

Polychlorinated Biphenyls in the Global Ecosystem

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Polychlorinated biphenyls are widely dispersed in the global ecosystem, and are powerful inducers of hepatic enzymes which degrade oestrodin. Together with other chlorinated biocides, such as DDT, they could account for a large part of the aberration in calcium metabolism which has been observed in many species of birds since the Second World War.

Declining populations of raptorial and fish-eating birds in Great Britain¹ and North America² have produced thin-shelled eggs since the period after the Second World War. A widespread change in the chemical environment which affected the calcium physiology of these species evidently occurred at that time. The chlorinated hydrocarbons, which came into general use in the 1940s, may now be the most abundant synthetic pollutants present in the global environment^{3,4}. Thin eggshells have been found only in species which accumulate high concentrations of these compounds; relatively uncontaminated populations of these species continue to produce normal eggs^{5,6}.

Calcium metabolism in birds is intimately related to reproductive metabolism and is to a large extent regulated by steroids such as oestrogen and vitamin D⁷. The deposition of medullary bone, the chief source of calcium during egg and eggshell formation, is controlled by the steroid sex hormones^{8,9}, and hence deficient in vitamin D starve with low "eggshell weights". Steroids are hydroxylated and thereby degraded in vivo and in vitro by hepatic enzymes induced by exogenous, lipid-soluble substances, including the chlorinated hydrocarbons¹⁰⁻¹⁴. The relatively small amounts of chlorinated hydrocarbons required to produce this effect^{15,16,17,18}, and the discovery that small amounts of some of the DDT compounds are oestrogenic^{19,20}, have made irrelevant much of the parts per million approach to pollutant ecology based on toxicity data alone.

In both Great Britain²¹⁻²⁴ and North America²⁵ it was the decline of the peregrine falcon which initiated concern about the extent of the harmful effects of environmental contamination. In the United States the eastern population was extinct before competent observers were aware of a general, widespread decline²⁶. Breeding peregrines persist in apparently normal numbers in British Columbia²⁷ and in the Arctic^{28,29}.

In 1967 we collected an unhatched, abandoned egg of a peregrine falcon in south-western North America, where a small remnant population remains (unpublished work of M. N. K., R. W. R. and S. G. H.). Analysis of this egg (Table 1) showed that it contained almost 5 mg of p,p'-DDE (dichloro-2,3-bis(p-chlorophenyl) ethylene). Unknown peaks present in the chromatograms of the extract of the egg were unidentified until polychlorinated biphenyls (PCB) were detected in European wildlife³⁰⁻³². Positive confirmation of the identification was accomplished by mass spectrometry in Sweden³³. The retention times of the unknown peaks in the peregrine extract proved to be identical with those of several PCB compounds on DC-500 and QP-1 columns³⁴ and on a mixed SE-30:FF-1 column³⁵ of either species of birds and fish were subsequently analysed for PCB. The chlorine content of several extracts and standards was determined with a Dehman microcoulometric detector and a method of quantification of the PCB compounds was devised based on peak heights produced in the electron capture detector related to standard p,p'-DDE³⁶. The DDT compounds are destroyed by nitration³⁷ and p,p'-DDT (1,1,1-trichloro-

2,3-bis(p-chlorophenyl)ethane), DDD (1,1-dichloro-2,3-bis(p-chlorophenyl)ethane) and toxaphene are dehydrochlorinated by saponification with alcoholic KOH. PCB is not degraded by either procedure.

