



Persistent Organic Pollutants (POPs) and inorganic elements in predatory bird livers and eggs 2007 to 2009: a Predatory Bird Monitoring Scheme (PBMS) Report

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This report should be cited as:

Walker, L.A., Beith, S.J., Lawlor, A.J., Moeckel, C., Peréira, M.G., Potter, E.D. & Shore, R.F.¹ (2011). Persistent Organic Pollutants (POPs) and inorganic elements in predatory bird livers and eggs 2007 to 2009: a Predatory Bird Monitoring Scheme (PBMS) Report. Centre for Ecology & Hydrology, Lancaster, 30pp.

Suggested keywords: Annual report; Birds of prey; Environmental contamination; Monitoring; Pesticides; Pollution; Predatory birds; United Kingdom (UK)

Centre for Ecology and Hydrology Project Number: NEC04288

E-copies of this report: This report can be requested through the Natural Environment Research Council's Open Research Archive <http://nora.nerc.ac.uk/> or can be downloaded directly from the PBMS website <http://pbms.ceh.ac.uk/>

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Contents

1.	Executive Summary.....	4
2.	The Predatory Bird Monitoring Scheme.....	5
2.1.	Background.....	5
2.2.	Collection of carcasses and eggs for monitoring.....	6
2.3.	Post mortems of carcasses and processing of eggs.....	7
2.4.	The PBMS tissue and egg archive.....	8
2.5.	Description of samples that undergo chemical analysis.....	8
2.6.	Summary of analysis methods.....	9
2.7.	Data expression, format and analysis.....	9
2.8.	Outputs from the PBMS.....	10
3.	Persistent organic pollutants and metals in the livers of sparrowhawks.....	11
3.1.	Contaminant concentrations in birds that died in 2007 to 2009.....	11
3.2.	Long-term time trends.....	13
4.	Persistent organic pollutants and metals in merlin (<i>Falco columbarius</i>) eggs.....	15
4.1.	Contaminant concentrations in eggs collected in 2007 & 2008.....	15
4.2.	Long-term trends in contaminant concentrations in merlin eggs.....	17
5.	Persistent Organic Pollutants and Metals in golden eagle (<i>Aquila chrysaetos</i>) eggs.....	18
5.1.	Introduction.....	18
5.2.	Contaminant concentrations in eggs collected in 2007 & 2008.....	18
5.3.	Long-term trends in contaminant concentrations in golden eagle eggs.....	20
6.	Persistent Organic Pollutants and Metals in northern gannet (<i>Morus bassanus</i>) eggs.....	22
6.1.	Introduction.....	22
6.2.	Contaminant concentrations in eggs collected in 2007 and 2009.....	22
6.3.	Long-term trends in contaminant concentrations in gannet eggs collected from Ailsa Craig.....	24
7.	Persistent Organic Pollutants and Metals in sea eagle (<i>Haliaeetus albicilla</i>) eggs.....	25
7.1.	Introduction.....	25
7.2.	Results.....	25
8.	References.....	27

1. Executive Summary

The Predatory Bird Monitoring Scheme (PBMS; <http://pbms.ceh.ac.uk/>) is the umbrella project that encompasses the Centre for Ecology & Hydrology's National Capability contaminant monitoring and surveillance work on avian predators. By monitoring sentinel vertebrate species, the PBMS aims to detect and quantify current and emerging chemical threats to the environment and in particular to vertebrate wildlife.

Sparrowhawk livers were analysed for a range of persistent organic pollutants (POPs) and heavy metals. Sparrowhawks are studied because they have a wide distribution across the Britain and can be used as a sentinel species for the terrestrial environment. Mean PCB and mercury liver concentrations were below those thought to have an adverse effect on individual birds.

Pollutants, such as mercury and PCBs, can affect development and hatchability. Therefore, the PBMS also monitors the levels of contaminants in the eggs of a range of species including those of conservation concern, such as golden eagle and the re-introduced white-tailed sea eagle. Other species that are monitored are the northern gannet, which is used as a monitor of the marine environment, and merlin that hunts in upland habitats. The residues measured in the eggs of golden eagle and gannets collected between 2007 and 2009 were below those thought to have an adverse effect, but some residues in individual merlin eggs were above concentrations suggested to be indicative of no effect concentrations for birds generally. Few white-tailed sea eagle eggs are received for analysis by the PBMS but many of the eggs that have been analysed, including one of the eggs analysed for this report, have DDE, PCB and/or mercury concentrations above levels associated with adverse effects on bird embryos and hatching success.

In terms of long-term trends, there has been a decline in congener sum PCB contamination in the eggs of most of the species that have been monitored, except for coastal nesting golden eagles. In contrast however, there has been no significant decline over time in PCB concentrations in sparrowhawk livers and concentrations of 'Paris 10' congener sum and PCB-TEQ concentrations have also largely remained unchanged in both livers and eggs since monitoring began in 1996. Evidence for changes over time in mercury concentrations in predatory birds or their eggs is inconsistent across the species monitored. Where a decline has been detected, it has occurred before approximately 1990 and remained largely unchanged since then.

2. The Predatory Bird Monitoring Scheme

2.1. Background

The Predatory Bird Monitoring Scheme (PBMS; <http://pbms.ceh.ac.uk/>) is the umbrella project that encompasses the Centre for Ecology & Hydrology's long-term contaminant monitoring and surveillance work on avian predators. The PBMS is a component of CEH's National Capability activities.



By monitoring sentinel vertebrate species, the PBMS aims to detect and quantify current and emerging chemical threats to the environment and in particular to vertebrate wildlife. Our monitoring provides the scientific evidence needed to determine how chemical risk varies over time and space. This may occur due to market-led or regulatory changes in chemical use and may also be associated with larger-scale phenomena, such as global environmental change. Our monitoring also allows us to assess whether detected contaminants are likely to be associated with adverse effects on individuals and their populations.

Overall, the PBMS provides a scientific evidence base to inform regulatory decisions about sustainable use of chemicals (for example, the [EU Directive on the Sustainable Use of Pesticides](#)). In addition, the outcomes from the monitoring work are used to assess whether mitigation of exposure is needed and what measures might be effective. Monitoring also provides information by which the success of mitigation measures can be evaluated.

Currently, the PBMS has two key objectives:

- (i) to detect temporal and spatial variation in exposure, assimilation and risk for selected pesticides and pollutants of current concern in sentinel UK predatory bird species and in species of high conservation value
- (ii) in conjunction with allied studies, to elucidate the fundamental processes and factors that govern food-chain transfer and assimilation of contaminants by top predators.

Further details about the PBMS, copies of previous reports, and copies of (or links to) published scientific papers based on the work of the PBMS can be found on the [PBMS website](#).

2.2. Collection of carcasses and eggs for monitoring

The PBMS monitors contaminants principally in liver tissues and usually in addled or deserted eggs. The choice of sample depends on the likely site of toxicological significance for any particular compound and/or the species of bird monitored. For example, SGAR concentrations are measured in the liver because it is the target organ for these compounds where they interfere with the production of clotting factors) and contains high-affinity binding sites to which SGARs bind. The latest PBMS report relating to SGARs can be downloaded from the projects web site (<http://pbms.ceh.ac.uk/>). Conversely some contaminants are embryotoxins and monitoring residues of these compounds in eggs is more appropriate. Furthermore, the populations of some species, such as golden eagle (*Aquila chrysaetos*) and merlin (*Falco columbarius*), are at low densities in Britain and it is not possible to collect sufficient carcasses each year to obtain a statistically robust sample for monitoring liver concentrations. However, it is possible to collect reasonably large numbers of failed eggs for these species. Thus, monitoring contaminant levels in eggs allows us to study the impacts of chemicals on these rarer species.

The PBMS analyses the carcasses of birds that have died from natural causes. The collection of carcasses relies on members of the public who, in response to advertisements, send in dead predatory birds that they find. Taxidermists and wildlife rehabilitation centres also contribute significant numbers of specimens. However, only birds that have been free-living prior to death, or at most have been in captivity for 7 days, are used by the PBMS. Volunteers submit the carcasses through the post using approved, biohazard-safe packaging. Typically, most submissions are in autumn (September and October), and early spring (February to April inclusive) and, together, these five months account for approximately 60% of the birds submitted per year. Most of the birds submitted at this time are juveniles (birds that have hatched in the current or previous calendar year). Overall, approximately twice as many juvenile birds as adults (where adults are defined as individuals that have hatched before the previous calendar year) are submitted to the PBMS. On receipt, the details of each predatory bird is given a unique identifying code and the carcass is stored at -20°C until a macroscopic post-mortem examination is carried out.

