ALBATROSS SPECIES DEMONSTRATE REGIONAL DIFFERENCES IN NORTH PACIFIC MARINE CONTAMINATION

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Abstract. Recent concern about negative effects on human health from elevated organochlorine and mercury concentrations in marine foods has highlighted the need to understand temporal and spatial patterns of marine pollution. Seabirds, long-lived pelagic predators with wide foraging ranges, can be used as indicators of regional contaminant patterns across large temporal and spatial scales. Here we evaluate contaminant levels, carbon and nitrogen stable isotope ratios, and satellite telemetry data from two sympatrically breeding North Pacific albatross species to demonstrate that (1) organochlorine and mercury contaminant levels are significantly higher in the California Current compared to levels in the high-latitude North Pacific and (2) levels of organochlorine contaminants in the North Pacific are increasing over time. Black-footed Albatrosses (Phoebastria nigripes) had 370-460% higher organochlorine (polychlorinated biphenyls [PCBs], dichlorodiphenyltrichloroethanes [DDTs]) and mercury body burdens than a closely related species, the Laysan Albatross (P. immutabilis), primarily due to regional segregation of their North Pacific foraging areas. PCBs (the sum of the individual PCB congeners analyzed) and DDE concentrations in both albatross species were 130-360% higher than concentrations measured a decade ago. Our results demonstrate dramatically high and increasing contaminant concentrations in the eastern North Pacific Ocean, a finding relevant to other marine predators, including humans.

Key words: albatross; contaminants; DDE; mercury; North Pacific; organochlorines; PCB; seabirds; stable isotopes; temporal trends.

Introduction

The publication of Rachael Carson's Silent Spring (1962) raised awareness about the impacts of man-made contaminants and catalyzed efforts to reduce environmental pollution, such as the U.S. 1963 Clean Air Act and 1972 Clean Water Act. Despite this legislation, global, or nonpoint source, pollution still threatens human health and biodiversity (Colborn et al. 1996, Skaare et al. 2000, de Wit et al. 2004, Hites et al. 2004). Nonpoint source pollution, which cannot be traced back to its origin, accumulates in the world's oceans via atmospheric transport and is distributed by ocean currents. Consequently, nonpoint source contamination is ubiquitous in the marine environment (Tatton and Ruzicka 1967, Guruge et al. 2001b), and pollutants that biomagnify up food webs, such as organochlorines (e.g., polychlorinated biphenyls [PCBs], dichlorodiphe-

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nyltrichloroethanes [DDTs]) and mercury, are found at concentrations that negatively affect health and fertility in humans and marine wildlife (Reijnders 1986, De Guise et al. 1995, Simmonds et al. 2002, Tchounwou et al. 2003).

Given that organic contaminants biomagnify, spatial differences in contaminant concentrations in seawater produce significant regional differences in contaminant loads in marine predators (Prudente et al. 1997, Aguilar et al. 2002, Ueno et al. 2003). The importance of understanding geographic patterns of marine pollution as a factor in contaminant biomagnification is underscored by recent evidence that high organochlorine concentrations in farmed salmon can be directly linked to the regional differences in the contaminant load of the salmon's main food source, small pelagic fish (Hites et al. 2004). However, defining regional sources of contaminant exposure to pelagic predators is difficult because predators travel across vast oceanographic distances, and organic contaminants accumulate in an organism throughout its lifetime.





PLATE 1. (Left) A pair of Black-footed Albatrosses (*Phoebastria nigripes*), and (right) a pair of Laysan Albatrosses (*Phoebastria immutabilis*). Photo credit: B. S. Keitt.

Organochlorines and mercury are highly toxic compounds, causing neurological, immune, and reproductive impairment (Carson 1962, Fry 1995, National Research Council 2000, Tchounwou et al. 2003). Organochlorines have been introduced into the environment through a variety of anthropogenic activities, although their use has been banned in the United States and Europe since the 1970s because of their toxicity and environmental persistence (Carson 1962). Mercury concentrations in the environment have increased substantially over the last century due to industrial waste discharge and fossil fuel combustion (Tchounwou et al. 2003).

