

Organochlorine and PCB Residues in Tissues of Raptors from Illinois, 1966-1981

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Organochlorine insecticides have been documented to have adverse effects on the reproduction and survival of raptors (Wurster 1969; Stickel 1973; Wiemeyer et al. 1975; Henny et al. 1976; Klaas et al. 1978; Ohlendorf 1981; Fleming et al. 1982; Prouty et al. 1982; Blus et al. 1983; Henny et al. 1983). The organochlorine insecticides aldrin, heptachlor, and chlordane were introduced in Illinois in 1954 for control of corn insect pests (Metcalf and Sanborn 1975). The use of organochlorine insecticides reached a peak in Illinois during 1967 when they were applied to an estimated 5.6 million of the 10 million acres planted to corn (Steffey et al. 1984). Aldrin was applied to an estimated 44.9 million acres of corn soil from 1956 through 1977, heptachlor was used on an estimated 11.6 million acres from 1956 through 1978, and chlordane, which contains heptachlor, was applied to an estimated 2.5 million acres from 1956 through 1978 (Steffey et al. 1984). DDT was used on fields planted to corn from 1945 through 1965 (Steffey et al. 1984). Chlorinated hydrocarbon insecticides were no longer applied to corn soil in Illinois after 1978 (MacMonegle et al. 1984). Polychlorinated biphenyls (PCBs) are industrial pollutants that were introduced in 1929 and are currently present throughout the environment (Dustman et al. 1971).

Because of the persistence of these chemicals in the soil (Kearney et al. 1969; Nash and Woolson 1967; Pimentel 1971), their bioaccumulation in the upper levels of the food chain (Frank et al. 1977; Price 1977), and the difficulty in obtaining samples from protected species such as raptors, we monitored the occurrence of residues of selected organochlorine insecticides and PCBs in tissues of birds of prey to document the level of contamination near the end and following the period of maximum use of chlorinated hydrocarbons in Illinois. The residues examined were DDE (a metabolite of DDT), dieldrin (a metabolite of aldrin), heptachlor epoxide (a metabolite of heptachlor), and PCBs (quantified as Aroclor 1254).

MATERIALS AND METHODS

A total of 68 individuals representing 14 species of raptors were obtained from 1966 through 1981. Most birds were collected from

1972 through 1979. These raptors were killed on highways, confiscated by law enforcement officers, or found disabled after flying into windows or being shot. The specimens were frozen immediately upon possession. Tissue samples analyzed for pesticides were brain, liver, heart, breast muscles (pectoralis and supracoracoideus), and subcutaneous and visceral fat. Wet samples were homogenized, saponified, and then extracted with 50 ml of nanograde hexane. The hexane extract was washed three times with ultrapure water, poured through a funnel of anhydrous Na_2SO_4 to remove residual water, and then placed on a steam bath and reduced to a volume of 2-5 ml under a 3-ball Snyder column. Each sample was fractionated on a 30-gm florisil column. The first fraction was eluted with 90 ml of hexane to recover the PCBs; the second fraction, eluted with 10 percent ethyl ether/hexane, contained the remaining chlorinated hydrocarbons. The samples were then reduced to a volume suitable for gas chromatographic analysis. The analyses were performed using a Varian-Aerograph Series 2100 gas chromatograph with a ^{63}Ni electron capture detector operated at 250°C . The analytical column was a 6' x 1/4" O.D. x 2 mm I.D. glass column packed with 2.5% OV-210 and 1.0% OV-17 coated on a 100-120 mesh Supelcoport. The column temperature was maintained at 190°C and the injection port at 210°C ; the electrometer was operated at 4×10^{-10} amperes. The carrier gas was O_2 -free nitrogen at a flow rate of 40 ml/min. Retention times of sample peaks were compared with those of standard reference compounds to provide presumptive identification. Quantification was achieved by peak height comparison. Detection limits were calculated to be 0.005 ppm. Because of limited sample sizes and generally low levels of pesticides, statistical tests of differences were not attempted.

