# Kapitel 3

## Organochlorine contaminants in body tissue of free-ranging white-tailed eagles from northern regions of Germany

Norbert Kenntner, Oliver Krone, Günter Oehme,

Dietrich Heidecke & Frieda Tataruch

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Adulter Seeadler

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## Introduction

The white-tailed eagle (*Haliaeetus albicilla*) is a top predator of the aquatic food web, feeding mainly on fish and waterfowl and it is also known for its opportunistic scavenging.

The northern and central European population of white-tailed eagles actually recovers from critical levels caused by human persecution over centuries, habitat degradation and agricultural use of organochlorine pesticides and organomercury compounds as seed dressing (DEL HOYO et al. 1994; TUCKER & HEATH 1994). The widespread application of DDT in agriculture and forestry since the end of the 1940s caused detrimental effects on the reproduction of raptorial birds like white-tailed eagles, inducing embryo toxicity, eggshell thinning and breakage, followed by a long-term population decline. An extraordinary high exposure to environmental contaminants was reported for the Baltic population of white-tailed eagles (JENSEN et al. 1969; KOIVUSAARI et al. 1980; HELANDER et al. 1982; PAASIVIRTA et al. 1987). In contrast to the Fennoscandian peninsula (FRØSLIE et al. 1986; TARHANEN et al. 1982), Poland (FALANDYSZ 1984; FALANDYSZ 1986; FALANDYSZ et al. 1988; AMAROWICZ et al. 1988) and Japan (IWATA et al. 2000), little information is available about the body burden of persistent organochlorines (OC) in soft tissue of white-tailed eagles collected elsewhere. Furthermore, most long-term studies on the influence of environmental contaminants on European white-tailed eagles were conducted by analyzes of OC in egg contents and therefore represent only the status of reproductive active females (KOIVUSAARI et al. 1980; HELANDER et al. 1982; NYGÅRD & SKAARE 1998).

The present study reports OC concentrations in livers and partly in adipose tissue of 145 free-ranging white-tailed eagles found either dead or dying in the fields of northern Germany between 1979 and 2001. The primary aim of this study was to determine the OC exposure of white-tailed eagles found in Germany and to examine long-term trends. Due to detailed knowledge about the finding circumstances and necropsy for each analyzed specimen we examined the relationships between contaminant levels and age classes, sexes, body condition indices, and sampling date, respectively. Furthermore, we want to contribute environmental contaminant data of white-tailed eagles from Germany for international comparisons and discussions.

## Materials and Methods

### *Sample Collection*

All analyzed white-tailed eagles were free-ranging and found dead or moribund in the northern counties of Germany between 1979 and 2001. Most birds were necropsied in the Institute for Zoo and Wildlife Research (Berlin) and the Institute for Zoology at the University of Halle (Saxony-Anhalt), some samples of specimen were obtained from the Natural History Museum Görlitz (Saxony) and the Institute for Food, Drugs and Animal Diseases (Berlin). All birds were aged due to plumage and bill characteristics. We define juvenile birds as post-nestlings up to an age of half a year, immature birds were found after the 1$^{st}$ October of their first year and adult birds were at least five years of age. The sex was determined during necropsy. Separated organs were stored at –20°C until analysis. Body condition was determined and categorized into five levels from very poor to very good (KENNTNER et al. 2001; KRONE et al. in press). In two nestlings, one immature and two adult birds sex identification was impossible, therefore Table 1 reports the sex, age and origin of 140 analyzed white-tailed eagles. The years of sample collecting are given in Table 2.

**Table 1:** Origin[a], sex and age of 140 white-tailed eagles (*Haliaeetus albicilla*).

| sex | age | Bra | L-S | Sa | M-P | S-H | S-A | n |
|---|---|---|---|---|---|---|---|---|
| male | nestling | 1 | 0 | 0 | 0 | 0 | 0 | 1 |
| | juvenile | 0 | 1 | 0 | 1 | 0 | 0 | 2 |
| | immature | 7 | 0 | 0 | 10 | 1 | 0 | 18 |
| | adult | 22 | 0 | 1 | 18 | 3 | 0 | 44 |
| | n | 30 | 1 | 1 | 29 | 4 | 0 | 65 |
| female | nestling | 0 | 1 | 0 | 1 | 0 | 0 | 2 |
| | juvenile | 1 | 0 | 0 | 3 | 1 | 0 | 5 |
| | immature | 7 | 0 | 4 | 9 | 3 | 2 | 25 |
| | adult | 10 | 0 | 4 | 29 | 0 | 0 | 43 |
| | n | 18 | 1 | 8 | 42 | 4 | 2 | 75 |

[a]Bra = Brandenburg; L-S = Lower Saxony; Sa = Saxony; M-P = Mecklenburg-Western Pomerania; S-H = Schleswig-Holstein; S-A = Saxony-Anhalt

**Table 2:** Table of sampling years of 145 white-tailed eagles (*Haliaeetus albicilla*) found dead or moribund in Germany.

| year | 1979 | 1987 | 1989 | 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1998 | 1999 | 2000 | 2001 | Total |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| n | 3 | 2 | 2 | 3 | 7 | 7 | 8 | 11 | 7 | 10 | 11 | 8 | 19 | 28 | 19 | 145 |

## *Chemical Analysis*

Sample preparation and chemical analyses were performed at the Research Institute of Wildlife Ecology at the University of Veterinary Medicine Vienna in the years from 1999 to 2001. We analyzed routinely the three OC pesticides (p,p´-dichlorodiphenyltrichloroethane [DDT] and its main metabolite p,p´-dichlorodiphenyl-dichloroethylene [DDE], hexachlorobenzene [HCB], $\gamma$-hexachlorocylohexane [$\gamma$-HCH]) and seven polychlorinated biphenyl (PCB) congeners (PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, PCB 180). The PCBs are identified by their IUPAC nomenclature (BALLSCHMITER & ZELL 1980).

For the analysis of OC concentrations about 1 g of thawed liver tissue resp. 0.3 g adipose tissue was weighed for the nearest 0.1 mg and mechanically homogenized with 5 g of purified sea sand (Merck, Darmstadt, Germany) and 5 g anhydrous sodium sulphate (Merck, Darmstadt, Germany). Lipid extraction was performed by Soxleth extraction for 5 h using 50 ml n-hexane for residue analysis (Promochem, Wesel, Germany). The solvent was evaporated, the residue was weighed to determine the lipid contents, followed by a clean-up with 25 ml n-hexane on 4.0 g aluminum oxide 90 (Merck, Darmstadt, Germany).

