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Pelagic vs Coastal—Key Drivers of Pollutant Levels in Barents Sea Polar Bears with Contrasted Space-Use Strategies

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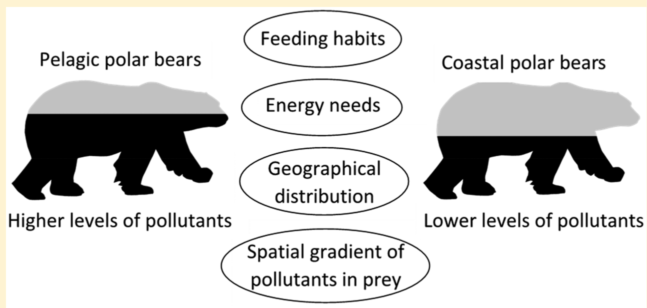
Supporting Information

ABSTRACT: In the Barents Sea, pelagic and coastal polar bears are facing various ecological challenges that may explain the difference in their pollutant levels. We measured polychlorinated biphenyls, organochlorine pesticides, polybrominated diphenyl ethers in fat, and perfluoroalkyl substances in plasma in pelagic and coastal adult female polar bears with similar body condition. We studied polar bear feeding habits with bulk stable isotope ratios of carbon and nitrogen. Nitrogen isotopes of amino acids were used to investigate their trophic position. We studied energy expenditure by estimating field metabolic rate using telemetry data. Annual home range size was determined, and spatial gradients in pollutants were explored using latitude and longitude centroid positions of polar bears. Pollutant levels were measured in harp seals from the Greenland Sea and White Sea−Barents Sea as a proxy for a West−East gradient of pollutants in polar bear prey. We showed that pelagic bears had higher pollutant loads than coastal bears because (1) they feed on a higher proportion of marine and higher trophic level prey, (2) they have higher energy requirements and higher prey consumption, (3) they forage in the marginal ice zones, and (4) they feed on prey located closer to pollutant emission sources/transport pathways.



1. INTRODUCTION

Persistent organic pollutants (POPs) are transported to remote places such as the Arctic through air and ocean currents in addition to river outflows.[1−6] Species at the top of the food web with lipid-rich diets, such as polar bears (*Ursus maritimus*), bioaccumulate relatively high concentrations of POPs.[7−11] Concomitantly, Arctic sea ice is declining at an unprecedented rate,[12] and loss of sea ice due to climate change is one of the greatest threats to polar bears.[13,14] Cumulative stress from habitat loss, reduced food availability, and exposure to pollutants could be of high significance in some polar bear populations.[15−17]

The Barents Sea polar bears experience high exposure to POPs compared to several other subpopulations.[18,19] In particular, concentrations of perfluoroalkyl substances (PFASs), mainly perfluoroalkyl acids that bind to proteins, have been detected at high concentrations in Barents Sea polar bears.[19,20] PFASs contain both emerging and legacy compounds and are broadly present in various consumer products, because of their surfactant and water-repellent properties.[21−23] The polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), followed by the polybrominated diphenyl ethers (PBDEs), are quantitatively the most abundant lipophilic compounds detected in Barents Sea polar bears.[24] PCBs and OCPs were extensively used in the past in various industrial and agricultural applications, and their use has been gradually regulated since 1970. PBDEs have been largely employed as brominated flame retardants, and their regulation has been ongoing for the past decade. Meanwhile, Arctic sea ice, which represents the main polar bear habitat for foraging, traveling, and mating,[14,25,26] is declining at the fastest recorded rate in the Barents Sea.[27] This polar bear subpopulation,

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Table 1. Estimated Pollutant Concentrations and Ecological Predictors in Pelagic and Coastal Adult Female Polar Bears from the Barents Sea (2011−2018)[a]

	n (pelagic/coastal)	estimated median ± SE for pelagic polar bears	estimated median ± SE for coastal polar bears	*p* value
		pollutants[b]		
∑CHLs (ng·g^{-1} lw)	14/24	**616.6 ± 93.0**	**375.1 ± 43.1**	**0.013**
∑PCBs (ng·g^{-1} lw)	14/24	2183.5 ± 388.3	1477.4 ± 200.2	0.089
α-HCH (ng·g^{-1} lw)	14/24	8.0 ± 1.8	9.0 ± 1.9	0.587
β-HCH (ng·g^{-1} lw)	14/24	**34.6 ± 4.3**	**24.9 ± 2.4**	**0.043**
mirex (ng·g^{-1} lw)	14/24	4.3 ± 1.0	2.7 ± 0.5	0.117
HCB (ng·g^{-1} lw)	14/24	63.1 ± 11.1	45.6 ± 6.2	0.149
p,p′-DDE (ng·g^{-1} lw)	14/24	**66.9 ± 18.5**	**30.8 ± 6.5**	**0.031**
∑PBDEs (ng·g^{-1} lw)	14/24	14.5 ± 2.1	10.3 ± 1.4	0.068
∑PFSAs (ng·g^{-1} ww)	15/25	**334.6 ± 63.4**	**224.1 ± 42.0**	**0.013**
∑PFCAs (ng·g^{-1} ww)	15/25	**121.2 ± 20.4**	**80.0 ± 13.4**	**0.003**
		ecological predictors		
δ^{13}C in RBCs (‰)	15/25	−19.4 ± 0.3	−20.9 ± 0.3	**<0.001**
δ^{13}C in hair (‰)	15/25	−18.2 ± 0.3	−18.9 ± 0.3	0.071
δ^{15}N in RBCs (‰)	15/25	**16.6 ± 0.4**	**15.3 ± 0.3**	**0.011**
δ^{15}N in hair (‰)	15/25	**18.4 ± 0.5**	**17.0 ± 0.4**	**0.030**
trophic level (from δ^{15}N-AA in RBCs)	15/25	3.1 ± 0.2	2.8 ± 0.1	0.099
trophic level (from δ^{15}N-AA in hair)	15/25	3.1 ± 0.2	2.6 ± 0.2	0.157
field metabolic rate (kJ·kg^{-1}·day^{-1})	15/25	**267.9 ± 5.7**	**207.1 ± 5.0**	**<0.001**
home range size (km^2)	15/25	**190 092 ± 52 865**	**63 452 ± 53 004**	**<0.001**
latitude centroid	15/25	**N 79.8 [79.1−80.4]**	**N 77.5 [76.6−78.3]**	**<0.001**
longitude centroid	15/25	**E 41.6 [38.9−44.7]**	**E 29.1 [27.7−30.6]**	**<0.001**
body condition index	15/25	−1.0 ± 0.1	−1.2 ± 0.1	0.280