Only individuals that are licensed by the relevant regulatory statutory body can collect bird eggs in the UK. Licensed collectors visit nests as part of other studies, predominantly ringing exercises, and send the PBMS eggs known to be addled or deserted.

The numbers of samples submitted to the PBMS vary year on year. This is because the samples are collected through volunteers and not through a standardized sampling protocol. Furthermore, it can be difficult to obtain samples, especially from remote areas. Usually, some 250 to 400 bird carcasses and 50-100 eggs are submitted to the PBMS each year. In 2007 to 2009, the PBMS received on average 324 birds and 119 eggs per year (Tables 2.1).

Table 2.1. Summary of birds and eggs submitted by volunteers to the PBMS between 2007 and 2009.

			Total number of	
			Carcasses ¹	Eggs ²
Owls	Barn Owl	<i>Tyto alba</i>	336*	17
	Eagle Owl	<i>Bubo bubo</i>	1	0
	Tawny Owl	<i>Strix aluco</i>	105	1
	Little Owl	<i>Athene noctua</i>	19	0
	Long-eared Owl	<i>Asio otus</i>	11	0
	Short-eared Owl	<i>Asio flammeus</i>	5	0
Falcons	Kestrel	<i>Falco tinnunculus</i>	51*	6
	Merlin	<i>Falco columbarius</i>	5	9*
	Peregrine falcon	<i>Falco peregrinus</i>	10	9
	Hobby	<i>Falco subbuteo</i>	9	1
Eagles	Golden Eagle	<i>Aquila chrysaetos</i>	0	9*
	White-tailed Sea Eagle	<i>Haliaeetus albicilla</i>	0	2*
Hawks & Vultures	Sparrowhawk	<i>Accipiter nisus</i>	121*	8
	Goshawk	<i>Accipiter gentilis</i>	7	6
	Buzzard	<i>Buteo buteo</i>	66	3
	Hen Harrier	<i>Circus cyaneus</i>	1	12
	Montague's Harrier	<i>Circus pygargus</i>	0	7
	Marsh Harrier	<i>Circus aeruginosus</i>	2	1
	Red Kite	<i>Milvus milvus</i>	52	9
Others	Heron	<i>Ardea cinerea</i>	8	0
	Kingfisher	<i>Alcedo atthis</i>	5	0
	Bittern	<i>Botaurus stellaris</i>	0	0
	Little Egret	<i>Egretta garzetta</i>	0	0
	Water Rail	<i>Rallus aquaticus</i>	0	0
	Gannet	<i>Morus bassanus</i>	-	8*
	Chough	<i>P. pyrrhocorax</i>	-	2
Mammals	Otter	<i>Lutra lutra</i>	108	-
Total			959	119

¹It is not always possible to take a liver sample from a bird and so the number of samples analysed may differ from the values in this table.

² Egg numbers may include more than one egg from a single clutch.

* indicates core species for which samples are taken for chemical analysis each year.

2.3. Post mortems of carcasses and processing of eggs

The post-mortem examination is conducted to determine age, using the European Union for Bird Ringing (EURING) classification (Speek et al., 2001), sex, body weight and putative cause of death. The most common causes of death (>70% of submissions) are road traffic accidents, other

types of collision, and starvation. Additional observations made on the carcasses include an assessment of the amount of fat reserves in the body, moult score (primaries, secondaries, tail and body feathers (Redfern and Clark, 2001)) and the presence of hemorrhaging in the body organs. Where possible, whole organs are excised, weighed, and various tissues (liver, brain, kidneys, pectoral muscle, subcutaneous fat) are archived in glass jars at and retained in the PBMS tissue and egg archive at -20°C.

Eggs that are submitted to the PBMS are measured (length, breadth, intact weight and weight of contents) and information on date of collection and provenance is also recorded. Each egg is cracked using a scalpel, the contents removed, an approximate degree of development assessed, and the contents are homogenised prior to archiving at -20°C in glass jars in the PBMS tissue and egg archive. The egg shell is rinsed, left to dry for 4 weeks at ambient temperature, and then weighed. An egg shell thickness index (Ratcliffe, 1970) is calculated as shell weight (mg)/length (mm) * breadth (mm).

2.4. The PBMS tissue and egg archive

The PBMS frozen (-20°C) archive contains samples dating back to 1968. It consists of approximately 20,000 tissue samples from 7,000 individual birds and 9000 egg contents; the majority (85%) of tissue samples are from 1980 onwards. This archive constitutes a valuable resource for a range of environmental toxicology studies and other conservation-orientated research work. The archive facilitates assessment of the exposure to and effects of chemicals in predatory birds that is wider in scope than the core PBMS activities (Jagannath et al., 2008, Pain et al., 2007) and can be used for piloting and refining new monitoring. It also provides a resource for projects that aim to assess the impacts of a range of non-chemical threats. In the past, these have included illegal trading and hunting (Natural Research, 2007) and quantifying the fine scale genetic structure of golden eagle populations (Bourke and Dawson, 2006).

2.5. Description of samples that undergo chemical analysis

At present, the core monitoring programme involves analysing contaminant concentrations in a sub-set of the livers and eggs that are archived each year. The numbers of samples available for analysis vary year on year. This is because samples are provided by amateur naturalists and other interested parties and not through a standardized sampling protocol. Furthermore, it can be difficult to obtain samples, especially from remote areas

The livers of a stratified sample of barn owls, and the livers of all kestrels (*Falco tinnunculus*) and red kites (*Milvus milvus*) received each year are analysed for four SGARs, difenacoum, bromadiolone, brodifacoum and flocoumafen. The monitoring data for these species are presented in a separate report (Walker et al., 2010).

The livers from a stratified sample of sparrowhawks received each year submitted to the PBMS are monitored for PCBs (35 individual congeners, sum PCB Toxic Equivalencies—PCB-TEQs, sum 'Paris 10' PCBs) and for a suite of non-essential and essential trace metals (mercury (Hg), arsenic (As), lead (Pb), cadmium (Cd), copper (Cu) and zinc (Zn)). The results of this monitoring are reported in [Section 3](#).

The eggs contents of one egg from each clutch of merlin, golden eagle and white-tailed sea eagle (*Haliaeetus albicilla*) eggs that have been received each year are analysed for the same compounds and the results of the monitoring are reported in sections [4](#), [5](#), and [7](#), respectively of this report. Approximately 10 gannet (*Morus bassanus*) eggs are collected biennially from colonies on Bass Rock (North Sea) and Ailsa Craig (Irish Sea). These eggs are also analysed for PCBs and metals

and the results are reported in section [6](#).

2.6. Summary of analysis methods

The analytical methods used to quantify PCBs and Hg are described in the [Addendum](#) to this report which can be downloaded from the PBMS website. Limits of detection for the different PCB congeners range typically between 0.407 and 1.026 ng/g wet weight in the sample and limit of detection for Hg was 0.2 µg/g dry wt. In addition to PCBs, contaminants originating from historic organochlorine insecticide use were also quantified as they are determined using the same analysis as that used for PCBs. The compounds quantified include DDE (from the insecticide dichlorodiphenyltrichloroethane (DDT)), HEOD (from the insecticides aldrin and dieldrin) and g-HCH (gamma-hexachlorocyclohexane). These compounds are no longer a focus for PBMS monitoring and the data are reported here only in the [Addendum](#) to this report.

Metal concentrations were measured in approximately 1 g sub-samples that were dried to constant weight at 80°C for 24 h, solubilised at room temperature overnight in 2 ml of (Analar) nitric acid, then heated at 90°C for 20 min followed by 120°C for 1 h. To further digest the organic matter, 0.5 ml of 30% hydrogen peroxide were added to the sample which was then heated at 120°C for 15 min. Samples were diluted with double-deionised water to known volume and a 10% acid strength. The resulting digests were analysed for a suite of trace metals and mercury by Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) using a Perkin Elmer DRCII ICP-MS with standard conditions.

2.7. Data expression, format and analysis

Throughout this report, liver and/or egg anticoagulant rodenticide, PCB and OC concentrations are reported as µg/g wet weight (wet wt). Sum PCB-TEQ concentrations (see below for explanation) are expressed as pg/g wet wt. All metal concentrations in livers and eggs are expressed on a µg/g dry weight (dry wt) basis. Residue data for individual birds are given in the tables within the [Addendum](#) to this report.

PCB concentrations in biological materials can be expressed in different ways. In the main body of the report, PCB concentrations are summarised as sum congener PCBs, Paris 10 congeners and sum PCB-TEQs.

The *sum congener PCB concentration* is the sum of the concentrations of each of the 35 PCB congeners that were individually measured. Data for individual congeners are given in the relevant [Addendum](#) to this report.