Identification and quantification of the OC compounds were performed by injecting a proper aliquot of final extract into a capillary gas chromatograph (Perkin Elmer Auto System XL, Norwalk, CT, USA) equipped with $^{63}$N electron capture detector, auto sampler and Turbochrome Navigator 4.1, using a 30 m x 0.25 mm high performance capillary column HP-5 (0.25 µm film cross linked 5%, phenyl methyl silicone phase, Hewlett Packard, Avondale, PA, USA) and argon/methane (10%) as a carrier gas. Standards were obtained from Dr. Ehrenstorfer (Augsburg, Germany). Minimal detection limit varied for OC between 0.1 to 0.3 µg*kg$^{-1}$ wet weight. All results were calculated as mg*kg$^{-1}$ (ppm) on a wet weight (wet wt) basis.

### *Statistical Treatment*

For statistical analyses all residue data were log transformed in an attempt to normalize the data. Kolmogorov-Smirnov test for goodness of fit and Levene´s -test for homogeneity were conducted before data analyses were performed with simple linear regression and Analysis of Variance (ANOVA) using Tukey´s multiple comparison procedure. The significance level was determined at $p \leq 0.05$. The statistical analyses were computed using SPSS 9.0 for PC.

## Results

None of the five eaglets or the seven juveniles white-tailed eagles were found in very poor body condition, as well as none of the juveniles was categorized for very good nutrition status. In the main sample of immature and adult birds the most obvious difference was found between the percentage of birds in poor and moderate body condition (Figure 1).

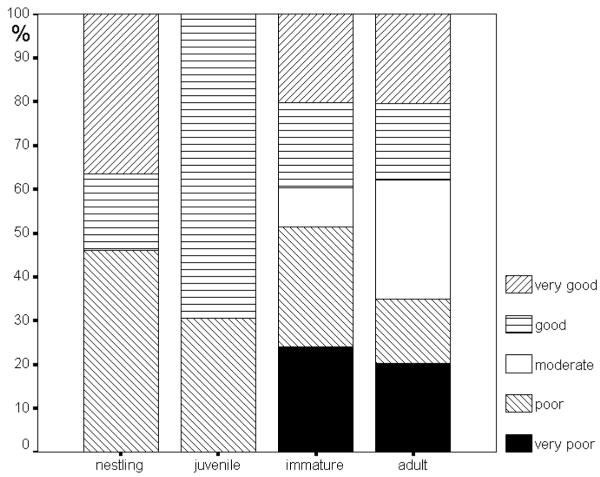


**Fig. 1:** Body condition of white-tailed eagles of different age classes.

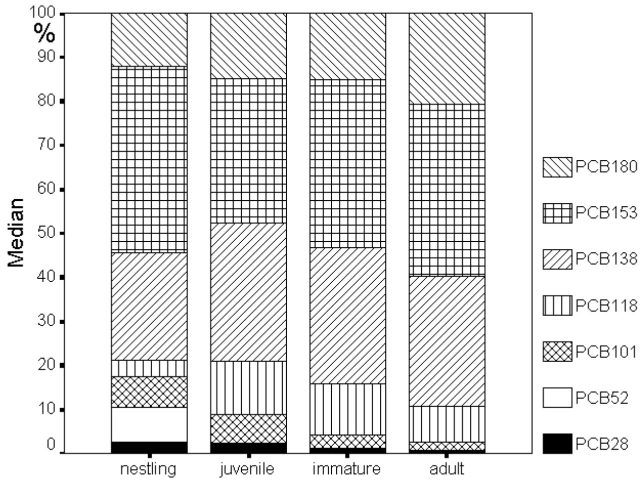


**Fig. 2:** Median percentage of hepatic PCB residue levels from white-tailed eagles of different age classes.

All samples had considerable levels of OC residues, but individual tissue concentrations varied up to several orders of magnitudes (Table 3). Concentrations of lindane, DDT, PCB 28 and PCB 52, respectively, were in general negligible and frequently below detection limit, therefore these compounds were excluded from further statistical analysis. However, concentrations of DDT, PCB 28 and PCB 52 were linked to ΣDDT+DDE (ΣDDT), and the sum of the analyzed seven PCB congeners (ΣPCB), respectively. Nevertheless, the eaglets had a higher percentage of the low substituted PCB 28 and PCB 52 compared with the older age classes, which was presumed to be biased by the lower concentrations of the higher chlorinated PCBs (Figure 2). The results for all OC concentrations in liver tissue and adipose tissue for each of the age-classes are summarized in Table 3.

The Box-plots in Figure 3 and Figure 4 show the 10, 25, 50 (median), 75 and 90 percent percentiles and the extreme values ("outliers"), respectively, of all ΣDDT and ΣPCB concentrations in immature and adult white-tailed eagles from 1979 to 2001.

There was a positive correlation for all OC concentrations between hepatic tissue and adipose tissue (Pearson, $n = 26$, $p < 0.001$) as shown in Figure 5 for HCB, ΣDDT and ΣPCBs. Likewise all hepatic OCs were correlated highly significant with each of the other hepatic OCs (Pearson, $n = 145$, $p < 0.001$).

**Table 3:** Range, median, mean and standard deviation of organochlorine concentrations in livers (n = 145) and adipose tissue (n = 26) from white-tailed eagles (*Haliaeetus albicilla*). Values are given in mg*kg$^{-1}$ (ppm) on a wet-weight basis[a].

