

## Organochlorine Residues in Gyrfalcons (*Falco rusticolus*) in Iceland

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Received: 29 April 1994/Accepted: 12 February 1995

Little information is available on the existence of persistent organochlorine (OC) compounds in Icelandic wildlife. Despite very limited local sources of contamination the pattern of global distribution of OCs suggests that these stable chemicals are as ubiquitous in Iceland as in other non-industrial environments (Gregor and Gummer 1989; Ballschmiter and Zell 1980; Iwata et al. 1993).

The levels of dichlorodiphenyl trichloroethane (DDT) and polychlorinated biphenyl (PCB) residues in airborne fallout and animals in Iceland were investigated by Bengtson and Södergren (1974), who came to the conclusion that Iceland at that time was still not very affected by OC pollutants. Helleberg et al. in 1979 investigated the presence of mercury, PCB and dichlorodiphenyl dichloroethene (DDE) in 37 gyrfalcons (*Falco rusticolus*) found in Iceland from 1966 to 1973. They found high levels of these chemicals in emaciated birds that even may have contributed to their deaths but healthy birds contained somewhat lower levels than birds in Norway (Holt et al. 1979). Sproul et al. (1975) investigated the levels of OCs in breast muscle of several species of Icelandic seabirds caught in 1973. They found lower levels of PCB, DDE and dieldrin, than in seabirds from industrial regions, but similar to those found in seabirds from North-Scotland and north of Britain. Skaftason and Jóhannesson (1979; 1982) investigated the levels of OCs in Icelandic reindeer, sheep, butter, trout and salmon fry and found very low levels of hexachlorobenzene (HCB),  $\alpha$ -hexachlorocyclohexane ( $\alpha$ -HCH) and DDT derived substances. Another study by Luckas et al. (1990) compared the levels of chlorinated hydrocarbons in seals from different marine regions and found that PCB levels doubled going from Spitzbergen in the Arctic to Southeast-Iceland, while seals caught in the North Sea were thirty times more contaminated than in the Arctic.

In order to further investigate the distribution of OCs in Icelandic biota we have studied the levels of these substances in samples from a number of gyrfalcons, one of the local top predators, collected during a period of more than 10 years. We have also included the analysis of a few samples of ptarmigan (*Lagopus mutus*), the major prey species of the gyrfalcon (Nielsen 1986).

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## MATERIALS AND METHODS

The falcon sample consisted of 59 birds (56 *F. r. islandus* and 3 *F. r. candicans*) found freshly dead or dying all over Iceland, mainly during 1979-1992. The carcasses were stored frozen at -20°C and thawed and sampled just prior to analysis. The total sample consisted of 34 females and 23 males, but two birds could not be sexed. The age of the birds was determined on plumage characteristics until their second fall (about 15 months of age). The age of older birds were known either exactly through birds banded as chicks or to a minimum age. The sample was divided into four age groups: 29 birds were 0-6 months, 16 birds 7-12 months, 9 birds 13-18 months, and 5 birds were at least 15-28 months, i.e. could not be aged exactly (3) or were 19 months or older (2). The cause of death was evidently accidental in twenty cases of which 15 were in the first age group.  $\alpha$ -,  $\beta$ -,  $\gamma$ -HCH, HCB, DDT and its metabolites DDE and dichlorodiphenyl dichloroethane (DDD), and 10 isomers of PCBs (#28, 31, 52, 101, 105, 118, 138, 153, 156, 180) were analysed in breast muscle of all birds and also in the liver of 13 birds for comparison. OCs were also determined in the breast muscle and liver of five newly caught ptarmigan.