The “*Paris 10 congeners*” are a selection of individual PCB congeners defined in the 1974 Paris Commission and are congeners 28, 31, 52, 101, 105, 118, 138, 153, 156, 180. The concentration of these congeners are summed to give the Paris 10 congener concentration.

The *sum PCB-TEQ concentrations* have been calculated using the appropriate Toxic Equivalence Factors (TEFs) as given in the 1997 World Health Organisation values that apply to birds (Van den Berg et al., 1998). TEFs have been assigned to the “dioxin-like” PCB congeners. Each individual PCB-TEQ is calculated by multiplying the wet weight concentration of the congener by the TEF for that congener. The PCB congeners for which TEQs have been calculated are congeners 77, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189. The sum PCB-TEQ concentration is the sum of the TEQs for these 11 congeners.

Previously, total PCB concentrations have been reported but these are likely to over-estimate the true level of PCB contamination because the measures are derived from a large number of peaks, some of which may not actually represent PCBs. Congener specific parameters such as congener sum and Paris 10 sum concentrations are more precise measures but are likely to underestimate the true total PCB contamination, because not all 209 PCB congeners are quantified. Furthermore, it is still unclear what the toxicological significance of these parameters are. PCB-TEQ concentrations give a relatively more accurate assessment of the potential effects of coplanar PCB residues.

When all summed PCB and PCB-TEQ concentrations were calculated, individual congener concentrations that were below the limit of detection (non-detected) were assigned a zero value.

Contaminant concentrations for birds that died in 2007 to 2009 are summarized as geometric means and geometric standard errors based on data for all birds, including non-detected residues. The number of birds or eggs analysed and reported in the [Addendum](#) tables may be greater than the number for which summary data are presented in the main sections of the report. This is because some carcasses or eggs may have been collected in earlier years but only submitted to the PBMS in the last three years.

Long-term temporal trends in contaminant concentrations were assessed using linear regression analysis of annual median values weighted for the number of birds analysed in each year (Minitab 15.1; Minitab Inc.). Median, rather than geometric mean, concentrations are used in time trend analyses as median values are unaffected by any small changes between years in limits of detection.

2.8. Outputs from the PBMS

The results of the monitoring work are published in annual reports and scientific papers (for example Jagannath et al., 2008, Knopper et al., 2007, Leslie et al., 2011, Pereira et al., 2009, Pereira et al., 2008, Shore et al., 2006a, Walker et al., 2007a, Walker et al., 2007b, Walker et al., 2008a, Walker et al., 2008b, Wienburg and Shore, 2004) and on a dedicated [PBMS website](#).

Volunteer collectors are sent a post mortem report (carcasses) or biometric data (egg) for each sample they submit and a summary of the contaminant levels for those samples that are analysed chemically. This level of feedback to collectors is essential in maintaining a strong relationship between the PBMS and the volunteer base that supports it.

3. Persistent organic pollutants and metals in the livers of sparrowhawks

Sparrowhawks (a sentinel species of the terrestrial environment) have been monitored for PCBs and Hg since the mid 1960s but since 2006 a wider set of metals have been included in the monitoring. For birds collected in 2007 to 2009, information on provenance, age, sex and condition is given in Table 2 in the [Addendum](#). Details of analytical methods and data format are given in sections [2.6](#) and [2.7](#) respectively.



3.1. Contaminant concentrations in birds that died in 2007 to 2009

The results for each individual liver are given in the [Addendum](#) to this report which is available on the PBMS website. The geometric mean sum congener PCB, sum PCB-TEQ concentrations and the concentrations of the Paris 10 congeners are presented in Table 3.1.

Table 3.1. Geometric mean (GM) and 95% confidence interval (95%CI) concentrations of polychlorinated biphenyls (PCBs) in the livers of sparrowhawks received in 2007 to 2009.

Analyte	Year					
	2007		2008		2009	
	GM	95%CI	GM	95%CI	GM	95%CI
Sum PCB-TEQ (pg/g ww)	5.291	1.984-14.11	0.749	0.266-2.107	0.767	0.312-1.882
Sum congener (µg/g ww)	0.762	0.364-1.594	0.429	0.231-0.794	0.384	0.227-0.651
Individual congeners (ng/g ww)						
PCB 28	0.280	0.084-0.930	0.164	0.058-0.466	0.286	0.093-0.882
PCB 31	0.070	0.032-0.152	0.037	0.024-0.058	ND	-
PCB 52	0.230	0.055-0.953	0.485	0.175-1.348	0.182	0.060-0.554
PCB 101	2.909	0.822-10.30	1.672	0.588-4.752	2.355	0.878-6.316
PCB 105	2.710	0.802-9.161	0.950	0.261-3.464	0.315	0.063-1.571
PCB 118	24.66	12.14-50.10	13.72	6.548-28.74	13.83	6.380-29.96
PCB 138	34.08	15.30-75.93	34.57	20.09-59.51	28.96	19.12-43.88
PCB 153	151.5	72.01-318.7	95.91	54.44-169.0	81.19	49.15-134.1
PCB 156	7.804	2.549-23.89	2.829	0.832-9.619	1.665	0.374-7.416
PCB 180	125.0	57.74-270.5	69.94	38.96-0.125	66.32	38.95-112.9

ND indicates not detected

The geometric mean total and sum congener PCB concentrations in sparrowhawk livers were relatively low and indicated that, although exposure to PCBs is wide-spread the magnitude, is generally low and below that thought to cause adverse effects in individual birds.

Liver TEQ concentrations varied markedly between individual sparrowhawks with concentrations ranging from non-detected to 136.7 pg/g wet wt. The geometric mean concentrations for birds that died in 2008 and 2009 were below 1 pg/g wet wt. while the geometric mean concentration for birds that died in 2007 was 5.29 pg/g wet wt. These values are within the ranges reported in the livers of various predatory bird species from Europe, the USA and Japan (Kannan et al., 2003, Coady et al., 2001, Senthilkumar et al., 2002). The toxicological significance of TEQ concentrations is less well established for livers than for eggs (Hoffman et al., 1996), but 25 ng/g on a lipid weight (lipid wt) basis has been reported as the lowest observed effect concentration (LOEC) for induction of cytochrome P450 enzymes and for a 50% reduction in plasma thyroxine levels in common tern (*Sterna hirundo*) chicks (Bosveld et al., 2000). The geometric mean TEQ concentration for sparrowhawks that died in 2007 to 2009 combined was 46.9 pg/g, when expressed on a lipid weight basis, approximately three orders of magnitude lower than the LOEC reported for tern chicks. The highest lipid weight TEQ concentration measured in the sparrowhawk livers (bird 15461, 186.7pg/g lipid wt.) was approximately two orders of magnitude below the LOEC for terns.

In general the hepatic metal concentrations in sparrowhawks were below those associated with adverse effects (Table 3.2). The proposed indicative liver mercury concentrations associated with mortality and effects on reproduction in birds is 20 µg/g wet wt. and 2 µg/g wet wt., respectively (Shore et al., 2011). These concentrations are equivalent to 70 µg/g and 7 µg/g dry weight in sparrowhawk eggs and are suggested values that are likely to be protective for most species; they are not specific for sparrowhawks. All sparrowhawks that died between 2007 and 2009 had liver concentrations below those associated with mortality, but seven birds had liver mercury concentrations greater than associated with effects on reproduction. Five of these birds were males, for which it is unclear whether these indicative concentrations are applicable. The other two were female, one adult and two juvenile, and so this suggests that some individual sparrowhawks are accumulating mercury liver concentrations that may cause impaired reproduction.

Table 3.2. Concentrations ($\mu\text{g/g}$ dry wt.) of selected inorganic elements in the livers of sparrowhawks that died between 2007 and 2009.

	Critical Concentration ^a	GM	95% CI	No. > Critical Concentration
2007 (N=24)				
Hg	70/7 ^c	1.996	1.323-3.012	0/3
As	40	0.049	0.037-0.066	0
Pb	12	0.166	0.086-0.319	0
Cd	133	0.263	0.168-0.410	0
Cu	-	19.99	16.73-23.89	-
Zn	-	129	101.1-164.6	-
2008 (N=30)				
Hg	70/7 ^c	1.708	1.224-2.383	0/1
As	40	0.024	0.017-0.034	0
Pb	121	0.195	0.127-0.301	0
Cd	133	0.308	0.212-0.448	0
Cu	-	18.24	14.85-22.40	-
Zn	-	118.8	93.41-151.1	-
2009 (N=25)				
Hg	70/7 ^c	2.080	1.288-3.358	0/3
As	40	0.076	0.054-0.106	0
Pb ^b	12			
Cd	133	0.255	0.145-0.448	0
Cu	-	17.13	14.04-20.91	-
Zn	-	90.26	73.14-111.4	-

^a values (converted to a dry weight basis assuming an average moisture content of 70%.) suggested to be associated with adverse effects for Hg (Shore et al., 2011), As(Erry et al., 1999), Pb(Franson, 1996) and Cd (Furness, 1996)

^b Pb was not quantified in the 2009 samples.