High concentrations of organochlorines and mercury are present in the marine environment, placing longlived marine predators that biomagnify contaminants at the greatest risk for toxic effects (De Guise et al. 1995, Guruge et al. 2001b, Aguilar et al. 2002, Tchounwou et al. 2003). One group of marine predators at risk is the albatrosses, large, long-lived predatory seabirds that travel great distances and forage over vast oceanographic areas (Tickell 2000). North Pacific albatrosses have the highest organochlorine concentrations of any albatross species studied, with concentrations up to an order of magnitude higher than southern-hemisphere albatrosses (Guruge et al. 2001b). Among North Pacific albatrosses, studies over the past three decades consistently show that Black-footed Albatrosses (Phoebastria nigripes) have higher organochlorine and mercury concentrations than Laysan Albatrosses (P. immutabilis) (Fisher 1973, Auman et al. 1997, Guruge et al. 2001b).

Similarities in behavioral and breeding ecology between Black-footed and Laysan Albatrosses (Whittow 1993a, b, Tickell 2000) make these reported differences in contaminant body burden surprising. Previous studies have suggested differences in diet are responsible for Black-footed Albatrosses having greater contaminant body burden concentrations than Laysan Albatrosses (Jones et al. 1996, Auman et al. 1997, Klasson-Wehler et al. 1998). However, these papers did not investigate foraging patterns or diet composition. Both species are

known to eat fish, fish eggs, squid, and crustaceans (Whittow 1993*a*, *b*), with past diet studies reporting conflicting relative proportions of each prey species consumed (Harrison et al. 1983, Gould et al. 1997).

We used these marine top predators, Black-footed and Laysan Albatrosses, to investigate the temporal and spatial patterns of North Pacific marine contamination. Plasma concentrations and correlation patterns of organochlorines and mercury, along with indices of trophic status (nitrogen stable isotopes) and foraging location (carbon stable isotopes and satellite telemetry) were evaluated to determine whether Black-footed and Laysan Albatrosses possess different contaminant concentrations, and if so whether these differences were due to a contaminated point source, trophic level, or regional differences in ocean contaminant concentrations. Temporal patterns were assessed by comparing our data with organochlorine data from Black-footed and Laysan Albatrosses collected in the early 1990s (Auman et al. 1997). Black-footed and Laysan Albatrosses are ideal species with which to examine temporal and spatial patterns of North Pacific marine contamination because (1) they breed sympatrically (Whittow 1993*a*, *b*), (2) they are closely related (Nunn et al. 1996), (3) they are high trophic-level species with a similar diet (Whittow 1993a, b), and (4) information is known about their foraging behavior (Shuntov 1974, Fernandez et al. 2001, Hyrenbach et al. 2002). Our study provides important information on persistent organic pollutants in marine predators in the North Pacific, an area of high biological and economic importance.

METHODS

Sample collection

Blood samples were collected from adult albatrosses during the breeding season in May of 2000 (n=9 Laysan, n=11 Black-footed) and 2001 (n=7 Laysan, n=15 Black-footed) on Sand Island, Midway Atoll (28°13′ N latitude, 177°13′ W longitude) in the Northwestern Hawaiian Islands (see Plate 1). Whole blood was stored frozen at -20° C for total mercury and stable

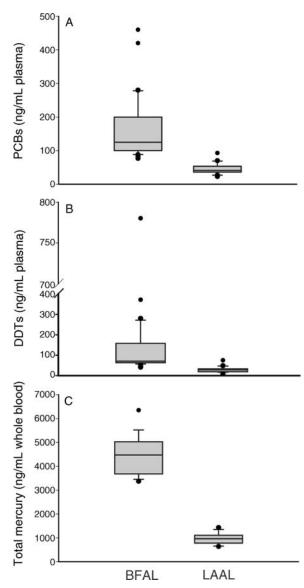


Fig. 1. (A) PCBs, (B) DDTs, and (C) total mercury concentrations in Black-footed and Laysan Albatrosses. Mean concentrations are significantly (\sim 400%) higher in Black-footed (BFAL) than in Laysan (LAAL) Albatrosses (P < 0.001 for PCBs, DDTs, and mercury, Mann-Whitney U; n = 26 BFAL, n = 16 LAAL [PCBs and DDTs]; n = 15 BFAL, n = 8 LAAL [total mercury]). The box plot shows the median (horizontal line within the box), 25th and 75th percentiles (lower and upper margin of the box), and 10th and 90th percentiles (lower and upper hatch marks), as well as outliers.