[a]Pelagic and coastal polar bears were compared using linear mixed-effect models with "sampling year" as a random factor. Significant differences are shown in bold. OCs and PBDEs have been measured in adipose tissue and PFASs in plasma. [b]Pollutants were ln transformed to meet model assumptions.

shared between Norway and Russia, is currently under multiple stressors that might act in synergy.[15,16,28,29]

There are two ecotypes of Barents Sea polar bears with distinct space-use strategies, individually stable movement patterns, and high site fidelity over years.[30,31] The "pelagic bears" undertake long annual migrations following the ice retreat toward the northeastern part of the Barents Sea, while the "coastal bears" stay on land or on land-fast ice year round at the western part of the Barents Sea in the Svalbard Archipelago.[30,32] The distribution of Barents Sea polar bears has shifted northward since the beginning of the 1990s due to changes in their habitat and in the abundance and distribution of their main prey.[14,25,33−36] Polar bears depend on sea ice as a platform for hunting and preferentially feed on ringed seals (*Pusa hispida*), bearded seals (*Erignathus barbatus*), and harp seals (*Phoca groenlandica*).[37−39] However, in the absence of sea ice, Barents Sea polar bears can feed opportunistically on alternative food sources such as ground-nesting bird, seabirds, bird eggs, reindeers, whale carcasses, algae, and even vegetation.[37,39−41] The two ecotypes of the Barents Sea are currently facing very different ecological challenges. The migration routes of pelagic bears following the marginal ice zone are getting longer, whereas longer ice-free periods in the Svalbard area force coastal bears to feed on land-based prey.

Previous studies have shown marked differences in pollutant levels between the two ecotypes, with the pelagic polar bears generally having higher pollutant levels than the coastal ones.[42−44] However, the underlying reasons for these differences in pollutant concentrations are largely unknown. Multiple factors can drive these differences including feeding habits, energy expenditure, proximity to emission sources, transport routes, and abiotic factors.[42,44−46] Tartu et al.[44] showed that pelagic females had a higher diet selectivity than the coastal females based on bulk stable isotope ratios of carbon (δ^{13}C) and nitrogen (δ^{15}N) in red blood cells. However, in order to correctly interpret stable isotope data in predators, the base of the food web (baseline) needs to be constrained. Determining and obtaining baseline stable isotope values can be problematic in animals that forage widely, such as polar bears. Nitrogen stable isotope of amino acids (δ^{15}N-AA) can overcome this issue by indirectly fingerprinting the base of the food web, as it conservatively traces δ^{15}N of primary producers. Simultaneously, trophic amino acids (trophic AA), which become enriched during trophic transfer, can be used to isolate a predator's trophic position.[47,48] In addition, pelagic bears occupy a wider home range,[30,42,44,49] and it has been proposed that this results in greater energetic costs, greater prey intake, and therefore higher pollutant levels.[42] Finally, higher levels of pollutants in the pelagic bears, which utilize the northeastern part of the Barents Sea to a greater extent, could be due to a spatial gradient in pollutant concentrations related to the proximity of emission sources, uptake, and/or transport routes of pollutants.[44−46]

In the present study, we investigated a suite of ecological drivers in order to decipher drivers of pollutant levels between the two ecotypes of Barents Sea polar bears. Specifically, the foraging habitat and diet were studied with bulk stable isotope ratios of carbon (δ^{13}C) and nitrogen (δ^{15}N) as proxies of feeding habits. We also used δ^{15}N-AA as a trophic indication and in order to estimate the polar bear trophic level. Using satellite telemetry data, we studied energy expenditure by estimating field metabolic rate (FMR). Annual home range