| | | | Liver | | | | | | Adipose | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| age | nestling | juvenile | immature | adult | total | ND (%) | juvenile | immature | adult | total | ND (%) |
| n | 5 | 7 | 44 | 89 | 145 | | 1 | 13 | 12 | 26 | |
| HCB | 0.001-0.016 | 0.001-0.273 | 0.001-0.325 | 0.001-1.19 | 0.001-1.19 | 0 | 0.085 | 0.041-9.77 | 0.115-1.15 | 0.041-9.77 | 0 |
| | 0.008 | 0.004 | 0.014 | 0.019 | 0.016 | | | 0.427 | 0.271 | 0.350 | |
| | 0.008±0.007 | 0.041±0.102 | 0.042±0.068 | 0.059±0.145 | 0.051±0.122 | | | 1.662±2.91 | 0.364±0.276 | 1.002±2.13 | |
| γ-HCH | ND-0.025 | ND-0.006 | ND-0.019 | ND-0.070 | ND-0.070 | 29.0 | 0.033 | ND-0.436 | ND-0.488 | ND-0.488 | 7.7 |
| | 0.002 | 0.001 | 0.001 | 0.001 | 0.001 | | | 0.024 | 0.022 | 0.024 | |
| | 0.006±0.011 | 0.001±0.002 | 0.002±0.004 | 0.002±0.008 | 0.002±0.007 | | | 0.053-0.116 | 0.068±0.135 | 0.059±0.121 | |
| DDT | ND-0.037 | ND-0.028 | ND-0.626 | ND-1.52 | ND-1.52 | 13.8 | 0.065 | 0.023-1.89 | 0.175-2.16 | 0.023-2.16 | 0 |
| | 0.002 | 0.005 | 0.013 | 0.028 | 0.016 | | | 0.558 | 0.746 | 0.732 | |
| | 0.013±0.017 | 0.007±0.010 | 0.036±0.102 | 0.103±0.252 | 0.075±0.208 | | | 0.817±0.605 | 0.886±0.577 | 0.820±0.589 | |
| DDE | 0.020-0.207 | 0.024-0.853 | 0.062-45.3 | 0.121-399.4 | 0.020-399.4 | 0 | 3.17 | 3.47-83.3 | 21.2-249.7 | 3.17-249.7 | 0 |
| | 0.162 | 0.133 | 0.588 | 2.383 | 1.278 | | | 33.0 | 63.1 | 45.7 | |
| | 0.118±0.087 | 0.211±0.287 | 2.41±7.07 | 12.2±44.7 | 8.23±35.5 | | | 37.5±30.5 | 101.3±82.1 | 65.6±67.8 | |
| ΣDDT | 0.022-0.232 | 0.030-0.880 | 0.069-45.9 | 0.132-400.2 | 0.022-400.2 | 0 | 3.24 | 3.49-85.2 | 22.7-250.4 | 3.24-250.4 | 0 |
| | 0.162 | 0.138 | 0.593 | 2.42 | 1.28 | | | 34.4 | 63.8 | 46.2 | |
| | 0.131±0.099 | 0.217±0.296 | 2.44±7.16 | 12.3±44.8 | 8.30±35.6 | | | 38.3±31.0 | 102.2±82.3 | 66.5±68.0 | |
| PCB28 | <0.001-0.144 | ND-0.065 | ND-0.109 | ND-0.555 | ND-0.555 | 7.6 | ND | ND-2.84 | ND-0.946 | ND-2.84 | 11.5 |
| | 0.004 | 0.002 | 0.010 | 0.012 | 0.010 | | | 0.206 | 0.199 | 0.204 | |
| | 0.045±0.063 | 0.011±0.024 | 0.019±0.027 | 0.038±0.079 | 0.031±0.066 | | | 0.459±0.773 | 0.293±0.280 | 0.365±0.578 | |
| PCB52 | ND-0.513 | ND-0.035 | ND-0.253 | ND-0.720 | ND-0.720 | 44.1 | ND | ND-0.993 | ND-2.01 | ND-2.01 | 42.3 |
| | 0.013 | <0.001 | <0.001 | 0.004 | 0.003 | | | 0.082 | 0.013 | 0.045 | |
| | 0.109±0.226 | 0.007±0.010 | 0.020±0.050 | 0.031±0.091 | 0.029±0.087 | | | 0.197±0.299 | 0.258±0.570 | 0.217±0.434 | |
| PCB101 | 0.002-0.052 | <0.001-0.168 | 0.001-1.124 | 0.004-1.59 | <0.001-1.59 | 0 | 0.059 | 0.102-3.52 | 0.170-7.52 | 0.059-7.52 | 0 |
| | 0.011 | 0.006 | 0.025 | 0.043 | 0.033 | | | 1.64 | 1.12 | 1.16 | |
| | 0.017±0.020 | 0.032±0.061 | 0.079±0.194 | 0.128±0.275 | 0.105±0.242 | | | 1.467±1.058 | 1.17±1.97 | 1.51±1.53 | |
| PCB118 | ND-0.069 | ND-0.113 | 0.002-4.93 | 0.005-9.43 | ND-9.43 | 1.4 | 0.204 | 0.296-17.0 | 1.37-24.3 | 0.204-24.3 | 0 |
| | 0.006 | 0.012 | 0.098 | 0.182 | 0.126 | | | 4.21 | 5.63 | 4.491 | |
| | 0.020±0.029 | 0.028±0.041 | 0.266±0.751 | 0.728±1.68 | 0.530±1.40 | | | 4.88±4.94 | 9.39±8.95 | 6.78±7.34 | |
| PCB138 | 0.012-0.180 | 0.003-0.571 | 0.009-23.6 | 0.058-38.2 | 0.003-38.2 | 0 | 0.603 | 0.840-51.7 | 5.51-89.1 | 0.603-89.1 | 0 |
| | 0.040 | 0.031 | 0.260 | 0.668 | 0.438 | | | 15.9 | 19.8 | 16.4 | |
| | 0.075±0.068 | 0.114±0.203 | 1.10±3.70 | 2.59±6.37 | 1.93±5.45 | | | 18.3±16.2 | 34.7±32.7 | 25.2±26.2 | |
| PCB153 | 0.012-0.119 | 0.013-0.605 | 0.010-38.6 | 0.068-47.9 | 0.010-47.9 | 0 | 0.707 | 0.767-69.2 | 2.58-120.9 | 0.707-120.9 | 0 |
| | 0.068 | 0.033 | 0.320 | 0.883 | 0.508 | | | 21.3 | 28.6 | 22.7 | |
| | 0.061±0.040 | 0.133±0.212 | 1.51±5.885 | 3.35±8.06 | 2.52±7.16 | | | 24.2±21.3 | 46.3±45.5 | 33.5±36.0 | |
| PCB180 | 0.012-0.046 | 0.004-0.255 | 0.004-19.5 | 0.030-27.8 | 0.004-27.8 | 0 | 0.384 | 0.194-30.6 | 3.641-94.5 | 0.194-94.5 | 0 |
| | 0.019 | 0.015 | 0.126 | 0.467 | 0.256 | | | 8.76 | 10.9 | 9.93 | |
| | 0.024±0.013 | 0.050±0.091 | 0.765±3.00 | 2.16±5.10 | 1.56±4.38 | | | 10.8±10.1 | 28.9±34.0 | 18.7±25.6 | |
| ΣPCB | 0.049-1.05 | 0.032-1.78 | 0.051-87.1 | 0.208-125.5 | 0.032-125.5 | 0 | 1.96 | 2.42-170.7 | 18.6-327.2 | 1.96-327.2 | 0 |
| | 0.220 | 0.118 | 0.883 | 2.471 | 1.50 | | | 51.3 | 67.5 | 55.1 | |
| | 0.352±0.402 | 0.375±0.628 | 3.76±13.4 | 9.02±21.5 | 6.71±18.6 | | | 60.3±52.4 | 121.4±121.2 | 86.3±94.9 | |