^c Liver Hg concentrations greater than 70 $\mu\text{g/g}$ dry wt. may indicate possible mortality due to Hg poisoning, while values greater than 7 $\mu\text{g/g}$ dry wt. may indicate possible impaired reproduction

3.2. Long-term time trends

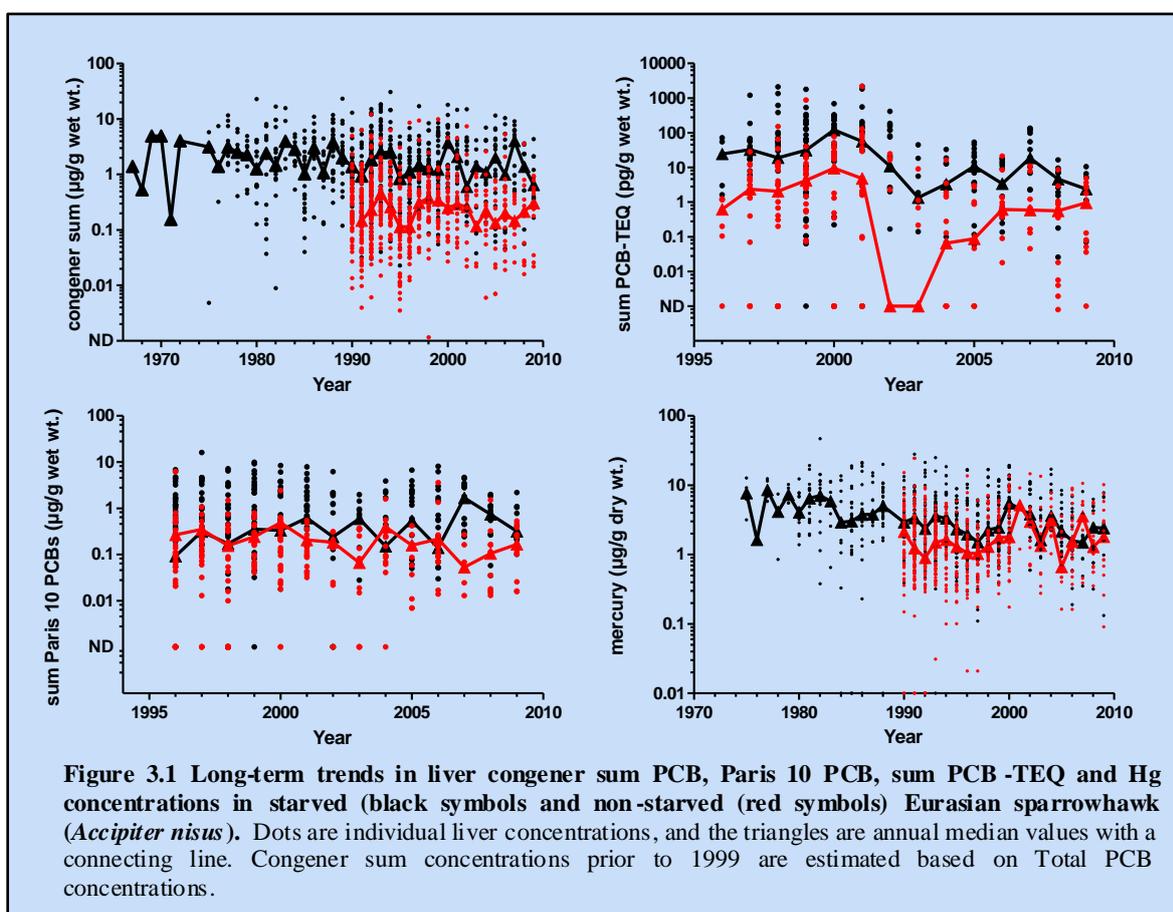
Previous studies (Shore et al., 2006, Wienburg & Shore, 2004) have demonstrated that contaminant concentrations, and in particular lipophilic contaminants such as PCBs, in birds on average are higher in starved individuals compared to non-starved individuals. Therefore, to reduce intra-year variation and increase our ability to detect long-term trends in PCB contamination, the sample has been split into birds that died from starvation (and by definition would have had no fat deposits) and those that died from other causes and had visible fat deposits. Data on extent of fat deposits was only collected after 1990 and so trends in liver PCB concentrations for birds that were not in a starved condition could only be determined for the period 1990-2009.

There was no significant change in PCB congener sum liver concentrations in non-starved sparrowhawk during the period 1990-2009 (Figure 3.1; $F_{1,18} = 0.0$ $P > 0.05$). There appeared to be a small decline in liver PCBs in starved birds over the longer period from 1967 to 2009 (Figure 3.1) but this was not statistically significant ($F_{1,39} = 1.46$, $P = 0.235$). The summed liver concentrations of the Paris 10 congeners and the TEQs have been monitored since 1996 (Figure 3.1). There was no significant change in Paris 10 congener concentrations in non-starved birds ($F_{1,12} = 2.65$, $P = 0.13$). although, rather surprisingly, there was a small, but statistically significant, increase in Paris 10

congener concentrations in starved sparrowhawks ($F_{1,12}=4.91$, $P=0.047$). No change in TEQ concentrations were evident in either starved or non-starved birds over this period ($F_{1,12}\leq 2.46$, $P\geq 0.143$).

The mean mercury concentration declined significantly in sparrowhawk livers between 1966 and 2009 in birds (Figure 3.1; $F_{1,32} = 12.08$, $P= 0.001$). However most of this decline appeared to occur before approximately 1990 and there was no significant decline in liver mercury between 1990 and 2009. Consistent with this, there was no significant decline in liver mercury concentrations in non-starved birds (Figure 3.1: $F_{1,18} = 0.75$, $P>0.05$) over the period 1990-2009.

Time trend analysis other metals is not presented here as monitoring of these elements has only been carried out for 4 years.



4. Persistent organic pollutants and metals in merlin (*Falco columbarius*) eggs

4.1. Contaminant concentrations in eggs collected in 2007 & 2008

Merlin eggs have been monitored long-term for PCBs and organochlorine compounds. The findings from previous analyses are reported elsewhere (Newton et al., 1999a, Newton and Haas, 1988, Newton et al., 1982) and in previous recent reports in this series (Shore et al., 2005a, Shore et al., 2005b, Shore et al., 2006b). As part of a strategic review of the monitoring program within the PBMS the monitoring of contaminants in merlin eggs was stopped from 2009 onwards. However, failed eggs will continue to be collected and archived to facilitate retrospective studies.



The analyses of 9 eggs (one per clutch) collected during 2007 and 12 eggs collected in 2008 are given in Table 4.1.

Table 4.1. Summary of PCB concentrations in merlin eggs received in 2007 & 2008.

Analyte	2007 (n=9)		2008 (n=12)	
	GeoMean	95%CI	GeoMean	95%CI
Sum TEQ (pg/g ww)	6.892	3.003-15.82	7.298	4.449-11.97
Congener sum (µg/g ww)	0.523	0.336-0.813	0.758	0.521-1.102
Selected congeners (ng/g ww):				
PCB 028	0.597	0.174-2.056	1.221	0.469-3.181
PCB 031	ND	-	ND	-
PCB 052	0.024	0.007-0.085	0.015	0.006-0.037
PCB 101	0.063	0.011-0.364	0.119	0.028-0.502
PCB 105	7.345	5.052-10.68	9.770	7.997-11.93
PCB 118	29.6	20.09-43.60	38.51	30.55-48.54
PCB 138	39.81	24.36-65.05	60.55	38.04-96.38
PCB 153	141.6	93.31-214.9	219.3	144.4-333.2
PCB 156	11.37	7.821-16.52	17.15	11.24-26.16
PCB 180	74.39	42.91-129.0	100.9	75.97-133.9

ND is not detected

The data for the eggs collected in 2007 and 2008 confirm that merlins eggs in Britain are still generally contaminated with PCBs. Congener sum PCBs and TEQ residues were detected in all eggs. Overall productivity is thought to be reduced in raptors when sum PCB concentrations in eggs are in excess of 24 µg/g wet wt. (Harris and Elliott, 2011). The concentrations of Congener sum PCBs in merlin eggs were generally low and are below this concentration. Harris & Elliot (2011) suggest that hepatic ethoxyresorufin-O-deethylase (EROD) induction be considered as a cross-species comparable lowest observed effect level (LOEL) for Dioxin-like contaminants which include some PCB congeners. Induced EROD activity is integral to the initiation of biochemical and physiological responses to dioxin-like contaminants and may be considered indicative of low grade effects in birds. In the field, EROD activity is induced in hatchlings of wild birds by threshold concentrations of 400-1600 pg

TEQs/g wet wt. The geometric mean and the maximum calculated TEQ concentration associated with PCB contamination in the merlin eggs received in 2007 and 2008 collectively was 10.29 pg/g wet wt and 43.72 pg/g wet wt, respectively. Therefore it is likely that the TEQ concentrations measured in the merlin eggs collected in these two years were not toxicologically significant.

