Influence of overwinter distribution on exposure to persistent organic pollutants (POPs) in seabirds, ancient murrelets (*Synthliboramphus antiquus*), breeding on the Pacific coast of Canada[☆]

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ABSTRACT

Assessing the fate of both legacy and newer persistent organic pollutants (POPs) is an ongoing challenge. Top predators, including seabirds, are effective monitors of POPs because they forage over a range of marine habitats, integrating signals over space and time. However, migration patterns can make unravelling contaminant sources, and potentially assessments of the effectiveness of regulations, challenging if chemicals are acquired at distant sites. In 2014, we fitted geolocators on ancient murrelets (*Synthliboramphus antiquus*) at four colonies on the Pacific Coast of Canada to obtain movement data throughout an annual cycle. All birds underwent a post-breeding moult in the Bering Sea. Around one-third then returned to overwinter on the British Columbia (BC) coast while the rest migrated to overwinter in waters along the north Asian coast. Such a stark difference in migration destination provided an opportunity to examine the influence of wintering location on contaminant signals. In summer 2015, we collected blood samples from returned geo-tagged birds and analyzed them for a suite of contaminants, including polybrominated diphenyl ethers (PBDEs), non-PBDE halogenated flame retardants, perfluoroalkyl substances (PFASs), organochlorines, and mercury. Feathers were also collected and analyzed for stable isotopes ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$). We found no significant differences in blood concentrations of any contaminant between murrelets from the two different overwinter areas, a result that indicates relatively rapid clearance of POPs accumulated during winter. Spatial variation in diet (i.e., $\delta^{13}C$) was associated with both BDE-47 and -99 concentrations. However, individual variation in trophic level had little influence on concentrations of any other examined contaminants. Thus, blood from these murrelets is a good indicator of recent, local contaminants, as most signals appear independent of overwintering location.

1. Introduction

The concept of the Anthropocene, whereby humans have become the dominant force shaping Earth's bio-geophysical composition and processes, includes impacts from production and release of an increasing array of synthetic chemicals (Rockström et al., 2009). Among the most problematic of these contaminants are commercial organic compounds, many of which are persistent, bioaccumulative, and toxic, presenting both an ecotoxicological and human health threat. A number of contaminant groups can be transported long distances via atmospheric and oceanic processes, and even transported and deposited via seabirds, causing contamination of ecosystems very distant from sources (Roosens et al., 2007; Choy et al., 2010; Shoji et al., 2019).

Ongoing monitoring of environmental contaminants is, therefore, essential to further understanding of the fate and consequences for biological systems of this pervasive contamination, and to advise regulators and industry and devise mitigation measures.

Several monitoring programs use seabirds as sentinel species because they integrate signals across space and time, including multiple environments from coastal to offshore, which are then brought back to a single place (e.g. Furness and Camphuysen, 1997; Elliott and Elliott, 2013; Miller et al., 2014a). Seabirds also meet many of the preferred criteria to qualify a species as suitable for contaminant monitoring, such as good knowledge of species biology and ecology (Moore, 1966; Furness and Camphuysen, 1997) and high trophic level (Moore, 1966; Coulson et al., 1972; Gilbertson et al., 1987). However, many seabird populations undertake complex movements throughout their annual cycle. Unless such migration patterns are known, identifying and measuring contaminants in an adult bird or their eggs may reveal little about where contaminant bioaccumulation occurred, because individuals were potentially exposed far from the site of sampling (Leat et al., 2013; Carr et al., 2017).

While overwintering location may influence environmental contamination recorded at breeding sites, other factors can also be involved. In particular, whereas capital breeders (species that mobilize reserves accumulated over the winter or during migration to breed) may have strong signals from overwinter grounds, income breeders (species that use current energy intake to breed) may more closely represent breeding ground signals (Stephens et al., 2009). For the latter species, reserves acquired from local resources would play an important role. Stable isotope analysis (SIA) is one tool for assessing the role of diet in contamination (e.g., Hoekstra et al., 2003; Braune, 2007; Bodin et al., 2008), and can improve understanding of species trophic relationships, dietary sources, and frequented habitats. In particular, $\delta^{15}N$ increases with trophic level, and serves as an index of trophic position (Jarman et al., 1997; Hop et al., 2002; Ruus et al., 2002, 2006). In contrast, $\delta^{13}C$ and $\delta^{34}S$ can provide indices of foraging habitat (Morrissey et al., 2010; Elliott et al., 2014; Miller et al., 2014b, 2015a, b; Elliott and Elliott, 2016).

Recent results of seabird monitoring data from the western Canadian coast have shown decreases for some contaminants but fluctuations for others (Miller et al., 2014b, 2015a, b). In some cases, it is unclear where contaminant uptake originates, and thus whether regulations and restrictions on various contaminants (e.g., the Canadian Environmental Protection Act) are having the desired outcome. Some persistent organic pollutants (POPs) continue to be produced in countries outside of North America, despite international regulations imposed on production, use, and distribution (e.g., the Stockholm Convention on Persistent Organic Pollutants [Stockholm Convention on Persistent Organic Pollutants, 2008], the Long Range Transboundary Air Pollution Protocol on POPs [Arnot et al., 2009]). Thus, knowledge about migration patterns is essential to determine if non-breeding season movements influence contaminant uptake and body burden (Burger and Gochfeld, 2004).

