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The role of diet on long-term concentration and pattern trends of brominated and chlorinated contaminants in western Hudson Bay polar bears, 1991–2007

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ABSTRACT

Adipose tissue was sampled from the western Hudson Bay (WHB) subpopulation of polar bears at intervals from 1991 to 2007 to examine temporal trends of PCB and OCP levels both on an individual and sum-(∑-) contaminant basis. We also determined levels and temporal trends of emerging polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD), polybrominated biphenyls (PBBs) and other current-use brominated flame retardants. Over the 17-year period, ∑DDT (and *p,p′*-DDE, *p,p′*-DDD, *p,p′*-DDT) decreased (−8.4%/year); α-hexachlorocyclohexane (α-HCH) decreased (−11%/year); β-HCH increased (+8.3%/year); and ∑PCB and ∑chlordane (CHL), both contaminants at highest concentrations in all years (>1 ppm), showed no distinct trends even when compared to previous data for this subpopulation dating back to 1968. Some of the less persistent PCB congeners decreased significantly (−1.6%/year to −6.3%/year), whereas CB153 levels tended to increase (+3.3%/year). Parent CHLs (*c*-nonachlor, *t*-nonachlor) declined, whereas non-monotonic trends were detected for metabolites (heptachlor epoxide, oxychlordane). ∑chlorobenzene, octachlorostyrene, ∑mirex, ∑$MeSO_2$-PCB and dieldrin did not significantly change. Increasing ∑PBDE levels (+13%/year) matched increases in the four consistently detected congeners, BDE47, BDE99, BDE100 and BDE153. Although no trend was observed, total-(α)-HBCD was only detected post-2000. Levels of the highest concentration brominated contaminant, BB153, showed no temporal change. As long-term ecosystem changes affecting contaminant levels may also affect contaminant patterns, we examined the influence of year (i.e., aging or "weathering" of the contaminant pattern), dietary tracers (carbon stable isotope ratios, fatty acid patterns) and biological (age/sex) group on congener/metabolite profiles. Patterns of PCBs, CHLs and PBDEs were correlated with dietary tracers and biological group, but only PCB and CHL patterns were correlated with year. DDT patterns were not associated with any explanatory variables, possibly related to local DDT sources. Contaminant pattern trends may be useful in distinguishing the possible role of ecological/diet changes on contaminant burdens from expected dynamics due to atmospheric sources and weathering.

1. Introduction

Persistent organic pollutants (POPs) in the Arctic, such as PCBs and organochlorine pesticides (OCPs), are largely transported from lower latitudes and tend to accumulate in northern wildlife (Letcher et al., 2010). Recent research has demonstrated increasing complexity in the organohalogen residues in these biota, including various brominated flame retardants (BFRs), polyfluorinated surfactants, current-use pesticides and persistent metabolites of legacy POPs such methylsulfone-($MeSO_2$-)PCBs (de Wit et al., 2010; Letcher et al., 2010; Weber et al., 2010). Regarding BFRs, polybrominated biphenyls (PBBs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) have been detected in various Arctic and subarctic biota (de Wit et al., 2010). However, less is known about the presence of new or replacement BFRs, such as pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), hexabromobenzene (HBB), 1,2-*bis*(2,4,6-tribromophenoxy)ethane (BTBPE) and decabromodiphenyl ethane (DBDPE) (de Wit et al., 2010).

The polar bear (*Ursus maritimus*) is a top predator in Arctic and subarctic marine food webs. High levels of POPs in polar bears have been strongly implicated as health stressors in subpopulations in East Greenland, Svalbard and southern and western Hudson Bay (Letcher et al., 2010). Although cause–effect relationships have not been

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established, several contaminant-biomarker correlative studies on the former two subpopulations have found associations between levels of certain organohalogens and, in particular, immune, reproductive and endocrine system biomarkers of effects (e.g., Haave et al., 2003; Lie et al., 2004; Skaare et al., 2001).

To address elevated exposures and associated risks from both legacy and emerging chemicals of concern, it is important to monitor trends and effects in sentinel species including polar bears. Currently, temporal trends have not been reported for any individual PBDEs or non-PBDE BFRs in polar bears. For PCBs and OCPs, the most extensive temporal trend data sets are from the western Hudson Bay (WHB) subpopulation. Analysis of archived WHB female adipose tissues collected in 1968, 1984 and most years between 1989 and 2002 demonstrated that levels of certain chlorinated contaminants were highest in 1968 and/or 1984, and that levels of most contaminants either declined or remained unchanged from 1989 to 2002 (Braune et al., 2005; Norstrom 2001; Verreault et al., 2005). Throughout the 1990s, PCB and/or OCP levels in East Greenland and Svalbard polar bears declined somewhat faster than in WHB bears (Dietz et al., 2004; Henriksen et al., 2001). Extended temporal trends for polar bear subpopulations that encompass more recent time points are lacking.

Particularly for current and future Arctic and subarctic trends studies, climate change may be an important factor influencing and confounding our interpretation of variation in POPs levels and patterns (Macdonald et al., 2005). For instance, Gaden et al. (2009) found that temporal trends of mercury in western Canadian arctic ringed seals (*Pusa hispida*) were correlated not with year but with length of ice-free season. Increasing trends of PCBs and mercury in western Canadian arctic burbot (*Lota lota*) were linked with increased primary productivity, likely as a result of warming temperatures (Carrie et al., 2010). Regarding polar bears, recent temporal trends in sum-($\sum$-)concentrations of selected POPs in the WHB subpopulation were partially explained by sea ice-associated changes in the marine diet/food web (McKinney et al., 2009). Thus, contaminant levels in polar bears reflect not only altered atmospheric sources and releases over time, but also variation and potential long-term ecosystem changes due to climate change. To our knowledge, changes in patterns of the congeners/metabolites in polar bears in relation to these diet/food web changes have not as yet been investigated.

Here, we expand on our previous work on WHB contaminant trends, and in particular, the influence of diet/food web changes (McKinney et al., 2009). Previously, we reported the annual change for select contaminant classes and the relationship to sea ice-associated diet/food web changes. In the current study, our first objective was to assess the actual individual and $\sum$-contaminant concentrations, congener and metabolite patterns and temporal trends for a comprehensive suite of legacy POPs in WHB adipose tissues. We compared these findings to previous longer-term WHB trends to provide one of the longest-term temporal comparisons reported to date for any arctic species. Next, we reported for the first time on the concentrations and temporal trends of numerous BFRs in polar bears. Finally, we tested the hypothesis that congener/metabolite patterns are influenced not only by atmospheric sources and "weathering" (i.e., aging of the contaminant residue pattern), but also by diet/food web variation.

2. Materials and methods

2.1. Sample details

Sampling and sample storage were described by McKinney et al. (2009). We used archived samples collected from live captured and subsistence harvested WHB polar bears in 7 years from 1991 to 2007 (Table 1). Samples chosen were mainly collected in September–November, which likely reduced seasonal variation in contaminant concentrations (Dietz et al., 2004). Initial efforts focused on adult (>4 year old) females to minimize effects of sex-related variation in contaminant levels and because age effects on PCB, OCP and PBDE concentrations are minimal for this group (Muir et al., 2006; Norstrom et al., 1998). However, the inclusion of samples from adult males and subadults (3–4 years old) in years post-2000 was unavoidable in order to attain adequate sample sizes.

2.2. Organohalogen contaminant and dietary tracer analysis

Contaminants were extracted from approximately 0.5 g of adipose tissue (0.2 to 0.3 g for biopsies) and analyzed by gas chromatography-single quadrupole mass spectrometry (GC-MS) as described by McKinney et al. (2009). Samples were spiked prior to extraction with internal standards (ISs): a mixture of $^{13}C_{12}$-PCBs, $^{13}C_6$-chlorobenzenes (ClBz) and $^{13}C_{12}$-*p,p′*-DDE, as well as 3-CH_3SO_2-2-CH_3-2′,3′,4′,5,5′-pentaCB ($MeSO_2$-PCB IS) and BDE30 (BFR IS). After accelerated solvent extraction and gel permeation chromatography, two fractions were isolated using pre-conditioned silica solid phase extraction cartridges. The first fraction was eluted with 8 mL of 5% DCM in hexane and contained (if present) all PCBs, OCPs and BFRs listed below. The second fraction was eluted with 8 ml of 100% DCM and contained (if present) 3-$MeSO_2$-*p,p′*-DDE and all $MeSO_2$-PCBs listed below. Both fractions were spiked prior to analysis with a performance standard, $^{13}C_{12}$-CB138. Samples were analyzed for 8 non-PBDE BFRs (PBT, PBEB, HBB, BB101, BB153 (co-eluted with BDE154), total-(α)-HBCD, BTBPE and DBDPE) and 37 PBDE congeners, BDEs 28/33, 54, 75, 49, 47, 66, 77, 100, 119, 99, 116, 85, 155, 154, 153, 139, 138, 140, 156, 184, 183, 191, 180, 181, 190, 171, 202, 201, 197, 203, 196, 205, 208, 207, 206 and 209. In addition to monitoring all BFRs in SIM for $[Br]^-$ anions, $[C_6Br_5O]^-$ anions were also used to monitor nonaBDEs and BDE209. To address the co-elution of BDE154 with BB153, BDE154 was additionally monitored using $[M\text{-}2Br]^-$ anions, which were less abundant but allowed for separation from BB153. For PCBs, 74 congeners were monitored, CBs 18, 17, 16/32, 31, 28, 33/20, 22, 52, 49, 47/48, 44, 42/59, 64/41, 74, 70/76, 66, 56/60, 95, 92, 101/90, 99, 97, 87, 85, 110, 118, 114, 105, 151, 149, 146, 153, 141, 130, 137, 138, 158, 128, 167, 156, 157, 179, 176, 178, 187/182, 183, 174, 177, 171, 172, 180, 170/190, 189, 202, 200,

Table 1
Biometric data from western Hudson Bay polar bears sampled from 1991–2007.

	1991	1992	1994	1995	2001	2003	2007
Total *n*	14	15	15	15	9	12	12
Adult female (>4 years)[a]	14	15	15	15	3	3	1
Adult male (>4 years)	0	0	0	0	0	9	6
Subadult (<5 years)	0	0	0	0	6	0	5
Collection date range	1 September–20 September	6 September–19 September	29 August–16 September	1 September–15 September	28 August–30 November	2 November–29 November[b]	1 November–22 November[b]
Adipose % lipid (± SE)	78 ± 2	83 ± 2	75 ± 2	80 ± 2	78 ± 3	74 ± 3	78 ± 3

[a] Age group classification followed Norstrom et al. (1998).
[b] Except for two samples collected 27 February and 19 March (2004) for 2003 and two samples collected 22 May and 26 May (2008) for the 2007 sampling period. The inclusion of these samples did not affect the conclusions of this study. For instance, their mean $\sum$DDT concentrations (previously reported to vary with collection month; Dietz et al., 2004) were similar (within 1 SD unit) from the mean of the other samples from that year.

