Levels and implications of polychlorinated hydrocarbons in male Australasian harriers

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Abstract

Breast tissues from four male Australasian harriers *(Circus approxmans)* were analysed for polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) pesticides at the AgriQuality Environmental ultra-trace analytical unit. The harriers were injured birds collected and euthanased over the period from 1998 to 1999 by Bird Rescue Wanganui-Manawatu.

The total toxic equivalents (I-TEQ) considering all the PCDD/F and PCB congeners, ranged from 3.7 to 47.7 pg/g wet weight. While these I-TEQ values are higher than previously reported for New Zealand avian species, the values are relatively low when compared to the concentrations in raptors that have been reported internationally.

The I-TEQ values in Australasian harriers were dominated by the PCB congeners, contributing approximately 65% of the I-TEQ. This finding is consistent with previous studies on New Zealand avian species and recent studies examining the congener compositions of raptor samples.

The dominant PCB congeners in the harrier samples were congeners #153, #138 and #180. The highest concentration for a PCB was 65 ng/g wet weight for congener #153. The only congener that was below detection limits in all samples was #81. The PCB congeners with the highest toxic equivalency factor (TEF) values - #126, #77 and #169 were present at concentrations in the approximate range of 0.1 to 0.01 ng/g wet weight.

The dominant PCDD/F congeners were OCDD and 1,2,3,6,7,8-HxCDD, while 1,2,3,4,6,7,8-HpCDD and OCDF were also found in high concentrations compared to the remaining congeners. The highest concentration for an individual congener was 24 pg 1,2,3,6,7,8-HxCDD /g wet weight. The only analysed congener that was below detection limits in all samples was 1,2,3,7,8,9-HxCDF.

Although three of the four sample residues in this study exceeded the adopted reference dose of 7 pg I-TEQ /g, it is unlikely that the current levels of PCDD/Fs and PCBs in Australasian harriers are causing population level effects. Nevertheless, the data suggest that there is little environmental assimilative capacity for I-TEQ.

1. Introduction

PCDDs, PCDFs and PCBs are a group of chemicals that have attracted considerable attention due to their high toxicity, persistence in the environment, and ability to bioaccumulate. The combination of these properties means that organisms such as raptors at the upper levels of the food chain can potentially accumulate concentrations sufficient to cause adverse effects. In the

relatively polluted North American Great Lakes system, these chemicals are suspected of causing reproductive impairment in bald eagles *(Haliaeetus leucocephalus)* (Bowerman et al. 1995) and other avian species (Yamashita et al. 1993).

PCDDs and PCDFs, of which there are respectively 75 and 135 possible congeners, are not produced commercially and have no known uses. They are principally formed as by-products during the production of other chemicals. The environmental release of PCDD/Fs predominantly occurs through the use of `contaminated' chemicals and also in emissions from incineration reactions. Chemicals that have been found to contain relatively high concentrations of PCDD/Fs include the pesticides 2,4,5-T and pentachlorophenol. The PCBs, of which there are 209 possible congeners, were used extensively in industrial applications as dielectrics (electrical transformer fluids), heat exchange fluids and for a variety of other uses. The use of PCBs in New Zealand was banned in 1995 (MfE 1998).

While PCDD/Fs and PCBs generally have low acute toxicity, they can produce a diverse suite of chronic adverse effects. These effects are all considered to be caused through a common mode of action. The mechanism involves binding to a particular receptor complex (*Ah* receptor) leading to a range of subcellular responses. Key amongst these responses are alterations in gene expressions leading to changes in the levels of compounds such as hormones and vitamins, and subsequent impacts including reproductive impairments. While there is a diverse range of adverse effects, there is a clear pattern to the effects that occur. The major difference between the various PCDD/F and PCB congeners is their potency at causing the characteristic suite of effects (Giesy et al. 1994). 2,3,7,8-TCDD is widely regarded as the most toxic of the PCDD, PCDF and PCB congeners. In comparison, the OCDD congener is only considered to have 1/1000th the potency of 2,3,7,8-TCDD.