Ancient murrelets (*Synthliboramphus antiquus*) breed along the British Columbia coast and are a component of long-term monitoring schemes (Miller et al., 2014b, 2015a,b; Elliott and Elliott, 2016); in fact, they are the Pacific species with the earliest eggs deposited in the Canadian National Wildlife Specimen Bank (Elliott, 1985). However, they have a unique migration strategy whereby all individuals molt in the Bering Sea, but then almost two thirds of individuals winter off of Asia and the rest return to winter off of British Columbia (Gaston et al., 2015, 2017). This unusual east-west migration means that some individuals are exposed to contaminants from Asia (Shoji et al., 2019) and could explain variation in contaminant concentrations. For instance, Miller et al. (2014b,

2015b) argued that high levels of hexabromocyclododecane (HBCDD) and low levels of PBDEs (polybrominated diphenyl ethers) in ancient murrelet eggs, relative to other species, originated from individuals wintering off Asia. Here, we examined differences in concentrations of various environmental contaminants in plasma of ancient murrelets based on migration patterns. We also attempted to correlate values with stable isotopes ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{34}S$) in feathers, although feathers were grown during the non-breeding period in the Bering Sea, to elucidate if migration direction influences contaminant concentration.

2. Materials and methods

2.1. Species

The ancient murrelet (hereafter murrelet) is a colonial burrow-nesting bird (Wilbur, 1969; Wilson & Manuwal 1986) with breeding colonies found in remote parts of coastal British Columbia (Gaston et al., 2015, 2017). It is a sub-surface feeder that preys primarily on zooplankton and small, schooling fish (Sealy, 1975). Spatially intensive telemetry data recently collected from birds tagged on the breeding colonies showed that foraging was all in local Canadian coastal waters. Recent geolocator data shows this species is migratory, with movements across the entire North Pacific Ocean to Asia (Gaston et al., 2015). Adult feeding thus occurs across a vast expanse of North Pacific waters (Sealy, 1975). This species is primarily a capital breeder (Shoji et al., 2013). Each breeding pair lays two eggs per year (Wilbur, 1969; Gaston, 2010). Chicks are precocial and leave the nest shortly after hatching, following the adult from the burrow to the ocean (Sealy, 1975).

2.2. Sites and geolocator attachments

In total 150 murrelets at four remote offshore breeding colonies in Haida Gwaii, British Columbia (Frederick Island, George Island, Hippa Island, and Reef Island; Table 1, Fig. 1) had leg-mounted geolocators attached during the breeding season of 2014. Geolocators of 43 birds that had usable tracking data were retrieved the following year during the breeding season. Details of the geolocators used, deployment methods, and data retrieval can be found in Gaston et al. (2015, 2017), including permit numbers. All islands are located in open coastal positions, distant from large urban centres.

2.3. Analysis of tracking data

Details of tracking data analysis have been reported earlier (Gaston et al., 2017). Briefly, data were downloaded and processed using IntiProc v1.03. The loggers worked by calculating sun elevation angles to estimate sunrise and sunset based on light levels recorded when birds were at sea. Occasionally, unknown shading conditions (e.g., influenced by bird behaviour) impacted the light sensor. Some data were excluded because of similar day lengths during spring and fall equinoxes (e.g., most data between 22

Table 1
Coordinates for ancient murrelet sampling sites; number of geolocators deployed and recovered at each site (from Gaston et al., 2017).