199, 196/203, 208, 195, 207, 194, 206 and 201. A suite of 23 $MeSO_2$-PCB congeners were monitored: 3-$MeSO_2$-CB52, 3′-$MeSO_2$-CB49, 4-$MeSO_2$-CB52, 4′-$MeSO_2$-CB49, 3-$MeSO_2$-CB64, 4-$MeSO_2$-CB64, 3-$MeSO_2$-CB91, 4-$MeSO_2$-CB91, 3-$MeSO_2$-CB70, 3′-$MeSO_2$-CB101, 4-$MeSO_2$-CB70, 4′-$MeSO_2$-CB101, 3′-$MeSO_2$-CB87, 3-$MeSO_2$-CB110, 3-$MeSO_2$-CB149, 4-$MeSO_2$-CB110, 4′-$MeSO_2$-CB87, 4-$MeSO_2$-CB149, 3′-$MeSO_2$-CB132, 4′-$MeSO_2$-CB132, 3′-$MeSO_2$-CB141, 4′-$MeSO_2$-CB141and 4′-$MeSO_2$-CB174. Finally, 19 OCPs were monitored: ClBzs (1,2,4,5-tetraClBz, 1,2,3,4-tetraClBz, pentaClBz and HCB), α- and β-hexachlorocyclohexane (HCH), octachlorostyrene (OCS), chlordanes (CHL; heptachlor epoxide, oxychlordane, *t*-chlordane, *c*-chlordane, *t*-nonachlor and *c*-nonachlor), DDTs (*p,p*′-DDE, *p,p*′-DDD and *p,p*′-DDT), dieldrin and mirex compounds (photomirex and mirex).

Carbon stable isotope ratios ($\delta^{13}C$) and fatty acids (FAs) were assessed as dietary tracers as described by McKinney et al. (2009). Briefly, 150–300 μg of lipid extract from gravimetric determination of lipid content was analyzed for $\delta^{13}C$ using an elemental analyzer coupled to a continuous flow isotope ratio mass spectrometer (Environmental Isotope Laboratory, University of Waterloo, Waterloo, ON, Canada). FAs were extracted from an inner sample (10–20 mg) of adipose tissue using 2:1 chloroform:methanol containing the antioxidant 2,6-di-*t*-butyl-4-methylphenol, then methylated via the Hilditch reagent, extracted and finally analyzed by GC with a flame ionization detector (FID). Factor scores from the first two significant principal components from a principal components analysis (PCA) of all dietary fatty acids (those that are incorporated relatively unchanged or predictably from prey to predator adipose tissues; Iverson et al., 2004) are herein referred to as FA-Index1 and FA-Index2 and considered representative indices of the FA variation in WHB polar bears (McKinney et al., 2009).

2.3. Quality control for contaminant and dietary tracer data

Quality control for dietary tracer analysis and some of the contaminant analysis were described by McKinney et al. (2009). Recoveries of the internal standards (IS) $^{13}C_6$-ClBzs, $^{13}C_{12}$-PCBs, $^{13}C_{12}$-*p,p*′-DDE, 3-CH_3SO_2-2-CH_3-2′,3′,4′,5,5′-pentaCB and BDE30 were 59 ± 7%, 85 ± 5%, 88 ± 8%, 91 ± 17% and 87 ± 12%, respectively. PCBs and OCPs were quantified by an external standard method, and ClBzs were also recovery corrected to account for volatility-related lower recoveries. $MeSO_2$-PCBs and BFRs were quantified by an internal standard method based on the relative response factor of the analyte versus the appropriate internal standard, and thus concentrations were inherently recovery and performance corrected.

The National Institute of Standards and Technology (NIST) pilot whale blubber SRM1945 was extracted and analyzed with each sample batch to assess accuracy and precision of the method. The average deviation in ∑PCB, ∑OCP and ∑PBDE concentrations from the certified values in repeated analyses of SRM1945 was 5 ± 4%, 7 ± 3% and 14 ± 7%, respectively. This SRM does not currently have certified values for $MeSO_2$-PCBs or non-PBDE BFRs. The average deviation on duplicate analyses of 6 polar bear adipose samples was 7%, 15%, 21% and 12% different for ∑PCB, ∑OCP, ∑PBDE and ∑$MeSO_2$-PCB.

A sodium sulfate method blank was analyzed with each batch of samples to assess any background contamination. Low and relatively constant blank responses were subtracted from sample responses on a batch-by-batch basis. However, BDE209 and DBDPE blank values were more variable and some were similar to sample values. Therefore, the method limits of quantification (MLOQs) for these compounds were set to the blank average plus 3× SD, and were higher than for other compounds. Reported BDE209 and DBDPE concentrations thus have a high degree of uncertainty, but most likely reflect the presence of these compounds in the samples since they were detected beyond the 99% confidence levels of the blanks. The MLOQs for all other analytes were based on a signal-to-noise ratio of 10.

2.4. Data analysis

Contaminant concentrations were calculated on a lipid weight basis. To minimize sex-related variation prior to statistical analyses, PCB, CHL and DDT concentrations (which consistently vary between sexes) in males were converted to female equivalents (Norstrom et al., 1998). Measures of central tendency were only calculated if >70% of the values in a given year were above the MLOQ, and in these cases, any non-detects were assigned to a random value between zero and one-half the MLOQ for statistical analysis. Similarly, trends were calculated only if >70% of the values in every year were above the MLOQ.

Temporal trends were based on the analytical approach of the International Council for the Exploration of the Sea (ICES) (Nicholson et al., 1998) and calculated by the PIA temporal trends statistical application (A. Bignert, 2007; developed for use by the Arctic Monitoring and Assessment Program (AMAP); available at www.amap.no). PIA analyzes the annual index values for (log) linear and non-linear (3-year running-mean smoother) trends. Medians were chosen as annual index values due to insensitivity to low numbers of extreme values and non-detects. Geometric means were additionally reported for selected contaminants for comparison to previous contaminant data sets for WHB bears. PIA also tests trends using the non-parametric Mann–Kendall test, which is less affected by endpoint values in the time series but has lower power to detect trends. Thus, trend direction was reported, and the *p*-value marked by an asterisk, only if at least a marginally significant trend ($p<0.10$) was also indicated by the non-parametric test. Annual % change ($(1-10^b)\times 100\%$, where *b* is the slope), r^2 and *p*-values from the linear regression portion were reported for reference even if there was no significant trend, a non-monotonic trend or a non-linear trend, but should not be considered meaningful in a quantitative sense.

For all contaminants, we tested whether the trend results (using PIA) were confounded by effects of age/sex group (i.e., subadult/adult male/adult female; hereafter referred to as biological group) by analysis of covariance (ANCOVA) with year and biological group (adjusted to female equivalents for PCBs, CHLs, DDTs). For contaminants known to vary between biological groups and for those where biological group was significant in the ANCOVA, annual median values for the 2000s years were also reported separately for each biological group and time trends were repeated on the adult female group. Time trend analyses repeated using PIA on the annual geometric mean and by simple linear regression on the individual concentration values using Statistica (StatSoft, 2003; Tulsa, OK, USA) were consistent with the reported trends. Contaminant concentrations were $\log(x+1)$-transformed to better approximate normal distribution (assessed by Shapiro–Wilk *W* test) prior to determination of interferential statistics.

For contaminant classes that comprised a mixture of individual compounds, contaminant patterns were investigated using the percent (%) composition of the major individual compounds to $\sum_{15}$PCB, $\sum_{15}MeSO_2$-PCB, ∑CHL, ∑DDT or $\sum_4$PBDE (specific congeners listed in Fig. 2). The % composition values were examined by PCA to determine correlations between congener proportions and to reduce congener patterns to a small number of representative variables. Significant PCs were considered those with an eigenvalue greater than one. Since most % composition values were normally distributed, PCA is quite robust to deviations from normality (Norman and Streiner, 2000), and to aid in subsequent interpretations, the analysis was performed on the untransformed values. The covariance matrix was used as all data were in the same units and had similar variance. To address possible factors contributing to variation in contaminant patterns, we performed a multiple regression of individual PC factor scores with year (considered as representative of changes in atmospheric sources and "weathering" of the contaminant residue pattern), biological group and the three dietary tracers ($\delta^{13}C$, FA-Index1 and FA-Index2). All statistical tests were deemed significant at $p<0.05$ unless otherwise indicated.

3. Results and discussion

3.1. Influence of biological factors on organohalogen concentrations

The influence of age and sex on organohalogen levels was assessed on ∑-contaminant classes except for HCH, as α- and β-HCH possess differing physico-chemical properties that lead to differences in environmental persistence and biomagnification (Braune et al., 2005). The proportion of subadults, adult females and adult males varied among years (Table 1). The only demographic difference was a tendency ($p = 0.09$) towards higher ∑CHL levels in subadults than in adult males (in 2007). With a larger circumpolar data set, Norstrom et al. (1998) previously reported higher levels of ∑CHL, but not ∑PCB, *p,p′*-DDE and dieldrin, in cubs/subadults (0–4 years old) than in adults (>4 years old). That study and two smaller studies on Svalbard and East Greenland bears (Bernhoft et al., 1997; Dietz et al., 2004) found higher ∑CHL levels in adult females than in adult males. Polischuk et al. (2002) reported that subadult and adult male polar bear body burdens for ∑CHL (driven by the metabolite oxychlordane) declined significantly during fasting, whereas those in females did not. The mechanism by which males are able to effectively metabolize CHLs is unknown, but may be related to male-specific cytochrome P450s or higher induction (or inducibility) of specific enzymes. ∑PCB, ∑CHL and ∑DDT levels in males were adjusted to female equivalents prior to analysis, as they have consistently shown sex-related concentration differences in larger data sets (Norstrom et al., 1998). Both before and after sex-adjustment, demographic differences in ∑PCB, ∑CHL and ∑DDT levels were generally small (1–18%) and were within the range of the analytical variation (7–21% between duplicates), but sex-adjustment generally reduced within-year contaminant concentration variation. All other OCPs, $MeSO_2$-PCBs and PBDEs were not sex-adjusted and showed no demographic differences except for mirex levels, which were slightly higher (13%) in females than males (in 2003).

To determine contaminant temporal trends, we wanted to assess confounding inter- as well as intra-annual variation. ANCOVA results (Table 2) generally showed significant effects of year for contaminants that also demonstrated significant time trends (i.e., in the PIA analysis) (Tables 3–5). That is, the time trends were not generally confounded by biological group differences. The exceptions to this were ∑$MeSO_2$-PCB and dieldrin levels, which were not significant with year by ANCOVA, but showed significantly increasing and decreasing time trends, respectively. ∑$MeSO_2$-PCB and dieldrin trends (Tables 4 and 5) were thus confounded by between-year biological differences, and should be regarded with caution. For ∑PCB, ∑PBDE and α-HCH levels, although temporal trends were consistent, biological group and the interaction was also significant by ANCOVA (Table 2). Thus, the magnitude of the reported trends may have been influenced by biological group, and the annual median values were additionally reported separately for each biological group for years not exclusively comprised of adult female samples, and temporal trends were also calculated on adult females only (Tables 3 to 5). These additional values were also provided for ∑CHL and ∑DDT for reference (e.g., Norstrom et al., 1998). However, they should be treated with caution due to very low adult female sample sizes in 2001, 2003 and 2007 ($n = 3$, 3 and 1, respectively).