Table 1. CHLORINATED HYDROCARBONS IN NORTH AMERICAN PEREGRINE FALCONS

Sample	De-chloro	Total DDT*	Per-centage DDE	PCB	DDT/PCB
1. Unhatched egg High California (wet)	0-11	102	97	10-5	10
2. Redwood forest migrant from Arctic					
Breast muscle (wet)	NM	98	94	54	5-5
Breast muscle (dry)	NM	200			
Brain (wet)	NM	85	96	51	6-7
Carcass (wet)	0-07	107	92	10-7	5-5
Carcass (lipid)	02-5	1,500		1,430	
3. Immature female from Arctic					
Brain (wet)	NM	11-5	90	5-4	1-5
Liver (wet)	NM	7-5	96	4-5	1-7
Brain (wet)	0-04	9-5	94	4-5	1-9
Brain (lipid)	0-04	35		10-3	
Carcass (wet)	0-11	20-5	96	10-0	1-9
Carcass (lipid)	0-11	300		100	
4. Adult, California					
Breast muscle (wet)	NM	137	97	90	1-5
Liver (wet)	NM	77	90	87	1-4
Brain (wet)	0-31	50-5	96	54-5	1-4
Brain (lipid)	0-07	100		100	
Carcass (wet)	1-7	85	97	85	1-5
Carcass (lipid)	50	2,500		1,500	
5. Immature migrant from Arctic					
Breast muscle (wet)	NM	7-5	91	0-16	14
Breast muscle (dry)	NM	2-0		0-16	
Liver (wet)	NM	1-0	92	0-10	10
Brain (wet)	0-4	0-45	92	0-37	10
Body fat (wet)	NM	50-5	92	0-18	18
Carcass (wet)	0-44	60-7	92	0-50	12
Carcass (lipid)	0-44	60-7		0-50	
6. Immature female from Arctic					
Breast muscle (wet)	NM	1-9	96	0-4	5-4
Breast muscle (dry)	NM	0-0		1-9	

Concentrations in parts per million wet weight, dry weight or lipid weight.

* DDT residues include: p,p'-DDT, p,p'-DDE, p,p'-DDD (p,p'-DDB, p,p'-DDMU, p,p'-DDT and p,p'-DDE). NM, Not measured.

† Chlorinated hydrocarbon contents of the egg were: 4,700 µg p,p'-DDT; 4,700 µg p,p'-DDE; 50 µg p,p'-DDT; 50 µg p,p'-DDE; 37 µg p,p'-DDD; 40 µg p,p'-DDD; 12 µg heptachlor epoxide. Concentrations were calculated by assuming a volume of 0.5 ml for the egg. The peregrine chick was found alive at the time of capture from California in the Museum of Vertebrate Zoology, University of California, Berkeley, and by assuming a density of 1.0 g/ml. A second year female, captured in October on the Texas coast during migration, died in captivity shortly afterwards, no apparent cause of death. No body fat. Total body content of chlorinated hydrocarbons: 50 mg DDT, 10 mg PCB, 0-44 mg dieldrin, 0-18 mg heptachlor epoxide. Total body lipid, 0-50 g.

†† Female characteristic of both 3 and 4 were intermediate between those of typically resident California birds and the Arctic male p.p. females, as described by Wilbur³⁸. It was an immature female of the year trapped in the southern San Francisco Bay area in the winter of 1968. Observed feeding in the area for a week before capture. Died suddenly after eating a dead gull. Reddish body fat. Total body chlorinated hydrocarbon content: 10 mg DDT, 0-8 mg PCB, 0-07 mg dieldrin, 0-09 mg heptachlor epoxide. Total body lipid, 0-50 g.

††† Adult female. Trapped in the southern San Francisco Bay area in the winter of 1968. Observed feeding in the area for a month before capture. Died shortly afterwards with no apparent cause of death and no body fat. Total body chlorinated hydrocarbon content: 50 mg DDT, 40 mg PCB, 1-9 mg dieldrin, 0-50 mg heptachlor epoxide. Total body lipid, 110 g.

†††† First year female trapped on the Texas coast in October. Shot at Pt. Mugu, California, the following January and was shot by a hunter 3 weeks later. Abundant body fat. Total body content: 1 mg DDT, 0-5 mg PCB, 0-44 mg dieldrin, 0-17 mg heptachlor epoxide. Total body lipid, 110 g.

††††† First year female trapped on the Texas coast in October. Died shortly afterwards of heart pneumonia.

In Table 1 are presented the results of analyses of peregrine falcons which died from a variety of causes

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directly after being trapped for falconry. Significant amounts of PCB were present in Arctic peregrines only a few months old (Nos. 4 and 6), but higher residues were present in a second year Arctic bird (No. 2) and exceptionally high residues were present in an adult trapped in California (No. 4). In birds 2 and 4 the total lipid reserves were very low, and in both the brain concentrations of DDE and PCB were high, perhaps at toxic levels. Fat mobilization during reproduction or in times of starvation or stress could be expected to cause significant changes in the internal distribution of chlorinated hydrocarbons. Steroid hydroxylase activity in the liver might increase at this time. Dieldrin concentrations were lower than in the peregrines analysed in Britain, but DDE concentrations were approximately comparable.

