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Spatial and temporal trends in brominated flame retardants in seabirds from the Pacific coast of Canada

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) are bioaccumulative flame retardants. PBDEs increased in many ecosystems during the late 20th century, but recently have declined in some environments. To examine trends in the northern Pacific, we analysed PBDEs, HBCDD and carbon and nitrogen stable isotopes ($\delta^{13}C$ and $\delta^{15}N$) to account for dietary effects in archived eggs of three seabird species from British Columbia, Canada, 1990–2011 (rhinoceros auklets, *Cerorhinca monocerata*; Leach's storm-petrels, *Oceanodroma leucorhoa*; ancient murrelets, *Synthliboramphus antiquus*, 2009 only). PBDEs increased until approximately 2000 and then decreased, while HBCDD increased exponentially throughout the examined period. No significant changes in dietary tracers were observed. HBCDD and ΣPBDE levels varied among species; ΣPBDE also varied among sites. Temporal changes in contaminant concentrations are unlikely to have been caused by dietary changes, and likely reflect the build-up followed by decreases associated with voluntary phase-outs and regulations implemented in North America to control PBDEs.

1. Introduction

Despite efforts to reduce the release of chemical contaminants from sources, for example land based manufacturing and use or ocean dumping and incineration, large inputs still accumulate in oceans, thus there is a need for ongoing monitoring. Seabirds have proven to be among the most effective tools for such monitoring (Elliott and Elliott, 2013). Brominated flame retardants (BFRs) are produced to ostensibly reduce the fire risk of various materials e.g., plastics, rubbers, construction materials and textiles (Alaee et al., 2003; Gauthier et al., 2007; Sjödin et al., 2003). Some BFRs have become ubiquitous, being found in sediment, sewage sludge, air and biota (see Chen and Hale, 2010; Darnerud, 2003; Daso et al., 2010; De Wit, 2002; De Wit et al., 2010; Kefeni et al., 2011; Sellström et al., 2003) and are typically persistent, bioaccumulative and lipophilic (De Wit, 2002). There are five main classes of BFRs, which include polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCDD), tetrabromobisphenol A (TBBA), polybrominated biphenyls (PBBs) and other brominated flame retardants (European Food Safety Authority, 2014).

PBDEs were produced commercially at three different degrees of bromination – penta-BDE, octa-BDE and deca-BDE (Alaee et al., 2003; De Wit, 2002). Generally, lower brominated congeners have a longer range transport potential similar to polychlorinated biphenyls (PCBs) (Wania and Dugani, 2003), and bioaccumulate due to their persistent, lipophilic nature (De Wit, 2002). Organisms feeding at higher trophic levels tend to have a greater exposure potential (Braune et al., 2007) but see Elliott et al. (2009). Several congeners have been reported to cause various detrimental effects to birds in laboratory studies, including endocrine disruption, particularly on the thyroid hormone system (Darnerud, 2008), and developmental anomalies (Eng et al., 2012; Winter et al., 2013). HBCDD is produced as a mixture of three stereoisomers – α, β and γ (Alaee et al., 2003; Covaci et al., 2006). HBCDD has been used for approximately 30 years and has a high bioaccumulation potential (De Wit, 2002). As with PBDEs, detrimental effects of HBCDD have been observed in laboratory studies (Covaci et al., 2006; Crump et al., 2010). PBDEs and HBCDD pose an ecotoxicological risk to environmental and human health.

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In Canada, PBDEs were declared toxic under the Canadian Environmental Protection Act 1999 (Environment Canada, 2004). Penta- and octa-BDE mixtures were voluntarily phased out in Canada in the early 2000s, which were soon followed by regulatory restrictions (Canadian Gazette, 2006), and in 2009 were included under the Stockholm Convention on Persistent Organic Pollutants (POPs) (Stockholm Convention on Persistent Organic Pollutants, 2008). Manufacture of the tetra-to deca-BDE congeners and use, sale and import of tetra-, penta- and hexa-BDE congeners in Canada are prohibited (Environment Canada, 2008). At the time of writing, HBCDD was under consideration for addition to the Stockholm Convention on POPs (Stockholm Convention on Persistent Organic Pollutants, 2008), and the Long Range Transboundary Air Pollution Protocol on POPs (Arnot et al., 2009). The Canadian Government has proposed implementing regulations in line with these international regulations (Environment Canada and Health Canada, 2012).

Besides regulations, biological factors have been suggested to contribute to observed temporal changes in PBDE concentrations in biota (Lavoie et al., 2010). Stable isotope analysis can be used as a proxy to evaluate changes in trophic position and, therefore, allows consideration of whether changes in contaminant concentrations are due to diet or contaminant abundance. Aquatic bird eggs are a widely used matrix for environmental contaminant monitoring (Burger and Gochfeld, 2004; Crosse et al., 2012; Elliott and Elliott, 2013; Furness and Camphuysen, 1997; Mondreti et al., 2013), and can be used for stable isotope analysis, which are commonly used to examine source factors (Braune et al., 2002; Elliott et al., 2014; Morrissey et al., 2010) and have been effective in demonstrating that variance in egg contaminant concentrations can also be influenced by changes in diet rather than changes in contaminant concentrations in the environment (Hebert and Weseloh, 2006; Hebert et al., 2000).