Site	Coordinates	Geolocators deployed, 2014	Geolocators recovered, 2015
Frederick Island	53°93′N; 133°17′W	50	20
Hippa Island	53°26′N 132°59′W	25	4
George Island	52°33′N; 131°20′W	50	18
Reef Island	52°86′N; 131°50′W	25	7
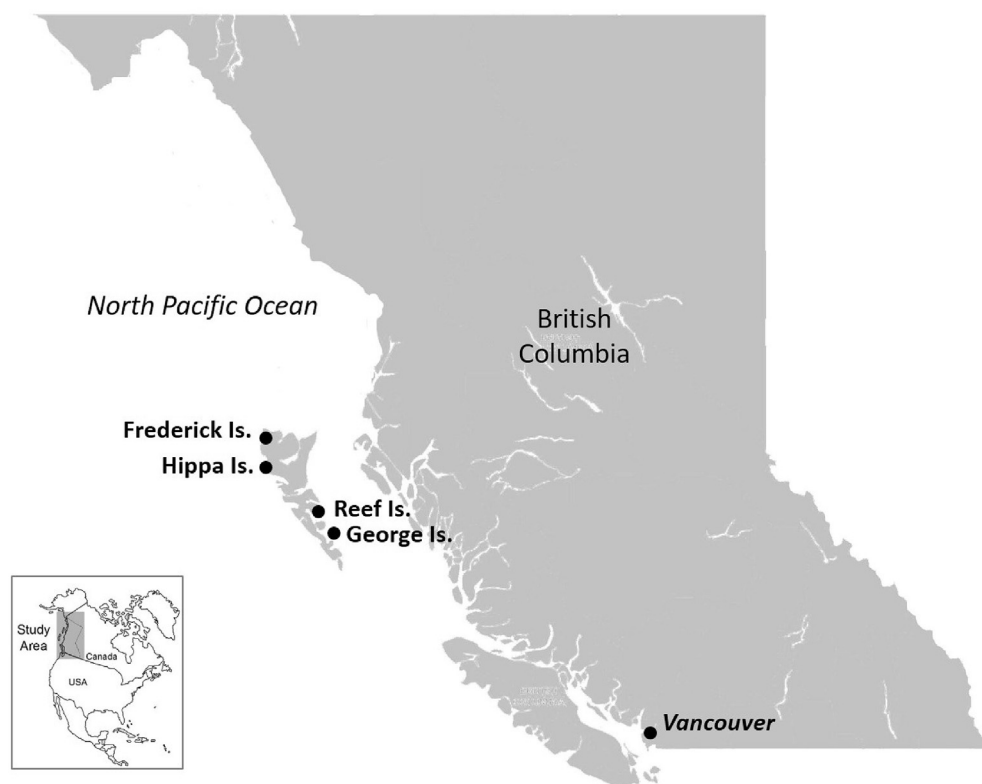
Fig. 1. Ancient murrelet geolocator placement and blood sampling sites, coastal British Columbia, Canada. Inset: North America. The shaded grey area represents the sampled area.

September - October 27, 2014 and 23 February - March 30, 2015 when latitudinal variance was high).

2.4. Sampling

In homeotherms, blood levels of slowly metabolized lipid-soluble POPs rapidly equilibrate with concentrations in the brain and other lipid-rich tissues due to high perfusion rates. Thus, blood is a suitable matrix for examining overall body burden of such POPs (Soine et al., 1982, Brown and Lawton 1984; Elliott and Norstrom, 1998; Elliott et al., 1998; Bustnes et al., 2001). Whole blood samples from both male and female adult murrelets that had geolocators attached in 2014, and returned to the breeding colonies in 2015, were collected from the four colonies (Fig. 1). Those birds were re-captured from their burrows while they were incubating eggs, geolocators were removed, and the birds were then weighed. Six breast feathers were hand plucked from the breast of each bird, and a small volume of blood (approximately 1 mL) was collected from the brachial vein using a 27-1/2" gauge needle and a 3 mL syringe, and stored in a vacutainer with sodium heparin, as per methods in Gaston et al. (2017). Two small circles of whole blood were dotted on filter paper (~10 μL blood/circle) (Protein Saver Snap Apart, Whatcom 903, GE Healthcare), air-dried and stored at room temperature. A small amount of formalin (ratio 1:20, formalin:blood) was added to the remaining blood to preserve the sample. Filter paper samples were used to determine sex from DNA using a PCR method (Fridolfson and Ellegren, 1999) at the National Wildlife Research Centre, Environment and Climate Change Canada, Ottawa, ON.

2.5. Stable isotope analysis

Because of the small volume of blood available, stable isotope

analysis (SIA) was carried out using feathers. SIA was conducted at the G.G. Hatch Stable Isotope Laboratory, Ottawa University (https://isotope.uottawa.ca/). Briefly, for samples analyzed for $\delta^{13}C$ and $\delta^{15}N$ isotopes, 1 mg sub-samples were freeze-dried, loaded into tin cups and analyzed using a Vario EL Cube elemental analyzer (Elementar, Germany) interfaced to an isotope ratio mass spectrometer (Delta Advantage manufactured by Thermo, Germany). For $\delta^{34}S$ isotopes, sub-samples were analyzed using an Isotope Cube (Elementar, Germany) elemental analyzer interfaced to a Delta Plus XP isotope ratio mass spectrometer (ThermoFinnigan, Germany).

During analysis, samples were interspersed with several replicates of laboratory standards (C-51 Nicotiamide [0.07,-22.95]; C-52 mix of ammonium sulphate and sucrose [16.58,-11.94]; C-54 caffeine [-16.61,-34.46]; and a blind standard of C-55: glutamic acid [-3.98, −28.53]). The final delta values are reported in parts per thousand (‰) relative to international standards Vienna PeeDee Belemnite and Vienna Cañon Diablo Troilite ($\delta^{13}C$) and air ($\delta^{15}N$), respectively. There was not enough $\delta^{34}S$ data to monitor results for in-house quality control materials. To check for homogeneity of samples, one random sample was analyzed in duplicate for every ten samples.