Table 2 presents the results of analyses of peregrine prey species, remains of which were collected at eyries in Baja California, Mexico. With the exception of the remains of one mourning dove (*Zenaidura macroura*) and of three fish bats (*Pteropus vivesi*), prey material found at four eyries consisted of sea birds. Of three, the cormorant (*Phalacrocorax*), the black petrel (*Loonania melanotos*) constituted 32 and 28 per cent, respectively, of the remains.

Black petrels, like other petrels and shearwaters (Table 3), contained especially high concentrations of both DDT and PCB. Reproductive success in this population of peregrines seems to be abnormal. In 1968 no pairs were observed to hatch or fledge more than a single young. In the past, two to four young, the number normally produced by healthy peregrines^{11,17}, were raised by each pair (L. W. Walker, personal communication). The thickness of fragments of a peregrine eggshell, with its membranes, collected in 1968 below an eyrie where one young hatched, was 0.24 mm, a decrease of 34 per cent from the mean thickness of 0.34 mm ± 0.015 mm (90 per cent confidence level) in twenty-three eggs collected in the area before 1947. The region is wilderness, with little or no human interference, and is remote from sources of pollution.

In California, numbers of breeding peregrines have been reduced by at least 80 per cent in recent years. The remaining few pairs, however, seem to be reproducing normally and rear, when undisturbed, between two and four young each year. They are found, like most of the surviving pairs in Great Britain^{11,17}, in a relatively uncontaminated region and seem to be feeding on birds

which contain low concentrations of chlorinated hydrocarbons. A pair which fledged three young in 1968 fed chiefly on passerines and columbiformes during the breeding season. Both prey groups are relatively uncontaminated (M. N. K., R. W. H., and S. G. H., in preparation). A clutch of eggs collected in this region weighed as much as the eggs obtained in previous years, whereas other peregrine eggs from California collected since the Second World War have been diminished (e.g. 2, and D. W. Anderson, D. Hickey and R. F. Christensen, in preparation). Despite official protection the surviving birds are still subjected to shooting by sportsmen and to harassment at the eyries. If these could be effectively reduced the population might yet survive.

In Table 3 are presented the results of analyses of marine and terrestrial birds and of three species of freshwater fishes for PCB and DDT content and concentration. From the ratio of total DDT to PCB, it is apparent that regional fallout patterns exist. In most of the birds from San Francisco Bay which have been analysed, including peregrine falcons (Table 1) and eggs of the western gull, the Caspian tern and the black-crowned night heron (Table 3), the ratio was between one and two. Another black-crowned night heron egg had a typically "oceanic" profile, suggesting that the adult female had wintered along the coast. In most of the birds from the Farallon Islands, which are 37 miles west of the Golden Gate Bridge, this ratio was between 2 and 5. In the Gulf of Panama, where PCB contamination might come from the Canal Zone and industrial areas in Panama City, the ratio is between 1 and 3 (Table 3).

In the Gulf of California, a region relatively remote from the sources of either DDT or PCB contamination, the ratio was in most cases approximately 5 or 10. This was true in the eggs of the peregrine falcon (Table 1), in all of the black petrels and least petrels analysed, in the (Crawley's) murrelets (Table 2), in five of six osprey eggs and in six of seven western gull eggs (Table 3). Among the exceptions the fish bat, with low concentrations of both DDT and PCB, was the most divergent, with a ratio of 43 (Table 2). It is not clear whether this reflects differences between avian and mammalian physiology or a fundamental difference in feeding habits. The other exceptions include species which are present in the area only during the breeding season (Table 2).

In sea birds from the Pacific the ratio was usually between 5 and 10 (Table 3). PCBs were not found in eggs of the Arctic peregrine from Laysan Island, Anderson et al.