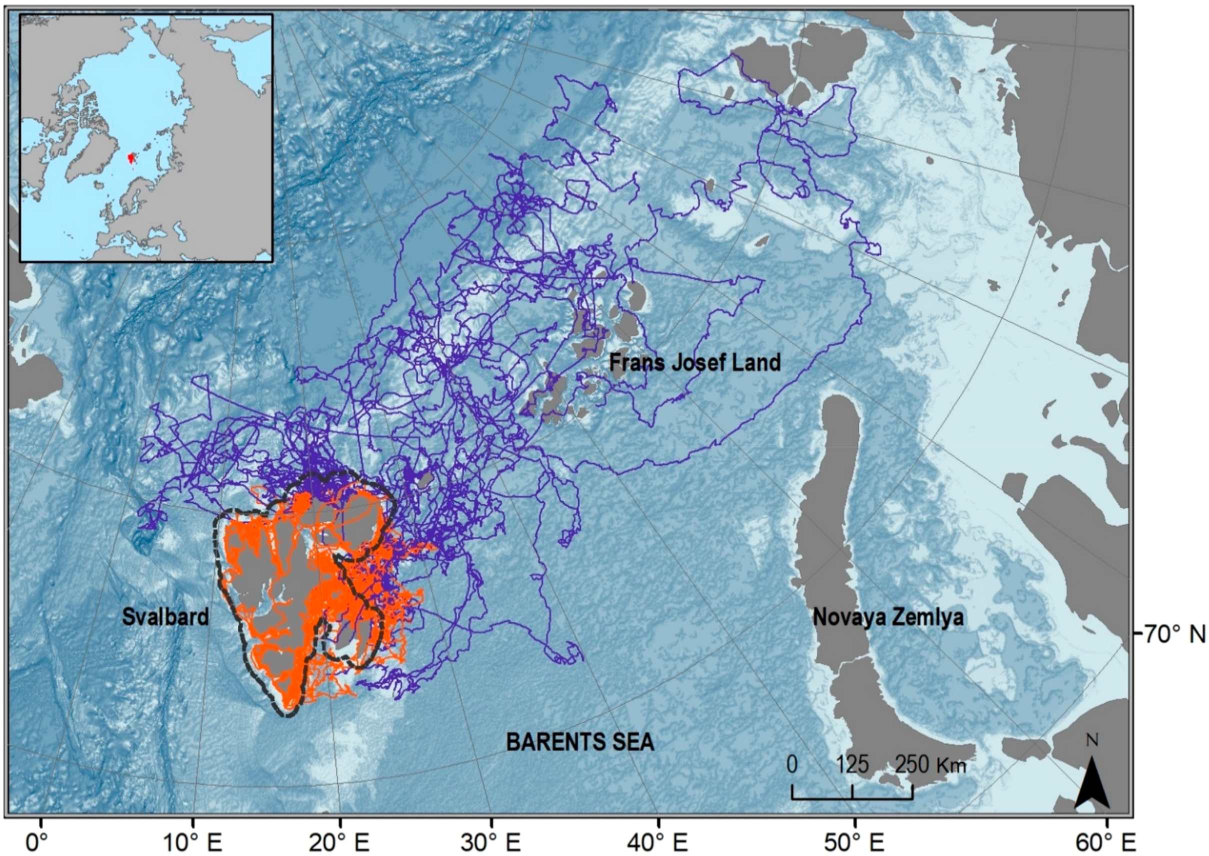

Figure 1. Map of the study area including the tracks of 40 adult female polar bears. Tracks are color coded according to their ecotype: pelagic (n = 15 in blue) or coastal (n = 25 in orange). Staple black line represents the coastal region around the Svalbard area. (Inset) Location of the Svalbard Archipelago (in red).

(HR) size was also determined, and potential spatial gradients in pollutants were explored using latitude and longitude centroid positions of polar bears. Finally, pollutant levels were measured in adult harp seals from the Greenland Sea stock and White Sea−Barents Sea stock as a proxy for a West−East gradient of pollutants in polar bear prey.

2. MATERIAL AND METHODS

Fieldwork. Adult female polar bears (n = 40; 15 pelagic and 25 coastal) from the Barents Sea were captured throughout the Svalbard Archipelago in spring (from March 29 to April 24) between 2011 and 2018 (Table S1). One female was captured twice, in 2016 and 2017, while the others were captured only once. Immobilization, sampling, and handling procedures followed standard protocols,[50,51] and are, together with methods for determination of body condition, age and reproductive status, further described in the Supporting Information (SI). As concentrations of pollutants are related to body condition and reproductive status,[24] we selected individuals with similar body condition (Table 1) and reproductive status (Table S1) for both ecotypes to avoid confounding effects of these factors.[44]

Blood and adipose tissue samples of adult harp seals of the Greenland Sea stock were collected in April 2017 (n = 3) and March 2018 (n = 7) in the pack ice of the Greenland Sea (geographical range N 69°10′−72°30, W 16°−20°). Blood and adipose tissue samples of harp seals from the White Sea−Barents Sea stock were collected in April 2018 (n = 11) in the Pechora Sea (geographical position N 69°52′, W 50°36′). Procedures for sampling and estimation of body condition are described in the SI.

Determination of Ecotype, Home Range, and Field Metabolic Rate. Annual HR size defined as the 50% minimum convex polygon (MCP) and the location of its centroid were calculated for each bear (detailed in the SI). We assigned each bear to an ecotype ("pelagic" or "coastal") based on the percentage of overlap between MCP of each individual and the Svalbard area. The Svalbard area was defined as the 4 largest islands in the Svalbard archipelago (Spitsbergen, Nordaustlandet, Edgeøya, and Barentsøya) and a 20 km buffer around each island (Figure 1). A bear was deemed "coastal" if at least one-half of its 50% yearly HR was included within the polygon (n = 25; Figure 1). By contrast, if at least 50% of the bear's HR was outside of this polygon, the bear was deemed "pelagic" (n = 15; Figure 1). Ecotype attribution was checked and validated after visual inspection of each track. The daily speed of each bear was corrected for sea ice drift following the approach taken by Durner et al.[52] (detailed in the SI). FMR was calculated for each bear based on average daily speed corrected for sea ice drift (as detailed in Blanchet et al., submitted for publication) and following the relationship in Pagano et al.:[53] Daily FMR = 167.3 × speed + 153, where daily FMR is in $kJ \cdot kg^{-1} \cdot day^{-1}$ and speed in $km \cdot h^{-1}$. Because denning events and their duration vary substantially between individuals and years, we only investigated FMR in the period between May 1 and September 30 when polar bears do not den.