isotope analysis. Plasma samples were isolated from whole blood by centrifugation within six hours of collection and stored frozen in kilned glass vials at -20° C until evaluated for organochlorine content.

Organochlorine analysis

Organochlorine determination was performed according to methods previously published (Newman et al.

1994, Jarman et al. 1996). Polychlorinated biphenyls (PCBs) are reported as the sum of 60 PCB congeners evaluated (Appendix A). DDTs are reported as the sum of seven dichlorodiphenyltrichloroethane (DDT) metabolites (Appendix B, o,p'-DDD, o,p'-DDE, o,p'-DDT, p,p'-DDD, p,p'-DDE, p,p'-DDMU, p,p'-DDT). Approximately 1 mL of plasma was spiked with PCB 207 (as a recovery standard), extracted, and fractionated by Florisil® chromatography. Both fractions were spiked with an internal instrument standard (to account for volume differences) and analyzed on a Hewlett Packard 6890 Series II capillary gas chromatograph (Hewlett Packard, Palo Alto, California, USA) utilizing electron capture detectors (GC/ECD). Two 60-m, 0.25 mm ID, 0.25 (film thickness), DB-5 and DB-17 columns provided dual-column confirmation. National Institute of Standards and Technology (NIST) solutions assured accuracy of the calibration curves. PCB 207 recoveries were $88\% \pm 8\%$ (mean \pm sD), and all samples were recovery corrected.

Total mercury analysis

Whole-blood samples (collected in 2001 only) were analyzed for total mercury concentrations by En Chem, Inc. (Green Bay, Wisconsin, USA). Samples were prepared and analyzed by the EPA SW-846 Method 7471 (Rev. 1, 1994) on a Flow Injection Mercury System (FIMS) 100 (PerkinElmer Life and Analytical Sciences, Boston, Massachusetts, USA). Standard reference material (DOLT-3, dog-fish liver) and laboratory biological control total mercury spike recoveries were between 88 and 94%. Matrix (albatross blood) total mercury spike recoveries were between 104 and 105%, and the difference between blind duplicate samples was 1–4%.

Carbon and nitrogen stable isotope ratios

Whole blood was transferred to tin capsules, dried, and analyzed for stable carbon and nitrogen isotope ratios (δ^{13} C and δ^{15} N) at the Stable Isotope/Soil Biology Laboratory, Institute of Ecology, University of Georgia, Athens, Georgia, USA.

Satellite data

Satellite telemetry data were collected in 1998 from adult Laysan (n=14) and Black-footed Albatrosses (n=15) during their breeding season (January through July) in the Northwestern Hawaiian Islands, as previously described in Fernández et al. (2001). In order to elucidate the different foraging patterns of Black-footed and Laysan Albatrosses for this study, satellite data were reanalyzed using a geographic information system (ArcGIS version 8.3, ESRI 2001) to create albatross foraging distribution maps (Fig. 4).

Data analysis

All statistical tests were performed using Systat (2000). Statistical significance was reported for P <

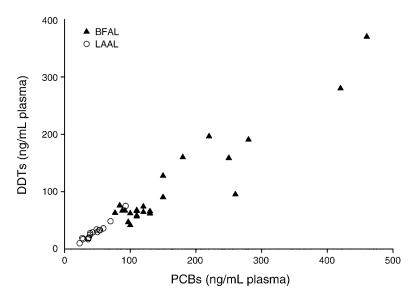


Fig. 2. A plot of PCBs vs. DDTs for Black-footed (BFAL) and Laysan (LAAL) Albatrosses. BFAL (n = 26, r = 0.79) and LAAL (n = 16, r = 0.96) had significantly correlated levels of PCBs and DDTs (P < 0.001, Pearson correlation of log-transformed data; a noninfluential BFAL data point was removed from the graph for clarity). This indicates that the contaminant exposure is diet driven from prey containing similar relative amounts of both PCBs and DDTs, as opposed to a common point source (e.g., soil contamination at their breeding site).