To enable comparisons of the toxicity that these chemicals have in mixtures, toxic equivalency factors (TEFs) were developed. They relate the toxicity of each congener to 2,3,7,8-TCDD. By multiplying the concentration of each congener with its TEF, and summing for all congeners, a value (I-TEQ) can be derived which gives a measure of the `total' toxicity relative to 2,3,7,8-TCDD. The I-TEQ for a particular congener class, e.g. PCBs, can be calculated by summing the contribution of only those congeners. It is important to differentiate between the I-TEQ, and the sum of congeners which does not correct for relative toxicity.

The Australasian harrier feeds on carrion and live prey (small mammals and birds, large insects) and through this position at the top of the food chain would be expected to accumulate relatively high concentrations of organochlorine compounds. In a recent study, Reid & Jones (1999) examined the levels of organochlorine pesticides in Australasian harriers. While the majority of pesticides were present at low levels, the concentrations of p,p'-DDE and dieldrin were approaching levels where adverse effects may have been expected.

While `remote' regions of New Zealand could be expected to have relatively low inputs of PCDD/Fs and PCBs due to a lack of industrialisation, the pres-

ence of organochlorine compounds in these areas cannot be discounted. Atmospheric redistribution from more polluted areas will still result in some inputs. This mechanism of long-range transport and deposition is demonstrated by the contamination occurring in polar and other remote regions (SETAC 1998).

1.1 STUDY OBJECTIVES

This study was designed primarily to:

- quantify the concentrations of PCDD/Fs and PCBs in male Australasian harriers collected from the Wanganui region;
- enable comparison of the concentrations with levels reported in other New Zealand raptors and in overseas species; and
- determine whether the concentrations are sufficiently high that they would be expected to cause adverse effects on Australasian harrier populations.

2. Methods

2.1 SAMPLING

Injured harriers were presented by members of the public to Bird Rescue Wanganui-Manawatu, and severely injured birds were euthanased with carbon dioxide. The birds were then stored frozen at -20°C. For analysis, the frozen carcasses were thawed and breast muscle tissue dissected into solvent pre-cleaned glass jars.

2.2 ANALYSIS

PCDD/F and PCB quantification was conducted at the AgriQuality Environmental analytical laboratory. For analysis, each muscle sample was thoroughly homogenised and a portion extracted to provide the total concentration of contaminants in the original sample. Samples were analysed by standard isotope dilution procedures for a selected PCDD/F and PCB congeners. Before extraction a number of isotopically labelled internal standards were added to each sample. The extracts were then subjected to a range of chemical and chromatography clean-up procedures to remove interfering substances.

After clean-up, analytes were determined by high-resolution gas chromatography with high-resolution mass spectrometry for identification and quantification of compounds of interest. Full analytical details are available on request, as are details of the quality assurance procedures used in the laboratory. All

analyses were performed under the laboratory's ISO 9002/ISO Guide 25 - IANZ (formerly Telarc) accreditation.

3. Results and discussion

The details for each sample, including collection information, estimated harrier age, total body weight, breast muscle weight and lipid content are described in Table 1.

The summary of I-TEQ, PCDD/F I-TEQ and PCB I-TEQ for each sample is described in Table 2. In the four harrier muscle samples examined, the I-TEQ ranged from 3.7 to 47.7 pg/g wet weight. Congeners that were below detection limits made very little impact on the derivation of the I-TEQ. As an example, for the sample with the greatest number of non-detected congeners, including the non-detects (at a concentration half their limit of detection (LOD)) made less than 1.5% difference to the final value.

There have been relatively few studies of the I-TEQ concentrations in New Zealand wildlife with which to compare the data from this study. Recently, the mean I-TEQ for albatross eggs and chicks (10 samples) was reported as approximately 9 pg/g wet weight (Reid & Jones 1999), a value slightly over half the average level found in harrier breast muscle. However, the relatively higher lipid fraction in eggs compared with harrier breast muscle means that on a lipid basis, the albatross I-TEQ was less than a quarter that of the harrier levels. Jones (1998) also recently reported levels in eggs from other species of New Zealand sea birds as being approximately half those of the harrier on a wet weight basis.