Table 2. PCB AND DDT IN PREY SPECIES OF PEREGRINE FALCONS IN THE GULF OF CALIFORNIA

	N	DDT (ppm)	DDT (p.p.m.)	Percentage DDE	PCB (ppm)	PCB (p.p.m.)	DDT/PCB
Least petrel (13)							
Whole body	8	—	0.28-0.36	97	NM†	—	—
Least petrel (10)							
Whole body	8	810 (800-1,266)	0.2 (W)	81	—	1.0 (W)	0.2
Least petrel (8)							
Whole body	8	89	0.2 (W)	82	—	0.86 (W)	0.8
Egg	8	80	—	84	1.2-4.0	—	10
Fish bat (2)							
Whole body	7	88 (10-31)	0.71 (W)	88	0.58 (0.46-1.08)	0.08 (W)	43
Crawley's murrelet (2)							
Egg (one clutch)	2	820 (177-324)	80 (L)	80	—	4.4 (L)	0.7
Whole bird, adult	1	371	0.31 (W)	85	—	0.60 (W)	7.0
Whole bird, adult	1	285	0.4 (W)	85	—	0.20 (W)	0.2
Elegant tern (1)							
Egg	1	11.8 (0.4-31.3)	0.0 (L)	90	—	1.8 (L)	0.0
Hermann's gull (1)							
Egg	3	105 (24-278)	48 (L)	85	—	0.1 (L)	0.0

Content in µg of whole bodies and eggs. Concentrations in p.p.m. wet weight (W) or lipid weight (L).
 † All specimens were collected in the vicinity of four peregrine eyries in Baja California. Numbers in parentheses are the number of remains of each prey species which were found at the eyries. Also found were remains of one cormorant (*Phalacrocorax* sp.), one red phalarope (*Phalaropus fulicarius*), one northern phalarope (*Phalaropus lobatus*) and one mourning dove (*Zenaidura macroura*), but local specimens of these species were not analysed. Eggs of least petrel, elegant tern and Hermann's gull were from different clutches. Least petrel: *Loonania melanotos*; fish bat: *Pteropus vivesi*; Crawley's murrelet: *Endomyscus crawleyi*; elegant tern: *Thalasseus elegans*; Hermann's gull: *Larus harrisi*.

† 1.0. Recovered. † Interfered peaks on chromatograms.
 ‡ Both eggs also contained 0.2 p.p.m. dieldrin (lipid weight) and 0.17 p.p.m. endrin (lipid weight).

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In sea birds from the Pacific the ratio was usually between 5 and 10 (Table 3). PCB was not found in eggs of the Adélie penguin from Cape Crozier, Antarctica.

Table 2 (continued)

19. <i>Spizella socialis</i> . Malibu Park, San Diego, February 1968. First year female, third of trichomoniasis.	
20. <i>Lepus arizonae</i> . San Luis Obispo Co., Calif., April 1968. Unhatched egg in nest where one young was raised. Egg also contained 4.7 µg of dieldrin, 1.9 µg of heptachlor epoxide, but no PCB.	
21. <i>Passer domesticus</i> . Gulf of California, Mex. March 1968. One egg also contained 0.10 p.p.m. dieldrin and 0.25 p.p.m. heptachlor epoxide.	
22. <i>Falco sparverius</i> . Imperial Valley, Imperial Co., Calif., December 1967. Eggs from Davis, California, 1968.	
23. <i>Ardea herodias</i> . Range from different dieldrin, San Francisco Bay, May 1967.	
24. <i>Larus argentatus</i> . Standard error, 56 per cent confidence limits.	
25. <i>Struthio fuscus</i> . San Diego Bay, May 1967.	
26. <i>Heteropoda tubicola</i> . 1967.	
27. <i>Phalaropus fulicarius</i> . Monterey Bay, November 1, 1968.	
28. <i>Felis concolor</i> . Farallon Islands, May 1967.	
29. <i>Myiarchus cinerascens</i> . Farallon Islands, April 1968.	
30. <i>Condalia bicolor</i> . Monterey Bay, November 1, 1968.	
31. <i>Tringa melanoleuca</i> . Monterey Bay, November 1, 1968.	
32. <i>Ardea herodias</i> . San Diego Bay, May 1967.	
33. <i>Felis concolor</i> . Gulf of Santa Cruz Co., Calif., March 1968. Chetoh B from Yolo Co., Calif., April 1968.	
34. <i>Merula migratoria</i> . Davis, Calif., December 1967.	

