

Choose your poison – Space-use strategy influences pollutant exposure in Barents Sea polar bears

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METHODS

Field sampling

Females were immobilized by remote injection of tiletamine hydrochloride and zolazepam hydrochloride (Zoletil Forte Vet®; Virbac, France), delivered by a dart fired from a helicopter (Eurocopter AS350 Ecureuil). Immobilization and handling procedures followed standard protocols^{1,2}, and were approved by the National Animal Research Authority (Norwegian Animal Health Authority, P.O. Box 8147 Dep., N-0033 Oslo, Norway). We collected 50-100 ml of blood from the femoral vein using vacutainers (9-10 ml) with Lithium-Heparine to avoid clotting. We kept samples cool and out of sunlight until centrifuged within 10 h (3500 rpm, 10 min). Red blood cells and plasma were transferred to separate cryotubes and kept at -20 °C until analyses.

Females were classified into one of four groups according to their reproductive status: solitary (i.e., alone or together with a male in spring), with cubs-of-the-year (COYs; cubs < 1 year old), with yearlings (offspring 1 to 2 years old) or with older offspring (2 to 3 years old). The bears were aged using a vestigial premolar tooth (P1) following standard methods³, or based on known age for bears originally captured as COYs. Only adult-sized polar bears (≥ 4 years)¹ were included in the study. Body condition index (BCI, $n=150$) was calculated as described for polar bears⁴, for females not weighed in the field and for which body measurements were available ($n=38$), body mass was estimated¹ before BCI calculation.

Space-use strategy

The seasons in the annual cycle of polar bears are the six month long winter season (December to May), followed by the shorter spring (June-July), summer (August-September) and autumn (October- November) seasons⁵. Out of 27 recaptures, we only observed three cases of strategy shifts during winter and one during summer. We also observed one case of a female using an

offshore strategy in summer, whereas she was coastal during the rest of the year. Based on these few observations of space-use strategy shifts recorded over the 13-year time period, we can confidently assume that space-use strategies are stable over time as reported in previous studies^{6–9}.

Analyses of pollutants

Pollutant concentrations in plasma were determined from blood samples taken between March 26th and April 27th in 2000 and from 2002 to 2014. Pollutants were analysed at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences (NMBU), Oslo, Norway. Pollutants included in this study were obtained from different projects, they were therefore analysed in different batches (**Table S2**) and for some projects, the pollutants analysed varied according to study objectives. Concentrations of chlorinated and brominated compounds were blank corrected. Values below limits of detection (LOD) were assigned 0.5×LOD. LOD and recovery rates for PCBs, OCPs and BDE-47 are given in **Table S2**. Detailed QA/QC for PFAS data used in this study have been published previously¹⁰.

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Table S1: Overview of the data available obtained from female polar bears captured from the Barents Sea subpopulation between 2000 and 2014. Of the 152 collars, were deployed 93 Telonics, 13 SMRU, four Sir Track Prox and 21 ATS tags. Nitrogen and carbon stable isotope ratios were measured in red blood cells (rbc), polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), hydroxylated PCBs (OH-PCBs) and per- and polyfluoroalkyl substances (PFASs) were assessed in plasma. The numbers represent the number of individuals available each year for each variable. Further analyses were conducted on subsets of this dataset, descriptive annual values were obtained from tracks that covered $\geq 90\%$ of the year and mixed model analyses testing the effects of space-use strategy on stable isotopes and pollutants were conducted on tracks that covered $\geq 30\%$ of the year. Sample size (offshore/coastal) according to the season covered: $\geq 90\%$ (n=19/n=31), $\geq 60\%$ (n=26/n=70) and $\geq 30\%$ (n=33/n=93).

	2000	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	Total
Collars deployed	6	8	10	2	8	13	9	19	14	9	12	21	11	10	152
Body condition index	6	8	9	2	8	13	9	19	13	9	12	21	11	10	150
Stable isotopes (rbc)	2	1	10	2	4	4	9	16	8	7	12	21	11	9	116
PCBs, OCPs (plasma)	2	0	10	2	3	5	9	16	7	7	12	21	11	8	113
OH-PCBs (plasma)	2	0	10	2	3	4	9	13	7	7	12	21	11	8	109
PFASs (plasma)	2	1	9	2	2	5	2	3	7	6	12	21	11	9	92
Track covers $\geq 90\%$ of the year	6	4	5	1	0	4	1	12	6	5	0	0	2	4	50
Track covers $\geq 30\%$ of the year	6	7	10	1	5	8	9	19	13	8	9	13	8	10	126
Tag type	Telonics (GPS Gen II, Konv. ST-14, Konv ST-3)	Telonics (GPS Gen III)	Telonics (GPS Gen III)	Telonics (GPS Gen III)	SMRU	SMRU, Telonics (GPS TGW-3680, Konv.ST-14)	Telonics (Konv. A-3610)	Sir Track Prox, Telonics (Konv. A-3610)	Telonics (GPS TGW-3680, Konv. A-3610)	Telonics (GPS TGW-4680H, Konv. TAW-4610H)	Telonics (GPS, GPS TGW-4680H)	ATS Iridium	Telonics iridium GPS	Telonics iridium GPS	

Table S2: Median concentrations (ng/g wet weight) and range of pollutants measured in plasma of female polar bears captured in the Barents Sea between 2000 and 2014.