Table 2
Results (p-values[a]) from an analysis of covariance (ANCOVA) testing the influence of biological group (adult female/adult male/subadult) and year on adipose organohalogen concentrations in polar bears from western Hudson Bay, 1991–2007.

Contaminant	Biological group	Year	Biological group × year interaction term
∑PBDE	<0.001*	0.01*	<0.001*
∑PCB	0.02*	0.39	0.02*
∑$MeSO_2$-PCB	0.06	0.39	0.06
∑ClBz	0.46	0.76	0.46
α-HCH	0.01*	<0.001*	0.01*
β-HCH	0.07	0.05*	0.07
∑CHL	0.71	0.35	0.70
∑DDT	0.25	0.01*	0.25
Dieldrin	0.29	0.21	0.29
∑Mirex	0.08	0.63	0.08

[a] Statistical significance ($p<0.05$) is indicated by an asterisk.

3.2. Legacy contaminant levels and trends

In all years, the highest level legacy contaminants were ∑PCB followed by ∑CHL (both >1 ppm) with at least an order of magnitude lower levels of ∑DDT, ∑ClBz, ∑HCH, ∑$MeSO_2$-PCB, dieldrin, ∑mirex and OCS (Tables 4 and 5). The 3-$MeSO_2$-*p,p′*-DDE metabolite was not consistently detected. Levels of α-HCH and ∑DDT decreased and β-HCH increased (log-linearly) from 1991 to 2007. There were no clear temporal trends for ∑PCB, ∑ClBz, OCS, ∑CHL and ∑mirex. Since changes in dieldrin and ∑$MeSO_2$-PCB levels were related to between-year biological group differences, they likely showed no significant time trends as well. The power to detect an annual concentration change of 5% with the number of years in the current time series was generally only between 13% and 27%, although above 60% for ∑CHL and ∑mirex. This low power was similar to other time series in a recent meta-analysis of temporal trends in Arctic biota (Rigét et al., 2010). Henriksen et al. (2001) found the most effective action to improve statistical power would be annual sampling. Nonetheless, combining existing data sets to provide long-term series may clarify or substantiate increasing or decreasing time trends. The current results were compared to temporal trends of PCBs and OCPs in WHB polar bears from 1968 to 2002 (Fig. 1; Braune et al., 2005; Norstrom 2001; Verreault et al., 2005). Generating quantitative overall trends was not warranted due to possible variation from differences in pre-analysis storage times, extraction and analytical techniques and data analysis (de Solla et al., 2010). Nonetheless, when our 1990s data overlapped with the previous data set, the same-year means generally agreed (Fig. 1). The major differences were for α-HCH and ∑ClBz, likely due to greater analytical variation from volatility-related recovery losses relative to other OCPs.

Over the four decades comprising the combined studies, there was a five-fold increase in β-HCH levels (Fig. 1). Decreasing β-HCH levels have been observed in other northern biota, but slower declines or increases have been observed west of the European Arctic (Rigét et al., 2010). Although divergent same-year means prevented a long-term assessment, α-HCH levels declined in this and previous polar bear studies, and thus likely declined overall. The difference in α-HCH and β-HCH trends has resulted in a substantial shift in ∑HCH composition in WHB polar bears, from mainly α-HCH in 1968 (>80%) to mainly β-HCH in 2007 (78%). This pattern has been reported in other northern wildlife (Rigét et al., 2010). It is related to greater partitioning of β-HCH into water resulting in the slower arrival to higher latitudes, and to α-HCH being more readily metabolized and thus less bioaccumulative. We previously found that the rate of β-HCH increase (+8.3%/year) from 1991 to 2007, which was higher than all other increasing β-HCH trends reviewed by Rigét et al. (2010), was in part related to sea ice-associated diet/food web change (McKinney et al., 2009). This change did not influence the decline rate (−11%/year) of the less recalcitrant α-HCH, which was similar to decline rates reported in other species (Rigét et al., 2010).

Levels of ∑DDT have declined by an order of magnitude over the past four decades. From 1991 to 2007, the measured decline was fairly consistent across DDT compounds (Table 5). The rate of decline of ∑DDT (−9.6%/year) was more than double the mean rate measured in other northern biota (−4.4%/year; Rigét et al., 2010). We previously showed that this decline would have been slower if diet/food web changes had not occurred in WHB bears (McKinney et al., 2009). More rapid decline than in other regions may also have been related to local spraying of DDT in the 1950s and 1960s and more immediate subsequent declines (de Wit et al., 2004).

∑CHL levels showed no directional change from 1968 to 2007 (Fig. 1). Levels were highest in the early 1980s, but our data and the

Table 3
Concentrations (ng/g lipid weight) and temporal trends of BFRs detected[a] in adipose of polar bears from western Hudson Bay, 1991–2007.

	Annual median concentration (range)							Temporal trends				
Contaminant (Br_x)	1991	1992	1994	1995	2001	2003	2007	Trend[b]	Annual % change	95% CI	r^2	p[c]
$\sum_{37}$PBDE	6.8 (3.4–10.1)	7.1 (5.1–12.0)	10.3 (5.8–20.4)	10.0 (7.0–19.9)	29.3 (19.3–56.5)	50.8 (26.1–194)	38.8 (18.1–120)	↑ (L)	+13	+8.6 to +18	0.91	<0.001*
Adult female					34.5 (26.1–46.2)	57.9 (47.4–81.2)	120	↑ (L)	+19	+16 to +21	0.99	<0.001*
Adult male					–	42.8 (26.1–194)	42.7 (22.5–53.0)					
Subadult					27.8 (19.3–56.5)	–	34.7 (18.1–116)					
Geometric mean (95% CI)	6.6 (5.7–7.6)	7.3 (6.4–8.3)	10.5 (8.7–12.6)	10.9 (9.4–12.8)	32.2 (25.4–40.8)	53.0 (38.7–72.4)	42.1 (30.2–58.6)					
$\sum_4$PBDE	6.1 (3.4–9.1)	6.4 (4.7–8.9)	9.6 (5.2–18.8)	9.6 (6.5–18.0)	27.9 (17.5–51.7)	41.2 (23.5–172)	35.4 (16.7–115)	↑ (L)	+13	+8.8 to +17	0.93	<0.001*
Geometric mean (95% CI)	5.9 (5.2–6.8)	6.4 (5.7–7.2)	9.5 (7.9–11.5)	10.0 (8.7–11.6)	30.1 (23.8–38.1)	44.1 (32.8–59.6)	38.5 (27.5–54.0)					
BDE28/33 (Br_3)	(<0.05–0.3)	0.1 (<0.05–0.8)	(<0.05–1.3)	(<0.05–0.8)	0.3 (0.1–0.8)	0.4 (<0.05–3.5)	0.1 (<0.05–0.5)	NC				
BDE54 (Br_4)	(<0.05)	(<0.05–0.8)	(<0.05)	(<0.05)	(<0.05–0.2)	(<0.05)	(<0.05–2.5)	NC				
BDE47 (Br_4)	3.1 (1.9–4.0)	3.8 (2.7–5.4)	4.9 (2.6–8.7)	5.8 (3.8–10.1)	16.3 (9.2–22.0)	10.4 (4.3–60.4)	16.9 (6.7–36.6)	↑ (L)	+13	+8.8 to +17	0.93	<0.001*
BDE100 (Br_5)	0.3 (0.1–0.5)	0.5 (0.2–1.3)	0.7 (0.3–1.9)	0.4 (0.2–0.9)	1.6 (1.0–2.9)	2.8 (1.8–17.0)	1.8 (0.7–8.0)	↑ (L)	+13	+5.7 to +20	0.81	0.006*
BDE99 (Br_5)	0.8 (<0.05–2.0)	0.6 (0.3–1.2)	0.9 (0.4–2.0)	0.7 (0.2–1.9)	2.9 (1.7–5.0)	3.2 (1.6–62.3)	3.0 (1.0–13.1)	↑ (L)	+12	+6.4 to +17	0.86	0.003*
BDE116 (Br_5)	0.6 (<0.05–0.9)	0.8 (0.4–1.3)	0.7 (0.5–1.6)	0.5 (<0.05–2.1)	0.7 (0.5–2.3)	1.9 (1.1–4.8)	0.6 (<0.05–3.3)		+3.3	−3.5 to +10	0.24	0.27
BDE154 (Br_6)	(<0.4)	(<0.4)	(<0.4)	(<0.4)	0.6 (<0.4–1.9)	(<0.4–2.5)	0.6 (<0.4–3.0)	NC				
BDE153 (Br_6)	2.1 (1.2–3.1)	1.4 (0.9–2.5)	2.8 (1.9–6.3)	2.7 (1.2–5.1)	9.4 (5.4–26.0)	27.7 (14.7–55.5)	13.4 (3.8–69.2)	↑ (L)	+17	+8.2 to +25	0.84	0.004*
BDE208 (Br_9)	(<0.05)	(<0.05)	(<0.05)	(<0.05)	(<0.05)	(<0.05–1.9)	(<0.05)	NC				
BDE207 (Br_9)	(<0.05)	(<0.05)	(<0.05)	(<0.05)	(<0.05)	(<0.05–10.2)	(<0.05)	NC				
BDE209 (Br_{10})	(<1)	(<1)	(<1)	(<1)	(<1)	(<1–26.2)	(<1–10.2)	NC				
PBEB (Br_5)	(<0.05)	0.1 (<0.05–1.4)	0.1 (<0.05–0.3)	(<0.05)	(<0.05–0.1)	0.1 (<0.05–0.9)	(<0.05–1.7)	NC				
HBB (Br_6)	(<0.05)	0.2 (<0.05–3.4)	(<0.05–0.5)	(<0.05)	(<0.05)	0.1 (<0.05–2.6)	(<0.05–2.6)	NC				
BB101 (Br_5)	0.2 (<0.05–0.5)	0.6 (0.3–1.9)	0.6 (0.3–1.7)	(<0.05–1.7)	0.2 (<0.05–1.2)	1.6 (0.6–4.5)	(<0.05–0.7)	NC				
BB153/BDE154 (Br_6)	17.9 (9.4–32.5)	31.1 (18.7–49.2)	41.7 (30.2–102.5)	16.4 (7.8–57.3)	24.9 (12.0–75.7)	130.9 (52.2– 334)	34.9 (6.0–144)		+5.5	−6.0 to +17	0.23	0.27
Total-(α)-HBCD (Br_6)	(<0.3)	(<0.3–4.0)	(<0.3–15.4)	(<0.3)	3.9 (<0.3–6.8)	11.7 (6.1–287)	3.1 (1.3–20.4)	NC				
Geometric mean (95% CI)	( )	( )	( )	( )	2.9 (1.6–4.9)	14.9 (8.3–27.0)	3.9 (2.3–6.9)					
DBDPE (Br_{10})	(<1)	(<1)	(<1)	(<1)	(<1)	(<1)	(<1–2.0)	NC				

[a] BFRs monitored, but below MLOQ: PBT (co-elutes with BDE28/33) and BTBPE.
[b] Trends reported as ↓ (L) = (log-)linear decrease; ↑ (L) = (log-)linear increase; ↓(N-L) = non-linear decrease; ↑ (N-L) = non-linear increase; ↕ = non-monotonic trend; — = no trend; NC = not calculated.
[c] Statistical significance (p<0.05) is indicated by an asterisk.

