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Organochlorine Pesticides, PCBs, Dioxins, and Metals in Postterm Peregrine Falcon (*Falco peregrinus*) Eggs from the Mid-Atlantic States, 1993–1999

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Abstract Peregrine falcons were extirpated from the eastern United States by 1964 due to the effects of dichloro-diphenyl-trichlorethane (DDT) (Peakall and Kiff 1988). As a result of restoration efforts, peregrines have largely recovered in the region but remain a barometer of environmental contamination. In the course of monitoring nests, biologists in the mid-Atlantic states collected peregrine falcon eggs that failed to hatch. In the period 1993–1999, 93 eggs were collected from 66 nests in 31 locations in New Jersey, Pennsylvania, Delaware, Maryland, and Virginia. We analyzed eggs for organochlorine pesticides, PCBs, and metals, and calculated toxic equivalencies (TEQs) for dioxins and furans. Organochlorine contaminants were detected in eggs from all parts of the region. Although nest success in all parts of the region was good, the PCB TEQ in the Atlantic–New Jersey region was significantly related to nest success, and the regionwide PCB TEQ was nearly significant for nest success across the five-state area. dichloro-diphenyl-dichloroethylene (DDE), DDT, and total PCBs were negatively correlated with eggshell thickness, although eggshell thinning (10.4%) was not at a level associated with deleterious population effects. The five states represented in this study are productive for peregrine falcons and have contributed to the recovery of this species. However, the results suggest that Atlantic coastal peregrines might be subject to contaminant burdens that have the potential to decrease nest success and productivity.

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In the early 1960s, the peregrine falcon (*Falco peregrinus anatum*) appeared to be extirpated from the eastern United States (Berger et al. 1969). By 1970, it was gone from east of the Rocky Mountains, and throughout much of the western hemisphere the populations had declined drastically from those of the early 1900s. The peregrine falcon was listed as endangered on the US Federal List of Endangered and Threatened Wildlife (50 CFR 17.11-17.12) in June 1970. The 1965 Madison (Wisconsin) Peregrine Conference resulted in targeted research, and the persistent organochlorine compounds, specifically dichloro-diphenyl-trichlorethane (DDT) and its breakdown products, emerged as probable causative agents of eggshell thinning associated with peregrine falcon population decline (Anderson and Hickey 1972). Following a ban on the use of organochlorine compounds and the reintroduction of falcons into the wild (Cade 2003), the population has rebounded nationally as well as in the eastern United States in the past half-century. The peregrine falcon was removed from the US Federal List of Endangered and Threatened Wildlife on August 25, 1999. At the time, the population was estimated at 193 pairs in the eastern United States, about two-thirds of the 350 pairs estimated in the pre-DDT population (US Federal Register 1999). The population in the mid-Atlantic states of New Jersey, Pennsylvania, Delaware, Maryland, and Virginia numbered 56 pairs in 1999 and 85 pairs by 2007 (KEC). Historic populations within these states are not well known, due to the lack of documentation prior to the peregrine's decline.

In the mid-1990s, the US Fish and Wildlife Service's (USFWS) annual peregrine falcon nesting success records (i.e., production of young) for the eastern North American breeding population showed that production for many pairs remained below national levels. That review, combined with data from a limited number of analyses of peregrine falcon eggs and prey base from the mid-Atlantic states, indicated that contaminant monitoring of peregrine falcon eggs was still warranted (Steidl et al. 1991; USFWS 1994).

During the 1990s, postterm peregrine falcon eggs from nests in the mid-Atlantic states were collected and archived, and the contents were analyzed for polychlorinated biphenyls (PCBs), dioxinlike compounds, organochlorines, and metals. Eggshell thickness was measured and compared to pre-DDT-era peregrine falcon eggshells. This article presents those results and examines the relationships among contaminant levels, eggshell thickness, and nest success in the region. These results inform the postdelisting monitoring program and help direct peregrine falcon management.

Materials and Methods

Study Area, Sample Collection, and Eggshell Thickness Measurement

Between 1993 and 1999, 93 peregrine falcon eggs were collected postterm from 66 nests in 31 locations in New Jersey, Pennsylvania, Delaware, Maryland, and Virginia (Fig. 1). Eggs were wrapped in aluminum foil and refrigerated or frozen until processed in 1998 and 1999. For eggs collected from New Jersey and Pennsylvania, contents were harvested within 30 days after collection; others were frozen whole until contents were harvested. Whole-egg length, width, and mass were measured. Eggs were scored and the contents weighed and frozen prior to chemical analysis. Eggshells were rinsed with distilled water and air-dried for several months at room temperature in preparation for measuring thickness. Eggshell thickness was measured using a Starrett Dial Indicator Pocket Micrometer (0–9 mm). Measurements were obtained by averaging four measurements (for New Jersey and Pennsylvania, eight total, four from each eggshell half) taken in units of 0.01 mm along the eggshell equators. Measurements included shell membranes. In those instances in which the membrane was separated from the shell, the membrane was measured separately, averaged, and then added to the eggshell thickness. Eggshell fragments were not used for measurements.

Reproductive data were obtained from the state natural resource agencies. The number of birds fledged from a nest was recorded at the time of banding. Because the survey methods and timing were not similar across states, there was some uncertainty in records of total number of eggs laid. Therefore, the reproductive parameter used in the statistical analyses is nest success, defined as yes (fledging one or more young) or no (fledging no young).