0.05. Variables were log-transformed if normality and equality of variance were improved. Nonparametric methods were used when the data did not meet the assumptions of parametric analyses. We pooled data from blood samples collected in 2000 and 2001 since no annual effect was detected (P = 0.423, δ^{13} C; P = 0.306, δ^{15} N; P = 0.840, sum PCBs; P = 0.784, sum DDTs).

Temporal trend evaluation

Sum plasma PCB and DDE concentrations from adult Black-footed and Laysan Albatrosses collected in the fall of 1992 and the fall, winter, and spring of 1993 (Auman et al. 1997) were used for comparison with our data to evaluate temporal trends. A one-sample *t* test was used to determine if our mean values were different from those of Auman et al. (1997).

RESULTS AND DISCUSSION

Contaminant differences between species

Black-footed Albatrosses had 370–460% higher concentrations (ng/mL) of PCBs, DDTs, and mercury than Laysan Albatrosses (PCBs = 170 vs. 46 ng/mL plasma; DDTs = 130 vs. 30 ng/mL plasma; mercury = 4500 vs. 970 ng/mL whole blood; P < 0.001 for all three comparisons, Mann-Whitney U, Black-footed n = 26 [PCBs and DDTs], n = 15 [mercury]; Laysan n = 16 [PCBs and DDTs], n = 8 [mercury]; Fig. 1). The extreme differences observed between Black-footed and Laysan Albatross contaminant concentrations is not likely due to differences in contaminant metabolism or excretion rates given that both species are closely related genetically (Nunn et al. 1996), highly sympatric and

known to interbreed (Fisher 1972), and have similar blood protein composition (Brown and Fisher 1966). Previous studies have found comparable PCB congener profile patterns (Jones et al. 1996, Guruge et al. 2001a) as well as PCB metabolite formation (hydroxylated and methylsulfonyl polychlorinated biphenyls) (Klasson-Wehler et al. 1998) in Black-footed and Laysan Albatrosses, suggesting these two species metabolize organochlorine compounds in the same way. Organic contaminant concentrations are also heavily influenced by the amount of lipid in the tissue evaluated (Clark et al. 1987). Black-footed and Laysan Albatrosses had identical plasma lipid concentrations (BFAL 0.009 ± $0.003 \text{ g/mL}, n = 16; \text{ LAAL } 0.010 \pm 0.003, n = 14; P =$ 0.38, t test), demonstrating that plasma lipid content was not responsible for the observed differences in organic contaminant concentrations between these two species.

We believe the route of contaminant exposure for both albatross species is via their diet (nonpoint source) as opposed to a contaminated point source because the measured organochlorines (DDTs and PCBs) were strongly correlated with one another (P < 0.001, Pearson correlation of log-transformed data; n = 16 Laysan, n = 26 Black-footed; Fig. 2). Exposure from a common point source (e.g., contaminated soil at their breeding site) is improbable since albatrosses with high concentrations of DDTs also had high concentrations of PCBs (Fig. 2), and PCBs, an industrial product, are not applied or discharged into the environment with DDTs, a pesticide. Instead, high correlations between PCBs and DDTs within individual birds are indicative of a nonpoint source, or dietary, exposure in which both

