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Organochlorine Compounds in the Plasma of Peregrine Falcons and Gyrfalcons Nesting in Greenland

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ABSTRACT. Levels of organochlorine compounds in the blood plasma of after-second-year female peregrine falcons (*Falco peregrinus*) were determined from samples collected from southern Greenland in 1985 and western Greenland from 1983 to 1989, and from adult and nestling gyrfalcons (*Falco rusticolus*) from western Greenland in 1989 and 1990. Samples were taken during nesting. Levels of p,p'-DDE (DDE) in peregrine plasma were 140 µg/kg wet weight geometric mean (*GM*) for the southern samples, and 220 µg/kg *GM* for the western samples. Calculated levels of DDE for eggs from plasma levels are below those associated with declining peregrine populations. Only DDE was found in the gyrfalcon plasma; levels were below 20 µg/kg. In addition to DDE, other organochlorines quantified in peregrine plasma included p,p'-DDD, p,p'-DDT, polychlorinated biphenyls, six chlordane compounds, β-HCH, HCB, and Mirex. There was no trend over time for any of the compounds reported in peregrines except HCB, which decreased over the study period. There was no significant difference in the levels of the compounds reported between the regions. Females, which were sampled more than once, showed no clear trends with respect to increasing or decreasing residues.

Key words: peregrine falcon, gyrfalcon, DDE, organochlorine, blood plasma, Greenland

RÉSUMÉ. On a déterminé le niveau des composés organochlorés dans le plasma sanguin de faucons pèlerins (*Falco peregrinus*) femelles ayant plus de deux ans à partir d'échantillons provenant du Groenland méridional en 1985 et du Groenland occidental de 1983 à 1990, ainsi que de faucons gerfauts (*Falco rusticolus*) adultes et au nid prélevés dans le Groenland occidental en 1989 et 1990. Les échantillons ont été prélevés durant la période de nidification. Les niveaux de p,p'-DDE (DDE) dans le plasma des faucons pèlerins avaient une moyenne géométrique de 140 µg/kg de poids frais pour les échantillons provenant de la zone méridionale, et de 220 µg/kg pour les échantillons provenant de la zone occidentale. Les niveaux de DDE pour les oeufs, calculés à partir des niveaux de plasma, sont inférieurs à ceux que l'on associe avec les populations de faucons pèlerins en déclin. Seul le DDE a été trouvé dans le plasma du faucon gerfaut; les niveaux étaient inférieurs à 20 µg/kg. En plus du DDE, les autres organochlorés que l'on a quantifiés dans le plasma du faucon pèlerin comprenaient le p,p'-DDD, p,p'-DDT, les polychlorés biphényls, six composés du chlordane, le β-HCH, le HCB et le mirex. On n'a décelé de tendance pour aucun des composés trouvés dans les faucons pèlerins, sauf le HCB qui a diminué au cours de la période d'étude. On n'a noté aucune différence significative dans les niveaux des composés rapportés entre les diverses zones. Les femelles, qui ont fourni des échantillons plus d'une fois, n'ont pas montré de tendance nette en ce qui concerne une augmentation ou une diminution des résidus.

Mots clés: faucon pèlerin, faucon gerfaut, DDE, organochloré, plasma sanguin, Groenland

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INTRODUCTION

Organochlorine insecticides (e.g., DDT and Dieldrin) were introduced into the environment in large quantities beginning in the 1940s (Ware, 1989). By the early 1960s dramatic declines in certain bird populations had been noted, particularly in fish-eating birds and raptors (review in Risebrough, 1985). Reports of the disappearance of peregrine falcons (*Falco peregrinus*) in Great Britain (Ratcliffe, 1963), along with the observation of declining peregrine numbers in the eastern United States (Berger et al., 1969), led to an international conference in Madison, Wisconsin in 1965, which examined peregrine populations worldwide (Hickey, 1969). Shortly after this conference, the relationship between declining populations and eggshell thinning in falcons and fish-eating birds was established (Ratcliffe, 1967), and the correlation between eggshell thinning and residues of DDE (a metabolite of p,p'-DDT) was confirmed (Hickey and Anderson, 1968).

