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Three decades (1983–2010) of contaminant trends in East Greenland polar bears (*Ursus maritimus*). Part 1: Legacy organochlorine contaminants

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ABSTRACT

Legacy organochlorine contaminants were determined in adipose tissues from 294 polar bears (*Ursus maritimus*) sampled in East Greenland in 23 of the 28 years between 1983 and 2010. Of 19 major legacy contaminants and congeners (ΣPCB, 4 PCB congeners (CB153, 180, 170/190), ΣDDT, *p,p′*-DDE, *p,p′* -DDD and *p,p′*-DDT, α- and β-hexachlorocyclohexane (HCH), HCB, octachlorostyrene, dieldrin, oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, heptachlor epoxide and BB-153), 18 showed statistically significant average yearly declines of −4.4% (range: −2.0 to −10.8%/year) among subadult polar bears (i.e. females<5 years, males<6 years). For example, the ∑PCB concentrations declined 2.7 fold from 22730 ng/g lw (95% C.I.: 12470–32990 ng/g lw) in 1983–1986 to 8473 ng/g lw (95% C.I.: 6369–9776 ng/g lw) in 2006–2010. Similar but fewer statistically significant trends were found for adult females and adult males likely due to smaller sample size and years. Despite declines as a result of international regulations, relatively high levels of these historic pollutants persist in East Greenland polar bear tissues.

1. Introduction

The polar bear (*Ursus maritimus*) is an important species for assessing levels and trends of organohalogen contaminants (OHCs), including legacy organochlorines, within arctic marine ecosystems (Norstrom et al., 1998). As a consequence of the polar bear's trophic position at the top of the Arctic food web, it serves as an important sentinel species of long-range transported contaminants. Polar bears are reliant on a high fat (lipid) diet, mainly from their primary prey, ringed seal (*Phoca hispida*), and to a lesser extent from bearded seals (*Erignathus barbatus*) (Smith, 1980; Stirling and Archibald, 1977). Other prey such as harp seals (*Phoca groenlandica*), hooded seals (*Crystophora cristata*), white whales (*Delphinapterus leucas*), narwhal (*Monodon monoceros*), and walrus (*Odobenus rosmarus*) are sometimes taken (Derocher et al., 2004). As many OHCs are lipophilic, this is also a high contaminant diet (Letcher et al., 2010).

Reproductive performance and immunotoxicity are considered among the most important possible negative population-level impacts of OHC and mercury exposure in polar bears (Letcher et al., 2010; Sonne, 2010). Impacts from OHCs on reproductive and immunological systems have been suggested for polar bears at Svalbard (Derocher et al., 2003; Lie et al., 2004, 2005) and East Greenland (Letcher et al., 2010; Sonne, 2010; Sonne et al., 2006a). These health effects have been proposed to be mediated via disruptions of hormone and vitamin pathways (Bechshøft et al., 2012; Letcher et al., 2010; Sonne, 2010; Villanger et al., 2011). Also, lesions in internal organs such as liver, kidney, and thyroid glands, have been suggested to be associated with high OHC concentrations (Sonne, 2010; Sonne et al., 2005, 2006b, 2007, 2008, 2011). Controlled studies on other Arctic carnivores (i.e. Greenland sledge dogs, *Canis familiaris*, and farmed Arctic foxes, *Vulpes/Alopex lagopus*) have been conducted (Letcher et al., 2010; Sonne, 2010). The results from these studies support the hypothesis of a cause–effect relationship between the high OHCs levels measured in polar bears from East Greenland and Svalbard and the observed impairment of endocrine and immune systems in the same animals.

Given the growing body of evidence of associations between OHC exposure and biomarkers showing deleterious effects, it is important to determine how OHC concentrations in the polar bears have fluctuated over time. Global and regional conventions have been developed with the goal of reducing or even eliminating OHC emissions. Banned or regulated OHCs are referred to as 'legacy' OHCs, as present exposure is largely a 'legacy' of past releases (AMAP, 1998; Rigét et al., 2010). Despite restrictions in the use of many legacy OHCs, determining the levels of these persistent compounds in nature and their potential influence on the health of the polar bears still remains of high importance.

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Temporal trends have previously been reported in 316 Arctic OHC data series, in addition to a summary of the literature from an additional 162 time series (Rigét et al., 2010). However, these time series were generally too short to provide a clear picture of the temporal trends, generally ranging from 6 to 16 years in length/duration with a maximum 19 years.

Here, we examine longer-term and retrospective temporal trends in the adipose tissue concentrations of nineteen legacy contaminant classes and major congeners (PCB, CB153, 180 and 170/190, ΣDDT, *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT, α- and β-HCH, HCB, octachlorostyrene, dieldrin, oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor and heptachlor epoxide; Table 1) in 294 East Greenland polar bears sampled between 1983 and 2010. A parallel study is presented in this issue on brominated flame retardants (BFRs) from the same group of polar bears from East Greenland (Dietz et al., 2012). The present study together with the study by Dietz et al. (2008, 2011, 2012) represents the longest Arctic time series for contaminants to date, with 23 years of data sampled over a duration of 28 years.

2. Materials and methods

2.1. Sample details

A total of 294 polar bears were sampled from 1983 to 2010 in the Ittoqqortoormiit/Scoresby Sound region of East Greenland between ca. 69° to ca. 74° N (Table S1). Adipose samples were collected during native subsistence hunts. After sampling and during shipment, samples were kept frozen. At the Department of Bioscience Specimen Bank, samples were stored at −20 °C until further processing.

2.2. Age determination

Ages were determined by counting annual growth layer groups (GLGs) in the cementum of the lower right I3 using established methods (e.g. Dietz et al., 2004). Age classifications were as follows: adult males ≥ 6 years of age, adult females ≥ 5 years, and subadults consisted of all other bears (Rosing-Asvid et al., 2002). Subadults of both sexes were pooled for statistical analyses of OHC levels (Dietz et al., 2004).

2.3. Organohalogen contaminant analysis

Samples (n = 204) collected in 1983–1996, 2003, 2004, 2006, 2007, 2008, 2009, and 2010 were analysed for PCBs and OCs based on methods previously described by Letcher et al. (2009) and McKinney et al. (2009, 2010, 2011). Samples (n = 90) collected in 1999, 2000, and 2001 were analysed for PCBs and OCs as described in Verreault et al. (2005). Details on preparation, chemical analysis, quality assurance and control are provided in the supplemental information.

