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Polychlorinated Biphenyls in the Global Ecosystem

by

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Polychlorinated biphenyls are widely dispersed in the global ecosystem, and are powerful inducers of hepatic enzymes which degrade oestradiol. 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 Britain[1] and North America[2] 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]. 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[1,2].

Calcium metabolism in birds is intimately related to reproductive metabolism and is to a large extent regulated by steroids such as oestrogen and vitamin D[4]. The deposition of medullary bone, the chief source of calcium during egg and eggshell formation, is controlled by the steroid sex hormones[1-7], and hens deficient in vitamin D lay eggs with lower eggshell weights[8]. Steroids are hydroxylated and thereby degraded *in vivo* and *in vitro* by hepatic enzymes induced by exogenous, lipid-soluble substances, including the chlorinated hydrocarbons[9-14]. The relatively small amounts of chlorinated hydrocarbons required to produce this effect[9,11,15-18], and the discovery that small amounts of some of the DDT compounds are oestrogenic[9,19,20], have made irrelevant much of the parts per million approach to pollutant ecology based on toxicity data alone.

In both Great Britain[21-24] and North America[25] 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 decline[25]. Breeding peregrines persist in apparently normal numbers in British Columbia[26] and in the Arctic[27,28].

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-2,2-*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 wildlife[29-31]. Positive confirmation of the identification was accomplished by mass spectrometry in Sweden[30]. The retention times of the unknown peaks in the peregrine extracts proved to be identical with those of several PCB compounds on *DC*-200 and *QF*-1 columns[3] and on a mixed *SE*-30:*QF*-1 column. Other species of birds and fish were subsequently analysed for PCB. The chlorine content of several extracts and standards was determined with a Dohrman 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'*-DDE[3]. The DDT compounds are destroyed by nitration[32] and *p,p'*-DDT (1,1,1-trichloro-2,2-*bis*(*p*-chlorophenyl)ethane), DDD (1,1-dichloro-2,2-*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	Dieldrin	Total DDT*	Percentage DDE	PCB	DDT/PCB
1. Unhatched egg†					
Baja California (wet)	0·11	102	97	10·2	10
2. Second year‡ migrant from Arctic					
Breast muscle (wet)	NM	99	94	28	3·5
Breast muscle (dry)		296		84	
Brain (wet)	NM	85	98	21	4·7
Carcass (wet)	0·87	70	93	19·7	3·5
Carcass (lipid)	62·5	5,000		1,420	
3. Immature, California§					
Breast muscle (wet)	NM	14·4	90	9·4	1·5
Liver (wet)	NM	7·7	92	4·5	1·7
Brain (wet)	0·04	2·8	89	1·5	1·9
Brain (lipid)	0·50	36		19·3	
Carcass (wet)	0·11	20·2	92	10·8	1·9
Carcass (lipid)	1·6	300		160	
4. Adult, California‖					
Breast muscle (wet)	NM	127	87	98	1·3
Liver (wet)	NM	77	80	57	1·4
Brain (wet)	0·31	49·5	86	34·6	1·4
Brain (lipid)	3·7	595		415	
Carcass (wet)	1·7	85	87	65	1·3
Carcass (lipid)	50	2,600		1,980	
5. Immature¶ migrant from Arctic					
Breast muscle (wet)	NM	2·3	81	0·16	14
Breast muscle (dry)		7·8		0·54	
Liver (wet)	NM	1·0	92	0·10	10
Brain (wet)	NM	0·43	83	0·037	12
Body fat (wet)	NM	50·3	82	3·2	16
Carcass (wet)	0·07	9·3	82	0·80	12
Carcass (lipid)	0·44	63·7		5·5	
6. Immature Arctic migrant**					
Breast muscle (wet)	NM	1·9	89	0·6	3·4
Breast muscle (dry)		6·0		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'*-TDE), *p,p'*-DDMU, *o,p'*-DDT and *o,p'*-DDE. NM, Not measured.

† Chlorinated hydrocarbon contents of the egg were: 4,700 μg *p,p'*-DDE; 40 μg *o,p'*-DDE; 29 μg *p,p'*-DDT; 7·4 μg *p,p'*-DDD; 37 μg *p,p'*-DDMU; 5 μg dieldrin; 12 μg heptachlor epoxide. Concentrations were calculated by assuming a volume of 47·3 ml., the average value obtained by measuring eleven clutches of peregrine eggs from California in the Museum of Vertebrate Zoology, University of California, Berkeley, and by assuming a density of 1·0.

‡ 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: 35 mg DDT, 10 mg PCB, 0·41 mg dieldrin, 0·18 mg heptachlor epoxide. Total body lipid, 7 g.

