

Patterns and Trends of Chlorinated Hydrocarbons in Nestling Bald Eagle (*Haliaeetus leucocephalus*) Plasma in British Columbia and Southern California

Lillian S. Cesh · Tony D. Williams ·
David K. Garcelon · John E. Elliott

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Abstract Patterns and trends of chlorinated hydrocarbons were assessed in bald eagle nestling plasma from sites along the west coast of North America. Eagle plasma was sampled from four areas in southwestern British Columbia (BC), a reference site in northern BC, and from Santa Catalina Island, off the coast of California. Sites were chosen to reflect variation in contaminant exposure due to differing recent and/or historic anthropogenic activities. Santa Catalina Island had significantly greater mean concentrations of *p,p'*-DDE, 41.3 µg/kg wet weight (ww), than other sites, and Nanaimo/Crofton, BC had the greatest mean concentration of total PCBs, 28.9 µg/kg ww. Contaminant levels measured in 2003 in BC were compared to levels measured in 1993; over that ten year span, concentrations and patterns of chlorinated hydrocarbons have not significantly changed. There were no significant differences in levels of *p,p'*-DDE or hexachlorobenzene between 1993 and 2003, but significant decreases were found for *trans*-nonachlor and PCBs at BC sites. Levels of total PCBs and *trans*-nonachlor in the central Fraser Valley and Nanaimo/Crofton area have significantly decreased. Mean concentrations of *p,p'*-DDE measured in bald eagle nestling plasma samples in 2003 exceeded published criteria for

effects on bald eagle reproduction at Santa Catalina Island and Barkley Sound, more than 30 years since heavy usage restrictions were imposed.

Introduction

Bald eagles (*Haliaeetus leucocephalus*) are an important sentinel species for monitoring ecosystem contaminant concentrations and effects of contaminant exposure on wildlife in the North American environment (Bowerman et al. 1998, 2003; Donaldson et al. 1999; Elliott and Harris 2001/2002; Elliott and Norstrom 1998). Since the 1960's, levels of chlorinated hydrocarbons in North America have been monitored in this species (Elliott and Harris 2001/2002). Due to their position at the top of the food chain and their scavenging behavior, bald eagles bioaccumulate a wide array of lipophilic contaminants, including chemicals such as DDT (dichlorodiphenyl-trichlorethane), its metabolite *p,p'*-DDE (1,1-dichloroethylene bis[*p*-chlorophenyl]), PCBs (polychlorinated biphenyls), PCDDs (polychlorinated dibenzo-*p*-dioxins), and PBDEs (polybrominated diphenyl ethers) (Elliott and Harris 2001/2002; McKinney et al. 2006; Roe 2004). Most commercial uses of DDT and PCBs were severely restricted in the United States and Canada during the 1970s due to concerns over environmental persistence, bioaccumulation, and toxicity to humans and wildlife (Elliott and Harris 2001/2002). In bald eagles, these contaminants have been linked to effects such as reduced eggshell quality, decreased breeding success, cytochrome P450 enzyme induction, mortality, and possibly developmental deformities (Bowerman et al. 1994, 1995; Elliott et al. 1996a, 1996c; Elliott and Norstrom 1998; Garcelon and Thomas 1997; Gill and Elliott 2003;

L. S. Cesh (✉) · T. D. Williams
Department of Biological Sciences, Centre for Wildlife Ecology,
Simon Fraser University, Burnaby, British Columbia, V5A 1S6,
Canada
e-mail: lilly.cesh@gmail.com

D. K. Garcelon
Institute for Wildlife Studies, Arcata, CA 95518, USA

J. E. Elliott
Canadian Wildlife Service, Environment Canada, Delta,
British Columbia, V4K 3N2, Canada

Wiemeyer et al. 1984, 1993). Although the overall contaminant levels in North American bald eagles have decreased since the 1970s, there are geographic regions, including southern California and parts of the Great Lakes region, where contaminant exposure remains high and nest success of bald eagles is lower than expected (Bowerman et al. 2003; Elliott and Harris 2001/2002; Garcelon 1994).