Table 4
Concentrations (ng/g lipid weight) and temporal trends of PCBs and $MeSO_2$-PCBs in adipose of polar bears from western Hudson Bay, 1991–2007.

Contaminant (Cl_x)	Annual median concentration (range)							Temporal trends				
	1991	1992	1994	1995	2001	2003	2007	Trend[a]	Annual % change	95% CI	r^2	p[b]
$\sum_{74}$PCB	3519 (2088–5299)	3381 (2118–5873)	3415 (5126–11286)	2899 (1673–6304)	3745 (2279–10585)	5250 (2873–11892)	3652 (1200–19868)		+1.6	−1.3 to +4.5	0.29	0.21
Adult female					3745 (3556–10585)	7578 (6156–11892)	8319	↑ (N-L)	+6.0	+1.8 to +10	0.73	0.01
Adult male-adjusted					–	4581 (2873–8095)	3526 (1430–5894)					
Subadult					3628 (2279–9800)	–	3608 (1200–19868)					
Geometric mean (95% CI)	3448 (2996–3970)	3412 (2948–3950)	3591 (2913–4426)	3144 (2650–3729)	4392 (3067–6287)	5435 (4298–6874)	3589 (2264–5689)					
AMAP $\sum_{10}$PCB	2413 (1438–3707)	2267 (1437– 4052)	2397 (1441–7945)	1984 (1112– 4326)	2729 (1647– 7572)	3871 (2118– 8580)	2559 (863–14878)		+2.0	−1.2 to +5.2	0.33	0.17
CB47/48 (Cl_4)	28.1 (17.4–56.3)	29.5 (17.0–56.1)	30.4 (17.8–86.0)	28.1 (18.1–48.3)	21.5 (14.3–58.4)	19.1 (11.0– 54.7)	16.6 (8.5–61.6)	↓ (L)	−3.8	−5.0 to −2.6	0.93	<0.001*
CB99 (Cl_5)	386 (225–701)	379 (230–697)	421 (243–1595)	350 (235–701)	405 (281–1377)	467 (274–918)	387 (153–1995)		+0.6	−1.0 to +2.2	0.14	0.41
CB85 (Cl_5)	34.3 (22.6–54.0)	34.7 (21.3–63.1)	32.5 (19.5–107)	36.0 (24.1–59.5)	27.8 (13.9–54.8)	19.5 (9.3–39.1)	16.9 (8.6–50.3)	↓ (N-L)	−4.7	−6.7 to −2.7	0.88	0.002*
CB118 (Cl_5)	52.3 (35.4–60.9)	71.5 (44.0–111)	56.0 (36.3–113)	57.2 (38.2–95.2)	41.9 (26.0–65.0)	23.6 (16.4–77.1)	25.8 (6.2–90.2)	↓ (L)	−6.3	−9.7 to −2.8	0.82	0.006*
CB146 (Cl_6)	44.6 (32.9–63.3)	50.8 (32.4–86.9)	46.8 (27.2–121)	46.9 (37.7–69.7)	40.2 (25.2–83.6)	43.5 (23.2–70.2)	27.3 (13.4–104)	↓ (L)	−2.8	−5.0 to −0.5	0.67	0.03*
CB153 (Cl_6)	1393 (832–2231)	1262 (791–2318)	1406 (800– 5125)	1153 (600–2662)	1762 (1088– 5241)	2612 (1486– 5525)	1761 (524–10498)		+3.3	−0.2 to +6.8	0.55	0.06
CB130 (Cl_6)	29.3(17.1–45.3)	26.8 (18.5–48.2)	29.8 (15.0–97.9)	26.2 (15.7–49.6)	29.4 (19.0–78.8)	29.3 (16.8–63.6)	24.1 (8.0–111)		−0.5	−1.9 to +0.9	0.14	0.41
CB138 (Cl_6)	354 (219–551)	363 (238–634)	370 (193–1222)	353 (221–571)	323 (204–831)	338 (190–613)	266 (95.3–1049)	↓ (L)	−1.6	−2.7 to −0.5	0.74	0.01*
CB156 (Cl_6)	21.9 (12.2–32.5)	23.3 (13.9–44.9)	22.6 (12.8–56.3)	19.4 (10.3–27.1)	24.9 (19.5–71.6)	41.7 (20.4–72.1)	27.9 (8.7–110)		+2.7	−0.8 to +6.2	0.45	0.10
CB187/182 (Cl_7)	18.8 (13.4–27.6)	20.2 (12.3–34.3)	18.7 (10.3–40.0)	17.4 (14.4–31.1)	16.5 (6.8–23.9)	10.4 (6.0–25.5)	9.9 (5.0–40.0)	↓ (L)	−4.4	−6.6 to −2.2	0.84	0.004*
CB183 (Cl_7)	34.1 (20.3–55.0)	28.7 (18.9–49.8)	32.0 (14.0–106)	28.7 (16.7–50.8)	32.4 (16.3–63.5)	25.0 (13.6–78.9)	20.7 (6.5–92.2)	↓ (L)	−2.2	−4.3 to −0.2	0.60	0.04*
CB180 (Cl_7)	490 (310–819)	495 (293–888)	479 (323–1451)	391 (161–1190)	536 (273–1543)	847 (371–2345)	453 (121–3154)		+1.3	−3.0 to +5.6	0.11	0.46
CB170/190 (Cl_7)	212 (136–361)	194 (109–369)	179 (130–571)	162 (69.7–625)	228 (137–792)	462 (178–1074)	220 (63.8–1713)		+2.9	−2.6 to +8.4	0.27	0.23
CB194 (Cl_8)	103 (58.4–198)	105 (74.8–189)	118 (80.0–294)	94.9 (41.0–363)	88.3 (41.0–379)	211 (54.9–658)	71.2 (14.6–593)	↕	−0.3	−6.7 to +6.1	<0.01	0.88
CB206 (Cl_9)	38.0 (11.2–66.7)	34.7 (20.9–59.8)	37.4 (23.5–96.9)	26.6 (14.6–90.8)	22.2 (9.4–113)	44.3 (12.2–200)	18.5 (2.4–93.0)		−2.9	−8.0 to +2.2	0.29	0.21
$\sum MeSO_2$-PCB	153 (87.9–396)	165 (83.9–332)	131 (86.1–568)	158 (101–344)	235 (111–621)	402 (272–875)	314 (130–821)	↑ (L)	+6.0	+2.3 to +9.7	0.77	0.01*
Geometric mean (95% CI)	157 (127–194)	174 (144–210)	151 (119–191)	164 (139–193)	254 (173–373)	448 (361–557)	274 (195–386)					
3′-$MeSO_2$-CB49 (Cl_4)	11.0 (6.3–25.2)	14.0 (7.4–23.7)	11.7 (8.3–39.3)	12.0 (7.2–18.0)	13.3 (8.0–32.7)	21.8 (11.9–41.6)	18.9 (7.4–37.5)		+0.5	−1.8 to +2.8	0.06	0.60
4′-$MeSO_2$-CB49 (Cl_4)	10.9 (6.1–25.2)	13.3 (6.8–30.3)	10.5 (7.1–36.1)	10.7 (7.7–17.1)	12.1 (7.6–37.7)	25.6 (17.2–57.3)	15.4 (6.4–38.3)		+3.3	−1.3 to +7.9	0.40	0.13
4-$MeSO_2$-CB64 (Cl_4)	5.5 (3.5–21.0)	5.7 (3.7–11.3)	5.2 (2.4–23.9)	5.2 (4.4–9.8)	7.7 (3.7–17.5)	10.9 (5.9–17.5)	8.7 (4.9–22.9)	↑ (L)	+4.2	+1.4 to +7.0	0.75	0.01*
3-$MeSO_2$-CB70 (Cl_4)	5.3 (3.0–13.1)	6.1 (2.9–13.2)	4.7 (3.4–18.5)	4.9 (3.4–9.3)	6.6 (3.5–15.3)	12.1 (7.0–25.7)	10.0 (3.6–19.5)	↑ (L)	+4.9	+1.1 to +8.7	0.69	0.02*
4-$MeSO_2$-CB70 (Cl_4)	5.9 (3.9–17.3)	7.0 (3.3–18.4)	5.3 (3.9–20.5)	6.5 (4.1–9.4)	8.4 (4.4–21.8)	17.0 (9.7–24.2)	9.7 (5.3–27.2)		+4.8	−0.1 to +9.7	0.56	0.05
3′-$MeSO_2$-CB87 (Cl_5)	10.9 (6.0–29.2)	11.8 (5.7–33.7)	10.2 (5.3–49.5)	11.4 (7.2–34.0)	18.0 (8.4–49.3)	45.7 (26.5–92.0)	19.7 (9.4–62.8)		+6.5	+0.0 to +13	0.57	0.05
4′-$MeSO_2$-CB87 (Cl_5)	26.9 (15.2–72.0)	28.4 (12.3–76.6)	20.0 (11.6–92.8)	27.3 (15.9–79.2)	43.9 (18.3–130)	87.5 (55.0–222)	48.6 (16.3–164)	↑ (L)	+6.4	+0.6 to +12	0.62	0.04*
4-$MeSO_2$-CB91 (Cl_5)	4.1 (2.4–10.4)	4.0 (2.2–7.3)	3.7 (2.4–14.2)	4.4 (2.4–8.0)	5.5 (2.8–15.9)	7.2 (5.4–11.3)	6.3 (2.5–19.3)	↑ (N-L)	+3.8	+1.6 to +5.9	0.81	0.007*
3′-$MeSO_2$-CB101 (Cl_5)	17.2 (9.3–41.6)	17.6 (7.9–34.2)	16.2 (9.2–75.9)	19.6 (12.4–36.6)	27.3 (12.9–76.7)	34.5 (19.0–70.1)	36.4 (14.7–83.1)	↑ (N-L)	+5.4	+3.8 to +7.0	0.93	<0.001*
4′-$MeSO_2$-CB101 (Cl_5)	14.1 (8.0–36.2)	17.8 (9.3–36.4)	13.1 (7.9–63.8)	15.8 (9.2–42.2)	29.9 (11.8–81.9)	59.0 (35.5–139)	40.1 (15.5–126)	↑ (L)	+8.3	+3.2 to +13	0.78	0.009*
4-$MeSO_2$-CB110 (Cl_5)	4.4 (2.1–16.7)	5.1 (2.2–10.6)	4.0 (2.4–17.5)	5.8 (3.5–8.5)	8.1 (5.0–18.4)	9.4 (4.9–19.5)	7.9 (3.8–20.0)	↑ (L)	+4.7	+1.6 to +7.8	0.75	0.01*
4′-$MeSO_2$-CB132 (Cl_6)	5.6 (2.8–12.6)	5.2 (3.0–12.6)	4.1 (2.8–14.9)	4.8 (3.1–8.9)	7.5 (3.6–20.1)	12.0 (8.4–25.8)	8.4 (2.7–24.8)		+4.9	+0.7 to +9.2	0.64	0.03
4′-$MeSO_2$-CB141 (Cl_6)	3.2 (1.5–7.8)	4.4 (1.3–7.7)	2.5 (1.5–13.2)	3.2 (1.7–10.8)	6.3 (2.5–18.3)	11.2 (5.3–33.1)	6.4 (2.3–23.5)	↑ (L)	+6.6	+0.2 to +13	0.59	0.04*
3-$MeSO_2$-CB149 (Cl_6)	3.1 (1.9–5.7)	2.9 (1.7–4.1)	2.6 (1.5–7.3)	2.9 (2.3–4.5)	3.1 (2.3–7.1)	2.4 (1.3–3.7)	3.4 (1.2–6.1)	↕	+0.3	−1.9 to +2.5	0.03	0.72
4-$MeSO_2$-CB149 (Cl_6)	16.5 (10.2–38.5)	15.1 (9.5–24.8)	14.6 (8.5–47.9)	15.5 (10.5–26.4)	19.9 (10.0–61.2)	18.2 (13.1–29.8)	21.8 (8.6–67.9)	↑ (L)	+2.2	+0.9 to +3.5	0.79	0.008*