2.4. Data analysis

For detailed data presentation, 19 of the most important (highest concentration) legacy contaminants and their major congeners were selected among 47 PCBs and 20 OCPs (Table 1). Leaving out the more minor contaminants was justified by the observed significant auto correlation between most of the analysed groups of contaminants. The consecutive temporal trend analyses of these OHCs followed the ICES (International Council for the Exploration of the Sea) temporal trend assessment procedure (Nicholson et al., 1998). In short, the log-median OHC concentration was used as annual contaminant index value. The total variation over time of the OHCs and SIs was partitioned into a linear and a non-linear component. Linear regression analysis was applied to describe the linear component. A LOESS smoother (locally weighted scatter plot using a weighted quadratic least squares regression smoothing) with a window width of 7 years was applied to describe the non-linear component. The linear and non-linear components were tested by means of an analysis of variance (ANOVA). The theory behind the method is described in detail by Fryer and Nicholson (1999, 2002). Statistical analyses were performed using version 2.3.1 of the open source software R® (R development core team, 2010).

Table 1
Temporal trends of legacy organochlorine concentrations in three polar bear groups sampled in central East Greenland between 1983 and 2010. Significant trends are highlighted in bold.

Animal group:	Subadult (1983–2010)				Adult female (1984–2010)				Adult male (1989–2010)			
Contaminants	Annual change (%)	P (linear model)	P (nonlinear model)	n (years)	Annual change (%)	P (linear model)	P (nonlinear model)	n (years)	Annual change (%)	P (linear model)	P (nonlinear model)	n (years)
ΣPCB	**−4.0**	<**0.01**	**0.03**	23	**−2.2**	**0.02**	0.13	19	−1.8	0.11	0.24	16
PCB-153	**−3.6**	<**0.01**	<**0.01**	23	**−2.0**	**0.04**	0.10	19	−1.3	0.28	0.13	16
PCB-180	**−4.1**	<**0.01**	0.05	23	**−2.0**	0.12	0.45	19	−2.2	0.09	0.30	16
PCB-170/190	**−3.3**	<**0.01**	0.11	23	−0.8	0.51	0.54	19	−2.0	0.13	0.44	16
ΣDDT	**−4.4**	<**0.01**	0.09	23	**−6.1**	<**0.01**	0.61	19	−2.6	0.15	0.59	16
p,p′-DDE	**−4.0**	<**0.01**	0.10	23	**−5.6**	<**0.01**	0.59	19	−2.4	0.18	0.51	16
p,p′-DDD	**−5.1**	<**0.01**	**0.02**	23	**−7.5**	**0.01**	0.81	19	**−5.1**	**0.03**	0.40	16
p,p′-DDT	**−8.2**	<**0.01**	0.22	23	**−9.1**	<**0.01**	0.90	19	−4.4	0.07	0.28	16
α-HCH	**−10.8**	<**0.01**	<**0.01**	23	**−12.5**	<**0.01**	0.17	19	**−9.5**	<**0.01**	0.22	16
β-HCH	**−3.5**	<**0.01**	0.18	23	**−3.6**	<**0.01**	**0.05**	19	**−4.0**	<**0.01**	0.16	16
HCB	**−2.0**	<**0.01**	**0.04**	23	**−3.6**	<**0.01**	0.29	19	**3.4**	**0.02**	<**0.01**	16
Octachlorostyrene	**−2.7**	<**0.01**	0.13	23	**−2.0**	**0.03**	0.41	19	−1.6	0.06	0.10	16
Dieldrin	**−3.9**	<**0.01**	0.61	23	**−4.0**	<**0.01**	0.83	19	−0.2	0.79	0.56	16
Oxychlordane	**−2.9**	<**0.01**	0.29	23	**−2.8**	**0.03**	0.83	19	0.5	0.63	0.85	16
c-Chlordane	−2.3	0.13	0.48	20	**−4.1**	<**0.01**	0.93	16	−1.7	0.13	0.52	13
t-Chlordane	**−5.6**	<**0.01**	0.58	18	−2.9	0.30	0.80	15	−2.4	0.46	0.51	11
t-Nonachlor	**−4.6**	<**0.01**	0.37	23	**−4.8**	<**0.01**	0.14	19	−2.5	0.08	0.66	16
c-Nonachlor	**−3.7**	<**0.01**	0.18	23	**−5.9**	**0.01**	0.39	19	−2.0	0.34	0.45	16
Heptachlor epoxide	**−3.3**	<**0.01**	0.2	23	**−3.5**	**0.01**	0.96	19	0.4	0.67	0.56	16
Mean significant trends	**−4.4**	**18/19**	**5/19**		**−5.0**	**16/19**	**1/19**		**−3.8**	**4/19**	**1/19**	
Min significant trends	**−10.8**				**−12.5**				**−9.5**			
Max significant trends	**−2.0**				**−0.8**				**3.4**			

3. Results and discussion

Legacy contaminants were analysed in adipose tissue from 294 polar bears sampled in Central East Greenland between 1983 and 2010 (Table S1). The majority of the samples ($n = 156$) were obtained from subadult bears during a 23 years period (1983–2010), whereas samples from 61 adult females were obtained in 19 years during a 27 year period (1984–2010) and 77 males in 16 years during a 22 year period (1989–2010; Table S1).

3.1. Overall trend patterns

Temporal trend analyses were performed on nineteen legacy contaminant classes and major congeners: sum (Σ) PCB, 4 PCB congeners (CB153, 180 and 170/190), ΣDDT, *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT, α- and β-HCH, HCB, octachlorostyrene, dieldrin and CHLs (oxychlordane, *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor and heptachlor epoxide; Table 1). Of these, 18 showed significant declines of, on average, −4.4%/year (range: −2.0 to −10.8%/year) in subadult polar bears (Table 1). For instance, the average $\sum$PCB concentrations declined almost 3 fold from 22 730 ng/g lw (95% C.I.: 12 470–32 990 ng/g lw) from the 1983–1986 period to 8473 ng/g lw (95% C.I.: 7020–9926 ng/g lw) from 2006 to 2010 (Table S2). These declines are a consequence of bans on the production and use of PCBs and organochlorine pesticides during the 1970s and the following two decades (AMAP, 1998). Overall the temporal trends measured in East Greenland polar bears mirrored other documented temporal trends that reflect the effect of international regulations and conventions on long range trans-boundary transport of legacy OHCs.