2.6. Chemical analyses and quality assurance

Whole blood samples were analyzed for perfluoroalkyl substances (PFAS) at NWRC, Ottawa, Canada using an ultra-high performance liquid chromatography tandem mass spectrometry (UPLC-MS/MS) system. For polybrominated diphenyl ethers (PBDE) and non-PBDE halogenated flame retardants, twelve samples were analyzed at the Université du Québec à Montréal (UQAM; Montreal, QC, Canada) and eight samples were analyzed at the Great Lakes

Institute for Environmental Research (GLIER) at the University of Windsor, Windsor, ON, Canada. Plasma samples for organochlorines (OC) and polychlorinated biphenyls (PCB) were analyzed at the GLIER laboratory, while total mercury (THg) in murrelet plasma was analyzed in an accredited laboratory for metals analyses at the National Wildlife Research Centre (NWRC), Environment and Climate Change Canada. See E-supplement M1 for further details for all chemical analyses.

Lipid content of whole blood was determined colorimetrically using the sulfo-phospho-vanillin method as reported earlier (Frings et al., 1972; McKinney et al., 2006).

2.7. Statistical analyses

All organic contaminants data were log-transformed prior to statistical analyses, in order to obtain normal distributions. For the calculation of summed pollutant values (i.e., ΣPBDE), non-detects were treated as 0, while values less than the method limit of detection (<MLOD) or less than the method limit of quantification (<MLOQ) were assigned half the limit of detection (LOD) or limit of quantification (LOQ, calculated with a signal to noise ratio of 10:1) respectively. Where more than 50% of reported values were below MLOQ or MLOD, or were reported as non-detects, the congeners were removed from analyses (subsequent to calculation of summed pollutant values).

Where sample size allowed (i.e., for PBDEs) we conducted generalized linear models (GLM) (R Studio version 3.5.1). Contaminants (log-transformed wet weight basis) were the dependent variables, while migration (overwintering location, Asia or North America), site (breeding colony), sex, and stable isotopes (i.e., δ^{14}C, δ^{15}N, and δ^{34}S values for each bird) were the independent variables. We performed F tests followed by the suitable two-sample *t*-test (unpaired; equal, or unequal variances) using log transformed data for non-BDE HFRs and PFAS, to compare concentrations based on general migration direction. Pearson's correlation coefficient was used to examine the relationship between trophic position (δ^{15}N) and log transformed dominant or sum congeners (PBDEs, non-BDE HFRs, PFAS; Excel 2013). No statistical analyses were conducted for Hg, OCs, and PCBs due to the small number of birds analyzed or the small number of birds migrating to North America.

Geolocator data provided dates and locations for each bird during its migration. Birds that migrated in a similar direction were amalgamated, resulting in two groups: birds that wintered off Asia, and birds that wintered off North America (specifically southern British Columbia and California). These migration directions (i.e., Asia, North America) were used for the GLMs. The level of significance for any traditional statistical tests was set a $p < 0.05$.

3. Results

3.1. Migration

Migration data were available for 43 murrelets (Fig. 2). Just over two-third of the birds with usable migration data migrated across the North Pacific Ocean (n = 24), many going via the Bering Sea, the Sea of Okhotsk/Kamchatka Peninsula/Kuril Islands, and south to waters off of the east coast of Japan. A few individuals ventured into the East China Sea/Yellow Sea. Birds migrating in that direction were grouped as migrating to Asia. Those birds then typically returned to Haida Gwaii and their breeding colonies directly across the North Pacific, following the edge of sea ice en route. The rest of the birds (n = 20) migrated to various parts of the North Pacific Ocean in different directions e.g., the Chukchi Sea, Beaufort Sea, Aleutian Islands (Fig. 2) before returning to overwinter in North America, ranging from Southern California north to their breeding

colonies. These birds were grouped as migrating to North America.

3.2. Chemical analyses

Due to limited plasma volumes, we opted to stratify samples for each contaminant group with the goal of having a target sample size of six birds for two groupings: six birds for North America, and six that migrated to Asia. Thus, not all birds were analyzed for all contaminants (Table 2).

3.3. PFASs

After trace congeners were removed, seven PFAS congeners remained: PFHxA, PFNA, PFDA, PFUdA, PFDoA, PFTrDA, and PFOS. Of these, PFUdA, a PFCA compound, was most dominant, typically comprising between 30% and 50% of the summed PFAS (ΣPFAS), followed by PFOS, a PFSA compound, comprising between approximately 20%−40% of ΣPFAS. Of the 12 birds sampled for PFASs, 11 had corresponding migration and stable isotope data (see Table 1 in E supplement R1).

No significant difference was observed in PFUdA or PFOS concentrations, or in ΣPFCA or ΣPFSA between birds migrating to Asia or migrating to North America (*t*-test: df = 10, p > 0.05; Fig. 3). Pearson's correlation coefficient showed a moderate but non-significant positive correlation between trophic level (δ^{15}N) and PFUdA (r = 0.58, p = 0.06), and a positive but non-significant correlation with PFOS (0.31, p = 0.35).

3.4. PBDEs and non-PBDE HFRs

A total of 20 birds had HFR and migration direction data available. Stable isotope data (δ^{13}C, δ^{15}N, and δ^{34}S) was available for 17 of these birds. Dominant PBDE congeners were: BDE-47>BDE-99>BDE-49, which were all penta-BDEs. No octa- or deca-BDE congeners were among the dominant congeners. Of the non-PBDE HFRs analyzed (n = 12), only pentabromoethylbenzene (PBEB) and hexabromobenzene (HBB) remained after trace congeners were removed (concentration range 0.005−0.07 ng g-1 ww, and 0.005−0.057 ng g-1 ww, respectively; see Table 2 in E supplement R1).