RESULTS AND DISCUSSION

The 68 hawks and owls used in this study included 17 belonging to species considered as migrants and 51 belonging to species considered as permanent residents in Illinois (Table 1). With the exception of goshawks (Accipiter gentilis), species for which we had the largest sample sizes [barred owls (Strix varia), great-horned owls (Bubo virginianus), screech owls (Otus asio), and red-tailed hawks (Buteo jamaicensis)], are permanent residents (Table 1). All of the goshawks were collected during the winter of 1972-1973 when there was an unusually large influx of this rare winter resident. Percentages of all birds ($N=68$), of migrants ($N=17$), and of permanent residents ($N=51$) in which residues of the various contaminants were detected in at least one tissue sample are as follows: DDE--76.5%, 88.2%, 72.5%; dieldrin--85.3%, 64.7%, 92.2%; heptachlor epoxide (HE) --54.4%, 35.3%, 60.8%; and PCBs--26.5%, 17.6%, 29.4%. Thus, 76.5% of all birds of prey sampled were contaminated with residues of DDE, 85.3% with dieldrin, 54.4% with HE, and 26.5% with PCBs. DDE occurred in a higher percentage of migrants (88.2%) than permanent residents (72.5%), whereas detectable levels of dieldrin, HE, and PCBs occurred in a higher percentage of permanent residents. Dieldrin residues were found in 92.2% of the permanent residents examined. Seidensticker and Reynolds (1971) reported low levels of DDE,

dieldrin, and HE in an adult great-horned owl collected in Montana in 1967 and in 100% of 3 great-horned owl eggs and 5 red-tailed hawk eggs. These authors also reported low levels of DDE and dieldrin but no HE in all 9 nesting and fledgling red-tailed hawks examined. Springer (1980) analyzed 15 red-tailed hawk eggs collected in Ohio and Kentucky from 1967 through 1978 and found that 100% contained measureable amounts of PCBs and DDE, 87% contained dieldrin, and 80% had HE.

For many organochlorine pesticides and for PCBs, the brain is the best tissue for diagnostic determination of lethal contamination, whereas sublethal levels of exposure can be evaluated from residues in other tissues (Stickel 1973; Heinz et al. 1979). Diagnostic lethal residues in the brain are comparable for a wide range of animals (Stickel 1973; Heinz et al. 1979). Diagnostic lower lethal levels in brain tissues have been estimated to be 300 to 400 ppm (wet weight) for DDE (Stickel et al. 1970; Stickel et al. 1984a), 4 to 5 ppm for dieldrin (Stickel et al. 1969), near 8 ppm for HE (Stickel et al. 1979), and about 310 ppm for PCBs (Aroclor 1254) (Stickel 1975; Stickel et al. 1984b). Of the 57 brain samples analyzed from the 68 birds of prey, detectable levels of contaminants were found in the following percentages: DDE, 54.4%; dieldrin, 68.4%; HE, 29.8%; and PCBs, 5.3%.

Five of the 57 birds of prey whose brains were analyzed had dieldrin levels that approached or exceeded the diagnostic lethal level of 4 to 5 ppm and, thus, could have died or been adversely affected by the contaminant. These five individuals included 3 resident species and 1 migrant species: great-horned owl, 5.42 ppm; red-tailed hawk, 7.94 ppm; red-tailed hawk, 5.73 ppm; marsh hawk (Circus cyaneus), 7.42 ppm; and sharp-shinned hawk (Accipiter striatus), 4.01 ppm. No birds had diagnostically lethal levels of DDE, HE, or PCBs.

Two of the 5 red-tailed hawk brains examined had dieldrin levels above the 4-5 ppm diagnostic level and a third had a concentration of 2.41 ppm. Heinz and Johnson (1982) concluded that brain concentrations of dieldrin of about 1 ppm or more could present a grave hazard to some birds. Thus, it appears that of the 4 chemicals we examined in the birds of prey from 1966 through 1981, dieldrin was physiologically the most serious contaminant.