Some of the first studies documenting the decline of peregrine falcons, eggshell thinning, and its relationship to DDE were in the northern populations of Alaska and Canada (Cade et al., 1968, 1971; Berger et al., 1970; Fyfe et al., 1976). Alaskan arctic falcon populations experienced a significant decline (Cade et al., 1971), but are currently experiencing a population recovery (Kiff, 1988). The reports of the decline of Alaskan and Canadian peregrines provided an impetus to study falcon populations, eggshell thinning, and pollutants in other northern areas such as Norway (Nygård, 1983), Finland (Lindén et al., 1984), and Greenland (Walker et al., 1973; Burnham and Mattox, 1984; Springer et al., 1986; Mattox and Seegar, 1988). Arctic populations

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of peregrines still have relatively high organochlorine levels and low productivity in southern Finland (Lindén et al., 1984) and Russia (Henny et al., in press; Potapov, in press).

Although DDE levels and the associated eggshell thinning in peregrines reached critical levels in Greenland during the 1970s (Walker et al., 1973), no population decline was documented (Mattox and Seegar, 1988). More recent data from Greenland indicate a decrease in organochlorine levels, thicker eggshells, and a stable and healthy population (Mattox and Seegar, 1988). We evaluated the levels, trends, and patterns of organochlorine contaminants in plasma samples collected from Greenland peregrine falcons between 1983 and 1989, and from adult and nestling gyrfalcons (*Falco rusticolus*) from 1989 to 1990, to determine the possible sources and impact of these compounds on the populations.

Plasma was an ideal medium for this analysis because blood (whole blood, plasma, and serum) has been used to monitor organochlorine contaminants in a variety of avian species (Henny and Meeker, 1981; Henny et al., 1982, 1989; Court et al., 1990; Mora et al., 1993), and has been validated as an accurate measure of body levels of organochlorine contaminants in humans (Burse et al., 1980, 1989; Mes et al., 1992), rats (Mohammed et al., 1990), and birds (Henny and Meeker, 1981; Wiemeyer et al., 1984; Clark et al., 1987; Court et al., 1990). In this study blood plasma was used to determine organochlorine levels in peregrines and gyrfalcons. These levels were then used to predict the corresponding levels in the eggs using equations developed by Henny and Meeker (1981) and Court et al. (1990) for comparison to previously reported levels of organochlorine compounds in peregrines from the Arctic.

MATERIALS AND METHODS

Plasma samples were collected during a long-term population study of falcons in Greenland that began in 1972 (Burnham and Mattox, 1984; Falk et al., 1986; Falk and Møller, 1988; Mattox and Seegar, 1988). Results from two sampling areas, West Greenland and South Greenland (Fig. 1), are presented in this paper. A complete description of the individual study areas can be found in Falk et al. (1986) for the southern region, and Mattox and Seegar (1988) for the western region.

Falcons were captured and banded at the nest after egg-laying. A sample of approximately 2 ml of blood was taken from the brachial vein; 0.25 ml of this sample was placed in a buffered solution for subsequent DNA analysis (Longmire, 1988) and the remainder was centrifuged to remove the red blood cells. The plasma portion was used in the chemical analysis for organochlorine residues.

A complete description of the chemical analysis can be found in Jarman et al. (1993a). Briefly the method is as follows:

The plasma sample (approximately 0.5–1 ml) was ground with anhydrous sodium sulfate until free-flowing. The sample vial was weighed before and after the sample was removed to determine wet weight.

Samples collected prior to 1989 were extracted for six hours with 125 mls of methylene chloride in a soxhlet apparatus. Samples collected in 1989 and 1990 were extracted in a glass extraction column by adding 20 ml of 1:1 (v:v) hexane:methylene chloride, followed by elution with an additional 100 ml of 1:1 (v:v) hexane:methylene chloride. The sample was transferred to a kilned, solvent-rinsed, and pre-weighed 4 ml glass vial, evaporated under nitrogen gas to dryness, and weighed on an analytical balance to determine lipid weight.

The lipid extract was applied to a chromatographic glass column packed with 0.5% water-deactivated Florisil®. Two fractions were then eluted successively: fraction 1 contained HCB, Mirex, polychlorinated biphenyls (PCBs), o,p'-DDT (50%), o,p'-DDE, and p,p'-DDE, and fraction 2 contained the HCH compounds, chlordane compounds and metabolites, p,p'-DDD, p,p'-DDT, o,p'-DDT, Toxaphene, heptachlor epoxide, and Dieldrin.

Instrumental analyses were performed using a Hewlett-Packard 5890A gas chromatograph equipped with ^{63}Ni electron capture detector and a 60 m DB-5 column (0.25 mm inside diameter, 0.25 µm film thickness, fused silica capillary column, J&W Scientific). Levels of all compounds are reported as µg/kg (parts per billion, ppb) of the wet weight. PCBs were quantified as individual congeners (n = 12); total PCBs ($\sum$-PCBs) is the sum of these congeners.

