

Sea Ice-associated Diet Change Increases the Levels of Chlorinated and Brominated Contaminants in Polar Bears

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Two global environmental issues, climate change and contamination by persistent organic pollutants, represent major concerns for arctic ecosystems. Yet, it is unclear how these two stressors interact in the Arctic. For instance, the influence of climate-associated changes in food web structure on exposure to pollutants within arctic ecosystems is presently unknown. Here, we report on recent changes in feeding ecology (1991–2007) in polar bears (*Ursus maritimus*) from the western Hudson Bay subpopulation that have resulted in increases in the tissue concentrations of several chlorinated and brominated contaminants. Differences in timing of the annual sea ice breakup explained a significant proportion of the diet variation among years. As expected from climate change predictions, this diet change was consistent with an increase in the consumed proportions of open water-associated seal species compared to ice-associated seal species in years of earlier sea ice breakup. Our results demonstrate that climate change is a modulating influence on contaminants in this polar bear subpopulation and may pose an additional and previously unidentified threat to northern ecosystems through altered exposures to contaminants.

Introduction

There is an established link between recent climate change and phenological, geographical, and compositional changes to ecosystems across many regions of the world (1, 2). The magnitude of warming is regionally variable, though, and is nearly twice the global average in the Arctic (3, 4). There are reports of recent and drastic climate-related shifts in the composition of arctic and subarctic marine ecosystems. In northern Hudson Bay, there has been an increase in subarctic fish relative to arctic fish (as measured by the diet of thick-billed murre (*Uria lomvia*)) with decreasing July ice cover from 1980 to 2002 (5). Reduced sea ice, along with warmer air and ocean temperatures, has shifted the previously

benthic-dominated Northern Bering Sea ecosystem to a more pelagic, Southern Bering Sea-type (subarctic) ecosystem (6).

Environmental contamination by persistent organic pollutants (POPs) is also a major concern for northern ecosystems, despite substantial distances from source regions further south (7, 8). Contamination of subarctic and arctic regions by POPs occurs mainly through long-range transport from lower latitudes (9). Polychlorinated biphenyls (PCB) as well as organochlorine pesticides (OCP) including dichlorodiphenyltrichloroethane (DDT), hexachlorocyclohexane (HCH), and chlordane (CHL) have long been observed as contaminants in northern biota (10, 11). Lesser known, more recently monitored chemicals such as polybrominated diphenyl ether (PBDE) flame retardants and perfluorinated contaminants (PFCs) are now also a concern in the Arctic (12, 13). As upper trophic level consumers, polar bears, as well as humans consuming a local diet, accumulate high tissue concentrations of, e.g., PCB and CHL contaminants, which have been related to biological effects (14–16). For instance, endocrine, immune, and reproductive biomarkers have been correlated with tissue levels of PCBs and certain OCPs in polar bears (15, 16 and references therein).

Polar bears from the western Hudson Bay (WHB) subpopulation exist near the southern limit of the range of this species (Figure 1), and appear to be more impacted by recent climate warming than their more northerly located conspecifics (17). Hudson Bay polar bears (as well as those from the European Arctic) also have generally elevated adipose concentrations of organohalogen contaminants relative to other circumpolar populations (18). For this subpopulation, movement, mating and feeding depends in part on the timing and conditions of seasonal sea ice in the bay (19). In this region, the mean annual air temperature is on average about 1.5 °C warmer (as measured at Churchill, Manitoba, 58.3°N, 93.8°W) (20), and the summer sea ice breakup is around three weeks earlier (20), than thirty years ago in the western part of the bay. Over the same period, there has been no significant trend in the timing of fall sea ice freeze-up in western Hudson Bay (20). There have been no significant trends in winter maximum land-fast ice thickness and timing at Churchill from 1960 to 1987 (21). The increasingly earlier breakup of the summer sea ice has been linked to lower body condition, birth, and survival rates in WHB bears over the past two decades (19, 22). We hypothesized that this sea