The 7 goshawks collected during the 1972-1973 winter were the least contaminated of the various species represented by a sample size of 3 or more (Table 1). Although residues of DDE were detected in at least one tissue of all 7 goshawks, dieldrin and HE were only found in the liver of one specimen and no PCB residues were found. This low level of contamination undoubtedly is a reflection of the goshawks' association with northern nonagricultural habitat.

The highest concentrations of the contaminants measured in individual birds were DDE, 781.1 ppm in fat from a red-shouldered

Table 1. Residues of DDE, dieldrin, heptachlor epoxide, and PCBs in brain, fat, liver, breast muscle, and heart of 68 owls and hawks from Illinois, 1966-1981.

RESIDUES OF CONTAMINANTS

Species	Years	Tissue	DDE					DIELDRIN					HEPTACHLOR EPOXIDE					PCBs (1254)				
			Concentration in those contaminated, ppm wet weight*					Concentration in those contaminated, ppm wet weight*					Concentration in those contaminated, ppm wet weight*					Concentration in those contaminated, ppm wet weight*				
			Mean	Median	Range	% Contaminated		Mean	Median	Range	% Contaminated		Mean	Median	Range	% Contaminated		Mean	Median	Range	% Contaminated	
Barred owl**	1966-1979	Brain Fat	11 1.68	1.22	0.14-4.13	36.4	72.7	0.16	0.03	0.02-0.99	54.6	0.11	0.04	0.01-0.41	9.1	3.26	3.26	---	---	---	---	---
(Strix varia)		Liver	12 2.75	2.49	0.60-5.40	33.3	83.3	28.81	2.44	1.41-132.12	33.3	5.48	5.48	0.80-10.16	16.7	487.80	487.80	---	---	---	---	---
		Breast	13 2.44	0.96	0.30-8.43	38.5	76.9	0.82	0.06	0.02-3.74	66.7	0.45	0.04	0.01-1.40	8.3	6.91	6.91	---	---	---	---	---
		Heart	11 1.96	1.36	1.12-4.02	36.4	81.8	0.25	0.05	0.005-1.42	53.9	0.13	0.02	0.01-0.73	7.7	2.39	2.39	---	---	---	---	---
								0.88	0.08	0.02-4.22	54.6	0.04	0.03	0.02-0.09	18.2	3.69	3.69	0.02-7.36				
Great-horned owl**	1974-1981	Brain Fat	14 2.38	0.82	0.004-10.21	85.7	78.6	1.12	0.84	0.03-5.42	35.7	0.50	0.32	0.02-1.09	7.1	13.10	13.10	---	---	---	---	---
(Bubo virginianus)		Liver	16 2.80	0.48	0.04-10.52	43.8	93.8	14.22	12.08	0.70-32.00	66.7	1.59	1.59	0.15-3.04	25.0	16.17	16.17	---	---	---	---	---
		Breast	15 4.48	1.02	0.04-18.49	73.3	100.0	0.57	0.11	0.004-5.57	12.5	0.32	0.32	0.13-0.51	18.8	46.81	34.24	8.66-97.52				
		Heart	15 1.97	0.61	0.04-8.50	46.7	100.0	1.10	0.30	0.01-7.27	33.3	1.11	0.62	0.01-4.00	33.3	25.29	9.51	0.004-105.70				
								0.24	0.22	0.03-0.52	33.3	0.25	0.31	0.02-0.51	20.0	10.82	9.99	0.37-22.10				

Table 1 (cont.)