Extraction and cleanup of the samples was done basically according to the method of Jensen et al. (1983). Briefly, samples of tissue were weighed accurately, cut to pieces in a glass beaker and recovery standard (PCB #53) added. The samples were then homogenized in acetone/hexane (2.5:1) with a stainless steel Ultra-Turrax homogenizer (Janke & Kunkel, IKA - Germany). A mixture of hexane/diethyl ether (9:1) was added and the solvents filtered and pressed with nitrogen gas out of the homogenate. After repeating this step the combined extracts were washed with sodium chloride 0.9% in 0.1 M ortho-phosphoric acid. The extractable fat content was determined by evaporation to dryness at 62°C. Aliquots of fat were resuspended in 2 ml isooctane with internal standard (1,2,3,4-tetrachloronaphtalene) added, and cleaned in 10 ml of sulphuric acid (conc). The recovery of PCB #53 was in the range of 75-88%. All solvents were analytical grade from Merck, Germany.

The individual PCB congeners and pesticides were determined by gas chromatography against standard curves made from the corresponding individual standards from Promochem, Wesel, Germany. An HP5890 gas chromatograph with HP Ultra-1 or Ultra-2 (25 m, 0.20 mm i.d., 0.33  $\mu$ m film thickness) capillary columns, and an HP5970 mass selective detector (MSD) were used. The chemicals were detected by single ion monitoring (4 ions for each chemical or congener) and quantified from ion chromatograms of the single most abundant ion. Carrier gas was He (25 cm/s), splitless injection of 3 min, injector temp. 270°C, MSD-interphase 290°C. Temperature program: 85°C for 2 min, 30°C/min to 185°C, hold for 30 min, 2°C/min to 250°C, 7°C/min to 290°C, hold for 2 min. The laboratory has participated in 4 steps of the ICES/IOC/OSPARCOM intercomparison exercise on the analysis of PCBs and demonstrated that our method was of adequate quality. The limit of quantification was at least 1  $\mu$ g/kg for the pesticides and the individual PCB congeners. Total PCB ( $\Sigma$ PCB) was estimated by adding the levels of PCBs #138 and #153 and multiplying with 5, based on the fact that these isomers are approx. 20% of Aroclor 1260 (Luckas et al. 1990). The relative composition of the most abundant congeners in our samples, was also similar to A1260 (the ratio between isomers #153:180:138:118 was 1:0.6:0.5:0.3). However, it is possible that  $\Sigma$ PCBs are somewhat overestimated by this procedure.

Significant differences between medians in Tables 1 and 2, were found by the Kruskal-Wallis and Mann-Whitney tests, using Instat, version 2.01 for MacIntosh.

## RESULTS AND DISCUSSION

All samples contained high levels of PCBs that grew strikingly with age as shown in Fig. 1. Already at hatching the levels are about 0.1 mg/kg and have increased about hundredfold at 10 months, and nearly thousandfold at 20 months. DDT was not detected in any sample but DDE constituted 95-99% of the total DDT ( $\Sigma$ DDT), the rest being DDD. HCB was found in most samples whereas HCH was found only in 15 birds and then mostly as the  $\alpha$ -isomer (Table 1). The levels of  $\Sigma$ PCB,  $\Sigma$ DDT and HCB increase significantly ( $P < 0.05$ ) with each age group, as might be expected from the bioaccumulating properties of these chemicals. The PCB/DDT ratio is above 4 in all cases. It was found to be around 3 in eggs from the same species in Alaska (Walker 1977) and around 4 in the liver of gyrfalcons in Norway (Frøslie et al. 1986).

Because of the strong dependence on age, no effect of location, year or sex could be detected. However, a plot of  $\Sigma$ PCBs vs  $\Sigma$ DDT or HCB shown in Fig. 2, reveals a striking linear relationship between these chemicals. This indicates that the OCs originate from the same source, making the possibility of a local contamination less likely. Long range atmospheric and water transport has been considered to be the main source of OCs in remote areas (Ballschmiter and Zell 1980; Gregor and Gummer 1989; Bengtson and Sørdergren 1974). The gyrfalcon is sedentary in Iceland (Nielsen 1986) but the possibility of migratory prey species of the gyrfalcon becoming contaminated on their wintering grounds must nevertheless not be overlooked.