In these, however, the amount of DDT was very low; with a DDT:PCB ratio of 18, no PCB would have been detected. A larger amount of fat material from Antarctic organisms, which would contain more DDT, would therefore have to be analysed before concluding that PCB has not yet reached the Antarctic.

Individuals of species resident in industrial areas have, as expected, higher PCB levels than individuals of the same species from more remote regions. Analysis of two eggs from ten clutches of the western gull from the Farallon Islands showed an average PCB content of 130 ± 55 µg (95 per cent confidence level). PCB content of four eggs from three clutches from an island in San Francisco Bay averaged 805 µg with a range from 580 to 1,310 (Table 3). Seven eggs from seven clutches in the Gulf of California contained 45 ± 30 µg of PCB. The DDT content of the eggs from the Farallons was not significantly different from that of the Baja California eggs.

Birds from the Gulf of California also contain dieldrin and endrin (Table 2 and 3), but the number of analyses as yet insufficient to compare their relative abundance with that of DDT and PCB. Despite the inability of the Shell Chemical Company to find any chlorinated hydrocarbons in the Gulf of California and at the mouth of the Colorado River, which drains into the Gulf, it is likely that some do enter from the Colorado River and from agricultural areas in western Mexico. A significant fraction, however, must come from the atmosphere²², and air transport best explains the presence of PCB in remote areas. In extracts from the Gulf of California one PCB compound was present in relatively small concentrations. This compound is readily degraded by ultraviolet light irradiation in laboratory experiments, and may therefore be selectively degraded in the atmosphere. Although PCB is not soluble in water, it has a low but finite vapour pressure²³. Incorporation of materials containing PCB would greatly increase the rate of entry into the atmosphere. PCB is used in the manufacture of many industrial products, so the high amounts found in San Francisco Bay, Puget Sound²⁴ and San Diego Bay presumably result from direct discharge of industrial wastes into these waters and from local fallout.

The presence of PCB in the few land birds and freshwater fish analysed indicates that it is also distributed among continental biotopes in North America. Invertebrates could therefore acquire PCB, as well as the other chlorinated hydrocarbons, over all their global range. No PCB residue data are available for prey species from the Atlantic, but compounds which seem to be PCB have been isolated from seals²⁵. A second year male peregrine spent the winter of 1967-68 on Isla Páchoa, Panama, where the pelican, booby and frigate-bird eggs of Table 3 were collected. The Cooper's hawk which was analysed, a species which also preys upon birds, contained high residues of both DDT and PCB (Table 3). Cooper's hawks have declined in eastern North America and in some regions have produced thin-shelled eggs²⁶. Several

species of raptors do not accumulate high amounts of the organochlorine compounds (Table 3), a result of the very low residue levels usually found in their prey. American kestrels and barn owls are common residents of California cities. The white-tailed kite, which was near extinction 60 years ago in California, is now abundant in areas where insecticide use is intense, yet because it preys primarily upon the short-lived and herbivorous vole, *Microtus*, the species accumulates very little DDT or PCB (Table 3).

Previous work¹ has shown that both DDT and dieldrin induce hepatic enzymes in the pigeon which, in an *in vitro* preparation, increases the metabolism of progesterone and testosterone. This work has now been extended to study the metabolism of oestradiol by enzymes induced by p,p'-DDE, technical DDT (Dupont) and PCB (Aroclor 1248). The experimental procedure previously described¹ was followed except that the chlorinated hydrocarbons were injected intramuscularly rather than given orally, and in the separation of oestradiol and its metabolites the solvent system used was the upper layer of a mixture of benzene:heptane:methanol:water 7:3:8:3. The strips were monitored with an autoradiographic strip scanner. The profiles obtained for the various inducing agents are shown in Fig. 1. It will be noted that the profiles of the metabolites obtained after enzyme induction using DDE and DDT are identical, but that different metabolite is produced by the enzyme induced by PCB. The amount of metabolites formed was calculated from the radioactivity of the peaks. The results obtained are given in Table 4.

