ORGANOCHLORINES AND MERCURY IN OSPREY EGGS FROM THE EASTERN UNITED STATES

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ABSTRACT.—Organochlorine and mercury concentrations were determined in Osprey eggs collected from Maryland, Virginia, and Massachusetts during 1986–87. DDE concentrations were significantly different among locations. Median DDE concentrations did not decline significantly in eggs from Glenn L. Martin National Wildlife Refuge, Maryland, between 1973 and 1986. The median DDE residue for eggs from Martin Refuge in 1986 surpassed the value associated with 10% eggshell thinning, but was below the value associated with production of 1.0 young per active nest, a level assumed to represent a stable population. DDD, DDT, dieldrin, PCB, and mercury residues in all eggs appeared insignificant with regard to potential effects on shell thickness or reproduction. DDE and PCB residues were lower in eggs collected in 1986–87 than in those collected in the 1970s for each area. DDD, DDT, and dieldrin were not detected in Martin Refuge eggs in 1986, representing a significant reduction since 1973. DDD, DDT, and dieldrin levels in Massachusetts and Virginia eggs in 1986–87 were similar to those in eggs from the 1970s for each state. Mercury residues in eggs from Martin Refuge may be increasing and although not significant in this study, may warrant future monitoring.

Mercurio y compuestos organoclorados en huevos de Águila Pescadora del este de los Estados Unidos

EXTRACTO.—Concentraciones de mercurio, y compuestos organoclorados normalmente usados en pesticidas, fueron determinadas en huevos de águilas de la especie *Pandion haliaetus* colectados en Maryland, Virginia y Massachusetts durante 1986–87. Las concentraciones de DDE fueron significativamente diferentes de un lugar a otro. La media de las concentraciones de DDE no declinó significativamente en huevos colectados en el Refugio Nacional de Vida Silvestre Glenn L. Martin, Maryland, entre 1973 y 1986. La media de residuos de DDE, en huevos del Refugio Martin en 1986, sobrepasó el valor asociado con el 10% de disminución en el espesor de la cáscara; pero estuvo por debajo del valor asociado con la producción de una cría por nido activo, lo que es un nivel que se asume representa una población estable. Los residuos de DDD, DDT, dieldrine, bifenil policlorinado (PCB), y mercurio en todos los huevos parecían tener insignificantes potenciales efectos en el grosor de la cásara o en la reproducción. Los residuos de DDE y PCB en huevos colectados en 1986–87, fueron más bajos que los de aquellos colectados en los años 70 en cada área. DDD, DDT y dieldrine no fueron detectados en huevos del Refugio Martin en 1986, lo que representa una significativa reducción desde 1973. Los niveles de DDD, DDT y dieldrine en huevos colectados en Massachusetts y Virginia en 1986–87, fueron similares a aquellos de los colectados en los años 70 en cada estado. Puede que haya un incremento en los residuos de mercurio en huevos procedentes del Refugio Martin; y aunque no haya sido significativo para este estudio, puede que justifique futuros controles.

[Traducción de Eudoxio Paredes-Ruiz]

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Osprey populations in the eastern United States, including Chesapeake Bay, began to decline in the late 1950s, continuing through the 1970s (Ames 1966, Schmid 1966, Henny 1977, Reese 1977). During this period, high concentrations of organochlorines, including DDE, dieldrin, and polychlorinated biphenyls (PCBs), were found in eggs of populations

with poor reproductive success (Wiemeyer et al. 1975, Wiemeyer et al. 1978, Spitzer et al. 1978, Wiemeyer et al. 1988). By the early 1970s, productivity began to increase and continued through the 1970s in many portions of the Chesapeake Bay (Reese 1975, 1977). Also, preliminary data showed a decline in organochlorine levels found in Chesapeake Bay Ospreys in 1975–82 compared to 1971–73 (Wiemeyer et al. 1987). However, Osprey nestlings in areas such as Poplar Island, Tilghman Island, Glenn L. Martin National Wildlife Refuge and the mouth of the Choptank River, Maryland, have had decreased survival with mortality rates ranging as high as 40–75% (P.R. Spitzer unpubl.).

