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DRAMATIC DECLINES OF DDE AND OTHER ORGANOCHLORINES IN SPRING MIGRANT PEREGRINE FALCONS FROM PADRE ISLAND, TEXAS, 1978–2004

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ABSTRACT.—Peregrine Falcons (*Falco peregrinus*) captured in the spring at Padre Island, Texas, nest across the arctic and subarctic from Alaska to Greenland and winter throughout Latin America. Padre Island, located immediately north of the Mexican border, is the peregrines' first landfall in the U.S.A. after spending about 6 mo in Latin America. Blood plasma was collected from spring migrants at Padre Island between 1978 and 2004 to monitor trends in organochlorine (OC) pesticides and their metabolites. Geometric mean concentrations of p,p′-DDE (μg/g, ww) decreased throughout the study: 1978–1979 (0.879), 1980 (0.617), 1984 (0.551), 1994 (0.406) and 2004 (0.013). Most other OC pesticides, with detection limits used during the earlier portion of this study, were no longer detected during the last two sampling periods. The reduced concentrations of OC pesticides suggest that other pesticides (including carbamates, organophosphates and pyrethroids) are likely being used as replacements. These replacement compounds are not as persistent and cannot be readily evaluated at migration sites like Padre Island. However, concentrations of flame retardants (polybrominated diphenyl ethers; PBDEs) have recently increased in bird eggs in many regions and have been reported in blood plasma. Concentrations of PBDEs in peregrine plasma could be evaluated at Padre Island for assessment of trends in the Americas.

KEY WORDS: *Peregrine Falcon*; Falco peregrinus; *contaminants*; *DDE*; *DDT*; *migration*; *monitoring*; *Texas*.

DISMINUCIONES DRAMÁTICAS DE DDE Y OTROS ORGANOCLORADOS EN LOS HALCONES PEREGRINOS MIGRANTES DE PRIMAVERA DE LA ISLA PADRE, TEXAS, 1978–2004

RESUMEN.—Los halcones peregrinos (*Falco peregrinus*) capturados en primavera en la isla Padre, Texas, anidan a lo largo del Ártico y del Subártico desde Alaska a Groenlandia, e invernan a lo largo de América Latina. La isla Padre, ubicada inmediatamente al norte de la frontera con México, es el primer sitio de parada de los halcones peregrinos en EEUU luego de una estadía de cerca de seis meses en América Latina. Se colectó plasma sanguíneo de individuos migrantes de primavera en la isla Padre entre 1978 y 2004 para monitorear las tendencias en pesticidas organoclorados (OC) y sus metabolitos. Las concentraciones geométricas medias de p,p′-DDE (μg/g, peso a peso) disminuyeron a lo largo del estudio: 1978–1979 (0.879), 1980 (0.617), 1984 (0.551), 1994 (0.406) y 2004 (0.013). La mayoría de los otros pesticidas OC, con límites de detección usados durantes la porción inicial de este estudio, no fueron detectados de nuevo durante los dos últimos períodos de muestreo. Las concentraciones reducidas de pesticidas OC sugieren que otros pesticidas (incluyendo carbamatos, organofosfatos y piretroides) están siendo probablemente usados como reemplazantes. Estos compuestos de reemplazo no son tan persistentes y no pueden ser evaluados fácilmente en los sitios de migración como isla Padre. Sin embargo, las concentraciones de retardadores de llama (éteres difenilo polibrominados; PBDEs por sus siglas en inglés) han incrementado

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recientemente en los huevos de las aves en muchas regiones y han sido encontrados en el plasma sanguíneo. Las concentraciones de PBDEs en el plasma de los halcones peregrinos podrían ser estudiadas en la isla Padre para evaluar las tendencias en las Américas.