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Pollutant Measurements. Organochlorine compounds (OCPs and PCBs) and PBDEs were analyzed from polar bear (n = 38) and harp seal (n = 20) adipose tissue. PFASs were analyzed in polar bear plasma (n = 40) and harp seal plasma/serum (n = 20). All analyses were conducted at the Norwegian Institute for Air Research (NILU) in Tromsø, Norway, following Scotter et al.[54] and Hansen et al.[55] Analytical procedures and quality assurance are given in the SI. We quantified OCPs (*trans-* and *cis*-chlordane, *oxy*-chlordane, *trans-* and *cis*-nonachlor, α-, β-, and γ-hexachlorocyclohexane [HCH], mirex, hexachlorobenzene [HCB], *o,p*′- dichlorodiphenyltrichloroethane [DDT], *p,p*′-DDT, *o,p*′-dichlororodiphenyldichloroethane [DDD], *p,p*′-DDD, *o,p*′-dichlorodiphenyldichloroethylene [DDE], and *p,p*′-DDE), PCBs (−28, −52, −99, −101, −105, −118, −138, −153, −180, −183, −187, −194), PBDEs (−17, −28, −47, −49, −66, −71, −77, −85, −99, −100, −119, −126, −138, −153, −154, −156, −183, −184, −191, −196, −197, −202, −206, −207, −209), perfluoroalkyl sulfonic acids (PFSAs) with 4−10 carbons (C) (both linear and branched C_8), 4:2, 6:2, and 8:2 fluorotelomere sulfonate (FTS), perfluorooctanesulfonamide (FOSA), and C_{6-14} perfluoroalkyl carboxylic acids (PFCAs). Only compounds detected in at least 60% of the samples was used for further statistical analyses, and values below the limits of detection (LOD) were replaced by 1/2 LOD. The compounds remaining for further investigation included adipose tissue concentrations of $\sum_5$CHLs, α- and β-HCH (detected in ≥60% of polar bear samples only), mirex, HCB, *p,p*′-DDE, ∑PCBs (−99, −105, −118, −138, −153, −180, −183, −187, −194), ∑PBDEs (−47, −99, −100, −153) expressed in $ng \cdot g^{-1}$ lipid weight (lw), and plasma/serum concentrations of ∑PFSAs and ∑PFCAs expressed in $ng \cdot g^{-1}$ wet weight (ww) with the following carbon chain lengths: C_{5-8} PFSAs and C_{7-13} PFCAs for polar bears and C_{6-8} PFSAs and C_{8-13} PFCAs for harp seals.

Stable Isotope Analysis (SIA). SIA was carried out mostly at the Liverpool Isotope Facility for Environmental Research (LIFER) lab in the United Kingdom and partly (26 red blood cell [RBC] samples) at the University of Alaska Anchorage in the United States. The respective roles of foraging habitat and diet were investigated in RBCs and hair using bulk SIA ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$).[56] Bulk isotopes were used to investigate isotopic niche width as a proxy of the trophic niche.[57] The $\delta^{13}C$ of a predator reflects the origin of food sources, as there is generally a good discrimination between terrestrial and marine food sources.[7,58−61] The $\delta^{15}N$ is commonly used as an indicator of the trophic position of a consumer[7,58,59] owing to the large trophic fractionation of 2−5 per mil (‰) between each trophic level.[62] We also performed a principal component analysis (PCA) on $\delta^{15}N$-trophic AA as a proxy of polar bear trophic position. Finally, polar bear trophic level was estimated from $\delta^{15}N$-AA using phenylalanine as the "source amino acid" and glutamic acid as the "trophic amino acid".[47,63] This combined approach allowed for robust trophic level estimation, taking account of potential spatial variation of the $\delta^{15}N$ baseline. Trophic level was computed according to the formula developed by Chikaraishi et al.,[48,64] adapted for marine food webs[65] (β = 2.9‰), and based on a marine mammal trophic enrichment factor[66] (TEF = 4.3‰; Harbor seal [*Phoca vitulina*]: $TL_{Glu/Phe} = [^{15}N_{Glu} - \delta^{15}N_{Phe} - 2.9]/4.3 + 1$). Therefore, $\delta^{13}C$, $\delta^{15}N$, and $\delta^{15}N$-AA are used in the present study as relevant proxies of polar bear feeding habits. RBCs are a metabolically active tissue, having half-life ≈ 1.5 months for $\delta^{13}C$ and at least twice as long for $\delta^{15}N$ in polar bears.[67] As a metabolically inert tissue, hair provides information at the time of tissue synthesis, about 6−8 months before sampling in the case when the bears were sampled in April.[68] Thus, measuring stable isotopes in both RBCs and hair samples can provide a retrospective record of polar bear feeding habits in different seasons over a larger time scale. Sample preparation, instrumental analysis, and data processing are further described in detail in the SI.