Prior to restrictions, PBDE concentrations were increasing rapidly in eggs of great blue herons (*Ardea herodias*) and double-crested cormorants (*Phalacrocorax auritus*) from inner coastal and estuarine sites on the southern Pacific coast of Canada (Elliott et al., 2005). Here, we examine PBDE concentrations in eggs from rhinoceros auklets, Leach's storm-petrels and ancient murrelets that forage further offshore and away from point sources (cities, industries) during the period of egg formation, and therefore likely reflect temporal trends in food webs from the eastern North Pacific Ocean. The aim here is twofold: 1) to investigate how PBDE and HBCDD concentrations have developed both temporally and spatially since regulations were instigated in Canada to reduce emission, production and use of many PBDEs; and 2) to determine what relationship exists between diet and contaminant concentrations in these populations.

2. Materials and methods

2.1. Study species

The rhinoceros auklet, *Cerorhinca monocerata*, inhabits temperate waters of the northern Pacific (Bertram et al., 1991; Ydenberg, 1989). It is an epipelagic piscivorous feeder preying on schooling fish (Burger et al., 1993) and migrates south in winter (Vermeer, 1979). Leach's storm-petrel, *Oceanodroma leucorhoa*, is a planktivorous surface feeder distributed throughout the northern Atlantic and Pacific Ocean (Hedd and Montevecchi, 2006). Outside of the breeding season, petrels feed many hundreds of kilometres from the breeding colony, beyond the continental shelf edge (Hedd and Montevecchi, 2006). The ancient murrelet, *Synthliboramphus antiquus*, is an offshore, sub-surface feeder that preys on zooplankton and small, schooling fish (Sealy, 1975). Adults feed almost exclusively offshore when not on land to breed (Sealy, 1975). These species lay a single (auklets, petrels) or two eggs (murrelets) annually in a colonial burrow-nesting environment (Wilbur, 1969; Wilson and Manuwal, 1986).

2.2. Sites, sampling matrix and design

Eggs were sampled from four islands on the Pacific coast of British Columbia, Canada – Cleland Island (auklet, petrel); Lucy Island (auklet); Hippa Island (petrel); and Langara Island (murrelet) (Fig. 1). One fresh egg was removed per sampled nest at the beginning of the laying period. Nests were selected randomly. Eggs were stored frozen until preparation for analysis occurred. All islands are located in open coastal positions and are distant from large urban centres. Eggs were collected at four yearly intervals during spring and early summer (late April to early July). Not all sites were sampled in the same years due to cost and logistics (see Table S1, electronic supplementary material).

Fifteen eggs from individual nests were collected every four years for auklets and petrels. Eggs were analysed retrospectively as one pool of 15 eggs (to conserve archived samples) from 1990/1991 until either 2002/2003 or 2006/2007, from whence they were analysed as 5 pools of 3 eggs each, as well as being re-analysed as one group of 15, as per previous years. A total of 18 murrelet eggs from Langara

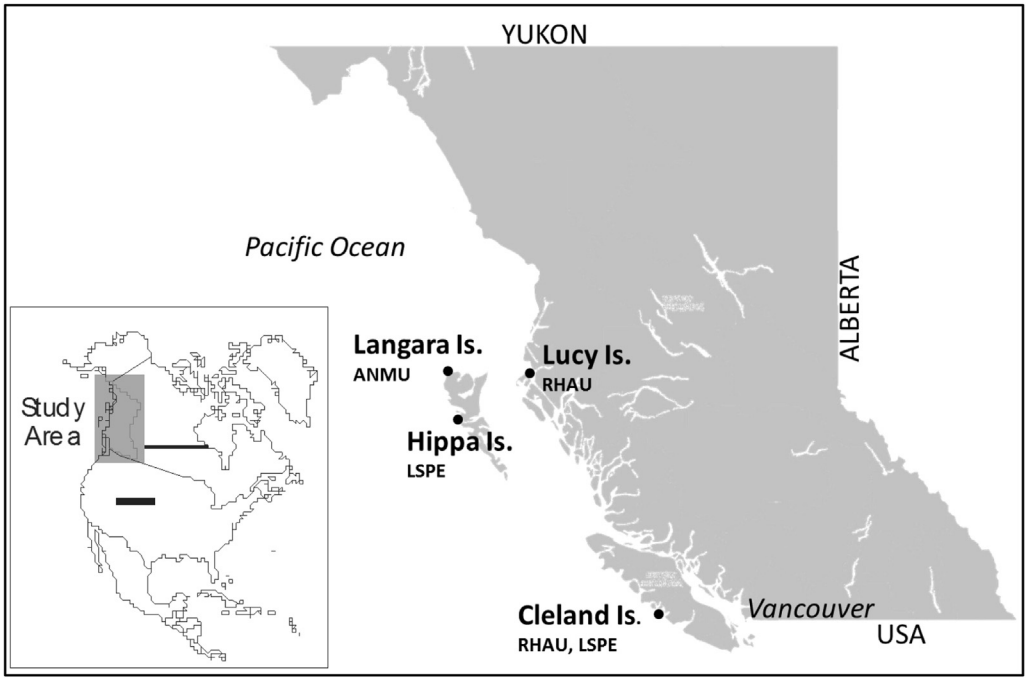

Fig. 1. Map indicating each island where a seabird colony was sampled within British Columbia, Canada. RHAU = rhinoceros auklet (*Cerorhinca monocerata*), LSPE = Leach's storm-petrel (*Oceanodroma leucorhoa*), ANMU = ancient murrelet (*Synthliboramphus antiquus*).

