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Choose Your Poison—Space-Use Strategy Influences Pollutant Exposure in Barents Sea Polar Bears

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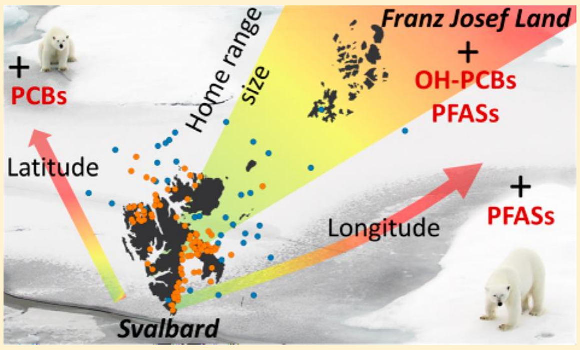
Supporting Information



ABSTRACT: Variation in space-use is common within mammal populations. In polar bears, *Ursus maritimus*, some individuals follow the sea ice (offshore bears) whereas others remain nearshore yearlong (coastal bears). We studied pollutant exposure in relation to space-use patterns (offshore vs coastal) in adult female polar bears from the Barents Sea equipped with satellite collars (2000−2014, $n = 152$). First, we examined the differences in home range (HR) size and position, body condition, and diet proxies (nitrogen and carbon stable isotopes, $n = 116$) between offshore and coastal space-use. Second, we investigated how HR, space-use, body condition, and diet were related to plasma concentrations of polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) ($n = 113$), perfluoroalkyl substances (PFASs; $n = 92$), and hydroxylated-PCBs ($n = 109$). Offshore females were in better condition and had a more specialized diet than did coastal females. PCBs, OCPs, and hydroxylated-PCB concentrations were not related to space-use strategy, yet PCB concentrations increased with increasing latitude, and hydroxylated-PCB concentrations were positively related to HR size. PFAS concentrations were 30−35% higher in offshore bears compared to coastal bears and also increased eastward. On the basis of the results we conclude that space-use of Barents Sea female polar bears influences their pollutant exposure, in particular plasma concentrations of PFAS.

INTRODUCTION

Anthropogenic activities have affected wildlife health and habitat at numerous levels. Industrialization has accelerated global warming (http://www.ipcc.ch) and is responsible for the release of toxic compounds into the environment that have become imbedded in food webs from tropical to polar ecosystems.[1] For higher trophic species, the main source of exposure occurs via diet and levels of persistent organic pollutants (POPs) are biomagnified in marine food webs.[2−5] Polar bears (*Ursus maritimus*) are among the most polluted animals,[6,7] and there are concerns about the negative impact of climate change on their population dynamics due to the recent decreases in Arctic sea ice coverage,[8−10] which constitute their main habitat for feeding, travel, and mating.[11] Habitat fragmentation and extended ice-free seasons associated with climate change may decrease prey encounter rates and increase energy expenditure during hunting and travel.[12] Polar bears preferentially feed on ringed seals (*Pusa hispida*), bearded seals (*Erignathus barbatus*), and harp seals (*Pagophilus groenlandicus*) but they are also opportunistic feeders who prey upon other various mammals and birds including terrestrial species such as reindeer (*Rangifer tarandus platyrhynchus*) and ground-nesting waterfowl.[13−21]

The distributions, geographic ranges, and therefore diets of species are largely influenced by climate, and the spatial and temporal patterning of the resources of the habitat.[22−24] Animals often display circannual seasonal movements, particularly in changing environments and in numerous instances, feeding

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strategies appear to be plastic.[25] For instance, when experiencing resource competition or abrupt environmental change, animals often transition to a more varied diet and use both optimal and alternative food sources,[25−27] which has been observed within populations in several mammals.[28−30] Individual specialization in diet, and in selection of habitat, can be beneficial if it confers higher or similar fitness in comparison to previous behavior[31−33] but can also influence the species negatively by reducing its energy intake, and increasing exposure to pathogens and anthropogenic pollutants.[28−30]

Polar bears display divergent space-use patterns within some of the 19 subpopulations found in the Arctic. In the Barents Sea area, home range size of offshore female polar bears, which migrate seasonally to follow the sea-ice retreat and advance, may be 100 times larger compared to that of coastal females that mostly remain on land or nearshore.[34,35] The offshore ecotype is used as the equivalent to what Mauritzen et al.[35] termed as "pelagic" polar bears. Repeatability of movement patterns over years indicate that an individual's specialization is a recurrent behavior.[34−36] Changes in the proportions of coastal versus offshore polar bears have been related to recent climate changes. For instance, in the Southern Beaufort and Chukchi sea subpopulations, the proportion of polar bears using the coastal strategy has increased from 10% to 35% and from 20% to 38%, respectively, between pre-2000 and post-2000 periods.[37,38] In the Southern Beaufort Sea subpopulation, the diet of coastal bears changed toward consumption of a larger proportion of bowhead whale (*Balaena mysticetus*) carcasses, while the diet of the offshore bears was consistently seal-dominated during the same period.[17] It is, however, unclear if the observed changes were due to behavioral plasticity (individuals adjusting their behavior in response to climate change) or to selection (higher reproductive success of one ecotype). In contrast, within the Barents Sea area, the number of coastal bears in Svalbard was similar in the autumns of 2004 and 2015, with an estimated number of ~250 bears in both years.[39,40]