	Median	Minimum	Maximum
Lipid %	1.29	0.783	1.97
HCB	0.889	0.141	5.30
Oxychlordan	6.18	<LOD	34.3
<i>p,p'</i> -DDE	0.260	<LOD	2.93
PCB118	0.285	<LOD	0.889
PCB-138	2.15	<LOD	13.2
PCB-153	15.8	3.15	104.2
PCB-180	9.33	2.32	74.6
4 OH-CB107	5.48	0.722	35.3
3'OH-CB138	0.609	<LOD	2.94
4 OH-CB146	26.4	2.61	84.7
4'OH-CB159	0.236	<LOD	3.11
3'OH-CB180	0.862	<LOD	3.065
4 OH-CB187	56.3	5.62	425.7
BDE47	0.163	<LOD	0.938
PFHxS	26.44	11.06	59.17
PFOS	229.2	53.3	602.1
PFOA	5.96	1.54	14.8
PFNA	33.6	11.7	91.6
PFDA	9.91	3.55	28.1
PFUnDA	31.2	7.41	84.4

Table S3: Limit of detection (LOD; ng/g) and recovery rate (%) of lipophilic pollutants and OH-PCBs in spiked reference material for polar bear batches analyzed in 2007, 2009, 2011, 2014 and 2015. OH-PCBs and BDE-47 (all batches) and, PCBs and OCPs (2007, 2009 and 2011) were measured by HRGC (Agilent 6890 Series with Agilent 7683 Series autosampler) connected to a quadrupole MS detector (Agilent 5973 Series). In 2014, PCBs and OCPs were measured using HRGC (Agilent 6890 Series with Agilent 7683 Series autosampler) connected two μ -ECD (Agilent 6890). In 2015 OCPs and PCBs were measured using HRGC (Agilent 6890 Series with Agilent 7683 Series autosampler) connected to a quadrupole MS detector (Agilent 5975 Series). n.a.: not analysed. *Because of large inter-batch variability of recovery rates, 4 OH-CB187 was corrected for recovery rate for statistical analyses.

	2007 (n=9)		2009 (n=16)		2011 (n=35)		2014 (n=32)		2015 (n=21)	
	LOD	Recovery	LOD	Recovery	LOD	Recovery	LOD	Recovery	LOD	Recovery
HCB	0.01	120	0.01	91	0.02	95	0.01	118	0.007	101
Oxychlordane	0.015	99	0.015	83	0.05	85	0.14	92	0.041	117
<i>p,p'</i> -DDE	0.025	107	0.025	100	0.03	97	0.07	105	0.126	123
PCB-118	0.01	104	0.04	99	0.06	102	0.015	95	0.015	97
PCB-138	0.02	90	0.03	103	0.06	111	0.02	97	0.011	119
PCB-153	0.025	97	0.025	108	0.07	82	0.015	95	0.009	118
PCB-180	0.02	91	0.02	104	0.06	108	0.015	99	0.009	95
4 OH-CB107	0.025	109	0.025	104	0.07	86	0.055	97	0.051	101
3'OH-CB138	0.06	109	0.06	103	0.06	84	0.115	92	0.123	96
4 OH-CB146	0.02	108	0.02	109	0.1	84	0.13	90	0.059	97
4'OH-CB159	0.12	88	0.025	90	0.02	96	0.053	101	0.017	100
3'OH-CB180	n.a.	n.a.	0.02	69	0.04	66	0.053	81	0.022	93
4 OH-CB187*	0.02	96	0.025	116	0.07	42	0.11	62	0.135	100
BDE-47	0.025	62	0.025	92	0.005	102	0.01	102	0.012	84

Table S4: Sensitivity tests for minimal track duration selection. We tested the effects of space-use strategy, average annual 95% home range, annual latitudinal, longitudinal centroid positions, body condition and diet proxies ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) on pollutant concentrations in plasma of female polar bears from the Barents Sea (2000-2014) by sub setting individuals followed for at least 30%, 60% or 90% of the year. Since the obtained results were mostly similar between the three subsets, we kept the largest one (individuals tracked at least 30% of the year, n=126) for statistical analyses including pollutants. Significance codes were obtained from generalized linear mixed models with female identity, sampling year (14 years) and reproductive status (solitary, with cubs of the year, with yearlings, with older cubs) used as random factors.