TABLE 4. INCREASE OF OESTRADIOL METABOLISM BY PIGEON LIVER HOMOGENATE FROM BIRDS TREATED WITH VARIOUS CHLORINATED HYDROCARBONS

Amount of polar metabolites formed in 30 minutes	
Control	22.2 ± 0.6
DDE (40 mg/kg)	76.1 ± 1.1
DDT (40 mg/kg)	96.1 ± 1.2
PCB (80 mg/kg)	100.0 ± 10.5

Each figure is the average with standard deviation of a group of four birds. In all cases 500 nmole of oestradiol-4,7,10 (100 mCi/mole) was present in the incubation mixture. Incubation time 30 min. Weight of microsomal fraction used 500 mg. All chlorinated hydrocarbons were injected intramuscularly into the pectoral muscle 7 days before death.

Birdy concentrations of 40 p.p.m. of p,p'-DDE significantly increased the rate of oestradiol degradation by the induced enzymes in the experimental conditions. On a weight basis the PCB preparation had an oestradiol-degrading potential approximately five times that of p,p'-DDE or technical DDT. Both DDE and PCB, which are apparently the most abundant of the chlorinated hydrocarbon pollutants in the global ecosystem, have therefore the capacity to produce substantial physiological effects in birds.

Studies on the activity of induced enzymes in the rat at various times after a single injection of DDT or dieldrin have been made by Ghazal et al.²⁷ He found that it took 70 days for the activity to fall to half its maximum value in the case of DDT and 16 days in the case of dieldrin. Experiments with dieldrin on pigeons

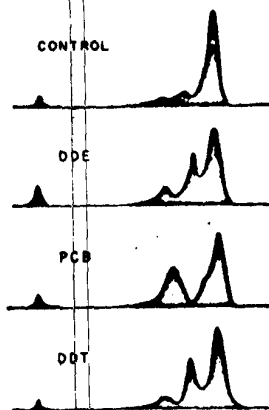


Fig. 1. Chromatographic separation of estradiol and its metabolites. The large peak is analyzed estradiol.

show that only a quarter of maximum activity remains after a month. Thus there is evidence that the effects of these induced enzymes can persist over a long period of time although more studies are needed to determine the steroid degrading potentials of livers of those species contaminated with chlorinated hydrocarbons. The profile of the metabolites should be some indication of the history of exposure to chlorinated hydrocarbons.

The reductions in eggshell thickness and eggshell weight increase the chances of egg breakage¹¹⁻¹³ and water intrusion, which affects incubation success¹⁴, might be impaired. The environment in which birds now exist is therefore no longer the same as that in which they evolved; it is unlikely that any species has the genetic capacity to meet the selection pressures resulting from the abrupt environmental change which has produced the thin eggshells. The peregrine falcon is a species long highly revered and respected. O. H. Thayer¹⁵ has described it as "the embodiment of noble respect and lonely freedom". An irony therefore exists in the fact that the peregrine may be the first species to be extirpated by global contamination.

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Tsunamis on the Moon?

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The spacing of the five annular mountain rings around Mare Orientale fits a dispersion curve for gravity waves on a 50 km "liquid" layer overlying a rigid basement.

This question whether the multiple ring-like mountain structures surrounding most of the lunar craters larger than 150 miles in diameter might be "Grazing tsunami" set in motion by the shock waves caused by impacting

planetesimals, was first brought to my attention by R. B. Baldwin (personal communication). Baldwin points out that in several cases (including the giant Mare Imbrium) two or more rings can be made out

begin of a large its to return current from two p caluul Mare are al The Moon's plan is materi bech calu calu pchit basin, the m later at expho- ragon km in the m of the into a the rest charact- in the crate w For me rigid ha posited mchons taneous ring-apo wave by.

Table 1.
Ring No.
Elevation
(m) ±
Radius
to Ocean
to Ring
to Coast

Mountain
The in- vided l ively (or Van Dorn polar sea placemen- taneous) . i by

where $\Delta\theta$ transform $2\pi/\lambda$ is length, ω The green number θ