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INFLUENCE OF CARBON AND LIPID SOURCES ON VARIATION OF MERCURY AND OTHER TRACE ELEMENTS IN POLAR BEARS (*URSUS MARITIMUS*)

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Abstract—In the present study, the authors investigated the influence of carbon and lipid sources on regional differences in liver trace element (As, Cd, Cu, total Hg, Mn, Pb, Rb, Se, and Zn) concentrations measured in polar bears (*Ursus maritimus*) ($n = 121$) from 10 Alaskan, Canadian Arctic, and East Greenland subpopulations. Carbon and lipid sources were assessed using $\delta^{13}C$ in muscle tissue and fatty acid (FA) profiles in subcutaneous adipose tissue as chemical tracers. A negative relationship between total Hg and $\delta^{13}C$ suggested that polar bears feeding in areas with higher riverine inputs of terrestrial carbon accumulate more Hg than bears feeding in areas with lower freshwater input. Mercury concentrations were also positively related to the FA 20:1n-9, which is biosynthesized in large amounts in *Calanus* copepods. This result raises the hypothesis that *Calanus glacialis* are an important link in the uptake of Hg in the marine food web and ultimately in polar bears. Unadjusted total Hg, Se, and As concentrations showed greater geographical variation among polar bear subpopulations compared with concentrations adjusted for carbon and lipid sources. The Hg concentrations adjusted for carbon and lipid sources in Bering–Chukchi Sea polar bear liver tissue remained the lowest among subpopulations. Based on these findings, the authors suggest that carbon and lipid sources for polar bears should be taken into account when one is assessing spatial and temporal trends of long-range transported trace elements. Environ. Toxicol. Chem. 2012;31:2739–2747. © 2012 SETAC

Keywords—Polar bear Mercury Trace elements Spatial trends Carbon and lipid sources

INTRODUCTION

The polar bear (*Ursus maritimus*) is an apex predator of circumpolar arctic marine ecosystems. Due to its trophic position, the polar bear may be exposed and in some cases have elevated levels of a wide variety of environmental contaminants [1,2]. These contaminants include trace elements such as mercury (Hg), but also essential (As, Cu, Mn, Se, and Zn) and nonessential (Cd, Hg, Pb, and Rb) elements of both natural and anthropogenic origin, many of which are toxic at elevated concentrations [3]. Mercury and Cd have been detected at high concentrations in species occupying the top of the arctic marine food webs including polar bears [3–6]. Toxicological effects of these trace element contaminants in the Arctic have been of concern in species such as polar bears, seals, beluga whales (*Delphinapterus leucas*), and bowhead whales (*Balaena mysticetus)* [7–12]. Of the different chemical forms of Hg, the most important from an environmental toxicology perspective is methylmercury (MeHg). Methylmercury biomagnifies through food chains, and more than 95% has been shown to be absorbed from the diet in exposed mammals [13]. In recent years, increasingly subtle but important biological effects have been documented, including behavioral, neurochemical, hormonal, and reproductive changes in predatory fish and wildlife exposed to environmentally relevant levels of MeHg [14].

Trace element concentrations show wide geographical variation among polar bear subpopulations. Studies published over the last 25 years have documented the highest concentrations of total Hg, Se, and As in polar bears from the Beaufort Sea and lowest in southern and western Hudson Bay and the Chukchi–Bering Sea [3,6,15–17]. In contrast, Cd concentrations in polar bears and ringed seals generally increase from the western to eastern Arctic [3,6,15–18]. Trace element concentrations in arctic biota are influenced by physical factors including riverine output and geology, as well as biological factors such as underlying food web structures that are manifested in the diet of higher trophic level wildlife [18–20]. The relative importance of these underlying factors in modulating geographical differences in trace element concentrations in polar bears and other marine mammals is not completely understood.

The food web length and diet composition of polar bears are known to vary considerably among Arctic subpopulations [21–23]. For example, polar bears feed predominantly on ringed seals (*Pusa hispida*), but depending on the subpopulation, bearded seals (*Erignathus barbatus*), harp seals (*Phoca groenlandica*), harbor seals (*Phoca vitulina*), hooded seals (*Cystophora cristata*), walruses (*Odobenus rosmarus*), narwhals (*Monodon monoceros*), belugas, bowhead whales, and sperm whales (*Physeter macrocephalus*) may also form part of their diet [21,24]. It is, however, unclear how such trophic factors may influence spatial variation in trace element levels.

All Supplemental Data may be found in the online version of this article.

