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BIOMONITORING OF CONTAMINANTS IN BIRDS FROM TWO TROPHIC LEVELS IN THE NORTH PACIFIC

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Abstract—The presence and accumulation of persistent contaminants at high latitudes from long-range transport is an important environmental issue. Atmospheric transport has been identified as the source of pollutants in several arctic ecosystems and has the potential to severely impact high-latitude populations. Elevated levels of contaminants in Aleutian Island avifauna have been documented, but the great distance from potential industrial sources and the region's complex military history have confounded identification of contaminant origins. We sampled bird species across the natural longitudinal transect of the Aleutian Archipelago to test three contaminant source hypotheses. We detected patterns in some polychlorinated biphenyl congeners and mercury that indicate abandoned military installations as likely local point sources. Carbon isotopes were distinct among island groups, enabling us to rule out transfer through migratory prey species as a contaminant source. The long-range transport hypothesis was supported by significant west-to-east declines in contaminant concentrations for most detected organochlorines and some trace metals. Although relatively low at present, concentrations may increase in Aleutian fauna as Asian industrialization increases and emitted contaminants are atmospherically transported into the region, necessitating continued monitoring in this unique ecosystem.

Keywords—Atmospheric transport Aleutian Islands Contaminants Birds Stable isotopes

INTRODUCTION

The North Pacific Ocean and Bering Sea host large numbers of seabirds and some of the world's most productive fisheries, making this an important region of high-latitude biodiversity and productivity. The Aleutian Archipelago defines the border between these two water bodies and provides nesting sites for some of the largest seabird colonies in North America and for many endemic bird populations [1]. Although remote and largely uninhabited by humans, the Aleutian Islands have been impacted by many anthropogenic activities, such as the harvest of natural resources [2], introduction of exotic species, military activities [3], and most recently, introduction of contaminants [4,5]. Aleutian birds are especially vulnerable to anthropogenic influences because of their restricted ranges, and in the case of some endemics, small population sizes [1]. Thus, these birds also can serve as important biomonitors in this biologically and economically important region.

Elevated concentrations of contaminants can cause acute and chronic health effects. Chronic effects may include impairment of reproduction, behavior, and neurological function, and suppression of immune function [6]. The most toxic organochlorines (OCs) are lipophilic compounds that accumulate in fatty tissues [7,8]. Year-round residents in arctic environments depend on fat reserves as a buffer against cold, stress, and periods of low food availability [7,9], a characteristic that may contribute to elevated contaminant concentrations in these organisms. Metabolism of fat stores may have a negative impact by releasing stored OCs into the blood or increasing the concentration in remaining fat to toxic levels [6]. Thus, the storage and metabolic dependence upon fat may make many nonmigratory Aleutian birds especially vulnerable, even when contaminants are present at relatively low levels in the environment [7].

Contaminants have been implicated in low productivity in nonmigratory bald eagles (*Haliaeetus leucocephalus*) on several Aleutian Islands [4,5,10]. Although the origin of contamination in the Aleutian Archipelago remains undetermined, three hypotheses exist regarding possible contaminant origins in this region: point sources, global transport, and migratory prey. Past military sites have been linked to high concentrations of polychlorinated biphenyls (PCBs) because of the improper disposal of electrical equipment [9] and are considered a potential source for contamination in the Aleutian Islands [4,10]. Bald eagle eggs from islands that were occupied by military installations had the highest PCB concentrations, a factor used to advance the hypothesis that historic military activities are point sources of Aleutian Island pollution [4,5].

Atmospheric and oceanic pathways play a large role in the global distribution of contaminants and have been implicated as a major source of persistent organic pollutants and mercury in Pacific [11,12] and arctic ecosystems [13–16]. Contaminants are transported in the atmosphere from sources in warm regions to colder climates, where they condense and precipitate into the ecosystem [16]. The Aleutian low pressure that dominates weather over the North Pacific and Bering Sea during winter months draws both storms and airborne contaminants from southeast Asia eastward along the Aleutian Archipelago [17,18]. A west-to-east decrease in contaminant concentrations along the Aleutian Archipelago is expected in atmospherically transported contaminants considering the prevailing weather patterns and the fact that atmospheric concentrations tend to decrease with distance from the source [13]. This hypothesis was forwarded to explain the high OC and dichlorodiphenyldichloroethylene (DDE) concentrations found in bald eagle eggs from western Aleutian Islands [4,5].

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Pollutants also may be introduced into the Aleutian food web through migratory piscine and avian prey species. Many Aleutian breeding birds winter in coastal areas in Washington, Oregon, and California (USA) [19], where contaminants have been documented in marine food webs [20], and contaminants may be transferred to predatory birds that prey upon them [6].