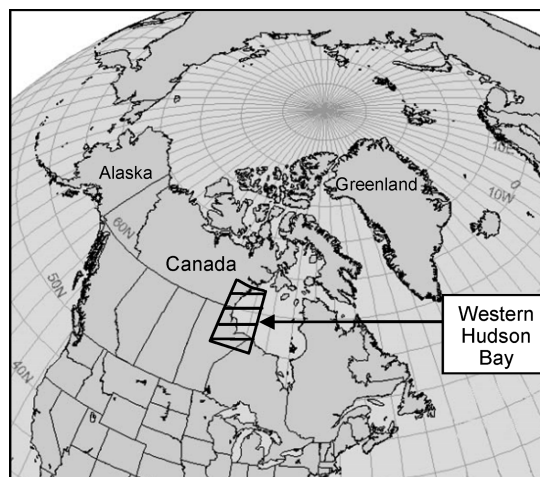


FIGURE 1. Map of study site. The western Hudson Bay polar bear subpopulation, indicated by the striped area, is bounded by 63.10°N, 88.30°W, and by the western Hudson Bay coastal region.

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ice change is also impacting WHB polar bear feeding ecology, as measured by stable isotope and fatty acid dietary tracers. Since diet is the main route of exposure to contaminants, we also hypothesized that feeding changes would, in turn, impact their tissue contaminant levels.

Experimental Methods

Study Site and Sample Details. We analyzed archived WHB polar bear adipose tissues from seven available years over the 17 year period of 1991–2007: 1991 ($n = 14$), 1992 ($n = 15$), 1994 ($n = 15$), 1995 ($n = 15$), 2001 ($n = 9$), 2003 ($n = 12$), and 2007 ($n = 12$). Supporting Information (SI) Table S2 provides further sample details. The 1990s samples were adipose biopsies taken during polar bear tagging operations, whereas the 2000s samples were mainly pieces of adipose tissue taken from bears harvested by Inuit hunters. It has been shown that there are no differences POPs patterns and concentrations or fatty acid composition among major adipose depots in polar bear (23, 24). All samples were initially and during shipment kept frozen and stored at -40°C over the long term at Environment Canada's Wildlife Specimen Bank. There appears to be no change in fatty acid concentrations and patterns between samples stored at -40 and -80°C (25). To focus on the effect of diet on the contaminant concentrations over time, we selected and corrected (see also the Data Analysis Section, SI Table S2) the data set to minimize confounding factors that could contribute to interindividual variation in contaminant levels. We selected samples collected in fall/winter, which were at least 3 years old and 80% of which were female to minimize seasonal, age and gender related variability.

Determination of Sea Ice Breakup Timing. We calculated the timing of annual sea ice breakup with established methods (19, 26). In brief, we determined the date when the ice cover in WHB (Figure 1) was at 50% by interpolation of weekly sea ice data from the Canadian Ice Service (<http://ice-glaces.ec.gc.ca>).

Diet Analysis. Traditional methods of studying feeding behavior, such as observation of kills and analysis of gut contents, may be biased and are limited to recently consumed items. We thus examined changes in polar bear feeding using two established dietary tracers (27, 28).

Stable carbon isotope ratios ($\delta^{13}\text{C}$, in ‰)

$$\delta^{13}\text{C} = \left[\left(\frac{^{13}\text{C}/^{12}\text{C}_{\text{sample}}}{^{13}\text{C}/^{12}\text{C}_{\text{Pee Dee belemnite standard}}} \right) - 1 \right] \times 1000$$

are not altered substantially through a food web, but may distinguish carbon sources (29). For instance, $\delta^{13}\text{C}$ is enriched in benthic versus pelagic food webs; France (30) reviewed literature on 876 marine and freshwater benthic and pelagic (phytoplankton) algae and found consistent $\delta^{13}\text{C}$ enrichment in benthic algae (and consumers) versus phytoplankton. We therefore used $\delta^{13}\text{C}$ variation as an indicator of changes in the proportions of benthic-foraging versus pelagic-foraging prey species in the polar bear diet. The lipid extract from the gravimetric lipid determination was used for $\delta^{13}\text{C}$ determination. Using the tip of a solvent-cleaned, unfolded paperclip, 150–300 μg of lipid was weighed accurately into a 5×3.5 mm tin capsule. Stable isotopes were combusted with an elemental analyzer and analyzed by a coupled continuous flow isotope ratio mass spectrometer (Environmental Isotope Laboratory, University of Waterloo, Waterloo, ON, Canada).