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Chemical tracers, such as nitrogen and carbon stable isotope (SI) ratios ($\delta^{15}N$, $\delta^{13}C$) and fatty acid (FA) composition, have been used to elucidate trophic relationships and food web structure [25,26]. For estimating relative trophic positions of animals within a food web, $\delta^{15}N$ is frequently used, whereas $\delta^{13}C$ delineates the major carbon sources of an organism, that is, benthic/pelagic, marine/terrestrial, and freshwater/marine [25]. The FAs may be used to assess the individual diet composition of animals [26]. In addition, transfer of energy from phytoplankton and zooplankton to top predators may be traced using FAs [27], because characteristic FAs synthesized in primary and secondary producers are transferred through food webs [28]. Carbon and nitrogen SIs have become powerful tools to study dietary exposure and biomagnification of persistent contaminants in marine ecosystems [29], whereas FA composition has been used in contaminant studies only recently, including studies of persistent organic pollutant variation among polar bear subpopulations [23,30–32].

We recently reported that concentrations of major bioaccumulative trace elements showed significant variation among subpopulations of polar bears from Alaska to East Greenland [3]. In the present study, we used SI and FA chemical tracers to test the hypothesis that among these same polar bear subpopulations, differences in trace concentrations of essential and nonessential elements are affected by variations in carbon and lipid sources. Before combining the information on trace elements and carbon and lipid sources, we investigated subpopulation differences in carbon and lipid sources assessed using $\delta^{13}C$ ratio values and fatty acid signatures, respectively.

MATERIALS AND METHODS

Sample collection and age estimation

Polar bear liver, muscle, and subcutaneous fat tissues were collected from eight subpopulations in the Canadian Arctic, as well as from the Alaska (Chukchi–Bering Sea) and East Greenland subpopulations over the years 2005 to 2008 (Fig. 1, Table 1). A vestigial premolar tooth was used to estimate the age of the bears [33,34]. Detailed information on sampling and biometric measurements has been comprehensively reported elsewhere [34].

Trace element analysis and quality control

All trace element analysis of the present polar bear liver samples was carried out at the National Wildlife Research Centre, Environment Canada, Carleton University. Polar bear liver samples were analyzed for As, Cd, Cu, total Hg, Mn, Rb, Pb, Se, and Zn. We have thoroughly and recently described [3] the analytical procedures and quality assurance and control used for trace element determination in the liver samples (see Supplemental Data). Briefly, total Hg, As, Cd, Cu, Mn, Pb, Rb, Se, and Zn were determined in polar bear liver samples using U.S. Environmental Protection Agency method 200.8 with modifications for biological samples. Concentrations of total Hg were determined by DMA-80 Direct Mercury Analyzer (Milestone), and other elements were determined by ELAN 9000 inductively coupled plasma mass spectrometry from Perkin Elmer. Recoveries of total Hg for certified reference materials varied between 83 and 111%. Average certified reference material recoveries of other elements ranged from 81% for Cu to 104% for Se. Standard deviations between duplicate results for random liver samples were from 1 to 11% for Hg, below 10% for Cd, Cu, Mn, Rb, Se, and Zn, from 1 to 19% for As, and from 1 to 14% for Pb.

Stable isotope and fatty acid analysis

Carbon source variation of individual polar bears was investigated using $\delta^{13}C$ ratio values. All carbon SI analyses were carried out by the Environmental Isotope Laboratory, University of Waterloo (Waterloo, ON, Canada). We have described the analytical procedures in comprehensive detail for all bears

Fig. 1. Polar bear subpopulation ranges throughout the circumpolar basin. Labeled subpopulations denote those examined in the present study: Bering-Chukchi Sea (CB), southern Beaufort Sea (SB), northern Beaufort Sea (NB), Lancaster/Jones Sound (LJS), Gulf of Boothia (GB), western Hudson Bay (WHB), southern Hudson Bay (SHB), Baffin Bay (BB), Davis Strait (DS), and East Greenland (EG). [Color figure can be seen in the online version of this article, available at wileyonlinelibrary.com]

Table 1. Subpopulation, number of individuals, age, and stable isotope ratio of carbon[a] of polar bears investigated for the influence of dietary tracers, sex, and age on trace element concentrations

Subpopulation	No. (males:females)	Median age (range)	$\delta^{13}C \pm SD$[a]
Alaska, Bering–Chukchi Sea (CB)	11 (7:4)	7.0 (2–22)	-16.8 ± 0.3
Southern Beaufort Sea (SB)	10 (7:3)	9.0 (4–20)	-19.1 ± 0.4
Northern Beaufort Sea (NB)	24 (17:7)	6.0 (3–24)	-19.2 ± 0.6
Gulf of Boothia (GB)	6 (4:2)	8.5 (3–24)	-17.5 ± 0.5
Lancaster/Jones Sound (LS)	12 (10:2)	6.0 (3–11)	-17.6 ± 0.5
Southern Hudson Bay (SHB)	12 (8:4)	9.0 (3–22)	-18.9 ± 0.3
Western Hudson Bay (WHB)	11 (8:3)	7.0 (3–29)	-18.8 ± 0.6
Baffin Bay, N.E. Baffin Island (BB)	10 (7:3)	5.0 (2–10)	-17.5 ± 0.6
Davis Strait, S.E. Baffin Island (DS)	5 (5:0)	4.0 (3–6)	-16.6 ± 0.7
E. Greenland, Scoresbysund (EG)	20 (14:6)	6.5 (3–19)	-18.7 ± 0.3
All females	34	7 (2–24)	-18.6 ± 1.0
All males	87	7 (3–29)	-18.2 ± 1.0