[a] Trends reported as ↓ (L) = (log-)linear decrease; ↑ (L) = (log-)linear increase; ↓(N-L) = non-linear decrease; ↑ (N-L) = non-linear increase; ↕ = non-monotonic trend; — = no trend.
[b] Statistical significance ($p<0.05$) is indicated by an asterisk.

Table 5
Concentrations (ng/g lipid weight) and temporal trends of organochlorine pesticides in adipose of polar bears from western Hudson Bay, 1991–2007.

Contaminant	Annual median concentration (range)							Temporal trends				
	1991	1992	1994	1995	2001	2003	2007	Trend[a]	Annual % change	95% CI	r^2	p[b]
∑ClBz	210 (143–634)	287 (176–540)	216 (167–443)	186 (140–266)	202 (144–378)	242 (138–779)	186 (149–569)		−0.92	−3.6 to +1.8	0.13	0.43
Geometric mean (95% CI)	217 (190–248)	298 (256–347)	228 (198–264)	186 (172–201)	210 (167–201)	259 (201–334)	221 (174–279)					
1,2,4,5-TetraClBz	118 (73.8–229)	150 (87.5–239)	120 (96.0–315)	113 (86.8–165)	96.1 (60.5–208)	138 (84.1–212)	86.3 (32.9–207)		−1.8	−4.8 to +1.1	0.34	0.17
1,2,3,4-TetraClBz	(<0.1)	(<0.1–0.5)	(<0.1–0.3)	(<0.1)	(<0.1)	0.1 (<0.1–0.3)	(<0.1–0.5)	NC				
PentaClBz	29.0 (19.4–47.4)	39.8 (21.4–54.1)	30.1 (24.5–47.5)	28.5 (21.9–40.8)	18.8 (14.4–42.8)	27.8 (13.2–60.9)	22.9 (9.2–48.5)		−2.6	−5.8 to +0.6	0.46	0.09
HCB	63.8 (49.8–191)	76.8 (52.7–406)	60.2 (44.0–270)	43.6 (30.8–67.7)	72.3 (50.4–157)	65.3 (40.8–545)	85.9 (48.7–487)		+1.6	−2.1 to +5.2	0.19	0.32
α-HCH	252 (181–374)	312 (254–410)	256 (189–337)	255 (193–370)	108 (68.0–131)	102 (40.0–132)	48.5 (18.2–105)	↓ (L)	−11	−14 to −7.9	0.94	<0.001*
Adult female					116 (82.2–130)	105 (40.0–132)	105	↓ (L)	−7.6	−11 to −4.6	0.82	0.006*
Adult male					–	102 (52.5–113)	47.1 (18.2–54.5)					
Subadult					97.3 (68.0–131)	–	60.5 (35.1–72.9)					
Geometric mean (95% CI)	253 (228–280)	318 (292–346)	253 (231–277)	253 (233–275)	98.6 (84.2–115)	85.4 (69.7–105)	48.8 (38.1–62.8)					
β-HCH	51.2 (29.5–113)	62.5 (47.4–110)	64.4 (54.7–163)	71.6 (48.1–94.6)	93.3 (53.3–237)	192 (118–441)	187 (76.3–313)	↑ (L)	+8.3	+5.2 to +11	0.90	<0.001*
Geometric mean (95% CI)	49.8 (41.7–59.6)	67.9 (60.0–76.9)	71.1 (61.3–82.4)	69.2 (63.2–75.8)	101 (71.5–143)	200 (159–252)	175 (139–221)					
OCS	9.9 (8.0–13.4)	12.7 (8.4–25.4)	11.2 (7.3–20.6)	9.0 (6.2–13.5)	8.0 (6.4–21.6)	11.0 (7.3–23.1)	8.7 (4.5–30.0)		−1.5	−4.1 to +1.2	0.28	0.22
Geometric mean (95% CI)	10.2 (9.3–11.1)	12.4 (10.9–14.1)	11.2 (9.8–12.9)	9.0 (8.1–10.1)	9.0 (6.9–11.6)	11.8 (9.7–14.4)	8.9 (6.4–12.3)					
∑CHL	2608 (1652–4344)	2710 (1661–4781)	2584 (1680–6080)	2445 (1471–4596)	2976 (1629–6282)	2505 (1497–5533)	2697 (1355–10012)	↕	+0.2	−0.9 to +1.4	0.05	0.88
Adult female					3052 (2090–3473)	4853 (4310–5533)	3612		+3.1	+0.2 to +6.0	0.60	0.04
Adult male-adjusted					–	1764 (1497–3454)	2157 (1355–2923)					
Subadult					2814 (1629–6282)	–	2891 (1851–10012)					
Geometric mean (95% CI)	2601 (2256–2998)	2740 (2388–3143)	2701 (2260–3227)	2562 (2212–2967)	2833 (2186–3671)	2588 (1978–3388)	2673 (1991–3591)					
Heptachlor epoxide	327 (219–598)	371 (237–622)	345 (248–951)	334 (223–650)	336 (208–681)	360 (211–657)	331 (190–952)	↕	−0.1	−1.0 to +0.7	0.02	0.73
Oxychlordane	1852 (1130–3309)	1916 (1145–3485)	1890 (1165–4385)	1656 (987–3316)	2253 (1287–5268)	1378 (686–4409)	1871 (923–8731)	↕	−0.4	−3.2 to +2.5	0.02	0.74
t-Chlordane	7.3 (4.7–14.1)	9.1 (4.9–17.7)	8.5 (5.4–12.4)	7.2 (4.5–13.0)	9.1 (4.3–28.6)	3.8 (1.8–26.0)	4.8 (2.0–48.9)		−3.8	−8.4 to +0.9	0.47	0.09
c-Chlordane	31.1 (22.3–44.1)	30.5 (20.6–53.8)	30.0 (16.9–82.4)	27.7 (16.4–50.1)	25.4 (13.7–53.2)	26.6 (12.3–49.3)	28.1 (15.5–80.2)	↕	−0.9	−1.9 to +0.1	0.52	0.07
t-Nonachlor	334 (265–430)	373 (245–591)	332 (224–880)	334 (226–606)	177 (107–534)	245 (105–618)	202 (143–805)	↓ (L)	−4.1	−6.9 to −1.3	0.74	0.01*
c-Nonachlor	10.9 (4.2–14.2)	11.6 (8.9–34.8)	9.2 (6.2–21.8)	10.0 (7.5–16.2)	3.7 (1.6–12.6)	4.5 (0.6–12.4)	2.9 (1.0–53.0)	↓ (L)	−9.2	−12 to −6.4	0.94	<0.001*
∑DDT	317 (194–490)	438 (319–783)	333 (194–814)	403 (311–706)	132 (57.4–278)	116 (44.3–259)	105 (19.0–540)	↓ (L)	−9.6	−14 to −5.4	0.87	0.003*
Adult female					132 (98.9–239)	142 (48.6–259)	125	↓ (L)	−8.4	−13 to −4.1	0.84	0.004*
Adult male-adjusted					–	90.9 (44.3–234)	81.4 (19.0–122)					
Subadult					114 (57.4–278)	–	123 (45.6–540)					
Geometric mean (95% CI)	329 (285–380)	461 (402–529)	378 (298–480)	431 (379–490)	134 (93.0–194)	113 (80.9–158)	89.9 (56.1–145)					
p,p′-DDE	261 (171–411)	384 (289–726)	310 (177–768)	361 (274–642)	103 (53.8–255)	105 (19.1–241)	91.8 (12.2–491)	↓ (L)	−9.7	−15 to −4.7	0.83	0.005*
p,p′-DDD	8.1 (2.6–14.4)	5.4 (3.1–17.6)	3.0 (1.6–5.8)	7.1 (4.2–12.8)	1.9 (0.5–6.0)	1.5 (0.3–3.8)	1.6 (0.2–18.6)	↓ (L)	−11	−17 to −4.2	0.79	0.008*
p,p′-DDT	46.5 (20.3–78.5)	39.8 (22.3–58.8)	24.8 (13.2–67.5)	42.3 (16.7–76.7)	10.6 (3.0–26.8)	8.4 (2.3–28.4)	6.7 (1.9–31.4)	↓ (L)	−13	−17 to −8.8	0.93	<0.001*
3-MeSO$_2$-*p,p′*-DDE	– (<0.9–2.1)	1.3 (<0.9–3.5)	– (<0.9–4.0)	2.3 (<0.9–3.2)	– (<0.9–1.9)	– (<0.9–3.0)	– (<0.9–3.9)	NC				
Geometric mean (95% CI)	– (–)	1.2 (0.6–1.8)	– (–)	1.7 (1.1–2.5)	– (–)	– (–)	– (–)					
Dieldrin	253 (175–356)	276 (211–556)	249 (184–730)	221 (157–387)	209 (132–306)	211 (108–652)	216 (133–751)	↓ (N-L)	−1.4	−2.6 to −0.3	0.66	0.03*
Geometric mean (95% CI)	253 (228–281)	294 (252–343)	281 (233–339)	224 (198–253)	201 (164–245)	229 (175–299)	244 (185–322)					
∑Mirex	38.6 (29.2–63.9)	40.2 (26.8–72.9)	44.5 (20.8–131)	39.3 (28.5–80.7)	48.8 (25.8–94.5)	53.3 (33.7–148)	44.7 (18.5–177)		+1.4	−0.2 to +3.0	0.52	0.07
Geometric mean (95% CI)	40.8 (36.2–46.0)	40.5 (35.2–46.6)	46.5 (36.7–59.2)	42.2 (35.7–49.9)	48.7 (37.0–64.2)	58.8 (46.7–74.1)	50.2 (34.5–73.5)					
Photomirex	29.7 (25.5–51.1)	31.2 (22.3–56.4)	34.2 (16.4–106)	31.9 (23.0–68.1)	40.4 (21.2–75.3)	41.6 (25.1–111)	36.4 (15.8–141)	↑ (L)	+1.7	+0.2 to +3.1	0.64	0.03*
Mirex	8.8 (3.6–15.3)	8.7 (4.5–16.5)	10.3 (4.5–25.1)	7.4 (5.3–15.0)	8.4 (4.6–19.6)	10.8 (7.9–36.2)	9.0 (2.8–35.5)		+0.5	−1.8 to +2.8	0.06	0.60

[a] Trends reported as ↓ (L) = (log-)linear decrease; ↑ (L) = (log-)linear increase; ↓(N-L) = non-linear decrease; ↑ (N-L) = non-linear increase; ↕ = non-monotonic trend; — = no trend; NC = not calculated.
[b] Statistical significance ($p<0.05$) is indicated by an asterisk.