On the southern California Channel Islands, bald eagles were extirpated in the early 1960s, in part due to high DDT and PCB exposure (Garcelon et al. 1989). Although still unable to naturally reproduce as a consequence of continued high DDT and PCB exposure, bald eagles were reintroduced to the island of Santa Catalina more than 20 years ago (Garcelon et al. 1989). Levels of contaminants in bald eagles on Santa Catalina Island have not been reported since 1993.

The number of bald eagles breeding on the south coast of BC has increased steadily since the mid-1980s. Elliott and Harris (2001/2002) showed that the regional rate of increase in the population size of bald eagles from 1966 to 1996 was greater than 1.5% each year in southwestern coastal BC and parts of Vancouver Island. Overall, on the BC coast, bald eagle nest success appears to be influenced mainly by ecological factors such as combined influences of food supply and weather (Elliott and Harris 2001/2002; Elliott et al. 1998, 2005), although there are some localized areas of poor nest success associated with exposure to relatively high concentrations of chlorinated hydrocarbons (Gill and Elliott 2003).

These previous bald eagle contaminant studies in California and BC form the historical baseline for the current study (Elliott et al. 1998b; Elliott and Norstrom 1998; Garcelon et al. 1989; Gill and Elliott 2003). The primary objectives of this study were to (1) evaluate patterns and trends of chlorinated hydrocarbons in nestling bald eagle plasma along the west coast of North America, including four areas in southwestern British Columbia (BC), a reference site in northern BC, Fort St. James (FSJ), and from Santa Catalina Island (SCI), off the coast of California, and (2) compare contaminants levels at sites in BC measured in 1993 and 2003. By continuing to monitor these legacy contaminants, information related to the health of bald eagles, status of the local environments, and fluctuations in contaminant concentrations at monitoring sites can be obtained.

Methods

Sample Collection

Nestling bald eagle blood samples were collected from five sites in British Columbia, Canada and one site in

California, USA, which were chosen to reflect variation in contaminant exposure due to differing current and/or historic anthropogenic activities (Fig. 1). Samples were collected in 2003 from southwestern BC and California sites, and in 2001 from the northern BC site. In BC, samples were obtained from Delta-Richmond (lower Fraser Valley, $n = 9$), Abbotsford-Chilliwack (central Fraser Valley, $n = 6$), Nanaimo/Crofton (southeast Vancouver Island, $n = 8$), Barkley Sound (southwest Vancouver Island, $n = 3$), and Fort St. James (northern BC, $n = 5$). In California, samples were taken from Santa Catalina Island (southern California, $n = 3$). BC eagles were first located by aerial or boating surveys and then removed from the nest by a professional tree climber. Body mass, other biometric data, and age were determined. California nestlings were collected from rocky cliffs as part of a bald eagle reintroduction program carried out by the Institute for Wildlife Studies (Arcata, CA). Detailed sampling procedures for the Fort St. James eagles are available in Weech et al. (2006). For all chicks, blood samples were collected (up to 24 mL) and immediately transferred to vacutainers containing sodium heparin. After <6 h on ice, plasma was extracted from whole blood by centrifugation and stored at -20°C .



Fig. 1 Location of study sites

Contaminant Analysis

Determination of organochlorines (OCs) and polychlorinated biphenyls (PCBs) were performed by staff at the Canadian Wildlife Service National Wildlife Research Center (NWRC; Ottawa, ON, Canada). The suite of organochlorines analyzed included: chlorobenzenes (tetra, penta, and hexa), hexachlorocyclohexanes, chlordane-related compounds (oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and heptachlor epoxide), *p,p'*-DDT and metabolites (*p,p'*-DDE and *p,p'*-DDD), mirex, photomirex, and dieldrin. Sixty-seven major PCB congeners were analyzed and summed to present the level of total PCBs. The PCB congeners analyzed for include the following: PCB 16/32, PCB 17, PCB 18, PCB 22, PCB 28, PCB 31, PCB 33/20, PCB 42, PCB 44, PCB 47/48, PCB 49, PCB 52, PCB 56/60, PCB 64, PCB 66, PCB 70/76, PCB 74, PCB 85, PCB 87, PCB 92, PCB 95, PCB 97, PCB 99, PCB 101/90, PCB 105, PCB 110, PCB 118, PCB 128, PCB 130, PCB 137, PCB 138, PCB 141, PCB 146, PCB 149, PCB 151, PCB 153, PCB 156, PCB 157, PCB 158, PCB 170/190, PCB 171, PCB 172, PCB 174, PCB 176, PCB 177, PCB 178, PCB 179, PCB 180, PCB 183, PCB 187, PCB 194, PCB 195, PCB 196/203, PCB 200, PCB 201, PCB 202, PCB 206, PCB 207, PCB 208. Of these PCB congeners, PCB 99, PCB 188, PCB 138, PCB 153, and PCB 180 had the highest mean concentrations.