The data were log transformed for statistical analysis. Differences among data sets were considered significant when the probability value (p) was < 0.05. Statistical analysis was performed on a IBM-compatible microcomputer using the software Minitab (Ryan et al., 1985).

RESULTS AND DISCUSSION

It has been reported that to obtain accurate lipid measurements from blood, 50 ml of blood must be used (Mes et al., 1992). In wildlife samples this volume is rarely obtainable. In birds, sample volumes of blood are usually less than 10 ml. Additionally, the concentration and composition of lipids in avian blood vary with stress, egg-laying, and diet (i.e., lipid composition of prey and time of last feeding) (De Freitas and Norstrom, 1974; Henny and Meeker, 1981; Anderson et al., 1984; Clark et al., 1987). In order to investigate the precision of our lipid extraction of small samples (between 0.5 and 1 ml), the total lipid extracted (between 0.0012 and 0.0042 g) and the wet weight of the samples were correlated. There was a significant correlation ($n = 50$, $r = 0.62$, $p < 0.05$) between the lipid amounts and the wet weights; therefore, even with small samples of blood, the efficiency of lipid extraction from the blood was consistent.

All of the compounds reported in this study are highly lipophilic and are associated with lipoproteins in the plasma (Clark et al., 1987; Mes et al., 1992). Because of the small amount of lipid extracted, it was important to determine whether the concentrations of organochlorines were dependent on the total lipid extracted (e.g., a small amount of extracted lipid would have a low concentration of DDE in the wet weight plasma). Since there was no correlation between the amount of lipid extracted and the DDE concentration ($n = 50$, $r = 0.25$, $p > 0.05$), the residues reported were not dependent on the sample size.

DDE was found in every after-second-year (ASY) female

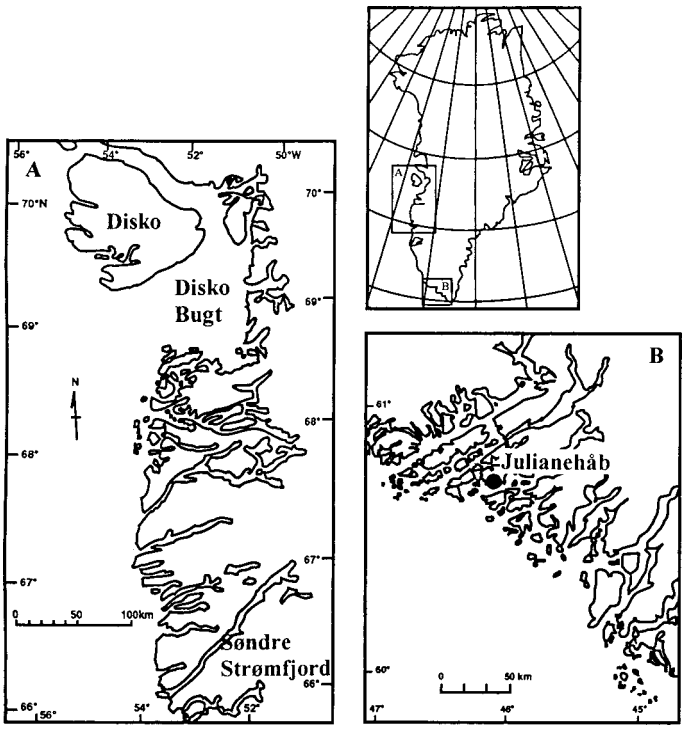

FIG. 1. General sample collection sites. West Greenland: peregrines, n = 41; gyrfalcons, n = 4. South Greenland: peregrines, n = 5

peregrine plasma sample analyzed; the levels were 140 µg/kg geometric mean (*GM*) in the southern birds, and 220 µg/kg *GM* in the western samples (Table 1). These levels are approximately one-third of those reported in the plasma of peregrines sampled after breeding in the Northwest Territories of Canada (Court et al., 1990), and approximately one-half of those found in the plasma of peregrines trapped during fall migration (Henny et al., 1982). All ASY females reported here were sampled after egg-laying. Both Court et al. (1990) and Henny and Meeker (1981) found lower levels of residues in the plasma of post-laying birds; so the levels reported here are probably an underestimate of the pre-laying levels.