No significant associations were observed between any of the dominant PBDE congeners or ΣPBDE and the independent variables. Akaike Information Criterion (AIC) scores were moderate for all models (between 35 and 45). We observed no significant difference in PBEB or HBB concentrations between birds that migrated to Asia and those that migrated to North America (*t*-test: p = 0.7, df = 5; p = 0.09, df = 10, Fig. 4).

3.5. Organochlorines, PCBs, and mercury

Organochlorine pesticides remaining after trace congeners were removed included: 1,2,3,4-tetrachlorobenzene, pentachlorobenzene, α-hexachlorohexane, hexachlorobenzene, oxychlordane, p,p-DDE, and mirex. These were dominated by p,p-DDE, which had considerably higher concentrations compared to the other OCs. Five birds analyzed for OCs had migration data available (see table 3 in E supplement R1). Given the small number of individual birds migrating to Asia or North America, it is difficult to make any inferences related to migration direction. Nonetheless, there appeared to be little variation between birds that migrated in these two directions (e.g., 1,2,3,4-tetrachlorobenzene and pentachlorobenzene; Fig. 5).

After trace congeners were removed, 13 PCB congeners remained, including: co-eluted PCB-28/31, PCB-52, PCB-99, PCB-101, PCB-105, PCB-118, PCB-138, PCB-149, PCB-153, PCB-180, PCB-
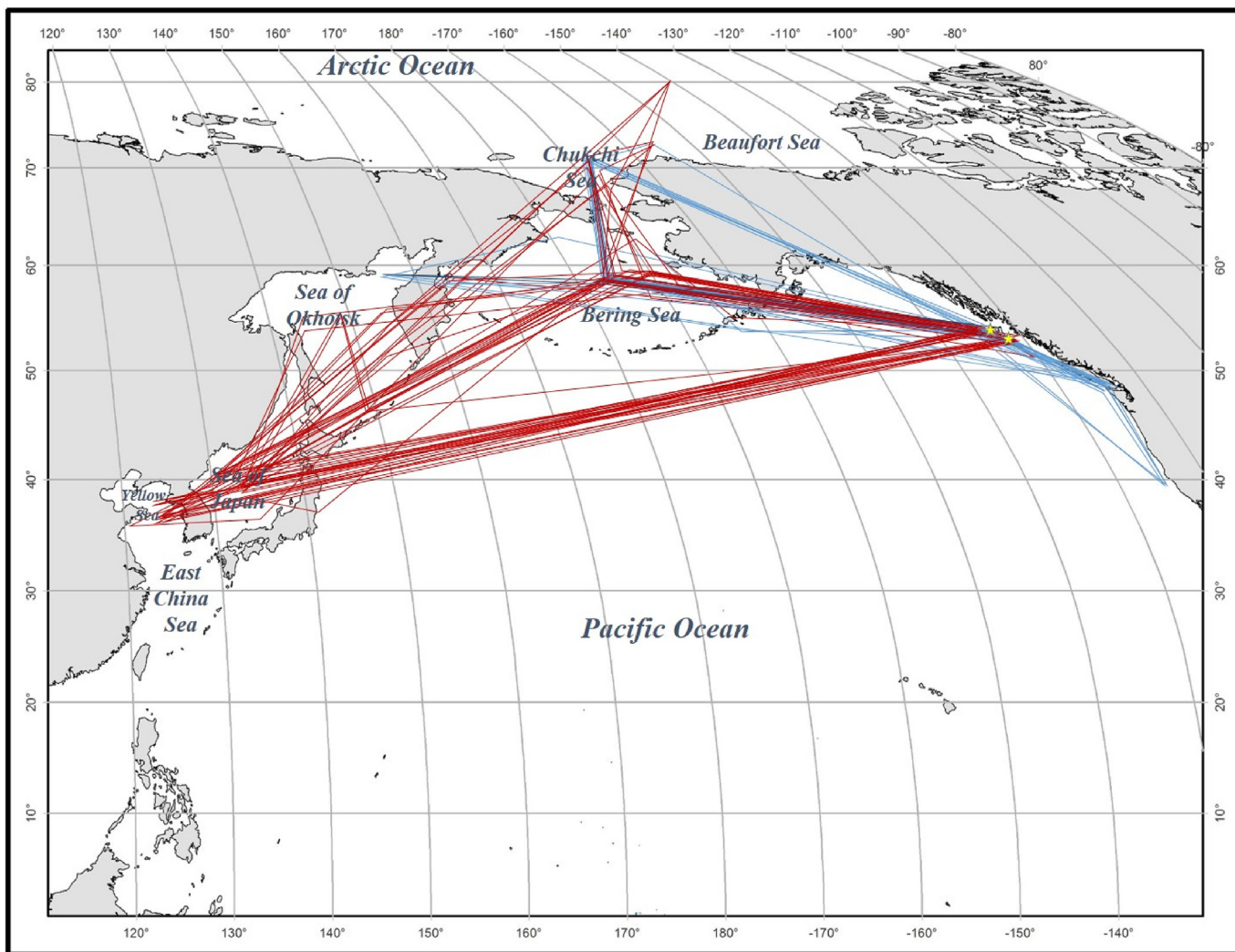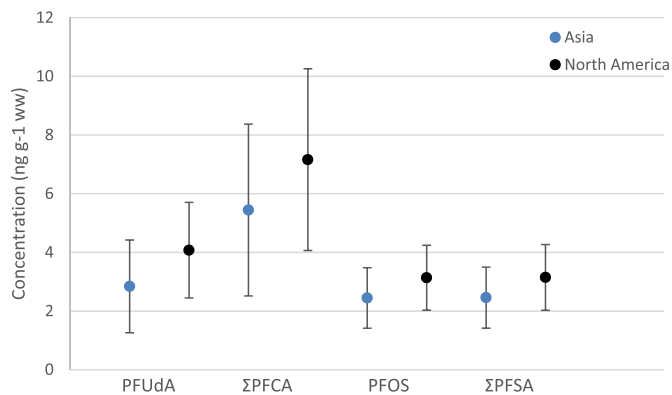
Fig. 2. Generalized migration direction, as determined from geolocator data, of ancient murrelets breeding at four colonies on coastal British Columbia, Canada. Yellow stars depict breeding colonies. Red lines show the general direction of murrelets that migrate via Asia. Blue lines depict the general movement pattern of murrelets that migrate via western North America. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 2