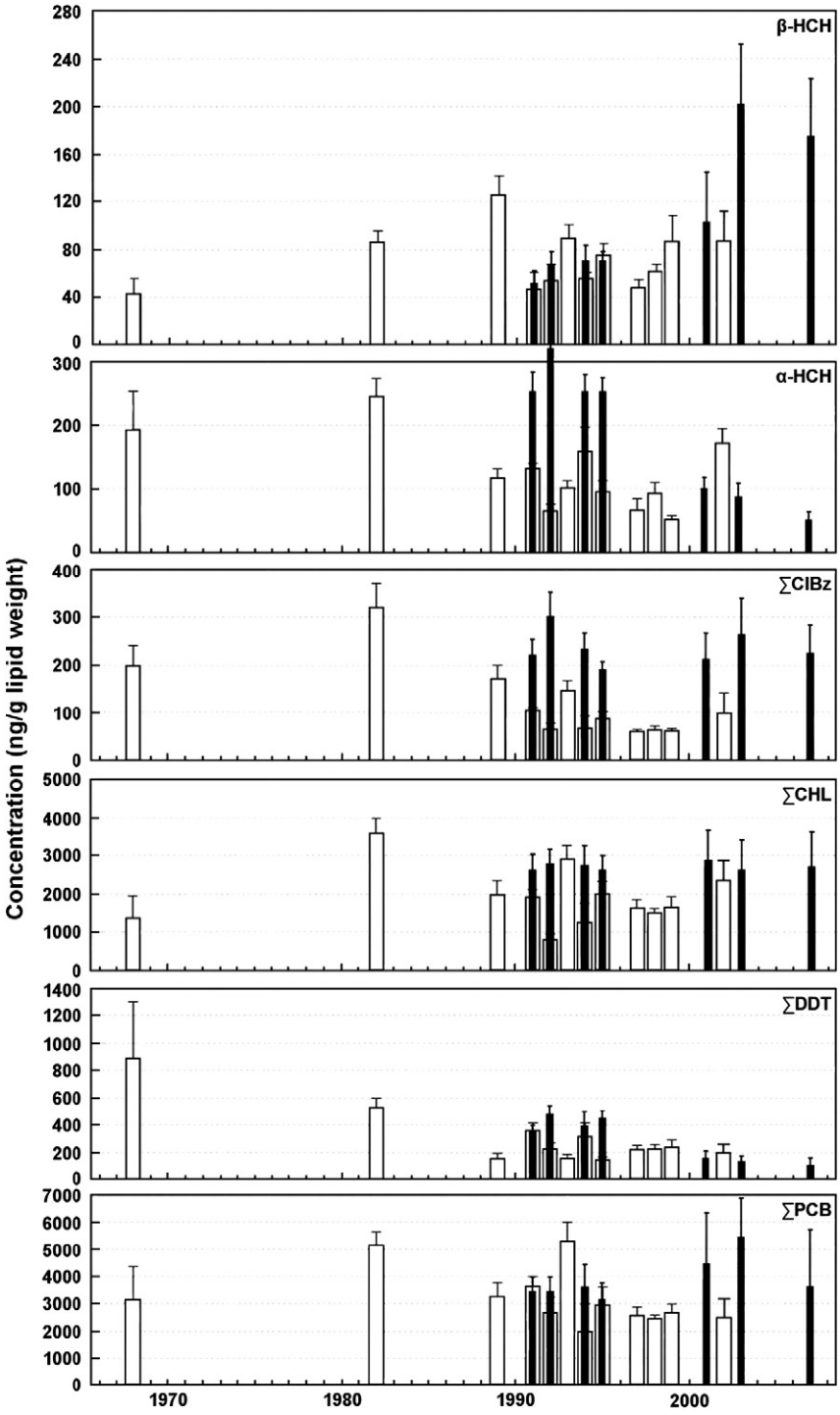

Fig. 1. Levels (geometric mean + 95% CI) of PCBs and organochlorine pesticides in adipose of polar bears from western Hudson Bay collected in 1968-2007. Figure adapted from de Wit et al. (2004) with permission. White bars are from de Wit et al. (2004) with original data from Norstrom (2001) and Verreault et al. (2005). Overlaid black bars are from the current study.

data from Verreault et al. (2005) indicated that there has been no unidirectional change over the past two decades. ∑CHL trends were not reflective of the behaviour of individual CHLs (Table 5). Within our data set, the major CHL compound was oxychlordane, followed by lower levels of heptachlor epoxide and *t*-nonachlor. Oxychlordane and heptachlor epoxide, which are CHL metabolites, appeared to drive the lack of trend in ∑CHL. In contrast, the parent compounds of oxychlordane, *t*-nonachlor and particularly *c*-nonachlor, both decreased significantly from 1991 to 2007. This increasingly weathered pattern suggested that recycled residues are responsible for sustained high inputs of CHL into this ecosystem. However, from 1986 to 2006, ∑CHL levels decreased in ringed seals from four Canadian locations including western Hudson Bay (Muir et al., 2007). Verreault et al. (2005) previously found lower ∑CHL levels in two-point temporal comparisons of all examined polar bear subpopulations except WHB. This discrepancy may be related to recent feeding changes, as we previously found that after diet-adjustment, WHB polar bear ∑CHL residues did in fact have a declining tendency (although not statistically significant) from 1991 to 2007 (McKinney et al., 2009). We hypothesized that the polar bear dietary tracer results indicated

increased proportional consumption of harbor and harp seals and a relative decrease in bearded seals. Bearded seals in other regions have been relatively less contaminated than other seals (e.g., Hoekstra et al., 2003). Harp seals are migratory and may be a vector for transporting a more contaminated North Atlantic organohalogen signature to WHB polar bears (McKinney et al., 2009). A recent study using stable nitrogen isotope ratios and mercury as dietary tracers found that harbor seals in western Hudson Bay feed at a higher trophic level than ringed and bearded seals (Young et al., 2010). Thus, proportionally less bearded seal and more harbor and/or harp seal consumption is consistent with the anomalous organohalogen time trends in WHB polar bears. This explanation remains speculative and could be addressed by the collection of dietary tracer and contaminant data in western Hudson Bay food web studies.

Along with $\sum$CHL, $\sum$PCB was the other organohalogen of most concern from a quantitative perspective. Levels of $\sum$PCB showed no obvious trends over the four decades (Fig. 1), other than a small and not significant increasing tendency from 1991 to 2007 (Table 4). The AMAP $\sum_{10}$PCB (sum of CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156 and CB180) results were similar (Table 4). After considering the influence of biological group, it is likely that $\sum$MeSO$_2$-PCB levels were temporally unchanging as well. This stagnant temporal trend for $\sum$PCB and $\sum$MeSO$_2$-PCB levels was not reflective of individual congener trends. The 15 highest concentration PCBs as well as MeSO$_2$-PCBs comprised on average 96% (range: 89–98%) and 94% (range: 89–98%) of $\sum_{74}$PCB and $\sum_{23}$MeSO$_2$-PCB, respectively (Table 4). Unchanged CB180 and CB194 levels paralleled the lack of $\sum$PCB trends, but levels of congeners such as CB118 and CB138 declined from 1991 to 2007. Levels of CB153 showed an increasing tendency ($p = 0.06$). These changes indicate a shift to a more weathered PCB signature, but similar to $\sum$CHL, there has been no concomitant drop in $\sum$PCB levels, possibly in part related to diet/food web changes.

Temporal trends in $\sum$ClBz levels were unclear. Norstrom (2001) reported declines in the 1990s. Similar to the current study, Verreault et al. (2005) found that 2002 levels were unchanged from the early 1990s. Individual ClBz levels were variable, but also showed non-significant temporal trends (Table 5). Consistent with our results, Norstrom (2001) did not detect a temporal change in dieldrin levels through the 1990s. Mirex and OCS levels were not reported in the earlier trend studies.

3.3. Flame retardant levels and trends

Adipose $\sum_{37}$PBDE levels increased from 1991 to 2007 (Table 3). $\sum_4$PBDE was also reported as only BDE47, BDE153, BDE99 and BDE100 were consistently detected, comprising on average 90% of $\sum_{37}$PBDE. These congeners and $\sum_4$PBDE increased similar to $\sum_{37}$PBDE. Lower PBDE levels in 2007 *cf.* 2003 (except for BDE47) may have indicated recently stabilizing or declining trends related to the late-2004 North American phaseout of pentaBDE and octaBDE. However, levels of certain legacy contaminants (e.g., $\sum$PCB) were also elevated in 2003 *cf.* 2001 and 2007, so apparent recent declines of $\sum$PBDE may partially be attributed to inter-annual variation rather than a directional change in trends. PBDE levels in western Hudson Bay ringed seal blubber increased from 1992 to 2006, but dropped in 2008 (Muir et al., 2009). To our knowledge, there are no published reports on PBDE temporal trends in other polar bear subpopulations, but ringed seals in other regions have been studied. There was no trend in East Greenland ringed seals from 1986 to 2004 (Rigét et al., 2006). Levels increased in western Canadian arctic ringed seals from 1981 to 2000, leveling off or declining in 2002 to 2003 (Ikonomou et al., 2005). Most temporal trends studies on Arctic air, sediment and biota have also demonstrated PBDE increases, but possible leveling off or declines in recent years (de Wit et al., 2010). Regardless, $\sum$PBDE levels in WHB bears were consistently two orders of magnitude lower than $\sum$PCB and $\sum$CHL levels.

