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Persistent organic pollutants and methoxylated polybrominated diphenyl ethers in different tissues of white-tailed eagles (*Haliaeetus albicilla*) from West Greenland

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ABSTRACT

We investigated polychlorinated biphenyls (PCBs), organochlorine pesticides (e.g. dichlorodiphenyltrichloroethane (DDT)), polybrominated diphenyl ethers (PBDEs) and methoxylated PBDEs (MeO-PBDEs), in six matrices (muscle, liver, kidney, adipose, blood, preen oil) of 17 white-tailed eagles from West Greenland sampled between 1997 and 2009. High inter-individual variation in contamination was found (PCBs: 0.49–1500 μg/g lipid weight (lw), DDTs: 0.23–910 μg/g lw, PBDEs: 0.01–24 μg/g lw, MeO-PBDEs: 0.001–0.59 μg/g lw), mostly due to age-related differences and not to temporal trends. One adult female (age > 5 years) displayed PCB levels up to 1500 μg/g lw in liver, which is the highest concentration ever reported in Arctic wildlife. Muscle generally contained the highest median levels, while adipose tissue displayed the lowest median levels on a lipid basis. No significant differences were found among tissues for MeO-PBDEs. Remarkably, we found distinct correlations ($0.62 \leq r \leq 0.98$; $<0.0001 \leq p \leq 0.17$) between levels of MeO-PBDEs and PBDEs, suggesting similar bioaccumulation pathways of PBDEs and MeO-PBDEs in white-tailed eagles.

1. Introduction

Persistent Organic Pollutants (POPs) have been the subject of many studies, as these compounds accumulate in lipid-rich tissues and biomagnify through the food chain, reaching potentially toxic concentrations in top predators (Chen and Hale, 2010; Golden and Rattner, 2003; Jaspers et al., 2006). In addition, most POPs have been distributed world-wide due to long-range transport (Simonich and Hites, 1995). They have even been able to reach the remote Arctic areas via air and water currents (e.g. Braune et al., 2005; Carlsson et al., 2012). Although legacy POPs, e.g. polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), have been banned, they still persist in the environment (e.g. Fängström et al., 2005; Letcher et al., 2010; Dietz et al., 2012a). Because of their high persistence and very slow degradation, continued monitoring is warranted to assess their levels in the environment. Emerging POPs, including certain brominated flame retardants (BFRs), are still largely employed and their environmental levels should be followed up closely. Polybrominated diphenyl ethers (PBDEs), an important class of BFRs, have recently been banned in Europe (Directive EEC, 2003; European Court of Justice, 2008), while in the USA some formulations are still produced and will be phased out by the end of 2012 (Hess, 2010). Recent studies have documented that BFRs such as ΣPBDE, BDE-100, BDE-153, and hexabromocyclododecane (HBCD) have been increasing over the last three decades in Greenland top predators like the polar bear (*Ursus maritimus*; Dietz et al., 2012b). However, some congeners like BDE-47and BDE-99 peaked between 2000 and 2004 and have now started to decline (Dietz et al., 2012b). Some studies have reported methoxylated PBDEs (MeO-PBDEs) in wildlife as well, but they are generally considered to be of natural origin and not metabolites of PBDEs (Malmvärn et al., 2008; Qiu et al., 2007; Rotander et al., 2012; Weijs et al., 2009).

Birds of prey have been used in many studies as biomonitoring species for local and regional studies of environmental contamination (Burger, 1993; Chen and Hale, 2010; Jaspers et al., 2011). They are ideal

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species to monitor POP pollution as they accumulate high concentrations at the top of the food chain (Chen and Hale, 2010). The white-tailed eagle (*Haliaeetus albicilla*, also referred to as white-tailed sea eagle, WTSE), is a large predatory bird from the northern part of Eurasia. It is a top predator in the aquatic food chain, where it mainly feeds on fish and marine bird species, but also carrion (Cramp and Simmons, 1983). The WTSE population in Greenland is distributed in the southwest with 150–170 breeding pairs in 1990 (Kampp and Wille, 1990) and has been expanding to Disko Bay during the last decade (D. Boertmann, pers. comm.). Yet, POPs contamination remains a significant threat today for humans and wildlife, including birds of prey, living in the Arctic regions (UNEP/AMAP, 2011). Furthermore, information from arctic birds of prey is limited compared to marine birds and high trophic marine species from more southern locations (Letcher et al., 2010).

Here, we investigate the accumulation of various POPs in six tissues of 17 WTSEs from Greenland sampled between 1997 and 2009. As sublethal exposure, metabolism and toxicokinetics of POPs include unique tissue-specific toxicities (Letcher et al., 2010), the information obtained on the tissue-specific accumulation of POPs in this study can be of interest for further risk assessment studies in Arctic wildlife. Furthermore, we also investigate the tissue-specific accumulation of MeO-PBDEs, since such information has not been available until now.

2. Materials and methods

White-tailed eagles ($n = 17$) found dead in West Greenland between March 1997 and January 2009, were analysed in this study (Table 1; Fig. 1). The birds were aged based on plumage characteristics as immature birds can be identified until the age of approximately 5 years (Glutz Von Blotzheim et al., 1971). Eight birds were juveniles, two birds in 2nd, one bird in 4th, one in 5th plumage and five birds were adults. The cause of death was either shooting, lead intoxication, trauma/shock, a broken wing or unknown (in 3 cases). Further information on the birds used in this study is summarized in Table 1. According to the legislation, dead birds that are not left in nature must be delivered to the Greenland Institute of Natural Resources, where they are stored at −18 °C. More details on the sampling area and procedures can be found in Krone et al. (2004). Muscle, liver and kidney were dissected and preen oil, blood and adipose tissue were collected when available in a sufficient amount for analysis. Feathers were sampled as well to investigate POPs variation among different feather types (Jaspers et al., 2011) and to study their suitability as biomonitoring matrices for perfluoroalkyl substances (Herzke et al., 2011). Samples were shipped with CITES permission from Greenland to Department of Bioscience Roskilde, (Aarhus University, Denmark and subsequently to the Toxicological Centre (University of Antwerp, Belgium) for POP analysis.

For the analysis of POPs, approximately 1 g of muscle, liver and kidney, 2.5 g of blood, 10–80 mg of preen oil, 90 mg of adipose tissue were weighed and spiked with internal standards (CB 143, BDE 77, BDE 128, and ε-HCH). Sample treatment and analysis were performed according to previously described methods (Covaci et al., 2008a,b; Roosens et al., 2008). Briefly, tissues were homogenised with anhydrous Na_2SO_4 and extracted with 100 ml hexane:acetone (3:1, v/v) in an automated hot Soxhlet extractor for 2 h. The lipid content was determined gravimetrically on an aliquot of the extract (1 h at 105 °C), while the rest of the extract was cleaned up on a column filled with 8 g acid silica and eluted with 15 ml hexane and 10 ml dichloromethane. The eluate was concentrated to 100 μl and transferred to an injection vial. PBDEs, MeO-PBDEs, HCHs and CHLs were analysed using an Agilent 6890-5973 GC–MS operated in electron capture negative ionization (ECNI) mode equipped with a 30 m × 0.25 mm × 0.25 μm DB-5 capillary column. Bromine isotope ions (m/z 79 and 81) and two most characteristic ions for HCHs and CHLs were acquired in selected ion monitoring (SIM) mode, with a dwell time of 50 ms. PCBs, DDTs and HCB were analysed on a GC/MS operated in electron ionization (EI) mode and equipped with a 25 m × 0.22 mm × 0.25 μm HT-8 capillary column (SGE, Zulte, Belgium). The mass spectrometer was used in SIM mode with the two most intense ions (typically from the molecular cluster) acquired for each homologue group or isomer.

All tissues were analysed for 38 PCB congeners (CB 18, 28, 31, 44, 47, 49, 52, 74, 87, 95, 99, 101, 105, 110, 118, 128, 132, 138, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 180, 183, 187, 194, 195, 196/203, 199, 205, 206 and 209), eight PBDE congeners (BDE 28, 47, 49, 99, 100, 153, 154 and 183), 2 methoxylated PBDEs (2′-MeO-BDE 68 and 6-MeO-BDE 47), dichlorodiphenyltrichloroethane (p,p'-DDT and o,p'-DDT) and their metabolites (p,p'-DDE, o,p'-DDE, p,p'-DDD and o,p'-DDD), hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs; α-, β- and γ-HCH), chlordanes (CHLs; cis-nonachlor (CN), trans-nonachlor (TN) and oxychlordane (OxC)). Concentrations are expressed on a lipid weight (lw) basis.