Total number of plasma samples of ancient murrelets analyzed for each chemical group based on migration direction.

Analyses	Migration Direction		Total
	Asia	North America	
PBDEs	15	5	20
Non-PBDE HFRs	9	3	12
PFASs	6	6	12
OC pesticides	3	2	8
PCBs	3	2	8
Total Mercury	10	2	12

Fig. 3. Selected perfluorinated compounds, PFUdA, ΣPFCA, PFOS, and ΣPFSA geometric means (ng g-1 ww) ± 95% confidence intervals, in ancient murrelets by migration direction.

187, PCB-194, and PCB-209. Dominant congeners amongst these were PCB-153>PCB-138>PCB-180>PCB-118 (see table 3 in E supplement R1). Migration data were available for three birds that migrated to Asia and two that migrated to North America.
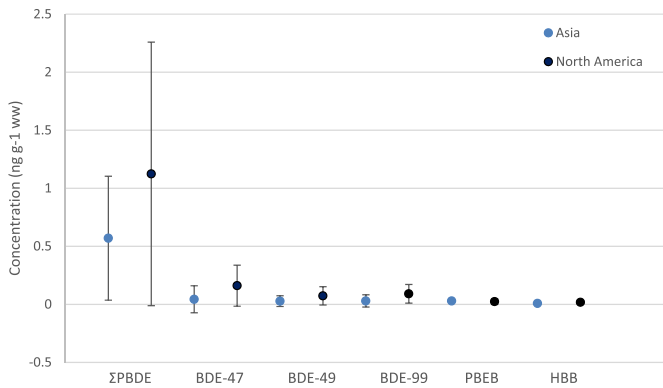
A total of 12 birds had total Hg and migration data available. Ten birds migrated to Asia and two migrated to North America (see table 4 in E supplement R1). Two individuals that migrated to Asia showed particularly high total Hg concentrations compared to all other birds.

4. Discussion

As reported previously (Gaston et al., 2015, 2017), almost two-

Fig. 4. Selected flame retardants, ΣPBDE, BDE-47, -49 and −99, and PBEB and HBB geometric means (ng g-1 ww) ± 95% confidence intervals, in ancient murrelets by migration direction.
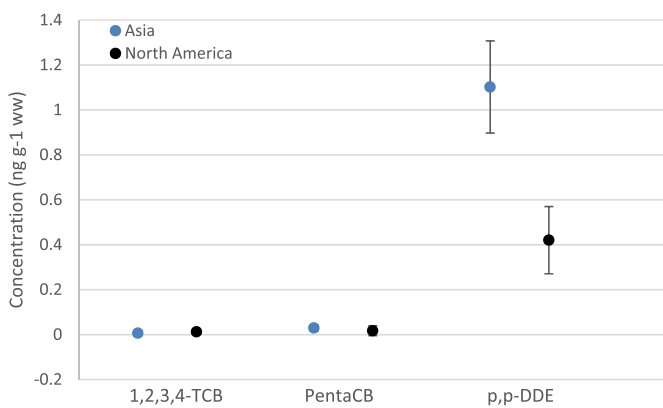
Fig. 5. Selected chlorinated hydrocarbons, 1,2,3,4-TCB (tetrachlorobenzene), pentaCB (pentachlorobenzene) and *p,p*'-DDE geometric means (ng g-1 ww) ± 95% confidence intervals, in ancient murrelets by migration direction.

thirds of the birds sampled here migrated to waters off Asia, while the rest moved into the North Pacific Ocean, before all birds returned to their respective breeding colonies along the Pacific coast of Canada. Despite their relatively small size (average weight for both adult males and females is approximately 205 g; Sealy, 1975), murrelets are capable of migrating across the entire width of the North Pacific Ocean during the non-breeding season. Prior to this study, a lack of knowledge about murrelet migration patterns meant that investigating associations between contaminant concentrations and non-breeding season movements was, at best, difficult (Miller et al. 2014a,b; 2015a,b).