[a]ND = not detectable

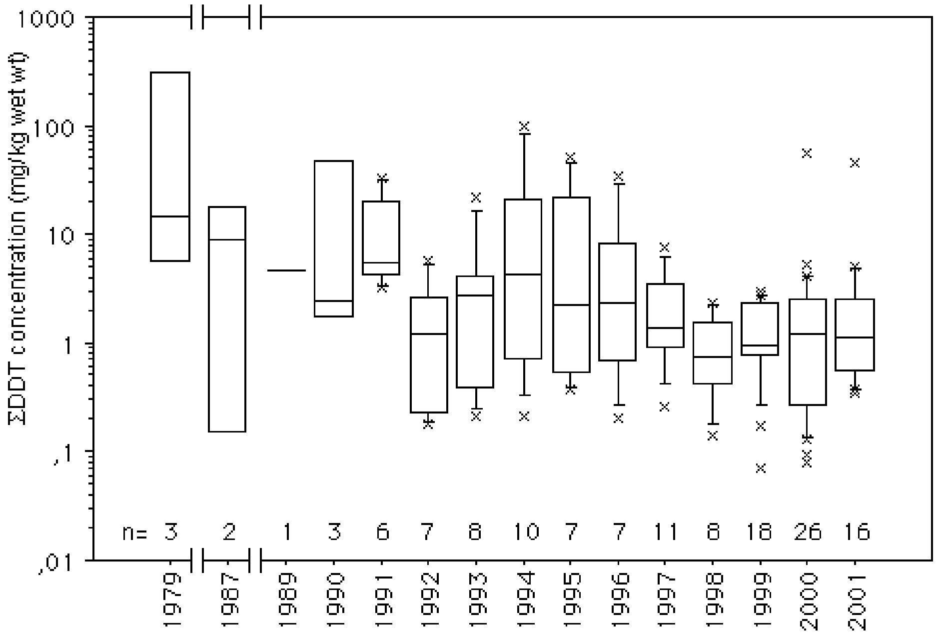


**Fig. 3:** Hepatic ΣDDT (mg/kg) concentrations of 133 adult and immature white-tailed eagles from Germany.

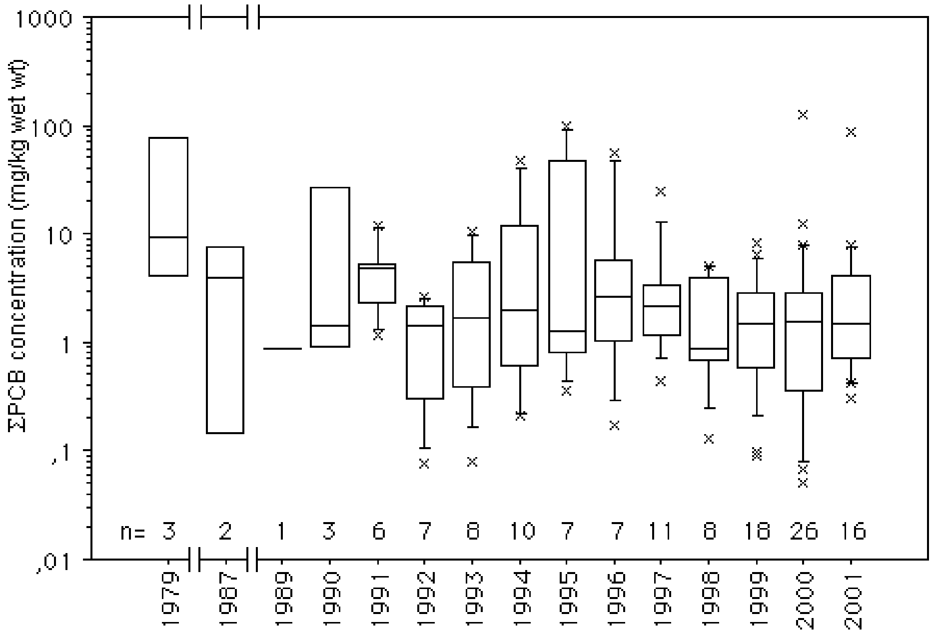


**Fig. 4:** Hepatic ΣPCB (mg/kg) concentrations of 133 adult and immature white-tailed eagles from Germany.

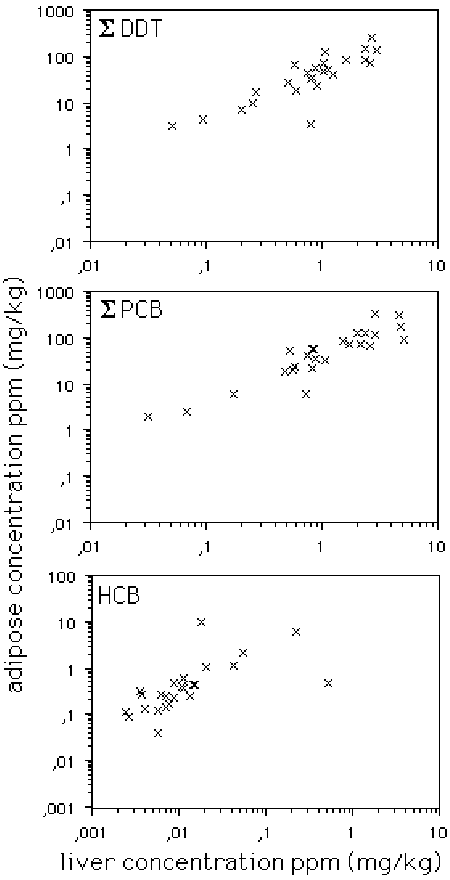


**Fig. 5:** $Log_{10}$ plotted scattergram for OC concentrations in adipose tissue vs. hepatic concentrations of 26 white-tailed eagles.

One-way ANOVA with Tukey adjusting for multiple tests indicated significant higher hepatic concentrations of ΣDDT, PCB 118, PCB 138, PCB 153, PCB 180 and hence also for linked ΣPCBs for adults with $p \leq 0.001$ compared with each of the other age classes. Likewise the liver concentrations for ΣDDT were significantly higher in immature birds than in juveniles ($p = 0.026$) or eaglets ($p = 0.018$), respectively. Whereas adult birds had also higher liver concentrations for PCB 101 ($p = 0.008$) than juveniles, the immature had higher hepatic concentrations for PCB 118 ($p = 0.005$), PCB 138 ($p = 0.020$), PCB 180 ($p = 0.029$) and ΣPCBs ($p = 0.041$) than juveniles, as well as immature birds had higher concentrations for PCB 118 ($p = 0.003$) than eaglets. There were either no significant differences between any age-class for HCB, nor between eaglets and juveniles for any analyzed OCs. Furthermore, the adult white-tailed eagles had higher concentrations of ΣDDT ($F = 7.981$, $p = 0.01$) in adipose tissue than immature birds. No significant differences between sexes were found, with the exception of HCB residues in adipose tissue, which are higher in males than females ($F = 4.266$, $p = 0.05$).