Pollutant levels in polar bears within European and Russian Arctic vary spatially. Studies conducted in 1987−1998 revealed that female polar bears from Franz Josef Land (belonging to the Barents Sea subpopulation) and the Kara Sea subpopulation (Figure S1 of the Supporting Information, SI) were among the most polluted with respect to polychlorinated biphenyls (PCBs), oxychlordane, *trans*-nonachlor, and dichlorodiphenylchloroethylene (DDE) compared to polar bears from other areas including Svalbard, East-Siberian Sea, and Chukchi Sea.[41,42] Furthermore, Olsen et al.[43] reported that PCB concentrations were highest in polar bears from the Barents Sea subpopulation exploiting eastern habitats and having larger annual home range size, while PCB concentrations were lowest in polar bears using northern habitats. The authors proposed that polar bears with large home range sizes in the eastern Barents Sea consumed more prey and consequently ingested more pollutants compared to bears with smaller home range sizes.[43] In contrast, in the 2000s, PCBs were neither related to home range size, longitude, nor latitude.[44] Van Beest et al.[44] also reported higher per- and polyfluoroalkyl substances (PFAS) concentrations in female polar bears from the Barents Sea using eastern habitats, but hydroxylated PCBs (OH-PCBs) and polybrominated diphenyl ethers (PBDEs) were higher in females using northern habitats. The discrepancies between these two studies[43,44] could be related to ongoing changes in sea ice conditions. Confounding factors not considered in these studies could also explain pollutant variation. For example, body condition index (BCI),[45] which represents the nutritional state of an individual, is a stronger predictor than diet for the concentrations of lipophilic pollutants such as organochlorine pesticides (OCPs), PCBs, and PBDEs in polar bears.[46] In contrast, feeding habits (inferred from stable isotope ratios) were strong predictors of PFAS concentrations in polar bears.[47]

The aim of the present study was to investigate if space-use strategy influences pollutant concentrations in polar bears in the Barents Sea. Our first hypothesis was that offshore bears with larger home ranges, located further east, ingest a larger proportion of higher trophic level and/or marine prey (inferred from nitrogen [δ^{15}N] and carbon [δ^{13}C] stable isotope values) compared to coastal bears which may ingest a larger proportion of lower trophic level and/or terrestrial food. In addition, the habitat advantages conferred to offshore bears could be offset by ongoing climate change, they would therefore expend more energy to encounter their prey and have lower body condition, as compared to coastal bears. Yet, if climate change does not modify prey encounter probability, then we predict that offshore bears would be in better condition than coastal bears. Our second hypothesis was that offshore bears, compared to coastal bears, would have (1) higher concentrations of lipophilic pollutants and their metabolites (PCBs, OCPs, PBDEs, OH-PCBs) as a consequence of larger home ranges which have a higher energetic demand, resulting in lower body condition, and (2) higher PFAS concentrations, as higher energetic demands involves greater intake and potentially greater exposure to pollutants as a consequence of a more marine diet.

■ METHODS

Field Sampling. One hundred fifty-two adult female polar bears (estimated age 4−28 years) from the Barents Sea subpopulation were captured throughout Svalbard between March 26th and April 27th in 2000 and from 2002 to 2014 (Figure S2, Table S1). Immobilization, blood collection and conservation, age determination, and female classification according to reproductive status are detailed in the SI. BCI (n = 150) was calculated as described for polar bears,[45] for females not weighed in the field and for which body measurements were available (n = 38), body mass was estimated[48] before BCI calculation. The females, all with body weights >100 kg, were collared with satellite transmitters (Table S1).

Space-Use Strategy. We obtained 152 polar bear tracks of varying duration (1 month to 1 year) in 2000−2014 (excluding 2001 as no satellite collars were deployed that year). The 152 samples represented 112 individual females, among which 17 were captured in two different years, eight were captured during three different years and two during four different years. Due to different sampling regimes, we resampled all tracks to a 24 h resolution to achieve a common temporal scale across all years. For statistical analyses, we either used the entire data set or we used subsets with females that were tracked for ≥30% or ≥90% of the year when annual home range size and position were included in the analyses (detailed in Statistics, for sample sizes see Table S1). Seasonal split is detailed in the SI (Methods-*Space-Use Strategy*, Figure S3).

Annual home range size was calculated using 50%, 75%, and 95% minimum convex polygons (MCP), which represent the smallest convex polygon enclosing all daily locations of an individual. The 50% MCPs were used to attribute an offshore or coastal space-use strategy for each seasonal or annual track, based on the geographic overlap between the MCP of each individual and the Svalbard polygon. This polygon includes the four biggest islands in the Svalbard archipelago (Spitsbergen, Nordaustlandet, Edgeøya, Barentsøya) and a 20 km buffer around each island. A bear was assumed to be coastal if >50% of its home range was within the Svalbard polygon, and offshore if this condition was not met. Attribution to offshore or coastal strategy was thereafter checked using individual annual track maps. In this study, annual home ranges and geographical locations were not significantly related to reproductive status and the age distribution was not related to space-use strategy ($p > 0.35$ for all tests).

Analyses of Pollutants. Plasma samples were analyzed for PCBs, OCPs, PBDEs ($n = 113$), OH-PCBs ($n = 109$), and PFASs ($n = 92$). Methods for lipophilic pollutants, OH-PCBs, and PFAS determination in plasma and quality assurance have been detailed elsewhere.[46,49−53]

Only pollutants that were analyzed and detected in ≥60% of the individuals were considered for statistical analyses. This included three OCPs: hexachlorobenzene (HCB), oxychlordane, *p,p′*-dichlorodiphenyldichloroethylene (*p,p′*-DDE); four PCB congeners: PCBs-118, -138, -153, -180; six phenolic compounds: 4 OH−CB107, 3′OH−CB138, 4 OH−CB146, 4′OH−CB159, 3′OH−CB180, 4 OH−CB187; one PBDE: BDE-47; two perfluoroalkyl sulfonates (PFSAs: perfluorohexanesulfonate PFHxS and perfluorooctanesulfonate PFOS); and four perfluoroalkyl carboxylates (PFCAs: perfluorooctanoate PFOA, perfluorononanoate PFNA, perfluorodecanoate PFDA, perfluoroundecanoate PFUnDA). Concentrations for these compound groups are given in Table S2, and QA/QC are detailed in Table S3. For statistical analyses, we used concentrations in lipid weight (ng/g lw) for lipophilic pollutants, whereas proteinophilic pollutants (PFASs, OH-PCBs) concentrations are given in wet weight (ng/g ww).