Island were collected in 2009, and analysed as 6 pools of 3 eggs. The whole egg was homogenised. Approximately 1 g from each egg pool was subsampled and sent for chemical analysis. Subsamples were archived individually and as equal weight pools at −40 °C at the Canadian Wildlife Service National Wildlife Specimen Bank (Elliott et al., 1988). Moisture and lipid content were recorded for pooled samples. Because eggs were collected fresh and stored frozen, contaminant concentrations were not corrected for egg moisture content (Peakall and Gilman, 1979).

2.3. Chemical analysis and Quality Assurance

Egg homogenates were analysed at the National Wildlife Research Centre as per methods described earlier (Chen et al., 2013). Briefly, approximately 1.5 g wet weight (ww) of aliquots were homogenized with anhydrous sodium sulphate (Na_2SO_4) followed by a neutral extraction with DCM:Hexane (1:1). The homogenate was spiked with labelled internal standards (BDE-30, BDE-156 and/or ^{13}C-BDE-209). A portion of the resulting extract was removed for lipid determination. Lipids and biogenic materials were removed from the extract, with further clean-up by Florisil column chromatography. Purified sample extracts were analysed for BDE/BFRs using capillary gas chromatograph (Agilent 6890N), coupled with a mass selective detector (Agilent 5973N). A 30 m long DB-5 fused-silica column (25 mm ID, 0.25 μm film thickness, J&W Scientific, Agilent Tech) was used. The injector was operated in splitless injection mode, held at 260 °C. Initial oven temperature was held at 100 °C for 3 min; increased by 20 °C/min to 180 °C; then increased by 5 °C/min to 325 °C (held for 4 min). The determination of α-HBCDD by GC–MS is representative of total-HBCDD as any β- and γ-HBCDD residues are thermally isomerized to α-HBCDD at temperatures exceeding 160 °C in the injection port. Contaminant recovery values for BDE-30 were 43–69%; for BDE-156 41–75%; and for ^{13}C-BDE-209, recovery values were 13–41%.

All analytical samples were monitored for interferences and contamination. A standard reference material (Lake Michigan fish tissue homogenate from the National Institute of Standards and Technology) was analysed with each batch of samples. All reported PBDE congeners were within ±20% of the certified values, except for co-eluted BDE 154/BB 153. Duplicate extractions, clean ups and injections were performed. Data were verified to ensure test method criteria were met. In most years the method limit of detection (LOD) was <0.10 ng/g for all congeners, except for HBCDD (1.0 ng/g) and BDE 209 (from 1.0 to <5.0 ng/g). LOD was pre-determined by method validation. Concentrations were determined in eight replicate chicken egg samples spiked with native standards with concentrations near the instrument detection limit. The detection limit (95% probability of detection) was obtained by multiplying the standard deviation by three. BDE-15 and β-TBECH, and BDE-154 and BB-153 coeluted. Non-PBDE flame retardants analysed for included alpha and beta tetrabromoethylcyclohexane (α-TECH, b-TECH/BDE 15), hexabromobenzene (HBB), HBCDD, brominated biphenyl (BB) 101, 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), and stereoisomers of dechlorane plus (*syn*-DP, *anti*-DP).

2.4. Stable isotope analysis

Stable isotope analysis was carried out using the same egg homogenate as used for chemical analyses and described elsewhere (Elliott et al., 2014). Briefly, 1 mg subsamples were freeze-dried, loaded into tin cups and analysed using a PDZ Europa ANCA-GSL elemental analyser interfaced to a PDZ Europa 20-20 isotope ratio mass spectrometer (IRMS; Sercon Ltd., Cheshire, UK) at the Stable Isotope Facility, University of California, Davis (http://stableisotopefacility.ucdavis.edu). Samples were analysed for ^{13}C/^{12}C and ^{15}N/^{14}N isotopes. During analysis, samples were interspersed with several replicates of at least two different laboratory standards. The final delta values were presented in parts per thousand (‰) relative to international standards Vienna PeeDee Belemnite and Vienna Cañon Diablo Troilite (δ^{13}C) and air (δ^{15}N), respectively. The δ^{13}C values were lipid normalised as variation in lipid content can obscure variation in δ^{13}C (Elliott et al., 2014).

2.5. Statistical analysis

Concentrations are presented on a lipid weight basis to allow inter-species and spatial comparisons due to variation in egg lipid content between species. Values below the method limit of detection (LOD) were replaced by the LOD divided by the square root of two (Helsel, 1990). Arithmetic means were calculated for each year where multiple pooled samples were available for species and site. The sum of HBCDD isomers (α,β,γ) are presented as HBCDD.

A principal component analysis (PCA) was used to examine the pattern of distribution of dominant congeners and HBCDD between auklet and petrel eggs at all sites for all years ($n = 6$ for each species/site combination). Simple linear regression was conducted on egg moisture and lipid content over time. The proportion contribution of each dominant congener to ΣPBDE was calculated for the most recent year of sampling. General linear models (GLM) were used to examine the relationship of log-transformed HBCDD or log-transformed ΣPBDE with species (auklets, petrels), site (south–Cleland Island; north–Lucy and Hippa Islands) and year.