In a corresponding investigation on the same polar bears brominated flame retardants (BFRs) were also analysed (Dietz et al., 2012). Most BFRs exhibited increasing trends, unlike the legacy OHCs. That is, significant linear increases were found for ΣPBDE, BDE100 and 153, and HBCD, showing average increases of 5.0% per year (range: 2.9 to 7.6%/year) among the subadult polar bears. Concentrations of BDE47, BDE99 and ΣPBDE showed significant non-linear trends, peaking during the years 2000–2004. Similar linear trends were found for adult females and adult males although fewer significant temporal trend correlations were found in adult females and the fewest in adult males (Dietz et al., 2012). Only one of the BFRs showed decreasing trends, namely BB153, with declines ranging from −1.1 to −2.2%/year depending on age and sex group. PBBs show a different time trend than the other BFRs, as hexaBB was banned earlier due to an accidental mixing of hexaBB into cattle feed in Michigan, USA , in 1973. OctaBB and decaBB production continued in the US until 1979, but in France it was used until 2000 (Alaee et al., 2003).

3.2. Specific temporal trends

3.2.1. PCBs

For subadult polar bears, decreasing temporal trends averaging −3.8%/year (range: −3.3 to −4.1%/year) were observed in $\sum$PCB and the three highest concentration congeners (CB153, 180 and 170/190), and all linear trends were statistically significant (Table 1; Fig. 1). Only two out of four correlations were significant for adult female bears and none were significant for the adult males. The significant female trends showed a lower rate of decline (mean: −2.1; range: −2.0 to −2.2%/year) than in subadults. Previously documented temporal trends in $\sum$PCB concentrations varied among species, subpopulations and time periods. Similarly, subadult ringed seals from the same East Greenland region showed significant decreasing temporal trends for PCBs of −4.5%/year (range: −3.5 to −5.9%/year) between 1986 and 2010 (Vorkamp et al., 2011). As also found in the present study, the adult ringed seals showed a smaller number of significant trends than the subadults. Only two out of six tested congeners were significant for adult ringed seals with a slightly lower rate of decline averaging −4.0%/year (range: −3.6 to −4.4%/year) (Vorkamp et al., 2011).

Dietz et al. (2004) earlier reported a decline of 74.5 to 81.2% for $\sum_{10}$PCB (CB99, 149, 118, 146, 153, 138, 183, 180, 170/190 and 194) and 5 separate major congeners (CB199, 153, 138, 180 and 170/190) over the 10 year period from 1989–1991 to 1999–2001 in East Greenland polar bears. This decline is equivalent to yearly declines of −12.8 to −15.4%/year, which is significantly ($p < 0.0001$) more rapid than declines we found in the present study. One of the reasons that the decline between 1989–1991 and 1999–2001 is somewhat steeper than the decline between 1983 and 2010 is that a levelling off or moderate increase in concentrations occurred post-2000. In addition, the 1989–1991 analyses from Dietz et al. (2004) were carried out at a different lab (Norstrom et al., 1998). The Σ10PCB concentrations reported in Norstrom et al. were significantly higher (27.0% higher, $p = 0.044$) than our current analysis on in the same animals collected between 1989 and 1991. This difference was mainly due to the congener CB170/190, which was 112% higher than in our current analysis.

Norstrom et al. (1998) could not document any consistent temporal trends in ΣPCB in Canadian Arctic polar bears during the 1980s, as concentrations were higher in four areas, lower in five areas, and the same in three areas for the period 1982–1984 compared to 1989–1993. Beside the limited data, some analytical inconsistencies and varying sex and age composition of the samples were provided as reasons for the inconclusive patterns (Norstrom et al., 1998).

A comparison of age-adjusted Σ_{42}PCB concentrations for polar bear data reported in Norstrom et al. (1998) and Verreault et al. (2005) revealed a consistent decrease of 32–75% over the period from 1989–1993 to 1996–2002. These decreases are equivalent to yearly declines of −1.5%/year at Northern Baffin Island to −12.8%/year in East Greenland. Similarly, Henriksen et al. (2001) reported decreasing Svalbard polar bear blood plasma CB153 levels of 36% over four years (from 1991–1994 to 1995–1998) equivalent of a yearly decline of −10.5%/year throughout the 1990s, which was slightly steeper than our calculated decline between 1983 and 2000.

When we looked at our East Greenland data for the period 2000 to 2010, there was no further decline, but an insignificant increase ($p = 0.831$) for $\sum$PCB of 1.3%/year, and a similar insignificant increase ($p = 0.171$) for PCB-153 of 2.9%/year. McKinney et al. (2011) analysed $\sum_{42}$PCB polar bears sampled from the entire Arctic (except Russia) in 2005–2008 and compared these with bears from 1996 to 2002 (Verreault et al., 2005). Decreasing patterns (here, recalculated to yearly trends) were only observed for Alaska–Chukchi/Bering Sea (−4.3%/year) and Svalbard (−1.5%/year) bears. Bears from the 7 other subpopulations (northern Beaufort Sea, Gulf of Boothia, Lancaster/Jones Sound, Baffin Bay, Davis Strait, western Hudson Bay, East Greenland) showed a yearly +8.8% mean increase ranging from +2.5% in Baffin Bay to +15.3% in the northern Beaufort Sea region (McKinney et al., 2011).

In conclusion, most $\sum$PCB and individual congener concentrations seem to have levelled off in the 2000s in most polar bear subpopulations, subsequent to the declines observed in the 1990s. Although many studies have found declining PCB trends in Arctic biota depending on the time periods and number of years analysed, others have not (Rigét et al., 2010). Of 40 Arctic time series of CB153 (the predominant PCB congener in East Greenland polar bears), the mean annual decrease was −1.2%. The lower annual decrease compared to the one found in the present study is most likely caused by the shorter and more recent time series of the datasets, which are reflective of a recent slowing in the declining trends. A reduction of PCB declines has likewise been reported in UK harbour porpoises (*Phocoena phocoena*) between 1991 and 2005 (Law et al., 2010). Also, PCB trends in Arctic air have shown only slight declines over the period 1998–2006 (Hung et al., 2010).