Court et al. (1990) and Henny and Meeker (1981) provide equations to predict concentrations of DDE in eggs from the blood plasma levels (Table 2). Using the geometric mean levels of DDE in the plasma of the western and southern Greenland samples, the calculated egg levels using Court et al. (1990) would be 2600 µg/kg and 1800 µg/kg respectively. Using a theoretical concentration of DDE in the eggs of 20 000 µg/kg (a critical level associated with population declines, Peakall et al., 1975), critical blood levels for DDE (using Court et al., 1990) would be 3200 µg/kg; this is more than an order of magnitude higher than the mean levels found in this study.

Walker et al. (1973) reported levels of 18 000 and 15 000 µg/kg (assuming 5% lipid in the eggs) of DDE in the eggs of peregrines collected in western Greenland in 1972. Burnham and Mattox (1984) reported a mean DDE level of 14 300 µg/kg wet weight (n = 8) in eggs from western Greenland collected between 1972 and 1978. Springer et al. (1986) found DDE levels of 12 000 µg/kg (n = 7, arithmetic mean, assuming 80% water) in the eggs collected between 1978 and 1979. Mattox and Seegar (1988) reported a highest DDE level of 9100 µg/kg wet weight for eggs collected in 1981 and 1982. The highest calculated levels of DDE for eggs in this report are not as high as levels reported previously, so it appears that levels of DDE in Greenland peregrines have decreased since 1980.

TABLE 1. Levels of organochlorine compounds in ASY female peregrine falcon plasma from Greenland. Concentrations reported as µg/kg (ppb) in wet weight.

	South Greenland			West Greenland		
	Geometric mean	Range ± 1 SD	n[1]	Geometric mean	Range ± 1 SD	n[1]
p,p'-DDE	140	100–190	5	220	63–790	41
p,p'-DDT	5.8	3.6–9.3	5	4.9	1.1–23	31
p,p'-DDD	1.8	1.8–2.8	3	3.5	1.4–9.1	13
β-HCH	2.8	1.9–4.1	5	2.6	0.72–9.5	34
oxychlordane	26	12–57	5	18	7.6–43	37
heptachlor epoxide	31	14–71	5	17	4.5–68	40
MC-2	3.5	1.4–8.9	5	2.8	1.3–6.1	19
MC-5	6.2	2.4–16	5	4.8	1.6–14	22
trans-nonachlor	3.3	1.5–7.4	5	3.2	1.2–8.2	34
cis-nonachlor	2.4	0.78–7.6	5	2.4	0.94–6.2	17
HCB	5.3	4.1–6.9	5	3.9	1.5–10	34
Mirex	18	9.1–37	5	10	3.8–27	39
PCBs						
99	14	7.8–25	5	6.9	2.2–22	40
118	12	6.6–21	5	6.7	1.9–24	36
153	120	63–220	5	47	14–150	40
138	30	16–57	5	5	5.1–44	40
187	18	9.1–37	5	8.5	2.7–26	35
180	43	23–82	5	17	5.9–51	40
170,190	18	9.5–35	5	8.2	2.9–23	40
201	6.8	3.1–15	5	4.0	1.5–11	40
196,203	12	5.4–25	5	5.0	1.9–13	39
195	4.7	2.1–11	5	2.2	0.79–6.1	36
194	8.5	4.2–17	5	3.3	1.2–6.1	39
206	8.0	2.9–22	5	3.0	0.94–9.3	37
Σ–PCBs	300	160–570	5	130	43–380	40

[1] n is the number of samples that contained residues above the detection limit (DDE, PCBs, and Mirex 3.0 µg/kg, all other compounds 1.0 µg/kg).

TABLE 2. Predicted levels of DDE in the eggs of peregrine falcons from the blood plasma levels. Both equations based on post egg-laying. Concentrations reported as µg/kg (ppb) of the wet weight.

	South Greenland		West Greenland	
	Geometric mean	Range ± 1 SD	Geometric mean	Range ± 1 SD
DDE	140	100–190	220	63–790
Calculated level in eggs				
Court et al. 1990[1]	1800	1400–2300	2600	890–7700
Henny and Meeker 1982[2]	1000	760–1400	1700	450–6200

[1] Log DDE in egg contents = 9.723 + 0.8506 (log DDE in plasma)

[2] DDE in egg contents = 6.243 (DDE in plasma) $^{1.0033}$

Levels of Σ-PCBs (the sum of the individual PCB congeners) in the plasma were 300 and 130 µg/kg *GM* for southern Greenland and western Greenland respectively (Table 1). Henny et al. (1982) reported generally low levels of PCBs in peregrine

plasma, but did not report actual values. Court et al. (1990) found mean levels of PCBs at 530 μg/kg *GM*. Levels of Σ-PCBs in birds are generally a factor of two lower than values reported for PCBs in birds when quantified by Aroclor mixtures (Turle et al., 1991). This observation would make the values between this study and Court et al. (1990) similar. Profiles of the PCB congeners in Greenland are similar to those reported in falcon eggs from California (Jarman et al., 1993b), probably indicating a similar pattern of accumulation and elimination rather than similar sources.