Fatty acid (FA) patterns in mammalian predators reflect the FA composition of their diet (28). The FA patterns of potential polar bear prey species from a large number ($n = 20$ –281) of individuals collected between 1992 and 2004 across the Canadian Arctic and Subarctic was recently published; despite some intraspecies geographic differences, the differences in FA patterns between species were larger

(31). Thus, we used changes in polar bear FA patterns over time, in conjunction with this published prey FA data, to infer alterations in the proportions of prey species consumed. Extraction and analysis of FAs from adipose tissue has been described previously. In short, 10–20 mg of inner adipose tissue was used to avoid potentially oxidized outer tissue (32). Lipids were extracted thrice using 2:1 CHCl_3 :MeOH (33) containing 0.01% 2,6-di-*t*-butyl-4-methylphenol as antioxidant. 5- α -Cholestane was used as internal standard. The extract was evaporated to dryness under N_2 , and lipid was redissolved in toluene. FAs were methylated via the Hilditch reagent (32). After addition of 2% KHCO_3 , the organic layer was collected and fatty acid methyl esters (FAMES) were completely collected by two further extractions with hexane. FAMES were analyzed by GC-FID equipped with a Supelco-2560 bis-cyanopropyl column and quantified against a Supelco 37 component FAME external standard (25). Here, we report only on the “dietary” FAs (i.e., those that are incorporated relatively unchanged from prey to predator adipose tissues for a monogastric predator) (28, 32) that were available for quantification based on the external standard. Each FAME was calculated as the % of total dietary FAME.

Organohalogen Contaminant Analysis. We extracted brominated and chlorinated contaminants from polar bear adipose tissue as described previously (34) with modifications outlined here. We homogenized around 0.5 g of fat with sodium sulfate and spiked it with a mixture of ^{13}C -PCBs, ^{13}C -*p,p'*-DDE and BDE30 internal standards. We then extracted the contaminants with 1:1 DCM:hexane using an accelerated solvent extractor at 100°C and 1500 psi for one cycle. A 10% portion of the extract was used for gravimetric determination of lipid content. From the remainder, lipids and other bioorganics were removed by automated gel permeation chromatography. The extract was further cleaned up on a preconditioned (6 mL of 10% MeOH in DCM, followed by 8 mL of 5% DCM in hexane) silica (500 mg) solid phase extraction cartridge with 8 mL of 5% DCM in hexane (35). Analysis of PCBs and OCPs by GC-MS in electron impact ionization mode has been detailed elsewhere (36). PBDEs were analyzed by GC-MS in electron capture negative ionization mode as per Gauthier et al. (37), except that the source temperature was set to 250°C . Each contaminant concentration (or sum of contaminant class) was calculated on a lipid weight basis.

Quality Control for Diet and Contaminant Data. For $\delta^{13}\text{C}$, duplicate analysis was performed on 13% of the samples; the mean difference between repeated measurements was 0.16‰ . For FA analysis, the internal standard recovery was $107 \pm 5\%$. A blank, duplicate and two reference materials, a Great Lakes herring gull egg pool and the NIST Pilot Whale blubber SRM1945, were extracted with each batch. The %RSD for all dietary FA values was on average 5% for the herring gull egg. The pilot whale blubber FA values were on average within 15% of our laboratory results from the 2007 NIST/NOAA Interlaboratory Comparison Exercise Program for Organic Contaminants in Marine Mammal Tissues. The dietary FA values were on average 6% different for the polar bear duplicate analyses.

For contaminant analysis, the internal standard recoveries were $85 \pm 5\%$, $88 \pm 8\%$, and $87 \pm 12\%$ for ^{13}C -PCBs, ^{13}C -*p,p'*-DDE, and BDE30, respectively. ΣPCB , ΣOCP , and ΣPBDE concentrations in repeated analyses of SRM1945 were within $5 \pm 4\%$, $7 \pm 3\%$, and $14 \pm 7\%$, respectively, of the NIST certified values (38, 39). In the same order, polar bear duplicate analyses were on average 7%, 15%, and 21% different. For the individual or sum of contaminant classes reported in this study, there were no values below the detection limits.

Data Analysis. As it is the overall pattern of FA that provides diet information (28), we performed principle components analysis on a suite of dietary FAs: linoleic acid

(18:2n-6), γ -linolenic acid (18:3n-6), α -linolenic acid (ALA; 18:3n-3), *cis*-11,14,17-eicosatrienoic acid (ETA; 20:3n-3), arachidonic acid (ARA; 20:4n-6), *cis*-5,8,11,14,17-eicosapentaenoic acid (EPA; 20:5n-3), *cis*-7,10,13,16,19-docasapentaenoic acid (DPA; 22:5n-3), and *cis*-4,7,10,13,16,19-docasahexaenoic acid (DHA; 22:6n-3). The values of the first and second principal components, which accounted for 51% of the total FA variation, are herein referred to as FA-Index1 and FA-Index2 and used in subsequent analyses.