Statistical Analysis. All statistical analyses were performed using R version 3.5.1. In order to compare pollutant concentrations in both ecotypes, we used linear mixed-effect models (LMEs, "nlme" R-package, developed by Pinheiro et al.[69]) with ∑CHLs, ∑PCBs, α- and β-HCH, mirex, HCB, *p,p*′-DDE, ∑PBDEs, ∑PFSAs, and ∑PFCAs as response variables. Pollutants were ln transformed to meet model assumptions. "Sampling year" was included in each model as a random factor to account for temporal variation of pollutant levels in Barents Sea polar bears.[70,71] As suggested by Zuur et al.,[72] we used the restricted maximum likelihood estimation (REML) method to avoid any potential biased estimations. Similarly, we compared $\delta^{13}C$ and $\delta^{15}N$ signatures (in RBCs and hair), PC1 scores of $\delta^{15}N$-trophic AA (in RBCs and hair), estimated trophic level (in RBCs and hair), FMR, HR size, latitude and longitude centroids, and BCI in pelagic vs coastal polar bears. The PC1 scores of $\delta^{15}N$-trophic AA were extracted from a PCA performed on 5 trophic AA inferred from RBCs (alanine, valine, leucine, aspartic acid, glutamic acid) and 4 trophic AA from hair (alanine, proline, aspartic acid, glutamic acid). Prior to PCA, we subtracted the $\delta^{15}N$ of phenylalanine from the $\delta^{15}N$ of each trophic AA to remove potential bias due to variation in the baseline and scaled the baseline-corrected $\delta^{15}N$ values of each trophic AA using a z transformation. Higher PC1 scores indicate increasing trophic positions of polar bears. Isotopic niche widths (inferred from $\delta^{13}C$ and $\delta^{15}N$ in RBCs and hair) of both ecotypes were illustrated by standard ellipses (containing ∼95% of the data) on an isotopic biplot (Figures 2 and S1) using "SIBER" R-package.[73] The areas of the resultant ellipses were then computed using both the maximum likelihood (SEAc, adjusted for small sample size) and the Bayesian approaches (SEAb; parametrized as detailed in Jackson et al.[73]) (Figures 2 and S1). Estimated SEA values were directly compared in a probabilistic manner in terms of similarity between pelagic and coastal bears.[73] Pollutant levels

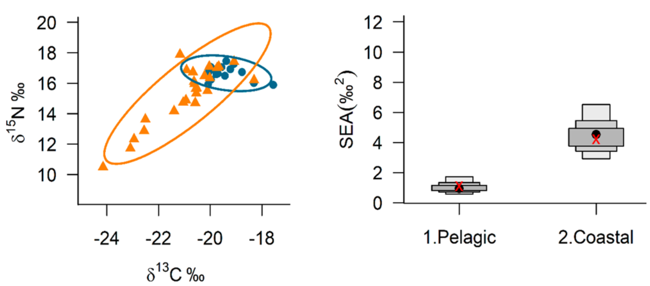

Figure 2. (A) Isotopic niche width (inferred from $\delta^{13}C$ and $\delta^{15}N$ in RBCs) illustrated by standard ellipses (containing ∼95% of the data and computed with "SIBER" R-package) for both pelagic (blue point) and coastal (orange triangle) Barents Sea polar bears (n = 40 adult females). (B) Comparison of the standard ellipse area (SEA) according to the ecotype. SEA_b is illustrated with a black point and SEA_c with a red cross.

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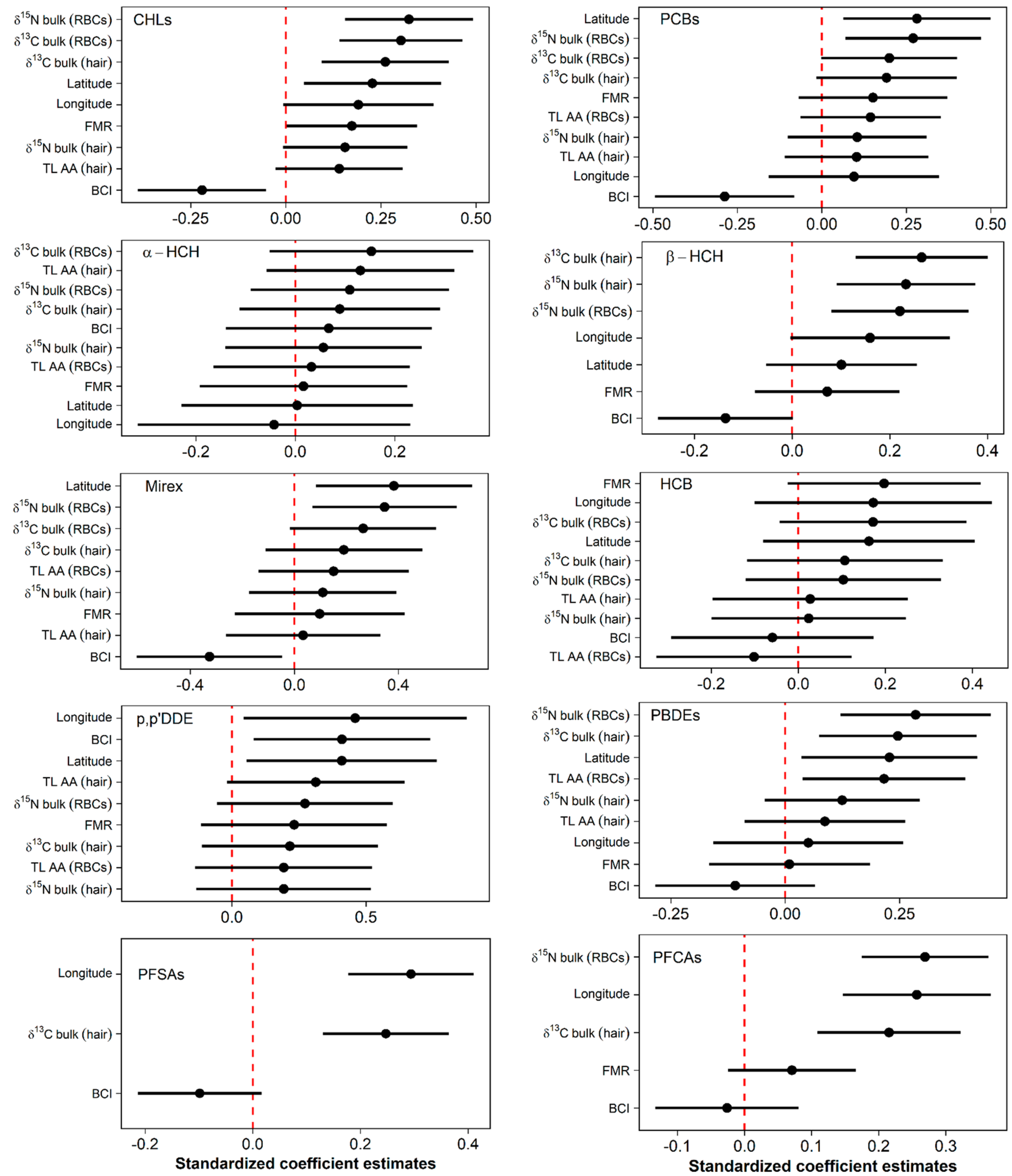