Asian countries on the Pacific Rim are increasingly important producers of synthetic chemicals (C&EN, 2019), including manufacturing sites of precursors of specific compounds, such as PFUdA (Buck et al., 2011). Regardless of the potential greater exposure on the Asian Pacific coast, there were no significant differences in blood concentrations of PFASs, PBDEs, and non-PBDE HFRs between birds that migrated to Asia and those that wintered on the North American coast. Inference of trophic levels from δ^{15}N values provided no insight into accumulation of targeted contaminants. However, sample sizes were low, which in combination with a high degree of variability in exposure, limited statistical power. For Hg, OCs, and PCBs, no comparisons were possible due to the limited number of birds with both contaminant, stable isotope, and migration data available—although some OCs, such as *p,p*'-DDE, appeared lower in birds wintering off North America.

Results of other studies examining environmental contaminants

in migratory and resident birds using blood samples and/or geo-locator data have been contradictory, which may be due to variability in species examined, methods used, or even contaminants examined. For example, some studies showed feeding while at wintering locations affected contaminant concentrations measured in spring (Henny et al., 1982; Leat et al., 2013; Fort et al., 2014), while others did not (Elliott and Shutt, 1993; Henny et al., 1996; Elliott et al., 2007; Maldonado et al., 2017; Hill, 2018).

The low sample sizes meant we only detected relatively weak associations between wintering areas and contaminant concentrations. In a previous study using a study design of five pools of three eggs, we determined significant differences among species, including ancient murrelets, for many of the same compounds tested here (Miller et al. 2014a,b; 2015a,b). That egg monitoring study design does reduce variability and increase statistical power over a design of N = 5 or 6 individuals (Gilbertson et al., 1987). Numerous other causes of variation can influence contaminant levels, making comparison difficult. For example, blood typically has a low lipid content compared to a more lipid-rich matrix e.g., egg, which will influence the relative concentrations of lipophilic compounds such as PBDEs and non-PBDE HFRs, and potentially increase within sample variance. Additionally, auks may be able to switch from capital to income breeding (Shoji et al., 2019), which would affect contaminant concentrations.

4.1. PFASs

No difference in PFAS concentrations were determined between birds that migrated to Asia or North America. The dominant PFASs were PFUdA, a long-chain, odd-numbered PFCA compound, (C_{11}) and PFOS (C_8), a compound that was not only phased out in 2000 by 3 M, a major global producer (US EPA, 2000), but was also restricted in various jurisdictions in the US (USEPA, 2006), by Canadian Environmental and Health Authorities (Environment Canada, 2010), and the European Union (Directive, 2006/122/EC, 2006). Since 2009, PFOS is one of the nine new persistent organic pollutants listed under the Stockholm Convention. Both PFUdA and PFOS have been noted as dominant compounds in other marine organisms e.g., Canadian Arctic seabirds (Braune and Letcher, 2013); seabirds on the Pacific coast of Canada (Miller et al., 2015b); beluga whales (*Delphinapterus leucas*) in Alaska (Reiner et al., 2011).

PFUdA increased significantly in eggs of thick-billed murres (*Uria lomvia*) and northern fulmars (*Fulmarus glacialis*) from Prince Leopold Island in the Canadian Arctic between 1975 and 2011 (Braune and Letcher, 2013). Similarly, the total contribution of PFUdA to PFASs was observed to be increasing between 1990 and 2011 in rhinoceros auklets (*Cerorhinca monocerata*) and Leach's storm-petrels (*Oceanodroma leucorhoa*) breeding on the coast of BC (Miller et al., 2015b). Storm petrels migrate up to thousands of kilometers beyond the continental shelf (Pollet et al., 2014), and it is likely that Pacific coast birds move very far west, likely close to coastal Asia. In 2003, China began large-scale production of PFOS, with many producers located in coastal China, potentially offsetting any progress made by the restriction in PFOS use and production in other countries (Chen et al., 2009; Han, 2009; Lim et al., 2011).

4.2. PBDEs and non-PBDE HFRs

Dominant PBDE congeners were all penta-BDEs that have a longer-range transport potential compared to higher-brominated congeners (Wania and Dugani, 2003). BDE-47 and -49 were significantly associated with site, in this case, Reef Island, located on the east coast of Haida Gwaii i.e., between Haida Gwaii and the west coast of mainland British Columbia, in the Queen Charlotte Sound.