The analysis of contaminants followed standard procedures and is described in full in the CWS Laboratory Service Methods Manual (Environment Canada 2003). Briefly, the plasma samples were denatured with formic acid (1:1 v/v) after the addition of internal standards. OCs/PCBs were extracted with activated C18 cartridges, eluted with DCM/hexane (1:1), and cleaned up by Florisil column chromatography. Samples were quantitatively analyzed by capillary gas chromatography coupled with a mass selective detector operated in the selected ion monitoring mode. The instruments used were the HP 5890 GC with HP Mass Selective Detector HP 5971A for PCB analysis, and Agilent 6890 GC with HP Mass Selective Detector Agilent 5973 for OC analysis. As part of the quality control, blanks and CWS reference material (2003 Herring Gull QA) were run concurrently with the samples. The nominal detection limit for all compounds was 0.1 ng/g wet weight (ww). Residues were not corrected for internal standard recoveries, which were typically between 70% and 90%. Plasma lipids were determined colorimetrically following the CWS Laboratory Service Methods Manual (Cesh 2005; Environment Canada 2003).

Bald eagle plasma samples collected in 1993 (Elliott and Norstrom 1998) were also analyzed by staff at the NWRC.

NWRC has a strict QA/QC process in place to manage changes in instrumentation and methodology.

Statistics

All statistical analyses were performed using SAS 9.1 (SAS Institute Inc., 2002–2003). Data were log-transformed ($\log_{10} + 1$) to approximate a normal distribution. Site differences were determined using analysis of variance (ANOVA) and the Student–Newman–Keuls test for post hoc pairwise comparisons. The relationship between contaminants and plasma lipids was determined using linear regression models. To compare contaminant levels in 1993 to those found in 2003, we used a Wilcoxon rank test. Unless stated otherwise, results were considered significant if $p < 0.05$.

Results

Spatial Trends in Contaminant Levels

There was no significant linear relationship between total plasma lipids and *p,p'*-DDE ($p = 0.75$) or total PCBs ($p = 0.18$); therefore, concentrations were not normalized to plasma lipid content. Total lipids were significantly different among sites ($F = 2.97$, $p = 0.0291$, $df = 5$; Table 1).

Plasma samples from Santa Catalina Island had significantly higher levels of mean *p,p'*-DDE than all BC sites ($F = 3.15$, $p = 0.02$, $df = 5$; Fig. 2). Within BC, there was no significant differences among sites ($F = 2.54$, $p = 0.64$, $df = 4$). Total PCB concentrations varied significantly among sites ($F = 5.11$, $p < 0.002$, $df = 5$), with the greatest concentration in samples from Nanaimo/Crofton and Barkley Sound, followed by Santa Catalina Island (Table 1 and Fig. 2). Mean concentrations of individual PCB congeners generally followed the geographical pattern of the total PCBs; for example, highest concentration of PCBs 99 and 153 were also at Nanaimo/Crofton and Barkley Sound (Table 1).

Temporal Changes in Contaminant Levels

Temporal changes in contaminant concentrations were investigated by comparing between 1993 and 2003 at four of the same sites (central and lower Fraser Valley, Nanaimo/Crofton, and Barkley Sound; Fig. 3).