HBCDD and ΣPBDEs are presented for the entire time series. Natural log transformed dominant congener and (log)ΣPBDE concentrations were spilt into pre- and post-usage restrictions, circa-2000, providing three monitoring times points pre-restrictions (1990/1991 to 1998/1999), and three post-restrictions (2002/2003 to 2010/2011) and examined using simple linear regression. In all cases, $n = 3$ (yearly average concentrations for three years), hence the lack of data points means results must be interpreted with caution. Doubling times were calculated from the slope of the regression line and assume first-order (exponential) increases (Elliott et al., 2005). Only data from 2009 was available for murrelets, thus temporal trends were not examined.

Temporal trends of stable isotopes of δ^{13}C and δ^{15}N were examined using simple linear regression. An outlier was removed (petrel, Hippa Island, δ^{15}N, 0.32‰; δ^{13}C, −29.57‰). Dominant congeners and ΣPBDE (wet weight basis) from auklet and petrel eggs collectively, and by species and site, as well as from murrelet eggs, were examined for any relationship with δ^{13}C, δ^{15}N and lipid percentage and year using multiple linear regression. Statistical analyses were conducted in Excel 2010 and R v3.0.3.

3. Results

3.1. Biological parameters

No significant change in average moisture or lipid content was seen over time in rhinoceros auklet or Leach's storm-petrel eggs at any site. Lipid content in murrelet eggs was almost twice as high as for the other two species (Table S2, electronic supplementary material).

3.2. Dominant PBDE congener trends

In auklet eggs, dominance usually occurred in the order of BDE 47 > BDE 100, BDE 99 > BDE 154/BB 153 (Table 1). BDE 154/BB 153 was more dominant than the other penta-BDE congeners in petrel eggs in most years. For murrelet eggs dominant congeners were BDE 47 > BDE 99 > BDE 100 > BDE 154/BB 153 (Table 1). PCA showed dominant PBDE congeners grouped together, while HBCDD formed a separate group, with 90% of the variance explained by the first two axes (Fig. S1, electronic supplementary material).

3.3. Non-PBDE flame retardants

Since 1995, HBCDD was the dominant congener out of all PBDE and non-PBDE flame retardants measured in petrel eggs at Hippa Island and since 1998 in petrel eggs at Cleland Island. By 2010, HBCDD was second dominant in auklet eggs from Cleland Island, and dominant in auklet eggs from Lucy Island. In murrelet eggs, HBCDD was second dominant (Table 1). In 2011, HBB levels in petrel eggs at both Cleland and Hippa Islands were 0.86 and 1.1 ng/g lw, contributing 1.4% and 2.1% respectively to the sum of all monitored flame retardants. Murrelet eggs from Langara Island showed quantifiable levels of HBB (range 1.1–3.7 ng/g lw, mean 2.4 ± 0.4), while HBCDD concentrations were higher than many PBDE congeners. All other targeted non-PBDE flame retardants were detected, although most were below LOQ, and are thus not presented or discussed further.

3.4. Spatial patterns

For the most recent sampling year at each site (see Table S1), the highest mean ΣPBDE, BDE 99 and BDE 154/BB 153 concentrations were observed in murrelet eggs from Langara Island (Table 1, Fig. 2a, b). Mean ΣPBDE for auklet eggs at Cleland Island was 110.0 ng/g lw (Fig. 2a), higher than auklet eggs at Lucy Island. Concentrations of penta-BDE congeners (lw) were higher in auklet eggs than petrel eggs (Table 1). HBCDD concentration was highest in petrel eggs from Hippa Island, followed by petrel eggs from Cleland Island (Fig. 2a). BDE 47 and HBCDD were the two dominant congeners at each site in the most recent year (Table 1). However, in murrelets, BDE 99 also showed a comparatively high proportion (Fig. 2b), contributing ~22% to the ΣPBDE. Dominant congener

Table 1
Arithmetic mean of lipid and moisture percentages, dominant congener, ΣPBDE of 14 congeners (17, 28, 47, 49, 66, 85, 99, 100, 138, BDE 154/BB 153, 153, 183, 190, 209), and HBCDD concentrations (ng/g lw) for eggs of all three species from examined colonies in each monitored year. All values are presented to 1 decimal place. A – denotes years where HBCDD was detected but not quantified.

Species	Rhinoceros auklets											
Location	Cleland Island						Lucy Island					
Year	1990	1994	1998	2002	2006	2010	1990	1995	1999	2003	2006	2010
N	15[a]	10[a]	11[a]	15[a]	15	15	16[a]	15[a]	15[a]	15	15	13
Lipid %	11.6	6.5	8.5	7.0	16.5	12.0	12.0	11.2	10.7	11.8	12.3	9.5
Moisture %	69.5	69.2	68.2	69.0	70.9	69.4	70.4	69.3	68.9	68.0	70.9	61.8
BDE 47	38.2	139.1	150.0	179.3	31.3	45.6	14.7	28.0	55.9	27.5	23.3	24.3
BDE 99	27.9	80.0	45.8	84.4	13.0	11.4	12.3	23.2	33.9	21.2	13.4	10.6
BDE 100	23.7	70.1	96.7	100.7	17.7	19.0	9.4	21.6	25.6	16.7	14.3	11.2
BDE154/BB153	33.1	69.6	39.9	49.7	6.6	9.8	13.4	17.1	14.9	7.4	6.9	5.4
ΣPBDE	153.4	444.6	405.5	486.4	83.9	110.0	8.0	12.9	17.3	11.6	14.5	83.3
HBCDD	–	–	–	–	7.6	26.2	–	–	–	2.6	7.4	36.2