[Traducción del equipo editorial]

Peregrine Falcon (*Falco peregrinus*) populations throughout much of the world declined following the widespread use of DDT in the late 1940s, primarily due to eggshell thinning and poor reproductive success (Hickey 1969, Ratcliffe 1980). The use of DDT in the United States was banned in 1972; however, by that time *F. p. anatum* was extirpated from the eastern United States (Berger et al. 1969) and the highly migratory *F. p. anatum* and *F. p. tundrius* from the arctic and subarctic range of species were at low numbers (see summary, Kiff 1988). Peregrines were still observed at autumn migration sites along the Atlantic and Gulf Coasts (e.g., Ward et al. 1988). One such site is Padre Island, Texas (Hunt and Ward 1988). Systematic peregrine counts were made at autumn migration sites to assess population changes over time, with many birds captured and banded. In 1976, autumn-captured peregrines were first sampled and their plasma analyzed for DDT, DDE (the most persistent metabolite of DDT) and other organochlorine (OC) contaminants at Assateague Island, Maryland/Virginia and Padre Island, Texas by Henny et al. (1982). The peregrines captured on both Padre Island and Assateague Island in the autumn were mostly young of the year (hatch year, HY) and only a few months old, with generally low pesticide concentrations. Therefore, older age classes were of more interest from a contaminant perspective.

A spring concentration of migrating peregrines was first discovered by F. Ward at Padre Island in April 1978 (Hunt and Ward 1988). These peregrines, after spending about six months wintering throughout Latin America (Yates et al. 1988), made their first landfall in the United States at this barrier island immediately north of the Mexico border. Thus, contaminant burdens in this long-distance migrant reflect the condition of the environment in both North and South America. Spring-captured peregrines at Padre Island included adults (after second year, ASY) and second year (SY) birds, both of contaminant interest. A limited number were captured in 1978 and 1979, but by 1980 procedures for locating and capturing the spring migrants had been refined, and large numbers of peregrines were sampled for contaminants.

Previous contaminant concentrations were published in a series of papers that documented accumulation of DDE on the wintering grounds (i.e., compared concentrations in autumn-captured HY birds vs. spring-captured SY birds; Henny et al. 1982), and contaminant concentration declines over time were reported more recently (Henny et al. 1988, 1996). The latter paper also reported nesting and wintering localities for the migrant peregrines based on satellite telemetry that supported earlier banding data indicating that spring-captured birds at Padre Island nested across the arctic and subarctic from Alaska to Greenland and wintered throughout Latin America.

We here report OC concentration changes between 1994 and 2004. During this interim, *F. p. tundrius* was removed from the Endangered Species list in 1994 (Federal Register 59:50796) and *F. p. anatum* in 1999 (Federal Register 64:46541), although continued monitoring for contaminants was required for both subspecies as part of the delisting process.

METHODS

Peregrine Falcons were captured throughout this long-term study using lures with noose jackets, as described by Ward and Berry (1972). We collected blood samples from the brachial veins of captured peregrines, and processed samples as described by Henny et al. (1982). The plasma samples from 1978–1979, 1980, 1984, and 1994 were analyzed at Patuxent Wildlife Research Center; details were described in Cromartie et al. (1975), Kaiser et al. (1980), and Henny et al. (1996). Polychlorinated biphenyls were estimated as Aroclors, but considered as not comparable over the time series, and excluded from this analysis. Concentrations of p,p′-DDE (DDE), p,p′-DDD (DDD), p,p′-DDT (DDT), dieldrin, heptachlor epoxide, oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, endrin, toxaphene, hexachlorobenzene, mirex, hexachlorocyclohexane (alpha, beta, and gamma isomers), o,p′-DDE, o,p′-DDD and o,p′-DDT were determined in 1994. The latter four were not evaluated in 1984 or earlier. The recovery of spiked samples during this time period averaged 88% in 1978–1980, 94% in 1984, and 95% in 1994 with no adjustment for recovery.

Plasma samples from 2004 were analyzed for OC pesticides at Mississippi State Chemical Lab according to SOP 1.260 (EPA Method 3545) for extraction procedures. SOP 1.255 and 1.265 was used for quantification of residues by megapore column electron capture gas chromatography (GC). Determinations were run on a Varian 3600 GC with a Varian Star Data System and a Varian 8200 Autosampler. The GC was equipped with dual DB-608 and DB-5 30 M megapore columns. All compounds were calculated using a three-point standard curve forced through the origin using external standards (SOP 1.267). The same OCs were analyzed in 2004 as in 1994. The recovery of spiked OC samples in 2004 averaged 81%, but with 70% for DDE, again with no initial adjustment made for recovery.