Analytical Chemistry

Individual egg contents were analyzed for organochlorine pesticides, PCB congeners, dioxins/furans, and metals at the Geochemical and Environmental Research Group, Texas A&M University (College Station, TX). For pesticide, PCB, and dioxin/furan analyses, tissue extraction followed MacLeod et al. (1985) with minor revisions (Brooks et al. 1989; Wade et al. 1988). Samples were extracted with dichloromethane in the presence of sodium sulfate (drying agent) by maceration. After concentration and exchange to hexane, pesticide and PCB extracts were cleaned up using silica gel/alumina column chromatography and further purified by gel permeation chromatography. The extracts were analyzed for pesticides and PCBs by gas chromatography with an electron capture detector (GC/ECD) with analysis conditions (90-min run) to separate the pesticides and PCBs. Dioxin/furan analysis followed Tondeur (1987) and US Environmental Protection Agency Method 1613b (USEPA 1990). Extracts were processed through acid-silica swirl, mixed-bed silica columns, and charcoal columns; samples were analyzed by GC on a high-resolution mass spectrometer. For most metals, egg tissue was digested in screw-cap Teflon Bombs with concentrated nitric acid. Mercury (Hg) was determined by cold vapor atomic absorption spectrometry (AAS). Arsenic, selenium, cadmium, and lead were determined by graphite furnace AAS. The remaining elements were determined by atomic emission (Taylor and Presley 1998).

Quality assurance and quality control (QA/QC) procedures were USFWS Analytical Control Facility standards. Procedural blanks, standard reference samples, duplicates, and spiked recovery samples were run on 6% of the total number of samples for each analyte. Duplicates and spiked recovery results were within acceptable ranges, with the exception of endosulfan II, for which the results were not used for statistical analysis and interpretation.

Data Analysis

Following Steidl et al. (1991) and USFWS (1994), the mean eggshell thickness was compared to the pre–1947 mean eggshell thickness of 0.375 mm (Anderson and Hickey 1972) for peregrines of the eastern United States using the Student's *t*-test. The temporal trend of eggshell thickness was explored using the nonparametric Kendall τ-b concordance analysis, with individual eggshell thickness and duration between 2004 and the egg collection year as

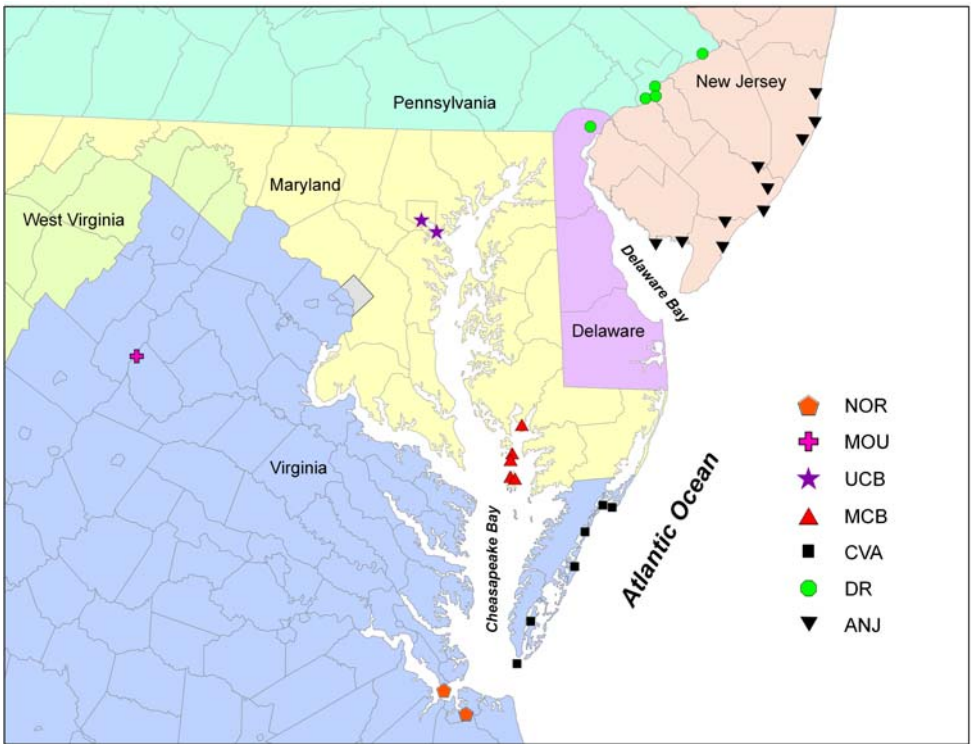

Fig. 1 Map of the mid-Atlantic study region showing locations of nests from which eggs were taken, 1993–1999, and their assignment of nests to localized areas

variables. Nests were assigned to regions based on watersheds (Fig. 1), and if the sample size within a region was large enough (eight or more), one-way analysis of variance (ANOVA) and post-ANOVA (Student–Newman–Keuls) were used to test the effect of watershed on eggshell thickness.