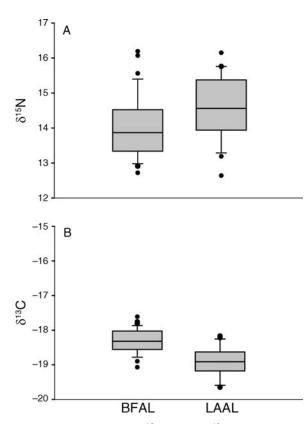


Fig. 3. Levels of (A) δ^{15} N and (B) δ^{13} C in whole-blood samples from Black-footed (BFAL) and Laysan (LAAL) Albatrosses. (A) Mean δ^{15} N values for BFAL were not higher than for LAAL (one-tailed t test). (B) Mean δ^{13} C values of BFAL were significantly higher than for LAAL (t test, P < 0.001). The box plot shows the median (horizontal line within the box), 25th and 75th percentiles (lower and upper margin of the box), and 10th and 90th percentiles (lower and upper hatch marks), as well as outliers (n = 31 BFAL, n = 17 LAAL).

species are eating prey with similar relative amounts but different absolute concentrations of PCBs and DDTs.

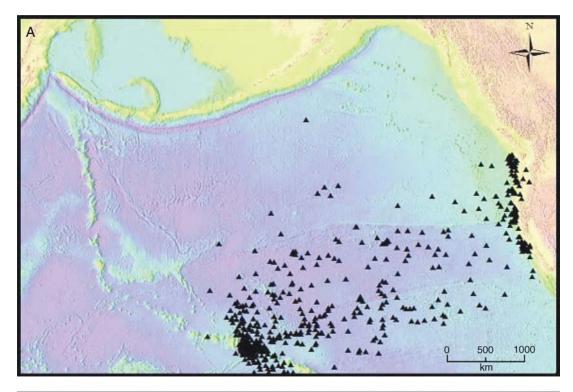
Trophic position can explain diet-driven differences in contaminant body burdens in pelagic marine species from the same oceanographic region (Jarman et al. 1996, Hobson et al. 2002). Past studies have suggested that differences in trophic-level feeding are responsible for the elevated concentrations of contaminants in Blackfooted compared to Laysan Albatrosses (Auman et al. 1997, Guruge et al. 2001b). However, because they appear to have similar diets (Harrison et al. 1983, Gould et al. 1997), the extreme differences in contaminant loads in these two albatross species are unlikely due to feeding at different trophic levels.

Stable isotopes can be used, in addition to diet comparisons, to investigate the trophic ecology of seabirds (Hobson et al. 1994, Kelly 2000). Nitrogen stable isotopes in whole blood of Black-footed and Laysan Albatrosses confirm that trophic feeding position is not responsible for the differences in contaminant load between these two species. Nitrogen stable isotopes

 $(\delta^{15}N)$ increase predictably with respect to trophic position and are used to determine trophic level in marine species, including seabirds (Hobson et al. 1994). Black-footed Albatrosses should have higher $\delta^{15}N$ values than Laysan Albatrosses if trophic position alone were the predominant determinant of their greater contaminant body burdens. In reality, blood $\delta^{15}N$ values (mean \pm se) for Black-footed Albatrosses were not higher than those for Laysan Albatrosses (Blackfooted, 14.03 ± 0.16 , n = 31; Laysan, 14.58 ± 0.24 , n = 17; one-tailed t test; Fig. 3A), indicating that trophic feeding position does not explain the Black-footed Albatross's higher contaminant concentrations.

The finding that Black-footed and Laysan Albatrosses feed at similar trophic levels is contradicted by a previous study of muscle tissue from birds killed in the North Pacific driftnet fisheries that found Laysan Albatrosses had lower $\delta^{15}N$ values than Black-footed Albatrosses (Gould et al. 1997). A more recent study in another high trophic-level North Pacific predator, the northern fur seal, demonstrated that $\delta^{13}C$ and $\delta^{15}N$ are both affected by latitudinal gradients and decrease as latitude increases (Burton et al. 2001). The latitudinal influence on $\delta^{15}N$ values may explain why the more northern-foraging Laysan Albatrosses had lower δ¹⁵N values than Black-footed Albatrosses (Gould et al. 1997). Our whole-blood samples are likely less influenced by latitude than Gould et al.'s (1997) samples because they represent a time period when the sympatric breeding Black-footed and Laysan Albatrosses begin their foraging trips from the same location (Northwestern Hawaiian Islands).