During the period from 1990 to 2001 there was a significant decline for hepatic OCs of immature and adult white-tailed eagles ($n = 127$) for HCB ($F = 7.620$; $\beta = -0.240$; $R^2 = 0.057$; $p = 0.007$) and ΣDDT ($F = 11.570$; $\beta = -0.290$; $R^2 = 0.085$; $p = 0.001$). The declining ratio of ΣDDT:ΣPCB ($F = 35.247$; $\beta = -0.469$; $R^2 = 0.22$; $p < 0.001$), calculated from the non-transformed data, proved the disproportional change of ΣDDT and ΣPCB during the 1990s up to the year 2001 (Figure 6).

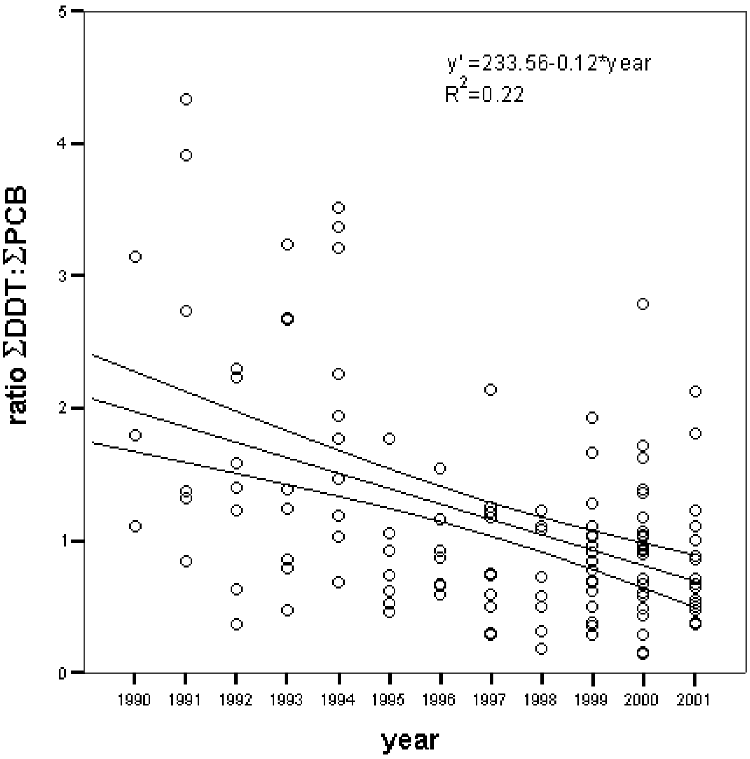


**Fig. 6:** Linear regression for the ratio of ΣDDT:ΣPCB of 127 immature and adult white-tailed eagles from 1990 to 2001. The outer lines show the 95% confidence interval for the linear regression.

Body condition was strongly associated with OC concentrations in livers (ANOVA; $p \leq 0.001$), but not with OC concentrations of adipose tissue. Hepatic HCB, ΣDDT and ΣPCBs concentrations were higher in white-tailed eagles in very poor condition than in good ($p = 0.011$; $p = 0.003$; $p = 0.005$) and very good ($p < 0.001$; $p < 0.001$; $p < 0.001$) condition, and likewise in birds in moderate condition than in very good condition ($p = 0.001$; $p = 0.008$; $p = 0.004$), respectively (Figure 7). However, whereas the indices given for body condition were associated well with the extractable liver fat, and were not biased by sample year or age structure we have no explanation for the overall lower OC concentrations for the birds categorized for poor body condition.

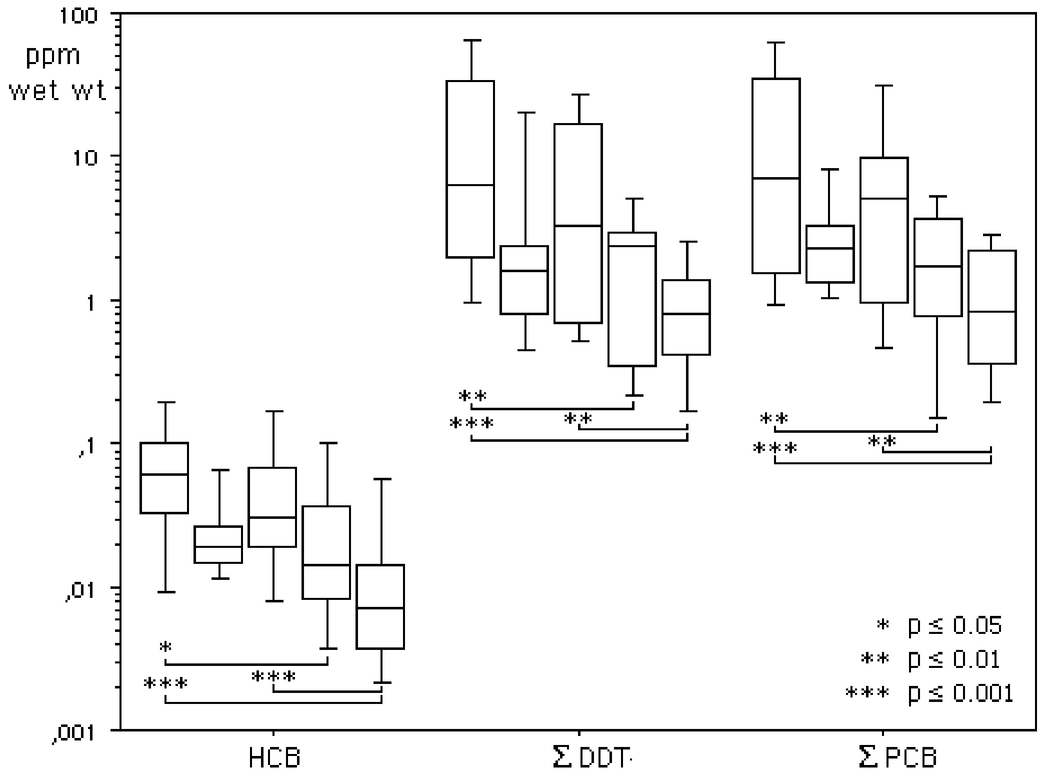


**Fig. 7:** Hepatic OC concentrations of 123 immature and adult white-tailed eagles (1990 - 2001) of very poor (n = 14), poor (n = 15), moderate (n = 15), good (n = 38) and very good (n = 41) body condition, each from left to right. The Box-plots show the 10, 25, 50 (median), 75 and 90 percent percentiles. Significant levels were computed on log transformed data.

## Discussion

Concentrations of the pesticides HCB, ΣDDT and the ratio of ΣDDT:ΣPCB in livers of immature and adult white-tailed eagles in northern Germany decreased significantly from 1990 to 2001. However, no time trend for hepatic ΣPCB concentrations during this period could be proved for the same material. Highest residues of OCs were found in liver tissue of the few birds collected in 1979 and the 1980s, with one female specimen found in debilitated condition in 1979, with hepatic ΣDDT and PCB concentrations of 400.23 ppm and 99.10 ppm which is indicative for lethal ΣDDT exposure (GARCELON & THOMAS 1997).