We used simple correlation analysis to investigate the relationship of the dietary tracers ($\delta^{13}\text{C}$, FA-Index1 and FA-Index2) to ice breakup date. However, due to the limited number of years of available data, we also tested these associations by performing correlation analysis on the ranked data (Table S1).

We then determined the impact of dietary changes on the concentrations of POPs in WHB bears by comparing the annual % change in the concentration of each contaminant before and after diet correction. Prior to these analyses, for contaminants consistently observed to differ in concentration between sexes (ΣPCB , ΣCHL , ΣDDT), data from the small number of male samples was converted to female equivalents (36). In addition, all contaminant concentrations were $\log(x + 1)$ -transformed to approximate normal distribution (tested using Shapiro-Wilk W). Annual % change controlling for diet was calculated as $(1 - 10^b) \times 100\%$, where b is the slope of the relationship between the contaminant concentration and the year from a multiple regression of the log-transformed contaminant concentration versus the dependent variables, $\delta^{13}\text{C}$, FA-Index1, FA-Index2 and year. Actual annual % change (i.e., without diet correction) was calculated as $(1 - 10^b) \times 100\%$, where b is the slope of the simple regression of the log-transformed contaminant concentration versus year. To determine whether the diet-controlled versus actual annual % change for each contaminant were significantly different, we compared the two b -values (slopes) using a Student's t -test. All tests were deemed statistically significant at $p < 0.05$, marginally significant at $p < 0.10$ and were two-tailed. We used Statistica V6.0 (StatSoft, 2003; Tulsa, OK) for all statistical analyses. Here, we report only on the change in contaminant concentrations; actual contaminant concentrations and congener patterns are outside the scope of this study.

Results and Discussion

Relationship of Diet to Ice Breakup Date. The mean annual $\delta^{13}\text{C}$ values were significantly correlated with the annual ice breakup date ($r = 0.92$, $p = 0.004$; Figure 2a). The timing of the ice breakup explained 84% of the variation in $\delta^{13}\text{C}$ in WHB polar bears over the 1991 to 2007 time period. Decreasing (more negative) $\delta^{13}\text{C}$ values, which suggest decreased proportions of benthic-foraging versus pelagic-foraging prey items in the polar bear diet, were associated with earlier ice breakup dates. Recent FA research has demonstrated that the WHB polar bear diet consists of two ice-associated prey, ringed seal (*Pusa hispida*) and bearded seal (*Erignathus barbatus*), and two open water-associated prey, harbor seal (*Phoca vitulina*) and harp seal (*Phoca groenlandica*) (40). As bearded seal is the only one of these seal species that forages predominately on benthic items (31), the change in polar bear $\delta^{13}\text{C}$ suggests a lower consumption of bearded seals and consequently a relative increase in consumption of one or more of the other seals in years with a shortened period of ice cover.

Decreasing $\delta^{13}\text{C}$ values with earlier breakup of the sea ice could also in part be influenced by other factors: increased terrestrial feeding, longer fasting, $\delta^{13}\text{C}$ variation at the base of the food web, or incorporation of $\delta^{13}\text{C}$ -depleted CO_2 emitted from fossil fuel combustion into marine food chains. However, terrestrial feeding is an unlikely explanation, as