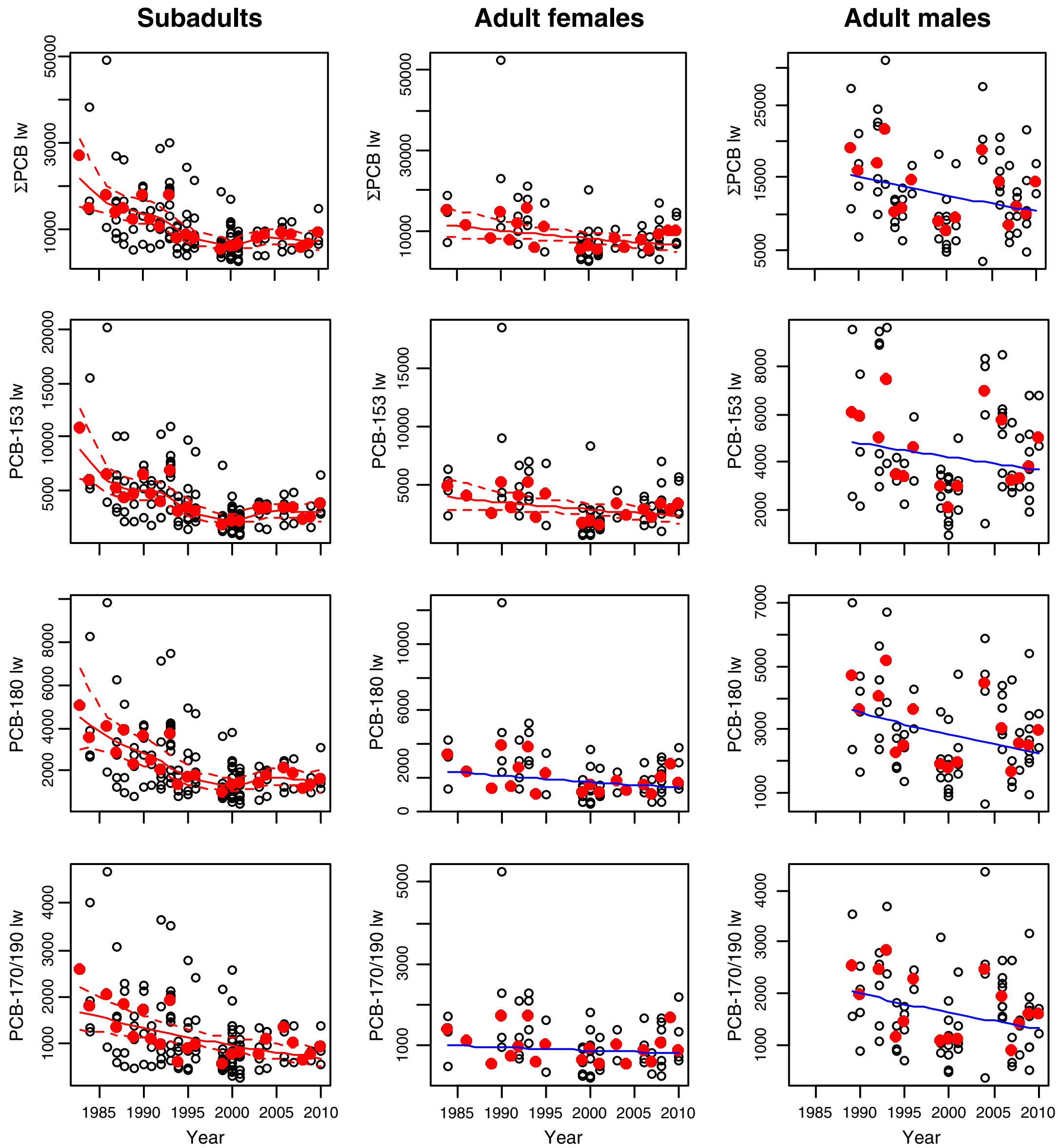

Fig. 1. Log-linear temporal trend of ΣPCB, CB-153, CB-180 and CB-170/190 in a retrospective time trend study on subadult, adult female and adult male polar bears from East Greenland. The filled dots are median values. Lines accompanied by dotted lines showing the 95% confidence intervals (C.I.) indicate significant trends ($p \leq 0.05$). Lines without C.I. indicate non-significant trends. Trend and significance levels are provided in Table 1. All units of the graphs above are given in ng/g lw.

3.3. Other legacy OHCs

3.3.1. DDTs

Temporal trends of −4.4%/year for ΣDDT and −4.0, −5.1 and −8.2%/year for the congeners *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT, respectively, were measured in subadult polar bears (Table 1; Fig. 2). All linear models were found to be significant for the subadults. For adult females, the yearly linear declines were likewise significant ($p \leq 0.01$; range: −5.6 to −9.1%). Adult male bears showed less steep trends than the females and subadults of both sexes (range: −2.4 to −5.1%/year) and only the *p,p′*-DDE decline was significant at −5.1%/year. Dietz et al. (2004) reported a decline of −33.7% for ∑DDT and −28.9% for *p,p′*-DDE over the 10 year period from 1989–1991 to 1999–2001. These trends are equivalent to between −4.0 and −3.4%/year, similar to the present study. Norstrom et al. (1998) suggested that *p,p′*-DDE declined in most Canadian Arctic regions in the late 1980s, as mean concentrations in composite adipose tissue in 10 Canadian management zones were 0.40 ± 0.34 μg/g lw in 1982–1984 (Norstrom et al., 1988) compared to 0.26 ± 0.21 μg/g in 1989–1993.

Rigét et al. (2004) investigated temporal trends of ΣDDT in four horned sculpin (*Myoxocephalus quadricornis*) and ringed seals from Ittoqqortoormiit in 1994, 1999 and 2000 (2001 only for sculpins), but no clear temporal trends were found given such limited time periods. The *p,p′*-DDE levels decreased from 1989–1993 to 2005–2008 in most polar bear subpopulations reported by McKinney et al. (2011). For the Western Hudson Bay subpopulation, DDT congeners showed a steeper decline compared to temporal patterns in most other subpopulations (McKinney et al., 2009, 2010). The decline was somewhat reduced (from −11 to -6.7%/year for ΣDDT) when correcting for changes in diet over the period (McKinney et al., 2009).

In a review of 40 and 19 Arctic time-series, the mean annual decrease was −1.9 and −4.4%/year for *p,p′*-DDE and ΣDDT, respectively (Rigét et al., 2010). In addition, *p,p′*-DDT was found to have declined