Overall, there was no significant difference in the level of *p,p'*-DDE in British Columbia from 1993 to 2003 when pooling all BC sites ($Z = 1.82$, $p = 0.07$, $df = 1$) or when

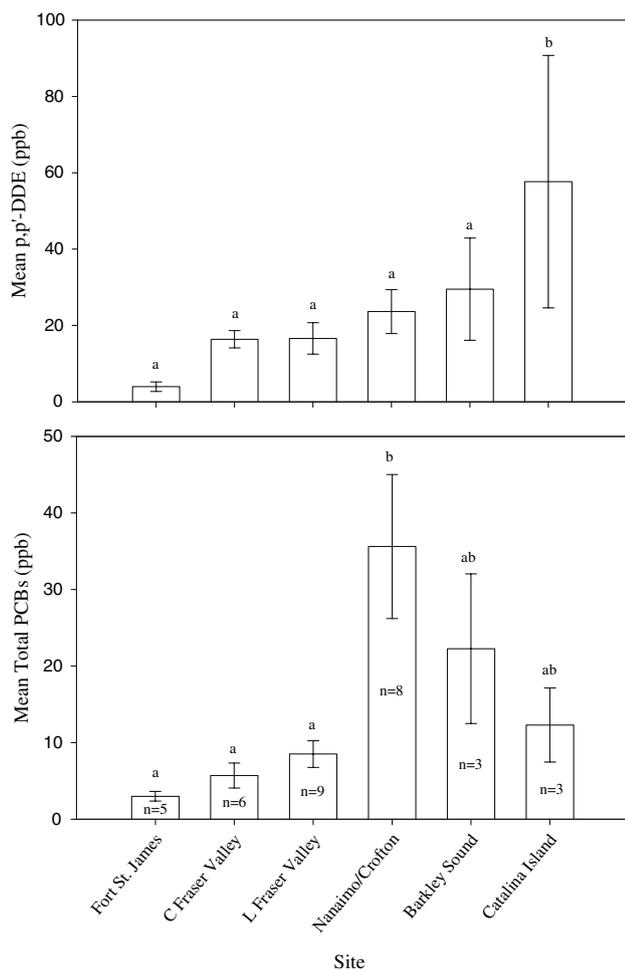


Fig. 2 Residue levels of p,p' -DDE and total PCBs in plasma samples of bald eagles nestlings collected in British Columbia and in southern California. Means that do not share the same lowercase letter were significantly different ($p < 0.05$)

sites were compared individually. Levels of *trans*-nonachlor significantly decreased over the 10 years, pooling data from the four BC sites (Fig. 3B; $Z = -2.20$, $p = 0.03$, $df = 1$), but levels of HCB did not (Fig. 3C; $Z = -0.33$, $p = 0.74$, $df = 1$). Concentrations of total PCBs significantly declined between 1993 and 2003 in BC ($Z = 2.17$, $p = 0.03$, $df = 1$; Fig. 3D, pooling all four sites). The largest decrease in total PCBs was noted in the central Fraser Valley ($Z = 2.2874$, $p = 0.0452$, $df = 1$).

Discussion

In North America, many studies have monitored contaminant levels in plasma of bald eagle nestlings (Dominguez et al. 2003; Elliott and Norstrom 1998; Roe 2004). Concentrations of p,p' -DDE and total PCBs in bald eagle plasma are shown to be variable among the sites compared

in Canada and the United States (Table 2). Santa Catalina Island has the highest p,p' -DDE concentration of the five areas compared; however, Michigan (specifically the Great Lakes area) has the highest reported total PCB concentration (Table 2). Both Santa Catalina Island and the Great Lakes region are considered hot spots for these chemicals due to past contamination (Garcelon et al. 1989; Roe 2004).

In most cases, the site-specific patterns of contaminant concentrations in nestling bald eagle plasma samples match patterns of land-use development, the degree of urbanization, and the level of industrial development at each of the sites sampled. Santa Catalina Island had the highest mean p,p' -DDE level, which is consistent with the known source of contamination in that area (Garcelon et al. 1989; Lee 1994). Between 1947 and 1961, an estimated 37–53 million liters of DDT-contaminated acid sludge, containing 348–696 tons of DDT, was disposed of in an ocean dump site 16 km northwest of Santa Catalina Island. In addition, it was estimated that another 1800 tons of DDT and PCBs was discharged from the Joint Water Pollution Control Plant outfall, 3.3 km offshore of Palos Verdes Peninsula (Lee 1994). Surveys completed in 1992 and 1993 by the US Geological Survey found more than 100 tons of p,p' -DDT and 10 tons of PCBs remaining in the sediment on the ocean bottom of the Palos Verdes Shelf (Lee 1994). These levels are still environmentally significant and are affecting the survival and breeding of wildlife in the area, especially bald eagles (Sharpe 2006).