Mercury was detected in all of the merlin eggs received in 2007 and 2008 (Table 4.2). The geometric mean and the maximum concentration were 2.94 µg/g dry wt and 8.72 µg/g dry wt and was typical of concentrations recorded in previous years (Shore et al., 2005a). Total mercury concentrations greater than approximately 0.6 µg/g wet wt have been proposed as an indicative value associated with impaired reproduction in birds (Shore et al., 2011). When expressed on a wet wt basis, the maximum mercury concentration in the merlin eggs received in the two years studied was 1.46 µg/g wet wt (E9047), and a further nine eggs had mercury concentrations greater than 0.6 µg/g wet wt. All three eggs with mercury concentrations exceeding 1 µg/g wet wt. were collected from nests in the Orkney isles, while the other six eggs with concentrations between 0.6 and 1 µg/g wet wt. were from both the Orkney isles and the North Pennines/Yorkshire Dales area. It is possible that mercury contamination may have had an adverse effect on the development of these eggs.

Both lead and cadmium are not thought to accumulate in avian eggs (Furness, 1996) and this is reflected in the low concentrations measured in the merlin eggs received in 2007 and 2008 (Table 4.2). Studies in Spain on a range of raptor species have measured concentrations of non-detected to 4.54, and 0.001 to 1.12 µg/g ww for lead and cadmium concentrations, respectively (Garcia-Fernandez et al., 2008). Therefore lead and cadmium concentrations in merlin eggs are consistent with those reported in raptor eggs from Spain. Both copper and zinc are essential metals and so their concentrations are expected to be homeostatically controlled.

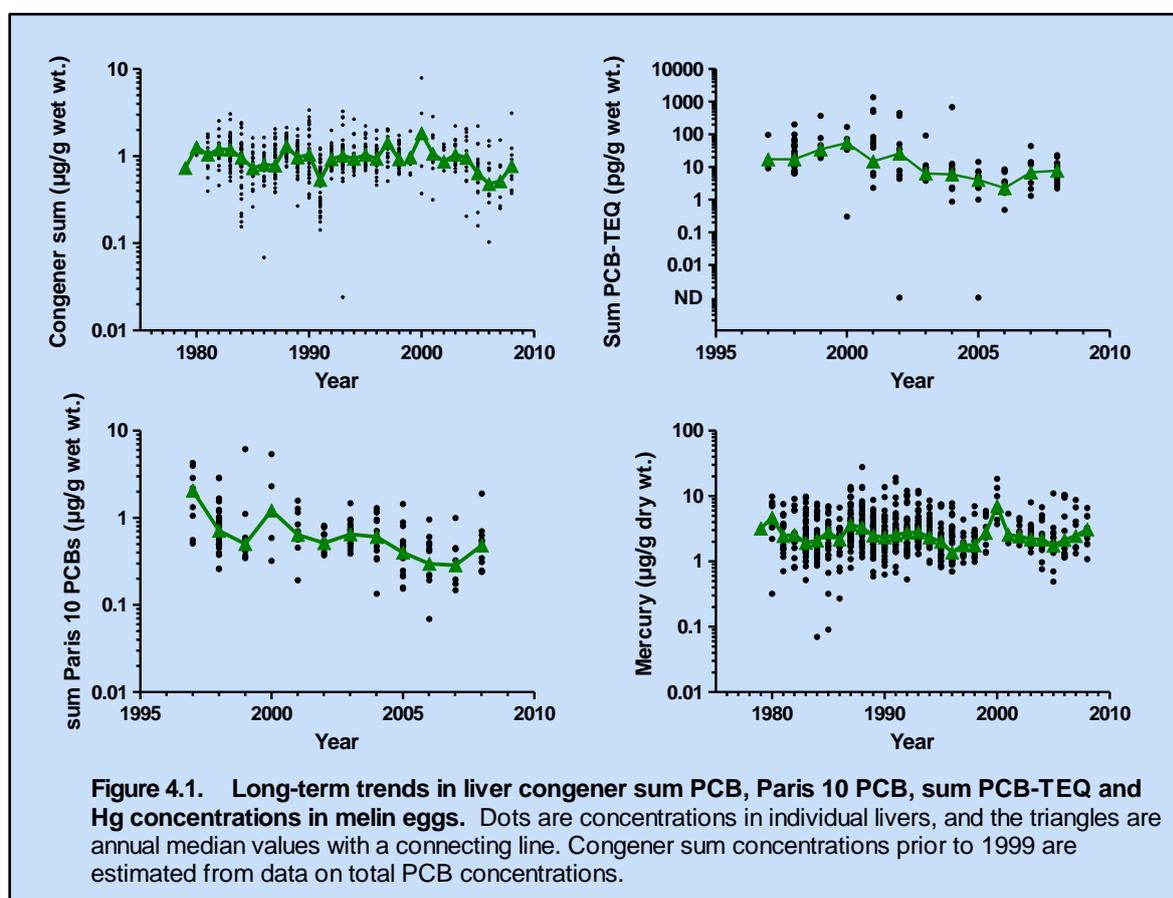
Arsenic has not been widely studied in raptor eggs. Previous studies in Spain have measured wet weight arsenic concentrations in raptor egg contents of up to 0.032 µg/g, measured in black kite (*Milvus migrans*) eggs, which are thought to be low and not related to reproductive effects (Blanco et al., 2003). As the maximum concentration in the merlin eggs received in 2007 and 2008 collectively was 0.031 µg/g wet wt., it is unlikely that arsenic would have an adverse effect on these merlin eggs.

Table 4.2. Summary of metal concentrations (µg/g dry weight) in merlin eggs received in 2007 and 2008.

Analyte	2007 (n=9)			2008 (n=12)		
	GeoMean	lower 95%CI	upper 95%CI	GeoMean	lower 95%CI	upper 95%CI
mercury (Hg)	2.973	1.798	4.914	2.910	2.123	3.990
arsenic (As)	0.068	0.044	0.107	0.065	0.047	0.088
lead (Pb)	0.009	0.007	0.014	0.010	0.007	0.014
cadmium (Cd)	0.001	0.001	0.003	0.002	0.001	0.006
copper (Cu)	2.467	2.238	2.720	2.847	2.482	3.267
zinc (Zn)	28.63	24.08	34.04	36.69	30.07	44.77

4.2. Long-term trends in contaminant concentrations in merlin eggs

There has not been a statistically significant change in PCB congener sum concentrations in merlin eggs during the period 1979-2008 ($F_{1,28}=2.05$, $P=0.164$; Figure 4.1). However, there have been significant declines (Figure 4.1) in the liver concentrations of the Paris 10 congeners ($F_{1,10}=7.07$, $P=0.024$) and in PCB-TEQs ($F_{1,10}=6.15$, $P=0.033$) as measured from 1997, the first year in which these concentrations could be calculated from the analytical data. The PCB congener sum concentration also declined in merlin eggs over this shorter time period ($F_{1,10}=8.96$, $P=0.013$). This suggests that either PCB concentrations in merlins have only started to decline in more recent years or that the total PCBs concentrations in eggs prior to 1999 (the data used to estimate congener sum PCBs concentrations for eggs laid before 1999) included compounds that were not PCBs and which obscured any decline in PCB levels in the eggs.



Mercury concentrations have not significantly changed during the period 1979-2008 ($F_{1,28}=0.69$, $P=0.414$). During this monitoring period the annual geometric mean concentrations of mercury have ranged between 1.74 and 7.72 $\mu\text{g/g dw}$, and the level of mercury contamination quantified in eggs collected in 2007 and 2008 are consistent with these observations. Time trend analysis other metals is not presented here as monitoring of these elements has only been carried out for 4 years.