§ Plumage characteristics of both 3 and 4 were intermediate between those of typically resident California birds and the Arctic race *F.p. tundrius*, as described by White[33]. No. 3 was an immature female of the year trapped in the southern San Francisco Bay area in the winter of 1966. Observed feeding in the area for a week before capture. Died suddenly after eating a dead gull. Moderate body fat. Total body chlorinated hydrocarbon content: 13 mg DDT, 6·8 mg PCB, 0·07 mg dieldrin, 0·09 mg heptachlor epoxide. Total body lipid, 43 g.

‖ Adult female. Trapped in the southern San Francisco Bay area in the winter of 1966. 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: 52 mg DDT, 40 mg PCB, 1·0 mg dieldrin, 1·0 mg heptachlor epoxide. Total body lipid, 20 g.

¶ First year female trapped on the Texas coast in October. Lost at Pt Mugu, California, the following January and was shot by a sportsman 3 weeks later. Abundant body fat. Total body content: 7 mg DDT, 0·6 mg PCB, 0·05 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 heat prostration.

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

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Table 8. PCB AND DDT IN THE GLOBAL ECOSYSTEM

Species	N	Total DDT (µg)	Total DDT (p.p.m.)	Percentage DDE	PCB (µg)	PCB (p.p.m.)	DDT/PCB
White crapple (1)	1	—	1·83 (W)	6	—	0·004 (W)	475
Black crapple (2)	1	—	2·10 (W)	6	—	0·003	690
Bluegill (3)	1	—	5·5 (W)	6	—	0·005	1,200
Adelie penguin (4)							
Eggs	5	0·78 (0·59–1·04)	0·128 (L)	74	< 0·044 µg/egg		> 18
Western grebe (5)							
Breast muscle	1	—	26·4 (W)	5	—	0·098 (W)	270
Fulmar (6)							
A	1	—	0·41 (W)	76	—	0·08 (W)	5
B	1	—	3·4 (W)	89	—	0·34 (W)	10
C	1	10,475	17·5 (W)	96	3,900	6·5 (W)	2·7
Pink-footed shearwater (7)	1	2,000	3·0 (W)	93	277	0·42 (W)	7·2
Sooty shearwater (8)							
A	1	—	12·3 (W)	04	—	1·2 (W)	10
B	1	—	10·3 (W)	86	—	0·9 (W)	12
C	1	1,205	2·3 (W)	85	NM		
Slender-billed shearwater (9)	1	—	32·0 (W)	92	—	2·1 (W)	15
Ashy petrel (10)	3	2,158 (1,644–2,826)	59·3 (W)	95	389 (298–482)	9·8 (W)	5·5
Brown pelican (11)							
Eggs, Panama	6	59 (18–183)	11·5 (L)	61 (47–78)	23·1 (18–30)		1·55 (0·97–3·2)
Eggs, Baja California	2	53 (47–59)	10·0 (L)	81	10·4 (9·0–11·7)	—	6·5, 4·0
Frigate-bird (12) eggs, Panama	3	9·6, 8·7, 30·0	—	88	8·4, 5·7, 84·0	—	1·1, 1·5, 0·4
Brown booby (13) eggs, Panama	4	20·8 (16·4–24·5)	8·2 (L)	89	12·2 (6·5–18·9)	4·8 (L)	1·7
Brandt's cormorant (14) eggs	17	326	—	91	113	—	2·9
Pelagic cormorant (15) eggs	2	128 (125–130)	—	90	62 (48–75)	—	2·1
Cinnamon teal (16)	1	4,340	10·9 (W)	70	—	0·91	12
White-tailed kite (17)							
Eggs, clutch *A*	4	11·4 (9·8–12·9)	0·76 (W)	77	6·4 (5·5–7·8)	—	1·8
Unhatched egg *B*	1	5·1	0·34 (W)	82	0·84	—	6·0
Unhatched egg *C*	1	5·3	0·35 (W)	73	3·6	—	1·5
Clutch *D*	2	8·3/egg	—	82	4·3/egg	—	2·0
Clutch *E*	3	10·2/egg	9·0 (L)	57	4·0/egg	—	2·6
Clutch *F*	4	31·0/egg	—	80	3·8/egg	—	8·5
Cooper's hawk (18)	1	8,500	25·2 (W)	90	—	6·3	4·0
Golden eagle (19) egg	1	150	2·0 (W)	98	17·5	0·23 (W)	8·5
Osprey (20), Baja California	6	127 (30–264)	55 (L)	85	(7·3, 3·0, 103) (24, 7·0, 19)	—	8·5, 10·1, 1·3, 11·2, 9·7, 11·1
Merlin (21)	1	435	2·9 (W)	94	55·4	0·39 (W)	12·7
American kestrel (22)							
Whole body, adult	1	5·1	0·044	39	3·7	0·031	1·4
Eggs, two clutches	6	2·4/egg	0·20 (W)	93	1·0/egg	0·09 (W)	2·3
Black-crowned night heron (23)							
Egg	1	541	—	89	330	—	1·6
Egg	1	869	—	99	23	—	36
Western gull (24)							
Eggs, San Francisco Bay, three clutches	4	803 (632–1,123)	—	85	805 (589–1,310)	—	1·0
Eggs, Farallon Is., ten clutches	10	412 ± 102	—	89	136 ± 55	—	3·0
Eggs, Baja Calif., seven clutches	7	385 ± 230	—	97	45 ± 30	—	9·1, 10·5, 5·0, 12·3, 10·6, 10·7, 10·5
Forster's tern (25) eggs	2	665 (598–732)	—	89	114 (91–137)	—	5·8
Caspian tern (26)							
Eggs, San Francisco Bay	2	1,269 (1,216–1,322)	—	89	805 (660–950)	—	1·7
Eggs, San Diego Bay	5	1,430 (901–2,430)	—	88	1,010 (550–1,600)	—	1·4
Red phalarope (27)	1	—	0·78 (W)	79	—	0·10	8
Common murre (28) eggs	6	1,945 (932–3,621)	151 (L)	90	558 (364–1,010)	45 (L)	3·5
Cassin's auklet (29)	1	—	5·8 (W)	98	—	0·16	36
Ancient murrelet (30)	1	—	0·75	90	—	0·15	5
Rhinoceros auklet (31)	1	—	2·7	97	—	0·36	8
Mourning dove (32)	2	12·0, 33·1	0·19 (W)	77, 93	Not detected	—	—
Barn owl (33)							
Eggs, one clutch	3	27·7/egg	1·25 (W)	95	10·4/egg	0·47 (W)	2·7
Eggs, one clutch	2	143/egg	6·6 (W)	96	14·4/egg	0·66 (W)	10
Meadowlark (34)	2	21·2, 418	0·18, 3·3 (W)	77, 93	2·5, 28·1	—	8·4, 15·9