	HCB			Oxychlorane			$\Sigma_3\text{PCB}$			$\Sigma_6\text{OH-PCB}$			$\Sigma_2\text{PFSA}$			$\Sigma_2\text{PFCA}$		
	90 %	60 %	30 %	90 %	60 %	30 %	90 %	60 %	30 %	90 %	60 %	30 %	90 %	60 %	30 %	90 %	60 %	30 %
$\delta^{15}\text{N}$.									**	***	***			*			*
$\delta^{13}\text{C}$	*			*						**	***	***						*
Home range centroid latitude								.	*									
Home range centroid longitude													**	***	***		*	**
BCI	*	.	*	*	**	*	***	***	***									
95% Home range	*									**	.	*	**	**	***	*	**	***
Space-use strategy (ref: Coastal)										*	.	.	*	*	*	*	**	***
Sample size (offshore/coastal) according to season covered: $\geq 90\%$ (n=19/n=31), $\geq 60\%$ (n=26/n=70) and $\geq 30\%$ (n=33/n=93)																		
Significance codes: $p < 0.001$ (***), $0.001 < p < 0.01$ (**), $0.01 < p < 0.05$ (*), $0.05 < p < 0.10$ (.)																		

Table S5: Effect of space-use strategy (coastal or offshore) on space-use components (average annual home ranges, average annual longitudinal and latitudinal position), body condition (BCI) and diet proxies ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) in female polar bears from the Barents Sea (2000-2014). We obtained estimates from mixed models with female identity, sampling year (14 years) and reproductive status (solitary, with cubs of the year, with yearlings, with older cubs) as random factors. In bold are variables that significantly vary with space-use strategy.

Effect of space-use strategy on:	n (coastal/offshore)	Intercept	Estimate [95% CI]
50% Home range (ln)	50 (31/19)	6.96 [6.11; 7.82]	2.6 [1.46; 3.75]
75% Home range (ln)	50 (31/19)	7.85 [7.18; 8.53]	2.7 [1.84; 3.56]
95% Home range (ln)	50 (31/19)	8.95 [8.4; 9.5]	2.33 [1.63; 3.02]
Home range centroid longitude (ln)	50 (31/19)	2.97 [2.87; 3.06]	0.27 [0.13; 0.42]
Home range centroid latitude (ln)	50 (31/19)	4.36 [4.36; 4.37]	0.01 [0.0001; 0.0161]
BCI	150 (106/44)	-1.55 [-1.75; -1.35]	0.37 [0.15; 0.6]
$\delta^{15}\text{N}$	116 (86/30)	16.11 [15.74; 16.49]	0.47 [-0.11; 1.06]
$\delta^{13}\text{C}$	116 (86/30)	-19.89 [-20.09; -19.69]	0.28 [-0.09; 0.64]
Reference level: Coastal. Random effects: female identity, reproductive status and year			

Table S6: Redundancy analysis (RDA) scores on the 1st and 2nd RDA axes. The first two RDA axes accounted for 79.1 % of the total variance (RDA1: 59.6 %, RDA2: 19.5 %). In bold we show variables which contribute the most to RDA1 and RDA2.

	RDA1	RDA2
Constraining variables		
BCI	0.49	0.69
Longitude	-0.26	0.67
Latitude	-0.24	-0.11
MCP50	-0.45	0.51
MCP75	-0.56	0.48
MCP95	-0.50	0.48
$\delta^{15}N$	-0.58	0.42
$\delta^{13}C$	-0.67	0.27
Constrained variables		
HCB	-0.57	-0.14
Oxychlordan	-0.69	-0.25
<i>p,p'</i> -DDE	0.12	0.15
PCB118	-0.05	0.32
PCB138	-0.75	-0.22
PCB153	-0.79	-0.42
PCB180	-0.68	-0.49
4OH-CB107	-0.41	0.15
3'OH-CB138	-0.52	0.08
4OH-CB146	-0.72	0.20
4'OH-CB159	-0.67	-0.12
3'OH-CB180	-0.34	-0.40
4OH-CB187	-0.56	0.10
BDE47	-0.38	-0.05
PFHxS	-0.63	0.40
PFOS	-0.47	0.49
PFOA	-0.65	0.36
PFNA	-0.27	0.49
PFDA	-0.18	0.38
PFUnDA	-0.35	0.28

Table S7: Akaike information criterion (AIC) for the eight candidate models to explain ln-transformed pollutant concentrations in female polar bears from the Barents Sea (2000-2014). Female identity, sampling year (14 years) and reproductive status (solitary, with cubs of the year, with yearlings, with older cubs) were used as random factors for generalized linear mixed models. Predictors in bold represent top rank models ($\Delta AIC_c \leq 2$).