Figure 3. Effects size of δ^{13}C (in RBCs and hair), δ^{15}N (in RBCs and hair), trophic level (from δ^{15}N-AA in RBCs and hair), field metabolic rate (FMR), latitude and longitude centroids, and body condition index (BCI) on pollutant levels in adult female polar bears from the Barents Sea (2011−2018; n = 38 for OCs/PBDEs and n = 40 for PFASs). Figures illustrate model averaging outputs (conditional averaged estimates and 95% confidence interval) from the selected models. Values of pollutants were ln transformed

and body condition between the Greenland Sea and the White Sea−Barents Sea harp seals were compared with linear models.

To investigate the influence of the ecological drivers on pollutant concentrations in Barents Sea polar bears, we tested and quantified the effects of feeding habits (δ^{13}C, δ^{15}N, and estimated trophic level from δ^{15}N-AA), energetic cost (FMR), spatial gradient in pollutants (latitude and longitude centroid positions), and BCI on pollutant concentrations, regardless of which ecotype they belonged to. We used LMEs with ln-transformed ∑CHLs, ∑PCBs, *α*- and *β*-HCH, mirex, HCB, *p,p′*-DDE, ∑PBDEs, ∑PFSAs, and ∑PFCAs as response variables and δ^{13}C (both RBCs and hair), δ^{15}N (both RBCs and hair), trophic level (both RBCs and hair), FMR, latitude and longitude centroids, and BCI as predictors. "Sampling year" was included in each model as a random factor. All predictors were standardized (scaled to mean = 0 and standard deviation = 1) to facilitate the comparison of their effect size.[74] We generated a model set containing ecologically relevant submodels from the set of predictors of interest and including an intercept model (null model). Significantly correlated predictor variables were not included within the same model to minimize any collinearity concerns[75] (Table S2). This resulted in a final set of 44 competitive models (Table S3). Models (parametrized with the maximum likelihood estimation as suggested in Zuur et al.[72]) were first ranked using an information−theoretic approach based on the Akaike's

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information criterion corrected for small sample size (AICc).[76] The AIC weight (w_i) was estimated and can be interpreted as the probability that the model i is the best fit given the candidate set of models.[77] We then performed conditional model averaging (parametrized with the REML estimation as suggested by Zuur et al.[72]) from the selected models (cutoff value = cum [$\sum w_i \leq 0.95$]) as described in Grueber et al.[78] This method produces averaged estimates of all predictors, weighted according to their w_i.[76,79] For all of the predictor variables considered in the selected models, we finally determined conditional parameter-averaged estimates and 95% confidence intervals (CI). CIs provide information about the range in which the true estimate value lies with a certain degree of probability as well as the strength and direction of the demonstrated effect.[80] As a general guideline, if CIs do not cross zero, it can be assumed that the predictor significantly affects the response variable. Diagnostic plots were assessed on residuals to test whether the data met the assumptions of LMEs.

3. RESULTS AND DISCUSSION

Pollutant Levels: Pelagic vs Coastal Polar Bears. Pelagic polar bears generally had higher levels of pollutants than coastal bears (Tables 1 and S4). Median concentrations of ∑CHLs, β-HCH, *p,p*′-DDE, ∑PFSAs, and ∑PFCAs were 64%, 39%, 117%, 49%, and 52% higher in pelagic bears than in coastal bears (Table 1). With the exception of α-HCH, all other compounds investigated were higher in the pelagic bears, although these differences were not significant (Table 1). Previous studies have already highlighted similar differences in concentrations of pollutants between pelagic and coastal polar bears from the Barents Sea.[42−44] However, no such differences were reported for the lipophilic compounds measured in plasma.[44] Concentrations of lipophilic POPs are strongly related to body condition, and as Tartu et al.[44] observed that pelagic bears were fatter than coastal bears, body condition may have masked potential differences between these two ecotypes.[44]