The Aleutian Islands provide a natural experimental longitudinal transect across the North Pacific to test hypotheses of contaminant sources. The objective of our study was to quantify organic and trace metal contaminant concentrations in birds from two Aleutian food webs to test the three contaminant source hypotheses forwarded by Anthony et al. [4]. We used contaminant concentrations in nonmigratory birds from two food webs to assess the impact of point sources and long-range transport at five islands along the Aleutian Archipelago. Stable isotopes were used to examine the possible confounding factor of migratory prey species in the top trophic feeders studied. We hypothesized that the regionally distinct isotopic values in Aleutian plankton [21] would be detected in predators feeding in local food webs, because isotopic values are transferred up the food chain in a predictable, step-wise manner [22].

MATERIALS AND METHODS

Focal species

Pelagic cormorants (*Phalacrocorax pelagicus*) and red-faced cormorants (*Phalacrocorax urile*) are year-round, nonmigratory residents in the Aleutian Islands, where they often co-occur in breeding colonies and foraging flocks [23,24]. Both species are exclusively marine, but they prefer inshore and coastal habitats where they feed mainly on benthic, solitary fish [23,24]. Both cormorant species were used to assess contaminant loads in top predators of coastal Aleutian Island food webs. Collection efforts focused at the generic level (*Phalacrocorax* spp.) because of the ecological similarity of the two species. Subsequent statistical analyses detected no differences in contaminant concentrations between the two species, confirming our decision to pool the two as ecologically similar (see below).

The Aleutian rock sandpiper (*Calidris ptilocnemis*) also is a nonmigratory resident in the Aleutian Islands [25], and this species was used to assess contaminant concentrations at lower trophic levels. Rock sandpipers feed on inshore benthic invertebrates, except during breeding, when birds forage predominantly on their tundra territories for terrestrial invertebrates [25]. Specimens were collected during the breeding season only from inshore habitats.

Study area

The Aleutian Archipelago includes more than 200 islands, extends more than 3,500 km from the Alaska mainland, and defines the border between the North Pacific Ocean and the Bering Sea. The study area consisted of five islands in this archipelago, representing a west-to-east geographical gradient (Attu, Kiska, Adak, Amlia, and Amak/Alaska Peninsula, USA; Fig. 1). Logistical constraints precluded the collection of rock sandpipers on Kiska Island.

Sample collection and preparation

Pelagic and red-faced cormorants ($n = 76$) were collected with steel shot, and rock sandpipers ($n = 38$) were collected with lead shot in July to August 2000 and May to June 2001. Each specimen was weighed, tagged, and frozen until preparation. In the laboratory, birds were thawed and morphological measurements (wing chord, tarsus, tail, bill, and skull lengths; bill width; and bill height) were taken to the nearest 0.1 mm. After harvesting tissue samples for chemical and isotopic analyses (described below), all specimens were preserved and archived at the University of Alaska Museum, Fairbanks (AK, USA) as skins, skeletons, and tissue and stomach samples (after Winker [26]).

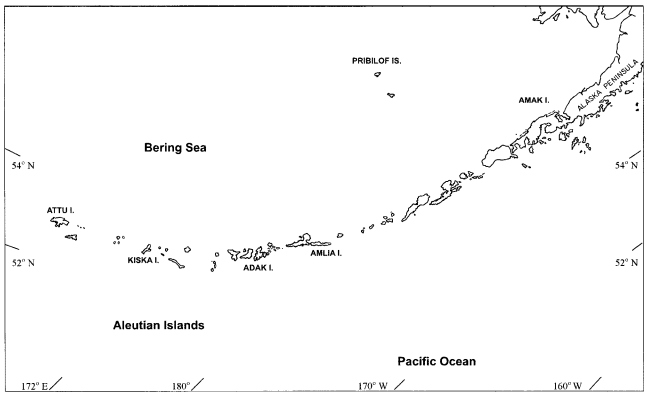

Fig. 1. Sampled sites in the Aleutian Archipelago, Alaska, USA.

Contaminant analyses

Liver and breast muscle samples were removed from specimens by using chemically clean instruments (hexane–acetone wash) and new, sterile, stainless steel scalpel blades were replaced after each sample was removed. Because of the small body size of rock sandpipers, liver and muscle tissues from two individuals from the same site were randomly combined to obtain adequate sample mass of each tissue for analyses. Samples were placed in separate, chemically clean, glass jars (I-CHEM, Rochester, NY, USA) and frozen at −20°C until shipped to the analytical laboratory. Environmental Research Institute (Storrs, CT, USA) performed the elemental and OC analyses.

Elemental analyses

Samples were analyzed for nine different metals. Arsenic, cadmium, chromium, copper, nickel, lead, selenium, and zinc were analyzed via inductively coupled plasma–mass spectrometry by using a modified National Oceanic and Atmospheric Administration ORCA 130 method [27]. The method detection limits for these techniques vary by target analyte, but ranged from 0.03 to 0.5 mg/kg dry weight. Tissues were analyzed for total mercury by cold vapor atomic absorption spectroscopy by Environmental Protection Agency Method 245.6. The method detection limit for this method was 0.2 μg/kg. Percent moisture was calculated as the percent difference between the wet (initial) weight and the dry (final) weight of a homogenized subsample that was dried for at least 16 h at 105°C.