One possible cause of these isolated declines in nestling survival is contaminant accumulation in eggs or young. We present data on contaminants in Osprey eggs collected in 1986 from Martin Refuge, which supports one of the largest Osprey concentrations on the east coast. This area represents an Osprey "colony" where the cause of recent decreased nestling survival is unknown. Data on contaminants in Osprey eggs from the eastern United States collected in 1986–87 and Martin Refuge in 1973 are also presented. Our objective was to determine if concentrations of contaminants found in Osprey eggs from the eastern United States were at levels associated with adverse effects to reproduction, including nestling survival.

MATERIALS AND METHODS

Martin Refuge is located on the northern end of Smith Island, Somerset County, Maryland, and is bordered to the west by Chesapeake Bay, to the north by Kedges Straits, and to the east by Tangier Sound. Five freshly laid Osprey eggs were collected from randomly selected active nests in the spring of 1986. The eggs were double wrapped in aluminum foil, placed in plastic bags, and refrigerated soon after collection. Eggs were prepared for analysis in cooperation with staff of the Patuxent Analytical Control Facility of the U.S. Fish and Wildlife Service. The contents of each egg were emptied into separate chemically cleaned jars. Addled eggs collected from coastal Massachusetts (between Narragansett Bay and Buzzards Bay), and Virginia (York River area, Mobjack Bay, and Rappahannock River) as part of the 1986–87 U.S. Fish and Wildlife Service's "Northeast Bird Egg and Tissue Project" were prepared for analyses in a similar manner as Martin Refuge eggs collected in 1986. Field collection techniques and contaminant analyses for Osprey eggs collected at Martin Refuge in 1973 were described by Wiemeyer et al. (1988).

Eggs collected in 1986–87 were analyzed by laboratories under contracts administered by the Patuxent Analytical Control Facility, Laurel, Maryland, which monitored per-

formance and assured quality. Organochlorines were analyzed by Weyerhaeuser Analytical and Testing Services, Tacoma, Washington. Briefly, portions of homogenized samples were mixed with sodium sulfate and extracted for 20 hr with petroleum ether. Lipid cleanup of extracts was by gel permeation chromatography. Analysis was conducted with a Hewlett Packard 5880A gas chromatograph with dual columns (DB1 and DB 1701) and dual electron capture detectors. Lower limits of detection, before corrections for dehydration, that varied among samples were <0.1 ppm for pesticides and PCBs, except for PCBs in Maryland eggs where the limit was ≤0.6 ppm. In addition to the contaminants reported here, the samples were also analyzed for chlordane isomers and metabolites, heptachlor epoxide, endrin, hexachlorobenzene, mirex, and several other compounds, none of which were detected. The Osprey eggs were analyzed in a batch process with other lots. The batch size for soxhlet extraction was 12 (11 samples and 1 blank). No analytes were detected in the blank at concentrations greater than 0.5 ppb. Duplicate analysis of one of the Martin Refuge eggs collected in 1986 resulted in standard deviations of 0.21 and 0.13 for DDE and PCBs, respectively. Duplicate analysis of one of the Northeast Egg and Tissue Project eggs resulted in standard deviations of 0.19, 0.12, 0.12, 0.05, and 0.50 for DDE, DDD, DDT, dieldrin and PCBs, respectively. Mercury was analyzed by Environmental Trace Substances Research Center, Columbia, Missouri, using cold vapor atomic absorption with a Perkin Elmer Model 403 AA. The limit of detection for mercury was 0.02 ppm. Duplicate analysis of a Martin Refuge egg collected in 1986 and a Northeast Egg and Tissue Project egg resulted in standard deviations of 0.07 and 0.04, respectively. Spike recoveries of individual eggs were 97% for eggs collected at Martin Refuge in 1986 and 107% for eggs collected for the Northeast Egg and Tissue Project. Eggshell thickness was not measured in eggs collected in 1986–87.