Species	Leach's storm-petrel												Ancient murrelet
Location	Cleland Island						Hippa Island						Langara Island
Year	1990	1994	1998	2002	2006	2011	1991	1995	1999	2003	2007	2011	2009
N	15[a]	15[a]	16[a]	16[a]	15	18	15[a]	15[a]	10[a]	15[a]	15	15	18
Lipid %	11.8	3.1	11.4	10.2	12.2	11.3	10.2	10.0	12.0	12.5	9.9	11.1	19.4
Moisture %	70.9	72.7	71.3	70.7	71.9	71.9	73.7	70.0	70.7	70.8	71.3	72.7	61.2
BDE 47	3.5	32.2	20.1	17.1	3.0	8.8	3.1	4.8	5.4	3.1	6.2	3.7	42.8
BDE 99	2.5	30.4	15.5	10.7	2.4	4.6	1.3	1.9	2.4	2.2	4.9	2.0	31.5
BDE 100	2.6	36.8	20.0	16.2	4.6	7.3	1.1	2.4	3.5	2.9	4.4	3.7	14.3
BDE154/BB153	16.2	86.3	23.3	15.2	3.7	5.4	12.1	8.7	11.2	3.6	8.9	5.2	13.4
ΣPBDE	36.3	252.4	102.0	79.9	40.1	60.1	30.5	35.4	37.0	25.2	41.1	51.7	139.1
HBCDD	–	45.9	35.0	62.4	18.2	53.3	11.0	34.5	110.7	20.6	213.3	88.9	32.7

[a] Denotes years when eggs were a single pooled sample.

concentrations in the most recent sampling year were highest in auklets (BDE 47, BDE 100) or murrelet eggs (BDE 99, BDE 154/BB 153).

3.5. Temporal trends

Log (HBCDD) increased with year ($t_{20} = 4.03$, $p < 0.001$) and varied with species (GLM, $t_{20} = -6.09$, $p < 0.001$) and but not site ($t_{20} = -0.47$, $p = 0.65$), while log (ΣPBDE) varied with site (GLM, $t_{19} = 3.34$, $p < 0.01$) and species ($t_{19} = 4.58$, $p < 0.001$) but not with year ($t_{19} = -0.75$, $p = 0.46$). The majority of dominant congeners in auklet eggs at both sites and petrel eggs from Cleland Island tended to increase pre-2000s and decrease post-2000 (Table S3, electronic supplementary material). However, in petrel eggs at Hippa Island, only BDE 99 showed a decreasing tendency post-2000. All other dominant congeners and ΣPBDE tended to increase at this site; however ΣPBDE concentration was the lowest here of all sites examined (Fig. 3a). HBCDD was below LOD in petrel eggs from Cleland Island in 1990, and in auklet eggs at both Cleland and Lucy Island until 2002 and 1999 respectively (Fig. 3a–d), thus no regression was conducted pre-2000 for HBCDD in auklet eggs. HBCDD showed a significant increasing trend ($p < 0.01$) pre-2000 in petrel eggs at Hippa Island (Table S3). Post-2000, a weak, decreasing tendency in HBCDD was observed in petrel eggs at Cleland Island. At the other examined sites, HBCDD tended to increase (Table S3).

3.6. Stable isotopes

There was little temporal variation in C or N stable isotope ratios in auklet and petrel eggs from any site, with $\delta^{15}N$ values in both species between 12‰ and 16‰. $\delta^{13}C$ values in auklet eggs were more enriched (−17.3‰ to −19.4‰) than murrelets (−19‰ to −19.3‰) and petrels (−20.7‰ to −23.7‰). Petrel eggs from Hippa Island had the lowest $\delta^{13}C$ values (Fig. S2a, electronic supplementary material), while petrel eggs from Cleland Island had the

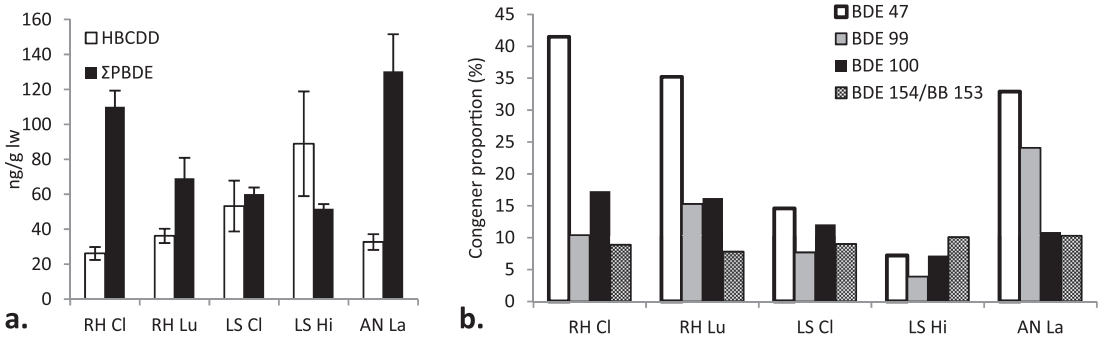

Fig. 2. a) ΣPBDE and HBCDD (ng/g lw) ± SEM and b) dominant congeners as a proportion (%) of ΣPBDE for each species at each site in the most recent sampling year. RHAU Cl = rhinoceros auklet (*Cerorhinca monocerata*), Cleland Island (2010), $n = 5$; RHAU Lu = rhinoceros auklet, Lucy Island (2010), $n = 5$; LSPE Cl = Leach's storm-petrel (*Oceanodroma leucorhoa*), Cleland Island (2011), $n = 6$; LSPE Hi = Leach's storm-petrel, Hippa Island (2011), $n = 5$; ANMU La = ancient murrelet (*Synthliboramphus antiquus*), Langara Island (2009), $n = 6$.