Our findings for declining OC pesticide concentrations in livers of white-tailed eagles agree with the different periods of their usage in Germany and elsewhere.

Whereas the use of DDT was completely banned in the Federal Republic of Germany (FRG) in 1972 (agriculture) resp. in 1976 (forestry) its application was only continuously reduced in the former German Democratic Republic (GDR) during the 1970s. In 1983 and 1984 during a massive appearance of the moth *Lymantria monacha* in forests DDT was applied by airplanes in high amounts. Finally it was banned in 1988 before the German reunion in 1990.

The fungicide HCB was applicated until 1977 and 1984 in the FRG and the GDR, respectively (BAUM & CONRAD 1978; BEITZ et al. 1991; HEINISCH & WENZEL-KLEIN 1994). With the exception of the ten white-tailed eagles from Lower Saxony and Schleswig-Holstein (Table 1) all birds were collected in regions of the former GDR, where white-tailed eagles still have their highest population density all over Germany (HAUFF 1998). Therefore our data represent mainly tissue concentrations of birds collected in regions were DDT was banned several years after the international measures against DDT during the early 1970s (BIGNERT et al. 1998).

Adult white-tailed eagles had highest hepatic concentrations for all higher chlorinated PCBs (Penta-, Hexa-, Heptachlorobiphenyls) and ΣDDT, which are well known for their persistence and low excretion rate in birds (STICKEL et al. 1984a; STICKEL et al. 1984b). However, our findings did not prove significant differences during ageing for HCB residues. But generally hepatic HCB residues are low and excluding the nestling birds there is a non-significant trend for higher means and medians during ageing.

The concentrations of ΣDDT as well of all higher chlorinated PCBs are highly correlated with body condition, with higher concentrations in birds in very poor and moderate body condition. These higher OC concentrations in emaciated birds were also reported by several other investigations. During catabolic situations the body lipid reserves are depleted associated with contaminant mobilization. The lipophilic OCs were redistributed through the blood stream to organs with high metabolic activity, e.g. the liver (BOGAN & NEWTON 1977; COOKE et al. 1982; FRØSLIE et al. 1986; ELLIOTT et al. 1996). However, the inverse relationship between extractable lipids and OC concentrations proved the liver as target organ for lipophilic OC during periods of food deprivation. Physico-chemical binding of OC to hepatocytes or hepatic macromolecules may enhance the liver concentrations during metabolism of hepatic lipid reserves (PEREG et al. 2001).

Only two papers on liver residues of OCs in white-tailed eagles in Germany have been published so far. KOEMAN et al. (1972) analyzed eight eggs from 1969 to 1971 and one liver sample of an adult white-tailed eagle found dead in 1969 in Schleswig-Holstein with hepatic residues of 4.0 ppm, 35.0 ppm and 0.36 ppm for DDE, PCBs and HCB, respectively (wet wt). OEHME (1980) reported ΣDDT and PCBs residues in 31 livers and 20 adipose tissue samples of white-tailed eagles from the northern region of the former GDR. Mean ΣDDT residues in livers declined from 68 ppm wet wt to 3.2 ppm from 1966 to 1977, whereas hepatic ΣPCB as well as ΣDDT and ΣPCB in adipose tissue remained high during this period.

Most data of OCs in hepatic tissue and other body tissues of white-tailed eagles were reported from Norway and Poland (Table 4), but as most other Baltic and European surveys on this species the hepatic PCB concentrations were not reported congener-specific. However, assuming comparable chemical analyses and calculations for ΣPCBs, our data are within the range reported from these European surveys carried out in the 1980s.

**Table 4:** Organochlorine concentrations in livers of white-tailed eagles from Europe and Hokkaido/Japan. Values are given in ppm on a wet-weight basis[a].

| country | sample years | n | HCB | γ-HCH | DDE | PCB | authors |
|---|---|---|---|---|---|---|---|
| Germany | 1969 | 1 | 0.36 | NA | 4.00 | 35.0 | KOEMAN et al. 1972 |
| Poland | 1981 | 1 | 0.84 | 0.53 | 25.00 | 180.0 | FALANDYSZ&SZEFER 1983 |
| Poland | 1982 | 1 | 2.20 | NA | 300.00 | 880.0 | FALANDYSZ 1984 |
| Norway | 1965-1983 | 24-72 | <0.01-0.52 | <0.01-0.05 | <0.1-89.0 | 0.1-180.0 | FRØSLIE et al. 1986 |
| Poland | 1984 | 2 | 0.03+0.36 | ND+0.02 | 4.40+13.0 | 6.90+13.0 | FALANDYSZ 1986 |
| Finland | 1984-1985 | 4 | 0.06-1.62 | ND | 0.24-22.2 | 3.03-462.0 | TARHANEN et al. 1989 |
| Poland | 1987 | 1 | NA | 0.03 | 0.92 | 1.11 | AMAROWICZ et al. 1989 |
| Poland | 1986+1987 | 4 | 0.02-1.2 | ND-0.15 | 3.60-350.0 | 1.90-540.0 | FALANDYSZ et al. 1988 |
| Japan | 1997 | 2 | 0.01+0.01 | NA | 0.59-2.50 | 1.60+6.00 | IWATA et al. 2000 |
| Germany | 1979-2001 | 145 | 0.001-1.19 | ND-0.07 | 0.02-399.4 | 0.03-125.5 | this study |

[a]ND = not detectable; NA = not analyzed

Congener-specific analysis of 44 PCBs were reported from breast muscles of 10 white-tailed eagles from Poland indicating that the IUPAC Nos 99, 118, 138, 153, 180, 187 and one unidentified heptachlorobiphenyl contribute the major portion to the total PCBs (FALANDYSZ et al. 1994). Nearly the same PCBs pattern was also found in muscles of three female white-tailed eagles collected between 1981 and 1991 in Finland (KOISTINEN et al. 1997). In a second paper on OCs concentrations in breast muscles of 14 white-tailed eagles collected in Poland from 1991 to 1995 FALANDYSZ et al. (2000) reported a range of 0.0047 - 300 ppm and 0.0018 - 490 ppm for ΣDDT and ΣPCB (wet wt), respectively, with higher contaminant burden of eagles from the coastal area of the Baltic than birds collected in inland regions.