In 1979 and 1980, ASY female peregrines were captured on Padre Island between 5 April and 4 May (Henny et al. 1982). DDE concentrations showed little difference from 5 April–15 April, 16 April–25 April, and 26 April–4 May (geometric means 0.65, 0.75, and 0.66 μg/g, respectively). Blood plasma samples in 2004 were collected between 15–25 April, which was during the middle of the earlier sampling period. It is doubtful that the timing of capture had any affect on the contaminant concentrations reported.

Henny et al. (1982, 1988) reported concentrations of all OCs detected in earlier years. In this study, we summarize only those OCs occurring in at least 10% of the samples for any year. The lower limit for reportable residues for pesticides was 0.01 μg/g in 1994; however, for temporal contaminant comparisons we used 0.02 μg/g to be comparable with earlier datasets. The few sample non-detections (two of 156) between 1978 and 1994 were assigned a value of half the detection limit for DDE. A lower detection limit (0.002 μg/g) was obtained for 2004, but for the tabular data, we again used the 0.02 μg/g for direct comparisons. The exception was DDE, for which only seven of 27 samples contained ≥0.02 μg/g in 2004. For these DDE data, we used the lower detection limit and calculated a geometric mean with only one of 27 samples below the lowered detection limit. An analysis of variance (ANOVA) was used to compare the log-values of DDE residues among years. Differences found in the ANOVA were quantified with Tukey's studentized range test ($P \leq 0.05$) to compare means (SAS Institute 1999).

If a contaminant was detected, but did not occur above the detection limit in at least 75% of the samples, Fisher's exact test ($P \leq 0.05$) was used to compare occurrence during the years 1978–1979, 1980, 1984, and 1994 (previously reported in Henny et al. 1996). Again, as in 1994, none of the contaminants in 2004 which were listed earlier in this group (DDT, DDD, heptachlor epoxide, dieldrin, oxychlordane, mirex) were above the detection limit. Thus, no additional statistical tests were warranted, although 2004 information (number of non-detections) was summarized. All contaminant concentrations are presented on a wet weight (ww) basis.

RESULTS AND DISCUSSION

Temporal Trends in DDE Concentrations. In 1978–1994, both SY and ASY female peregrines were sampled at Padre Island (Henny et al. 1996), but sampling in 2004 was limited to ASY females. In early years, more ASY than SY peregrines were sampled, and, as an age class, ASY birds generally had greater DDE concentrations than SY birds. From 1978–1994, only two of 156 blood samples contained DDE concentrations below the detection limit (0.02 μg/g), whereas in 2004, only seven of 27 were above it. Geometric mean blood plasma DDE concentrations showed a decrease over time from 1978–1979 to 2004, with a 26% decrease from 1984 to 1994, and a 97% decrease from 1994 to 2004 (Table 1). The recent decrease was greater than anticipated, and even with an adjustment for the lower percent recovery of spiked samples in 2004 vs. 1994 (see Methods, 70 vs. 95% for DDE), the decrease in DDE concentrations was large (96%). However, this decrease in DDE was similar to changes in DDE levels in Osprey (*Pandion haliaetus*) eggs from the upper Willamette River, Oregon, between 1993 and 2006 (geometric mean 2.35 vs. 0.21 μg/g ww; 91% decrease; Henny et al. 2008).

The highest DDE concentration in 1984 (4.3 μg/g) was accompanied by the only detection of the parent compound DDT and another metabolite, DDD. In this sample, 14% of the total DDTs was DDT and DDD. This residue profile suggests that a small percentage of these peregrines (one in 27 of our 1984 samples) had been exposed to less-degraded mixtures of technical DDT, perhaps recently applied as an insecticide, on their wintering grounds, despite the general downward trend of the metabolite DDE in the remainder of the samples. No DDT or DDD was detected in samples collected in 1994, and only one sample (same bird) in 2004 (at the lower detection limit) contained p,p′-DDD (0.016 μg/g), o,p′-DDD (0.005 μg/g) and

Table 1. Concentrations of p,p′-DDE (μg/g, ww) in blood plasma of adult female Peregrine Falcons captured in the spring at Padre Island, Texas, 1978–2004.