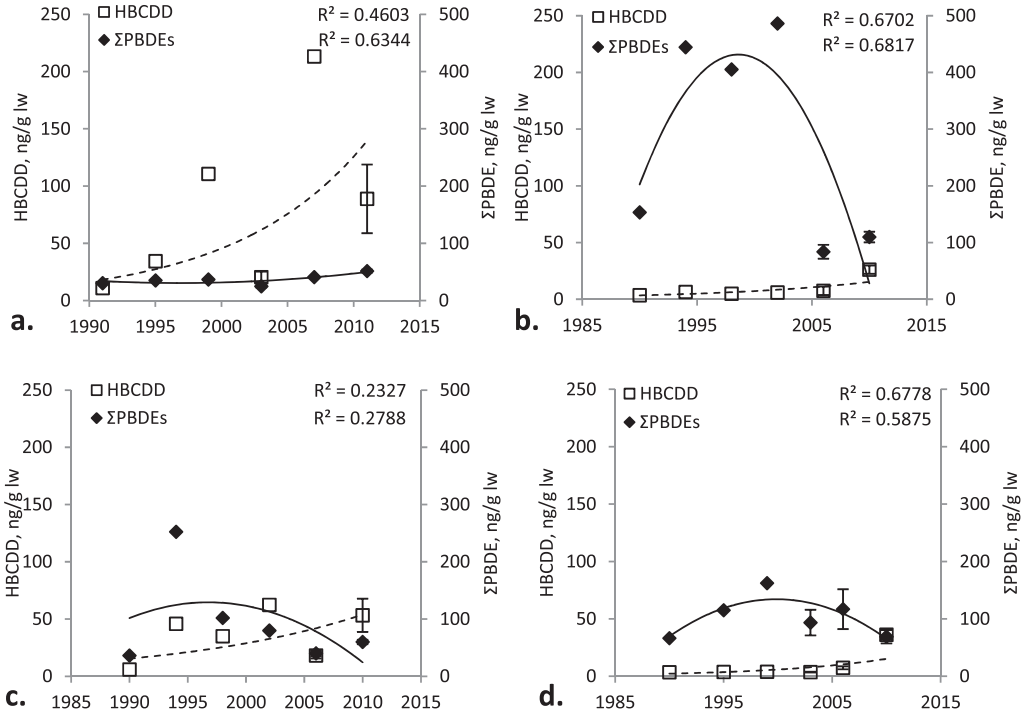

Fig. 3. HBCDD (*y* axis) and ΣPBDE (secondary *y* axis, ng/g lw) over time in a. Leach's storm-petrel eggs from Hippa Island; b. rhinoceros auklet (*Cerorhinca monocerata*) eggs from Cleland Island; c. Leach's storm-petrel (*Oceanodroma leucorhoa*) eggs from Cleland Island; d. rhinoceros auklet eggs from Lucy Island. Standard error of the mean is shown in years where multiple pooled samples were taken; $n = 6$ for all species and sites.

highest $\delta^{15}N$ values (Fig. S2b, electronic supplementary material). When auklet and petrel eggs at all sites were combined, a significant positive relationship was observed between ΣPBDE and $\delta^{13}C$, a significant negative relationship was observed with lipid percentage ($p < 0.001$ and $p < 0.03$ respectively), but no significant relationship with $\delta^{15}N$ was seen. This pattern held true for all dominant PBDE congeners, except BDE 100, where no significant relationship with lipid percentage was observed. For HBCDD, a significant negative relationship was observed only for $\delta^{13}C$ ($p < 0.01$). However, in most cases these relationships did not hold when examined at the level of individual species and site. The exception was auklet eggs at Cleland Island, which showed a significant relationship between ΣPBDE, BDE 100 and 99 with either or both $\delta^{13}C$ ($p < 0.05$) and lipid percentage ($p < 0.02$). Murrelet eggs showed no significant relationships between dominant congeners and stable isotopes or lipid percentage.

4. Discussion

Since the North American industry implemented voluntary phase out of penta-BDE mixtures, dominant PBDE congener concentrations and ΣPBDE have generally been decreasing in rhinoceros auklet eggs from colonies at both Cleland and Lucy Islands, and Leach's storm-petrel eggs at Cleland Island. However, that trend was not apparent for most dominant congeners from petrel eggs collected at Hippa Island, the population feeding farthest from North America and closest to Asia, although doubling times of these congeners slowed considerably post-2000. The lack of temporal changes in $\delta^{15}N$ and $\delta^{13}C$, and the lack of effect of trophic level ($\delta^{15}N$) with dominant congeners and ΣPBDE for individual species and sites, once spatial variance ($\delta^{13}C$) was accounted for, indicates a stable diet over time, hence no effect of trophic level at this scale. Thus, temporal changes in contaminant concentrations in these species are unlikely to be directly attributable to diet, and almost certainly due to restrictions imposed on PBDE usage (Canadian Gazette, 2006; Environment Canada, 2008, 2004).