BDE-47 and -99 were significantly associated with $\delta^{13}C$, while $\delta^{15}N$ showed limited association with those contaminants. Additionally, migration direction had little influence on PBDE congener concentrations. BDE-47 and -99, as well as BDE-100, are the most common congeners found in fish, while PBDEs with a higher degree of bromination are more typically associated with species feeding predominantly in the terrestrial environment. Miller et al. (2014b) observed a similar pattern of dominant congeners (BDE-47, -99, and −100) in rhinoceros auklet, Leach's storm-petrel, and ancient murrelet eggs sampled from remote breeding locations along the BC coast between 1990 and 2011. Similar to present results, spatial variation in diet ($\delta^{13}C$) was found to be an important variable predicting ΣPBDEs, but trophic level ($\delta^{15}N$) was not.

In light of regulations and restrictions imposed on PBDE use and production, use of other HFRs, including PBEB and HBB, has increased (McGrath et al., 2017). Here, only PBEB and HBB were detected above MLOD levels in murrelet plasma. Migration direction did not influence PBEB or HBB concentrations, though there was some tendency for HBB concentrations to be lower in birds that migrated to Asia compared to North America. HBB is used in a range of materials, such as hard plastics and textiles, to meet flammability standards and was reported as being widely deployed in Japan and produced in China (Covaci et al., 2011). However, as noted earlier, many factors can influence concentrations, including breeding strategy and pharmacokinetic factors. The half-life of HBB in male Wistar rats (whole body) was 48 days (Yamaguchi et al., 1986). Half-lives were shorter in kidney (5.2 days) and adipose tissue (7.3 days) in rats administered HBB via parenteral injection (Ogino, 1984 in EPA 2009). Thus, murrelets may have cleared most of the HBB accumulated during overwintering in Asia, before blood samples were collected.

4.3. Organochlorines, PCBs, and mercury

Only seven organochlorine pesticides remained after trace compounds were removed. These were dominated by *p,p'*-DDE, the very persistent, although not the most acutely toxic, metabolite of DDT. Nonetheless, DDE is an endocrine disruptor (Wong et al., 2015). It also affects egg shell quality in some species of birds, particularly raptorial and fish-eating species, which led to declines of some marine populations, including brown pelicans (*Pelecanus occidentalis*) and double-crested cormorants (*Phalacrocorax auritus*) on the California coast, and northern gannets (*Sula bassana*) on the Atlantic coast of Canada (Anderson et al., 1975; Elliott et al., 1998; Blus, 2011). In 2001, DDT was banned for agricultural use as one of the original 12 POPs under the Stockholm Convention, but may still be used as a pesticide in parts of the world (Tsygankov et al., 2017). A Decision Statement released by the Conference of the Parties to the Stockholm Convention (2013) notes that continued use of DDT is sanctioned for control of malaria vectors. Additionally, dicofol, a miticide synthesized from technical DDT and which contains DDT as an impurity, is in use in some Asian countries such as China and India (Qiu and Zhu, 2010; Sharma et al., 2014). There are also many hotspots of DDT contamination from past manufacturing and intensive use, including on the Pacific coast within the annual range of these murrelets (Glaser and Connolly, 2002).

PCBs are complex mixtures of congeners and can cause both arylhydrocarbon (Ah)-receptor mediated toxicity and endocrine disruption (Harris and Elliott, 2011). In murrelets, the dominant congeners were PCB-153>PCB-138>PCB-180>PCB-118. All four are listed as mandatory contaminants that should be analyzed within both the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR; https://www.ospar.org/) and the Baltic Marine Environment Protection Commission (HELCOM; http://www.helcom.fi/) conventions. Of the four, PCB-118 is a mono-*ortho* with some weak affinity for the Ah-receptor, including in birds (McKinney et al., 1985; Harris and Elliott, 2011; Nacci et al., 2016 and references therein). PCB-153 has been correlated with circulating thyroid hormones in bald eagles collected at sites along the Pacific coast (Cesh et al., 2011). In Canada, due to the highly persistent and toxic nature of PCBs, the Canadian Environmental Protection Act (CEPA 1999) has set specific dates for the destruction of PCBs in storage and in service (Environment Canada, 2017).

Atmospheric transport of mercury to Japan from the Asian continent has been blamed for spikes in gaseous elemental mercury in the Kyushu Islands, western Japan (Marumoto et al., 2015), which may impact total mercury concentrations. Total Hg concentrations from murrelets do not appear unusually high. However, due to limited sample size, no comparisons could be made between total Hg concentration, and thus inferences about the influence of migration pattern cannot be made.

5. Conclusions

Due to difficulties in tracking migration, few studies have systematically examined seasonal movements in sentinel species, which can confound information regarding the location of contaminant accumulation. However, due to recent technological advances, such as satellite trackers and geolocators, studies can now examine migration jointly with contaminant exposure. Here, using geolocators coupled with blood samples of tracked birds, we were able to examine both migration pattern and contaminant concentrations in murrelets breeding along the BC coastline. None of the contaminants exhibited a clear influence of non-breeding season movement or trophic level, thus possible increases or decreases during the non-breeding period may be lost due to clearance of the chemicals by the following spring.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.113842.

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