All individual standards for PCBs and OCPs were obtained from Dr. Ehrenstorfer Laboratories (Augsburg, Germany), while PBDE standards were obtained from Wellington Laboratories (Ontario, Canada) and solvents of pesticide grade were employed (Merck, Darmstadt, Germany). Procedural blanks and a certified reference material (SRM1945) were analysed for supporting quality control. More details about QA/QC are given in the SI.

For each analyte, the average blank value was subtracted from the measured value. The limit of quantification (LOQ) was fixed on 3 × SD of the procedural blanks. For pollutants not detectable in the blanks, the LOQ was calculated from a signal to noise ratio of 10. LOQs for the analysed pollutants varied from 1 to 4 ng/g lw. Data below LOQ of samples were assigned a value according to $f \times$ LOQ where f is the detection frequency or the proportion of samples > LOQ (Voorspoels et al., 2002). Compounds with ≥50% of the measurements < LOQ were not taken into account for statistical analysis. For PCBs, only CB 18, CB 44, CB 132 and CB 205 were <LOQ in more than half of the cases, while all PBDE congeners were above LOQ in more than 50% of the samples, except for BDE 183 (detected only in 28% of the cases). γ-HCH was only detected in 36% of the samples and was therefore not included in further statistics. LOQs for the different compounds and tissues on a wet weight basis (ww) are presented in Table SI-1 of the Supporting Information.

Statistical analyses were performed using XLSTAT (version 2011.2.02; Addinsoft™) and SAS 9.2 for Windows (SAS Institute Inc., Cary, NC, USA). To meet the

Table 1
Sample information of the white-tailed eagles (*Haliaeetus albicilla*) from South West Greenland (1997–2009) analysed in the present study (partly adopted from Jaspers et al., 2011). Weights between brackets are approximate values due to a high moisture content or decomposition of the carcass. JUV: juveniles + birds in 2nd plumage, AD: Adults + birds in 4th and 5th plumage. M: male, F: female, nd: not determined.

NERI ID#	Finding date	Finding place	Estimated age[a]	Plumage	Age class	Sex	Cause of death	Weight (g)	Condition
39640	1998	Nanortalik	6 months	Juvenile	JUV	M	Trauma	5100	Very good
39641	1998	Paamiut	4 months	Juvenile	JUV	M	Trauma	6010	Very good
39643	Unknown	Maniitsoq	+5 years	Adult	AD	F	Lead intoxication (Krone et al., 2004)	5130	Moderate
39644	1997	Maniitsoq	Almost 5 years	5th	AD	F	Trauma	(6340)	Moderate
39645	1998	Nuuk	5 months	Juvenile	JUV	M	Shot	5550	Very good
39646	1998	Kobberfjord	+5 years	Adult	AD	M	Trauma	5395	Very good
39647	1999	Paamiut	11 months	Juvenile	JUV	M	Trauma	(5280)	Very good
39648	1999	Paamiut	One year	Juvenile	JUV	M	Shock	3450	Bad
39649	1999	Unknown	3 years 4 months	4th	AD	F	Shot	(5970)	Very good
39650	1998	Nuuk	+5 years	Adult	AD	M	Unknown (infection)	5140	Very bad
39651	2000	Nuuk	1.5 years	2nd	JUV	F	Lead intoxication (Krone et al., 2004)	4680	Bad
39652	Unknown	Unknown	Max. 1 year	Juvenile	JUV	F	Unknown	nd	nd
39653	2001	Nuuk	Almost 2 years	2nd	JUV	F	Unknown	nd	nd
39654	2007	Nuuk	+5 years	Adult	AD	F	Probably shot	nd	nd
39655	1999	Narsaq	1 year	Juvenile	JUV	M	15 leadshots found in body	nd	nd
39656	2009	Paamiut	6 months	Juvenile	JUV	M	Broken wing	nd	nd
39657	2009	Godthåbsfjord	+5 years	Adult	AD	F		nd	nd

[a] From hatching and provided that chicks hatch in late May.

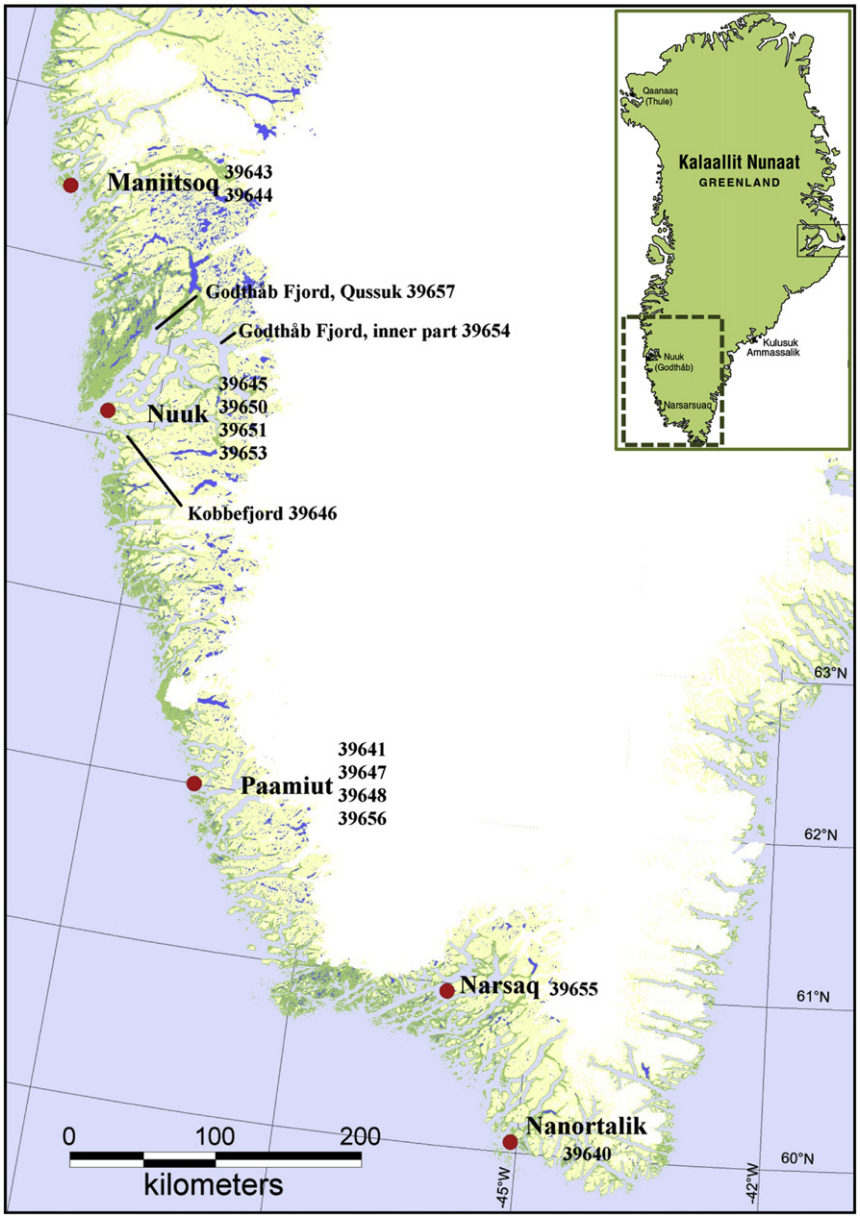

Fig. 1. Map of West Greenland with indication of sampling sites for white-tailed eagle carcasses from 1997 to 2009 analysed in the present study (specified with their ID #, adapted from Jaspers et al., 2011). Two birds with ID # 39649 and ID # 39652 could not be located on the map because the exact location was not known.

requirement of normality, all POP data were log-transformed according to $Y = \log10 (X + 1)$. Multi way repeated measures mixed models were calculated to investigate significant differences in concentrations of POPs between tissues, taking into account age and sex of the bird, sampling year and their interactions. Post hoc comparisons employed the Tukey correction for repeated measures. To include the factor "age" in the statistical analyses, juveniles and birds in 2nd plumage were grouped (JUV, $n = 10$), while birds in 4th and 5th plumage were grouped with the adult birds (AD, $n = 7$; Table 1). One adult female WTSE (NERI ID# 39654 or "GR54") had extremely high tissue concentrations (up to 1500 μg/g lw in liver) and was considered an outlier compared to the other birds. Therefore this individual was excluded from further statistical analyses. Profiles were constructed using the relative contribution of a compound class to the total sum of POPs or the relative contribution of specific congeners to the sum of PCBs and sum of PBDEs. Profiles were investigated by Principal Component Analysis (PCA) to reduce the complexity of the data into fewer independent factors. As such, the underlying relationships of the pollutants between tissues are clarified (Hobbs et al., 2002). To eliminate the effect of each individual pollutant, concentration data were first normalised by subtracting the mean of the original data and dividing by the standard deviation (Echols et al., 2000). Distance biplots were generated for the first two principal components and both factor loadings (red dots), representing the correlation of the congeners with the components, as factor scores (blue diamonds) were plotted. Differences between tissues were tested by performing non-parametrical Kruskal–Wallis tests with Bonferroni corrected significance level on calculated factor scores of the PCA. Pearson correlations were performed on the log transformed data. Correlations could not be investigated between preen oil and blood, because samples from both tissues were only available for three individuals. Furthermore, only data from three adipose tissues were available, so these were not included in the correlation analyses either. The level of significance was set at $\alpha = 0.05$ throughout this study.