No significant temporal trend for ΣDDT and ΣPCB was found for 72 liver samples of white-tailed eagles from Norway for the period from 1973 to 1983 (FRØSLIE et al. 1986), but different age classes were not considered. In contrast to these liver results, a significant decline of DDT and DDE was proven for eggs of Baltic white-tailed eagles from Finland and Sweden between 1974 to 1978, and 1965 to 1978, respectively (KOIVUSAARI et al. 1980; HELANDER et al. 1982; HELANDER 1983). NYGÅRD & SKAARE (1998) proved a significant decline not only for ΣDDT but also for ΣPCBs in eggs of white-tailed eagles from the Norwegian Atlantic coast from 1974 to 1994. Their findings additionally indicate a general lower OC exposure of the Norwegian white-tailed eagles compared with the population of the Baltic.

Numerous investigations have reported the OC exposure of the conspecific bald eagle (*Haliaeetus leucocephalus*) from North America since the 1960s up to nowadays, indicating a negative correlation between declining ΣDDT residues in organs or egg contents, and the population recovery of bald eagles throughout the past decades (GRIER 1982). However, in comparison with studies conducted throughout the 1960s and 1970s the first decline of OC concentrations in total carcasses and cerebral tissue of Bald eagles was obvious since 1975 (KAISER et al. 1980).

The OC levels in livers of 75 bald eagles from British Columbia/Canada, found between 1989 and 1994, were similar with those found in our samples between 1990 and 2001. This study also proved that PCB 118, PCB 138, PCB 153 and PCB 180, as well as PCB 99 and PCB 182 are the dominating congeners in livers of bald eagles (ELLIOTT et al. 1996).

Our data of declining ΣDDT throughout the 1990s, together with the fact of extraordinary high concentrations in samples from 1979 and the 1980s, may support the hypothesized relationship for depressed breeding success through high DDT exposure for white-tailed eagles in Germany until the mid 1980s. Actually the recent population recovery of white-tailed eagles in Germany is assumed for its important contribution to the expanding of this species to yet unoccupied habitats in western Europe. Therefore all white-tailed eagles found in the fields should be examined for their exposure of contaminants, either with respect to wildlife poisoning, e.g. lead poisoning or carbofuran (TATARUCH et al. 1998; KENNTNER et al. 2001; KRONE et al. in press), or to biomagnification of environmental pollutants. Due to the position at the top of the aquatic food web and their public interest, the white-tailed eagle is suggested as a sensitive species for monitoring the exposure of wildlife to environmental hazards.

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## Abstract

Concentrations of the organochlorine pesticides HCB, $\gamma$-HCH, DDT and its metabolite DDE, and of seven PCB congeners were analyzed in livers and adipose tissue samples of 145 white-tailed eagles (*Haliaeetus albicilla*) found dead or moribund in northern states of Germany from 1979 to 2001. Most birds were found in the fields of the region of the former German Democratic Republic where the insecticide DDT was used until 1988. Therefore our samples represent mainly residue data of specimen following the ban of DDT in these regions.

Contaminant levels of 127 immature and adult birds found between the years 1990 and 2001 were in general below threshold levels known for detrimental effects for birds of prey. The highest level of ΣDDT was detected in an adult bird found dead in 1979. Residues of most organochlorines were highly significantly correlated between hepatic and adipose tissue. Concentrations of ΣDDT increase during ageing, whereas only the levels of the higher chlorinated PCBs were higher in tissues of adult birds compared with the younger age classes. Hepatic residues of ΣDDT,

HCB and the ratio of ΣDDT:ΣPCB, respectively, were significantly declining from the year 1990 to 2001. The indices given for body condition of specimen were significantly correlated with liver concentrations, indicating higher residues in more emaciated birds.

## References

AMAROWICZ R, SZYMKIEWICZ M, GLAZEWSKI R, KORCZAKOWSKA B, MARKIEWICZ K, MELLIN M (1989): Content of chlorinated hydrocarbons, polychlorinated biphenyls and heavy metals in tissue of the white-tailed eagle and lesser spotted eagle. *Przeglad Zoologiczny* 33: 613-617

BALLSCHMITER K & ZELL M (1980): Analysis of polychlorinated biphenyls (PCB) by glass capillary gaschromatography. *Fresenius Zeitschrift für Analytische Chemie* 302: 20-31

BAUM F & CONRAD B (1978): Greifvögel als Indikatoren für Veränderungen der Umweltbelastung durch chlorierte Kohlenwasserstoffe. *Tierärztliche Umschau* 12: 661-668

BEITZ H, SCHMIDT HH, HÖRNICKE E, SCHMIDT H (1991): *Erste Ergebnisse der Analyse zur Anwendung von Pflanzenschutzmitteln und ihren ökologisch-chemischen und toxikologischen Auswirkungen in der ehemaligen DDR*. Mitteilungen der Biologischen Bundesanstalt für Land- und Forstwirtschaft, Berlin-Dahlem: Heft 274, Berlin, 123 pp

BIGNERT A, OLSSON M, PERSSON W, JENSEN S, ZAKRISSON S, LITZÉN K, ERIKSSON U, HÄGGBERG L, ALSBERG T (1998): Temporal trends of organochlorines in Northern Europe, 1967 - 1995. Relation to global fractionation, leakage from sediments and international measures. *Environmental Pollution* 99: 17-198

BOGAN JA & NEWTON I (1977): Redistribution of DDE in sparrowhawks during starvation. *Bulletin of Environmental Contamination and Toxicolology* 18: 317-321

COOKE AS, BELL AA, HAAS MB (1982): *Predatory Birds, Pesticides and Pollution*. Institute of Terrestrial Ecology, Cambridge, UK, 74 pp

DEL HOYO J, ELLIOTT A, SARGATAL J (1994): *Handbook of the birds of the world, Vol. 2: New world vultures to guinafowl*. Lynx Edicions, Barcelona, Spain

ELLIOTT JE, WILSON LK, LANGELIER KW, NORSTROM RJ (1996): Bald eagle mortality and chlorinated hydrocarbon contaminants in livers from British Columbia, Canada, 1989-1994. *Environmental Pollution* 94: 9-18

FALANDYSZ J (1984): Metals and organochlorines in a female white-tailed eagle from Uznam Island, Southwestern Baltic Sea. *Environmental Con*servation 11: 262-263

FALANDYSZ J (1986): Metals and organochlorines in adult and immature males of white-tailed eagles. *Environmental Conservation* 13: 69-70

FALANDYSZ J, JAKUCZUN B, MIZERA T (1988): Metals and organochlorines in four female white-tailed eagles. *Marine Pollution Bulletin* 19: 521-526

FALANDYSZ J, STRANDBERG L, MIZERA T, KALISINSKA E (2000): The contamination of white-tailed sea eages with organochlorines in Poland. *Roczniki Panstwowego Zakladu Higieny* 51:7-13

FALANDYSZ J, YAMASHITA N, TANABE S, TATSUKAWA R, MIZERA T, JAKUCZUN B (1994): Congener-specific analysis of polychlorinated biphenyls in white-tailed sea eagles (*Haliaeetus albicilla*) collected in Poland. *Archives of Environmental Contamination and Toxicology* 26: 13-22