Manufacture and use of decaBDE (mainly BDE209) is on-going in North America. Although BDE209 is a major congener in Arctic air (Hung et al., 2010), BDE209 and nonaBDEs were not consistently detected in WHB polar bears (<9% of all samples). Similarly inconsistent detection and/or low levels of BDE209 and nonaBDEs have been reported in other northern wildlife including ringed seal (blubber; Muir et al., 2007) and seabirds from northern Norway (eggs; Helgason et al., 2009). Low to non-detectable levels in biota versus the surrounding environment may be related to lower bioavailability (Sandholm et al., 2003) and/or greater metabolism of highly brominated PBDEs in biota (Stapleton et al., 2004).

To our knowledge, HBCD is the only other BFR for which temporal trends in Arctic biota have been previously investigated. Total-(α)-HBCD was largely not detected in WHB bears in the 1990s, but it was detected post-2000 at levels around five- to ten-fold lower than $\sum$PBDE (Table 3). Similarly, most HBCD time series published to date in Arctic biota could not determine trends due to large fluctuations in HBCD concentrations (de Wit et al., 2010). The PBB, BB153 (co-eluting with BDE154), was detected at similar levels to $\sum$PBDE. In the limited number of samples wherein BDE154 was quantifiable by $[M\text{-}2Br]^-$ anions, BDE154 comprised on average 4% (range: 1–13%) of the coeluting BB153/BDE154 signal. Therefore, BB153 was the predominant (individual) brominated contaminant detected in all years. The lack of temporal decreases in BB153 attested to its persistence, as PBBs have not been used in North America since the 1970s (de Boer et al., 2000). BB101 was infrequently found at low levels. PBEB, HBB and DBDPE were the only other BFRs detected, but detection was very infrequent and in the low- to sub-ppb range. PBEB and HBB were or are only low volume flame retardants, whereas DBDPE is a current, high production product. Because of its large molecular size, DBDPE may not be very mobile or bioavailable or it may be readily degradable (de Wit et al., 2010).

3.4. Contaminant patterns in relation to diet, year and biological group

Contaminant level and pattern trends in polar bears (Fig. 2) are partially due to changes in source signatures and pathways, as well as abiotic and biological weathering. Other recognized (confounding) factors include age, reproductive status, biotransformation capacity, lactation, seasonal fasting, body size, lipid content and habitat use (Borgå et al., 2004). Trends may also be related to changes in food web pathways (McKinney et al., 2009). To address some of these possibilities, we performed PCAs on the congener/metabolite proportions for major contaminant classes (Fig. 3). Multiple regression of PC factor scores with year, biological group and the three dietary tracers often resulted in low multiple r^2 values (below 0.5; Norman and Streiner, 2000) (Table 6). So the explanatory variables were not always good predictors of contaminant patterns, but reasonable explanations for certain contaminant patterns could be discussed.

The PCA on PCB congener proportions showed four clusters across two significant PCs (Fig. 3). PC1 and PC2 accounted for 43% and 35% of the pattern variation, respectively. Congeners in cluster A loaded positively on PC1 and consisted mainly of lower (tetra- to hexa-) chlorinated PCBs. Cluster B loaded negatively on PC1 and consisted of CB153. Polar bear PC1 factor scores were negatively correlated year and δ^{13}C and positively correlated with FA-Index1 (Tables 6 and 7). That is, a proportional increase in CB153 and decrease in lower chlorinated PCBs occurred over time (i.e., weathering) were also related to diet, consistent with lower proportional consumption of bearded seals and higher proportions of harbor or harp seals (see McKinney et al., 2009 for detailed dietary tracer interpretation).

For PCB-PC2, four highly (hepta- to nona-) chlorinated congeners loaded negatively (cluster C) and CB99 loaded positively (cluster D). PC2 factor scores were related to δ^{13}C, FA-Index2, biological group and their interactions. Highly chlorinated congeners were thus influenced by dietary exposure in all demographic groups, but in females and subadults the influence of diet may be moderated by lactational

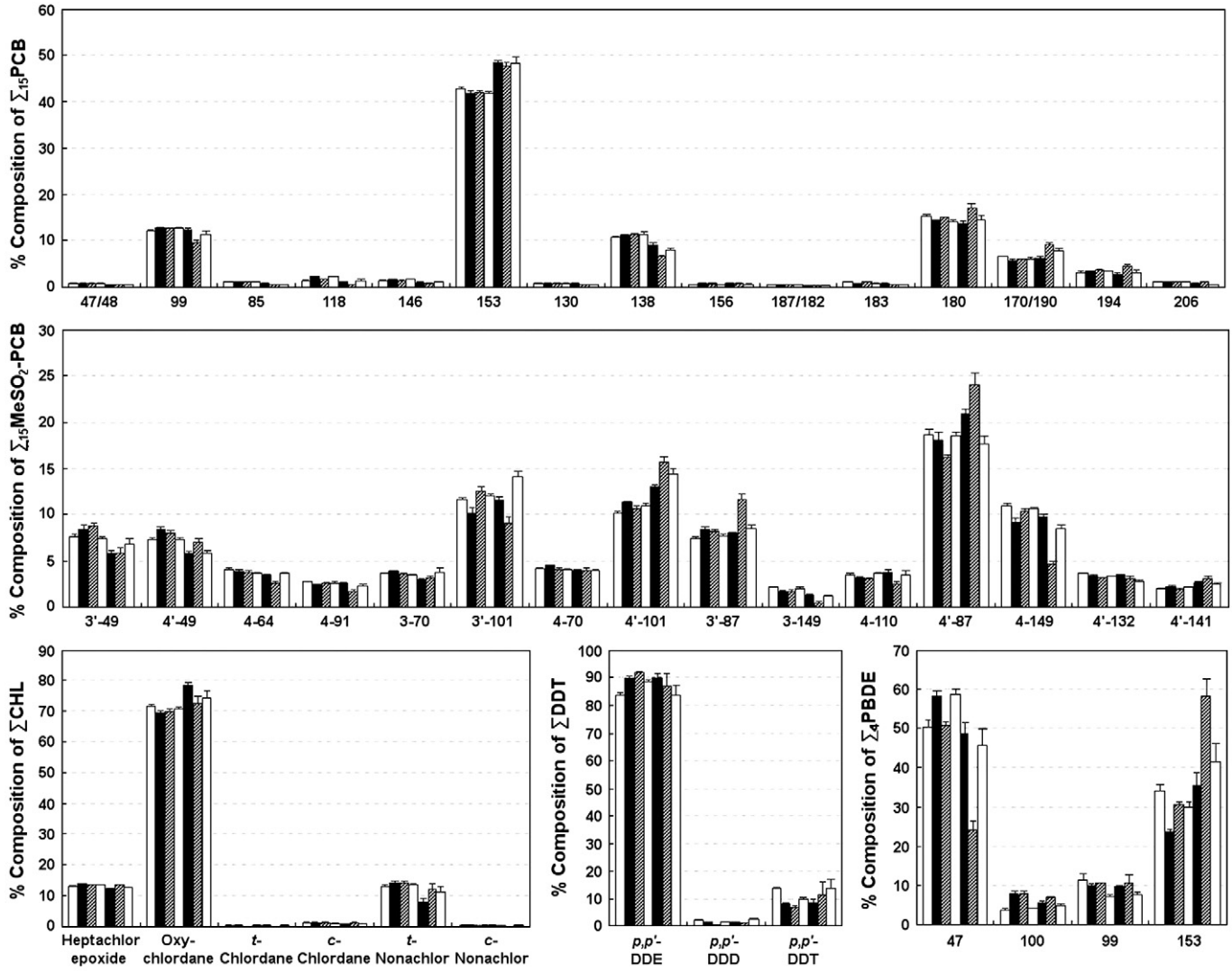

Fig. 2. Percent composition (+SE) of major contaminant classes in adipose of polar bears from western Hudson Bay (left to right): 1991 (first white bars), 1992 (first black bars), 1994 (first striped bars), 1995 (second white bars), 2001 (second black bars), 2003 (second striped bars) and 2007 (third white bars). PCB, $MeSO_2$-PCB and PBDE congener labels are further abbreviations from those listed in Tables 3 and 4.

transfer. Diet differences between male, female and subadult polar bears have been reported (Thiemann et al., 2008). Bernhoft et al. (1997) found higher levels of these same four highly chlorinated PCBs in adult male polar bears than in adult females/young/subadults from Svalbard. The proposed explanation was that lactational loss results in lower proportions of recalcitrant, highly chlorinated congeners in adult females versus males, but has less effect on more metabolizable, lower chlorinated PCBs. Lower proportions of highly chlorinated PCBs in subadults versus adult males may be because lactational transfer of highly chlorinated PCBs is limited.

BDE47 and BDE153 contributed to variation in PBDE-PC1, which accounted for 48% of the variance (Fig. 3). Higher and lower proportions of BDE153 and BDE47, respectively, were associated with adult males (versus adult females/subadults) and with higher FA-Index1 (higher proportions of harp or harbor seals, and lower proportions of bearded seals). Interaction terms between biological group (adult males versus adult females) and $\delta^{13}C$, FA-Index1 and FA-Index2 were significant, implying that the influence of dietary uptake in females and subadults was moderated by lactational transfer. Similar to the PCB pattern explanation (Bernhoft et al., 1997), greater lactational transfer of the lower brominated BDE47 versus the higher brominated BDE153 may have resulted in higher proportions of BDE47 in subadults. However, adult females still had relatively lower proportions of BDE153 than adult males because of lactational transfer. Year was not significant in explaining the PBDE congener pattern, suggesting that it is too early to detect weathering of the signature, i.e., increased proportions of the more persistent congener, BDE153 (Muir et al., 2006).

The CHL PCA found only one significant PC, accounting for 58% of the variation. CHL-PC1 separated oxychlordane from a cluster containing heptachlor epoxide, *c*-chlordane, *c*-nonachlor and *t*-nonachlor (Fig. 3). Higher proportions of oxychlordane and lower proportions of the clustered compounds were best explained by individual diet differences (depleted $\delta^{13}C$ and higher FA-Index1) (Table 6). Biological group and the interactions between biological group and dietary tracers also contributed. Adult females had higher proportions of oxychlordane versus subadults and adult males. Polischuk et al. (2002) found that oxychlordane decreased in adult and subadult male polar bears during fasting, but not in adult females, indicating sex-specific metabolism. Higher oxychlordane proportions were also associated with later years, suggesting that the CHL pattern has changed due to weathering.