Organochlorine analyses

Samples were analyzed for 28 chlorinated organic compounds (aldrin; alpha-, beta-, delta-, and gamma-benzene hexachloride; alpha- and gamma-chlordanes; *cis*- and *trans*-nonachlor; dieldrin; endosulfan I and II; endosulfan-sulfate; endrin, endrin aldehyde, and ketone; heptachlor; heptachlorepoxide; hexachlorobenzene; methoxychlor; mirex; oxychlordane; *o,p′*-dichlorodiphenyldichloroethane [DDD]; *o,p′*-dichlorodiphenyldichloroethylene [DDE]; *o,p′*-dichlo-

rodiphenyltrichloroethane [DDT]; *p,p′*-DDD; *p,p′*-DDE; *p,p′*-DDT) and 20 PCB congeners (8, 18, 28, 44, 52, 66, 77, 101, 105, 118, 126, 128, 138, 153, 170, 180, 187, 195, 206, and 209). Tissues were extracted for organic target compounds by automated solvent extraction (ASE 2000, Dionex, Sunnyvale, CA, USA) with methylene chloride. Extracts were analyzed for pesticides and PCB congeners by a modified National Oceanic and Atmospheric Administration ORCA 130 method [28]. A gas chromatograph was equipped with a dual microelectron capture detector, a liquid autosampler, and two capillary columns. The method detection limits for this method varied by target compound, but ranged between 0.00025 and 0.0025 mg/kg wet weight. Percent lipid was determined by drying two weighed subsamples of extract at 105°C for 30 min and reweighing. The percent lipid was calculated as the percent lipid of wet sample weight.

Analytical accuracy was assessed by using duplicate analyses for 10% of the tissue samples. Spiked sample recovery and procedural blanks were used for 5% of the samples. Data were considered acceptable if the spiked recoveries were ≥85% and ≤115% of expected values and the relative percent difference between a sample and its corresponding duplicate was ≤20%.

Isotopic analysis

Breast muscle samples were placed in separate, clean, plastic tubes (cryotubes) and frozen at −20°C until analysis. Before analyses, tissues were dried at 60 to 70°C for 48 h and then ground to a fine powder with a mortar and pestle. Samples were weighed (1–1.5 mg) into tin cups and analyzed for $\delta^{13}C$ and $\delta^{15}N$ at the mass-spectrometry facility, University of Alaska–Fairbanks, by using a Europa 20/20 continuous-flow isotope ratio mass spectrometer. Samples were analyzed in duplicate and results were accepted if the variance between the duplicates was not greater than the variance of the peptone standard [29]. Isotopic ratios are reported as per mil (‰) deviation from the standard as defined by

$$\delta X = [(R_{sample}/R_{standard}) - 1] \times 1{,}000$$

where X is ^{13}C or ^{15}N, and R is the ratio $^{13}C/^{12}C$ or $^{15}N/^{14}N$. Standards were PeeDee Belemnite (C) and atmospheric N_2 (N).

Data analyses

Organochlorine concentrations in breast muscle and trace metal concentrations in livers were compared within each species among the five sites. Analytes with more than 50% of all samples below the limit of detection (i.e., nondetect) were not statistically analyzed. Nondetect values of statistically analyzed compounds were substituted with normally distributed random numbers between zero and the limit of detection for each sample [30]. In addition to site differences, species (cormorants), sex (cormorants only; sandpiper samples were pooled randomly), year, and interaction terms were included in initial models. Terms were included in the final model only if univariate p values were <0.05. Data were not log transformed and were predominantly nonnormal ($p < 0.05$, Shapiro Wilks' test), but homoscedastic ($p > 0.05$, Bartlett's test), and because linear models are robust under these conditions [31], we used parametric statistical tests for all analyses.

Identifying possible point sources required testing for differences among sites. For these analyses, we used a multivariate analysis of covariance design with site as a factor, percent lipid as a covariate, and contaminant concentrations as the response variables for OCs. We used a multivariate analysis of variance with site as the main factor and contaminant concentrations as the response variables for trace metals. If an analyte had more than 50% of samples below the limit of detection at one site, that site was excluded from the analysis. If the overall multivariate model showed significant differences among sites, Bonferroni-adjusted post hoc comparisons were performed on analytes with significant ($p < 0.05$) univariate F statistics to determine significant differences among sites. Correlation analyses (Spearman's ρ) were used to detect any association among morphological characters and contaminants.

Patterns suggesting atmospheric transport (i.e., west-to-east decline in contaminant concentrations) could only be tested through linear regression. We used linear regression models to investigate relationships between relative longitude (longitudinal differences among sites with Attu set artificially to 0°) and concentrations of atmospherically transported contaminants and stable isotope values. To correctly test this hypothesis, regressions were conducted with all sites (including sites with >50% nondetects) included in the analyses under the presumption that low rates of detection were correlated with low concentrations. For these analyses, all nondetects were substituted with random numbers between zero and the limit of detection for each sample. All critical p values were adjusted for multiple comparisons by using the Bonferroni correction procedure [31].