[a] McKinney et al. [23].

in the present study [23]. For SI analysis, polar bear muscle tissues were homogenized and lipids were removed and prepared for analysis by standard protocols (e.g., Hebert et al. [35]). Carbon SIs were determined with an elemental analyzer coupled to a continuous flow isotope ratio mass spectrometer. The SI results were generally corrected using carbon standards International Atomic Energy Agency-CH6 (sugar), Environmental Isotope Laboratory-72 (cellulose), and Environmental Isotope Laboratory-32 (graphite). The error for clean ball-milled standard material was $\pm 0.2\%$. Mean deviation of duplicate SI analysis on 10% of the polar bear samples was 0.07%. Carbon compositions were calculated based on Carlo Erba Elemental Standards B2005, B2035, and B2036 with an error of $\pm 1\%$. See Supplemental Data for more detail.

All FA analyses for the polar bear subcutaneous fat tissue and quality control samples were carried out by the Organic Contaminants Research Laboratory at the National Wildlife Research Centre and are described elsewhere [23,30] (see Supplemental Data). Extraction and analysis of FAs was from 10 to 20 mg of inner adipose tissue of a collected polar bear sample. The 5-α-cholestane was used as internal standard. Extracted FAs were methylated via the Hilditch reagent to fatty acid methyl esters (FAMEs). The FAMEs were determined by gas chromatography–flame ionization detection, with quantification against a Supelco, 37-component FAME external standard. Here, we report only on the dietary FAs, that is, those that are incorporated relatively unchanged from prey to monogastric predator adipose tissues [26] that were available for quantification based on the external standard. Each FAME was calculated as the mass percentage of total dietary FAME. The 12 FAs used in the present study included linoleic acid (18:2n-6), γ-linolenic acid (18:3n-6), *cis*-11-eicosenoic acid (20:1n-9), α-linolenic acid (ALA; 18:3n-3), *cis*-11,14-eicosadienoic acid (20:2n-6), cis-8,11,14-eicosatrienoic acid (20:3n-6), erucic acid (22:1n-9), *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).

A blank, duplicate, and two reference materials, Great Lakes herring gull (*Larus argentatus*) egg pool and the National Institute of Standards and Technology pilot whale blubber standard reference material 1945, were extracted with every batch of 20 FA samples. Relative differences in duplicate analyses of samples were on average 6 and 7%, respectively. The relative standard deviation of dietary FAs averaged 6% for the herring gull egg pool. The SRM1945 dietary FA values were on average within 15% relative standard deviation of our laboratory results from the 2007 National Institute of Standards and Technology/National Oceanic and Atmospheric Administration Interlaboratory Comparison Exercise Program for Organic Contaminants in Marine Mammal Tissues. Recovery of 5-α-chlolestane was $100 \pm 10\%$. See Supplemental Data for more details.

Data analysis

Statistical analysis were carried out using R Version 2.11.1 [36]. To investigate subpopulation differences in lipid sources, FA composition was explored by correspondence analysis run on the 12 FAs [37]. The FA indexes ($FA_{ind}1$ and $FA_{ind}2$) were generated for further analysis from the first and second axis of a correspondence analysis run on the 12 FAs. Sample scores were plotted by subpopulation, sex, and season. Season refers to autumn (October–November), winter (December–March), and spring (April–May). Variation of carbon source using $\delta^{13}C$ as a tracer, among polar bear subpopulations, was tested by an analysis of variance. To explore whether the differences in trace concentrations of essential and nonessential elements are affected by variation in carbon and lipid sources, we visualized the relationships between chemical tracers and trace element concentrations using the multivariate redundancy analysis (RDA) [38]. The analysis was made on R package ade4 and based on the covariance matrix of centered log-transformed trace element concentrations. Prior to analysis, the explanatory measures of lipid and carbon source variation ($FA_{ind}1$, $FA_{ind}2$, $\delta^{13}C$) were standardized. Age was also added as an explanatory variable because preliminary analysis revealed significant relationships between age and trace element concentrations. Because Pb was detected at concentrations close to the minimum detection limit in 45% of the samples, it was excluded from the final RDA to avoid variables associated with high uncertainty dominating the ordination. The RDA model was highly significant based on the Monte-Carlo permutation test (1,000 replicates, RV coefficient 0.18, $p = 0.001$). Sample scores were plotted by subpopulation, sex, and season. Correlations based on ln-transformed data are shown in the text as Pearson correlation coefficients (r) with 95% confidence intervals.