3. Results and discussion

3.1. POP levels

Table 2 presents concentrations on a lipid weight basis. Concentrations on a wet weight basis and according to age (juveniles versus adults) can be found in Table SI-2 and Table Si-3 of the Supporting Information.

Table 2
Median concentrations and ranges (**μg/g** lipid weight) of POPs in the tissues of Greenland WTSE.

	Muscle	Preen oil	Liver	Kidney	Blood	Adipose tissue
n	17	13	6	6	5	3
Lipid %	1.5–11	16–96	3.4–6.2	3.0–6.8	0.2–1.5	94–96
Sum PCBs	36 (1.5–930)	14 (0.83–170)	11 (0.62–1500)	9.1 (0.49–1000)	6.9 (1.4–96)	1.8 (1.8–1.9)
Sum DDTs	18 (0.7–530)	12 (0.44–160)	7.2 (0.33–910)	6.3 (0.23–630)	5.1 (0.6–53)	1.1 (0.84–1.2)
Sum CHLs	7.8 (0.36–160)	5.2 (0.38–77)	3 (0.20–210)	2.6 (0.13–190)	2.1 (0.33–18)	0.52 (0.41–0.53)
HCB	1.2 (0.11–10)	1.1 (0.14–6)	0.78 (0.08–12)	0.74 (0.06–13)	0.88 (0.15–1.5)	0.17 (0.12–0.20)
Sum HCHs	0.26 (0.02–3.7)	0.23 (0.02–4.3)	0.22 (0.02–8.6)	0.16 (0.01–6.1)	0.23 (0.03–0.62)	0.05 (0.03–0.05)
Sum PBDEs	0.42 (0.03–15)	0.19 (0.02–3.2)	0.18 (0.01–24)	0.15 (0.01–18)	0.15 (0.04–1.7)	0.03 (0.03–0.04)

Sum PCBs includes 34 congeners (CB 28, 31, 47, 49, 52, 74, 87, 95, 99, 101, 105, 110, 118, 128, 138, 146, 149, 151, 153, 156, 170, 171, 172, 174, 177, 180, 183, 187, 194, 195, 196/203, 199, 206 and 209), Sum DDTs includes *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT, Sum CHLs includes OxC, TN and CN, Sum HCHs includes α- and β-HCH, Sum PBDEs includes 7 congeners (BDE 28, 47, 49, 99, 100, 153 and 154).

Legacy POPs were found in high levels in all tissues (Table 2), except in adipose tissue (on a lipid weight basis). High variation in contamination loads was found among individuals with more than a 100 fold difference in concentrations. No significant differences in concentrations were found between male and female birds ($F_{1,315} = 1.48$, $p = 0.22$), but significant differences were found between juveniles and adults ($F_{1,315} = 120.12$; $p < 0.0001$), according to collection year ($F_{6,315} = 37.72$, $p < 0.0001$) and a significant interaction between age and year ($F_{2,314} = 50.36$, $p < 0.0001$) was likewise found. Further visual inspection of the data revealed that the high variability in levels was probably due to age differences of the birds and not to temporal trends of POPs (see Figure SI-1 in the Supplementary Information). Indeed, extremely high levels were quantified in individual GR54, which was an adult female bird sampled in August 2007. Concentrations of sum PCBs were found up to 1500 μg/g lw in the liver of that bird, which is the highest level ever reported in Arctic wildlife to our knowledge. A high age (>5 years) may be an important cause of the high POPs concentrations found in this bird. This individual could not be sampled for blood and adipose tissue, which could explain lower maximum levels in blood and adipose tissue compared to the other tissues (Table 2). After removing individual GR54 from the mixed model, the interaction between year and age stayed significant ($F_{2,283} = 56.06$, $p < 0.0001$), but post hoc tests revealed that for the adults only a significant difference was found between years 1999 and 2009 (adjusted *p*-value (adj *p*) < 0.0001), while concentrations in the juveniles differed significantly from year to year (adj $p < 0.0001$ in all cases, except for a non-significant difference between years 1999 and 2001; $p = 0.96$). Besides age, variation in POP levels could also be related to temporal variation or spatial variation, as some of the sampling locations are more than 500 km apart (Fig. 1).

Significant differences of overall POP levels between tissues ($F_{5,283} = 9.05$, $p < 0.0001$) were found for adipose tissue with blood, liver (adjusted *p*-value < 0.05 for both cases) and muscle (adj $p < 0.0001$), between muscle and kidney (adj $p < 0.001$) and between muscle and preen oil (adj $p < 0.0001$). The highest median levels of POPs were found in muscle, while adipose tissue displayed the lowest median levels on a lipid weight basis (but $n = 3$ only; Table 2). Median concentrations of sum PCBs decreased in the order muscle (36 μg/g lw) > preen oil (14 μg/g lw) > liver (11 μg/g lw) > kidney (9.1 μg/g lw) > blood (6.9 μg/g lw) > adipose tissue (1.8 μg/g lw). However, these differences were not found significant, which is likely due to the high inter-individual variation within the relatively low sample size. In comparison, median concentrations of sum PCBs in feathers from the same individuals were previously found to be several orders of magnitude lower and ranged from 20 to 420 ng/g dry weight (dw) in different feather types (Jaspers et al., 2011).

When we compare our results to data published on WTSEs of Eastern Germany from 1979 to 1998 (Kannan et al., 2003) similar ranges were found for PCB concentrations in liver (64–89,400 ng/g ww compared to 21–86,000 ng/g ww in the present study). However, the concentrations of *p,p′*-DDE were one order of magnitude higher in the German WTSEs (maximum 686,000 ng/g ww compared to 51,000 ng/g ww in the present study; Table SI-2: DDE presents 99% of sum DDTs). Excluding individual GR54 increased the geographical trend, making the Greenland WTSE concentrations up to two orders of magnitude lower for *p,p′*-DDE (11–3100 ng/g ww) and one order of magnitude for PCBs (21–5400 ng/g ww) compared to the German WTSEs. Probably the proximity of local DDT sources (e.g. Kronimus et al., 2006; Schwarzbauer et al., 2003) in Eastern Germany has been responsible for the higher concentrations found in that study, but the differences in the reported periods may also contribute as the POP loads were higher between 1979 and 1998 (Kannan et al., 2003) compared to 1997–2008 (present study). In comparison to WTSEs from Japan collected between 1986 and 1999 (Iwata et al., 2000; Sakamoto et al., 2002), maximum concentrations of both PCBs and *p,p′*-DDE were found lower in the Greenland WTSEs as well (excluding GR54). Still, levels were higher in WTSE GR54 from Greenland. Finally, PCB levels (3800 μg/g lw) reported in muscle for one Steller's sea eagle (*Haliaeetus pelagicus*) from Japan (Kunisue et al., 2008) were higher than the maximum levels reported in muscle for the Greenland WTSE (930 μg/g lw).

A few studies reported PCB and OCP levels in blood of WTSE nestlings (Olsson et al., 2000; Eulaers et al., 2011a, 2011b). Sum PCB concentrations in the blood of the juvenile WTSEs from Greenland (3.5–98 ng/g ww; Table SI-3a) are similar to blood plasma levels in nestling WTSEs from Northern Norway (Eulaers et al., 2011a), but were in the lower range of the whole blood levels found in the WTSE nestlings from Sweden (74–160 ng/g ww for nestlings more than 8 weeks old; Olsson et al., 2000). On the contrary, DDE, HCB and HCHs showed higher maximum concentrations in the Greenland WTSEs compared to the other studies. This might be due to differences in the atmospheric transport of these compounds to Greenland (UNEP/AMAP, 2011). For example, different OCP patterns have been found between polar bear (*Ursus maritimus*) populations spanning the Arctic and subarctic regions, with the Greenland population displaying the highest sum DDTs concentrations (Verreault et al., 2005a), indicating that wildlife in Greenland may be subject to a high exposure risk. When the blood levels in the juvenile WTSEs are compared to POPs concentrations found in blood of one adult WTSE from Greenland (Table SI 3-b), concentrations in the Swedish nestlings were always lower, which can be explained by the increased accumulation of POPs over time in adult birds.