Polar Bear Trophic Position. The trophic level estimates based on δ^{15}N values of phenylalanine and glutamic acid suggested that the Barents Sea polar bears occupy trophic level ≈ 3 (i.e., secondary consumer; Table 1), which is lower than expected for an apex predator.[7,58] δ^{15}N-AA have not been investigated in polar bears before, and so a TEF from another marine mammal species was used (i.e., Harbor seal[66]) to determine trophic level. However, TEFs have been shown to vary greatly between species,[81] and previous studies reported consistent underestimation of trophic levels inferred from δ^{15}N-AA across a range of diverse wild marine predators, likely due to the use of inappropriate TEFs.[65,66,82−86] In addition, we assumed that polar bears from this study fed mainly on marine prey and determined trophic level based on an equation developed for marine food webs. However, coastal polar bears from the Barents Sea also consume terrestrial prey,[39,41,87,88] and the use of an equation developed for terrestrial food webs would have led to higher trophic level estimations.[64,65] According to the formula developed by Chikaraishi et al.[64] for terrestrial C3 plant food webs, we found an alternative estimate for trophic level ≈ 3.5 for coastal polar bears (compared to ∼2.7). Despite the notable underestimation of polar bear trophic level, we report very high correlations between the estimated trophic level and PC1 scores of δ^{15}N-trophic AA (Figure S2), suggesting that the trophic level based on δ^{15}N values of phenylalanine and glutamic acid is a reliable trophic indicator in the present study. However, further studies are needed to define appropriate TEF and β values for polar bears.

Role of Feeding Habits. The trophic level estimates based on δ^{15}N values of phenylalanine and glutamic acid tended to be higher in the pelagic bears, but the differences were less than one trophic level (Table 1). There were no significant differences in the δ^{15}N-trophic AA scores of PC1 scores between bears from each ecotype (Figure S3, LMEs; $p = 0.142$ for RBCs and $p = 0.190$ for hair), suggesting that coastal and pelagic polar bears maintain similar trophic levels. However, δ^{13}C and isotopic niche width differed significantly between the two ecotypes (Table 1; Figures 2, S1, and S3; probability = 1 for hair and RBCs). The higher δ^{13}C values and the restricted isotopic niche of pelagic polar bears suggest a selective diet essentially or exclusively composed of marine prey (i.e., seals), whereas the lower δ^{13}C values and the wider isotopic niche of coastal polar bears suggest a mixed diet including marine and terrestrial prey. The presence of terrestrial prey in polar bears diet from Svalbard has also been shown by earlier studies.[39,41,87,88] In addition, model-averaged estimates indicated that trophic levels and diet composition determined from δ^{15}N-AA, δ^{15}N, and δ^{13}C signatures were important predictors of pollutant levels in Barents Sea polar bears (Figure 3). Concentrations of ∑CHLs, ∑PCBs, β-HCH, mirex, ∑PBDEs, and ∑PFCAs increased significantly with δ^{15}N in RBCs. Similarly, concentrations of β-HCH increased significantly and ∑CHLs tended to increase with δ^{15}N in hair (Figure 3). We also found positive trends between trophic levels inferred from δ^{15}N-AA in hair and ∑CHLs and *p,p*′-DDE, whereas ∑PBDEs increased with trophic level in RBCs (Figure 3). Finally, concentrations of ∑CHLs, β-HCH, ∑PBDEs, ∑PFSAs, and ∑PFCAs increased significantly with δ^{13}C in hair and/or RBCs, whereas concentrations of ∑PCBs and mirex tended to increase with δ^{13}C in hair and/or RBCs (Figure 3). For example, median concentrations of ∑CHLs were about 3.5 times higher in bears with a predominantly marine diet at the highest trophic level compared to the bears with a mixed diet at the lowest trophic level. Our results are in agreement with previous findings, which indicated that bears with a predominantly marine diet and higher trophic level accumulated higher concentrations of pollutants than bears at a lower trophic level, which fed on a mixed diet including terrestrial prey.[20,24,44,70]

Role of Energy Expenditure. FMR reflects the energy expenditure of polar bears during both resting and active times such as feeding and movements. FMR in pelagic polar bears was 29% higher than FMR in coastal individuals (Table 1). This is consistent with the use of larger areas as shown by the size of their HR, which were 200% larger compared to HR occupied by coastal individuals (Table 1). Pelagic polar bears have greater energy expenditure (detailed in Blanchet et al., submitted for publication), presumably because they spend more time in motion in order to reach their foraging habitat and because they hunt for seals over larger areas than coastal bears, which live in more confined areas, feeding opportunistically on an alternative locally distributed diet (e.g., coastal ringed seal, whale carcass, seabird colonies, algae). Consequently, pelagic polar bears have higher energy requirements and thus higher food consumption. In addition, model-averaged estimates indicated that ∑CHLs concentrations were 2 times higher in bears with the highest FMR compared

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to those with the lowest FMR (Figure 3). Similar but less pronounced and nonsignificant tendencies were found for ∑PCBs, HCB, and ∑PFCAs (Figure 3). This supports the previous assumption made by Olsen et al.,[42] suggesting that polar bears with larger HR have greater energetic costs, greater food intake, and consequently higher pollutant assimilation.

Ice Edge Effect. Pelagic polar bears were distributed further north compared to coastal polar bears (Figure 1; Table 1). Moreover, model-averaged estimates indicated significantly increasing concentrations of ∑CHLs, ∑PCBs, mirex, *p,p′*-DDE, and ∑PBDEs with latitude centroid, being 2.5−5.2 times higher in the northernmost compared to the southernmost bears (Figure 3). Higher pollutant levels in polar bears using higher latitudes, in line with recent findings,[43,44] are likely related to the location of the sea ice edge, which is for most of the year north of Svalbard. Indeed, it has been proposed that when sea ice melts and retreats during spring and summer, pollutants deposited on snow and stocked in ice are released in large quantities into the water column and subsequently bioaccumulate within the lipid-rich and low ice-associated food web.[89,90] Once assimilated, POPs biomagnify in upper trophic consumers until reaching elevated concentrations in seals, which are then eaten by polar bears in spring and early summer.[91] Interestingly, concentrations of PCBs have been shown to be negatively related to latitude in Barents Sea polar bears monitored in the 1990s, which has also been related to the location of the sea ice edge.[42] However, the marginal sea ice zone was located much further south in the Barents Sea in 1990s than during our study period.[92−94]