Carbon stable isotopes and satellite telemetry data were used to test the hypothesis that divergent foraging areas between Black-footed and Laysan Albatrosses explain their different contaminant loads. Carbon stable isotope ratios (δ^{13} C) in marine systems are an indication of geographic location (Rau et al. 1982, Goericke and Fry 1994) and decrease in the North Pacific as latitude increases (Burton et al. 2001). Blood δ^{13} C mean values for Black-footed Albatrosses were significantly higher than for Laysan Albatrosses (Black-footed, -18.31 ± 0.06, n = 31; Laysan, -18.90 ± 0.11 , n = 17; P < 0.001, t test; Fig. 3B), suggesting that Black-footed Albatrosses forage at lower (more southern) latitudes than Lavsan Albatrosses. Satellite telemetry data from the breeding season (January–July) support the carbon isotope data and demonstrate that Black-footed and Laysan Albatrosses forage in two distinctly different regions of the North Pacific (Fig. 4). Both species lengthen their foraging trips in duration and distance from the colony as the breeding season progresses (Fernández et al. 2001). Black-footed Albatrosses travel northeast toward the west coast of North America, a region that has a long history of contaminant discharge through industrial and agricultural practices, whereas Laysan Albatrosses head northwest toward the northern and western



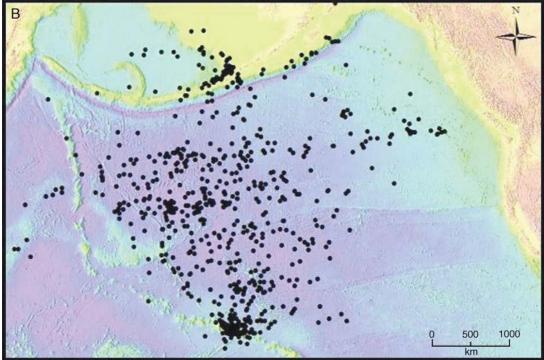


Fig. 4. Satellite-determined locations of (A) Black-footed (black triangles, n = 15) and (B) Laysan (black circles, n = 14) Albatrosses in the North Pacific Ocean. Satellite tracking data indicate that albatrosses forage in different areas within the North Pacific. The satellite transmitters were attached to birds on their breeding colony in the Northwestern Hawaiian Islands, which is the starting point for each track. Each point represents the location of a bird on a particular day (collected closest to 12:00) with 49 ± 9 d (points, mean \pm sE) represented per bird. Data were collected during the 1998 breeding season (January–July). The background map was supplied courtesy of USGS (http://walrus.wr.usgs.gov/infobank/gazette/jpg/regions/fr_np.jpg).

Table 1. Temporal trends of PCBs and DDE in Black-footed (BFAL) and Laysan (LAAL) Albatrosses.

	Contaminant level (ng/mL plasma)						
Contaminant and species	1992–1993			2000–2001			
	Mean ± sE	Range	n	Mean ± sE	Range	n	P
PCBs							
BFAL	110 ± 17	10-450	36	160 ± 19	77-460	26	0.009
LAAL	35 ± 2	12-76	39	46 ± 5	23-93	16	0.023
DDE							
BFAL	35 ± 4	0–98	36	125 ± 28	39-730	26	0.004
LAAL	16 ± 2	3-46	40	27 ± 3	9–67	16	0.007

Notes: Data from 1992 and 1993 are from Auman et al. (1997), *P* values are a result of one-sample *t* tests against 1992–1993 published mean values.

regions of the North Pacific. Year-round ship-based survey data collected between 1959 and 1968 (Shuntov 1974) further corroborate that these two species have a geographical separation in their foraging ranges. We believe that this geographical segregation explains the ~400% higher contaminant loads observed in Blackfooted compared to Laysan Albatrosses.