All of the concentrations reported here are on adjusted wet weight basis. Egg residue results were corrected for volume loss due to dehydration using the formula developed for American kestrel (*Falco sparverius*) eggs by Wiemeyer et al. (1986) and assuming a specific gravity of 1.0 (Stickel et al. 1973): fresh weight, grams = 0.55 × [length (cm) × breadth (cm^2)] – 0.175. Egg masses were not taken for 10 eggs collected in 1993–1994 from New Jersey. To cope with this missing metric, field notes on the condition of the eggs were reviewed and it was determined the eggs were intact. As the Kendall τ-b concordance analysis showed no significant egg mass trend over time ($\alpha = 0.05$), the egg mass geometric mean assigned to the 10 eggs was determined from all other peregrine falcon eggs from New Jersey in this sample set as well as eggs collected in a 1990–1991 study (US Fish and Wildlife Service and New Jersey Division of Fish, Game and Wildlife 1998).

If more than one egg was collected from a clutch, individual chemical analyses were performed on each egg. After assessing normality and variance homogeneity of the log-transformed data, the interclutch variation was determined to be significantly larger than the intraclutch variation (one-way ANOVA, $\alpha = 0.05$). Therefore, clutch residue and eggshell thickness geometric means were used to represent the clutches. The discussion here focused on analytes known or suspected to be biologically important to birds and/or, specifically, peregrine falcons: Hg, Se, *p,p′*-dichloro-diphenyl-dichloroethylene (*p,p′*-DDE), DDT, heptachlor epoxide, oxychlordane, *trans*-nonachlor, mirex, HCB (hexachlorobenzene), dieldrin, total PCBs, and dioxinlike compounds.

Analytes detected in >60% of the total number of samples were summarized as geometric means, ranges, and percent detects. The Helsel Robust method was used to generate reasonable substitute values for those below the detection limits (DL; UNCENSOR shareware at www.vims.edu/env/research/software/vims_software.html). The normality of the log-transformed data for each analyte was determined using the Shapiro–Wilks test before using the method. If there were multiple DLs reported by the lab, the analyte DL was set at the highest reasonable DL.

To evaluate the exposure from dioxinlike chemicals, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) toxic equivalent (TEQ) concentrations were calculated for specific PCB, polychlorinated dibenzo-*p*-dioxin (PCDD), and polychlorinated dibenzo-*p*-furan (PCDF) analytes and summed for each sample following the approach presented in US EPA (2003) and Van den Berg et al. (1998). TCDD was assigned a toxicant equivalent factor (TEF) value of 1 and the concentration of a compound multiplied by its established TEF results in its TEQ. PCBs 81, 123, and 157

were not reported by the laboratory and, therefore, were not included in the calculation. All TEQs were calculated three ways to cope with below detection values and to bracket actual TEQs: residue results below detection substituted at the detection limit, at one-half the detection limit, and at zero. There was little difference among TEQ means (~8%) using the three methods. For reporting here, we set residues below the DL at ½DL. The TEQ difference by region was analyzed using ANOVA and post-ANOVA (Student ~ Newman ~ Keuls).

The Kendall τ-b concordance analysis was used to explore concordance between eggshell thickness and contaminants that have potential eggshell thinning effects (i.e., total PCBs, *p,p′*-DDE, and total DDT). The relationships between independent variables of eggshell thickness and contaminant concentration, with the response variable of nest success, were explored using binary logistic regression (SAS LOGISTIC; SAS Institute 1999). Five eggs with some content loss were deleted from the residue analysis data sample set.

Results

Eggshell Thickness

Eggshell thickness showed no evidence of deviation from normality (Shapiro–Wilks test, $p = 0.134$; SAS UNIVARIATE; SAS Institute 1999). The mean thickness was 0.336 mm (standard error = 0.003, $n = 93$) and was significantly thinner (10.4%; Student *t*-test, $\alpha = 0.05$) than the pre-1947 mean eggshell thickness of 0.375 mm. There was no significant eggshell thickness change from 1993 to 1999 ($p = 0.581$). The mean eggshell thicknesses of Delaware River (DR) and middle Chesapeake Bay (MCB) were significantly greater than those from Atlantic–New Jersey (ANJ), and the DR thickness was significantly greater than the Coastal-VA (CVA) eggs ($\alpha = 0.05$). Eggshells from Norfolk-area (NOR), Upper Chesapeake Bay (UCB), and Mountain (MOU) were not found to be different from the other four areas (Fig. 2).

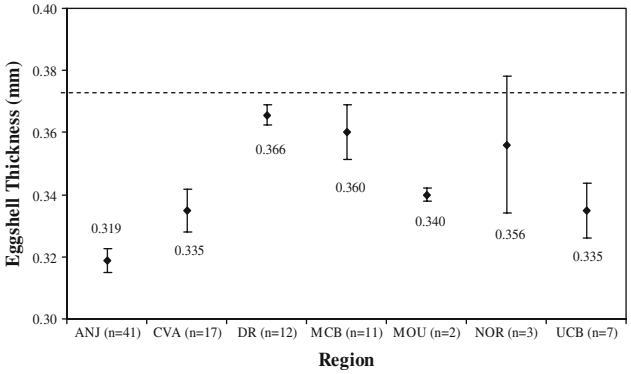

Fig. 2 Eggshell thickness means and standard errors by region. The dashed line represents the pre-DDT eggshell thickness of 0.375 mm. Sample sizes for the regions are shown in brackets (ANJ: Atlantic-New Jersey; CVA: Coastal-Virginia; DR: Delaware River; MCB: Middle Chesapeake Bay; MOU: Mountain; NOR: Norfolk area; UCB: Upper Chesapeake Bay)

