant correlation between feather $\delta^{13}\text{C}$ and wintering latitude provides support that adult rhinoceros auklet breast feathers represent a spatially and temporally integrated winter diet. Hobson et al. (1997) reported ^{13}C -depletion in high-latitude surface water compared to low-latitude surface water, and consistent with that report, auklets wintering at lower latitudes had ^{13}C -enriched feathers compared to conspecifics wintering at higher latitudes. Stable isotope biplots of rhinoceros auklet eggs indicated more variation in breeding season diet compared to wintering diet, but overall diet was comparable among breeding colonies. The absence of consistent significant relationships between stable isotopes in eggs and contaminant concentrations may be related to limited dietary variation among individual auklets.

The absence of a significant relationship between egg $\delta^{15}\text{N}$ and

some highly bioaccumulating chemicals, such as DDE, PCBs and PBDEs may be in part caused by the limited variation in egg $\delta^{15}\text{N}$ among breeding locations. Some studies have reported that PBDEs did not increase with $\delta^{15}\text{N}$, such as in bald eagles (Elliott et al., 2009), possibly because baseline variation in $\delta^{15}\text{N}$ may be large enough to obscure any relationship between $\delta^{15}\text{N}$ and trophic level (Elliott and Elliott 2016; Grenier et al., 2020). The capacity of seabirds to metabolize some OCs, e.g., α -hexachlorocyclohexane and certain chlordanes-related compounds, could explain the absence of trophic enrichment of some compounds (Borgå et al., 2005). Concentrations of a number of compounds were significantly lower in ^{15}N -enriched eggs. That unexpected relationship for PCBs and several OC pesticides likely does not indicate an absence of trophic enrichment, given their high capacity to bioaccumulate (Borgå et al., 2004; Fremlin et al., 2020), and could be explained by the negative correlation between egg $\delta^{15}\text{N}$ and adult wintering latitude. Eggs laid by adults wintering at lower latitudes tended to be ^{15}N -enriched and PCBs and OCs were measured at higher concentrations in adults wintering at lower latitudes. A decrease of $\delta^{15}\text{N}$ with latitude has been reported in Southern Hemisphere albatrosses, for example (Jaeger et al., 2010). A variety of processes could alter ^{15}N baselines and variation in ^{15}N independent of trophic position. They include: zooplankton feeding on detritus from higher trophic level predators, prey feeding on food webs influenced by agricultural input (typically enriched in ^{15}N) or feeding in river plumes (freshwater is depleted in ^{15}N), all of which could influence food chains leading to seabirds (Montoya et al., 2002; Grenier et al., 2020). However, those local-scale phenomena likely do not explain the decrease in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ with latitude. Rather, N_2 fixation by bacteria in oligotrophic waters, such as the subtropical waters off California, decreases baseline $\delta^{15}\text{N}$ across large spatial gradients (Montoya et al., 2002), and similar processes may be at work for $\delta^{13}\text{C}$.

4.3. Influence of breeding location

Breeding colony location was a significant predictor of egg concentrations of ΣPCBs and $\Sigma\text{chlordanes}$, but not other contaminants. Elevated concentrations of those legacy POPs were measured in the limited, and thus potentially biased, sample of two eggs from Pine Island, located at the northern end of Vancouver Island, in an area with relatively little industrial and virtually no agricultural development, making the finding difficult to interpret. Harris et al. (2005) identified small, but significant latitudinal gradients in several OC pesticides and PCBs in cormorant eggs from colonies along the southern Pacific Coast of Canada, with areas of the southern Strait of Georgia more contaminated than the less populated northern strait, a finding consistent with proximity to centres of human activity.

5. Conclusions

Some legacy and emerging POP and Hg concentrations in rhinoceros auklet eggs are influenced by variation in contaminant uptake at adult wintering locations related to anthropogenic inputs and transport, rather than dietary differences during winter or breeding periods. Thus, non-breeding season movements can significantly influence contaminant uptake and concentrations in seabirds migrating along the Pacific coast of North America. The data also show the long term contamination of POPs on regional environments. The relatively greater contamination of the US Pacific coastline by both legacy POPs, DDTs and PCBs continues five decades after being originally highly restricted, while newer compounds, such as PBDEs despite being banned from commerce in North America, continue to contaminate the offshore coastal

system. These data on seasonal movements and contaminant exposure reveal that rhinoceros auklet eggs are broad indicators of contamination by persistent chemicals of the continental shelf ecosystem along the Pacific Coast of North America.

Credit author statement

John E. Elliott: Conceptualization, Writing – revised draft; review & editing, Project, Administration, Funding acquisition. Mark Drever: Conceptualization, Formal analysis, Writing; Katharine Studholme: Sample Preparation, Investigation; Veronica Silverthorn: Initial formal analysis, Writing – original draft. Aroha A. Miller Writing – initial draft; Kyle H. Elliott: Visualization, Writing – review & editing. Ken G. Drouillard: Investigation. Sandi Lee: Investigation, Data curation; Abde Idrissi: Investigation; Emily Porter: Investigation; Glen T. Crossin: Conceptualization; J. Mark Hipfner: Conceptualization, Funding acquisition, Project Management, Writing – editing and reviewing.

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

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