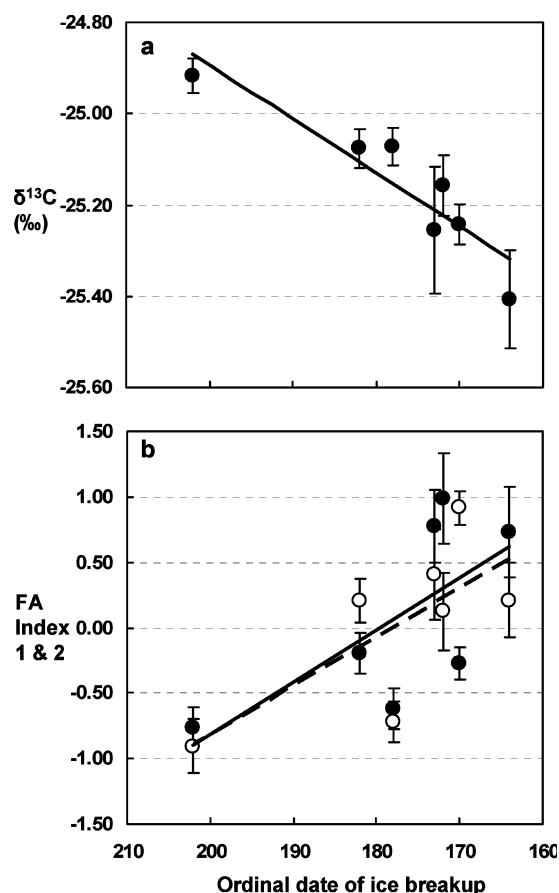


FIGURE 2. Mean (\pm SE) annual polar bear dietary tracer values versus ordinal date (January 1 = ordinal date 1) of annual sea ice breakup in western Hudson Bay from 1991 to 2007. (a) Carbon stable isotopes ($\delta^{13}\text{C}$). (b) Fatty acid (FA)-Index1 (●, solid line) and FA-Index2 (○, dashed line).

although feeding on, e.g., berries, has been observed in WHB polar bears during the ice-free season, $\delta^{13}\text{C}$ signatures in WHB bears, ringed seals and berries have consistently demonstrated that terrestrial input is not a significant contributor to the WHB bear diet (41). There are limited reports on the effects of fasting on dietary tracers. Yet, Polischuk et al. (42) found no difference in milk lipid $\delta^{13}\text{C}$ values between WHB polar bear females between fall and spring (encapsulating our sample collection dates), although spring values were more variable. FA composition in black bear (*Ursus americanus*) milk was also relatively constant during and after winter dormancy (43). Taken together, these findings suggest that the dietary tracer changes with ice breakup are not largely driven by changes in fasting. Baseline food web $\delta^{13}\text{C}$ changes due to variation in nutrients or productivity has been observed (44, 45), and without knowledge of possible temporal variation in baseline $\delta^{13}\text{C}$, we cannot rule out that this could, in part, result in $\delta^{13}\text{C}$ variation in the bears. Atmospheric CO_2 has become depleted in $\delta^{13}\text{C}$ due to the contribution of isotopically light CO_2 from fossil fuel emissions (46). Relevant to this study, the depletion was around 0.4‰ over the period of 1991 to 2006 (47). We found a nonsignificant decrease in WHB polar bear $\delta^{13}\text{C}$ from 1991 to 2007 ($r = 0.63$, $p = 0.13$). A correction factor for this confounding variation has been developed for temporal studies on terrestrial ecosystems, but the authors noted that this depletion would be less in marine systems, as dissolved inorganic carbon (DIC) is not in equilibrium with atmospheric CO_2 and atmospheric CO_2 is not the sole source of DIC to aquatic systems (48). Further research on the extent to which

TABLE 1. Contaminant Temporal Trends in Western Hudson Bay Polar Bears With and Without Correction for Diet^a

contaminant	simple regression with year			multiple regression with year and diet tracers						
	<i>r</i> ²	<i>p</i>	annual % change (1 – 10 ⁶)	<i>r</i> ²	<i>p</i>	year	standardized regression coefficients (β) $\delta^{13}\text{C}$	FA-Index1	FA-Index2	annual % change (1 – 10 ⁶)
log(α -HCH)	0.83	<0.0001	–12%	0.83	<0.0001	–0.91	0.01	–0.003	0.006	–12%
log(Σ PCB)	0.04	0.05	+1.7%	0.36	<0.0001	–0.11	0.02	0.60	0.23	–0.92%
log(Σ CHL)	0.00003	0.96	+0.04%	0.14	0.01	–0.16	–0.17	0.30	–0.36	–1.1%
log(Σ PBDE)	0.77	<0.0001	+12%	0.82	<0.0001	0.67	–0.15	0.29	–0.12	+9.4%
log(β -HCH)	0.60	<0.0001	+7.7%	0.64	<0.0001	0.64	–0.15	0.20	–0.19	+6.4%
log(Σ DDT)	0.57	<0.0001	–11%	0.68	<0.0001	–0.46	0.21	–0.43	0.15	–6.7%

^a Trends without diet correction (i.e., actual trends) are those from the simple regression of each contaminant with year; trends with diet correction are those from the multiple regression results of each contaminant with year, $\delta^{13}\text{C}$, FA-Index1 and FA-Index2. Significant correlations are indicated in bold.

this confounding factor impacts $\delta^{13}\text{C}$ studies of marine food webs is warranted.