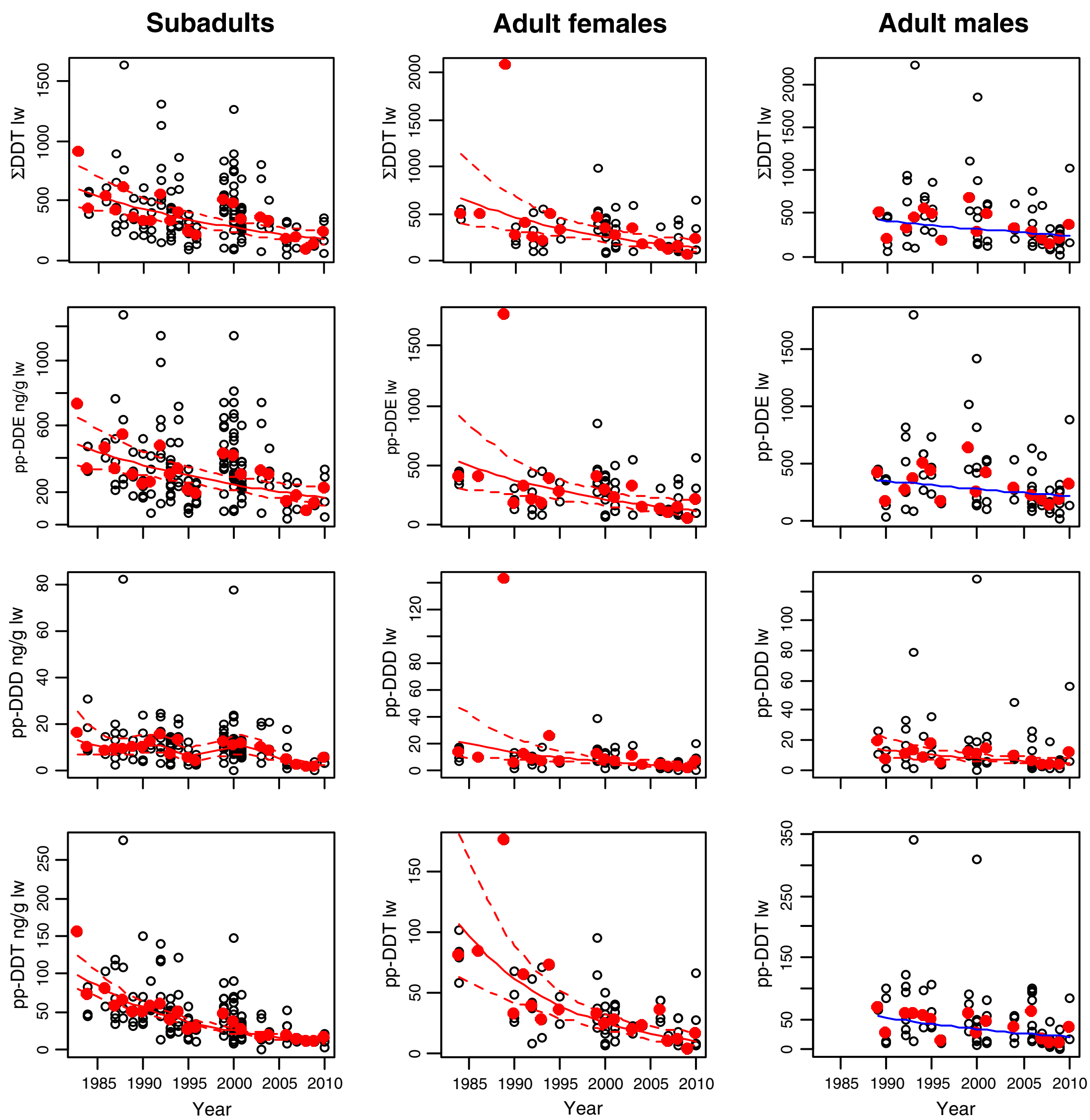

Fig. 2. Log-linear temporal trend of ΣDDT, *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT in a retrospective time trend study on subadult, adult female and adult male polar bears from East Greenland. The filled dots are median values. Lines accompanied by dotted lines showing the 95% confidence intervals (C.I.) indicate significant trends ($p \leq 0.05$). Lines without C.I. indicate non-significant trends. Trend and significance levels are provided in Table 1. All units of the graphs above are given in ng/g lw.

significantly in ringed seals from the Beaufort Sea (−3.3%/year; 7 years; 1972–2010), Lancaster Sound (−2.8%/year; 16 years; 1972–2010), Hudson Bay (−7.2%/year; 12 years; 1986–2010), and Baffin Bay (−3.1%/year; 6 years; 1986–2006) (AANDC, in press).

3.3.2. α-HCH

A temporal trend of −10.6%/year for α-HCH was found for the subadult bears, which was intermediate between the declines found in adult females (−12.5%/year) and the adult males (−9.5%/year). All linear models were significant ($p<0.01$; Table 1; Fig. 3). Dietz et al. (2004) reported a decline of 45.4% for α-HCH over the 10 year period from 1989–1991 to 1999–2001, which is an equivalent yearly trend of −5.9%/year, a less steep decline than reported in the present study. Rigét et al. (2008) analysed HCHs in ringed seals from Greenland waters from the same region as the current study (Ittoqqortoormiit region). Significant ($p<0.01$) decreasing trends for α-HCH of −10.4%/year in subadult (1986–2006) and −11.7%/year in adult (1994–2006) ringed seals were within the adult polar bear time trend range. Rigét et al. (2008, 2010) analysed a total of 32 α-HCH time-series and found an average annual yearly decrease of −7.4%. Also, a Western Hudson Bay study from 1991 to 2007, wherein archived samples were simultaneously analysed, documented significantly decreasing α-HCH levels of −11%/year (McKinney et al., 2010). In contrast, Addison et al. (2009) analysed α-HCH (and γ-HCH) in ringed seals at Ulukhaktok, Canada over the period 1978–2006, but found no significant trend.

3.3.3. β-HCH

A much weaker decline was observed for β-HCH compared to the α-HCH isomer described above; the temporal trend for the subadult bears was −3.5%/year and similar for adult females (−3.6%/year) and adult males (−4.0%/year). Again all linear models were significant (Table 1; Fig. 3). Dietz et al. (2004) reported a decline of 64.7% for β-HCH over the 10 year period from 1989–1991 to 1999–2001, which is equivalent to yearly trends of −9.9%/year, and a steeper decline than reported in the present study. The β-HCH trends in ringed seals from Ittoqqortoormiit were likewise weaker than for α-HCH with a decline of −1.4%/year in subadults and −3.1%/year in adults, although none of these trends were significant ($0.13<p<0.39$) (Rigét et al., 2008). According to Rigét et al. (2010), the mean temporal trends of β-HCH in 21 Arctic time-series showed an average