We compared the levels of OCs in the breast muscle and the livers of 13 birds (Table 2) and found that these levels were not significantly different ( $P > 0.05$ ). Holt et al. (1979) found similar levels of OCs in both tissues in various predators.

It has been suggested that redistribution of fat deposits during periods of starvation may rapidly increase the levels of OCs in vital organs of birds and thereby at least contribute to their death (Walker 1990; Wiemeyer and Cromartie 1981). To investigate this hypothesis we examined the effect of nutritional condition on the OC levels (Table 3). To control for age, only birds of age groups 7-18 months were used for this analysis. In the leaner birds with  $< 3\%$  fat in their breast muscles, the levels of  $\Sigma$ PCB and  $\Sigma$ DDT were 2-3 times higher than in birds with more fat. This effect is obviously more pronounced when the results are presented on a fat basis, then the difference is almost tenfold. Not much difference in the levels of HCB was observed. The median age of both groups was the same, 10 months, and the mean age  $11.0 \pm 3.7$  months for the leaner birds and  $10.7 \pm 3.0$  months for the birds in better condition. The mobilization of fat depots, where most of the whole body OCs are usually stored, may well have at least contributed to the death of the leaner birds, by critically raising the levels of OCs in vital organs (cf. Walker 1990). We will later report on the pathological examination of the sample birds.

It is difficult to compare our results to those found by others elsewhere. Other investigators may analyse different tissues, different age groups, or use different methods of PCB quantification, so any comparison is tentative at best, especially for PCBs. In Table 4 we attempt to compare a few values from the literature to our own. First we compare our results to the unpublished data of Helleberg et al.

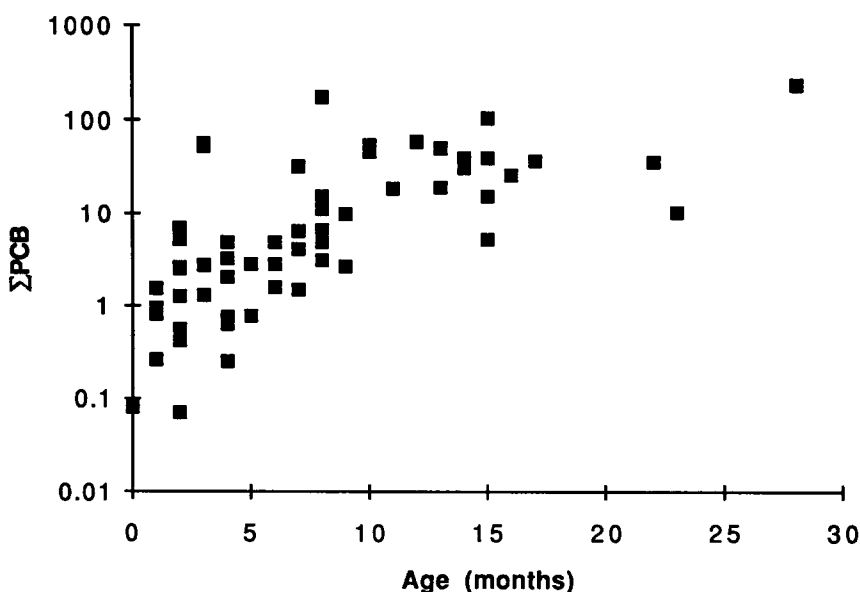


Figure. 1. Levels of  $\Sigma$ PCB (on a log-scale) vs. age in breast muscle of 59 gyrfalcons (mg/kg wet weight).

1979, who examined Icelandic gyrfalcons from the period 1966-1973. Taking the effect of age into account the levels of  $\Sigma$ PCBs and  $\Sigma$ DDT in Iceland appear to be very similar during the two periods despite a temporal difference of about 15 years. Although no conclusions can be drawn from only three birds, all *F. r. candicans* which originate in Greenland but found in Iceland show higher levels of OCs than the Icelandic falcons of the same age group. This indicates a higher level of OC contamination in Greenland than in Iceland. Further, if it is assumed that most of the Norwegian gyrfalcons analysed, fall into our first two age groups, contamination levels appear to be similar in Norway and Iceland. For Alaskan gyrfalcons we could only find results from fat biopsies and eggs. Compared to the data from a single bird in Norway the level of OC contamination in Alaska is low.