5. Persistent Organic Pollutants and Metals in golden eagle (*Aquila chrysaetos*) eggs

5.1. Introduction

The findings from the long-term monitoring of contaminants in golden eagle eggs carried out as part of the PBMS have been reported in previous reports by Newton and Galbraith (1991) and were recently summarised as part of the series of reports for the PBMS (Shore et al., 2005a). As part of a strategic review of the monitoring program within the PBMS the monitoring of contaminants in golden eagle eggs was stopped from 2009 onwards. However, failed eggs will continue to be collected and archived to facilitate retrospective studies. Eggs from 7 and 12 clutches were received in 2007 and 2008, respectively. The results of the chemical analyses are given in Table 5.1.



5.2. Contaminant concentrations in eggs collected in 2007 & 2008

The PCB and Hg concentrations (Tables 5.1 and 5.2, respectively) in the eggs were below concentrations thought to impair reproduction (Harris and Elliott, 2011)Shore et al, 2011). Overall productivity is thought to be reduced in raptors when sum PCB concentrations in eggs are in excess of 24 µg/g wet wt. (Harris and Elliott, 2011). The concentrations of Congener sum PCBs in golden eagle eggs were generally low and are below this concentration. Harris & Elliot (2011) suggest that hepatic ethoxyresorufin-O-deethylase (EROD) induction be considered as a cross-species comparable lowest observed effect level (LOEL) for Dioxin-like contaminants which include some PCB congeners. Induced EROD activity is integral to the initiation of biochemical and physiological responses to dioxin-like contaminants and may be considered indicative of low grade effects in birds. In the field, EROD activity is induced in hatchlings of wild birds by threshold concentrations of 400-1600 pg TEQs/g wet wt. The maximum calculated TEQ concentration associated with PCB contamination in the golden eagle eggs received in 2007 and 2008 collectively was 29.72 pg/g wet wt, respectively. Therefore it is likely that the TEQ concentrations measured in the golden eagle eggs collected in these two years were not toxicologically significant.

Lead and cadmium concentrations measured in golden eagle eggs in 2007 and 2008 are low and consistent with those reported in raptor eggs in various studies from Spain (Garcia-Fernandez et al., 2008). Arsenic concentrations are similar to those previously reported in black kite (*Milvus migrans*) eggs from Spain (Blanco et al., 2003). The maximum residue in the golden eagle eggs was 0.043 µg/g wet wt. and so it is unlikely that arsenic would have adverse effects on these eggs.

Table 5.1. Geometric mean concentrations of PCBs in the egg contents of golden eagles received in 2007 and 2008.

Analyte	2007 (N=7)			2008 (N=12)		
	GeoMean	lower 95%CI	upper 95%CI	GeoMean	lower 95%CI	upper 95%CI
Sum TEQ (pg/g ww)	0.307	0.016	5.987	0.841	0.389	1.815
Congener sum (µg/g ww)	0.180	0.035	0.925	0.220	0.103	0.469
Selected congeners (ng/g ww):						
PCB 028	0.120	0.015	0.981	0.011	0.011	0.011
PCB 031	0.062	0.016	0.245	0.026	0.026	0.026
PCB 052	0.023	0.003	0.185	0.013	0.007	0.023
PCB 101	0.064	0.005	0.756	0.091	0.021	0.399
PCB 105	0.210	0.012	3.685	2.229	1.214	4.091
PCB 118	3.839	0.696	21.18	7.269	3.015	17.53
PCB 138	3.546	0.784	16.03	10.13	3.717	27.6
PCB 153	34.34	4.97	237.2	41.60	15.16	114.1
PCB 156	0.400	0.014	11.45	3.705	1.911	7.185
PCB 180	28.66	4.773	172.0	37.44	15.62	89.7

Table 5.2. Geometric mean concentrations of selected inorganic elements in the egg contents of golden eagles received in 2007 and 2008.

Analyte	2007 (n=7)			2008 (n=12)		
	GeoMean	lower 95%CI	upper 95%CI	GeoMean	lower 95%CI	upper 95%CI
mercury (Hg)	0.140	0.054	0.366	0.207	0.113	0.380
arsenic (As)	0.091	0.048	0.169	0.077	0.051	0.117
lead (Pb)	0.029	0.008	0.101	0.021	0.012	0.036
cadmium (Cd)	0.003	0.001	0.007	- ¹	-	-
copper (Cu)	3.902	3.015	5.051	3.939	2.989	5.192
zinc (Zn)	28.66	15.70	52.29	35.33	26.72	46.70

¹ Only one egg collected in 2008 had a detectable concentration of Cd.

5.3. Long-term trends in contaminant concentrations in golden eagle eggs

Trends in residues in eggs from western and eastern areas have previously considered separately because regional variation in exposure of golden eagles to organochlorine pesticides has been noted previously (Lockie et al., 1969). This was thought to be due to sheep comprising a greater proportion of the diet of golden eagles in hilly western Scotland where sheep are abundant; the use of DDT and dieldrin in sheep dips up to the mid-1960s was probably one of the most important routes of exposure of eagles to these compounds (Newton and Galbraith, 1991 and references therein). However, as organochlorine insecticides are not currently the focus of the PBMS these regions are no longer considered separately. However, trends in residues of PCBs and mercury in eggs from coastal and inland territories have been considered separately where possible as seabirds can accumulate high levels of PCBs and mercury (Thompson, 1996) and are likely to be a more important component of the diet in coastal than inland nesting eagles. For the purposes of this analysis, coastal territories have been defined as those known to border the sea, or where such information was lacking, as sites within 3 km of the coast.

In eggs from inland nests (Figure 5.1), the PCB congener sum concentrations have significantly declined over time between 1973 and 2008 ($F_{1,33}=6.85$, $P=0.013$). There are relatively few data on which to assess long-term trends for Paris 10 congener and PCB-TEQ concentrations. However, there are significant declines in the concentrations of both these measures of PCB contamination ($F_{1,9}\geq 5.06$, $P\leq 0.05$) over time, although the decline in PCB-TEQ concentrations is very much influenced by the relatively high concentration measured in eggs in 1997. Annual median mercury concentrations range from non-detected to approximately 2 ug/g dry wt., and there are no clear progressive increases or decreases over time.

PCB congener sum, Paris 10, PCB-TEQ and mercury concentrations are all significantly higher in the eggs of eagles nesting within 3 km of the coastline than in birds nesting inland (Mann-Whitney U test, $P<0.001$ in all cases; Figure 5.2). There are no statistically significant time trends in contaminant concentrations in the eggs of coastal nesting birds (Figure 5.2).

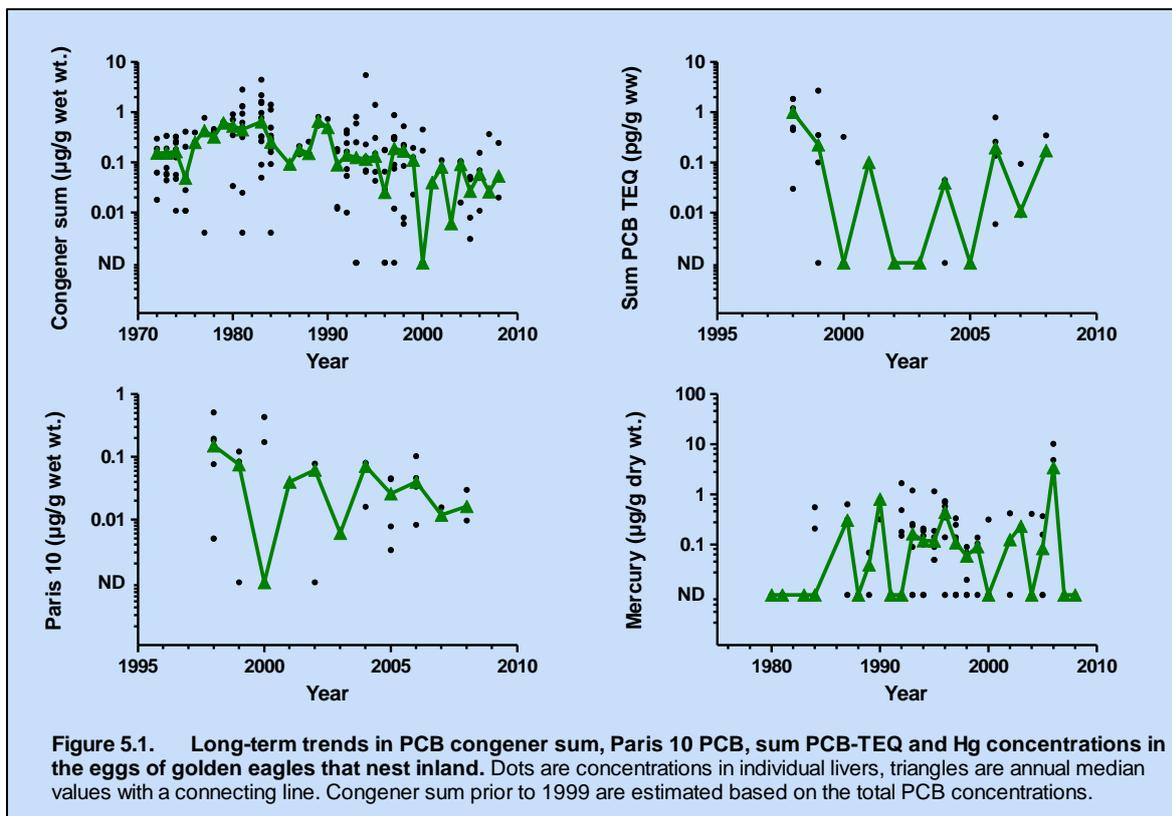


Figure 5.1. Long-term trends in PCB congener sum, Paris 10 PCB, sum PCB-TEQ and Hg concentrations in the eggs of golden eagles that nest inland. Dots are concentrations in individual livers, triangles are annual median values with a connecting line. Congener sum prior to 1999 are estimated based on the total PCB concentrations.