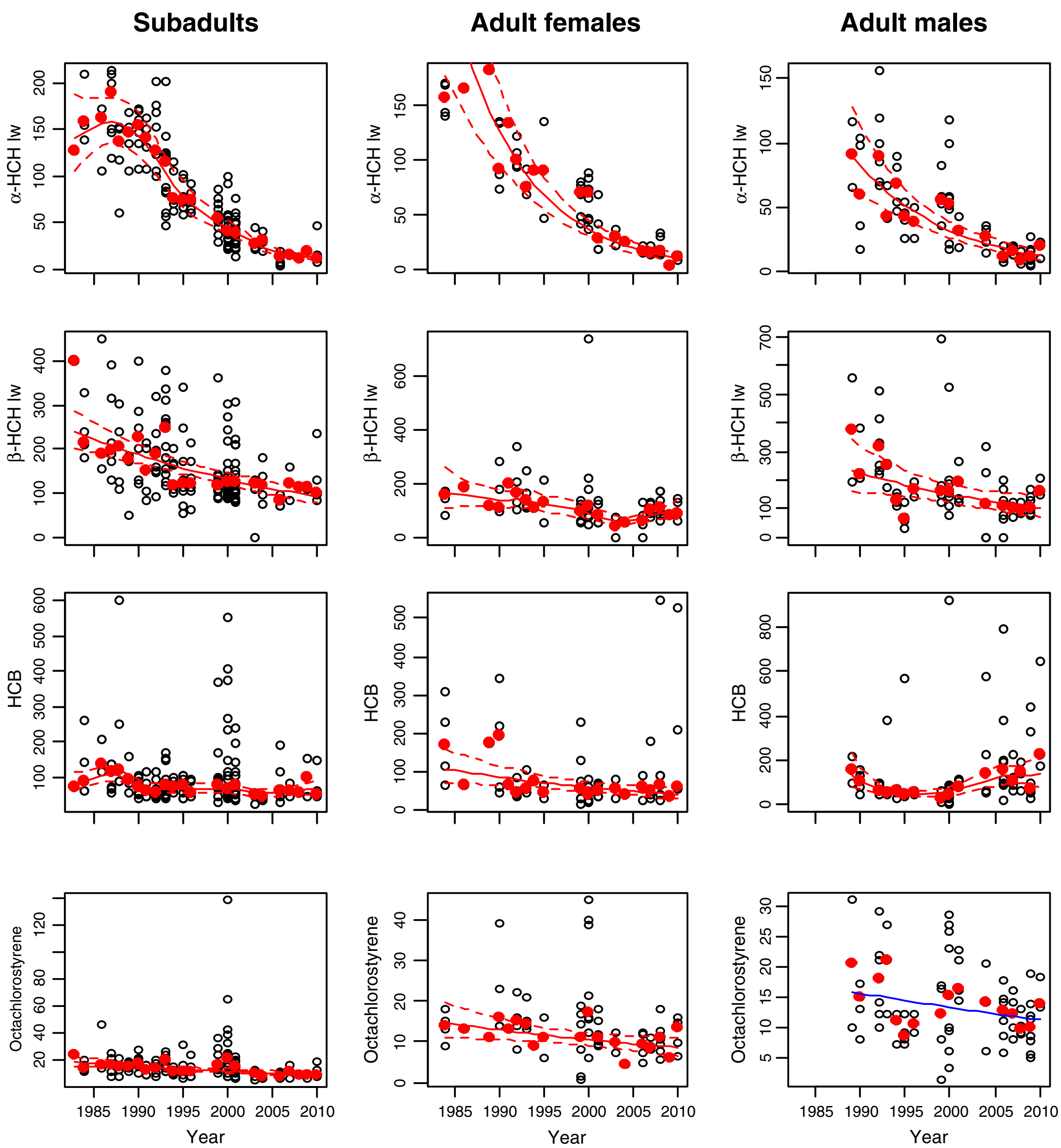

Fig. 3. Log-linear temporal trend of α-HCH, β-HCH, HCB and octachlorostyrene in a retrospective time trend study on subadult, adult female and adult male polar bears from East Greenland. The filled dots are median values. Lines accompanied by dotted lines showing the 95% confidence intervals (C.I.) indicate significant trends ($p \leq 0.05$). Lines without C.I. indicate non-significant trends. Trend and significance levels are provided in Table 1. All units of the graphs above are given in ng/g lw.

annual decrease of −2.9%/year. For 25% of the time-series, the decreasing trend was significant. In contrast, McKinney et al. (2010) found a significant log-linear increase in β-HCH levels of 8.5% in polar bears from the Western Hudson Bay study from 1991 to 2007. Similarly, Addison et al. (2009) showed that β-HCH was continuing increase in ringed seals at Ulukhaktok over the period 1978–2006, about 8 to 10-fold in females and 4 to 5-fold in males. The finding of increasing β-HCH and decreasing α-HCH levels may be explained by greater partitioning of β-HCH into sea water resulting in the slower arrival to northern latitudes via oceanic currents (Addison et al., 2009).

3.3.4. HCB

A significantly decreasing trend of −2.0%/year for HCB was found in subadult bears and a somewhat steeper decline was found in adult females (−3.6%/year). However, the adult males showed a significant linear increase of 3.4%/year (Table 1, Fig. 3). Rigét et al. (2004) investigated temporal trends of HCB in sculpin and ringed seals from Ittoqqortoormiit/Scoresby Sound in East Greenland in 1994, 1999 and 2000 (2001 only for sculpins), but for these few years no temporal trends were discernible. Rigét et al. (2010) reported an annual decrease averaging −2.5%/year in 40 HCB time series. Barber et al. (2005) found that HCB concentrations peaked in the 1960s, followed by a consistent downward trend of HCB levels in the environment over the past 20 years. However, some opposite trends in abiotic media, such as lake sediments, ice core and precipitation have been observed in the Arctic, as polar regions may still act as sinks for globally emitted HCB (Barber et al., 2005). Hung et al. (2010) reported that HCB concentrations decreased in Arctic air through the 1990s, yet over the period of 2000 to 2005 HCB concentrations slightly increased at the Arctic stations Alert and Zeppelin. This increasing trend cannot be directly attributed to increased emissions as HCB was largely banned as a pesticide in the 1970s. However, it is a known contaminant in some current-use pesticides, which have been increasingly used since 2000. Hung et al. (2010) hypothesized that increasingly ice-free Arctic sea-water may be acting as a HCB source to the atmosphere, influencing the atmospheric trend. The trend in East Greenland male polar bears is the first example of Arctic biota showing such an increase.

3.3.5. Octachlorostyrene (OCS)

A significantly decreasing trend of −2.7%/year for OCS was found in subadult bears and significant trends of the same magnitude were found in adult females (−2.0%/year). The adult males showed a less steep and non-significant decline of −1.6%/year (Table 1; Fig. 3). Only few Arctic time trend data sets were found in the literature for OCS. McKinney et al. (2011) found lower OCS levels in 2005–2008 relative to 1996–2002 in polar bears from Alaska, East Greenland and Svalbard. For other polar bear subpopulations, OCS was largely not detected in 1996–2002, whereas OCS was detected in all 2005–2008 samples from the 11 polar bear subpopulations. According to McKinney et al. (2011), this could be due to analytical variability between studies confounding the temporal comparisons.