When making comparisons between wildlife residue values, some caution needs to be used before concluding that a particular environment is relatively clean or polluted. The comparisons between wildlife values are complicated by factors that include: variations in analytical methods, the collection of samples over different seasons or years, sampling of different tissues, e.g. egg versus muscle, and variations in the presentation of data, e.g. /g wet weight or /g lipid. The wildlife studied may also vary in the ecological niches they fill, and thus the extent to which they are exposed to, and accumulate residues.

In contrast to the limited number of New Zealand studies, the I-TEQ in overseas avian species has received a great deal of attention. The comparison with these latter studies suggests that the current concentrations (3.7 - 47.7 pg/g) in Australasian harriers are relatively low. The mean concentrations of I-TEQ in Canadian bald eagle eggs ranged from approximately 100 to 300 pg/g wet weight depending on collection site (Elliott et al. 1996a). Note that, as described above, when comparing muscle to egg residues it needs to be considered that the egg concentrations could be approximately double the residues in muscle tissue (due to higher lipid contents in eggs). In bald eagle eggs from the relatively polluted Great Lakes region in North America, concentrations as high as 1650 pg/g have been recorded (cited by Bowerman et

al. 1995). I-TEQ levels ranging from 350 to 2800 pg/g wet weight were also reported in double-crested cormorant *(Phalacrocorax auritus)* and Caspian tern *(Hydroprogne caspia)* eggs from the Great Lakes region (Yamashita et al. 1993).

The I-TEQ values in Australasian harriers were dominated by the PCB congeners which contributed approximately 65% of the I-TEQ. This finding is consistent with previous studies on New Zealand avian species, with PCB congeners contributing approximately 85% of the I-TEQ in albatross eggs and chicks (Jones 1998; Reid &Jones 1999). Other studies have shown a similar pattern, with Yamashita et al. (1993) and Bowerman et al. (1995) suggesting that PCBs were the primary contribution to the I-TEQ in avian species from the Great Lakes. However, in Canadian bald eagle eggs, while PCBs were generally the dominant contributor, in some locations the PCDD/F contribution was similar to or greater than that of the PCBs (Elliott et al. 1996a).

The total PCB concentrations (not corrected for relative toxicity) in Australasian harriers ranged from 17.1 to 295 ng/g wet weight. These values appear relatively low in comparison with previous reports of total PCB concentrations in New Zealand birds of prey. Bennington et al. (1975) reported a concentration of approximately (assuming 5% muscle lipid) 4.4 Mg/g wet weight in a New Zealand falcon *(Falco novaeseelandiae)* egg. Solly and Shanks (1976) reported total PCB levels in 2 Australasian harrier muscle samples of 0.25 and 1.62 Mg/g wet weight. These authors also measured a total PCB concentration of 0.46 Mg/g muscle wet weight in a morepork *(Ninox novaeseelandiae)* collected from Wellington. The levels historically reported for total PCB concentrations in New Zealand seabirds are closer to those currently found in Australasian harriers. Bennington et al. (1975) measured levels generally in the order of 0.05 Mg/g wet weight, although concentrations approaching 1 Mg/g wet weight were recorded in some samples.

Total PCB concentrations ranging from 1 to 28 Mg/g wet weight were reported in muscle from American kestrels *(Falco sparverius)* collected in Florida between 1971 and 1981 (Sundlof et al. 1986). The values for muscle samples from red-shouldered hawk *(Buteo itneatus)* were lower, ranging from 0.13 to 7.9 Mg/g wet weight. The median levels of total PCBs in breast muscle of raptors collected from Illinois over 1966-1981 were 4.37 and 0.14 Mg/g wet weight respectively for the red-tailed hawk *(Buteo jamaicensis)* and the rough legged hawk *(Buteo lagopus)* (Havera & Duzan 1986). The median levels of total PCBs in Canadian bald eagle livers was reported as ranging from approximately 0.4 to 3.4 Mg/g wet weight. The concentrations of total PCBs in Canadian bald eagle eggs ranged from approximately 2 to 5 Mg/g wet weight (Elliott et al., 1996a). These comparisons suggest the total PCB levels in Australasian harriers are at the low end of the range of values that have been reported internationally for raptor species.