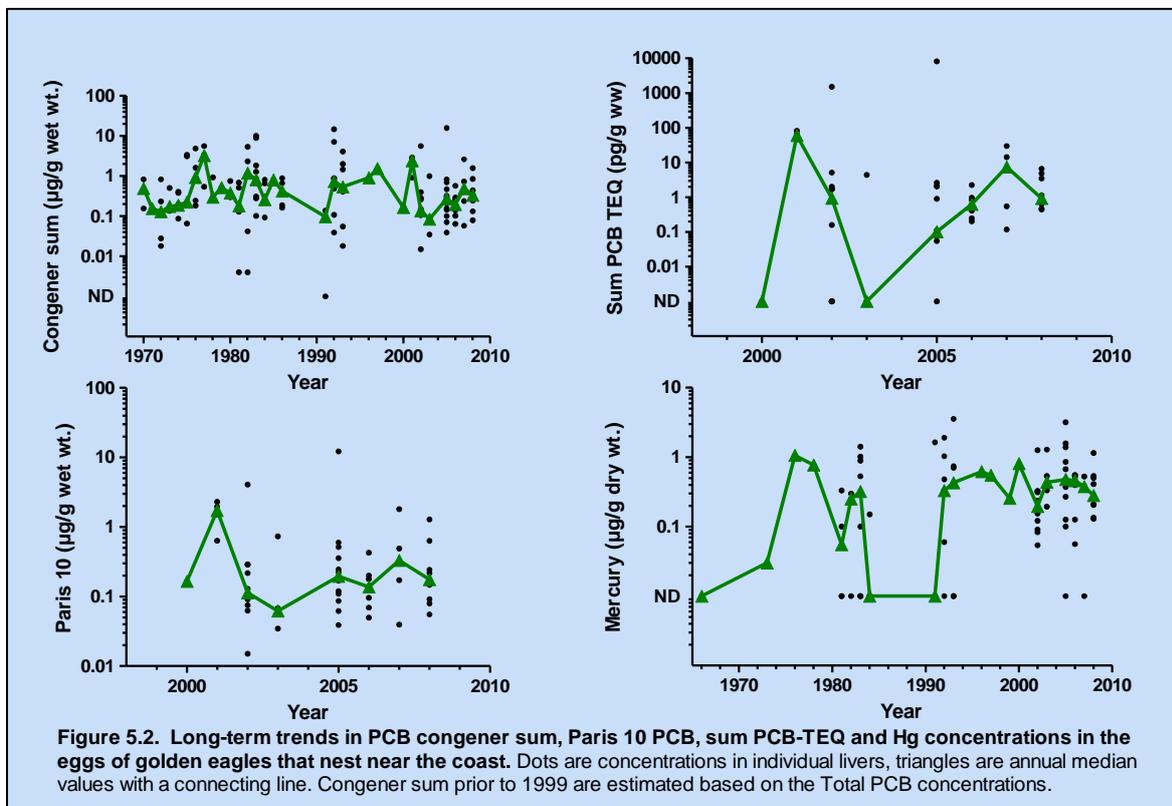


Figure 5.2. Long-term trends in PCB congener sum, Paris 10 PCB, sum PCB-TEQ and Hg concentrations in the eggs of golden eagles that nest near the coast. Dots are concentrations in individual livers, triangles are annual median values with a connecting line. Congener sum prior to 1999 are estimated based on the Total PCB concentrations.

6. Persistent Organic Pollutants and Metals in northern gannet (*Morus bassanus*) eggs

6.1. Introduction

The results of monitoring of gannet eggs examined up to 1988 were published by Newton et al., (1990a) and subsequent data, together with analysis of long-term trends, were recently summarised by Pereira et al. (2009). Gannet eggs are typically collected during visits to the colonies of Bass Rock and Ailsa Craig off the east and west coasts of Scotland, respectively. Collections are made during the laying or the early incubation period, and approximately ten eggs, each from a separate nest, are taken. Collections are made from each colony roughly every two years. In 2007 and 2009 eggs were collected from the Ailsa Craig colony which is situated 13km off the Ayrshire coast. Due to weather conditions during the laying period eggs were not sampled from Bass Rock during 2008; the most recent review of contaminants in eggs from the Bass Rock colony can be downloaded from the [PBMS website](#).



6.2. Contaminant concentrations in eggs collected in 2007 and 2009

PCB congener sum and PCB-TEQ residues were detected in all 20 eggs (Table 6.1). The geometric mean PCB congener sum concentrations were within the range (0.055 and 3.89 $\mu\text{g/g}$ wet wt.) previously reported for eggs from Ailsa Craig. Overall productivity is thought to be reduced in raptors when sum PCB concentrations in eggs are in excess of 24 $\mu\text{g/g}$ wet wt. (Harris and Elliott, 2011). The concentrations of Congener sum PCBs in gannet eggs were generally low and are below this concentration. Harris & Elliot (2011) suggest that hepatic ethoxyresorufin-O-deethylase (EROD) induction be considered as a cross-species comparable lowest observed effect level (LOEL) for Dioxin-like contaminants which include some PCB congeners. Induced EROD activity is integral to the initiation of biochemical and physiological responses to dioxin-like contaminants and may be considered indicative of low grade effects in birds. In the field, EROD activity is induced in hatchlings of wild birds by threshold concentrations of 400-1600 pg TEQs/g wet wt. The maximum calculated TEQ concentration associated with PCB contamination in the gannet eggs received in 2007 and 2009 collectively was 28.19 pg/g wet wt, respectively. Therefore it is likely that the TEQ concentrations measured in the gannet eggs collected in these two years were not toxicologically significant.

The geometric mean concentration of selected metals is given in Table 6.2. Mercury was detected in all 20 eggs collected in 2007 and 2009. The geometric mean and maximum concentration was 2.27 $\mu\text{g/g}$ dry wt and 4.12 $\mu\text{g/g}$ dry wt respectively, and was typical of concentrations recorded in previous years (Walker et al., 2010). When expressed on a wet wt basis, the maximum mercury concentration in gannet eggs received in 2007 and 2009 was 0.81 $\mu\text{g/g}$ wet wt, and this was the only egg that exceeded 0.6 $\mu\text{g/g}$ wet wt, the indicative threshold value associated with impaired hatching that has been proposed for birds (Shore et al., 2011). Of the other non-essential elements, concentrations of lead and cadmium, which are not thought to accumulate in avian eggs (Furness, 1996), and were low and similar to those previously reported. The geometric mean arsenic concentration was similar to the range (0.073-0.382 $\mu\text{g/g}$ dw) reported for common tern (*Sterna*

hirundo) measured in the gannet eggs from Bass Rock (Burger et al., 2009, Burger and Gochfeld, 2003) but higher than that (0.048 µg/g dw) measured in glaucous-winged gulls (*Larus glaucescens*).

Table 6.1. Geometric mean concentrations of PCBs in the egg contents of gannet collected in 2007 and 2009 from Ailsa Craig.

Analyte	2007 (N=10)			2009 (N=10)		
	GeoMean	lower 95%CI	upper 95%CI	GeoMean	lower 95%CI	upper 95%CI
Sum TEQ (pg/g ww)	3.152	1.777	5.590	4.268	1.967	9.261
Congener sum (µg/g ww)	0.501	0.379	0.660	0.534	0.349	0.816
Selected congeners (ng/g ww):						
PCB 028	1.188	0.8431	1.674	0.971	0.294	3.208
PCB 031	0.332	0.143	0.770	0.026	0.026	0.026
PCB 052	0.236	0.0425	1.306	0.813	0.258	2.559
PCB 101	1.268	0.3878	4.145	3.878	2.582	5.826
PCB 105	8.387	6.252	11.25	11.58	7.811	17.18
PCB 118	26.43	20.67	33.8	31.23	20.12	48.47
PCB 138	51.44	36.91	71.69	53.61	34.43	83.48
PCB 153	138.8	104.3	184.7	140.0	92.31	212.5
PCB 156	6.655	5.156	8.590	1.13	0.106	12.06
PCB 180	73.09	55.19	96.78	102.0	66.72	155.8

Table 6.2. Geometric mean concentrations of selected inorganic elements in the contents of gannet eggs collected in 2007 and 2009 from Ailsa Craig.