DDE			DIELDRIN				HEPTACHLOR EPOXIDE				PCBs (1254)								
Species	Years	Tissue	N	Contaminated		Range	%		Contaminated	Mean	Median	Range	%		Contaminated	Mean	Median	Range	
				%	Contaminated		%	Contaminated											
Screech owl (Otus asio)	1967-1979	Brain	7	71.4	2.40	0.29	0.06-10.50	57.1	1.19	0.84	0.10-3.00	14.3	0.42	0.42	0	25.0	104.36	104.36	---
		Fat	4	75.0	14.07	9.86	9.82-22.52	50.0	0.41	0.41	0.05-0.77	25.0	0.11	0.11	---	10.0	3.81	3.81	---
		Liver	10	60.0	3.97	0.33	0.01-22.27	70.0	0.40	0.46	0.01-0.71	40.0	0.44	0.46	0.02-0.83	20.0	0.13	0.13	0.12-0.13
		Breast	8	87.5	0.59	0.57	0.01-1.59	50.0	0.53	0.59	0.02-0.94	12.5	0.44	0.44	---	0	---	---	---
		Heart	9	55.6	1.46	0.92	0.40-3.79	50.0	0.69	0.37	0.04-2.41	33.3	0.29	0.24	0.08-0.54	0	---	---	---
Long-eared owl (Asio otus)	1977-1979	Brain	4	25.0	0.36	0.36	---	75.0	0.33	0.02	0.0002-0.96	50.0	0.52	0.52	0.08-0.97	25.0	1.96	1.96	---
		Liver	3	66.7	28.14	28.14	0.54-55.74	100.0	0.07	0.01	0.004-0.21	33.3	0.01	0.01	---	33.3	7.22	7.22	---
		Breast	3	33.3	1.61	1.61	---	33.3	0.19	0.19	---	33.3	0.11	0.11	---	0	---	---	---
		Heart	3	33.3	214.97	214.97	---	33.3	0.70	0.70	---	0	---	---	0	---	---	---	---
Short-eared owl (Asio flammeus)	1975	Brain	1	0				100.0	0.10	0.10	---	0	---	---	0	---	---	---	---
		Liver	1	0				100.0	0.06	0.06	---	0	---	---	0	---	---	---	---
		Breast	1	0				100.0	0.17	0.17	---	0	---	---	0	---	---	---	---
		Heart	1	0				100.0	0.09	0.09	---	100.0	0.06	0.06	---	0	---	---	---
Saw-whet owl (Aegolius acadicus)	1976	Brain	1	100.0	3.67	3.67	---	0				0	---	---	0				
		Liver	1	0				100.0	0.77	0.77	---	0	---	---	0				

Table 1 (cont.)

Species	Years	Tissue	DDE					DIELDRIN					HEPTACHLOR EPOXIDE					PCBs (1254)				
			% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range
Red-tailed hawk***	1971-1978	Brain	40.0	4.10	4.10	0.88-7.31	100.0	3.23	2.41	0.002-7.94	40.0	0.001	0.001	0.0003-0.002	0				0			
		Liver	33.3	0.32	0.32	0.21-0.43	100.0	2.58	1.53	0.006-7.42	33.3	0.42	0.42	0.003-0.84	0				0			
(Buteo jamaicensis)		Breast	66.7	0.85	0.14	0.002-3.10	100.0	1.54	0.66	0.04-4.27	33.3	0.02	0.02	0.004-0.04	16.7	4.37	4.37	---				
		Heart	50.0	0.16	0.14	0.004-0.34	100.0	3.36	2.49	0.004-11.74	16.7	0.74	0.74	---	0				0			
Goshawk***	1972-1973	Brain	33.3	0.94	0.94	0.76-1.11	0				0				0				0			
(Accipiter gentilis)		Fat	100.0	8.26	10.00	0.29-12.85	0				0				0				0			
		Liver	57.1	11.48	11.04	1.35-20.58	14.3	4.50	4.50	---	14.3	1.50	1.50	---	0				0			
		Breast	71.4	3.33	1.04	0.50-11.76	0				0				0				0			
Red-shouldered hawk***	1967-1975	Brain	100.0	1.85	1.85	0.43-3.27	100.0	0.46	0.46	0.05-0.87	0				0				0			
		Fat	100.0	781.10	781.10	---	100.0	195.30	195.30	---	100.0	1.42	1.42	---	0				0			
		Liver	50.0	0.36	0.36	---	50.0	0.21	0.21	---	0				0				0			
(Buteo lineatus)		Breast	100.0	2.90	2.90	0.93-4.86	100.0	0.84	0.84	0.41-1.27	50.0	0.10	0.10	---	0				0			
		Heart	50.0	4.54	4.54	---	100.0	0.16	0.16	0.05-0.27	0				0				0			
Rough-legged hawk***	1977-1978	Brain	0				100.0	0.39	0.39	0.04-0.74	50.0	0.29	0.29	---	0				0			
		Fat	100.0	4.32	4.32	---	100.0	3.21	3.21	---	0				100.0	0.31	0.31	---	100.0	0.31	0.31	---
		Liver	0				100.0	0.02	0.02	0.017-0.02	50.0	0.05	0.05	---	50.0	3.02	3.02	---	50.0	3.02	3.02	---
(Buteo lagopus)		Breast	50.0	0.02	0.02	---	100.0	0.003	0.003	0.001-0.004	50.0	0.001	0.001	---	50.0	0.14	0.14	---	50.0	0.14	0.14	---
		Heart	0				100.0	0.07	0.07	---	100.0	0.04	0.04	---	0				0			