Given the variety of other factors that may affect $\delta^{13}\text{C}$, we also studied FA patterns in WHB polar bears. Increasingly earlier ice breakup date explained 46% and 53% of the increases in FA-Index1 ($r = -0.68$, $p = 0.09$) and FA-Index2 ($r = -0.73$, $p = 0.07$), respectively (Figure 2b). The main FAs that loaded positively on Index1 (ALA and DPA) and Index2 (DHA) have been found in higher proportions in harbor and harp seal than in bearded seal (31). The main FAs that loaded negatively on FA-Index1 (EPA, γ -linoleic acid) have been found at lower proportions in harbor and harp seal than in bearded seal (31). However, these FA patterns did not suggest changes in the proportion of ringed seal consumed. These results, in combination with the $\delta^{13}\text{C}$ changes, suggest a relative decrease in the proportion of bearded seals consumed and increases in the proportion of harbor and harp seals consumed in years with a longer period of open water. This shift in the proportions of ice-associated to open-water associated prey is largely consistent with an observed diet shift for WHB bears using FA tracers of the bears and of their prey over the 1994–2004 period which, though, also found relatively constant ringed seal consumption (40).

Changes in WHB polar bear consumption patterns could be related to changes in accessibility to prey and/or prey abundance with altered ice conditions in the region. Ringed seals and bearded seals are considered ice-seals; yet, land-fast ice is more important habitat for ringed seals compared to pack ice habitat for bearded seals (49). Land-fast ice in the region appears to be stable, but pack ice conditions have changed (20, 21). It is possible that accessibility of WHB polar bears to bearded seals (but not ringed seals) has changed as a result. Harbor seals in Hudson Bay are year-round inhabitants, but stay in areas of open water (50). Harp seals are migratory, moving from the North Atlantic to, among other regions, Hudson Bay when open-water conditions prevail (49). Archaeological evidence from Inuit settlements in Labrador showed a higher ratio of harbor seal to ringed seal in centuries with lighter ice conditions (51), suggesting that open-water seals may be more abundant in northern regions in years of lighter ice conditions. Further study of polar bear prey in Hudson Bay is required to address these possible explanations. It is important to note, however, that the changes in polar bear dietary tracers may reflect not only shifts near the top of this food web, but also more extensive ecosystem composition changes in Hudson Bay (5).

Influence of Diet Change on Chlorinated and Brominated Contaminant Concentrations. All contaminants (except α -HCH) were correlated with FA-Index1 and FA-Index2, and Σ DDT and Σ PBDE were also correlated with $\delta^{13}\text{C}$, implying that diet explained a significant amount of the variation in WHB polar bear contaminant levels (Table 1). Similarly, Bentzen et al. (52, 53) used stable carbon and

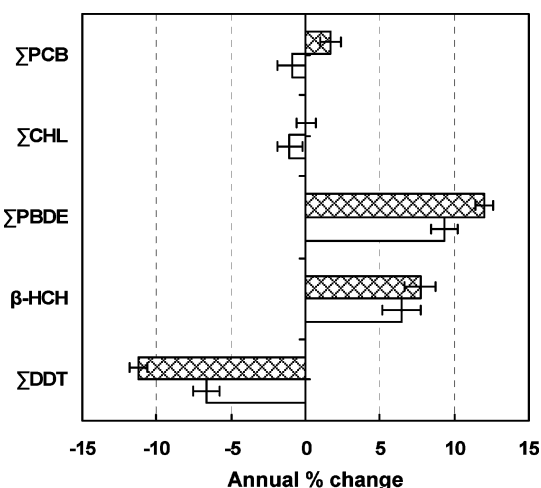


FIGURE 3. Annual % change (\pm SE) in the concentrations of persistent organic contaminants in western Hudson Bay polar bears, controlling for diet (white bars) and with the influence of diet included, i.e., the actual temporal trends (crosshatched bars).

nitrogen isotopes to demonstrate that southern Beaufort Sea polar bears show interyear differences in diet, and that $\delta^{15}\text{N}$ and to a lesser extent $\delta^{13}\text{C}$ explained a significant proportion of the interindividual variation in organochlorine concentrations.