We used linear models (multiple regressions) to quantify the relationships between trace element concentrations and carbon and lipid sources. In detail, we investigated the effect of SI and FA values on the concentrations of individual trace elements,

which were related to chemical tracers according to the RDA. We selected the most parsimonious linear models explaining the variance of trace element concentrations using likelihood ratio tests. Full models, including lipid and carbon source descriptors ($\delta^{13}C$ and $FA_{ind}1$, $FA_{ind}2$), sex, and age as explanatory variables, were simplified by eliminating interaction terms and variables if their removal did not result in a significant increase in deviance. Throughout the analysis, diagnostic plots of residuals were used to verify that the linear model assumptions were met, that is, most importantly, constant variance between residuals. The trace element concentrations were ln-transformed to meet model assumptions. The analytical diagnostics revealed one outlier in the model for As concentrations, and removal of the outlier did not affect the significance of the results. Parameter estimates (β) with 95% confidence intervals are given in the text.

Furthermore, we investigated whether polar bear subpopulation differences in trace element concentrations are affected by variation in the carbon and lipid sources. We adjusted the trace element concentrations for the lipid and carbon source descriptors and other variables (sex and age), if these were included in the most parsimonious models (Table 2). We then compared the adjusted element levels with those we reported previously for the unadjusted levels in the same individuals [3]. We tested the effect of carbon and lipid source variation on pairwise differences between subpopulations by running a post hoc Tukey's honestly significant difference test after and before adjusting the trace element concentrations for carbon and lipid source descriptors. After adjusting for carbon and lipid source descriptors, this means that the trace element concentrations were adjusted according to all the variables included in the most parsimonious models (Table 2). Before adjusting for carbon and lipid source descriptors, this means that the trace element concentrations were adjusted only to other variables (age and sex) if these were included in the most parsimonious models (Table 2). Level of significance was set to $\alpha \leq 0.05$.

RESULTS AND DISCUSSION

Regional variation in carbon and lipid sources

The $\delta^{13}C$ signatures were depleted in the northern and southern Beaufort Sea, Hudson Bay, and East Greenland subpopulations compared with the remaining subpopulations (analysis of variance, $F_{9,111} = 40$, $p < 0.001$, Table 1). The $\delta^{13}C$ values are enriched going from terrestrial/freshwater organic matter to pelagic phytoplankton to ice algae and benthos [25]. The depleted $\delta^{13}C$ signatures in the polar bears from the Beaufort Sea and Hudson Bay may originate from input of terrestrial organic carbon by rivers including the Mackenzie River, running into the Beaufort Sea, and several rivers feeding the Hudson Bay basin [39,40]. Although Hudson Bay polar bears spend prolonged seasonal periods on land, incorporation of terrestrial-based carbon from feeding on berries has been suggested to be a minor part of their carbon bulk [41]. The depleted $\delta^{13}C$ values in East Greenland polar bears may be related to the phenomenon that freshwater from the Arctic Ocean originating mainly from Russian and Canadian river runoff is strongly confined nearly to the East Greenland coast, as most of the polar sea ice is transported southward along the East Greenland shores, where it melts as it meets with warmer ocean currents [42].

Table 2. Explanatory variables including degrees of freedom (*df*) and r^2 for the most parsimonious linear models for $\log_e$-transformed trace element concentrations

	$\delta^{13}C$	$FA_{ind}1$	$FA_{ind}2$	Age	*df*	r^2
log(As)	X	X	X		3,115	0.29
log(Cd)			X		1,118	0.09
log(Hg)	X	X	X	X	4,115	0.31
log(Se)	X	X	X	X	4,115	0.28

FA_{ind} = fatty acid index.

Major FAs included 20:1n-9, 20:5n-3, 22:5n-3, 22:6n-3, and 18:2n-6 as presented in detail by McKinney et al. [23]. These FAs originate from pelagic herbivorous plankton and phytoplankton such as *Calanus* copepods, diatoms, dinoflagellates, and *Phaeosystis pouchetii* [28]. The FA composition in polar bears thus suggests that polar bear diet is coupled mainly to the pelagic marine food web, which is in agreement with previous carbon SI estimations reported by Hobson et al. [43]. The first axis explaining 64% of the FA variation distinguished mostly between 20:1n-9 and 22:1n-9, and 20:5n-3 (Fig. 2A). Correspondence analysis indicated that FA composition differed among the polar bear subpopulations, whereas the variation between seasons and sexes was minor (Fig. 2B–D). This is similar to previous results reported by McKinney et al. [23]. The first axis separated Hudson Bay and Chukchi Sea polar bears mainly from the other subpopulations, and showed a strong correlation with latitude. The bears from lower latitude Hudson Bay and Chukchi Sea populations had lower proportions of 20:1n-9 and 22:1n-9 compared with the polar bears from higher latitudes. The FAs 20:1n-9 and 22:1n-9 are biosynthesized by *Calanus* copepods and used as specific markers for this taxon [28], which is the major zooplankton taxa in the high Arctic [44,45]. Our results suggest that *Calanus* copepods are proportionally greater in the polar bear food webs in higher latitudes compared with those from lower latitudes. This is in accordance with differences reported elsewhere in zooplankton composition among Hudson Bay, Chukchi Sea, and the remaining areas [44–47].