Table 1 (cont.)

DDE				DIELDRIN				HEPTACHLOR EPOXIDE				PCBs (1254)			
Species	Years	Tissue	N	% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range	% Contaminated	Mean	Median	Range
Marsh hawk***	1975	Brain	1	100.0	94.27	94.27	---	100.0	7.42	7.42	---	0			0
		Liver	1	100.0	11.43	11.43	---	100.0	2.41	2.41	---	0			0
(Circus cyaneus)		Breast	1	100.0	41.24	41.24	---	0				0			0
		Heart	1	100.0	17.42	17.42	---	100.0	1.74	1.74	---	0			0
Sparrow hawk**	1977	Brain	1	0				0				0			0
(Falco sparverius)		Liver	1	0				100.0	0.04	0.04	---	0			0
		Breast	1	0				100.0	0.03	0.03	---	0			0
		Heart	1	0				100.0	0.04	0.04	---	0			0
Cooper's hawk**	1973	Brain	1	0				100.0	0.01	0.01	---	0			0
(Accipiter cooperii)		Liver	1	100.0	0.15	0.15	---	100.0	0.01	0.01	---	0			0
		Breast	1	100.0	0.47	0.47	---	100.0	0.04	0.04	---	100.00	0.002	0.002	---
		Heart	1	0				100.0	0.003	0.003	---	0			0
Sharp-shinned hawk***	1979	Brain	1	100.0	12.31	12.31	---	100.0	4.01	4.01	---	0			0
(Accipiter striatus)		Liver	1	0				100.0	3.71	3.71	---	0			0
		Breast	1	100.0	4.73	4.73	---	100.0	1.82	1.82	---	0			0
		Heart	1	0				100.0	2.88	2.88	---	0			0

* Standard error values of concentrations available upon request

** Generally permanent residents in Illinois

*** Generally migrants in Illinois

hawk (Buteo lineatus); dieldrin, 195.3 ppm in fat from the same red-shouldered hawk; HE, 10.16 ppm in fat from a barred owl; and PCBs, 487.8 ppm in fat from another barred owl (Table 1). Ohlendorf (1981) reported that concentrations of DDE, dieldrin, and PCBs appeared to be declining in bald eagles (Haliaeetus leucocephalus). Recent investigations of organochlorine contamination of potential avifauna prey of raptors in Illinois have also indicated a decrease in the concentrations of residues. Warner et al. (1984) reported that although the frequency of occurrence of HE, DDE, and dieldrin in ring-necked pheasant (Phasianus colchicus) chicks collected in intensively cultivated areas of east-central Illinois during 1976 and 1977 were similar to those found in pheasant chicks collected in 1968 (Anderson et al. 1970), the concentrations were less in 1976-1977. Edwards et al. (1983) reported that residues of organochlorine insecticides in woodcock (Philohelo minor), mourning doves (Zenaidura macroura), and robins (Turdus migratorius) collected in east-central Illinois during 1978-1979 probably remained widespread but at low and decreasing levels. As environmental concentrations of organochlorine insecticides decline, residues in raptors from the Midwest should correspondingly decrease below those reported in this study.

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