PBDE levels were in general approximately 100 fold lower than levels of PCBs and DDTs. PBDEs could be quantified in all tissues (median 0.03–0.42 μg/g lw) with maximum concentrations found in the liver up to 24 μg/g lw (Table 2), but no significant differences were

found between tissues for concentrations of PBDEs. The PBDE levels reported in muscle for the Steller's sea eagle from Japan (Kunisue et al., 2008) were similar (11 μg/g lw) to the maximum level (15 μg/g lw) in the current study. In comparison to PBDE levels reported in various tissues from terrestrial birds of prey from Belgium (Voorspoels et al., 2006), concentrations in the WTSE tissues from Greenland were within the same range, with also similar maximum concentrations in liver as reported for the sparrowhawk (*Accipiter nisus*) liver (26 μg/g lw; Voorspoels et al., 2006).

Further, we can compare our tissue results to PBDEs reported in WTSE eggs from Sweden and peregrine falcon (*Falco peregrinus*) eggs from South Greenland. The concentrations in the Greenland WTSE tissues show lower median levels, but comparable maximum concentrations to the WTSE eggs from the Baltic (up to 8.4 μg/g lw; Nordlöf et al., 2010) and the peregrine falcon eggs from South Greenland (up to 13 μg/g lw; Vorkamp et al., 2005). Median PBDE concentrations in WTSE tissues were also comparable to median PBDE levels reported in yolk sac from kittiwakes (*Rissa tridactyla*) collected in Norway (150–446 ng/g lw; Murvoll et al., 2006). One study reported PBDE levels in plasma of young bald eagles (*Haliaeetus leucocephalus*), a closely related species from North America (McKinney et al., 2006). Although concentrations in that study were generally found higher than in the blood of the Greenland WTSEs (up to 31 ng/g ww in the highest contaminated site of Santa Catalina Island), similar concentrations were found at the reference site (ranging from <LOQ – 1.25 ng/g ww), which was geographically separated from large urban and industrial zones (McKinney et al., 2006).

3.2. POPs profiles

As shown in Fig. 2, PCBs have the highest contribution to the total sum of POPs (around 50%), followed by DDTs (30%), CHLs (15%) and HCB (5%). HCHs and PBDEs contribute only less than 5% to the total POPs load. The POPs profile is similar in all tissues, except for a slightly higher contribution of sum CHLs in preen oil. The PCB profile (Figure SI-2 in the Supplementary Information) in all tissues was dominated by CB 153 (around 30%), followed by CB 138 (14–16%), CB 180 (11–14%) and CB 118 (6–8%). A similar pattern was observed in the blood of nestling WTSEs from Sweden (Olsson et al., 2000) and Norway (Eulaers et al., 2011a, 2011b), with the highest levels for CB 153 and CB 138, followed by CB 180 and CB 118. Furthermore, CB 153 was the highest in all muscle samples from WTSEs and Steller's sea eagles (*Haliaeetus pelagicus*) from Japan, followed by CB 138, CB 118, and CB 180. In preen oil, we found a higher contribution of lower chlorinated congeners (e.g. CB 118: 10%) and a lower contribution of the higher congeners (e.g. CB 180: 10%) compared to the other tissues. This is consistent with findings of previous studies on preen oil (Jaspers et al., 2008; Yamashita et al., 2007).

The PBDE profile of the Greenland WTSEs was clearly dominated by BDE 47 in all tissues (45–70% of the total sum PBDEs), followed by BDE 99, BDE 100 (10–15%) and BDE 153 and BDE 154 (5–10%). BDE 28 and BDE 49 contributed less than 5% to the total sum of PBDEs (see Figure SI-3 in the Supplementary Information). In comparison, BDE 47 was also found to be the dominant congener in eggs from Swedish WTSEs (40–60%; Nordlöf et al., 2010), in plasma of North American West Coast Bald Eaglet (around 50%; McKinney et al., 2006) and in plasma, body feathers and preen oil of WTSE from Norway (40–70%; Eulaers et al., 2011a, 2011b). Furthermore, BDE 47 was the only congener that could be quantified in the feathers of the Greenland WTSEs (Jaspers et al., 2011). The current results are in line with the general dominance of BDE 47 in studies on fish-eating birds from Europe and North America (reviewed by Kunisue et al., 2008) and also agrees with the results found for fish-eating coastal birds from Japan (Kunisue et al., 2008). In contrast, the dominance of BDE 153 has been reported in open seabirds (e.g. albatrosses) and inland predatory birds, illustrating the importance of both food habits and habitat (Kunisue et al., 2008).

No significant differences were found in the factor scores of the PCA for the PCBs (KW = 7.97 $p = 0.16$ for factor 1; KW = 11.08, $p = 0.0498$ for factor 2) and PBDEs (KW = 9.22, $p = 0.10$ for factor 1; KW = 6.58, $p = 0.25$ for factor 2), only for factor 2 of the PCBs between preen oil and muscle ($p = 0.011$). This is illustrated in Fig. 3, with lower chlorinated (tri- to penta-) PCBs clustering with preen oil and higher chlorinated (hexa- to nona-) congeners to muscle. Although adipose tissue is also clustering with the lower chlorinated PCBs on Fig. 3, this was not found significant, probably due to the low sample size ($n = 3$). However, as shown in Fig. 4, the PBDE profile in adipose tissue is deviating to some degree from the other tissues, with a higher contribution of lower brominated congeners. Furthermore, multiple pairwise comparisons using the Dunn's procedure revealed significant differences between concentrations in adipose tissue and concentrations in muscle ($p = 0.01$), blood ($p = 0.04$) and kidney ($p = 0.02$), but not after Bonferroni correction. Taking the small sample size ($n = 3$) for adipose tissue into account, this may be an indication for a significant difference with a larger sample size. Overall, we can conclude that the accumulation of POPs in the different tissues occurs in

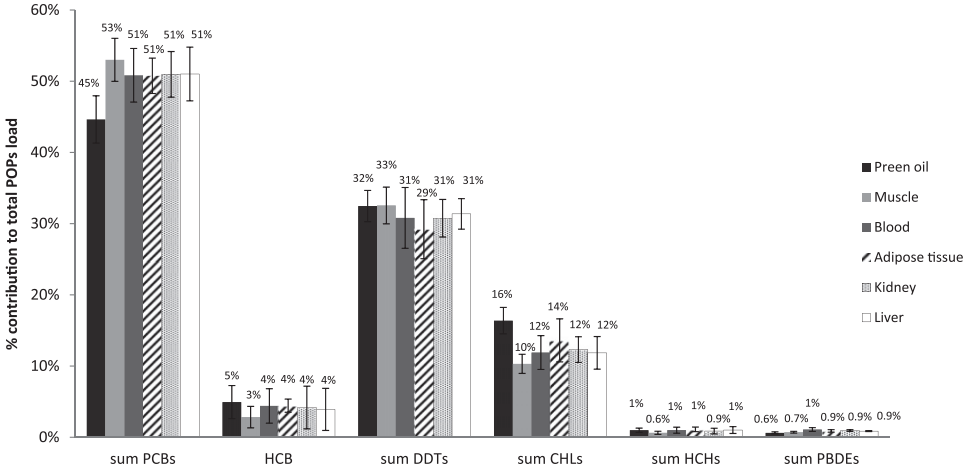

Fig. 2. POPs profile (mean % ± 2SE) in tissues of white-tailed eagles from Greenland 1997–2009.