Relationships between carbon and lipid sources and trace elements

Concentrations of Hg, Se, As, and Cd were related to FAs, $\delta^{13}C$, or both based on the ordination plot derived from the RDA (Fig. 3A). Plotting sample scores revealed that the relationships between trace elements were mainly related to subpopulation differences (Fig. 3B), whereas sex and season had minor influence on these relationships (Fig. 3C,D).

Hg concentrations were negatively related to $\delta^{13}C$ (Fig. 3A; $\beta = -0.27$ [-0.45, -0.09]), which is in accordance with a previous report on Hg in polar bears from western Hudson Bay and southern Beaufort Sea [22]. As mentioned earlier, depleted $\delta^{13}C$ signatures may originate from terrestrial organic carbon transported by rivers. Riverine transport is also a major source of Hg to the Arctic Ocean [48]. Thus, polar bear food webs rich in river-exported carbon may lead to elevated total Hg concentrations in polar bears. Concentrations of total Hg also showed a positive relationship with $FA_{ind}1$ (Fig. 3A; $\beta = 1.5$ [0.9, 2]), which was positively loaded with the *Calanus* FA marker 20:1n-9 (Fig. 2A). This raises the question of whether food webs rich in *Calanus* accumulate more Hg compared with food webs deficient in *Calanus*. Previous studies on beluga whales found positive correlations between total Hg concentrations in liver and muscle and long chain monounsaturated fatty acids (20:1n-7, 20:1n-9, 22:1n-9, and 22:1n-11) [32], which are biosynthesized in *Calanus*-copepods [28]. To our

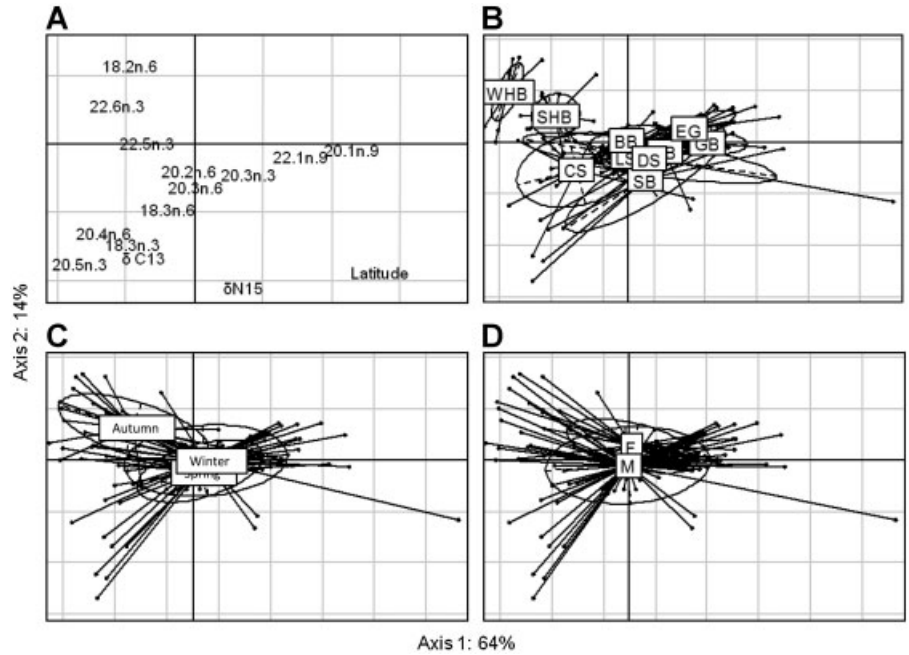

Fig. 2. Ordination plots from correspondence analysis based on mass percentage of total dietary fatty acids (FAs) in polar bear subpopulations (**A**). Sample scores are grouped by subpopulation (**B**), season (**C**), and sex (**D**). The first and second axes explained 64 and 14% of the total variation, respectively. Latitude and $\delta^{13}C$ are shown as supplementary variables. See Figure 1 for abbreviation definitions.