3.3.6. Dieldrin

Significantly decreasing log-linear temporal trends were found for dieldrin in subadult (−3.9%/year) and adult female bears (−4.0%/year) (Table 1; Fig. 4). Adult males showed a non-significant log-linear decline of −0.2%/year (Table 1; Fig. 4). For adult male, female, and subadult East Greenland polar bears, dieldrin levels decreased significantly by between 27.0 and 69.5% from 1989–1991 to 1999–2001 (Dietz et al., 2004). These changes were equivalent to yearly decreases of −3.6 to −12.2%, which was somewhat higher than found in the present study. Norstrom et al. (1998) likewise documented an almost tenfold decrease in mean dieldrin concentrations across Canadian Arctic polar bears in comparisons between 1982 and 1984 (960 ± 260 ng/g lw) (Norstrom et al., 1988) and 1989–1993 (98 ± 59 ng/g lw). Similarly, Muir and Norstrom (2000) reported a significant decrease in dieldrin concentrations in polar bears from Barrow Strait and Queen Maud Gulf in the central Canadian Arctic archipelago from 1984 to 1989, whereas no changes were detected in bears from northern Baffin Bay in the same period. Also, during the 1990s, no temporal trend was detected in bears from the Hudson Bay (de Wit et al., 2004). McKinney et al. (2011) also found no declines in dieldrin within the 11 polar bear management zones from Alaska, Canada, Greenland, and Svalbard in 2005–2008 compared to with earlier published trans-Arctic polar bear dieldrin data from 1989–1993

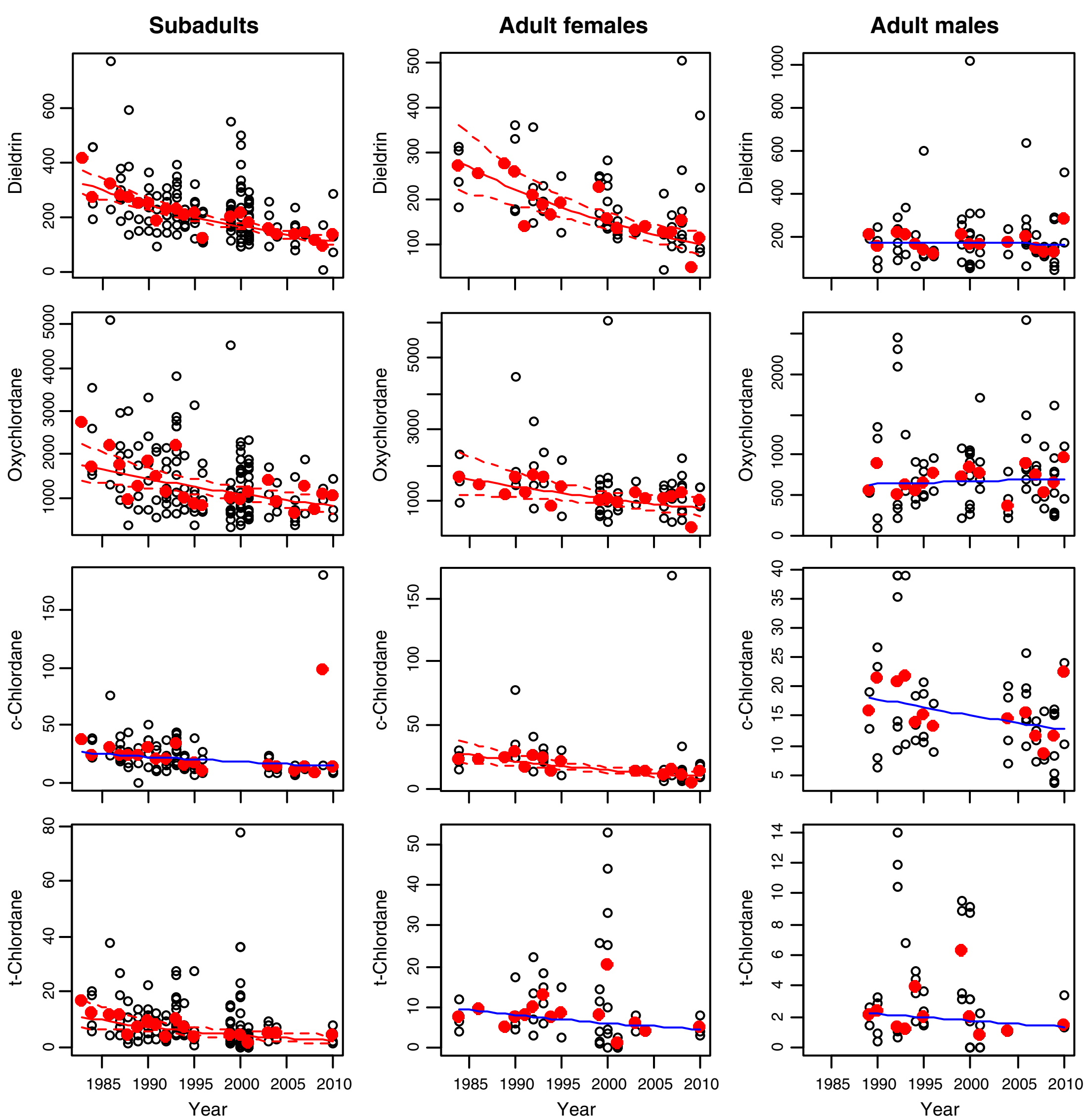

Fig. 4. Log-linear temporal trend of dieldrin, oxychlordane, *cis*-chlordane, and *trans*-chlordane in a retrospective time trend study on subadult, adult female and adult male polar bears from East Greenland. The filled dots are median values. Lines accompanied by dotted lines showing the 95% confidence intervals (C.I.) indicate significant trends ($p \leq 0.05$). Lines without C.I. indicate non-significant trends. Trend and significance levels are provided in Table 1. All units of the graphs above are given in ng/g lw.

(Norstrom, 2001) and 1996–2002 (Verreault et al., 2005). The insignificant trends in earlier polar bear studies are likely due to less robust time series compared to those presented in this study.

The mean change for 11 dieldrin time-series from both Canada and Greenland was an average annual decrease of −2.1%/year (Rigét et al., 2010). The decreases in biota are consistent with decreasing trends observed in Arctic air during the late 1990s at two stations in the Canadian Arctic (Hung et al., 2010).