The dominant PCB congeners in the Australasian harrier samples were congeners #153, #138, and #180 (Table 3). The highest measured concentration for a PCB congener was 65 Mg /g wet weight for #153. The only reported congener that was below detection limits in all samples was #81. The PCB congeners with the highest TEF values - #126, #77 and #169 were present at concentrations in the approximate range of 0.1 to 0.01 Mg/g wet weight. The

dominance of the #153, #138 and #180 congeners reflects the patterns observed in albatross eggs and chicks (Reid & Jones 1999). It is also consistent with the general pattern described in Great Lakes sea birds, with hexachlorobiphenyls, and particularly #153, dominating the congener profile (Yamashita et al. 1993). Elliott et al. (1996a, 1996b) also reported that PCBs #153, #138, and #180 were the dominant congeners in Canadian bald eagle egg and liver samples.

The dominant PCDD/F congeners were OCDD and 1,2,3,6,7,8-HxCDD, while 1,2,3,4,6,7,8-HpCDD and OCDF were also found in high concentrations compared to the remaining congeners (Table 4).The highest concentration for an individual congener was 24 pg /g wet weight for 1,2,3,6,7,8-HXCDD. The only analysed congener that was below detection limits in all samples was 1,2,3,7,8,9-HxCDF. This profile for the PCDD/F congeners is similar to that reported for New Zealand northern royal albatross *(Diomedea sanfordi)* (Reid & Jones 1999). However, an interesting difference is that the 2,3,7,8-PCDD congener was found in harrier muscle at approximately double the concentration of the 2,3,7,8-PCDF congener. In contrast, in albatross eggs, the 2,3,7,8-PCDF congener was at levels several times that of 2,3,7,8-PCDD.

Similarly to the Australasian harrier, Canadian bald eagle eggs were reported to have 1,2,3,6,7,8-HxCDD as a dominant PCDD congener (Elliott et al. 1996a). However, 1,2,3,7,8-PnCDD and 2,3,7,8-TCDD appeared to be more significant contaminants than in the Australasian harriers. Consistent with the harrier data was the finding of Elliott et al. (1996a) that 1,2,3,4,6,7,8-HpCDD and OCDD were significant contaminants and that the furans were generally at lower levels.

4. Risk assessment of PCDD/F and PCBs

The considerable international attention paid to PCDD/Fs and PCBs means that reference doses for conducting risk assessments are readily available. However, one difficulty in deriving reference closes for PCDD/Fs and PCBs is that avian species show a wide range of sensitivities to I-TEQ (Hoffman et al., 1998). The literature includes `no observed adverse effect concentrations' (NOAECs) from 1 to 114 pg/g egg, with the chicken being considered a relatively sensitive species.

With regard to the reproductive effects associated with I-TEQ, Bowerman et al. (1995) and Giesy et al. (1995) considered a NOAEC of 7 pg I-TEQ/g egg as protective for bald eagles. This was based on egg lethality and while the authors considered that the value was relatively conservative, they also noted that it was "near the median for the NOAEC values collected from the literature on the toxicity of TCDD to avian species". The United States Environment Protection Agency (US EPA) proposed a higher value, a NOAEC of 80 pg I-TEQ/g wet weight for American kestrel eggs (US EPA 1997).

The NOAEC is the upper concentration of a chemical in eggs that is considered not to be associated with adverse effects for that species. It contrasts with the `lowest observed adverse affect level' (LOAEL) which is a higher value representing the lowest concentration found to be associated with adverse effects for that species. The relationship between the level of risk and LOAEL is often expressed as a hazard index through the formula:

Hazard Index = Residue concentration (mg/kg) / LOAEL (mg/kg).

A derived hazard index of greater than 1 generally results in some concerns regarding the potential adverse effects of the chemical. However, population-level effects are not expected at a hazard index of 1. Depending on the slope of the dose-response relationship, hazard index values of 10 to 20 may begin to be related to population level effects (Bowerman et al. 1995).