Nitrogen and Carbon Stable Isotopes in Red Blood Cells. Nitrogen and carbon stable isotope ratios (δ^{15}N and δ^{13}C) were determined in red blood cells ($n = 116$) as described elsewhere.[17] The combustion analyses were conducted at the Environment and Natural Resources Institute—Stable Isotope Laboratory at the University of Alaska, Anchorage (http://www.uaa.alaska.edu/enri/labs/sils). QA/QC for the data used in this study is reported elsewhere.[53] Because δ^{15}N values increase with increasing trophic level, they reflect trophic position of individual polar bears.[54,55] In contrast, δ^{13}C varies marginally as a function of trophic level but rather indicates the sources of primary production in the particular food web, for example marine vs terrestrial, pelagic vs benthic, and inshore vs offshore.[54,55] Thus, polar bears with high δ^{15}N values have been feeding at a higher trophic level than bears with low δ^{15}N values. In addition, low δ^{13}C values indicate a larger proportion of terrestrial prey in polar bears diet in comparison with bears with high δ^{13}C values. In polar bear red blood cells, half-life for δ^{13}C is ∼1.5 months, whereas the half-life for δ^{15}N is at least twice as long.[56] Polar bear red blood cells provide a retrospective record of diet sources over several months.[17,20]

Statistics. We conducted statistical analyses using R version 3.2.5.[57] First, we examined the effect of space-use strategy (coastal or offshore) on mean annual home ranges size and position, body condition and feeding habits in female polar bears that were tracked ≥90% of the year ($n = 50$, see Table S1). Specifically, we used generalized linear mixed models (GLMM, R-package *nlme* version 3.1−121[58]) with 50%, 75%, and 95% MCPs, longitude and latitude of home range centroids, BCI, δ^{15}N, and δ^{13}C as response variables, and offshore vs coastal strategy as a predictor variable. We included sampling year and reproductive status (solitary, with COYs, with yearlings, or with older cubs) as random factors to account for temporal variation in feeding habits and fluctuations in body condition according to reproductive status.[53,59] We also added female identity as a random factor to account for repeated sampling. We used the following code in R "lme(log(*Response.variable*) ∼ 1+*Predictor.variable*, random = list(*Year* = ∼1, *Female.Identity* = ∼1, *Breeding.status* = ∼1), data = *data.set*, na.action = na.omit, method = "ML")", response variables were ln-transformed when necessary. In addition, in all individuals ($n = 152$) we tested if prey selectivity differed according to space-use strategy by performing Levene variance tests, *lawstat* R package[60] on δ^{13}C and δ^{15}N values in red blood cells and assuming a smaller variance within a group reflects a more specialized diet.

Second, we investigated how annual home range size, annual home range position, body condition, and feeding habits influenced pollutant concentrations of females that were tracked for at least 30% of the year ($n = 126$, see Tables S1 and S3). Sensitivity tests on the relationships between space-use strategy characteristics and pollutants were conducted to keep the largest sample size without modifying the results (Table S4). We performed a redundancy analysis, RDA, R-package *vegan* version 2.4−3,[61] to illustrate these relationships. RDA is a method to extract and summarize the variation in a set of constrained variables that can be explained by a set of constraining variables.[62,63] We performed the RDA on the 64 polar bears for which data on pollutants, space-use strategy, home range size, position, BCI, δ^{15}N, and δ^{13}C were available. Constraining variables included home range size (50%, 75%, and 95% MCPs), home range position (longitude and latitude of home range centroids), BCI, and stable isotope values, whereas concentrations of pollutants were constrained variables. We illustrated the effect of space-use strategy on the RDA axes 1 and 2 with an ordination plot.

We further tested and quantified the effects of space-use strategy (offshore vs coastal), home range size (95% MCP), home range position (latitude and longitude of centroids), BCI, and feeding habits (δ^{15}N and δ^{13}C) on pollutant concentrations using GLMMs on females that were tracked for ≥30% of the year ($n = 126$, see Tables S1 and S3). Continuous variables were standardized (mean = 0, SD = 1) before analysis to facilitate the comparison of effect sizes.[64] We defined sampling year, reproductive status, and female identity as random factors, to account for temporal and lactation-related variations of POP and PFAS concentrations[49,53,65,66] and variation in pollutant concentrations according to reproductive status.[46] To reduce the number of response variables, we selected pollutants with scores on RDA1 or RDA2 above |0.40| and summed the selected pollutants based on contaminant groups: ΣOH-PCBs, ΣPCBs, ΣPFSAs, and ΣPFCAs, whereas OCPs were analyzed individually. Pollutant concentrations were log transformed (ln) because of left-skewed distributions.

We used eight models with the following predictors: (1) space-use strategy, (2) 95% annual home range, (3) annual home range centroid longitude, (4) annual home range centroid latitude, (5) BCI, (6) δ^{15}N, (7) δ^{13}C, and (8) the null model. An information-theoretic approach[67] was used based on Akaike's information criterion corrected for small sample size (AICc, R package *MuMIn*[68]). We obtained the number of parameters (K), the difference in AICc values between the "best" model and the model at hand (ΔAICc) and a normalized weight of evidence in favor of the specific model, relative to the whole set of candidate models, derived by $e^{(-0.5(\Delta\mathrm{AICc}))}$ (AICc weights).