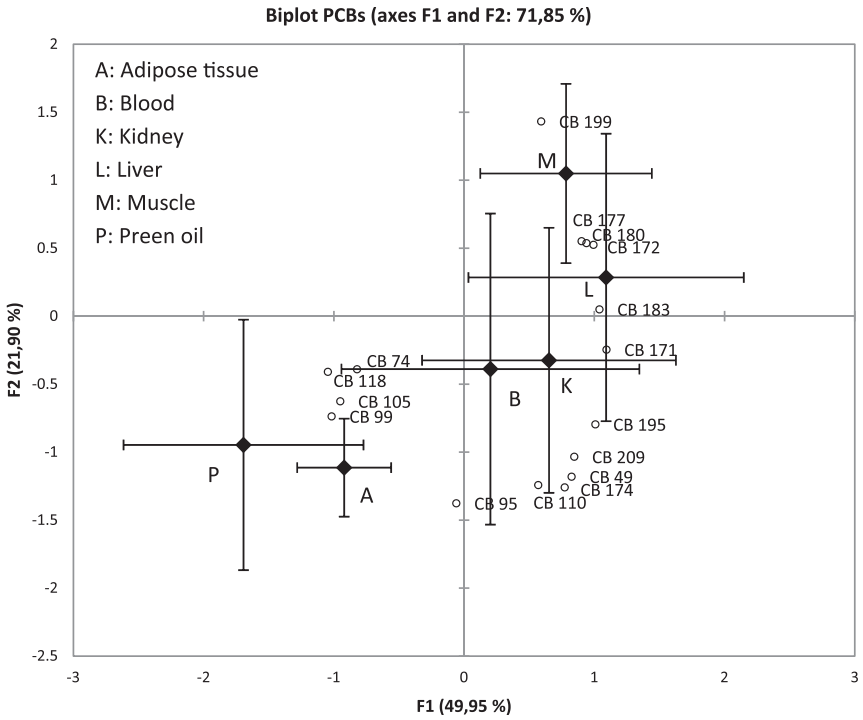

Fig. 3. PCA biplot of the PCB profile in tissues of white-tailed eagles from Greenland 1997–2009. Abbreviations used: M: Muscle, L: Liver, K: Kidney, A: Adipose tissue, P: Preen gland, B: Blood.

a similar way, although there may be slight differences for adipose tissue and preen oil that seem to accumulate more lower halogenated congeners. This is reflected in the PCB profile by a 2–5% higher contribution of CB 74, CB 99 and CB 118 in preen oil (and to some extent in adipose tissue) and in the PBDE profile by a 5–10% higher contribution in BDE 47 in adipose tissue (see Figures SI-2 and SI-3 in the Supplementary Information).

3.3. MeO-PBDEs

2′-MeO-BDE 68 and 6-MeO-BDE 47 could be quantified in all tissues (Table 3). For 2′-MeO-BDE 68, no significant differences were found between the tissues ($F_{5,22} = 1.85$, $p = 0.14$), but significant differences were found according to age ($F_{1,7} = 35.5$, $p < 0.001$) and year ($F_{6,7} = 7.44$, $p < 0.01$). Significantly higher

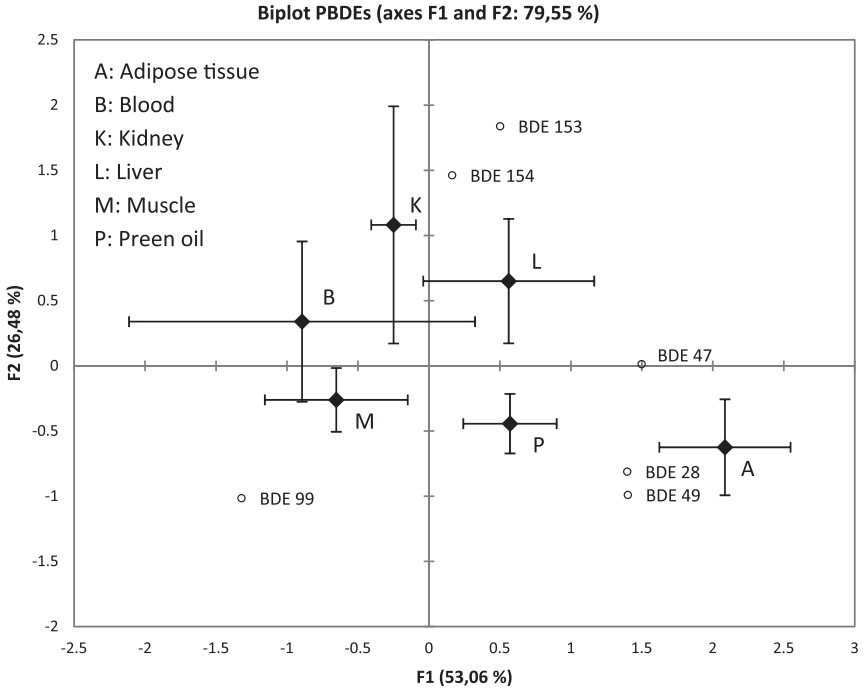

Fig. 4. PCA biplot of the PBDE profile in tissues of white-tailed eagles from Greenland 1997–2009. Abbreviations used: M: Muscle, L: Liver, K: Kidney, A: Adipose tissue, P: Preen gland, B: Blood.

Table 3
Median concentrations and ranges (**ng/g** lipid weight) of MeO-PBDEs in the tissues of Greenland WTSE. Levels of BDE 47 are given for comparison.

	Muscle	Preen oil	Liver	Kidney	Blood	Adipose tissue
n	17	13	6	6	5	3
Lipid %	1.5–11	16–96	3.4–6.2	3.0–6.8	0.2–1.5	94–96
2′-MeO-BDE 68	18 (1.8–340)	7.0 (1.5–91)	8.7 (1.0–590)	8.0 (<0.96–410)	6.5 (1.4–67)	2.2 (2.2–4.1)
6-MeO-BDE 47	23 (3.8–170)	9.3 (2.4–66)	20 (3.1–260)	18 (2.3–210)	15 (5.5–58)	6.6 (5.8–8.4)
BDE 47	213 (16–8700)	130 (8.4–2300)	98 (5.7–13000)	87 (3.7–10000)	89 (17–990)	22 (19–22)

concentrations were found in adults compared to juveniles (adj $p < 0.05$). Significant differences between years could be attributed to the levels in 1998 and 2009 being significantly lower than the levels in 2000 and 2007 (adj $p < 0.05$). For 6-MeO-BDE 47, no significant differences were found between the tissues ($F_{5,22} = 1.58$, $p = 0.21$), but a significant difference for age ($F_{1,7} = 10.19$, $p = 0.02$) and year ($F_{6,7} = 5.52$, $p = 0.02$) was observed. Post hoc tests revealed that adults had significantly higher concentrations than juveniles (adj $p < 0.05$) and a significantly higher concentration was observed in 1998 in comparison to 2000 (adj $p < 0.05$). All other years showed no significant differences (adj $p > 0.05$).

The present study is the first to report on levels of MeO-PBDEs in bird tissues. Therefore we can only compare our results with levels found for eggs and blood. In comparison to levels found in eggs from Swedish WTSEs (Nordlöf et al., 2010), concentrations of MeO-PBDEs in the Greenland WTSE are in the lower range, more similar to the inland than to the coastal samples from Sweden. The authors from that study pointed to the presence of MeO-PBDEs in the inland samples as from an as-yet unidentified source for MeO-PBDEs in the freshwater ecosystem (Nordlöf et al., 2010). 6-MeO-BDE 47 (2′-MeO-teBDE) was also quantified in blood of nestling WTSEs from the Swedish Baltic Coast (Olsson et al., 2000) and the authors found this compound at higher concentration, at least one order of magnitude higher, than the results for the Greenland WTSEs, but similar to the levels found in the WTSE eggs from the Baltic coast (Nordlöf et al., 2010). In comparison to other bird species, levels in blood of the Greenland WTSEs from the current study were comparable to concentrations of MeO-PBDEs found in blood plasma of glaucous gulls (*Larus hyperboreus*) from Svalbard in the Norwegian Arctic (Verreault et al., 2005b). However, 6-MeO-BDE 47 concentrations in blood from the Greenland WTSEs were one order of magnitude higher than concentrations measured in the blood serum of 8 bird species from South China (<LOD − 2.5 ng/g lw; Liu et al., 2010). This is probably due to the higher trophic position of the WTSE compared to the birds in the study of Liu et al. (2010), which were mostly feeding on plant material, seeds and insects. 2′-MeO-BDE 68 was not reported in the studies of Verreault et al. (2005b) and Liu et al. (2010) and can thus not be compared to our results. In contrast to the results for the WTSEs, no MeO-PBDEs could be detected in the plasma of North American bald eagles (McKinney et al., 2006), suggesting potential species differences in exposure and metabolism.

3.4. Correlations of POPs and MeO-PBDEs

We investigated the correlations between the levels of POPs in the different tissues, excluding the three adipose tissue samples. The results of the correlation analyses showed that sum PCBs concentrations were significantly correlated between all tissues ($0.97 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.013$). The same pattern was found for HCB ($0.96 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.008$) and sum PBDEs ($0.98 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.012$). For 6-MeO-BDE 47 and 2′-MeO-BDE 68 high correlation coefficients ($0.94 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.17$) were found between all tissues, although they were not significant for preen oil (p-value around 0.10). For sum DDTs ($0.97 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.033$), sum CHLs ($0.98 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.006$) and sum HCHs ($0.98 \leq r \leq 0.99$; $0.0001 \leq p \leq 0.042$) significant correlations were found between all tissues, again when excluding the three adipose tissue samples. This is also in accordance with the slightly deviating profiles that were found for adipose tissue (see supra). However, the low sample size for adipose tissue prevented definite conclusions.