To determine the effect of the measured feeding shift on the levels of contaminants in WHB polar bears, we compared the annual % change in the concentration of each contaminant (or sum of contaminant class) before and after controlling for diet change (Figure 3). When we controlled for diet change, the concentrations of Σ PCB, Σ CHL, and Σ DDT decreased over time in WHB polar bears, whereas Σ PBDE and β -HCH increased. However, when we examined the actual trends (i.e., not adjusted for diet change), the magnitude and in some cases direction of concentration changes over time was different. For Σ PCB, Σ CHL, Σ PBDE, and β -HCH, the diet change resulted in increased concentrations over time. This meant that the concentration of Σ PBDE increased at a significantly (Σ PBDE, $t_{0.05(2),10} = 2.45$, $p = 0.03$) and 28% faster rate than if the diet had not changed. β -HCH increased at a 20% faster rate (although not statistically faster than the diet corrected rate: $t = 1.17$, $p = 0.26$). For Σ PCB and Σ CHL, the trends actually switched from decreasing to increasing concentrations over time due to the change in diet, though only the Σ PCB switch was marginally statistically significant (Σ PCB: $t = 1.93$, $p = 0.08$; Σ CHL, $t = 0.95$, $p = 0.36$). Overall, these trends toward higher actual contaminant concentrations versus those if diet had remained constant are consistent with studies demonstrating lower contaminant levels in bearded seals relative to the other prey species

(54–56) and with the observed diet shift to less bearded seal consumption by WHB polar bears. Not unexpectedly, the diet shift had more impact on the concentrations of contaminants that are highly biomagnified in polar bears (e.g., Σ PCB, Σ PBDE) relative to contaminants that biomagnify to a lesser extent (e.g., β -HCH) (57, 58). Similarly, the change for α -HCH, which demonstrates relatively low persistence and bioaccumulation, remained at -12% per year regardless of diet correction (Table 1).

Surprisingly, Σ DDT concentrations decreased more rapidly (64% faster; $t = 2.69$, $p = 0.02$) in response to the polar bear diet change. Although the major burden of POPs in northern regions is from long-range transport (9), DDT was also sprayed in the 1950s and 1960s at military installations and communities within our study site. Resulting terrestrial runoff likely led to DDT contamination of the local marine benthos. For example, PCB use at DEW-line sites in the Canadian Arctic resulted in elevated PCB concentrations in nearby marine sediments and benthic species (59). In addition, a study of the White Sea in the Russian Arctic, a region much closer to urban and industrial pollution sources than the Canadian Arctic, found that benthic foragers (bearded seals) had higher Σ PCB and Σ DDT concentrations than more pelagic foragers (ringed and harp seals) (60). Thus in our study, the more rapid decrease in polar bear Σ DDT concentrations is consistent with the decreased contribution of benthic-foraging prey to their diet, as measured by $\delta^{13}\text{C}$ changes.

Adipose contaminant concentrations may be affected by decreased fatness (body condition), as has been documented in this subpopulation over the same time period (19). We were unable to investigate this possibility as accepted fatness indicators, e.g., Quetelet Index ($\text{mass}/\text{length}^2$), were not available for most samples. However, we did measure adipose lipid content in our analyses, which has recently been suggested to be an adequate marker of fatness in polar bears (61). In contrast, other studies have demonstrated that an increase in fatness of a polar bear is due solely to an increase in adipocyte number not volume (62), which would not lead to an increase in adipose lipid content. Regardless, we did not find a significant correlation of adipose lipid content with year or with ice breakup date.

We expect that this observed temporal change in the WHB polar bear diet/food web, which resulted in increased contaminant levels over the 1991–2007 period (relative to levels had diet not changed), may continue with increasingly warmer temperatures and diminished sea ice under predicted climate change scenarios (3). To date, the biological impact of climate change has largely focused on direct changes to ecosystems (2). Yet, our results indicate that there is a need to further investigate the interrelationships between climate change and other ecological stressors, and perhaps more urgently so in vulnerable populations and species of the Arctic and Subarctic.

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Supporting Information Available

Table S1 provides results of the ranked correlation analysis of dietary tracers versus ice breakup date. Table S2 provides biometric data, sample collection date, ice breakup date, and polar bear mean $\delta^{13}\text{C}$, FA-Index1, and FA-Index2 for the sampling years. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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