Conditional model averaging was used to make inference from all the models. This method produces averaged estimates of all predictor variables in the candidate model list, weighted using the AICc weights.[69,70] From this, we obtained conditional parameter-averaged estimates (β) and 95% confidence intervals (CIs) for all the predictors included in the models. To determine if parameters were significantly different from 0 at the 5% level, we used 95% CI of the model averaged estimates, 95% CI provide information about a range in which the true value lies with a certain degree of probability, and about the direction and strength of the demonstrated effect;[71] if it does not include the value of zero effect, it can be assumed that the result is statistically significant. Model fit was assessed by using residual diagnostic plots (Figures S4 and S5).

■ RESULTS AND DISCUSSION

Effects of Space-Use Strategy (Offshore or Coastal) on Home Range Size and Position, Body Condition, and Feeding Habits. Seventy-seven percent of the females (n = 152) were coastal. Among females for which track length covered ≥90% of the year (n = 50, 62% coastal), between 2000 and 2014, the 95% annual home range of coastal female polar bears from the Barents Sea subpopulation was 17 381 ± 4373 km^2 (mean ± standard error) ranging from 560 km^2 to 95 578 km^2, whereas offshore female polar bears had a 95% annual home range that was ~8-times larger (140 285 ± 32 404 km^2) ranging from 4930 km^2 to 514 377 km^2 (Figure 1A, Table S5).

Annual home range sizes of coastal and offshore females were comparable to those reported in this area between 1988 and 1998 (185–373 539 km^2).[35] Home range sizes of the present offshore females were comparable to the annual home range of polar bears from Hudson Bay (~260 000 km^2 in the 1990s, and ~350 000 km^2 in the 2000s),[72] Southern and Northern Beaufort sea (149 465 km^2 and 76 696 km^2, respectively)[73] and from the Canadian Archipelago (~125 100 km^2).[74] The mean annual home range position for coastal females was expectedly located on Svalbard Archipelago 78°43′N, 19°51′E, whereas it was located further north and east for offshore females (79°07′N, 26°84′E, Table S5). Long-term monitoring of mean annual home range position for each strategy could inform on whether space-use shifts can be measured over time.

BCI was measured in 150 females (Table S5), among which 71% were coastal. Offshore females had higher BCI than coastal females (Figure 1A), which suggests that although offshore females hunt over a larger area to find their key prey, the net energy intake of offshore bears is larger than that of coastal females. This is likely because offshore bears spend a larger proportion of the year in a hunting area with higher access to prey than coastal bears.[36] In addition, since 2010, habitat quality has been described as more optimal in the offshore area east of Svalbard than in habitats surrounding the coastline of Svalbard based on a resource selection function computing the number of days with optimal polar bear habitat.[75] This result suggests that climate change has not yet offset the advantages conferred to offshore polar bears. However, the diet of offshore females inferred from the $\delta^{15}N$ and $\delta^{13}C$ values did not differ from coastal females (n = 116, among which 74% were coastal, Figure 1A, Table S5). Nevertheless, variance tests on stable isotope values indicated that offshore females were more selective in terms of diet choices: $\delta^{15}N$ values had a narrower range in offshore than in coastal females (Levene statistic tests = 5.34, p = 0.023, Figure 1B) and a similar trend was indicated by the $\delta^{13}C$ values (Levene statistic tests = 3.75, p = 0.055, Figure 1B). Whereas coastal bears use lower trophic level and less marine