The volume of all eggs was measured by water displacement or estimated as described by Stickel et al. (1973). Contaminant concentrations were adjusted by egg wet weight to volume ratios (ppm) assuming a specific gravity of 1.0 (Stickel et al. 1966). To aid in quantitative data analyses, 0.05 ppm was used for eggs where a particular contaminant was not detected. However, when a contaminant was not found in any eggs for a particular location, residue levels were simply listed as "not detected."

Due to small sample sizes and uncertainty regarding the sampling distribution associated with our egg contaminant data, nonparametric statistical tests were used to differentiate between and among median contaminant concentrations. While median values and geometric means reported elsewhere in the literature are not directly comparable, both are valid measures of central tendency for a data set. Kruskal-Wallis tests (Chi-square approximation) were used to determine if differences existed among all locations with data on a particular contaminant (Sokal and Rohlf 1981). If significant differences in median values were found among locations, all pairwise multiple comparisons were made using Wilcoxon's signed-rank test for unpaired data (normal approximation, Sokal and Rohlf 1981). Statistical significance was assumed at $P < 0.05$, and Bonferroni's multiple comparison technique was used

Table 1. Median (and range) contaminant concentrations (ppm fresh wet weight) in Osprey eggs from several locations in the eastern U.S., 1973–87.

LOCATION AND COLLECTION YEAR	N	CONTAMINANT					
		DDE	DDD	DDT	DIELDRIN	PCB	MERCURY[a]
Maryland							
Glenn L. Martin NWR, 1973	11	3.4	0.44	0.14	0.05	2.8	0.05
		(1.3–5.9)	(0.27–1.3)	(n.d.[b]–1.2)	(n.d.–0.20)	(1.8–4.3)	(0.03–0.11)
Glenn L. Martin NWR, 1986	5	2.3	n.d.	n.d.	n.d.	1.0	0.11
		(0.82–3.0)				(0.59–2.3)	(0.70–0.24)
Virginia							
York River area, Mobjack Bay and Rappahannock River, 1987	5	0.65	0.05	0.13	0.01	3.7	0.11
		(0.38–0.83)	(0.04–0.11)	(0.11–0.80)	(0.01–0.02)	(2.2–5.7)	(0.05–0.21)
Massachusetts							
Between Narragansett Bay and Buzzards Bay, 1986	4	0.56	0.13	0.23	0.03	2.4	0.06
		(0.45–0.68)	(0.10–0.18)	(0.12–0.29)	(0.0[c]–0.04)	(2.16–2.50)	(0.05–0.23)

[a] Sample size for mercury analysis was five for Glenn L. Martin National Wildlife Refuge in 1973 and three for Massachusetts in 1986
[b] n d. = not detected.
[c] Actual value calculated as 0.001 but reported as 0.00 when rounded for consistency.

to control the Type I error rate at 0.05 (Miller 1981). All data analyses were performed using the PC version of SAS (SAS Institute, Inc. 1985). Statistical differences found between 1973 and 1986–87 data should be viewed with caution based on variations in chemical analytical techniques and laboratories used for these two separate data sets.

RESULTS AND DISCUSSION

Median contaminant concentrations and range (ppm) in Osprey eggs from each area and year are given in Table 1. Residues of DDE were detected in all eggs from all locations and collection periods. Median concentrations of DDE did not decline significantly in eggs from Martin Refuge between 1973 and 1986 ($Z = 1.47$, $P = 0.14$). However, DDE concentrations were significantly different among locations sampled in 1986–87 ($\chi^2 = 8.52$, df = 2, $P = 0.014$). Multiple comparisons did not reveal statistically significant differences between location pairs, although eggs from Martin Refuge appeared to contain higher DDE residues than those from either Virginia ($P = 0.06$) or Massachusetts ($P = 0.06$). Eggs from similar areas of Virginia collected in 1976–77 contained geometric mean DDE concentrations of 1.8 to 2.6 ppm with the lowest concentration being 0.92 ppm (Wiemeyer et al. 1988). Eggs from the Westport River, Massachusetts in 1972–73, an area within the region sampled in 1986, had a geometric

mean of 4.2 ppm DDE, with the lowest concentration being 2.0 ppm (Wiemeyer et al. 1988).