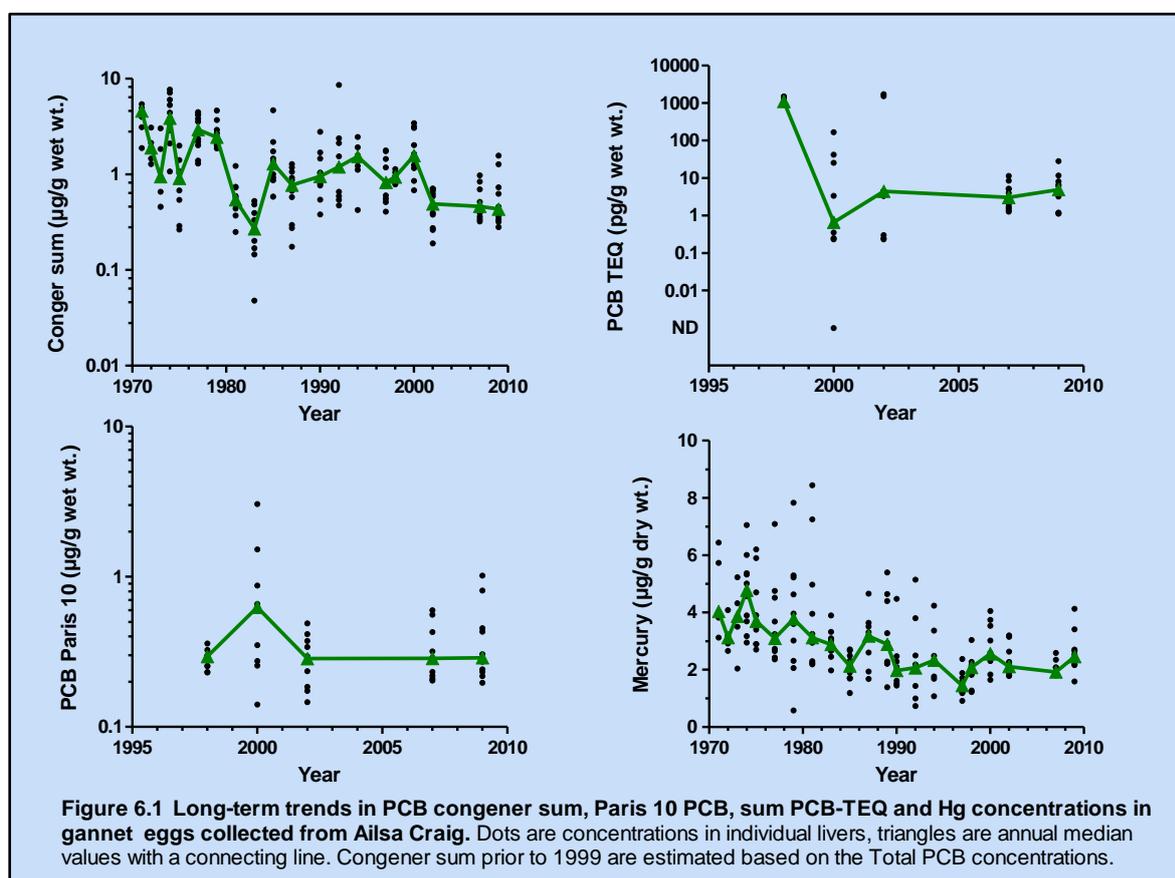
Analyte	2007 (n=10)			2009 (n=10)		
	GeoMean	lower 95%CI	upper 95%CI	GeoMean	lower 95%CI	upper 95%CI
mercury (Hg)	2.031	1.872	2.203	2.531	2.103	3.046
arsenic (As)	0.397	0.295	0.533	0.376	0.315	0.449
lead (Pb)	ND			- ¹	-	-
cadmium (Cd)	0.009	0.006	0.011	0.008	0.007	0.010
copper (Cu)	4.142	3.738	4.590	3.817	3.207	4.543
zinc (Zn)	30.63	24.05	39.01	37.73	30.29	47.00

¹ Pb was not quantified in samples from 2009.

6.3. Long-term trends in contaminant concentrations in gannet eggs collected from Ailsa Craig

There has been a statistically significant decline in PCB congener sum concentrations in gannet eggs during the monitoring period of 1969-2009 ($F_{1,19}=9.56$, $P=0.006$; Figure 6.1). Sum Paris 10 and TEQ concentrations (Figure 6.1) have only been quantified for the last five collections of eggs (1998, 2000, 2002, 2007 and 2009) and time-trend analysis for these measurements has not been conducted because of the relative paucity of data.

Mercury concentrations (Figure 6.1) have declined significantly during the period 1971-2009 ($F_{1,19}=27.95$, $P<0.001$), although most of this decline occurred before 1990 (Figure 6.1). This is the second year that other metals have been quantified in egg contents and so time trend analysis has not been carried out for these analytes.



7. Persistent Organic Pollutants and Metals in sea eagle (*Haliaeetus albicilla*) eggs

7.1. Introduction

Sea eagles were reintroduced to western Scotland between 1976 and 1985. They have had lower breeding success than individuals in some populations in continental Europe, although productivity has been similar to that of birds in Iceland. The relatively poor breeding success of the Scottish population is due to the number of total nest failures, and a few pairs persistently fail to rear young. One potential cause of breeding failure may be exposure to contaminants which the birds could acquire particularly from the marine component (various fish and seabirds) of their diet.



Some of the Scottish white-tailed eagles nest on inaccessible sea cliffs. This makes collection of samples difficult. Two failed eggs were collected in 2007 and a total of 15 eggs have been obtained and analysed during the course of this monitoring scheme. A summary of concentrations in eggs collected previously is given by Walker et al., (2008a).

As part of a strategic review of the monitoring program within the PBMS the monitoring of contaminants in white tailed sea eagle eggs was stopped from 2009 onwards. Failed eggs will continue to be collected and archived to facilitate retrospective studies.

7.2. Results

The wet weight contaminant concentrations in the egg collected in 2007 are given in Table 7.1.

The lipid weight equivalent concentrations of the DDE and PCB congener sum in the egg collected from the Isle of Rum were 535 $\mu\text{g/g}$ and 814 $\mu\text{g/g}$, respectively. Lipid DDE concentrations in sea eagle eggs of 30-50 $\mu\text{g/g}$ and 100-120 $\mu\text{g/g}$ have been suggested as the LOECs for eggshell thickness and productivity, respectively. Complete reproductive failure has been associated with a DDE concentration of 900 $\mu\text{g/g}$ lipid wt (Helander et al., 2002). Adverse effects on productivity due to PCBs appear to occur at lipid concentrations of about 300 $\mu\text{g/g}$ in sea eagle eggs, although, because there is a strong association between PCB and DDE residues, there is some uncertainty about whether such effects are due to PCBs or DDE (Helander et al., 2002). The lipid DDE concentration in the egg collected from Rum exceeded the LOEC for eggshell thickness and productivity. Overall productivity is thought to be reduced in raptors when sum PCB concentrations in eggs are in excess of 24 $\mu\text{g/g}$ wet wt. (Harris and Elliott, 2011). The sum congener PCB concentrations in the egg from Rum exceed this value. The egg had an egg shell index of 3.17, while there was not any embryo development apparent.

The mercury concentration in the egg collected from Rum is below the indicative value associated with impaired hatching in birds (Shore et al., 2011).

Table 7.1. Concentrations of organochlorine insecticides and PCB congener sum (all in µg/g wet wt), TEQs (pg/g dry wt), mercury (µg/g dw) and the shell indices (SI) for the white-tailed sea eagle egg received in 2007. Lipid wt concentrations for organochlorines and PCBs can be calculated by multiplying the wet wt concentrations by the conversion factor (CF).

Egg Number	Year	Location	SI	CF	pp'-DDE	HEOD	Con. Sum	PCB TEQ	Hg
Western Highlands									
E9001	2007	Argyll	-	14.97	3.402	0.188	5.294	51.27	- ¹
E9012	2007	Rum	3.17	13.29	40.26	0.475	61.26	179.5	0.658

¹ mercury was not measured in this egg

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