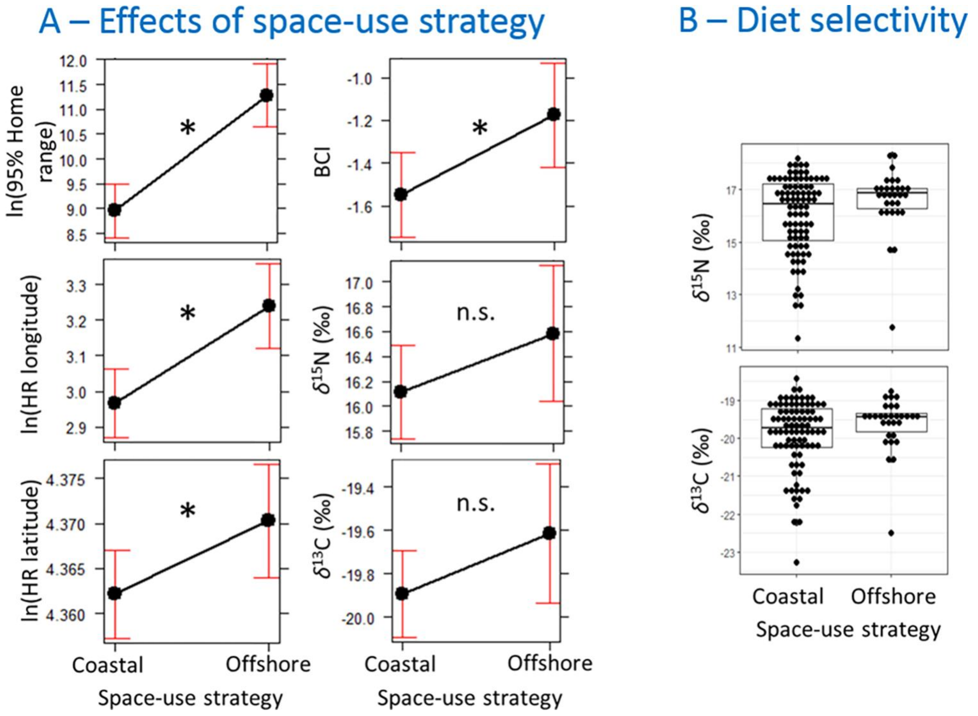

Figure 1. (A) Effect of space-use strategy on annual home range (HR) size and position (longitude, latitude), body condition (BCI), and feeding habits ($\delta^{15}N$ and $\delta^{13}C$). The values represent estimates and 95% confidence intervals derived from GLMM with sampling year, reproductive status and female identity as random factors. Asterisks denote significant differences between coastal and offshore females, whereas nonsignificant effects are noted as "n.s.". (B) Diet selectivity inferred from stable isotope values in red blood cells according to space-use strategy. Female polar bears were captured between 2000 and 2014 in the Barents Sea subpopulation.

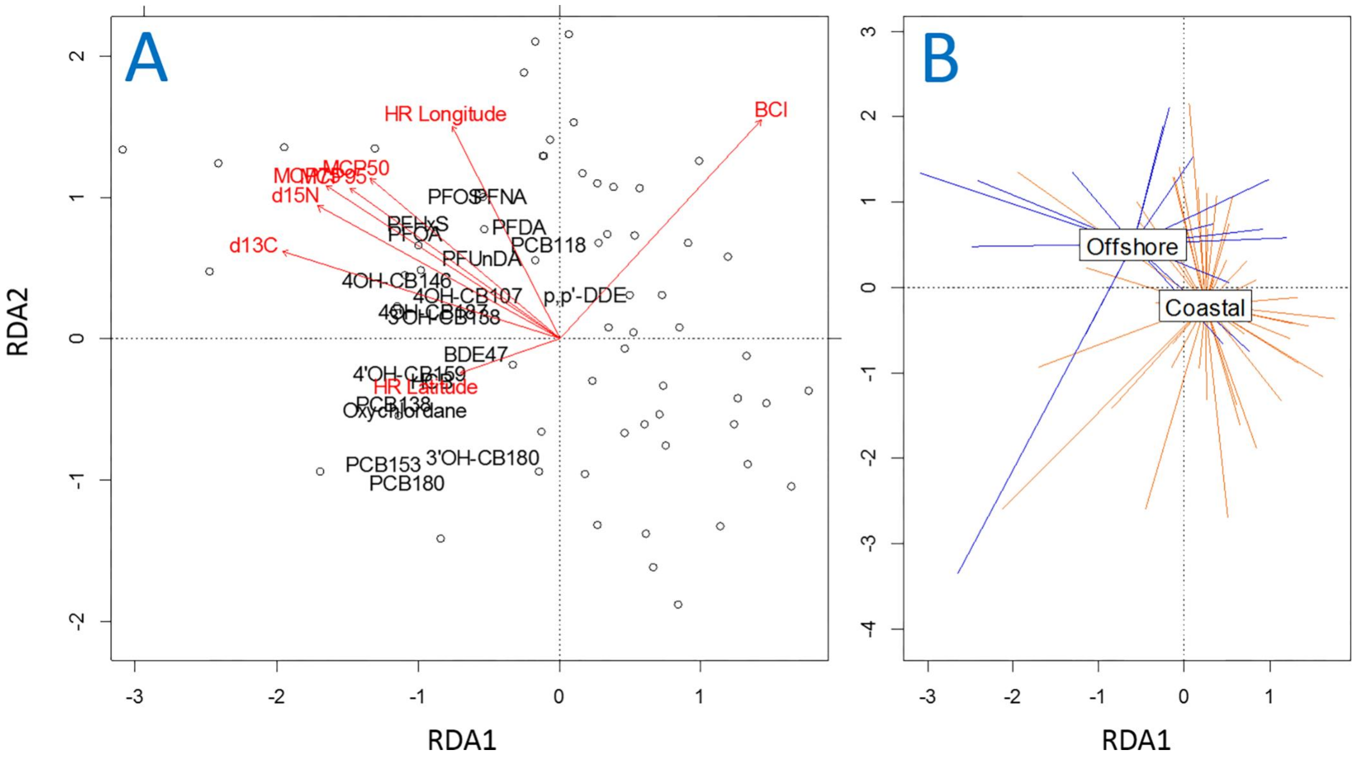

Figure 2. Relationships between feeding habits, body condition, home range size and position, and pollutants in female polar bears ($n = 80$) from the Barents Sea captured between 2000 and 2014. In the RDA scatter plot (A) constraining variables are represented in red (mean annual home range centroid latitude: HR Latitude; mean annual home range centroid longitude: HR Longitude; $\delta^{15}N$: d15N; $\delta^{13}C$: d13C; 50%, 75%, and 95% mean annual home ranges: MCP50, MCP75, and MCP95; body condition index: BCI), constrained variables (pollutants) in black and dots represent individuals. The ordination plot (B) separates individual RDA scores according to space-use strategy (offshore females in blue and coastal females in orange). The first two RDA axes accounted for 79.1% of the total variance (RDA1: 59.6%, RDA2: 19.5%). The contribution of each variable to RDA 1 and RDA 2 is given in in SI Table S6.

prey to their diet to meet energetic needs, offshore bears have access to seals through most of the year.