Existence of a West−East Pollutant Gradient. Pelagic polar bears were distributed further east compared to coastal polar bears (Figure 1; Table 1). Model-averaged estimates indicated significant increasing concentrations of *p,p′*-DDE, ∑PFSAs, and ∑PFCAs with longitude centroid, being 6.3, 3.2, and 2.8 times higher in the easternmost compared to the westernmost bears (Figure 3). Similar trends were found for ∑CHLs and *β*-HCH (Figure 3). Accordingly, harp seals from the White Sea−Barents Sea stock had generally higher levels of pollutants than those from Greenland Sea stock (Table 2 and S4). Median concentrations of ∑CHLs, ∑PCBs, HCB, *p,p′*-DDE, and ∑PFSAs were 53%, 82%, 62%, 70%, and 88% higher in White Sea−Barents Sea harp seals than in those from the Greenland Sea (Table 2). Our results, in line with recent findings,[43−46] indicate higher contaminant levels in the eastern part of the Barents Sea compared to more western areas. This suggests the existence of a pollutant gradient with increasing trends from Svalbard archipelago to western Russia. Such geographical pattern of pollutant levels could be related to the proximity to pollutant emission sources and transport pathways. Discharges of lipophilic POPs from large rivers outflows in the western Russian Arctic have been suggested as an important source of pollutants in this area.[6,95] Emissions of volatile PFAS precursors from the Russian and Chinese industry or elsewhere[96,97] can be transported to the eastern part of the Barents Sea through atmospheric currents and subsequently deposited on sea ice.[98] Due to a dilution effect, PFASs are generally more concentrated in surface snow than in seawater.[99,100] During melting periods, a considerable amount of pollutants is released, assimilated, and biomagnified within polar food webs, ultimately terminating in polar bears.

Implications. Our results indicate that pelagic polar bears from the Barents Sea are exposed to higher levels of pollutants than their coastal counterparts because (1) they feed on higher proportion of marine and high-trophic level prey, (2) they have higher energy requirements and subsequently higher prey consumption, (3) they forage in the marginal ice zones, and (4) they feed on prey located closer to pollutant emission sources/transport pathways. In this study, we selected pelagic and coastal polar bears with similar body condition to avoid confounding effects for our analyses. Larger studies based on random sampling on bears indicated that pelagic females are fatter than coastal females[44] (e.g., Blanchet et al., submitted for publication), and only concentrations of proteinophilic PFASs were reported to be higher in pelagic females.[44] Tartu et al.[44] concluded that the lack of difference in plasma concentrations of lipophilic POPs between coastal and pelagic polar bears was likely masked by the difference in body condition. Future studies should aim to predict how rapidly declining sea ice in the Barents Sea,[27] which is likely to challenge polar bears energetically,[101] will influence contaminant fate and exposure in Barents Sea polar bears.

Table 2. Estimated Pollutant Concentrations and Body Condition Index (BCI) in Adult Harp Seals from the White Sea−Barents Sea Stock (*n* = 10) and Greenland Sea stock (*n* = 10)[a]

variables	estimated median ± SE for White Sea−Barents Sea harp seals	estimated median ± SE for Greenland Sea harp seals	*p* value
∑CHLs	**195.4 ± 25.2**	**127.4 ± 16.4**	**0.030**
∑PCBs	**362.6 ± 55.7**	**199.2 ± 30.6**	**0.013**
α-HCH	**3.4 ± 0.3**	**5.1 ± 0.5**	**0.009**
mirex	3.9 ± 2.0	2.2 ± 1.1	0.431
HCB	**59.4 ± 10.7**	**35.3 ± 6.4**	**0.055**
p,p′-DDE	**265.8 ± 40.4**	**156.5 ± 23.8**	**0.024**
∑PBDEs	3.8 ± 0.5	3.6 ± 0.5	0.763
∑PFSAs	**39.7 ± 6.5**	**21.1 ± 3.4**	**0.013**
∑PFCAs	20.6 ± 2.9	18.0 ± 2.5	0.504
BCI	0.6 ± 0.1	0.7 ± 0.1	0.210

[a]White Sea−Barents Sea and Greenland Sea harp seals were compared using linear models. Values are expressed in ng·g^{-1} lw for OCs and PBDEs and in ng·g^{-1} ww for PFASs. Pollutants and BCI were ln transformed to meet model assumptions. Significant differences are shown in bold. OCs and PBDEs have been measured in adipose tissue and PFASs in plasma/serum.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.9b04626.

Ethic statement; fieldwork; stable isotope analysis; references; overview of GPS data collection and biological features associated with each adult female polar bear considered in this study; correlation matrix showing relationships between predictors considered in model averaging; list of the 44 candidate models used for model selection; measured pollutant concentrations in pelagic and coastal adult female polar bears from the Barents Sea and in adult harp seals from the White Sea-Barents Sea and Greenland Sea stocks; isotopic niche width illustrated by standard ellipses for both pelagic and coastal Barents Sea polar bears (*n* = 40 adult females), and comparison of the standard ellipse area according to the ecotype strategy; relationhips between trophic level and PC1 score for both pelagic and coastal Barents Sea polar bears (*n* = 40 adult females); principal component

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analysis of δ^{15}N-trophic AA values for both pelagic and coastal Barents Sea polar bears (PDF)

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