RESULTS

Organochlorines

Cormorants. Six compounds were detected in sufficient quantities to permit statistical comparisons in cormorants: hexachlorobenzene, *p,p′*-DDE, PCB-138, PCB-153, sum of all quantified PCB congeners (ΣPCBs), and *trans*-nonachlor (Table 1). Although uncommon among all sites (i.e., occurred in fewer than 50% of total samples), dieldrin was detected in more than 50% of the samples at Kiska and Adak, and PCB-180 was detected in more than 50% of samples from Attu and Adak (Table 1). Other OCs previously detected in the region [4,5] were rare (mirex) or not detected (*p,p′*-dichlorodiphenyldichloroethane).

Multivariate analysis indicated significant differences in contaminant levels in cormorants among sites ($F_{24, 228} = 2.98$, $p < 0.0001$; Wilks' lambda). No significant differences were found between cormorant species, sexes, or for the year × site interaction effect. Year was only marginally significant ($F_{6,45} = 2.37$, $p = 0.0446$) but was driven by low concentrations in Amak birds that were collected only in 2001. Therefore, year was not included in the subsequent univariate analyses.

The only OCs that differed significantly among sites were DDE, PCB-138, PCB-153, and ΣPCBs ($F_{5,70} = 2.36$, $p = 0.0486$; $F_{5,70} = 9.73$, $p < 0.0001$; $F_{5,70} = 7.45$, $p < 0.0001$; $F_{5,70} = 8.24$, $p < 0.0001$; respectively). Post hoc tests could not identify the source of variation for DDE, but all PCBs had the same pattern, with the highest concentrations in birds from Adak and Attu (Table 1). All detected OCs were positively correlated with each other ($p < 0.001$, Spearman's ρ); however, correlation analysis failed to detect any association between OC concentrations in muscle tissue and morphological characters or mass ($p > 0.05$, Spearman's ρ). Regressions of OCs on relative longitude were significant ($p < 0.05$) for four contaminants (hexachlorobenzene, DDE, *trans*-nonachlor, and

Table 1. Geometric means (and ranges) for organochlorine concentrations (mg/kg wet wt) in muscle tissue of cormorants (*Phalacrocorax* spp.) from the Aleutian Islands, AK, USA

Analyte[a]	Attu (n = 23)	Kiska (n = 12)	Adak (n = 12)	Amlia (n = 19)	Amak (n = 10)
Dieldrin	NA[b]	0.0012 (ND–0.003)	0.0022 (ND–0.005)	NA	ND[c]
Hexachlorobenzene	0.0033 (ND–0.029)	0.0023 (ND–0.005)	0.0026 (0.001–0.006)	0.0026 (ND–0.005)	0.0011 (ND–0.001)
p,p′-DDE[d]	0.0095 (ND–0.088)	0.0029 (ND–0.013)	0.0136 (0.009–0.024)	0.0090 (ND–0.028)	0.0017 (ND–0.003)
PCB[e]-138	0.0065 AB (ND–0.057)	0.0015 B (ND–0.006)	0.0172 A (0.005–0.036)	0.0023 B (ND–0.006)	ND
PCB-153	0.0085 AB (ND–0.106)	0.0015 B (ND–0.004)	0.0307 A (0.011–0.097)	0.0059 B (ND–0.018)	ND
PCB-180	0.0063 (ND–0.036)	NA	0.0149 (ND–0.005)	NA	ND
Total PCB	0.0260 AB (ND–0.275)	0.0023 B (ND–0.010)	0.0716 A (0.025–0.233)	0.0055 B (ND–0.033)	ND
trans-Nonachlor	0.0048 (ND–0.038)	0.0010 (ND–0.001)	0.0035 (0.003–0.005)	0.0029 (ND–0.011)	0.0012 (ND–0.002)
Percent lipid	1.68	2.95	2.66	2.20	0.77

[a] Analytes followed by different uppercase letters indicate significant differences at the $p < 0.01$ level.
[b] NA = not calculated because analyte was detected in <50% of samples (n = number of samples analyte detected).
[c] ND = not detected in any samples (limit of detection <0.0006 mg/kg).
[d] DDE = dichlorodiphenyldichloroethylene.
[e] PCB = polychlorinated biphenyl.

PCBs), and in all cases concentrations decreased from west to east (Fig. 2).

Rock sandpipers. Only *p,p′*-DDE was detected in enough rock sandpipers to permit an analysis of covariance test. Differences were found among sites in DDE ($F_{4,14} = 16.33$, $p < 0.0001$; analysis of covariance), with Adak birds having the highest concentrations among islands (Table 2). No relationship was detected between *p,p′*-DDE and relative longitude ($F_{1,17} = 0.02$, $r^2 = 0.001$, $p = 0.89$). The PCB-138 and PCB-153 were detected in all Adak and most (67%) Attu samples (Table 2).

Metals

Cormorants. Arsenic, cadmium, copper, chromium, mercury, selenium, and zinc were detected in enough samples to permit comparisons. Multivariate analysis of these metals indicated significant differences among sites ($F_{20,94} = 3.55$, $p < 0.0001$). Year, sex, and the interaction terms were not significantly different ($p > 0.05$).