The $MeSO_2$-PCB PCA found three significant PCs (PC1 and PC2 shown in Fig. 3), accounting for 39%, 17% and 12% of the variation. Two clusters of congeners (A and B) contributed to the variation along PC1. None of the explanatory variables (year, dietary tracers or biological group) significantly explained variation along this PC. Letcher et al. (1998) found that $MeSO_2$-PCBs could be present in polar bears due to

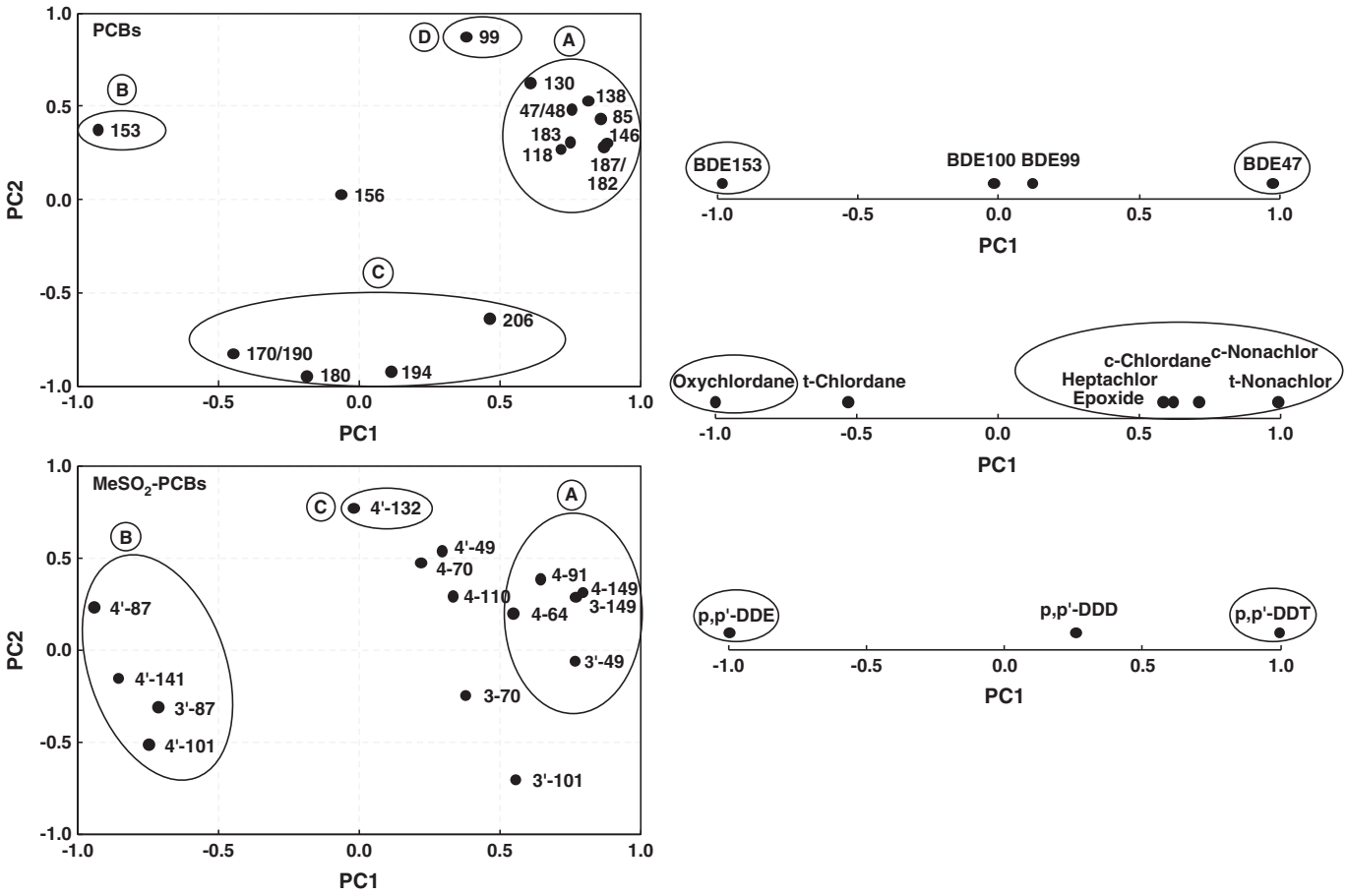

Fig. 3. Factor loadings from principle components analyses on the percent (%) composition of major contaminant classes in adipose of polar bears from western Hudson Bay sampled in 1991–2007. Encircled clusters contained compounds that loaded significantly (critical value of the correlation > 0.54) on the same PC, but were not significant on any other PCs. PCB and $MeSO_2$-PCB congener labels are further abbreviations from those listed in Table 4.

metabolic formation and/or accumulation from prey. As dietary tracers did not explain variation in PC1 factor scores, $MeSO_2$-PCBs that loaded significantly on PC1 were not simply bioaccumulated from their diet. Variation along PC1 may have been related to individual metabolic differences from, e.g., differences in exposure to enzyme-inducing contaminants or body condition, which were not considered in the model. Of the four $MeSO_2$-PCBs that loaded negatively on PC1 (cluster B), Letcher et al. (1998) demonstrated that 3′- and 4′-$MeSO_2$-CB87 and 4′-$MeSO_2$-CB141 were among the congeners most likely to be additionally present in polar bears due to biotransformation. On PC2, 4′-$MeSO_2$-CB132 loaded positively, one of the only $MeSO_2$-PCBs present in polar bears solely due to bioaccumulation (Letcher et al., 1998). Thus, it was not surprising that, in addition to being associated with year, higher scores on PC2 were significantly explained by dietary tracers. These findings suggested that some but not all $MeSO_2$-PCBs may be influenced by long-term diet/food web changes.

Table 6
Influence of year, biological group and the dietary tracers, $\delta^{13}C$, FA-Index1, FA-Index2, on contaminant patterns in adipose of polar bears from western Hudson Bay.

Contaminant pattern[a]	Multiple regression with the main effects year, biological group[b], dietary tracers and first-order interactions of biological group and dietary tracers		
	Multiple r^2	p	Significant explanatory variables (semipartial r)
PCB-PC1	0.64	<0.001	Main effects: $\delta^{13}C$ (0.18), FA-Index1 (−0.14), year (−0.18),
PCB-PC2	0.34	<0.001	Main effects: biological group (−0.25) Interactions: biological group × $\delta^{13}C$ (−0.24), biological group × FA-Index2 (−0.24)
$MeSO_2$-PCB-PC1	0.46	<0.001	None
$MeSO_2$-PCB-PC2	0.48	<0.001	Main effects: FA-Index1 (0.22), $\delta^{13}C$ (−0.16), year (−0.32)
$MeSO_2$-PCB-PC3	0.25	0.002	none
PBDE-PC1	0.66	<0.001	Main effects: biological group (−0.18), FA-Index1 (−0.21) Interactions: biological group × FA-Index1 (0.16), biological group × FA-Index2 (−0.14), biological group × $\delta^{13}C$ (−0.17)
CHL-PC1	0.47	<0.001	Main effects: biological group (0.19), year (−0.17), $\delta^{13}C$ (0.36), FA-Index1 (−0.41), Interactions: biological group × $\delta^{13}C$ (0.19), biological group × FA-Index1 (−0.17)
DDT-PC1	0.34	<0.001	None

[a] Contaminant patterns were represented by the factor scores of individual polar bears from a principle components analysis (PCA) on % composition of major individual contaminants to ∑-contaminant class.

[b] Results shown above were from testing biological group coding k-1 (k = 3 groups: adult female, adult male and subadult) dummy variables, with adult female as the reference category. To test for adult male versus subadult, the analysis was repeated with subadult as the reference category. For PCB-PC2 and PBDE-PC1, biological group was significant for adult male versus adult female (and adult male versus subadult). For CHL-PC1, biological group was significant for subadult versus adult female.

Table 7
Fatty acid profiles and carbon stable isotope ratios as dietary tracers in western Hudson Bay polar bear adipose from 1991 to 2007.

	Mean (SE) % of total dietary FAME							PCA factor loadings	
	1991	1992	1994	1995	2001	2003	2007	FA-Index1 (PC1)	FA-Index2 (PC2)
Linoleic acid (18:2n-6)	9.74 ± 0.34	9.57 ± 0.22	8.42 ± 0.15	8.37 ± 0.15	9.22 ± 0.51	9.22 ± 0.23	9.77 ± 0.66	0.23	−0.49
Gamma-linolenic acid (18:3n-6)	0.38 ± 0.02	0.41 ± 0.03	0.39 ± 0.02	0.38 ± 0.01	0.29 ± 0.02	0.28 ± 0.01	0.27 ± 0.01	**−0.76**	−0.28
Alpha-linolenic acid (ALA) (18:3n-3)	1.83 ± 0.16	1.99 ± 0.07	2.07 ± 0.12	1.62 ± 0.03	3.71 ± 0.58	2.66 ± 0.31	4.66 ± 0.37	0.52	−0.08
cis-11,14,17-Eicosatrienoic acid (ETA) (20:3n-3)	0.22 ± 0.01	0.35 ± 0.01	0.34 ± 0.01	0.21 ± 0.00	0.20 ± 0.02	0.24 ± 0.03	0.27 ± 0.02	−0.47	**−0.59**
Arachidonic acid (ARA) (20:4n-6)	0.89 ± 0.04	0.77 ± 0.04	0.78 ± 0.04	0.91 ± 0.04	0.83 ± 0.07	0.73 ± 0.04	1.18 ± 0.15	−0.30	0.41
cis-5,8,11,14,17-Eicosapentaenoic acid (EPA) (20:5n-3)	10.47 ± 0.52	10.94 ± 0.41	10.76 ± 0.53	12.81 ± 0.58	7.74 ± 1.07	7.76 ± 0.97	6.74 ± 1.07	**−0.84**	0.38
cis-7,10,13,16,19-Docasapentaenoic acid (DPA) (22:5n-3)	25.52 ± 0.64	24.91 ± 0.35	25.88 ± 0.41	27.69 ± 0.31	24.92 ± 0.77	26.03 ± 0.82	26.63 ± 1.16	**0.59**	−0.09
cis-4,7,10,13,16,19-Docasahexaenoic acid (DHA) (22:6n-3)	38.58 ± 0.67	35.92 ± 0.54	35.25 ± 0.42	39.73 ± 0.34	39.32 ± 0.85	39.24 ± 0.85	38.94 ± 0.69	0.18	**0.88**
Mean $\delta^{13}C$ (SE) (‰)	−25.08 ± 0.04	−24.92 ± 0.04	−25.07 ± 0.04	−25.24 ± 0.04	−25.26 ± 0.14	−25.41 ± 0.11	−25.16 ± 0.07		

The DDT PCA found one significant PC, accounting for 69% of the variation. DDT-PC1 was distinguished by positive loading of *p,p′*-DDT and negative loading of *p,p′*-DDE. None of the explanatory factors explained variation along DDT-PC1. Lack of biological group and dietary influences were consistent with substantial metabolism of DDTs in subadults, adult males and adult females (Polischuk et al., 2002). Like $MeSO_2$-PCB-PC1, perhaps DDT patterns were partly associated with individual differences in metabolic potential towards DDTs. The DDT pattern was not associated with year, although a more weathered pattern (proportionally higher DDE) over time was expected. Continued exposures to a more "fresh" local source of DDT may be from spraying in the study region in the 1950s and 1960s (McKinney et al., 2009).

Overall, chlorinated contaminant trends in WHB polar bears varied in comparison to those measured in other northern wildlife, for instance, more rapid DDT decline, more rapid β-HCH increase and no CHL decline. These unusual temporal trends may, in part, have been related to sea ice-associated diet and/or food web changes we previously reported in this subpopulation. Contaminant patterns were also suggestive of exposure changes that were related to source inputs and weathering and to diet/food web changes.

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