Effects of Space-Use Strategy on Pollutant Exposure. According to the RDA, variables related to space-use strongly explained (scores ≥ |0.40|, Table S6) concentrations of the following pollutants: HCB, oxychlordane, PCB-138, -153, -180, 4 OH−CB107, 3′OH−CB138, 4 OH−CB146, 4′OH−CB159, 3′OH−CB180, 4 OH−CB187, PFHxS, PFOS, PFOA, and PFNA. Specifically, as indicated in the RDA plot, PFOS, PFHxS, PFOA, PFNA, 4 OH−CB107, 3′OH−CB138, 4 OH−CB146, and 4 OH−CB187 were positively related to home ranges, the longitude of the home range centroid, $\delta^{13}C$ and $\delta^{15}N$ (Figure 2A). In contrast, HCB, oxychlordane, PCB-138, -153, -180, 4′OH−CB159, and 3′OH−CB180 were negatively related to BCI (Figure 2A). Pollutant signature differed between offshore and coastal bears according to the RDA (Figure 2B). The difference between the coastal and the offshore clusters seem to be driven by higher PFAS concentrations in offshore females. In further analyses, we summed pollutants that were the most related to space-use, feeding habits, and body condition (RDA score ≥ |0.40|). This resulted in Σ_3PCBs: PCBs-138, -153, -180; Σ_2PFSAs: PFHxS, PFOS; Σ_2PFCAs: PFOA, PFNA, Σ_6OH-PCBs: 4′OH−CB159, 3′OH−CB180, 4 OH−CB107, 3′OH−CB138, 4 OH−CB146, and 4 OH−CB187. Because 50%, 75%, and 95% home ranges were strongly correlated (Figure 2A), we used the largest home range (95%) in GLMMs.

Mixed models supported the relationships visually assessed from the RDA plots (Figure 2A,B, Tables 1 and S7). Specifically, when adjusted for sampling year, reproductive status, and female identity, we were able to identify two patterns according to the pollutant classes.

Lipophilic Pollutants and OH-PCB Concentrations According to Space-Use Strategy. According to model averaged estimates from GLMMs, concentrations of lipophilic pollutants were best explained by BCI, with higher pollutant concentrations in thinner bears (Tables 1 and S7). This is in accordance with Tartu et al.[46] showing that body condition is more important than diet (i.e., $\delta^{13}C$ and $\delta^{15}N$ values) to predict concentrations of lipophilic pollutants in female polar bears from the Barents Sea. Concentrations of lipophilic pollutants were not related to space-use strategy or longitude (Table 1), which contrasts with our hypothesis as well as previous findings on polar bears captured in the Barents Sea during the 1990s.[43] The lack of differences in concentrations of lipophilic pollutants between offshore and coastal females in our study is likely related to body condition (Figure 1, Table S5). In comparison to coastal females, offshore females likely have greater access to more contaminated prey for longer each year. Therefore, contaminant intake of offshore females should be higher, yet this effect could be masked by better body condition which may dilute lipophilic pollutants in the tissues. Olsen et al.[43] did not detect differences in body condition according to habitat use and home range size based on a subjective scale (ranging from 1 to 5), whereas BCI used in our study[45] provided a more precise body fat metric.

Model averaged estimates indicated that Σ_3PCB concentrations were higher in female polar bears foraging further north regardless space-use strategy (Table 1, Figure 3). In contrast,

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Table 1. Effects of Feeding Habits ($\delta^{15}N$ and $\delta^{13}C$), Annual Latitudinal and Longitudinal Home Range Position, Body Condition (BCI), Annual 95% Home Range Size, and Space-Use Strategy, on Pollutant Concentrations in Plasma of Female Polar Bears from the Barents Sea (2000-2014)[a]

predictors	HCB (n = 92)	oxychlordane (n = 92)	Σ_3PCB (n = 92)	Σ_6OH-PCB (n = 89)	Σ_2PFSA (n = 72)	Σ_2PFCA (n = 72)
intercept	**3.95 [2.47; 5.43]**	**5.54 [3.85; 7.22]**	**6.82 [6.47; 7.17]**	**10.7 [6.19; 15.21]**	**5.05 [4.74; 5.37]**	**2.66 [2.19; 3.13]**
$\delta^{15}N$	0.005 [−0.089; 0.099]	0.07 [−0.05; 0.20]	0.01 [−0.08; 0.11]	**0.18 [0.09; 0.27]**	**0.08 [0.001; 0.155]**	**0.06 [0.002; 0.116]**
$\delta^{13}C$	0.04 [−0.11; 0.18]	0.03 [−0.18; 0.23]	0.07 [−0.09; 0.22]	**0.33 [0.20; 0.47]**	0.09 [−0.04; 0.21]	**0.10 [0.01; 0.19]**
home range centroid latitude	−0.02 [−0.14; 0.09]	0.02 [−0.14; 0.17]	**0.14 [0.02; 0.26]**	0.05 [−0.07; 0.16]	−0.01 [−0.09; 0.07]	0.02 [−0.04; 0.08]
home range centroid longitude	−0.01 [−0.03; 0.01]	−0.01 [−0.04; 0.01]	−0.01 [−0.03; 0.01]	0.01 [−0.01; 0.03]	**0.025 [0.014; 0.035]**	**0.015 [0.006; 0.024]**
BCI	**−0.30 [−0.51; -0.09]**	**−0.42 [−0.71; −0.14]**	**−0.58 [−0.78; −0.39]**	−0.02 [−0.24; 0.19]	0.05 [−0.10; 0.20]	0.05 [−0.07; 0.17]
95% home range (km^2)	1.62×10^{-06} [-9.92×10^{-08}; 3.35×10^{-06}]	2.28×10^{-07} [-2.19×10^{-06}; 2.65×10^{-06}]	3.32×10^{-07} [-1.53×10^{-06}; 2.19×10^{-06}]	**1.97×10^{-06} [3.07×10^{-07}; 3.64×10^{-06}]**	**1.90×10^{-06} [8.88×10^{-07}; 2.92×10^{-06}]**	**1.46×10^{-06} [6.33×10^{-07}; 2.28×10^{-06}]**
space-use strategy (ref: coastal)	0.14 [−0.16; 0.45]	−0.08 [−0.51; 0.34]	0.05 [−0.28; 0.38]	0.30 [−0.01; 0.60]	**0.26 [0.06; 0.47]**	**0.30 [0.14; 0.46]**