Content in µg and concentrations in p.p.m. wet weight (W) or lipid weight (L).

Unless otherwise specified, analyses were of whole bodies. Endrin and dieldrin were identified on the basis of retention times on both *QF*-1 and *DC*-200 columns.

1. *Pomoxis annularis*, 284 g, Clear Lake, Lake Co., Calif., May 1968.
2. *Pomoxis nigromaculatus*, 212 g, Clear Lake, Lake Co., Calif., May 1968.
3. *Lepomis macrochirus*, 229 g, Clear Lake, Lake Co., Calif., May 1968.
4. *Pygoscelis adeliae*, Cape Crozier, Antarctica, October 1967.
5. *Aechmophorus occidentalis*, Clear Lake, Lake Co., Calif., May 1968.
6. *Fulmarus glacialis*. *A* and *B*: Monterey Bay, Calif., November 1, 1966. *C*: Point Reyes, Calif., December 1967. Fulmars breed in Alaska.
7. *Puffinus creatopus*. Breeds in Chile. Collected in May 1968, in the Gulf of California.
8. *Puffinus griseus*. Breeds in New Zealand and Chile. *A* and *B*: Monterey Bay, November 1, 1966. *C*: Gulf of California, May 1968. NM, Not measured, interfering peaks.
9. *Puffinus tenuirostris*. Breeds in Australia. Monterey Bay, December 1966.
10. *Oceanodroma homochroa*. Farallon Islands, Calif., May 1968.
11. *Pelecanus occidentalis*. Panama eggs were collected on Isla Pacheca and Isla Pachequilla, Gulf of Panama, February 1968. One egg contained 0·06 p.p.m. dieldrin and 0·05 p.p.m. endrin, another contained 0 16 p.p.m. dieldrin and 0·07 p.p.m. endrin (lipid weight). Baja California eggs were collected at Bahia de los Angeles, March 1968. One egg contained 0·24 p.p.m. dieldrin and 1·13 p.p.m. endrin (lipid weight).
12. *Fregata magnificens*. Isla Pacheca, Panama, February 1968.
13. *Sula leucogaster*. Isla Pacheca and Isla Pachequilla, Panama. February 1968. Two eggs were analysed for dieldrin and endrin. Dieldrin: 0·08, 0·18 p.p.m.; endrin: 0·06 and 0·611 p.p.m. (lipid weight).
14. *Phalacrocorax penicillatus*. Farallon Islands, May 1967.
15. *Phalacrocorax pelagicus*. San Mateo Co., Calif.
16. *Anas cyanoptera*. San Diego, April 1968. Adult male.
17. *Elanus leucurus*. *A*: Contra Costa Co., Calif., April 1968. *B* and *C*: Contra Costa Co., May 1968, two and three young raised, respectively. *D*: Abandoned, Contra Costa Co., May 1967. *E* and *F*: Destroyed nests, Contra Costa Co., March 1968.