Residues

The sample sizes, geometric means, ranges, and percent detections of the contaminants by region are listed in Table 1, as well as a summary of nest success. Six metals [copper (Cu), iron (Fe), mercury (Hg), magnesium (Mg), selenium (Se), and zinc (Zn)] were detected in >60% of all samples; all were detected in 100% of eggs except Hg (92%). Among all the regions, DR had the highest concentrations for all the metals except Hg. Fourteen organics were detected in >60% of all samples: beta BHC, *cis*-nonachlor, dieldrin, oxychlordane, *trans*-nonachlor, HCB, heptachlor epoxide, mirex, *o,p′*-DDD, *p,p′*-DDD, *p,p′*-DDE, *o,p′*-DDT, *p,p′*-DDT, and total PCBs. In 10 of the 14 organics, ANJ had the highest contaminant levels. The MOU area had the lowest contaminant levels for most of the organics (11 of 14).

The geometric mean of TEQs in all samples was 0.37 μg/kg, with a range of 0.03–1.84 μg/kg. The ANJ and CVA means were above the no-observed adverse effect level (NOAEL; USEPA 2003) of 0.23 μg/kg; all study site TEQs were below the lowest observed adverse effect level (LOAEL; US EPA 2003) of 3.39 μg/kg for American kestrel (*Falco sparverius*) (Fig. 3). The TEQ of MOU was significantly lower than the other regions, whereas that of ANJ was significantly higher than DR, NOR, UCB, and MOU.

Relations Among Eggshell Thickness, Contaminants, and Nest Success

Eggshell thickness was significantly negatively correlated with *p,p′*-DDE ($n = 60$, $p = 0.0098$), total DDT (*o,p′*-DDT and *p,p′*-DDT, $p = 0.0131$), and total PCB ($p = 0.0002$) concentrations (Kendall τ-b analysis, $\alpha = 0.05$).

On the whole, binary logistic regression did not show any significant effect of eggshell thickness, Hg, Se, *p,p′*-DDE, or total PCBs on peregrine falcon nest success ($\alpha = 0.05$). The TEQ was on the borderline of significance ($p = 0.0566$). The effects of the aforementioned toxicants and thickness on nest success by region were also explored: In ANJ, only the TEQ had a significant effect ($p = 0.0024$). In the other regions, none of the analytes was significantly related to nest success. Logistic regression also was performed using the logarithms of TEQs and residues, but less significance was obtained on the whole and by region.

Table 1 Sample size (*n*), geometric mean, range, and percent detection for analytes detected in >60% of peregrine falcon eggs (clutches)

Analyte	Mean	Region *n*	ANJ 31	CVA 10	DR 5	MCB 9	MOU 1	NOR 2	UCB 2
Cu	0.34	Mean	0.35	0.34	0.45	0.28	0.22	0.28	0.29
		Range	0.16–0.61	0.22–0.60	0.35–0.79	0.21–0.40	–	0.25–0.33	0.28–0.29
		Detect	100%	100%	100%	100%	100%	100%	100%
Fe	11.14	Mean	12.38	9.51	13.58	8.69	11.45	11.54	8.55
		Range	5.91–45.19	4.10–16.90	10.49–18.72	6.06–14.70	–	11.05–12.04	7.08–10.34
		Detect	100%	100%	100%	100%	100%	100%	100%
Mg	47.01	Mean	50.00	40.22	64.07	45.89	40.00	23.04	45.35
		Range	21.89–78.89	14.26–66.81	52.91–70.87	13.54–59.91	–	10.07–52.72	37.55–54.77
		Detect	100%	100%	100%	100%	100%	100%	100%
Hg	0.22	Mean	0.43	0.25	0.04	0.20	0.10	0.10	0.05
		Range	0.07–1.60	0.16–0.44	0.01–0.58	0.08–0.93	–	0.07–0.14	0.03–0.10
		Detect	100%	80%	60%	100%	100%	100%	100%
Se	0.26	Mean	0.25	0.30	0.29	0.26	0.18	0.24	0.25
		Range	0.11–0.42	0.21–0.45	0.25–0.33	0.18–0.39	–	0.23–0.24	0.18–0.34
		Detect	100%	100%	100%	100%	100%	100%	100%
Zn	5.83	Mean	6.00	5.90	6.80	4.81	6.14	6.33	5.08
		Range	2.20–9.66	4.30–9.61	5.51–8.37	3.13–6.94	–	6.09–6.58	4.44–5.83
		Detect	100%	100%	100%	100%	100%	100%	100%
Beta BCH	0.0035	Mean	0.0044	0.0028	0.0022	0.0027	0.002	0.0023	0.0033
		Range	0.0006–0.0163	0.0011–0.0050	0.0009–0.0043	0.0018–0.0072	–	0.0019–0.0029	0.0021–0.0051
		Detect	84%	90%	60%	100%	100%	100%	100%
Cis-nonachlor	0.026	Mean	0.041	0.020	0.006	0.035	0.003	0.018	0.012
		Range	0.009–0.148	0.002–0.105	0.001–0.061	0.020–0.084	–	0.007–0.043	0.008–0.020
		Detect	94%	90%	60%	100%	100%	100%	100%
Dieldrin	0.20	Mean	0.22	0.33	0.05	0.30	0.03	0.21	0.11
		Range	0.05–2.34	0.09–0.65	0.01–0.30	0.09–1.31	–	0.10–0.45	0.05–0.25
		Detect	100%	100%	100%	100%	100%	100%	100%
Oxychlordane	0.34	Mean	0.55	0.25	0.15	0.25	0.07	0.13	0.29
		Range	0.10–1.64	0.12–0.65	0.07–0.25	0.14–0.45	–	0.10–0.18	0.19–0.44
		Detect	100%	100%	100%	100%	100%	100%	100%
Trans-nonachlor	0.14	Mean	0.20	0.12	0.03	0.15	0.02	0.08	0.08
		Range	0.03–1.78	0.01–0.43	0.01–0.13	0.07–0.26	–	0.04–0.18	0.05–0.14
		Detect	100%	100%	80%	100%	100%	100%	100%