4.1. Dominant congeners and temporal trends

The three most dominant congeners here (BDE 47, BDE 99, BDE 100) are similar to those seen in other seabird eggs e.g., herring gull eggs from the North and Baltic Sea (Fliedner et al., 2012), ivory gull eggs (*Pagophila eburnea*) from the Canadian Arctic (Braune et al., 2007) and murre eggs from the Baltic Sea (Sellström et al., 2003). BDE 47 has the highest bioavailability potential of these congeners (De Wit, 2002). In 1999, North America constituted about 98% of global demand of commercial penta-BDEs used in polyurethane foam, of which BDE 47 was an important component (Hale et al., 2003). An increased use of penta-BDEs during that period may explain the observed trend. The potential debromination of higher-brominated congeners could also have contributed to the predominance of penta-BDE congeners (Gauthier et al., 2009, 2008, 2007), Additionally, BDE 47 has a higher aqueous solubility compared to e.g., BDE 99 (Tittlemier et al., 2002), indicating a greater environmental mobility of this lower brominated compound. Nonetheless, the dominance of BDE 47 in these species agrees with findings from eggs of double-crested cormorants, great blue herons, ospreys (*Pandion haliaetus*) and Leach's storm-petrel eggs sampled in British Columbia until 2002 (Elliott et al., 2005).

Dominant congener concentrations from auklet and petrel eggs during this period were considerably lower than those from double-crested cormorant and great blue heron eggs from the Pacific coast of British Columbia over the same time period (Elliott et al., 2005). To some degree the more offshore feeding habits of auklets and petrels (Burger et al., 1993; Elliott et al., 1989; Hedd and Montevecchi, 2006; Ydenberg, 1989) compared to the aforementioned bird species, likely contributed to differences in BFR

concentrations (Gauthier et al., 2008). Lower concentrations are likely due to these species feeding in pelagic food webs, where contaminants have been diluted to a greater degree compared to birds feeding in estuaries near large cities. Biota in coastal and land-based freshwater habitats normally have higher concentrations of environmental pollutants due to their proximity to urban areas and industrial activities (Elliott et al., 2005; Gauthier et al., 2008, 2007). It is also possible that, unlike PCBs and organochlorine insecticides, PBDEs and other BFRs have had less time to dissipate globally from highly urbanised and industrialised areas.

Across all sampling, ΣPBDE increased non-linearly until about 2000 followed by a non-linear decrease. When examined individually at each site pre-2000, when regulations did not exist to control PBDEs in Canada, the majority of dominant congeners showed consistent increases in individual species and sites, although these were non-significant most likely due to small sample sizes. HBCDD was below LOD in auklet eggs; however, by 2010, HBCDD featured prominently in auklet eggs at both sites (Table 1, Fig. 3a–d). Post-2000, concentrations of dominant congeners and ΣPBDE generally decreased, although these decreases were non-significant, again probably due to small sample sizes. Penta-BDEs continued to dominate in auklet eggs at both sites. However, HBCDD showed increasing concentrations in auklet eggs at both sites (Table 1, Fig. 3a–d) and remained dominant in petrel eggs from both sites. The general decreases observed in most dominant congeners began at or soon after voluntary restrictions on the use of dominant PBDE mixtures in North America. A study on barn owls (*Tyto alba*) at a site in Belgium and in France, showed HBCDD concentrations were surpassing PBDE concentrations, and that PBDE concentrations were decreasing between 2003/2004 and 2008/2009, in line with EU bans on penta- and octa-BDE commercial mixtures (Eulaers et al., 2014b).

In contrast to results here, ivory gull eggs collected in the Canadian Arctic between 1979 and 2004 showed increasing ΣPBDE trends (Braune et al., 2007); however, herring gull eggs from the Laurentian Great Lakes showed increases in penta- and octa-BDEs until 2000, and then no trend post-2000, likely due to voluntary restrictions and subsequent regulations (Gauthier et al., 2008). Comparably, murre eggs from Sweden showed rapid decreases post late-1980s in the wake of reduced emissions in EU production and use of these chemicals (Sellström et al., 2003). Herring gull eggs collected from the late 1980s until 2008 from three coastal sites in Germany showed decreases in penta- and octa-BDEs, but no decrease for deca-BDEs, in line with restrictions and regulations in Europe, where an EU-wide ban on deca-BDEs began in 2008 (Fliedner et al., 2012).

In contrast to the other colonies examined here, most dominant congeners in petrel eggs at Hippa Island post-2000 showed weakly increasing concentrations. Concentrations at this colony were low compared to the other colonies. HBCDD remained dominant at that site. Decreases in dominant congener concentrations were seen between 2007 and 2011. It is unclear whether these decreases are an ongoing trend and further sampling is required. Hippa Island is the most remote of the colonies examined here. Coastal or industrial influence is therefore unlikely due to its location. Thus, a lag period in observing decreases as a consequence of the regulations introduced may explain the observed concentrations. Increases in a variety of other Canadian Arctic wildlife (reviewed in Braune et al., 2005) have been observed, which supports the idea of a lag period due to distance from sources. Alternately, there may be some influence from the Alaskan gyre because ocean currents are a known transport route for many POPs (Schlosser et al., 1995; Wania, 2003).