Correlations among levels of PBDEs and MeO-PBDEs in the same tissue were also investigated (excluding adipose tissue). All PBDE congeners were strongly inter-correlated in the tissues we analysed ($0.82 \leq r \leq 0.99$; $<0.0001 \leq p \leq 0.052$), which could be expected on the basis of their common origin through the diet. The correlations of 6-MeO-BDE 47 and 2′-MeO-BDE 68 between the different PBDE congeners, were likewise found very strong and significant ($0.62 \leq r \leq 0.98$; $<0.0001 \leq p \leq 0.17$; Table 4; Fig. 5), which was surprising given the anticipated natural origin of the compounds. Therefore, the significant correlations found between PBDEs and MeO-PBDEs, explaining 63–90% of the variation in muscle (based on the r^2 value calculated from Table 4), suggest similar bioaccumulation pathways of PBDEs and MeO-PBDEs in WTSEs. In comparison, Verreault et al. (2005b) also found a positive relationship between 6-MeO-BDE 47 and BDE 47 in the glaucous gull, explaining 54% of the variation, and explained this by transfer of 6-MeO-BDE 47 through the food chain along with PBDEs. Wan et al. (2010) recently reported formation of MeO-PBDEs in fish through metabolism. This metabolic capacity of the fish may potentially explain the transfer of MeO-PBDEs up the food chain with the WTSE as the top predator. It might also explain the presence of MeO-PBDEs in the freshwater ecosystem as reported in the study of Nordlöf et al. (2010). This is the first study to suggest similarities in bioaccumulation of PBDEs and MeO-PBDEs and further research should be performed to evaluate this hypothesis and to identify the potential sources of MeO-PBDEs.

3.5. Risk assessment

The POP levels found in the Greenland WTSEs were lower than reported in previous studies on sea eagle species (*Haliaeetus* sp.) from contaminated areas around the world and similar to concentrations found in low contaminated areas. Olsson et al. (2000) found a negative impact of PCBs and DDE on the mean brood size in WTSEs from Sweden (only significant for CB 118), at mean PCB concentrations generally two-fold higher than the ones found in the present study. However, no effect was found on the average reproductive outcome and these levels were concluded not to pose a serious threat to the population of Swedish WTSEs. Likewise, median levels in the present study were below the PCB and DDE thresholds reported by Helander et al. (2002) for depressed productivity in WTSEs. Hence, the current levels of PCBs and OCPs in the Greenland WTSE are likely not of concern at the population level, although subclinical and developmental effects may still pose a potential threat at low concentrations (Grandjean and Landrigan, 2006). However, some birds may be at risk, as the maximum

Table 4
Results of the Pearson correlations (correlation coefficients) between PBDEs and 2′-MeO-BDE 68 and 6-MeO-BDE 47. Significant correlations are given in bold.

		Muscle	Preen oil	Liver	Kidney	Blood
n		16	12	5	5	5
2′-MeO-BDE 68	BDE 28	**0.93******	**0.92******	**0.95***	**0.94***	**0.93***
	BDE 49	**0.95******	**0.89*****	**0.97****	**0.95***	**0.97****
	BDE 47	**0.93******	**0.88*****	**0.97****	**0.96****	**0.95***
	BDE 100	**0.93******	**0.83*****	**0.97****	**0.98****	**0.95***
	BDE 99	**0.91******	**0.86*****	**0.97****	**0.97****	0.87^{T}
	BDE 154	**0.91******	**0.81*****	**0.94***	**0.94***	0.84^{T}
	BDE 153	**0.85******	**0.82*****	**0.94***	**0.95***	**0.91***
6-MeO-BDE 47	BDE 28	**0.84******	**0.73****	0.85^{T}	0.83^{T}	0.84^{T}
	BDE 49	**0.86******	**0.69***	**0.89***	0.86[a]	**0.90***
	BDE 47	**0.83******	**0.70***	**0.95***	**0.94***	**0.89***
	BDE 100	**0.82*****	**0.62***	**0.96***	**0.94***	**0.89***
	BDE 99	**0.80*****	**0.69***	**0.96***	**0.94***	0.78
	BDE 154	**0.82*****	**0.65***	0.88[a]	0.85[a]	0.72
	BDE 153	**0.81*****	**0.62***	**0.89***	0.87[a]	0.83[a]

[a] $p < 0.1$, $^{*}p < 0.05$, $^{**}p < 0.01$, $^{***}p < 0.001$, $^{****}p < 0.0001$ (p-values were not Bonferoni corrected because of low sample sizes).

concentrations found in this study were far above the reported Lowest Observed Effect Levels (LOELs) in Swedish WTSEs (500 μg/g lw for PCBs and 120 μg/g lw for DDE; Helander et al., 2002). The PCBs concentration found in the liver of GR54 (1500 μg/g lw) was even three times higher than the LOEL reported for PCBs in WTSE eggs that was associated with depressed productivity (Helander et al., 2002).

For PBDEs, Nordlöf et al. (2010) found no effect on the productivity of the WTSEs from Sweden, with concentrations in eggs similar or higher than the ones we found in the tissues of the Greenland WTSEs. Generally, PBDE levels in tissues of the Greenland WTSEs were also one to two orders of magnitude lower than the LOEL of 1000 ng/g ww in osprey eggs associated with impaired reproduction (Henny et al., 2009). However, this threshold was exceeded by the maximum values in liver (1400 ng/g ww) and preen oil (2200 ng/g ww; Table SI-2) found in the adult female GR54 from Greenland, suggesting that individual birds may be at risk from high PBDE contamination.

Overall, conventional POPs seem to be declining in Arctic regions (Rigét et al., 2010; Dietz et al., 2012a,b) and levels of POPs in the Greenland WTSE do not seem to have posed serious risks on the population in the last two decades. However, subclinical effects and synergism with other potential threats, e.g. heavy metal exposure and climate change driven food web and pathogen changes (Noyes et al., 2009; Sagerup et al., 2009) may be of concern. In particular, mercury has been documented to increase in Greenland WTSEs and other high trophic Arctic wildlife during the 20thcentury (Dietz et al., 2006, 2009, 2013). Also ΣPBDE, BDE-100, BDE-153, and HBCD have been increasing over the last three decades in East Greenland polar bears, whereas congeners like BDE-47and BDE-99 have recently started to decline (Dietz et al., 2012b). Furthermore, it may be possible that MeO-PBDEs in WTSEs are not only from natural origin and may be metabolic products from PBDEs produced in fish and subsequently ingested through the diet. However, this possibility should be further investigated in future studies. Since generally concentrations of MeO-PBDEs in fish are found higher than in the birds that prey on them (Zhang et al., 2010), further metabolism and transformation of MeO-PBDEs may take place in WTSEs and other bird species. Therefore, future risk assessment studies should take the potential metabolic origin and transformation of MeO-PBDEs into account.

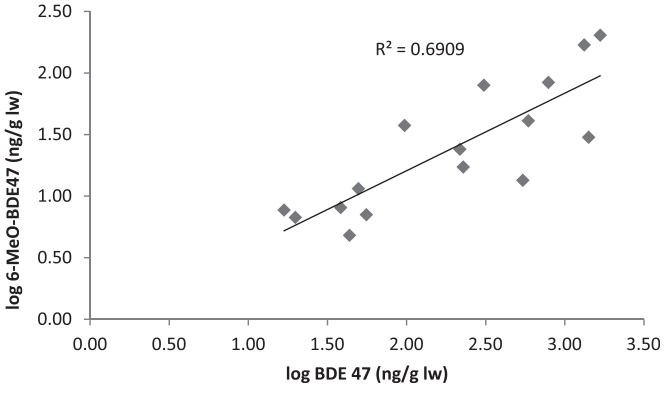

Fig. 5. Correlation plot between BDE 47 and 6-MeO-BDE 47 (log-transformed lw concentrations) in muscle tissue of white-tailed eagles from Greenland 1997–2009.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2012.12.023.

References

Braune, B.M., Outridge, P.M., Fisk, A.T., Muir, D.C.G., Helm, P.A., Hobbs, K., Hoekstra, P.F., Kuzyk, Z.A., Kwan, M., Letcher, R.J., Lockhart, W.L., Norstrom, R.J., Stern, G.A., Stirling, I., 2005. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: an overview of spatial and temporal trends. Science of the Total Environment 351, 4–56.

Burger, J., 1993. Metals in avian feathers: bioindicators of environmental pollution. Reviews in Environmental Toxicology 5, 203–311.

Carlsson, P., Cornelissen, G., Boggild, C.E., Rysgaard, S., Mortensen, J., Kallenborn, R., 2012. Hydrology–linked spatial distribution of pesticides in a fjord system in Greenland. Journal of Environmental Monitoring 14 (5), 1437–1443.