[a] The sample size used for each list of models is represented by "n". Values are parameter estimates and 95% confidence intervals derived from conditional model averaging of general linear mixed models that included female identity, sampling year (14 years), and reproductive status (solitary, with cubs of the year, with yearlings, with older cubs) as random factors. Pollutant concentrations were ln transformed. Values in bold are significantly different from 0 at the 5% level.

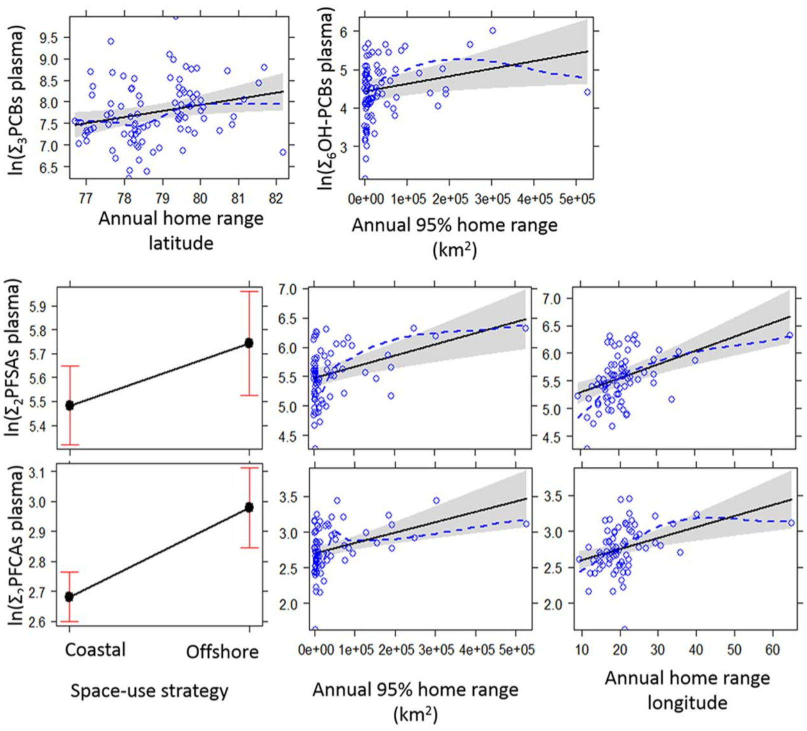
Σ_5PCBs (CB99, -153, -156, -180, and -194) was negatively related to latitudinal position in Barents Sea polar bears sampled in the 1990s.[43] The authors suggested that PCB concentrations were likely higher in polar bears feeding at the sea ice edge during spring and summer when sea ice is melting and pollutants are taken-up by the food web. The same hypothesis could also explain our results, as the spring/summer sea ice edge in the Barents Sea is moving northward.[76,77] It is noteworthy that the effect of latitude on Σ_3PCB concentrations disappears when reducing the sample size to bears for which tracks covered ≥90% of the year (Table S4). This may occur because fewer coastal females were included in this subset and the latitudinal gradient in PCB could be more pronounced around Svalbard. We are therefore cautious in interpreting this result.

The best predictor of Σ_6OH-PCBs was $\delta^{13}C$ values (Table S7). Model averaged estimates indicated that Σ_6OH-PCB increased with 95% annual home range size and with increasing $\delta^{13}C$ and $\delta^{15}N$ values indicating that bears with an intake of marine prey high in the food web had higher levels of PCB metabolites (Table 1). Furthermore, Σ_6OH-PCBs tended to be higher in offshore than coastal bears (0.30 [−0.01; 0.60]; Table 1). In polar bears, OH-PCBs mainly originate from biotransformation, as concentrations of these compounds in seal blubber are negligible.[78] According to the RDA plot (Figure 2A), 4 OH−CB107, 3′OH−CB138, 4 OH−CB146, and 4 OH−CB187 were the phenolic compounds that were best explained by polar bears' feeding habits. Parent compounds to these OH-PCBs such as PCB-105, -118, -138, -153, -187, and -183[49] are highly bioaccumulative.[79] We may therefore assume that high Σ_6OH-PCBs result from biotransformation of their parent compounds, which increase with marine prey that are at a higher trophic level. As indicated by the positive relationship between Σ_6OH-PCBs and the 95% annual home range size (Figure 3), gradually off the coasts of Svalbard, these parent compounds were likely more available, or their intake was higher due to larger net energy intake.

PFAS Concentrations According to Space-Use Strategy. Median PFSA and PFCA concentrations were 30% [6; 60] and 35% [14; 46] (values are exponential transformed estimates and 95% CI) higher in offshore than in coastal female bears. Moreover, PFAS concentrations increased from west to east (i.e., toward Russian territories) (Table 1, Figure 3). Plasma PFAS concentrations in polar bears were affected by diet.[47] We therefore hypothesized that offshore bears had higher concentrations of PFASs as a consequence of a higher proportion of marine items in their diet. Although in our study, $\delta^{13}C$ and $\delta^{15}N$ values did not significantly differ between offshore and coastal females (Table S5), variance analyses indicated a larger proportion of lower trophic level and terrestrial prey in coastal bears diet (Figure 1B). Considering the biomagnifying properties of PFASs in marine food web[2,80] the more varied diet of coastal females could contribute to their lower PFAS concentrations.