3.3.7. Chlordanes

Significant linear temporal decreases were observed for five out of the six monitored chlordane-related compounds in subadult polar bears, namely oxychlordane (−2.9%/year), heptachlor epoxide (−3.3%/year), *cis*-nonachlor (−3.7%/year), *trans*-nonachlor (−4.6%/year), and *trans*-chlordane (−5.6%/year) (Table 1; Figs. 4 and 5). A similar pattern was observed in adult females, where significant linear decreasing trends were observed in five out of the six chlordanes. Slower declines were observed in adult males, but none of these were significant (Table 1; Fig. 4). For subadult, adult female and adult male polar bears from East Greenland, ΣCHL levels decreased significantly by between −31.7 and −75.6% from 1989–1991 to 1999–2001 (Dietz et al., 2004). These changes were equivalent to a yearly decrease from −3.7 to −13.2%, indicating slightly steeper declines during the last ten years of the millennium than during the entire period of the present study (Table 1; Fig. 4). As reported by McKinney et al. (2011), ΣCHL levels decreased in most of the investigated polar bear subpopulations when comparing data from 1989 to 1993 to previous published data from 1996–2002 and 2005–2008. The 2005–2008 levels were lower relative to the Norstrom et al. (1998) results from 1989 to 1993 for all subpopulations with the exception of western Hudson Bay. A moderate ΣCHL increase of 0.04%/year was found for the longer-term studies in the western Hudson Bay polar bears, but these turned into a yearly decline of −1.1% when adjusting for changes in diet over time (McKinney et al., 2009). A significant ΣCHL increase of 0.2%/year was found for the same region and bears of western Hudson Bay by McKinney et al. (2010).

Norstrom et al. (1998) documented declining ΣCHL concentrations in polar bear adipose tissues between 1982–84 and 1989–1993 (Norstrom et al., 1988). This comparison did not take sex and age into account. However, Norstrom et al. (1988) also analysed ΣCHL in polar bears from three regions (southern Baffin Bay/northern Davis Strait, Fox Basin/Hudson Strait and western Hudson Bay) from 1969, in which a 4 fold increase took place relative to 1982–1984 (Norstrom et al., 1988). This indicates that ΣCHL concentrations peaked somewhere around the early 1980s.

Rigét et al. (2010) reported the mean change for 17 ΣCHL time-series to be an annual decrease of −1.8%. For 29 time-series of *trans*-nonachlor, the annual decrease was −1.0%. Chlordane-related compound concentrations were summarized in a recent Arctic assessment on ringed seals from 4 regions of Arctic Canada (AANDC, in press). Declines were observed in the Baffin Bay (−4.3%/year; 6 years; 1986–2006) and Hudson Bay (−8.2%/year; 12 years; 1986–2010) regions. However, the ΣCHL trends in the Beaufort Sea (−0.11%/year; 7 years; 1972–2010) and Lancaster Sound (0.89%/year; 16 years; 1972–2010) regions were not statistically significant, which the authors suggested could be related to continued inputs into these regions (AANDC, in press).

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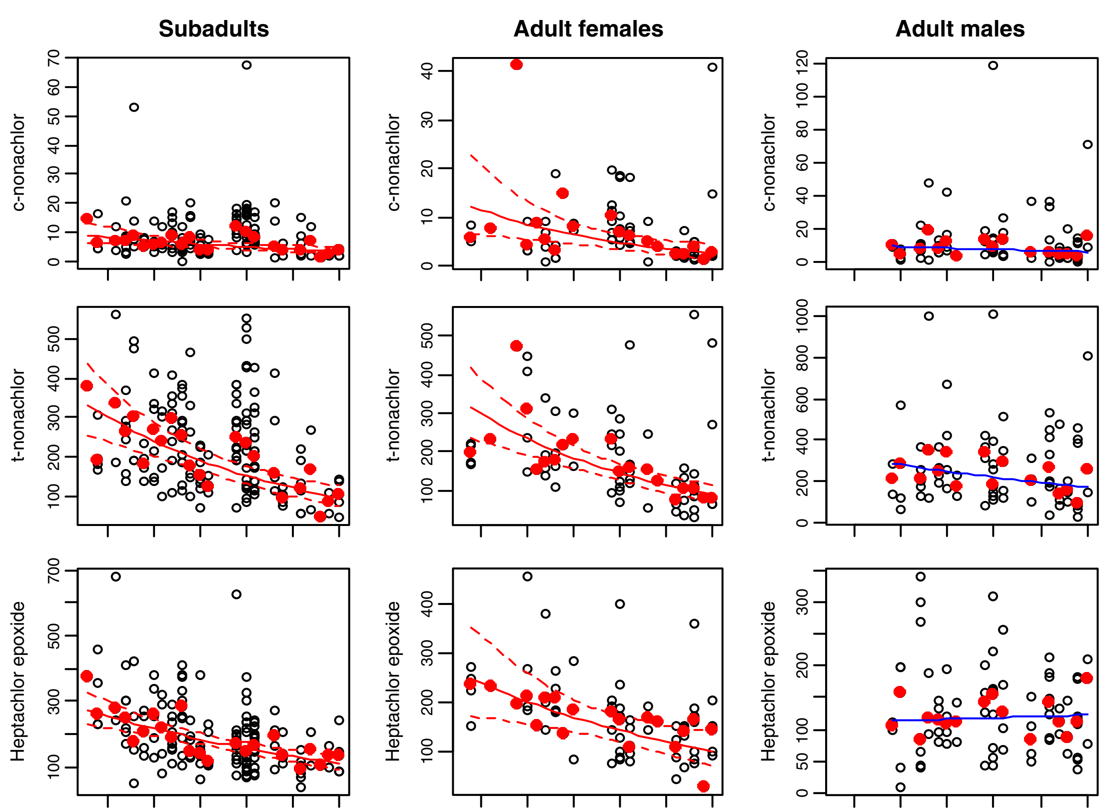

Fig. 5. Log-linear temporal trend of *cis*-nonachlor, *trans*-nonachlor and heptachlor epoxide in a retrospective time trend study on subadult, adult female and adult male polar bears from East Greenland. The filled dots are median values. Lines accompanied by dotted lines showing the 95% confidence intervals (C.I.) indicate significant trends (p ≤ 0.05). Lines without C.I. indicate non-significant trends. Trend and significance levels are provided in Table 1. All units of the graphs above are given in ng/g lw.

of the majority of polar bear fat/adipose tissue samples. The early sampling in Greenland was funded by the Greenland Institute of Natural Resources (Nuuk), Aage V. Jensen's Foundation. Samplings since 1999 and all chemical analyses were funded by a number of projects under the DANCEA (Danish Cooperation for Environment in the Arctic) programme including the CORE programme and the large scale IPY programme "BearHealth", which was also supported by KVUG (The Commission for Scientific Research in Greenland) and The Prince Albert II Foundation.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2012.09.004.

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