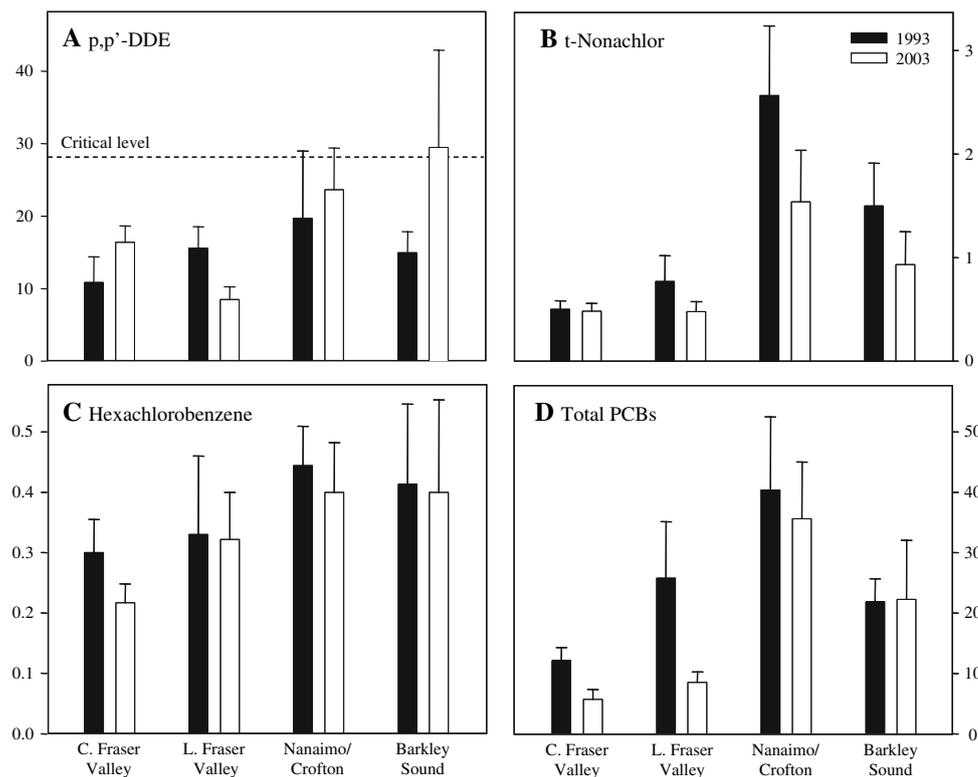
Elliott and Harris (2001/2002) derived a critical level of p,p' -DDE and total PCBs in bald eagle plasma that was based on declines in productivity below the minimal sustainable rate (0.7 young per occupied nest). The critical levels were developed using egg:plasma regressions for p,p' -DDE and total PCBs. For p,p' -DDE, the critical level was found to be 27.8 $\mu\text{g}/\text{kg}$, and for total PCBs, it was found to be 189 $\mu\text{g}/\text{kg}$. Recently, Strause et al. (2007) developed egg:plasma regressions for p,p' -DDE and total PCBs based on careful analysis of a broader range of comparative data using individual egg to plasma comparisons. Their p,p' -DDE regression has a similar slope, but the intercept was smaller than the relation produced by Elliott and Harris (2001/2002); therefore, for a given plasma value, a lower egg value would be predicted. For total PCBs, the regression has a steeper slope and the intercept is considerably lower. The equations derived in Strause et al. (2007) suggest that egg concentrations of p,p' -DDE would not exceed an effect level, whereas predicted PCB egg concentrations would be even lower than predicted by the equation in Elliott and Harris (2001/2002).