Chen, D., Hale, R.C., 2010. A global review of polybrominated diphenyl ether flame retardant contamination in birds. Environment International 36, 800–811.

Cramp, S., Simmons, K.E.L., 1983. Handbook of the Birds of Europe the Middle East and North Africa. In: The Birds of the Western Palearctic, vol. III. Oxford University Press.

Covaci, A., Losada, S., Roosens, L., Vetter, W., Santos, F.J., Neels, H., Storelli, A., Storelli, M.M., 2008a. Anthropogenic and naturally-occurring organobrominated compounds in two deep-sea fish species from the Mediterranean Sea. Environmental Science & Technology 42, 8654–8660.

Covaci, A., Voorspoels, S., Roosens, L., Jacobs, W., Blust, R., Neels, H., 2008b. PBDEs in liver and adipose tissue samples from Belgium. Chemosphere 73, 170–175.

Dietz, R., Riget, F., Olsen, M.T., Boertmann, D., Sonne, C., Kirkegaard, M., Asmund, G., Falk, K., Fjeldsaa, J., Egevang, C., Wille, F., Møller, S., 2006. Time trend of mercury in feathers of West Greenland birds of prey during 1859–2003. Environmental Science & Technology 40, 5911–5916.

Dietz, R., Outridge, P.M., Hobson, K.A., 2009. Anthropogenic contribution to mercury levels in present–day Arctic animals – a review. Science of the Total Environment 407, 6120–6131.

Dietz, R., Rigét, F.F., Sonne, C., Born, E.W., Bechshøft, T., McKinney, M.A., Letcher, R.J., 2012a. Part 1: Three Decades (1984–2010) of Legacy Contaminant Trends in East Greenland Polar Bears (*Ursus maritimus*). Environment International. http://dx.doi.org/10.1016/j.envint.2012.09.004.

Dietz, R., Rigét, F.F., Sonne, C., Born, E.W., Bechshøft, T., McKinney, M.A., Drimmei, R., Muir, D.C.G., Letcher, R.J., 2012b. Part 2: Three Decades (1984–2010) of Flame Retardant Trends in East Greenland Polar Bears (*Ursus maritimus*). Environment International. http://dx.doi.org/10.1016/j.envint.2012.09.008.

Dietz, R., Basu, N., Braune, B., O'Hara, T., Scheuhammer, T.M., Sonne, C., Andersen, M., Andreasen, C., Andriashek, D., Asmund, G., Aubail, A., Baagøe, H., Born, E.W., Chan, H.M., Derocher, A.E., Grandjean, P., Knott, K., Kirkegaard, M., Lunn, N., Messier, F., Obbard, M., Olsen, M.T., Peacock, E., Renzoni, A., Rigét, F., Skaare, J.U., Stern, G., Stirling, I., Taylor, M., Wiig, Ø., Aars, J., 2013. What are the toxicological effects of mercury in Arctic biota? Science of the Total Environment 443, 775–790.

Directive 2003/11/EC of the European Parliament and of the Council of 6 February 2003 amending for the 24th time Council directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (pentabromodiphenyl ether and octabromodiphenyl ether). Official Journal L 042, 15/02/2003.

Echols, K.R., Gale, R.W., Schwartz, T.R., Huckins, J.N., Williams, L.L., Meadows, J.C., Morse, D., Petty, J.D., Orazio, C.E., Tillitt, D.E., 2000. Comparing polychlorinated biphenyl concentrations and patterns in the Saginaw River using sediment, caged fish, and semipermeable membrane devices. Environmental Science & Technology 34 (19), 4095–4102.

European Court of Justice. Cases C–14/06 and C–295/06, Judgement of the Court, 1 April 2008, Directive 2002/95/EC and Commission Decision 2005/717/EC, 2008.

Eulaers, I., Covaci, A., Herzke, D., Eens, M., Sonne, C., Moum, T., Schnug, L., Hanssen, S.A., Johnsen, T.V., Bustnes, J.O., Jaspers, V.L.B., 2011a. A first evaluation of the usefulness of feathers of nestling predatory birds for non-destructive biomonitoring of persistent organic pollutants. Environment International 37 (3), 622–630.

Eulaers, I., Covaci, A., Hofman, J., Nygård, T., Halley, D.J., Pinxten, R., Eens, M., Jaspers, V.L.B., 2011b. A comparison of non–destructive sampling strategies to assess the exposure of white-tailed eagle nestlings (*Haliaeetus albicilla*) to persistent organic pollutants. Science of the Total Environment 410 (1), 258–265.

Fängström, B., Athanasiadou, M., Athanassiadis, I., Bignert, A., Grandjean, P., Weihe, P., Bergman, A., 2005. Polybrominated diphenyl ethers and traditional organochlorine pollutants in fulmars (*Fulmarus glacialis*) from the Faroe Islands. Chemosphere 60, 836–843.

Glutz Von Blotzheim, U.N., Bauer, K.M., Bezzel, E., 1971. Handbuch der Vögel Mitteleuropas. In: Seeadler, Band 4. Akademische Verlagsgesellschaft, Frankfurt am Main, ISBN 3-400-00069-8. 169–203.

Golden, N.H., Rattner, B.A., 2003. Ranking terrestrial vertebrate species for utility in biomonitoring and vulnerability to environmental contaminants. Reviews in Environmental Contamination and Toxicology 176, 67–136.

Grandjean, P., Landrigan, P.J., 2006. Developmental neurotoxicity of industrial chemicals. Lancet 368 (9553), 2167–2178.

Helander, B., Olsson, A., Bignert, A., Asplund, L., Litzén, K., 2002. The role of DDE, PCB, coplanar PCB and eggshell parameters for reproduction in the white-tailed sea eagle (*Haliaeetus albicilla*) in Sweden. AMBIO: A Journal of the Human Environment 31 (5), 386–403.

Henny, C.J., Kaiser, J.L., Grove, R.A., Johnson, B.L., Letcher, R.J., 2009. Polybrominated diphenyl ether flame retardants in eggs may reduce reproductive success of ospreys in Oregon and Washington, USA. Ecotoxicology 18 (7), 802–813.

Herzke, D., Jaspers, V.L.B., Boertmann, D., Rasmussen, L.M., Sonne, C., Dietz, R., Covaci, A., Eens, M., Bustnes, J.O., 2011. PFCs in feathers of white-tailed eagles (*Haliaeetus albicilla*) from Greenland and Norway, useful for nondestructive sampling? Organohalogen Compounds 73, 1337–1339.

Hess, G., 2010. Industry drops flame retardant. Chemical & Engineering News 88, 10.

Hobbs, K.E., Lebeuf, M., Hammill, M.O., 2002. PCBs and OCPs in male harbour, grey, harp and hooded seals from the Estuary and Gulf of St Lawrence, Canada. Science of the Total Environment 296, 1–18.

Iwata, H., Watanabe, M., Kim, F.Y., Gotoh, R., Yasunaga, G., Tanabe, S., Masuda, Y., Fujita, S. Contamination by Chlorinated Hydrocarbons and Lead in Steller's Sea Eagle and White-tailed Sea Eagle from Hokkaido, Japan. First symposium on Steller's and white-tailed sea eagles in East Asia, 2000. pp. 91–106.

Jaspers, V.L.B., Covaci, A., Voorspoels, S., Dauwe, T., Eens, M., Schepens, P., 2006. Brominated flame retardants and organochlorine pollutants in aquatic and terrestrial predatory birds of Belgium: levels, patterns, tissue distribution and condition factors. Environmental Pollution 139, 340–352.

Jaspers, V.L.B., Covaci, A., Deleu, P., Neels, H., Eens, M., 2008. Preen oil as the main source of external contamination with organic pollutants onto feathers of the common magpie (*Pica pica*). Environment International 34, 741–748.

Jaspers, V.L.B., Soler–Rodriguez, F., Boertmann, D., Sonne, C., Dietz, R., Covaci, A., Eens, M., 2011. Body feathers as a potential new biomonitoring tool in raptors: a study on organohalogenated contaminants in different feather types and preen oil of West Greenland white-tailed eagles (*Haliaeetus albicilla*). Environment International 37, 1349–1356.

Kampp, K., Wille, F., 1990. The white-tailed eagle population in Greenland. Dansk Ornitologisk Forenings Tidsskrift 84, 37–44.

Kannan, K., Kumar, K.S., Nakata, H., Falandysz, J., Oehme, G., Masunaga, S., 2003. Polychlorinated biphenyls, dibenzo–p–dioxins, dibenzofurans, and p, p′–DDE in livers of white-tailed sea eagles from eastern Germany, 1979–1998. Environmental Science & Technology 37 (7), 1249–1255.