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Table 3 (*continued*)

18. *Accipiter cooperii*. Balboa Park, San Diego, February 1968. First year female, died of trichomoniasis.
19. *Aquila chrysaetos*. 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 endrin.
20. *Pandion haliaetus*. Gulf of California, March 1968. One egg also contained 0·10 p.p.m. dieldrin and 0·25 p.p.m. endrin (L).
21. *Falco columbarius*. Immature, Utah, December 1967.
22. *Falco sparverius*. Adult was killed on road, Mendocino Co., Calif., December 1967. Eggs from Davis, California, 1968.
23. *Nycticorax nycticorax*. Eggs from different clutches, San Francisco Bay, May 1967.
24. *Larus occidentalis*. Standard errors, 95 per cent confidence limits.
25. *Sterna forsteri*. San Diego Bay, May 1967.
26. *Hydroprogne caspia*, 1967.
27. *Phalaropus fulicarius*. Monterey Bay, November 1, 1966.
28. *Uria aalge*. Farallon Islands, May 1967.
29. *Ptychoramphus aleutica*. Farallon Islands, April 1966.
30. *Synthliboramphus antiquum*. Monterey Bay, November 1, 1966.
31. *Cerorhinca monocerata*. Monterey Bay, November 1, 1966.
32. *Zenaidura macroura*. San Diego, July 1968.
33. *Tyto alba*. Clutch *A* from Contra Costa Co., Calif., March 1968. Clutch *B* from Yolo Co., Calif., April 1968.
34. *Sturnella neglecta*. 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 ten eggs from ten clutches of the western gull from the Farallon Islands showed an average PCB content of 136 ± 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 (Tables 2 and 3), but the number of analyse is 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[35], 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 atmosphere[36], 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 pressure[37]. Incineration 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 Sound[3] 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 ecosystems in North America. Peregrines 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 seals[33]. A second year male peregrine spent the winter of 1967–68 on Isla Pacheca, 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 eggs[34]. Several species of raptors do not accumulate high amounts of the organo-chlorine 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 40 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 work[18] has shown that both DDT and dieldrin induce hepatic enzymes in the pigeon which, in an *in vitro* preparation, increase 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 1262'). The experimental procedure previously described[18] 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 7:3:8:2. The strips were monitored with an autographic 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 a 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 HOMOGENATES FROM BIRDS TREATED WITH VARIOUS CHLORINATED HYDROCARBONS

	Amount of polar metabolites formed in mμmoles
Control	29·3 ± 6·5
DDE (40 mg/kg)	76·2 ± 13·1
DDT (40 mg/kg)	93·1 ± 11·2
PCB (20 mg/kg)	160·0 ± 10·5

Each figure is the average with standard deviation of a group of four birds. In all cases 500 mμmoles of oestradiol-6,7-T (500 mCi/mmole) was present in the incubation mixture. Incubation time 30 min, weight of microsomal fraction used 300 mg. All chlorinated hydrocarbons were injected intramuscularly into the pectoral muscle 7 days before death.

Body 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 sublethal 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.*[39]. He found that it took 70 days for the activity to fall to half its maximum value in the case of DDT and 15 days in the case of dieldrin. Experiments with dieldrin on pigeons

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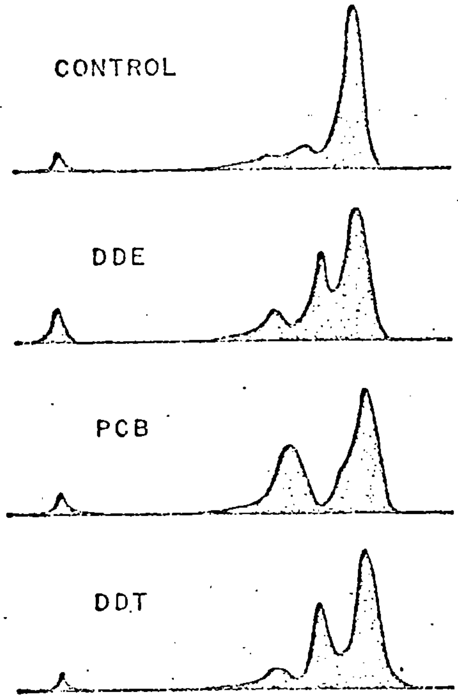

Fig. 1. Chromatographic separation of oestradiol and its metabolites. The large peak is unaltered oestradiol.

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 breakage[21–24], and water retention, which affects hatching success[40–41], 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. G. H. Thayer[42] has described it as "the embodiment of noble rapacity 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.

THE question whether the multiple ring-like mountain structures surrounding most of the lunar craters larger than 120 miles in diameter might be "frozen tsunamis", 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

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