Table 1 continued

Analyte	Mean	Region *n*	ANJ 31	CVA 10	DR 5	MCB 9	MOU 1	NOR 2	UCB 2
HCB	0.010	Mean	0.010	0.012	0.005	0.012	0.002	0.007	0.011
		Range	0.002–0.045	0.004–0.037	0.001–0.016	0.005–0.021	–	0.005–0.011	0.009–0.014
		Detect	94%	100%	60%	100%	100%	100%	100%
Heptachlor epoxide	0.10	Mean	0.12	0.13	0.05	0.09	0.03	0.09	0.14
		Range	0.02–0.47	0.06–0.34	0.03–0.09	0.06–0.17	0.03–0.03	0.07–0.11	0.07–0.27
		Detect	100%	100%	100%	100%	100%	100%	100%
Mirex	0.124	Mean	0.193	0.087	0.060	0.103	0.059	0.036	0.077
		Range	0.070–1.026	0.030–0.211	0.032–0.100	0.063–0.236	–	0.021–0.064	0.064–0.092
		Detect	100%	100%	80%	100%	100%	100%	100%
o,p′-DDD	0.0092	Mean	0.0196	0.0050	0.0016	0.0040	0.0008	0.0066	0.0032
		Range	0.0005–0.1404	0.0004–0.0575	0.0008–0.0041	0.0009–0.0148	–	0.0042–0.0102	0.0012–0.0062
		Detect	97%	80%	60%	67%	100%	100%	100%
p,p′-DDD	0.024	Mean	0.035	0.049	0.007	0.012	0.017	0.027	0.003
		Range	0.008–0.120	0.009–0.121	0.002–0.020	0.002–0.089	–	0.012–0.063	0.001–0.006
		Detect	97%	100%	60%	100%	100%	100%	100%
p,p′-DDE	4.26	Mean	6.52	3.56	1.81	3.39	1.06	2.14	1.28
		Range	1.75–31.87	0.81–9.23	0.62–3.29	2.15–5.05	–	1.15–3.95	0.94–1.75
		Detect	100%	100%	100%	100%	100%	100%	100%
o,p′-DDT	0.010	Mean	0.016	0.015	0.001	0.013	0.001	0.012	0.003
		Range	0.001–0.102	0.005–0.062	0.000–0.007	0.002–0.039	–	0.010–0.015	0.001–0.011
		Detect	74%	100%	20%	100%	100%	100%	100%
p,p′-DDT	0.0079	Mean	0.0155	0.0031	0.0061	0.004	0.0016	0.0004	0.0092
		Range	0.0007–0.3565	0.0003–0.1335	0.0022–0.0289	0.0003–0.0715	–	0.0003–0.0005	0.0039–0.0217
		Detect	87%	60%	60%	89%	100%	0%	100%
Total PCB	7.62	Mean	12.95	5.59	2.99	5.50	0.48	3.62	3.64
		Range	3.77–57.91	2.96–14.81	0.14–9.70	4.71–6.73	–	3.56–3.69	2.85–4.65
		Detect	100%	100%	100%	100%	100%	100%	100%
Clutch productivity		Eggs collected	41	17	12	11	2	3	7
		Clutches that produced	19	10	1	10	0	1	1
		Clutches that failed to produce	13	2	4	1	1	1	2

Note: Analyte residues are presented in mg/kg, adjusted fresh wet weight. Geometric means were calculated with values below DL substituted using the Helsel Robust method. Clutch productivity (yes/no) is listed at the end. ANJ: Atlantic–New Jersey, CVA: Coastal-VA, DR: Delaware River, MCB: middle Chesapeake Bay, MOU: Mountain, NOR: Norfolk area, UCB: Upper Chesapeake Bay

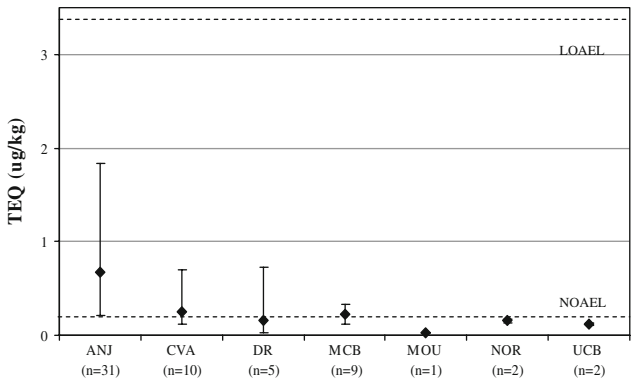

Fig. 3 The TEQ geometric means (adjusted fresh wet weight) for peregrine falcon eggs. Bars reflect range of eggs within each study region. Sample sizes for the regions are shown in brackets. The LOAEL (3.39 μg/kg) and NOAEL (0.23 μg/kg) are also shown