High HBCDD concentrations in petrel eggs from Hippa Island compared to conspecifics at Cleland Island and auklet eggs may be related to differences in feeding ecology, migration, trophic level, food sources and/or contaminant variation (Lavoie et al., 2010). HBCDD is not manufactured in Canada but is imported incorporated into various products (Environment Canada and Health Canada, 2012). HBCDD is produced in the Netherlands, the USA, Japan and China (UNEP, 2010). High concentrations have been found in coastal waters of Japan, China and Europe (UNEP, 2010). Stable isotope analysis (SIA) in petrels indicated they feed the farthest offshore of the three species here, and SIA from Hippa Island petrels indicate they generally feed even more remotely than conspecifics at Cleland Island, possibly explaining these differences in concentration. However, the overall increasing concentrations of HBCDD observed in seabird eggs here may be a result of leaching from HBCDD-containing products in Canada, although leaching can also indicate long-range transport (De Wit, 2002), or due to exposure near production sites or contaminated areas during the non-breeding season e.g., the north–west Pacific coast near Asia. However, as no data exists on the movements and feeding grounds of these species outside of the breeding season, this suggestion is only an hypothesis.

4.2. Stable isotopes

Neither species transcended more than one trophic level, assuming a fractionation factor of 3.2‰–3.4‰ represents an average for multiple trophic transfers (Post, 2002). $\delta^{13}C$ values show all three species are offshore pelagic feeders (Hobson et al., 1994), although petrels feed more offshore compared to auklets and murrelets. Only lipid percentage and $\delta^{13}C$ were important variables predicting ΣPBDE and dominant congener concentrations, indicating there was no significant effect of trophic level ($\delta^{15}N$) after accounting for spatial variance ($\delta^{13}C$). Few significant relationships were observed between either $\delta^{13}C$ or $\delta^{15}N$ and dominant congeners or ΣPBDE. Likewise, ΣPBDE concentrations were independent of trophic level in nesting bald eagles (*Haliaeetus leucocephalus*) (Elliott et al., 2009). The lack of relationships and of significant temporal changes in stable isotope values indicate little change in auklet or petrel diet over time, hence dietary changes are unlikely to be the primary factor driving changes in contaminant concentrations in these species. Similarly, changes in contaminant concentrations could not be attributed to dietary changes in ivory gulls from the Canadian Arctic (Braune et al., 2007). However, other studies have found that dietary changes do affect contaminant concentrations in various bird species (Burgess et al., 2013; Eulaers et al., 2014a; Hebert and Weseloh, 2006; Hebert et al., 2000; Leat et al., 2011; Sørmo et al., 2013; Zhang et al., 2011).

4.3. Spatial trends

Differences between species were observed for HBCDD and between both species and site for ΣPBDE. As site did not have a significant effect on HBCDD, as per the GLM, one possible hypothesis is that HBCDD is primarily obtained in non-breeding grounds, assuming mixture of both colonies in winter. Murrelet eggs from Langara Island had the highest concentration of ΣPBDE when investigating the most recent sampling year. The absence of murrelet leg band recoveries from western North America, in contrast to auklets, whose bands are often recovered off western North America, hints at the possibility that murrelets winter off Asia where they are observed in large numbers (A.J. Gaston, *pers. comm.*). Non-breeding ground exposure may be a factor contributing to the higher levels of BFRs in murrelets.

The influence of seasonal movements, particularly latitudinal migration, on contaminant exposure in birds has been explored with varying results (Baert et al., 2013; Elliott and Shutt, 1993; Elliott et al., 2007; Henny et al., 1996; Yates et al., 2010). Most

studies have examined legacy POPs, especially DDT and its metabolites, and whether e.g., DDE acquired while wintering in tropical regions can subsequently affect reproduction back at the breeding colony. DDE is very slowly metabolized and cleared from the body, for example half-lives in herring gulls have been calculated to be greater than a year (Clark et al., 1987), increasing the likelihood of carryover from wintering exposure to deposition in eggs. Similarly, wintering area has been shown to have a significant effect on the concentration of PBDEs in plasma of great skua (*Stercorarius skua*) (Leat et al., 2013). Half-lives of BDE 47, 99, 100 and 153 in herring gulls were estimated to be approximately 100 days (Norstrom et al., 2002). A half-life for HBCDD in non-occupationally exposed humans weighing approximately 70 kg has been given as 64 days (Geyer et al., 2004), and for γ-HBCDD, 17 days in mice (Szabo et al., 2010). Thus, more recalcitrant BFRs accumulated from non-breeding ground exposure could still be present in the female lipid pool and be maternally deposited to eggs.

Regardless, at Cleland Island inter-specific differences are likely due to species differences e.g., differences in post-breeding movements and feeding grounds, rather than physical site differences. Nevertheless, all colonies investigated here are distant from urban influences and industrial activities, meaning spatial differences are unlikely to be a result of local anthropogenic activities.

Voluntary restrictions and subsequent regulations implemented to control PBDE congeners in North America appear to have been effective in reversing or slowing the increasing trends of these contaminants in the examined seabird eggs collected from colonies along the Pacific coast of British Columbia. Rapid increases of HBCDD are however, cause for some concern.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2014.08.009.

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