Krone, O., Wille, F., Kenntner, N., Boertmann, D., Tataruch, F., 2004. Mortality factors, environmental contaminants, and parasites of White-Tailed Sea Eagles from Greenland. Avian Diseases 48, 417–424.

Kronimus, A., Schwarzbauer, J., Ricking, M., 2006. Analysis of non–extractable DDT-related compounds in riverine sediments of the Teltow Canal, Berlin, by pyrolysis and thermochemolysis. Environmental Science & Technology 40 (19), 5882–5890.

Kunisue, T., Higaki, Y., Isobe, T., Takahashi, S., Subramanian, A., Tanabe, S., 2008. Spatial trends of polybrominated diphenyl ethers in avian species: utilization of stored samples in the Environmental Specimen Bank of Ehime University (es-Bank). Environmental Pollution 154 (2), 272–282.

Letcher, R.J., Bustnes, J.O., Dietz, R., Jenssen, B.M., Jørgensen, E.H., Sonne, C., Verreault, J., Vijayan, M.M., Gabrielsen, G.W., 2010. Effects assessment of persistent organohalogen contaminants in Arctic wildlife and fish. Science of the Total Environment 408, 2995–3043.

Liu, J.A., Luo, X.J., Yu, L.H., He, M.J., Chen, S.J., Mai, B.X., 2010. Polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyles (PCBs), hydroxylated and methoxylated-PBDEs, and methylsulfonyl-PCBs in bird serum from South China. Archives of Environmental Contamination and Toxicology 59 (3), 492–501.

Malmvärn, A., Zebuhr, Y., Kautsky, L., Bergman, A., Asplund, L., 2008. Hydroxylated and methoxylated polybrominated diphenyl ethers and polybrominated dibenzo-p-dioxins in red alga and cyanobacteria living in the Baltic Sea. Chemosphere 72 (6), 910–916.

McKinney, M.A., Cesh, L.S., Elliott, J.E., Williams, T.D., Garcelon, D.K., Letcher, R.J., 2006. Brominated flame retardants and halogenated phenolic compounds in North American west coast bald eaglet (*Haliaeetus leucocephalus*) plasma. Environmental Science & Technology 40 (20), 6275–6281.

Murvoll, K.M., Skaare, J.U., Moe, B., Anderssen, E., Jenssen, B.M., 2006. Spatial trends and associated biological responses of organochlorines and brominated flame retardants in hatchlings of North Atlantic kittiwakes (*Rissa tridactyla*). Environmental Toxicology and Chemistry 25 (6), 1648–1656.

Nordlöf, U., Helander, B., Bignert, A., Asplund, L., 2010. Levels of brominated flame retardants and methoxylated polybrominated diphenyl ethers in eggs of white-tailed sea eagles breeding in different regions of Sweden. Science of the Total Environment 409, 238–246.

Noyes, P.D., McElwee, M.K., Miller, H.D., Clark, B.W., Van Tiem, L.A., Walcott, K.C., Erwin, K.N., Levin, E.D., 2009. The toxicology of climate change: environmental contaminants in a warming world. Environment International 35 (6), 971–986.

Olsson, A., Ceder, K., Bergman, A.k., Helander, B., 2000. Nestling blood of the White-Tailed Sea Eagle (*Haliaeetus albicilla*) as an indicator of territorial exposure to organohalogen compounds – an evaluation. Environmental Science & Technology 34, 2733–2740.

Qiu, X., Mercado–Feliciano, M., Bigsby, R.M., Hites, R.A., 2007. Measurement of polybrominated diphenyl ethers and metabolites in mouse plasma after exposure to a commercial pentabromo diphenyl ether mixture. Environmental Health Perspectives 115 (7), 1052–1058.

Rigét, F., Bignert, A., Braune, B., Stow, J., Wilson, S., 2010. Temporal trends of legacy POPs in Arctic biota, an update. Science of the Total Environment 408, 2874–2884.

Roosens, L., Dirtu, A., Goemans, G., Belpaire, C., Gheorghe, A., Neels, H., Blust, R., Covaci, A., 2008. Brominated flame retardants and organochlorine contaminants in fish samples along Scheldt River, Belgium. Environment International 34, 976–983.

Rotander, A., van Bavel, B., Riget, F., Auounsson, G.A., Polder, A., Gabrielsen, G.W., Vikingsson, G., Mikkelsen, B., Dam, M., 2012. Methoxylated polybrominated diphenyl ethers (MeO-PBDEs) are major contributors to the persistent organobromine load in sub-Arctic and Arctic marine mammals, 1986–2009. Science of the Total Environment 416, 482–489.

Sagerup, K., Savinov, V., Savinova, T., Kuklin, V., Muir, D.C.G., 2009. Persistent organic pollutants, heavy metals and parasites in the glaucous gull (*Larus hyperboreus*) on Spitsbergen. Environmental Pollution 157, 2282–2290.

Sakamoto, K.Q., Kunisue, T., Watanabe, M., Masuda, Y., Iwata, H., Tanabe, S., Akahori, F., Ishizuka, M., Kazusaka, A., Fujita, S., 2002. Accumulation patterns of polychlorinated biphenyl congeners and organochlorine pesticides in Steller's sea eagles and white-tailed sea eagles, threatened species, in Hokkaido, Japan. Environmental Toxicology and Chemistry 21 (4), 842–847.

Simonich, S.L., Hites, R.A., 1995. Global distribution of persistent organochlorine compounds. Science 269, 1851–1854.

Schwarzbauer, J., Ricking, M., Littke, R., 2003. DDT–related compounds bound to the nonextractable particulate matter in sediments of the Teltow Canal, Germany. Environmental Science & Technology 37 (3), 488–495.

UNEP/AMAP, 2011. Climate Change and POPs: Predicting the Impacts. Report of the UNEP/AMAP Expert Group. Secretariat of the Stockholm Convention, Geneva, 62 pp.

Verreault, J., Muir, D.C.G., Norstrom, R.J., Stirling, I., Fisk, A.T., Gabrielsen, G.W., Derocher, A.E., Evans, T.J., Dietz, R., Sonne, C., Sandala, G.M., Gebbink, W., Riget, F.F., Born, E.W., Taylor, M.K., Nagy, J., Letcher, R.J., 2005a. Chlorinated hydrocarbon contaminants and metabolites in polar bears (*Ursus maritimus*) from Alaska, Canada, East Greenland, and Svalbard: 1996–2002. Science of the Total Environment 351, 369–390.

Verreault, J., Gabrielsen, G.W., Chu, S., Muir, D.C.G., Andersen, M., Hamaed, A., Letcher, R.J., 2005b. Flame retardants and methoxylated and hydroxylated polybrominated diphenyl ethers in two Norwegian Arctic top predators: glaucous gulls and polar bears. Environmental Science & Technology 39, 6021–6028.

Voorspoels, S., Covaci, A., Maervoet, J., Schepens, P., 2002. Relationship between age and levels of organochlorine contaminants in human serum of a Belgian population. Bulletin of Environmental Contamination and Toxicology 69, 22–29.

Voorspoels, S., Covaci, A., Lepom, P., Jaspers, V.L.B., Schepens, P., 2006. Levels and distribution of polybrominated diphenyl ethers in various tissues of birds of prey. Environmental Pollution 144, 218–227.

Vorkamp, K., Thomsen, M., Falk, K., Leslie, H., Møller, S. r., Sørensen, P.B., 2005. Temporal development of brominated flame retardants in peregrine falcon

(*Falco peregrinus*) eggs from South Greenland (1986–2003). Environmental Science and Technology 39 (21), 8199–8206.

Wan, Y., Liu, F., Wiseman, S., Zhang, X., Chang, H., Hecker, M., Jones, P.D., Lam, M.H., Giesy, J.P., 2010. Interconversion of hydroxylated and methoxylated polybrominated diphenyl ethers in Japanese medaka. Environmental Science & Technology 44 (22), 8729–8735.

Weijs, L., Losada, S., Das, K., Roosens, L., Reijnders, P.J.H., Santos, J.F., Neels, H., Blust, R., Covaci, A., 2009. Biomagnification of naturally-produced methoxylated polybrominated diphenyl ethers (MeO-PBDEs) in harbour seals and harbour porpoises from the Southern North Sea. Environment International 35 (6), 893–899.

Yamashita, R., Takada, H., Murakami, M., Fukuwaka, M.-A., Watanuk, i Y., 2007. Evaluation of non-invasive approach for monitoring PCB pollution of seabirds using preen gland oil. Environmental Science & Technology 41, 4901–4906.

Zhang, K., Wan, Y., An, L., Hu, J., 2010. Trophodynamics of polybrominated diphenyl ethers and methoxylated polybrominated diphenyl ethers in a marine food web. Environmental Toxicology and Chemistry 29 (12), 2792–2799.