Discussion

Eggshell Thickness Trends

Eggshell thickness is an important indicator of reproductive health. Peakall and Kiff (1988) summarized eggshell thinning data and peregrine falcon population changes and reported that "…declining or extirpated populations were found in every instance where mean eggshell thinning exceeded 17%." The eggshells in this study were 10% thinner than that of the pre-DDT era, with a maximum thinning of 30%. Eggshell thickness was not significantly related to nest success across all sample area. Nor was a temporal trend of thickness evident, possibly due to the limited time period represented. Eggshell thinning reported in our sample set was similar to that reported by Burns et al. (1994), who found 11% thinning for addled peregrine falcon eggs ($n = 112$) collected in 1986–1988 from the East Coast of the United States. Steidl et al. (1991) reported 16.4% eggshell thinning in New Jersey peregrine falcon eggs ($n = 58$) in 1985–1988, slightly higher than the 12.5% thinning (range: 1.8% thickening to 30.2% thinning) observed in New Jersey (ANJ and DR) eggs currently. The USFWS (1994) reported 14.6% thinning (range: 4.8–26.9%) in 10 eggs collected from Virginia and Maryland in 1992, notably higher than our results of 8.6% in 1993–1999 for those two states (range: 21.30% thinning to 9.9% thickening). Comparing our results with previous studies, there might be some improvement of eggshell thickness with time in the mid-Atlantic area.

Metals

It is known that metal exposure in ovo can affect the immune system, brain, and hatchability (Bunn et al. 2000; Edens and Garlich 1983; Nyholm 1998). Hg can cause mortality and reproductive impairment in wild birds (Eisler 1987). Although the geometric mean of Hg (0.22 mg/kg) reported here was higher than the 0.10 mg/kg found in western North Carolina falcons (Augspurger and Boynton 1998), the means in all the regions were below the suggested critical level of 1 mg/kg associated with reproductive impairment in peregrine falcons (Peakall et al. 1990). Further, all individual eggs had Hg concentrations lower than this threshold except two eggs from ANJ. However, one-third of eggs in our sample set (most in ANJ) exceeded 0.5 mg/kg Hg, which Wolfe et al. (1998) suggested as the LOAEL for bird eggs, based on their extensive review of the literature. Although Thompson (1996) suggested that 0.5 mg/kg caused little detrimental effect on reproduction based on interpopulation studies, Heinz and Hoffman (2003) concluded that concentrations <1 mg/kg could harm mallard embryos. We did not detect a negative effect of Hg on nest success in any of the regions, including ANJ, where 59% of eggs had >0.5 mg/kg Hg concentrations. Se in birds has been associated with embryotoxicity, teratogenesis, and other reproductive impairment (Heinz 1996; Hoffman and Heinz 1988). The mean (0.26 mg/kg) and highest (0.45 mg/kg) concentrations of Se in this sample set were an order of magnitude below the suggested critical levels of 3–5 mg/kg for birds (Heinz 1996) and was not related to nest success significantly. USFWS (1994) reported a similar level of 0.29 mg/kg for eight peregrine falcon eggs in Virginia. Other metals in this sample set were below levels of significance.

Organochlorine Residues

Eggshell quality and embryo hatchability of peregrine falcons can be impaired by DDT and its primary metabolite DDE (Anderson and Hickey 1972). Peakall et al. (1990) suggested a critical level of DDE in eggs (15–20 mg/kg, wet weight) based on experimental cause–effect data. Although *p,p′*-DDE was detected in all of our samples, levels were below the critical range, except for one ANJ egg containing nearly 32 mg/kg. We found the geometric mean of DDE (4.26 mg/kg) to be lower than those reported from an earlier study in the eastern United States (7.8 mg/kg; Burns et al. 1994), Nunavut, Canada (7.6 mg/kg; Court et al. 1990), and California (12 mg/kg; Jarman et al. 1993) but higher than that of western North Carolina falcons (Augspurger and Boynton 1998). Lower levels were also observed in Ireland (2.1–2.3 mg/kg), but they ranged from 0.7 to 15.2 across Britain (Newton et al. 1989). Differences in DDE among regions might be explained by different prey availability. Within our four regions with more than two samples, DDE means ranged from 1.8 mg/kg in the DR to 6.5 mg/kg in ANJ, with MCB and CVA in between, with 3.4 and 3.6 mg/kg, respectively. Peregrines in the

relatively urban DR area forage primarily on pigeons (*Columba livia*) and short-distance migrant passerines such as blue jays (*Cyanocitta cristata*) and have less exposure to DDT derivatives. Coastal peregrines feed on an abundance of migrant shorebirds (Steidl et al. 1997) that migrate to South American countries, where DDT remains in use. Studies of ospreys in the Delaware Bay region have documented both a reduction in DDE over time (1980s to 1990s; Clark et al. 2001) and DDE levels that could still depress reproduction (Toschik et al. 2005). The fact that peregrines in this same region (DR) had the lowest level of DDE among regions with more than two samples suggests their diet is less contaminated, nonmigrant, or short-distance migrant birds. We suggest that the coastal peregrines continue to be exposed to contaminants, including DDE, at a higher level than inland peregrines as a result of their dominant prey of coastal resident and migrant birds.