In comparing the most conservative raptor NOAEC of 7 pg I-TEQ/g egg with the residues in Australasian harriers (3.7 to 47.7 pg I-TEQ/g muscle wet weight), it needs to be considered that analysis was conducted on male breast tissue rather than eggs. The extrapolation from muscle breast residues to egg concentrations is not particularly difficult since chlorinated aromatic compounds generally accumulate in tissues in proportion to the percentage of lipid (SETAC 1998). The lipid content in harrier muscle ranged from 1.8 to 6.5%, comparing with lipid levels in albatross eggs of 9 to 4.6 % (Reid & Jones 1999), in a New Zealand falcon egg of 5.6% (Bennington et al. 1975), and in bald eagle eggs of 3 to 6.2 % (Elliott et al. 1996a). This suggests that a conservative value (assuming transfer to the eggs on a lipid basis) would be for harrier egg concentrations on a wet weight basis to be approximately double breast tissue residues. This increase is balanced by the consideration that female breast residues would be expected to be lower than males since egg laying is a process that can substantially reduce female body burdens. The two contrasting factors suggest that male breast residues are likely to represent a reasonable `worst case' scenario for egg concentrations.

While three of the four samples exceed the NOAEC of 7 pg I-TEQ/g and are therefore of some concern, it seems unlikely that the current residues in Australasian harriers would be causing population level effects. This conclusion is based on:

- the acknowledged conservatism in the 7 pg I-TEQ /g value;
- the 10- to 20-fold factor between exceeding the NOAEC and the potential for population level effects (Bowerman et al. 1995);
- the likely conservatism in directly extrapolating male muscle residues to egg concentrations; and
- only one of the samples being substantially higher than the 7 pg/g value, the other samples being approximately 4, 8 and 11 pg I-TEQ /g.

In addition to reference values for I-TEQ, values have also been established based on total PCB concentrations. Bowerman et al. (1995) and Giesy et al. (1995) used a NOAEC for total PCBs based on egg lethality of 4 mg/kg. Havera

and Duzan (1986) referenced brain residues of 310 mg/kg as being diagnostic of acute PCB poisoning. These values are comfortably above the mean muscle residues in Australasian harriers of approximately 0.1 mg/kg. However, it is worth noting that Bowerman et al. (1995) based their NOAEC on reproductive effects (egg survival) but acknowledged that PCBs could potentially subtly affect adults at lower concentrations.

While it therefore appears unlikely that the current PCDD/F and PCB residues are causing population level effects, the fact that three of the four samples exceed the NOAEC of 7 pg I-TEQ /g supports previous comments that there is little environmental assimilative capacity for I-TEQ (Jones 1998).

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Table 1. Sample identification, tag information, harrier and breast tissue weights, age, and sample lipid content for Australasian harriers analysed for PCDD/Fs and PCBs.

Report harrier #	Analysis #	Available information on tag	Total body, breast weight (g)	Estimated age (years)	Percent lipid content
Harrier 2	990672/5	Adm, head injuries, in on 20/10/98, killed 5/11/98 (0.1 mg lead)	585, 57.7	8 +	2.8
Harrier 3	990672/6	Jane Nugent Oleary Marton Adm, 18/3/99	575, 58.3	8+	6.5
Harrier 6	990672/3	Halcombe 9/6/99	490, 56.5	3-4	1.8
Harrier 7	990672/4	Budge St Fordell, 26/3/99	475, 57.4	3-4	2.0

Note: Report harrier numbers are not presented as Harrier 1-4 to be consistent with identification with an earlier report using the same harrier samples (Reid, 1999).

Table 2. Summary of the total concentrations of PCDD/Fs and PCBs, and I-TEQ in Australasian harrier breast muscle. Values include1/2 LOD data.