DDE residues have been clearly associated with adverse effects on Ospreys including decreased reproductive success and associated population declines, whereas other organochlorine pesticides have not been associated with such effects (Wiemeyer et al. 1988). Median values for DDE reported from Virginia and Massachusetts in 1986–87 were well below reported values associated with biologically significant effects on eggshell thickness and reproductive success (Wiemeyer et al. 1975, 1988). The median residue for DDE from Martin Refuge in 1986 surpasses the 2.0 ppm DDE concentration associated with 10% eggshell thinning but is well below the 4.2 ppm DDE associated with 15% eggshell thinning (Wiemeyer et al. 1988). Also, the median residue value for Martin Refuge in 1986 is less than the 2.6 ppm DDE value associated with a production rate of 1.0 young per active nest and assumed to represent a healthy and stable population (Wiemeyer et al. 1988). A production rate of 0.8 young per active nest is considered necessary to maintain a stable population (Spitzer et al. 1983). Eggs collected at Martin Refuge in 1973 contained higher median DDE residues (3.4 ppm); eggshell thinning was 17% (Wiemeyer et al. 1988) and young produced was about 1.5 per active nest (S.N. Wiemeyer

unpubl.) which was considered excellent. Wiemeyer et al. (1988) had predicted these egg residues to be associated with about 14% thinning and a production rate of about 0.9 young per active nest. The equation estimating the relationship between DDE concentrations and brood size for eggs collected after failure to hatch, gave production estimates that were below actual levels of production in nearly all sampled populations (Wiemeyer et al. 1988) and should be used with caution.

Residues of DDD, DDT, and dieldrin were not detected in any eggs from Martin Refuge in 1986; therefore, this location and colletion period was assumed to have the lowest concentration of these contaminants and data analyses include only the other locations. The median concentration of DDD plus DDT was significantly higher in eggs from Massachusetts than in those from Virginia ($Z = 2.08$, $P = 0.037$). DDD and DDT residues were combined in the statistical analysis because DDT is metabolized to DDD during embryonic development (Abou-Donia and Menzel 1968) and reductive dechlorination occurs in embryonated eggs following death (Walker and Jefferies 1978). DDD plus DDT residues in Virginia and Massachusetts eggs collected in similar areas in the 1970s (Wiemeyer et al. 1988) were similar to those found in 1986–87. The DDD and DDT residues appear insignificant with regard to potential effects on shell thickness or reproduction.

The median concentration of dieldrin did not differ between eggs collected in Virginia and Massachusetts ($Z = 1.11$, $P = 0.27$). Dieldrin was seldom detected in Virginia eggs collected from similar areas in 1976–77, whereas eggs from the Westport River, Massachusetts, collected in 1972–73 contained a mean of 0.14 ppm. The median dieldrin values in the present study are similar to mean values reported to have no significant impact on Osprey productivity (Wiemeyer et al. 1988).

PCBs were detected in all eggs from all locations and collection periods. Significantly lower PCB residues were found in eggs collected at Martin Refuge in 1986 than in 1973 ($Z = 2.606$, $P = 0.01$), suggesting a decline in the loading of PCBs. Overall, median concentrations of PCBs were significantly different among locations sampled in 1986–87 ($\chi^2 = 8.63$, df = 2, $P = 0.01$). Multiple comparisons did not reveal statistically significant differences between location pairs, although eggs from Martin Refuge may have contained lower residues than eggs from Virginia ($P = 0.06$) or Massachusetts ($P = 0.11$).

The median PCB residue concentration for eggs from Virginia was the highest among all locations and collection periods reported in this study. Although this value is within the range of reported values for Osprey eggs collected from similar areas of Virginia in 1976–77, eggs from these areas contained mean concentrations of 5.0 to 9.2 ppm. Eggs collected from Westport River, Massachusetts, in 1972–73 contained a geometric mean of 8.3 ppm PCBs (range 2.2–23.0 ppm). PCB concentrations of the magnitude reported here have not been associated with adverse effects on Osprey reproduction (Wiemeyer et al. 1988). However, concentrations of highly toxic coplanar dioxin-like PCB cogeners and related compounds were not measured. These compounds have been implicated in reproductive impairment of fish-eating birds in other areas (Kubiak et al. 1989).