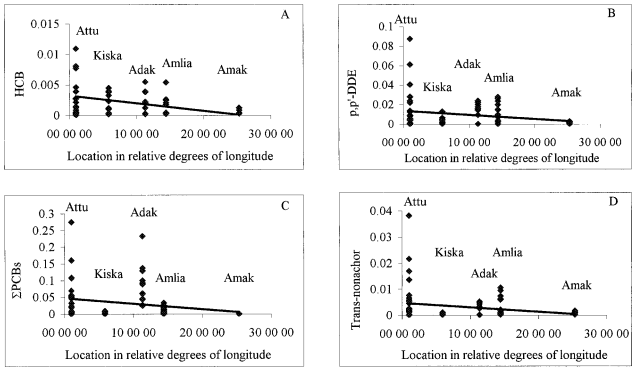

Fig. 2. Relationships between atmospherically transported organochlorines in muscle tissue (mg/kg wet wt; see Table 1 for sample sizes) of cormorants (*Phalacrocorax* spp.) and relative longitude of sampled Aleutian Islands, Alaska, USA. (**A**) Hexachlorobenzene (HCB) (HCB = -1.24×10^{-8}[longitude] + 0.03, $r^2 = 0.07$, $p = 0.0201$); (**B**) *p,p′*-dichlorodiphenyldichloroethylene (DDE) (DDE = -4.21×10^{-8}[longitude] + 0.01, $r^2 = 0.06$, $p = 0.04$); (**C**) total polychlorinated biphenyls (ΣPCB) (ΣPCB = -1.62×10^{-7}[longitude] + 0.05, $r^2 = 0.06$, $p = 0.035$); and (**D**) *trans*-nonachlor (*trans*-nonachlor = -1.67×10^{-8}[longitude] + 0.01, $r^2 = 0.06$, $p = 0.04$).

Overall, elemental metal concentrations were highly variable (Table 3). Univariate comparisons detected differences in cadmium ($F_{4,70} = 6.50$, $p = 0.0005$), mercury ($F_{4,72} = 6.50$, $p = 0.022$), and selenium ($F_{4,44} = 3.75.21$, $p = 0.010$) among sites. Cadmium concentrations were highest on Attu and significantly lower on Kiska and Amak (Table 3). Mercury concentrations were extremely variable (Table 3), and post hoc tests did not identify the source of variation. Although selenium concentrations were highest on Attu and Amak, they only differed significantly from Kiska (Table 3). Linear regressions detected significant relationships between relative longitude and arsenic and cadmium, but not mercury or selenium (Fig. 3). Mercury was positively correlated with selenium ($r = 0.4975$, $p = 0.0003$; Spearman's ρ).

Rock sandpipers. Six metals (arsenic, cadmium, chromium, nickel, mercury, and selenium) were detected in sufficient quantities to permit statistical analyses (Table 2). Multivariate analysis showed a significant difference among sites ($F_{3,21} = 4.98$, $p = 0.0242$), and subsequent univariate analyses detected significant differences among sites in concentrations of arsenic ($F_{3,15} = 8.09$, $p = 0.002$) and mercury ($F_{3,15} = 13.00$, $p = 0.000$; Table 2). Concentrations of arsenic and cadmium decreased significantly with relative longitude, but concentrations of mercury significantly increased with longitude (Fig. 4). Unlike the cormorants, mercury was not correlated with selenium in sandpipers ($r = 0.0374$, $p = 0.8991$; Spearman's ρ).

Trophic comparisons. As expected because of trophic level differences, the majority of detected contaminant concentrations in breast and liver tissue were significantly higher in cormorants than in rock sandpipers from the same location ($p < 0.01$; see Tables 1–3 for geometric means). Concordance between trophic levels among the four sites held in common between the two taxa thus required testing of patterns (i.e., how often did detects or nondetects agree between the two

Table 2. Geometric means (ranges) for elemental concentrations (mg/kg dry wt) in livers and *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE) and total polychlorinated biphenyl (ΣPCB) concentrations (mg/kg wet wt) in muscle of rock sandpipers (*Calidris ptilocnemis*) from the Aleutian Islands, AK, USA