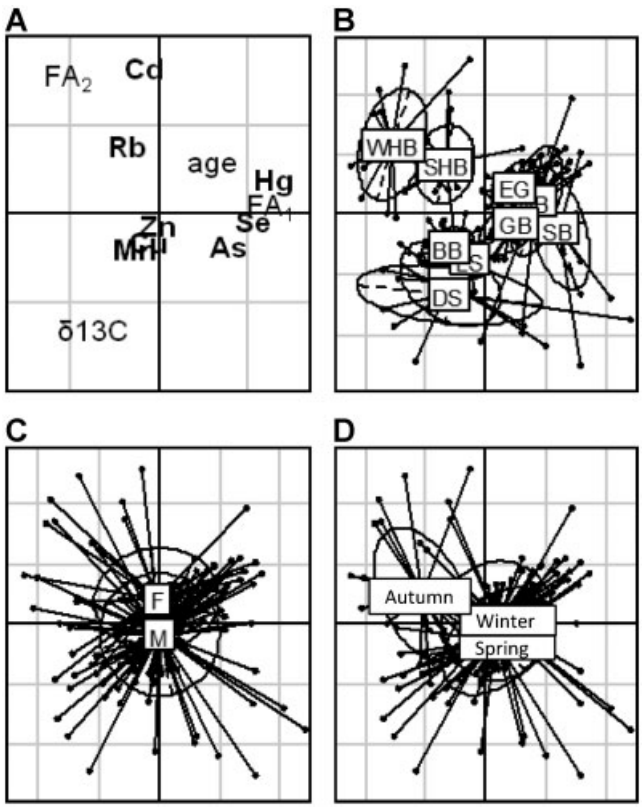

Fig. 3. Ordination plots from redundancy analysis (RDA) based on covariance matrix of log-transformed trace element concentrations in the liver of polar bears. The relationships are shown between response variables (trace element concentrations) and explanatory variables (chemical tracers and age) (**A**). The sample scores are grouped by subpopulation (**B**), sex (**C**), and season (**D**). The first linear combination of the explanatory variables explained 33% of the response variables and the second, 16%. The first axis explained 89% of the variation and second axis, 8.7%. See Figure 1 for abbreviation definitions.

knowledge, there is no comparative literature about Hg uptake by *Calanus* compared with the major zooplankton taxa from the subarctic. *Calanus* may be exposed to relatively high Hg levels due to its foraging ecology. The key *Calanus*-copepod in Arctic shelf seas, *C. glacialis*, times its foraging to the ice algal bloom in April, whereas their offspring feeds on the phytoplankton bloom following the sea-ice break-up [49]. Interestingly, Hg concentrations in sea-ice brines are highest in April and decrease with the progressing melting season when melt water flushes the brine into the underlying seawater [50]. Concentrations of Hg were positively related to age ($\beta = 0.0478$ [0.016, 0.080], which is in accordance with previous studies on polar bears [4,17].

Strong positive correlations of Se and As were found with total Hg ($r = 0.97$ [0.95, 0.98] and 0.80 [0.72, 0.85], respectively), which were thus positively related to $FA_{ind}1$ and negatively to $\delta^{13}C$. Concentrations of Se were also positively related to age. Strong correlations between Hg and Se have been reported by numerous wildlife studies, which are probably related to a detoxifying effect of Se on Hg [12]. Mercury forms the equimolar inert tiaminite Se complex Hg:Se [51], and the molar ratio of Hg:Se varied between 0.8 to 1.6 in the present polar bears, which is at a similar range as previously observed in polar bears [6,52]. The underlying reason for the positive correlation of As with either Hg or Se is not clear. First, biomagnification potency of As has not been demonstrated in contrast to Hg [53–56]. Second, although arsenite (As^{3+}) and Se may interact by forming an equimolar complex with glutathione [51], the major As form in marine mammals (seals) and birds is arsenobetaine [57], which to our knowledge is not known to interact with Se. However, different forms of As have not been investigated in polar bears, and further research is warranted to investigate the underlying reason for strong correlations of As with Hg and Se.

Concentrations of Cd were positively related to $FA_{ind}2$ (Fig. 3A; $\beta = 1.5$ [0.6, 2.3]), which loaded negatively with

FA 20:4n-6 (Fig. 2A). This suggests enrichment of Cd from near-shore to the pelagic environment, because 20:4n-6 is synthesized in macro-algae growing in shallow waters (<12 m) [58]. This is in agreement with increasing Cd levels from the inner fjord system toward the open sea in, for example, the Greenland environment [59]. In the case of Cu, Mn, Rb, and Zn, these were not related to carbon or lipid source descriptors (Fig. 3A). Thus, Cu, Mn, and Zn are all essential elements, and their uptake is naturally regulated by organisms, whereas the role of Rb as a micronutrient has been discussed [60].