Mercury was detected in eggs from all locations and collection periods. No significant difference in mercury concentrations between collection periods was noted for eggs from Martin Refuge ($Z = 1.67$, $P = 0.09$). Further, no significant differences were detected among locations for eggs collected in 1986–87 ($\chi^2 = 0.96$, df = 2, $P = 0.62$). The slightly higher mercury levels found at Martin Refuge in 1986 compared to 1973 suggest that an increase in mercury contamination may have occurred. Mercury is being increasingly used in gold mining in Brazil in the Amazon Basin, much of which pollutes the aquatic environment (Martinelli et al. 1988, Pfeiffer et al. 1989). This is an important wintering area for Ospreys that breed in the Mid-Atlantic and Northeast areas of the United States (Poole and Agler 1987). Mercury concentrations in Osprey eggs were below those associated with adverse effects on reproduction (Wiemeyer et al. 1988).

DDE and PCB residues were lower in Osprey eggs collected in 1986 than in 1973 at Martin Refuge. Further, residues of DDD, DDT, and dieldrin were not detected in 1986 leading us to assume that a significant reduction in these contaminants has occurred as well. Concentrations of DDE and PCBs also appear to have declined in eggs from Virginia and Massachusetts. Although not significant, mercury residues in Osprey eggs from Martin Refuge may be increasing and warrant future monitoring. The concentrations of contaminants found appear far too low to impact nestling survival.

Geometric mean DDE concentrations in Osprey eggs from the Atlantic Coast and Delaware Bay of New Jersey that were collected in 1985–89 (Steidl

et al. 1991) bracketed the median concentration in eggs from Martin Refuge in 1986, whereas the New Jersey eggs contained somewhat higher DDE concentrations than eggs from Massachusetts and Virginia in 1986 and 1987. Dieldrin concentrations in the New Jersey eggs were similar to those we found in Virginia and Massachusetts eggs, whereas the New Jersey eggs, especially those from Delaware Bay, contained higher PCB concentrations than the eggs we analyzed. The differences in residue concentrations in Osprey eggs among these areas are an indication of exposure of the adults on their breeding areas, for they share common wintering grounds (Henny and Van Velzen 1972, Poole and Agler 1987).

Osprey eggs from Eagle Lake, California, collected after failure to hatch in 1983–84 (Littrell 1986), contained DDE concentrations similar to those in eggs from Martin Refuge in 1986. The California eggs contained much lower PCB concentrations than our samples from Virginia and Massachusetts, possibly due to the remote location of the California site from industrial contamination.

The ratios of DDE to DDD + DDT in the recent eggs from Virginia and Massachusetts compared to that in eggs from a variety of areas in earlier years (Wiemeyer et al. 1988), and the presence of DDT in all eggs from these two states suggests that these Ospreys were recently exposed to low levels of unmetabolized DDT; however, the source is unknown.

Bald Eagle (*Haliaeetus leucocephalus*) eggs from Maryland and Virginia that were collected from 15 territories after failure to hatch in 1980–84, contained geometric means of 4.4 ppm DDE, 0.42 ppm DDD + DDT, 0.31 ppm dieldrin, 14 ppm PCBs, 0.07 ppm mercury, and low concentrations of a variety of other organochlorines (S.N. Wiemeyer unpubl.). The higher concentrations of organochlorines in these eggs reflects the higher position of Bald Eagles in the food chain than that of Ospreys. Also, Chesapeake Bay Ospreys are exposed to contaminants on their breeding grounds for only about one-half of the year due to their migration, whereas breeding pairs of Bald Eagles are resident on the Chesapeake Bay.

One Peregrine Falcon (*Falco peregrinus*) egg collected in 1984 from South Marsh Island, Maryland, just to the north of Smith Island and Martin Refuge, contained 14 ppm DDE, 0.36 ppm heptachlor epoxide, 0.75 ppm oxychlordane, and 8.2 ppm PCBs (Gilroy and Barclay 1988). These elevated concentrations are also an indication of the high position of this species in the food chain and its presumed resident status in the region.

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