On Santa Catalina Island, two of the three eagles measured in 2003 exceeded the p,p' -DDE threshold derived by Elliott and Harris (2001/2002), and one eagle by more than

Table 1 Geometric means and range of contaminants, as well as mean \pm SD and range of total lipids measured in bald eagle nestling blood plasma from sites on the Pacific coast of North America, 2003

Location	n	Residue levels ($\mu\text{g}/\text{kg}$ ww)									
		Total lipids (%)	<i>p,p'</i> -DDE	HCB	<i>Trans</i> -nonachlor	Total PCBs	PCB 99	PCB 118	PCB 153	PCB 138	PCB 180
Fort St. James area	5	0.71 \pm 0.11	3.00	0.07	0.05	2.64	0.10	0.05	0.36	0.05	0.18
		0.53–1.02	0.7–8.0	0.05–0.1	0.05–0.05	1.0–4.0	0.05–0.2	0.05–0.05	0.1–0.7	0.05–0.05	0.1–0.3
Central Fraser Valley	6	0.87 \pm 0.11	15.66	0.20	0.46	4.51	0.09	0.36	0.84	0.66	0.17
		0.47–1.10	11.4–25.7	0.1–0.3	0.3–0.8	1.5–12.7	0.05–0.2	0.05–1.5	0.4–2.1	0.5–2.0	0.05–0.6
Lower Fraser Valley	9	1.08 \pm 0.08	13.56	0.26	0.40	7.11	0.27	1.53	1.35	0.92	0.64
		0.82–1.60	5.4–44.5	0.1–0.7	0.2–0.9	2.8–18.6	0.1–0.8	0.7–3.4	0.5–3.0	0.4–2.3	0.4–1.3
Nanaimo/Crofton Area	8	1.03 \pm 0.07	19.12	0.35	1.33	28.87	0.78	5.04	5.27	3.55	2.40
		0.80–1.29	4.5–57.7	0.2–0.8	0.3–5.2	7.6–97.0	0.2–2.4	2.1–12.2	1.2–18.0	0.7–12.6	0.7–9.0
Barkley Sound	3	1.00 \pm 0.12	18.38	0.31	0.78	15.19	0.48	1.99	3.18	2.43	1.50
		0.79–1.21	3.5–48.2	0.1–0.6	0.3–1.3	3.6–36.6	0.1–1.1	0.5–4.4	0.8–7.6	0.7–5.4	0.4–3.5
Santa Catalina Island	3	0.65 \pm 0.07	41.25	0.08	0.52	10.66	0.39	2.24	2.02	1.49	0.62
		0.53–0.78	18.0–123.4	0.05–0.1	0.3–0.8	6.5–21.9	0.2–1.0	1.4–4.2	1.4–4.2	1.0–3.0	0.4–1.2

Fig. 3 Residue levels of mean *p,p'*-DDE (A), mean *trans*-nonachlor (B), mean HCB (C), and mean total PCBs (D) in plasma samples of bald eagle nestlings collected at sites in British Columbia in 1993 and 2003. All residue levels are presented in micrograms per kilogram wet weight. Dotted line on graph A indicates the critical level (27.8 $\mu\text{g}/\text{kg}$) of *p,p'*-DDE based on declines in productivity below the minimum sustainable rate (derived in Elliott and Harris, 2001/2002). Chemicals measured in 1993 are from Elliott and Norstrom (1998)



fourfold. Using the regression of Strause et al. (2007), the mean predicted egg concentration of *p,p'*-DDE on Santa Catalina Island would be less than published effect levels, which is not consistent with the poor reproductive success and poor shell quality for bald eagles on the island. One of the three samples would exceed any predicted egg concentration; however, the sample is too small to make a confident inference, particularly given the considerable

within-clutch and between-clutch variability in chlorinated hydrocarbon concentrations reported for raptors (Strause et al. 2007).