Analyte[a]	Attu ($n = 6$)[b]	Adak ($n = 6$)	Amlia ($n = 4$)	Alaska Peninsula ($n = 3$)
Arsenic	6.58 A (4.01–8.17)	2.09 B (1.36–3.27)	1.34 B (1.04–1.69)	3.52 AB (2.01–8.12)
Cadmium	7.61 (5.03–10.02)	5.96 (4.72–7.36)	6.88 (5.18–9.55)	4.65 (3.66–5.65)
Chromium	5.68 (ND[c]–7.77)	5.83 (5.26–6.54)	6.15 (5.65–7.05)	6.56 (ND–6.50)
Mercury	0.54 B (0.08–1.02)	1.12 B (0.14–3.87)	0.92 B (0.31–2.29)	8.48 A (4.13–13.95)
Nickel	1.03 (ND–3.57)	0.09 (0.05–0.17)	0.11 (0.07–0.17)	0.08 (ND–0.10)
Selenium	14.12 (5.25–24.59)	11.01 (6.54–22.35)	15.82 (5.56–35.36)	24.27 (12.16–60.94)
p,p′-DDE[d]	0.0015 B (ND–0.0089)	0.0031 A (0.0017–0.0045)	0.0008 B (ND–0.0014)	0.0012 B (ND–0.0015)
ΣPCB[e]	0.0031 (ND–0.0017)	0.0045 (0.0020–0.0120)	ND[f]	ND
Lipid (%)	0.96	1.68	1.36	1.29

[a] Analytes followed by different uppercase letters indicate significant differences at the $p < 0.025$ level.
[b] Sample sizes are two individuals combined for each value of *n* (see *Materials and Methods*).
[c] ND = not detected (limit of detection [LOD] < 0.59 mg/kg for Cr, LOD < 0.03 for Ni).
[d] LOD = 0.0006 mg/kg.
[e] Sum of PCB congeners 138 and 153.
[f] LOD < 0.0006 mg/kg.

taxa?). Concordance for within-site comparisons showed pattern agreement (detect–detect or nondetect–nondetect for population samples of each species) among 82% of all 37 contaminants measured. Concordance was lower (30%) among detected contaminants, but this value is more heavily biased by contaminant level differences between the two trophic levels (i.e., the higher trophic level cormorants are more likely to yield detections of the contaminants that are present).

Stable isotopes. Other studies have demonstrated that lipids tend to show depleted levels of $\delta^{13}C$ relative to other tissues [32,33]; therefore, isotope ratios in tissues may be affected by lipid content. Multivariate analysis of $\delta^{13}C$, $\delta^{15}N$, and lipid detected significant differences among sites ($F_{5,42} = 34.63$, $p < 0.0001$). Univariate tests detected differences among sites in both nitrogen and carbon ($F_{4,42} = 5.26$, $p = 0.002$ and $F_{4,42} = 25.64$, $p < 0.0001$, respectively). Although nitrogen isotope values in muscle tissue had a small range among islands (range 10.36–11.64‰), a significant difference was found between the highest $\delta^{15}N$ value from Adak and the lowest values from Kiska ($p < 0.01$). Location, lipid, and the interaction (lipid × location) had significant effects on $\delta^{13}C$ ratios ($F_{4,39} = 8.89$, $p = 0.0001$, $F_{1,39} = 5.22$, $p = 0.0278$, and $F_{4,39} = 4.28$, $p = 0.0057$, respectively). Carbon values were significantly lower on Attu and Kiska than values from Adak, Amlia, and Amak ($p < 0.0001$; Fig. 5). Regression analysis detected a significant increase in $\delta^{13}C$ values in cormorant muscle tissue with relative

Table 3. Geometric means (and ranges) for elemental concentrations (mg/kg dry wt) in cormorant livers from the Aleutian Islands, AK, USA

Metal[a]	Attu ($n = 23$)	Kiska ($n = 12$)	Adak ($n = 12$)	Amlia ($n = 20$)	Amak ($n = 10$)
Arsenic	1.18 (0.27–4.02)	0.74 (0.40–1.16)	1.13 (0.77–1.67)	1.14 (0.38–2.92)	1.58 (1.08–2.96)
Cadmium	3.04 A (ND[b]–12.93)	1.21 B (0.39–2.81)	2.05 AB (0.83–4.56)	1.92 AB (0.86–6.61)	0.85 B (0.33–2.01)
Chromium	4.98 (2.24–17.69)	3.77 (3.18–5.16)	4.07 (1.37–7.50)	4.40 (2.70–9.36)	5.40 (4.41–6.72)
Copper	20.30 (14.86–45.14)	20.53 (14.14–28.76)	17.33 (7.07–29.08)	19.97 (17.05–23.88)	NA[c]
Mercury	3.69 (0.16–27.66)	1.45 (0.49–5.63)	2.11 (0.89–5.03)	3.45 (0.34–14.15)	3.89 (1.45–7.51)
Selenium	16.95 A (4.25–39.64)	5.70 B (5.01–6.32)	13.67 AB (9.65–26.17)	12.86 AB (7.31–24.82)	19.33 A (11.63–39.07)
Zinc	86.04 (63.19–273.26)	88.82 (69.92–117.36)	68.79 (36.13–84.37)	77.21 (67.07–89.12)	NA

[a] Elements followed by different uppercase letters indicate significant differences at the $p < 0.01$ level.
[b] ND = not detected (limit of detection < 0.59 mg/kg).
[c] NA = not analyzed for this location.