In an attempt to find the source of OCs in the Icelandic gyrfalcons we investigated samples from breast muscle and liver of five ptarmigan. Ptarmigan are sedentary like the falcons and their major prey (Nielsen 1986). No OCs could be detected in these birds. However, OCs have been detected in ptarmigan in Greenland (Braestrup et al. 1974), Alaska (Walker 1977), but not Sweden (Lindberg et al. 1985).

Because the gyrfalcon is protected by law in Iceland our sample was not scientifically chosen and the results probably do not represent the average levels of OCs found in this species at large. Being a non-migratory top predator, the levels and trends, however, should give an indication of the status of possible contamination by these chemicals in Icelandic wildlife. As evident from the age distribution the young roaming birds seem to be most vulnerable or at least most easily found. The oldest bird in our sample was at least 28 months, but gyrfalcons become at least 9 years old in the wild (Icelandic Bird Ringing

Table 1. Organochlorines in breast muscle of gyrfalcons.  
Values are medians with range shown in parenthesis (mg/kg wet weight).

age group (months)	n	ΣPCB	ΣDDT	HCB	ΣHCH
0-6	28	1.42* (0.07-56.2)	0.34* (0.005-16.6)	0.02* (n.d.-0.58)	n.d. (n.d.-0.32)
7-12	14	8.88* (1.50-59.0)	1.63* (0.23-13.9)	0.10* (0.007-1.13)	n.d. (n.d.-0.39)
13-18	9	26.3* (5.08-103)	4.13* (1.02-19.7)	0.12* (0.01-0.70)	n.d. (n.d.-0.03)
15+ - 28+	5	37.1* (10.2-232)	9.15* (2.15-49.9)	0.18* (0.02-1.40)	n.d. (n.d.-0.16)

\*values are significantly different ( $p < 0.05$ ) between age groups. n.d.: not detected

Table 2. Levels of organochlorines in breast muscle and liver of 13 gyrfalcons.

Values are medians in mg/kg fat (mg/kg wet weight)

	muscle	liver	muscle/liver
ΣPCB	31.2*	23.6*	1.3 (2.5)
ΣDDT	18.6*	8.83*	2.1 (3.1)
HCB	0.90*	0.90*	1.0 (1.3)

\*values are not significantly different ( $p > 0.05$ ) between tissues.

Table 3. Effect of %fat on organochlorine levels in breast muscle of gyrfalcons aged 7-18 months.  
Values are medians with range shown in parenthesis.

% fat	n	ΣPCB	ΣDDT	HCB
mg/kg wet weight				
<3%	9	26.3 (5.18-103)	3.37 (1.50-19.7)	0.08 (0.01-1.13)
>3%	14	8.22 (1.50-59.0)	1.60 (0.23-13.3)	0.13 (0.007-0.70)
mg/kg muscle fat				
<3%	9	1300 (376-7935)	201 (107-1515)	3.40 (0.96-69.4)
>3%	14	140 (6.22-1233)	26.2 (0.74-280)	2.14 (0.10-13.1)