Court et al. (1990) also reported levels of heptachlor epoxide of 40 μg/kg *GM*, whereas the Greenland peregrines had 31 and 17 μg/kg *GM* for southern and western Greenland respectively. No other chlordanes were reported by Court et al. (1990). The relatively high levels of the chlordane compounds MC-2 and MC-5 in Greenland peregrine plasma are similar to those of other falcons (Jarman et al., 1993a), and as with the PCBs suggest a similar pattern of accumulation.

DDE was detected in three of the four gyrfalcon plasma samples analyzed, at levels below 20 μg/kg. At detection limits of 3.0 μg/kg for DDE, PCBs, and Mirex, and 1.0 μg/kg for all other compounds, no other compounds were detected in any of the gyrfalcons. These remarkably low levels probably reflect the generally non-migratory nature of both the gyrfalcon and its main prey, ptarmigan and hare (Platt, 1976), and the low trophic level of these prey species (Nielsen and Cade, 1990). Low organochlorine levels in gyrfalcons also indicate the relatively minor contribution from consumption of contaminated migratory prey and the lack of atmospheric contaminants in the gyrfalcons' main prey. The majority of contaminants in the peregrines probably originate from the consumption of contaminated, migratory prey on wintering and breeding grounds. This conclusion is supported by the work of Henny et al. (1982), who found that peregrines returning from wintering grounds in South America had higher levels of pesticides than those migrating from their breeding grounds.

Other organochlorine compounds found in the plasma of ASY female peregrines include p,p'-DDD, p,p'-DDT, six chlordane compounds, β-HCH, HCB, and Mirex (Table 1). The compounds Σ-DDT (DDE, DDD, and DDT), Σ-PCBs, Σ-chlordane (oxychlordane, heptachlor epoxide, MC-2, MC-5, *trans*- and *cis*-nonachlor), β-HCH, and HCB in peregrine plasma were examined for any increasing or decreasing trend over the years 1984–86 and 1988–89 (1983 was excluded because of the small sample size). The only significant trend found (ANOVA $p > 0.05$) was for HCB, which decreased during this period (ANOVA $F = 11.51$, $p < 0.05$). Also, six female peregrines were sampled more than once (Fig. 2). No trends over time can be