Abiotic conditions such as sea ice extent, concentration, and melting can influence the amount of PFAS released into the ocean, and thus affect the PFAS concentrations in offshore vs coastal bears. PFASs are more concentrated in surface snow than in seawater, due to a dilution effect.[81,82] When sea ice melts, large amounts of PFASs can be released in the ocean, accumulated in the phytoplankton which is concomitantly blooming, and thus biomagnified.[2,83,84] Consequently, in areas with more sea ice, such as those used by offshore bears, environmental PFAS levels were likely higher than in areas with less sea ice such as the coast of Svalbard.

A – Effects of space-use on pollutant concentrations



B – Schematic gradient of pollutant exposure according to space-use

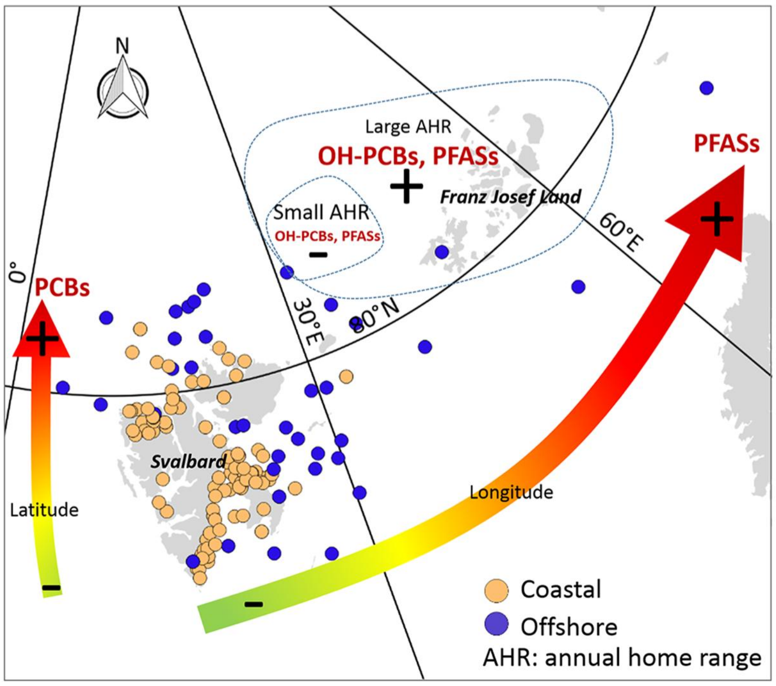


Figure 3. A – Significant relationships between pollutant concentrations in plasma, body condition (BCI), and space-use strategy components. Dots are partial residuals derived from mixed models with year, reproductive status, and female identity as random factors, blue dots are the partial residuals and dashed line a loess smooth of the partial residuals. The black solid line is the parameter estimate and the gray area represents its 95% confidence interval. Removal of the extreme value did not change the results. B – Schematic view of how space-use strategy can explain pollutant concentrations, the red end of the arrows represents high pollutant concentrations, blue dotted lines represent hypothetical annual home range extent with PFAS concentrations being lower in bears using small home ranges than those using large ones. Yellow and blue dots represent home range centroid positions in spring for coastal and offshore females, respectively.

The positive relationship between PFAS concentrations and home range longitude position in polar bears accords with a study that showed that PFOA, PFNA, and PFHxS concentrations in ivory gull (*Pagophila eburnea*) eggs from more eastern

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colonies at Franz Josef Land were slightly higher than concentrations in eggs from Svalbard.[85,86] The geographical differences could be related to locality of emission sources. Releases of PFCAs from fluoropolymer production sites in China, Russia, Poland, and India have been estimated to be the major contributors to global PFCA emissions in 2003−2015.[87] For example, two Russian factories situated ∼1000 km from the Arctic coast produced seven thousand tons of fluoropolymers in 2010 (http://www.halopolymer.com/about) and PFSA emissions from China have increased since 2003.[88] Emissions of volatile PFSA and PFCA precursors from Russia or China can be transported to the Arctic through air currents as shown for aerosols and black carbon.[89] The long-range transport of aerosols such as mineral dust and coal fly ash is a potential PFCA source to the Arctic.[90]

Implications. Offshore females were in better condition than coastal females, so we could assume that an offshore space-use strategy would be more advantageous in terms of fitness and that climate change to 2014 has not affected the condition of offshore bears. Yet, one has to remain cautious on this conclusion due to the difference between offshore and coastal bears with regard to time of sampling versus start-time for feeding. It is possible that the offshore bears were in better condition in spring because they built up more fat the year before since they spend a larger proportion of the year in a feeding habitat. Although offshore females were in better condition than coastal females, they were exposed to higher concentrations of PFASs. Information on the effects of PFAS in polar bears is scarce, however modeling and correlative field studies suggest that PFASs interact with polar bear physiology and metabolism at various levels.[91−93] Further studies examining the transport of legacy and emerging pollutants in the Arctic, as well as more precise measures for diet and metabolism of lipophilic POPs, would help clarify the absence of difference in lipophilic pollutant concentrations between coastal and offshore bears.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b06137.

> Biological information on the study animals, detailed method descriptions, overview of the available data, pollutant concentrations, quality assurance for pollutant analyses, statistical analyses testing the effects of space-use strategy, RDA scores, model selection tables, polar bear subpopulations distribution, sampling locations map, seasonal movements map, and diagnostic residual plots (PDF)

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Notes

The authors declare no competing financial interest.

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