FRØSLIE A, HOLT G, NORHEIM G (1986): Mercury and persistent chlorinated hydrocarbons in owls (*Strigiformes*) and birds of prey (*Falconiformes*) collected in Norway during the period 1965-1983. *Environmental Pollution* 11B: 91-108

GARCELON DK & THOMAS NJ (1997): DDE poisoning in an adult bald eagle. *Journal of Wildlife Diseases* 33: 299-303

GRIER JW (1982): Ban of DDT and subsequent recovery of reproduction in bald eagles. *Science* 218: 1232-1235

HAUFF P (1998): Population development of white-tailed eagle *Haliaeetus albicilla* in Germany since 1980 with a review of the past 100 years. *Vogelwelt* 119: 47-63

HEINISCH E & WENZEL-KLEIN S (1994): Produktion und Anwendung von Chlorkohlenwasserstoff-Pestiziden in der ehemaligen DDR. In: HEINISCH E, KETTRUP A, WENZEL-KLEIN S (Hrsg): *Schadstoffatlas Osteuropa: ökologisch-chemische und ökotoxikologische Fallstudien über organische Spurenstoffe in Ost-Mitteleuropa*. Ecomed Verlagsgesellschaft, Landsberg, Germany, pp 4-7

HELANDER B (1983): Reproduction of the white-tailed sea eagle *Haliaeetus albicilla* (L.) in Sweden, in relation to food and residue levels of organochlorine and mercury compounds in the eggs. Ph.D. thesis, Department of Zoology, Stockholm, Sweden, 27pp

HELANDER B, OLSSON A, REUTERGÅRDH L (1982): Residue levels of organochlorine and mercury compounds in unhatched eggs and the relationship to breeding success in white-tailed sea eagles in Sweden. *Holarctic Ecology* 5: 349-366

IWATA H, WATANABE M, KIM EY, GOTOH R, YASUNAGA G, TANABE S, MASUDA M, FUJITA S (2000): Contamination by chlorinated hydrocarbons and lead in Steller´s sea eagle and white-tailed sea eagle from Hokkaido, Japan. In: UETA M, MCGRADY M, FIRST J (Hrsg): *Symposium on Steller´s and White-tailed Sea Eagles in East Asia*, Wild Bird Society of Japan, Tokyo, Japan, pp 92-106

JENSEN S, JOHNELS AG, OLSSON M, OTTERLIND G (1969): DDT and PCB in marine animals from Swedish waters. *Nature* 224: 247-250

KAISER TE, REICHEL WL, LOCKE LN, CROMARTIE E, KRYNITSKY AJ, LAMONT TG, MULHERN BM, PROUTY RM, STAFFORD CJ, SWINEFORD DM (1980): Organochlorine pesticide, PCB, and PBB residues and necropsy data for bald eagles from 29 states - 1975-77. *Pesticides Monitoring Journal* 13: 145-149

KENNTNER N, TATARUCH F, KRONE O (2001): Heavy metals in soft tissue of white-tailed eagles found dead or moribund in Germany and Austria from 1993 to 2000. *Environmental Toxicology and Chemistry* 20: 1831-1837

KOEMAN JH, HADDERINGH RH, BIJEVELD MFIJ (1972): Persistent pollutants in the white-tailed eagle in the Federal Republic of Germany. *Biological Conservation* 4: 373-377

KOISTINEN J, KOIVUSAARI J, NUUJA I, VUORINEN PJ, PAASIVIRTA J, GIESY JP (1997): 2,3,7,8-tetrachlordibenzo-p-dioxin equivalent in extracts of Baltic white-tailed sea eagles. *Environmental Toxicology and Chemistry* 16: 1533-1544

KOIVUSAARI J, NUUJA I, PALOKANGAS R, FINNSLUND M (1980): Relationships between productivity, eggshell thickness and pollutant contents of addled eggs in the population of white-tailed eagles *Haliaëtus albicilla* L. in Finland during 1969 -1978. *Environmental Pollution* 23A: 41-52

KRONE O, LANGGEMACH T, SÖMMER P, KENNTNER N (in press) Causes of mortality in white-tailed eagles from Germany. *Proceedings*, Sea Eagle 2000, Swedish Society for Nature Conservation SNF, Project Sea Eagle, Björkö, Sweden, September 13-17, 2000

NYGÅRD T, AND SKAARE JU (1998): Organochlorines and mercury in eggs of white-tailed sea eagles *Haliaeetus albicilla* in Norway 1974-1994. In: Meyburg B-U, Chancellor RD Ferrero JJ (Hrsg): *Holarctic Birds of Prey*, World Working Group on Birds of Prey and Owls. Berlin, Germany, 501-524 pp

OEHME G (1980): Der Seeadler, *Haliaeetus albicilla* (L.), als biologischer Spitzenindikator für das Vorhandensein persistenter Biozide in der Biosphäre. In: SCHUBERT R, SCHUH J (Hrsg): *Bioindikation auf der Ebene der Individuen*, Wissenschaftliche Beiträge 1980/26 (P10), Martin-Luther-University, Halle-Wittenberg, pp 81-89

PAASIVIRTA J, TARHANEN J, JUVONEN B, VUORINEN P (1987): Dioxins and related aromatic chloroethers in Baltic wildlife. *Chemosphere* 16: 1787-1790

PEREG D, TAMPAL N, ESPANDIARI P, ROBERTSON LW (2001): Distribution and macromolecular binding of benzo[*a*]pyrene and two polychlorinated biphenyl congeners in female mice. *Chemico-Biological Interaction* 137: 243-258

STICKEL WH, STICKEL LF, DYRLAND RA, HUGHES DL (1984a): DDE in birds: Lethal residues and loss rates. *Archives of Environmental Contamination and Toxicology* 13: 1-6

STICKEL WH, STICKEL LF, DYRLAND RA, HUGHES DL (1984b): Arochlor 1254® in birds: Lethal levels and loss rates. *Archives of Environmental Contamination and Toxicology* 13: 7-13

TARHANEN J, KOISTINEN J, PAASIVIRTA J, VUORINEN PJ, KOIVUSAARI J, NUUJA I, KANNAN N, TATSUKAWA R (1989): Toxic significance of planar aromatic compounds in the Baltic ecosystem - new studies on extremely toxic coplanar PCBs. *Chemosphere* 18: 1067-1077

TATARUCH F, STEINECK T, FREY H (1998): Vergiftung durch Carbofuran bei Wildtieren (Greifvögeln, Singvögeln und Carnivoren) in Österreich. *Wiener Tierärztliche Monatsschrift* 85: 12-17

TUCKER GM & HEATH MF (1994): *Birds in Europe—Their Conservation Status*. BirdLife International, Cambridge, UK