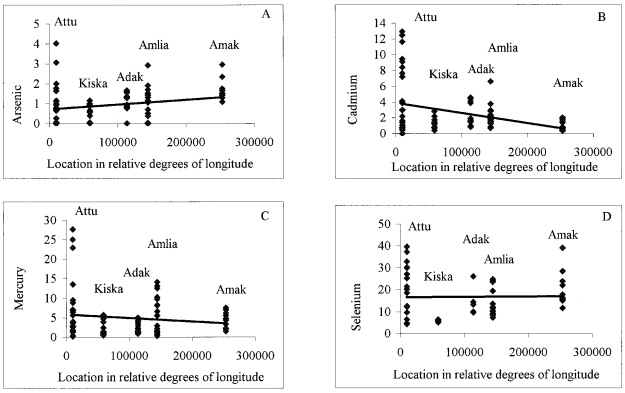

Fig. 3. Relationships between trace elements in liver tissue (mg/kg dry wt; see Table 3 for sample sizes) of cormorants (*Phalacrocorax* spp.) and relative longitude of sampled Aleutian Islands, Alaska, USA. (**A**) Arsenic (As = -2.4×10^{-6}(longitude) + 0.72, $r^2 = 0.05$, $p = 0.05$); (**B**) cadmium (Cd = -1.28×10^{-5}(longitude) + 3.89, $r^2 = 0.13$, $p = 0.001$); (**C**) mercury (Hg = -8.9×10^{-6}(longitude) + 5.83, $r^2 = 0.02$, $p = 0.26$); and (**D**) selenium (Se = 1.45×10^{-6}(longitude) + 16.64, $r^2 < 0.01$, $p = 0.59$).

longitude ($F_{1,47} = 78.89$, $p = 0.0001$, $r^2 = 0.63$; Fig. 5). No such relationship existed for δ^{15}N and longitude ($F_{1,47} = 0.00$, $r^2 = 0.0001$, $p = 0.96$). Arsenic and PCBs were the only analytes correlated with δ^{15}N or δ^{13}C, respectively ($p < 0.05$, Spearman's ρ), suggesting a slight biomagnification effect in cormorants for these compounds.

DISCUSSION

Our results reveal a repeated pattern of significant west-to-east declines among some organic contaminants in cormorants and rock sandpipers in the Aleutian Islands. Atmospheric transport is the only hypothesis proposed that explains these overarching results, but variation in the data suggests that point sources also may contribute to Aleutian contaminants. Nevertheless, this repeated west-to-east pattern suggests that this subarctic region is susceptible to similar atmospheric deposition phenomena occurring in some regions of the arctic [7,11–14,16,34,35]. Linear regressions were used only to test for longitudinal relationships with contaminant concentrations, not to serve as predictive models. The *p* values for some analytes were above the adjusted alpha levels for multiple comparisons, but the overwhelming percentage of significant relationships belies the possibility that these positive findings are a result of increased experimentwise error. Despite the high variability in contaminant concentrations among individual cormorants (i.e., low r^2 values), which is common in mobile organisms, examination of our OC data consistently revealed patterns concordant with the atmospheric transport hypothesis.

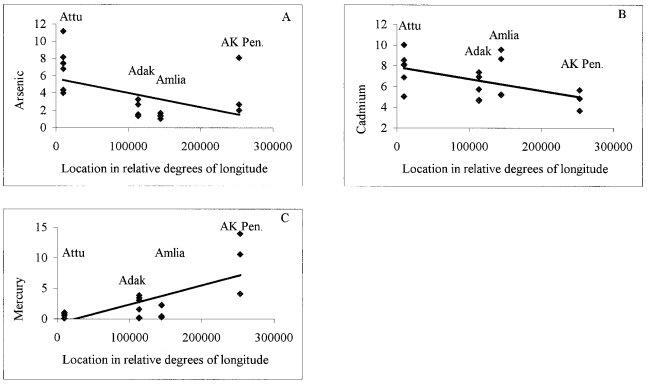

Fig. 4. Relationships between trace elements in liver tissue (mg/kg dry wt; see Table 2 for sample sizes) of rock sandpipers (*Calidris ptilocnemis*) and relative longitude of sampled Aleutian Islands, Alaska, USA. (**A**) Arsenic (As = -1.65×10^{-5}(longitude) + 5.69, $r^2 = 0.21$, $p = 0.05$); (**B**) cadmium (Cd = -1.15×10^{-5}(longitude) + 7.88, $r^2 = 0.27$, $p = 0.02$); and (**C**) mercury (Hg = -3.13×10^{-5}(longitude) + 0.77, $r^2 = 0.51$, $p < 0.001$).

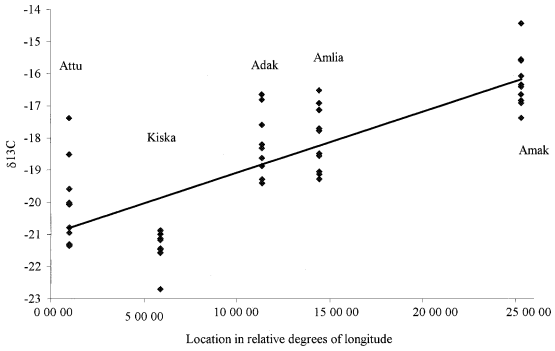

Fig. 5. Relationship between carbon isotope ratios (‰) in muscle tissue ($n = 10$ for all sites) of cormorants (*Phalacrocorax* spp.) and relative longitude of sampled Aleutian Islands, Alaska, USA. $\delta^{13}C = -20.989 + 0.001$(longitude), $r^2 = 0.63$, $p < 0.001$.