Lundholm (1997) reviewed the effects of *p,p′*-DDE on the Ca and prostaglandin metabolism of bird eggshell gland and suggested that *p,p′*-DDE is the only substance in the DDT family that produces eggshell thinning. We found a significant negative relationship between eggshell thickness and *p,p′*-DDE and total DDT. However, there was no statistically significant relationship of *p,p′*-DDE and regional nest success.

The geometric mean of dieldrin (0.2 mg/kg) is below the suggested screening value of 1–4 mg/kg for adverse effects in peregrine eggs (Peakall et al. 1990). However, there were two eggs (in ANJ and MCB) within that adverse effects range. Newton (1988) suggested a critical level in peregrine falcons of 0.7 mg/kg. Based on both references, some peregrines in ANJ and MCB might be at risk for dieldrin effects.

Chlordane and metabolites *cis*-nonachlor, oxychlordane, *trans*-nonachlor, and heptachlor epoxide were detected in >60% of our samples at levels below those associated with reproductive impairment (see summary by Wiemeyer, 1996). The mirex level (0.12 mg/kg) is comparable to that found by Burns et al. (1994) and is below levels associated with adverse effects in birds (Wiemeyer 1996). HCB is a fungicide that has been associated with adverse effects to reproduction in birds (Jarman et al. 1996). The HCB level in our sample set was 0.010 mg/kg, slightly below the mean of falcons from the eastern United States in 1986–1988 (0.023 mg/kg; Burns et al. 1994) and well below levels associated with adverse reproductive effects to birds (4 mg/kg; Peakall et al. 1990).

PCBs and 2,3,7,8-TCDD Toxic Equivalency

Interpreting PCB results is difficult given the wide range of effects to birds and residue levels reported in the literature. Peakall et al. (1990) suggested a critical PCB level of 40 mg/kg for effects to peregrine eggs. Although eggs in our study averaged well below that (geometric mean: 7.62 mg/kg), 1 egg had 57.91 mg/kg and 11 eggs had 19–22 mg/kg. All these elevated-PCB eggs were from ANJ. Burns et al. (1994) reported a similar total PCB average of 8.9 mg/kg in peregrine falcon eggs in the eastern Unites States in 1986–1988, as did Court et al. (1990), with an average of 8.7 mg/kg in Nunavut, Canada. Jarman et al. (1993) found 4.8 mg/kg (range: 1.4–13.0) in California peregrine falcons. Henny et al. (1994) found total PCBs of 7.3 mg/kg in eggs from the Kola Peninsula, Russia. Herzke et al. (2002) documented a total PCB concentration of 9.1 mg/kg in peregrine eggs in Norway, the highest among eight raptor species they analyzed. Much lower levels were documented from Zimbabwe, Africa, averaging 0.2–0.3 mg/kg (Hartley et al. 1995). In Britain, PCB levels were lower in peregrine eggs from inland regions (0.4–5.5 mg/kg) than those in coastal areas (3.6–13.7 mg/kg) (Newton et al. 1989). Regions within our sample area did not differ in PCB congener makeup: Across regions, PCB congener 118 accounted for 55–73% of total PCBs, and the sum of congeners 118 and 105 accounted for 76–88% of total PCBs. The previous study in the east by Burns et al. (1994) did not include congener analysis, so we cannot compare this for the same geographic area. Interestingly, total PCBs in peregrine eggs from the New Jersey coast in 1986–1988 (Burns et al. 1994) did not differ appreciably from our findings: They reported 14.0 mg/kg in the region where we found 12.95. There are no known point sources that might be causing elevated PCBs in southern New Jersey tidal marshes, but there are elevated contaminants in the lower Delaware River, 80 km to the west (Delaware River Basin Commission 1998).

As possible eggshell-thinning compounds, the total PCB concentration was found to be significantly correlated with eggshell thickness. A study on osprey eggs by Rattner et al. (2004) did not show such a relationship ($\alpha = 0.05$). Our finding might be a result of a correlation between DDE (known to cause eggshell thinning) and total PCBs.

Polychlorinated biphenyls have been reported to be embryotoxic and teratogenic to birds (see summary in Rice and O'Keefe,1995). The most toxic congeners in PCB mixtures are non-ortho-PCBs (Safe et al. 1982). The PCDDS, PCDFs, non-ortho PCBs, and mono-ortho PCBs share similar chemical structure, toxicant response pattern, and receptor-mediated mechanism (Poland and Knutson 1982), known to cause aryl hydrocarbon receptor-mediated toxicity to birds, such as embryotoxicity and reproductive and developmental impairment. As a result, the TEFs are based on the relative induction and toxicity of PCDDs, PCDFs, PCBs, and related compounds. The TEQ results are "order of magnitude" because of the uncertainty in TEFs for the mix of PCB congeners with dioxinlike toxic potency.