Influence of carbon and lipid sources on subpopulation differences in trace element concentrations

When adjusted for carbon and lipid sources as measured by $\delta^{13}C$ and FA tracers, total Hg concentrations were significantly different from concentrations that were not adjusted (Fig. 4). Mean total Hg concentrations were higher in southern and western Hudson Bay polar bears and lower in southern and northern Beaufort Sea subpopulations when adjusted for $\delta^{13}C$ and FA tracers compared with unadjusted concentrations. We recently reported that total Hg concentrations in polar bears from northern and southern Beaufort Sea were higher than in any other subpopulations except Lancaster/Jones Sound and the Gulf of Boothia, whereas total Hg concentrations were lower in southern and western Hudson Bay polar bears compared with any other subpopulation except those of the Davis Strait and Chukchi Sea [3]. The subpopulation differences of concentrations of total Hg adjusted for carbon and lipid sources were less pronounced than unadjusted trends among these subpopulations. Adjusted total Hg concentrations in southern and northern Beaufort Sea polar bears were generally not higher compared with other subpopulations ($0.12 < p < 1$; $p = 0.06$ for northern Beaufort Sea–East Greenland; southern Beaufort Sea–southern Hudson Bay, $p = 0.001$), except for Chukchi Sea ($p < 0.001$). Total Hg concentrations adjusted for carbon and lipid sources in polar bears from western Hudson Bay were similar to all the subpopulations ($0.15 < p < 1$) and higher than in Chukchi Sea polar bears ($p = 0.042$). Adjusted total Hg concentrations for the southern Hudson Bay subpopulation were still lower compared with polar bears from northern Beaufort Sea and Lancaster/Jones Sound ($p \geq 0.001$). Because Se and total Hg concentrations were strongly correlated, concentrations of Se adjusted for carbon and lipid sources were less pronounced compared with unadjusted trends among these subpopulations (Fig. 4). Concentrations of As adjusted for carbon and lipid sources were, in general, similar between subpopulations ($p > 0.095$; East Greenland–northern Beaufort Sea $p = 0.024$). However, subpopulation differences were observed in the unadjusted As concentrations [3] (Fig. 4). Prior to adjustments for lipid source, concentrations of Cd in polar bears generally increased from east to west [3]. However, after adjustments, this trend was less pronounced (Fig. 4).

Our results suggest that differences in Hg, Se, As, and Cd concentrations among polar bear subpopulations are partly explained by variation in carbon and lipid sources. Low concentrations of total Hg adjusted for carbon and lipid sources in Chukchi Sea polar bears may be related to Hg concentrations in water. Dissolved gaseous Hg in surface waters from the Chukchi–Bering Sea has also been reported to be low compared with the remaining Arctic [61]. The Chukchi–Bering Sea is influenced by inflow of water from the Pacific Ocean, where evasion of gaseous mercury is not blocked by ice cover [62].

Confounding factors—methylated Hg species in water column and food web length

A recent study comparing total Hg levels in the hair of polar bears from western Hudson Bay and the southern Beaufort Sea concluded that the differences in total Hg levels between these two subpopulations may be related to both the length of the food web and pelagic concentrations of MeHg concentrations [22]. Concentrations of MeHg in the water column vary between the Canadian Arctic archipelago and Hudson Bay, whereas total Hg concentrations are similar in the different areas within the Canadian Arctic and sub-Arctic [22,63]. Monomethylated Hg