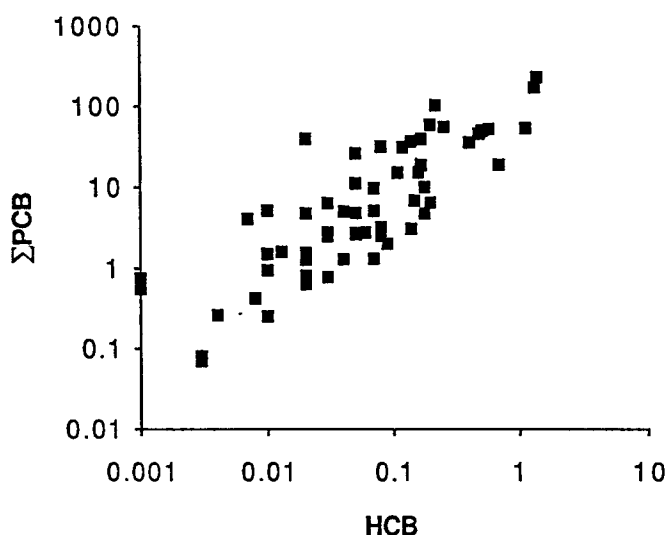
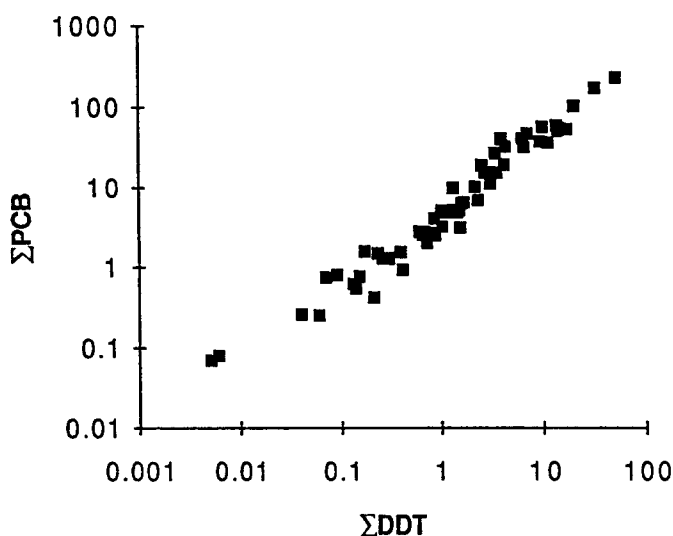


Figure 2. Relationship between the levels of  $\Sigma\text{PCB}$  and  $\Sigma\text{DDT}$  (above) and  $\Sigma\text{PCB}$  and HCB (below) in breast muscle of 59 gyrfalcons. Both figures are shown with log-scales, values are mg/kg wet weight.

Scheme, unpublished data). One can only speculate to what level the OCs have accumulated in the older birds, possibly lethal in some cases (Walker 1990). The next step will be to investigate the levels of OCs in several migratory and other prey species of the gyrfalcon, in an attempt to delineate to what extent they might contribute to the levels of OCs we have found in the Icelandic falcons.

Table 4. Organochlorines in gyrfalcons.  
Values are medians or (means in parenthesis) in mg/kg wet weight.

place-species year (ref.)	Tissue	n	Age (months)	ΣPCB	ΣDDT
Iceland- <i>rusticolus</i> <i>islandus</i> 1966-1973 (*)	muscle	29	<12	(17.0)	(3.5)
		6	>12		
	liver		"	(24.8)	(5.5)
			"	(111)	(22.5)
	adipose				
1979-1992 (#)	muscle	28	0-6	1.42 (5.72)	0.34 (1.42)
		14	7-12	8.88 (16.4)	1.63 (3.48)
		9	13-18	26.3 (32.9)	4.13 (6.61)
	liver	13	0-15	1.01 (2.00)	0.21 (0.39)
Iceland- <i>rusticolus</i> <i>candicans</i> 1983-1987 (#)	muscle	1	0-6	2.02	0.72
		2	7-12	46.4-173	7.0-31.2
Norway- <i>rusticolus</i>  1965-1983 (§)	muscle	2	not	0.1-11.0	0.1-3.9
	liver	12	stated	5.0	1.2
	egg	1		12.0	3.2
Alaska- <i>rusticolus</i>  1970-71 (§)	adipose	8	adult	23.9	12.3
	egg	14		1.6	0.8

\* Helleberg et al. 1979, # this paper, § Frøslie et al. 1986, § Walker 1977.

Acknowledgments. Funding provided by the Icelandic Council of Science and the Ministry for the Environment is gratefully acknowledged.

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