The TEQs in this study included PCDDs, PCDFs, and both non-ortho and mono-ortho PCBs on a wet weight basis. The mean TEQ (0.37 μg/kg) was just above the US EPA's NOAEL and well below the LOAEL for American kestrel. This result and those from similar studies are not directly comparable because of the differences in individual compounds included and the weight base (fat, wet, or dry weight) used in the calculation. Jarman et al. (1993) reported 0.12 μg/kg TEQ for California peregrine falcons, which included only PCDDs, PCDFs, and non-ortho PCBs. The TEQ for peregrine falcons in central Spain (0.047 μg/kg) was for PCDDs, PCDFs, and non-ortho- and ortho-PCBs (Merino et al. 2005). The TEQ (0.368 μg/kg) for peregrine falcons from Germany was on a fat basis (Malisch and Baum 2007). Across our sample area, TEQ toxicity and nest success approached significance ($p = 0.0566$), and we did find a significant correlation ($p = 0.0092$) between TEQ level and nest success in the ANJ region (TEQ: 0.791 μg/kg.) Given the uncertainty in the TEFs used to calculate TEQs, and the difficulty of accounting for individual and synergistic effects of PCBs, we suggest that these compounds might pose a risk to peregrine falcon productivity in the mid-Atlantic states.

Food Base, Nest Location, and Residues

Peregrine falcons are exposed to contaminants through their mostly avian food base, with more than 400 species of birds as prey recorded for North American peregrines (White et al. 2002). In urban areas and on the bridges that span large rivers, the diet consists of primarily resident passerines and Columbiformes (nonmigratory and low-trophic-level seed-eaters). Adult peregrine falcons in the mid-Atlantic region are mostly nonmigratory, remaining in their nesting territories year around; thus their eggs are a reflection of the prey available in those areas, subject to seasonal fluctuations. In our sample area, eggs from the more urban regions (DR, UCB, and NOR) generally showed less exposure to organochlorines, but DR eggs had higher metals (Cu, Fe, Mg, Se, Zn), which might reflect a local condition of the lower DR (NJDEP 2006). In coastal areas, the diet is chiefly one of migratory shorebirds and waterfowl (Steidl et al. 1997; White et al. 2002). Steidl et al. (1997) found that the peregrine diet in ANJ was composed of two-thirds migratory birds, which typically contain greater contaminant levels than resident birds (Lindberg et al. 1985). In the only analysis of peregrine prey in the region, they collected potential prey in southern New Jersey marshes in 1987–1988, reporting DDE ranging from 0.32 (blue jay) to 0.90 (fish crow) and total PCBs ranging from 0.1 (grackle) to 1.7 (fish crow); levels in resident prey were similar to willets, the only migrant species sampled, although all were taken from salt marsh habitats. We would expect peregrine diets to be similar in ANJ, CVA, and perhaps MCB, based on the tidal marsh habitats that dominate those areas, with an abundance of long-distance migrant shorebirds and water birds. However, ANJ stood out among the coastal areas as having the highest levels of contaminants, particularly PCBs, TEQs, Hg, and DDE. For some organics and Hg, CVA also had elevated levels, but all below those of ANJ and none that correlated with reduced nest success. Among the mid-Atlantic states, higher PCBs and DDE in coastal New Jersey peregrine eggs seems to be a continuing condition, as very similar residues were found in our results and the most comparable previous study by Burns et al. (1994). Thus, the differences between ANJ and the other areas might not be explained by what might be more contaminated prey available in coastal areas. There is also no cause to suspect that peregrines in the ANJ are older than those nesting elsewhere, age being a factor in the level of bioaccumulation of contaminants. We did not have sufficient data to examine age of breeding birds, but more recent data on individuals indicate a relatively even age distribution (KEC). On the whole, these results suggest there might be historic, possibly localized sources of organochlorines in southern New Jersey tidal marshes that continue to be mobilized in the food web. Additional research is needed to locate and control point sources, although historic contamination might now be categorized as nonpoint and more difficult to mitigate.

In a study of southern New Jersey ospreys, PCBs and DDE in eggs and prey fish decreased to moderate levels (1989 to 1998), but Hg increased in Atlantic coast samples. To the extent that the source of Atlantic coastal food-chain Hg is from air deposition to water (Porcella et al. 1995), all waters, including the Atlantic Ocean coast, are vulnerable to elevated Hg, a condition that might continue until more effective control of atmospheric Hg emissions is achieved. Another possible source is methylation that occurs in estuaries with high organic matter (Lambertsson and Nilsson 2006), such as salt marshes. Further, migratory peregrine falcons and their prey continue to be exposed to contaminants in areas of the world where organochlorine pesticides are still in use.

Conclusions

These results present the opportunistic analysis of contaminants data collected ancillary to the evaluation of peregrine falcon productivity and nest success. The results might be biased because only postterm eggs were collected. For peregrines represented in the sample area, organochlorine contaminants were detected in all parts of the mid-Atlantic region, although nest success was generally good.

The only statistically significant evidence of contaminant effects on nest success was with the PCB-related TEQ in the Atlantic–New Jersey region, although the regionwide PCB TEQ was nearly significant for nest success in the sample area as a whole. DDE, DDT, and total PCBs were negatively correlated with eggshell thickness, although eggshell thinning was not at a level associated with population effects.

The coastal areas of New Jersey, Virginia, and Maryland have been, and continue to be, productive nesting areas for peregrine falcons, contributing to the recovery of this species in those states and the eastern United States. As the current study indicates, peregrine falcons continue to be monitored for environmental contaminants that might affect wildlife. It is a goal to fully restore peregrines to their historic range in the East, in particular the Appalachian Mountains, where nesting continues to be sparse. The coastal peregrine populations can contribute to this cause, with the additional benefit that the inland mountain habitats appear to be associated with lower contaminant burdens in peregrines. Monitoring all of the mid-Atlantic nesting sites over time is necessary to discern trends in contaminant exposure and productivity and the associations that might exist.

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