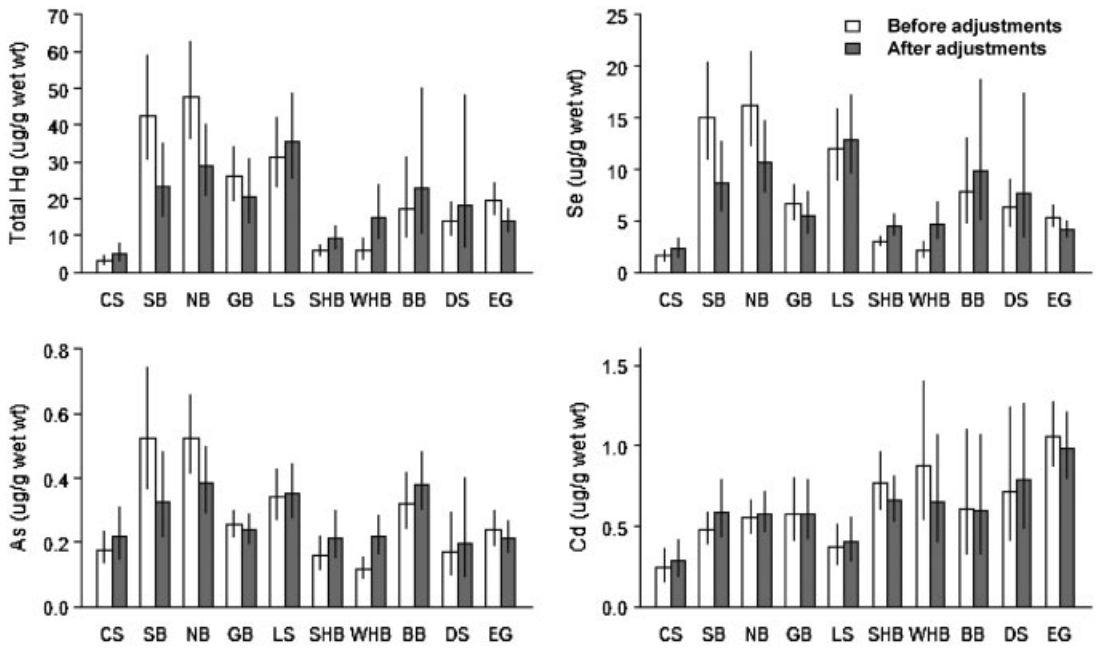

Fig. 4. Geometric mean concentrations (μg/g wet wt ± 95% confidence intervals) of total Hg, Se, As, and Cd in liver of polar bears from 10 subpopulations before [3] and after adjusting for lipid and carbon sources (see Table 2 for details). See Figure 1 for abbreviation definitions.

is the toxic form of Hg accumulating in the food web, whereas elemental Hg has poor bioaccumulation potential [64]. Although our results suggest that total Hg variation among polar bear subpopulations is mainly explained by food web differences, the results for the Canadian subpopulations may be partly confounded by regional variation in MeHg species in the water column [22,63]. We had reported that the highest concentrations of total Hg (corrected for sex and age) were in polar bears from the Beaufort Sea, Lancaster Sound, and Gulf of Boothia, followed by the Baffin Bay and Davis Strait; Hudson Bay bears had the lowest Hg concentrations [3]. Similarly, concentrations of MeHg in mid-depth and deep water column were reported to be highest in the southern Beaufort Sea and Lancaster Sound and lower for Davis Strait to Hudson Bay [22,63]. However, the geographical differences in MeHg concentrations in the water column were smaller compared with differences that we observed among the polar bear subpopulations. Furthermore, the Beaufort Sea and Baffin Bay were shown not to have different MeHg concentrations in the water column [22,63] as they did for total Hg concentrations in polar bears. Therefore, the difference that we presently observed in total Hg concentrations between Beaufort Sea and Baffin Bay subpopulations could be explained by differences in carbon and lipid sources between the Beaufort Sea and Baffin Bay.

Trophic biomagnification of Hg has been reported in both polar bears [22,65] and marine food webs [56,66]. We previously reported that $\delta^{15}N$ values varied among the polar bear subpopulations [23]. As part of the present study, we found that the $\delta^{15}N$ values were positively correlated with the concentrations of total Hg, Se, and As (Supplemental Data, Fig. S1). This finding suggests that regional differences in polar bear food web length play a role in explaining subpopulation differences in trace element concentrations. However, the trophic baseline of $\delta^{15}N$ values in Arctic marine food webs may vary significantly between geographical regions [67,68]. Thus, the differences in $\delta^{15}N$ values among polar bear subpopulations do not necessary reflect their trophic position in the food web. We recommend that future studies should investigate the role of the whole food web length in subpopulation variation of trace element concentrations in polar bears. This will require a thorough investigation of $\delta^{15}N$ values at lower trophic levels to properly adjust polar $\delta^{15}N$ values to possible variations in $\delta^{15}N$ baseline.

CONCLUSIONS

The present study demonstrates the importance of including information on carbon and lipid sources when interpreting the spatial trends of certain trace elements in polar bears. Subpopulation differences are partly explained by variation in carbon and lipid sources, but MeHg in the water column and food web length may also play an important role in total Hg concentrations in apex predators. It has been proposed that trace element concentrations in arctic apex predators may change during the next decades, as Hg and Cd emissions are expected to increase due to the increasing use of coal in Asia and worldwide [69,70]. Additionally, changing climate may be affecting the natural cycles and long-range transport of these elements [62,71], as well as access, abundance, and distribution of polar bear prey. Frequent monitoring of both polar bear food web structure and exposure to trace elements is thus important to detect possible and rapid changes.

SUPPLEMENTAL DATA

In the Supplemental Information section, comprehensive details are given on methods for stable isotopes, fatty acids, and total Hg and other elements under study. Supplemental Figure S1 shows the relationships between liver total Hg, Se, and As, and muscle stable nitrogen isotope ($\delta^{15}N$) values in polar bears from the 10 subpopulations. (37 KB DOC).

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