YEAR	*N*	GEOMETRIC MEAN DDE (μg/g)	HIGH DDE (μg/g)	SOURCES
1978–1979	21	0.879 A[1]	3.8	Henny et al. (1982)
1980	63	0.617 AB	3.1	Henny et al. (1982)
1984	27	0.551 AB	4.3	Henny et al. (1988)
1994	45	0.406 B	2.4	Henny et al. (1996)
2004	27	0.013 C	0.067	This study

[1] Means in columns sharing a letter are not significantly different.

o,p′-DDE (0.002 μg/g). The same peregrine also contained the highest p,p′-DDE (0.067 μg/g).

Other Pesticides. None of the other pesticides or metabolites were reported above 0.02 μg/g ww in 1994 or in 2004 (Table 2). However, with lower detection limits (0.002 μg/g) in 2004, some extremely low concentrations were recorded in individuals including: heptachlor expoxide (0.002, 0.004, 0.01), dieldrin (0.003 [two birds]), oxychlordane (0.005), mirex (0.002 [five birds], 0.003, 0.004, 0.005, 0.009, 0.018), *cis*-chlordane (0.005, 0.015), and hexachlorobenzene (0.002). In 2004, mirex was most common and was detected in 10 of 27 (37%) blood plasma samples.

Conclusions. The reduction of OC pesticides in peregrines, especially the 96–97% decline in DDE concentrations between the last two sampling periods (1994 and 2004), is encouraging because burdens in this long-distance migrant reflect the prevalence of these compounds in both North and South America. However, the reduced use of OC insecticides likely implies that other insecticides are being used, possibly including the organophosphate monocrotophos, which induced mass mortality of another North American migrant, the Swainson's Hawk (*Buteo swainsoni*), in Argentina in 1995–1996 (Goldstein et al. 1999). The alternative organophosphates and carbamates are often more toxic (which can result in direct avian mortality) than the OCs, but they are less persistent (Mineau et al. 1999). Therefore, they cannot readily be monitored at migration sites like Padre Island. Although OC pesticide concentrations in peregrine blood plasma were extremely low in 2004, it is noteworthy that the World Health Organization (WHO) reversed a 30-yr policy on 15 September 2006 against the use of DDT via indoor spraying for malaria control in Africa (WHO 2006). If this practice extends to Latin America, perhaps this series of plasma contaminant concentration data from peregrines can continue to be used to monitor long-term DDE residue trends in the Americas. Several emerging contaminants, such as the polybrominated diphenyl ether (PBDE) flame retardants, have dramatically increased in bird eggs at many locations in recent years (Elliott et al. 2005, Gauthier et al. 2007) and have been

Table 2. Organochlorine pesticides (μg/g, ww), excluding p,p′-DDE, detected in blood plasma of adult female Peregrine Falcons captured in the spring at Padre Island, Texas, 1978–2004.

		NUMBER WITH DETECTABLE RESIDUES (% OCCURRENCE)[1,2]					
YEAR	*N*	p,p′-DDT	p,p′-DDD	HEPTACHLOR EPOXIDE	DIELDRIN	OXYCHLORDANE	MIREX
1978–1979	21	4(19%)	2(10%)	6(29%)	9(43%)	2(10%)	4(19%)
1980	63	1(2%)	0	28(44%)	37(59%)	11(17%)	18(29%)
1984	27	1(4%)	1(4%)	9(33%)	15(56%)	4(15%)	7(26%)
1994	45	0	0	0	0	0	0
2004	27	0	0	0	0	0	0
Fisher's test (*P*)		0.003	0.016	<0.0001	<0.0001	0.008	0.0001

[1] Number of samples with ≥0.02 μg/g. Fisher's exact test used to determine whether differences in occurrence among years for 1978–1994 (reported in Henny et al. 1996). No test made with the additional 2004 data because all samples were non-detections.
[2] Highest individual value (between 1978 and 1984) for each contaminant (μg/g): p,p′-DDT 0.44, p,p′-DDD 0.28, heptachlor epoxide 1.40, dieldrin 0.65, oxychlordane 0.13, mirex 0.17.

documented in blood plasma of Bald Eagles (*Haliaeetus leucocephalus*; McKinney et al. 2006). Peregrine blood plasma from Padre Island could be used for a large-scale PBDE monitoring program in the Americas.

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