Report harrier #	Analyte concentrations					
	PCDD/F sum pg/g ww	PCB sum ng/g ww	PCDD/F I-TEQ pg/g ww	PCB I-TEQ pg/g ww	I-TEQ pg/g ww	I-TEQ pg/g lipid
Harrier 2	100	295	18.3	29.4	47.7	1703
Harrier 3	23.1	74.5	1.15	6.96	8.11	124
Harrier 6	34.3	76.5	3.07	8.29	11.36	631
Harrier 7	9.17	17.1	1.25	2.46	3.71	186
Mean values	41.6	115	5.94	11.8	17.7	661

Table 3. Concentrations of individual congeners, sum of congeners and I-TEQ for PCBs (pg/g wet weight unless otherwise stated) in Australasian harrier breast muscle.

Analyte	Harrier 2	Harrier 3	Harrier 6	Harrier 7
PCB#77	0.13	0.030	0.029	0.0068
PCB#126	0.19	0.033	0.053	0.016
PCB#169	0.080	0.024	0.070	0.037
PCB#28 + PCB31	0.83	< 0.1	0.18	< 0.1
PCB#52	0.24	0.12	0.044	< 0.04
PCB#49	0.11	0.091	0.024	< 0.02
PCB#44	0.033	< 0.02	< 0.03	< 0.02
PCB#74	0.68	0.23	0.21	0.076
PCB#70	0.31	0.11	0.048	< 0.02
PCB#81	< 0.01	< 0.02	< 0.021	< 0.01
PCB#101	1.5	0.97	0.36	0.085
PCB#99	3.2	2.2	0.94	0.27
PCB#110	0.18	0.27	0.10	< 0.04
PCB#123	0.38	0.31	< 0.2	< 0.2
PCB#118	11	9.6	3.7	0.74
PCB#114	0.24	0.19	0.078	0.016
PCB#105	3.1	1.8	0.97	0.23
PCB#153	65	18	18	4.4
PCB#138	51	14	12	2.8
PCB#167	11	3.4	2.4	0.54
PCB#156	8.2	2.7	1.6	0.35
PCB#157	1.2	0.40	0.24	0.060
PCB#187	28	3.6	5.3	1.2
PCB#183	8.3	1.4	2.5	0.45
PCB#180	56	8.1	15	3.2
PCB#170	25	4.5	6.3	1.3
PCB#189	1.3	0.19	0.32	0.029
PCB#202	0.69	0.087	0.16	0.045
PCB#196	7.0	0.98	2.2	0.38
PCB#194	7.3	0.91	2.1	0.33
PCB#208	0.27	0.064	0.049	0.017
PCB#206	1.5	0.31	0.45	0.11
PCB#209	1.6	0.23	0.28	0.12
PCB congener sum including1/2LOD (ng/g)	295	74.5	76.5	17.1
Total PCB I-TEQ: including 1/2 LOD (pg/g)	29.4	6.96	8.29	2.46

Table 4. Concentrations of individual congeners, sum of congeners and I-TEQ for PCDD/Fs (pg/g wet weight) in Australasian harrier breast muscle.

Analyte	Harrier 2	Harrier 3	Harrier 6	Harrier 7
2378 TCDF	3.0	0.20	0.48	0.14
2378 TODD	5.7	0.49	0.90	0.63
12378 PeCDF	1.7	< 0.1	0.35	0.074
23478 PeCDF	3.9	0.31	0.83	0.20
12378 PeCDD	12	0.46	1.9	0.52
123478 HpCDF	2.3	0.21	< 0.8	0.15
123678 HpCDF	3.5	0.17	< 0.7	0.17
234678 HpCDF	1.1	0.14	< 0.9	0.088
123789 HpCDF	< 0.2	< 0.2	< 0.3	< 0.1
123478 HxCDD	6.5	< 0.3	0.80	0.26
123678 HxCDD	24	1.1	3.7	1.2
123789 HxCDD	2.8	0.21	0.85	0.31
1234678 HpCDF	3.0	0.86	< 2	0.28
1234789 HpCDF	0.89	< 0.3	< 0.6	< 0.2
1234678 HpCDD	13	2.2	< 3.8	1.5
OCDF	12	1.3	< 2	< 1
OCDD	4.7	15	17	3.0
PCDD/F congener sum including 1/2 LOD	100	23.1	34.3	9.17
Total PCDD/F I-TEQ: including 1/2 LOD	18.3	1.15	3.07	1.25