Although many of the PCBs found at high latitudes are globally transported [7,9], past military installations are considered a major point source in the Aleutian Islands [4,10,35,36]. Indeed, examination of our data suggests that both sources may be responsible for PCB distribution in the Aleutians. The repeated significant relationships between contaminants and longitude do not generally show smooth west-to-east declines. The highest mean concentrations of PCBs in cormorants (Table 1) and the only detected PCBs in rock sandpipers (Table 2) were from Attu and Adak. Although the military histories of these islands are very different, these patterns suggest that military installations may contribute point source PCBs at two trophic levels. Significant differences in cadmium and selenium among sites in cormorants (Table 3) had no discernible pattern and therefore may reflect unknown point sources. Mercury concentrations increased significantly from west-to-east in rock sandpipers, a pattern opposite to that expected from long-range transport and a further indication that point sources may be involved in this system. High mercury concentrations in rock sandpipers from the Alaska Peninsula drive this relationship (Fig. 4C), and this may indicate an eastern point source, either from military or other human use [3] or from natural mercury deposits in the region [18].

Theoretically, the repeated, significant pattern of west-to-east decline among OCs is confounded by a concordant west-to-east decline in point source intensities. This does not seem likely, because the Aleutian Islands have experienced a westward expansion and subsequent west-to-east decline in modern development and occupation [3; D.A. Rocque and K. Winker, personal observations]. However, future work focusing on the role of point sources across the archipelago is necessary to decouple the relative contributions of these two sources of contaminants to this region.

Importantly, we found no evidence to suggest that contaminants were transported into local food webs by migratory prey species. The easterly enrichment of δ^{13}C values was significant in cormorant muscle (Fig. 5) and reflects the trend of west-to-east enrichment of δ^{13}C found in plankton along the Aleutian

Archipelago [21]. Although no direct comparison could be made between patterns in our study and patterns found by Schell et al. [21], the similar trend of $\delta^{13}C$ values between plankton and cormorants implied that, during this study, cormorant populations were feeding in nonmigratory, localized food webs.

Among the PCB congeners, only PCB-138 and PCB-153 were frequently detected in this study. These two congeners are prevalent in aquatic biota [37] and are routinely detected in marine and arctic food webs [9]. Toxicity thresholds for PCBs are congener-specific, and toxic equivalency factors enable direct comparisons among studies by converting toxicity relative to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin [38]. Low concentrations of few congeners in this study limited the utility of toxic equivalency factors for comparison. Total PCBs detected in this study did not exceed thresholds considered to be harmful to birds [8].

Organochlorine concentrations (geometric mean) in our study were lower and, specifically, total PCBs were a level of magnitude lower than concentrations in individual Amchitka cormorant breast muscle [35], but the high limit of detection (i.e., 0.01 mg/kg) in the Amchitka study limited comparisons between the two studies [35]. Other contaminant studies of Aleutian avifauna [4,5,10,35,36] generally had higher concentrations than detected in our study. These results may be due to biomagnification, because the majority of contaminant research in the Aleutian Islands has been conducted on avian predators that are trophically elevated with respect to cormorants and rock sandpipers. Differing tissue matrices also may explain the lower concentrations detected in our study. We analyzed OCs in muscle tissue, which has less lipid and therefore lower accumulated OCs than eggs, which were analyzed in other studies [4,5]. Sampling techniques also may be responsible for concentration differences among studies. Other studies documenting contaminants in Aleutian birds have been conducted opportunistically on carcasses and unhatched eggs [5]. Our sampling of living birds is more representative of contaminant concentrations in birds throughout the entire archipelago and provides baseline data for future comparison.

Trace metal concentrations in cormorant livers were also below those previously found in Amchitka cormorants and Adak bald eagles [10,35]. Mean levels of pollutants for our study and others conducted on Aleutian birds generally are below levels considered harmful [39,40]. However, declines in reproductive success in bald eagles [4,5] and elevated levels in some of the cormorants examined in our study suggest that contaminants are a problem in the Aleutian Islands.

Despite complex air and ocean currents, unknown point sources, and increasing ship traffic between the western United States and Southeast Asia, we detected patterns consistent with long-range transport at two trophic levels. Global transport, unlike point sources, cannot be reduced through mitigation. Contaminants in this region not only have implications for wildlife, but they also have the potential to impact one of North America's largest fisheries. Contaminant concentrations in high-latitude species are likely to rise with increasing industrial growth and emissions in Asia. Continued monitoring of sentinel species in this unique ecosystem is crucial as slowly degrading compounds accumulate through continued atmospheric transport and deposition in the North Pacific.

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