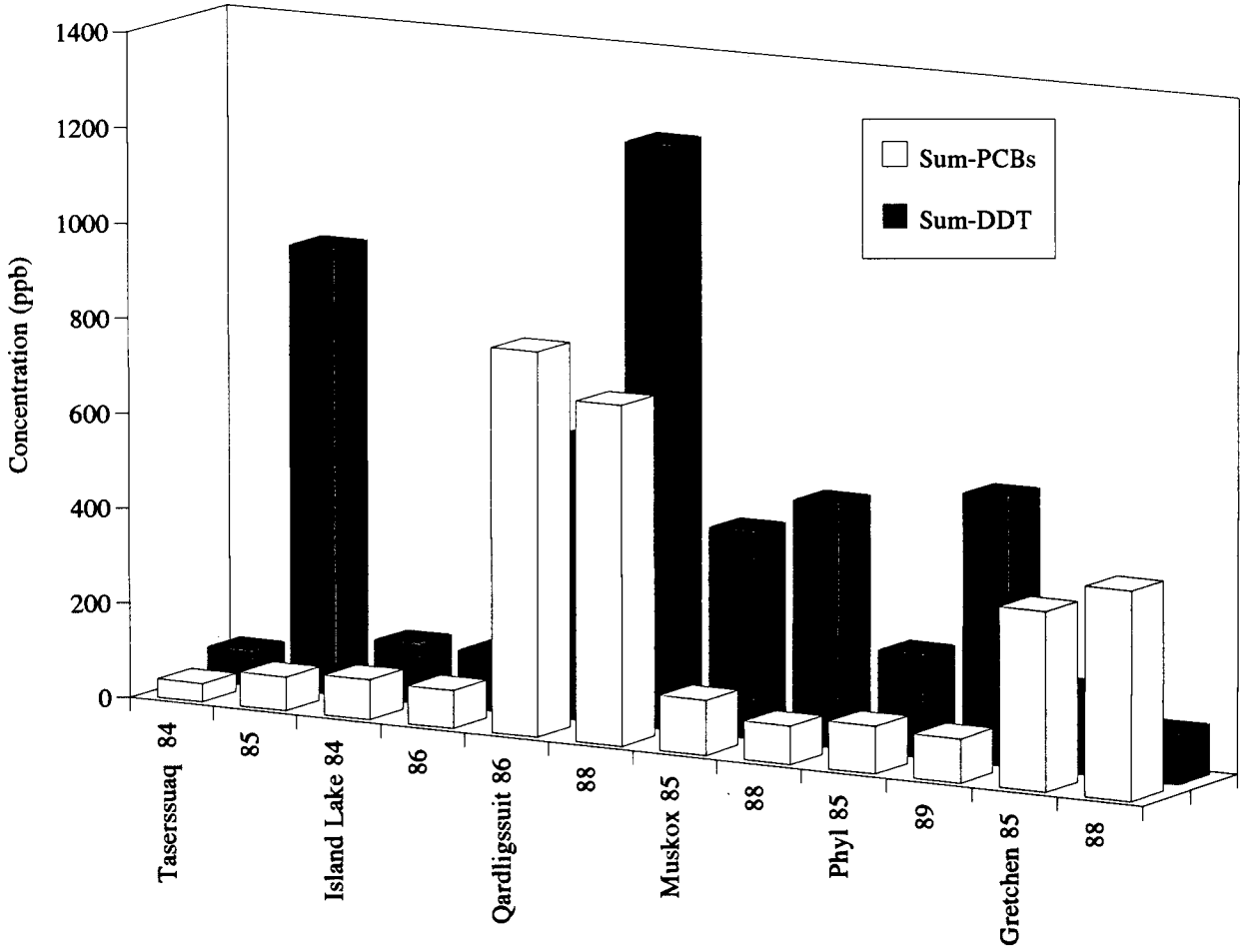

FIG. 2. DDE and PCB levels in six after-second-year-females sampled more than once. Concentrations reported in μg/kg wet weight.

observed for either of the major compounds present in the females (∑-DDT or ∑-PCBs) or for increasing or decreasing residues. Individual females, in general, tended to have constant relative proportions of ∑-DDT to ∑-PCBs over time.

When data were analyzed for differences between residues in peregrine samples from southern and western regions, no concentrations were found to be statistically different. Interestingly, DDE was higher in the western birds, while PCBs were higher in the southern birds. Although not statistically significant, these different concentrations might reflect a different prey base or different migratory or wintering habits of these peregrine falcon populations.

The ratio of DDT and DDD to DDE has been used in Alaskan and Greenland peregrine falcon eggs collected between 1977 and 1980 to determine possible differences in exposure to organochlorine contaminants (Springer et al., 1986). The ratios of DDT and DDD to DDE in the Alaskan and Greenland eggs were 0.9 and 1.4 respectively. The ratios for these same compounds in the plasma for the southern and western samples in this study were 5.4 and 3.8 respectively. According to Springer et al. (1986), higher ratios indicate relatively recent consumption of the parent compound DDT, exposure to recently applied DDT on the wintering grounds, or the consumption of migratory prey, or both. The high ratios in the Greenland birds we studied might also be explained by the differential partitioning of DDT and DDE into the plasma (De Freitas and Norstrom, 1974; Clark et al., 1987).

CONCLUSIONS

DDE levels in Greenland peregrine falcons have shown a decline beginning around 1980 to levels that are well below those associated with reproductive problems. This conclusion agrees with productivity data that show a healthy, increasing population of falcons in Greenland (Mattox and Seegar, 1988). However, levels of DDE in the plasma of Greenland peregrine falcons over the years 1984–86 and 1988–89 show no decline, probably reflecting the continued exposure to organochlorine compounds on the wintering ground and from migratory prey. Gyrfalcons had low levels of all organochlorine compounds (< 20 μg/kg). The similar patterns of organochlorine contamination (i.e., PCB and chlordane profiles) in peregrines from different regions (southern and western Greenland, California, Colorado, and the east coast of the United States) probably indicate a similar mechanism of accumulation. This study has shown that blood (even in small volumes) can be a good medium for monitoring organochlorine contamination in remote populations of wildlife.

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