The Fort St. James area in northern BC had the lowest level of contamination, most likely due to its remote inland location and limited industry in the area. However, we found that Barkley Sound had high levels of contaminants (especially *p,p'*-DDE), despite being fairly remote and

Table 2 Range of geometric means of *p,p'*-DDE and total PCBs in the plasma of bald eagle nestlings from a number of studies across North America

Contaminant ($\mu\text{g}/\text{kg}$)	Study location and year(s)				
	British Columbia (2001–2003) ^a ($n = 31$)	Santa Catalina Island, California (2003) ^a ($n = 3$)	Michigan (1999–2002) ^b ($n = 398$)	Ohio (1994–1997) ^b ($n = 147$)	Newfoundland (1996–1997) ^c ($n = 23$)
<i>p,p'</i> -DDE	3–19	41	4–37	3–6	2–9
Total PCBs	3–29	11	88–199	7–69	10–30

^a Current study^b Data from Roe (2004)^c Data from Dominguez et al. (2003)

away from any major urban and industrial development. There is some industrial forest industry activity in Port Alberni at the head of the inlet, but only relatively minimal increases in polychlorinated dioxin (PCDD) and furan (PCDF) contamination was found previously in bald eagle plasma and egg samples in Barkley Sound (Elliott and Norstrom 1998; Elliott et al. 1996b).

The Nanaimo/Crofton area showed high levels of PCBs, which is consistent with previous studies (Elliott and Harris 2001/2002; Elliott et al. 1998b). The southeastern portion of Vancouver Island, including the coastal area between Nanaimo and Crofton has had a considerable forest industry presence including lumber sorting yards, sawmills, and two large pulp and paper mill complexes. Discharge from those industries have been linked to releases of PCDDs and PCDFs into the local marine environment and bioaccumulation associated effects in fish-eating birds (Elliott et al. 1996b, 1998a). However, there has not been evidence of any substantial PCB inputs to the Nanaimo/Crofton area. Earlier surveys of chlorinated hydrocarbon contaminants in eggs and plasma samples of bald eagles did find that PCB concentrations were higher in samples from the Nanaimo/Crofton area, but also from the opposing mainland coast at Powell River (Elliott and Norstrom 1998; Elliott et al. 1996c). This may reflect some localized contamination inputs, however we believe that diet and food chain variation also plays an important role in determining the spatial contaminant patterns that we have measured here and in previous studies. It has previously been shown that patterns of PCB congeners in eggs of bald eagles varied among sites on the BC coast, which was attributed in part to dietary variation (Elliott et al. 1996c). In a related study, Weech et al. (2006) found a significant relationship between stable nitrogen isotope ratio ($\delta^{15}\text{N}$) and mercury in plasma of adult bald eagles from BC. Further work on the role of diet and trophic levels as an influence on contaminant concentrations in eagles is planned. This information will assist in explaining difference in contaminant concentrations and in determining the sources of contamination.

Temporal changes showed that the level of total PCBs and *trans*-nonachlor decreased in BC between 1993 and 2003 (data were pooled from the four BC sites), but the levels of *p,p'*-DDE and hexachlorobenzene did not. The greatest decrease in total PCBs and *trans*-nonachlor was in central Fraser Valley and the Nanaimo/Crofton area, respectively.

Some eagles sampled in 2003 in the Nanaimo/Crofton area and in Barkley Sound exceeded the *p,p'*-DDE threshold derived by Elliott and Harris (2001/2002), and there are likely to be some ongoing effects on egg shell quality, nest success, and thyroid hormone systems (Cesh 2005). During the early 1990s, the only contaminant measured that showed a significant association with nest success in BC coastal bald eagles was *p,p'*-DDE (Elliott and Norstrom 1998). Concentrations for total PCBs in 2003 were all below the critical level (Elliott and Harris 2001/2002). Bald eagles in BC have been well below the critical level for PCBs since 1993; therefore, this chemical is not likely to be having an effect on the population.

Overall, the levels of contaminants measured at the BC sites have not significantly changed from 1993 to 2003. Nevertheless, all sites (including remote locations) have detectable amounts of the measured contaminants, demonstrating the persistence of these chemicals and their ability to be transported long distances in the environment.

In conclusion, long-term monitoring of environmental contaminants continues to provide important information on the health of bald eagles, status of the local environments, and changes in environmental contaminant concentrations.

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