Temporal trends

DDE (the main breakdown product of DDT) and PCB concentrations in both Black-footed and Laysan Albatrosses were 130-360% higher in the plasma samples we collected in 2000 and 2001 than in plasma samples collected by Auman et al. (1997) in 1991-1992 and analyzed using similar techniques (Table 1). Blackfooted Albatrosses had a greater proportional increase in organochlorines over this time period, particularly for DDE (360% increase for Black-footed vs. 170% for Laysan). Temporal studies in the North Atlantic have shown that organochlorines in seabird eggs and beluga whales (Delphinapterus leucas) have decreased (Muir et al. 1996, Braune et al. 2001), while PCBs in minke whales (Balaenoptera acutorostrata) in the Antarctic and North Pacific have increased or remained stable (Aono et al. 1997). Due to differences in study species and study systems, comparisons of our results with other contaminant temporal trend studies are difficult. The environmental fate of persistent organic chemicals is complex and regulated by multiple processes, including temperature, chemical degradability, and location and quantity of discharge (Wania and Mackay 1995), characteristics that likely contribute to the regional differences of temporal patterns in marine contamination reported by Tanabe et al. (2003). Nevertheless, given that organochlorines are currently used in countries that border the Pacific Ocean (Voldner and Li 1995), and Black-footed Albatrosses have organochlorine levels believed to be of toxicological concern (Auman et al. 1997, Guruge et al. 2001a), the observed increase in organochlorines in North Pacific albatrosses in this study should cause alarm for future health risks from marine pollution in high trophic-level North Pacific species.

Conclusion

We found that two closely related (Nunn et al. 1996) and sympatrically breeding (Whittow 1993a, b) North Pacific marine top predators, Black-footed and Laysan Albatrosses, had 370-460% differences in organochlorine (PCBs, DDTs) and mercury body burdens. Evaluation of contaminant correlation patterns, nitrogen and carbon stable isotopes, and satellite telemetry data demonstrated that these contaminant differences were primarily due to regional segregation of the albatrosses' North Pacific foraging areas, not due to a contaminated point source or trophic feeding position. Furthermore, temporal patterns of PCBs and DDE concentrations in both albatross species indicate these contaminants are higher (130–360%, Table 1) than concentrations measured a decade ago. Our data provide important information on the temporal and spatial patterns of nonpoint source, or global, marine contamination.

Global contamination of the marine environment by persistent organic pollutants was first recognized in Adelie penguins (Pygoscelis adeliae) in Antarctica in 1966 (Sladen et al. 1966). Numerous studies describing marine contamination in seawater (Schulz-Bull et al. 1995) to high trophic-level species such as seabirds (Auman et al. 1997) have been published since these initial findings. Yet, the role of regional differences in the biomagnification of toxic compounds in wideranging pelagic predators remains difficult though important to understand, because marine contamination can negatively impact human health (Bjerregaard et al. 2001, Simmonds et al. 2002, Tchounwou et al. 2003, Hites et al. 2004). Elevated mercury concentrations in marine predators prompted the Environmental Protection Agency in March 2004 to advise women of childbearing age and young children to avoid eating large pelagic fish (e.g., shark, swordfish) and to restrict their intake of other marine species (U.S. Department of Health and Human Services and U.S. Environmental Protection Agency 2004).

Our results show that the North Pacific, a highly productive and economically important ocean basin, has critical regional differences in organochlorine and mercury concentrations. We also report that overall contaminant levels in the marine environment may be increasing over time. These findings have serious implications for contaminant exposure risks to long-lived high trophic-level species such as seabirds, marine mammals, and humans that are known to accumulate organochlorines and mercury through a marine diet (De Guise et al. 1995, Bjerregaard et al. 2001, Guruge et al. 2001b, Aguilar et al. 2002, Tchounwou et al. 2003).

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APPENDIX A

Polychlorinated biphenyl (PCB) congeners evaluated in Black-footed and Laysan Albatross plasma samples that were used to determine the sum of PCBs (*Ecological Archives* A016-028-A1).

APPENDIX B

Dichlorodiphenyltrichloroethane (DDT) metabolites measured in Black-footed and Laysan Albatross plasma samples (*Ecological Archives* A016-028-A2).