Response variables	Predictors	df	log Likelihood	AICc	ΔAIC_c	weight
HCB	BCI	6	-88.1	189.1	0	0.76
	95% annual home range	6	-90.2	193.3	4.17	0.09
	Null model	5	-91.9	194.5	5.4	0.05
	Space-use strategy	6	-91.5	195.9	6.8	0.03
	Home range centroid longitude	6	-91.6	196.2	7.11	0.02
	$\delta^{13}\text{C}$ red blood cells	6	-91.8	196.6	7.45	0.02
	Home range centroid latitude	6	-91.9	196.7	7.55	0.02
	$\delta^{15}\text{N}$ red blood cells	6	-91.9	196.8	7.67	0.02
Oxychlorthane	BCI	6	-117.4	247.8	0	0.86
	Null model	5	-121.6	253.9	6.15	0.04
	$\delta^{15}\text{N}$ red blood cells	6	-121.0	254.9	7.19	0.02
	Home range centroid longitude	6	-121.1	255.1	7.32	0.02
	Space-use strategy	6	-121.5	256.0	8.3	0.01
	$\delta^{13}\text{C}$ red blood cells	6	-121.6	256.1	8.37	0.01
	Home range centroid latitude	6	-121.6	256.2	8.4	0.01
	95% annual home range	6	-121.6	256.2	8.4	0.01
$\Sigma_3\text{PCBs}$	BCI	6	-80.6	174.1	0	1
	Home range centroid latitude	6	-92.7	198.5	24.3	0
	Null model	5	-95.6	201.8	27.7	0
	Home range centroid longitude	6	-94.8	202.5	28.4	0
	$\delta^{13}\text{C}$ red blood cells	6	-95.2	203.5	29.4	0
	95% annual home range	6	-95.5	204	29.9	0
	Space-use strategy	6	-95.5	204	29.9	0
	$\delta^{15}\text{N}$ red blood cells	6	-95.5	204	29.9	0
$\Sigma_6\text{OH-PCBs}$	$\delta^{13}\text{C}$ red blood cells	6	-79.2	171.3	0	0.96
	$\delta^{15}\text{N}$ red blood cells	6	-82.3	177.7	6.4	0.04
	95% annual home range	6	-87	187	15.6	0
	Space-use strategy	6	-87.8	188.7	17.4	0
	Null model	5	-89.7	190.2	18.9	0
	Home range centroid longitude	6	-89.4	191.8	20.5	0
	Home range centroid latitude	6	-89.4	191.9	20.6	0
	BCI	6	-89.7	192.4	21.1	0
$\Sigma_2\text{PFSA}$ s	Home range centroid longitude	6	-28.2	69.8	0	0.96
	95% annual home range	6	-31.6	76.5	6.7	0.03

	Space-use strategy	6	-34.9	83.1	13.4	0
	$\delta^{15}\text{N}$ red blood cells	6	-36.1	85.6	15.8	0
	Null model	5	-38.2	87.3	17.5	0
	$\delta^{13}\text{C}$ red blood cells	6	-37.2	87.7	17.9	0
	BCI	6	-38	89.3	19.5	0
	Home range centroid latitude	6	-38.1	89.5	19.7	0
$\Sigma_2\text{PFCAs}$	Space-use strategy	6	-12.6	38.5	0	0.59
	95% annual home range	6	-13.5	40.3	1.7	0.25
	Home range centroid longitude	6	-14	41.4	2.9	0.14
	$\delta^{13}\text{C}$ red blood cells	6	-17.1	47.4	8.9	0.01
	$\delta^{15}\text{N}$ red blood cells	6	-17.2	47.6	9.1	0.01
	Null model	5	-19.3	49.5	11	0
	BCI	6	-18.9	51.2	12.7	0
	Home range centroid latitude	6	-19.2	51.6	13.1	0



Figure S1: Circumpolar distribution of the polar bear. Abbreviations of delineated subpopulations include Viscount Melville Sound (VM), Norwegian Bay (NW), Kane Basin (KB), Lancaster Sound (LS), Baffin Bay (BB), Davis Strait (DS) Southern Hudson Bay (SH), Western Hudson Bay (WH), Foxe Basin (FB), Gulf of Boothia (GB), M’Clintock Channel (MC), Southern Beaufort Sea (SB), and Northern Beaufort Sea (NB). Source: IUCN/SSC Polar Bear Specialist Group (2006).

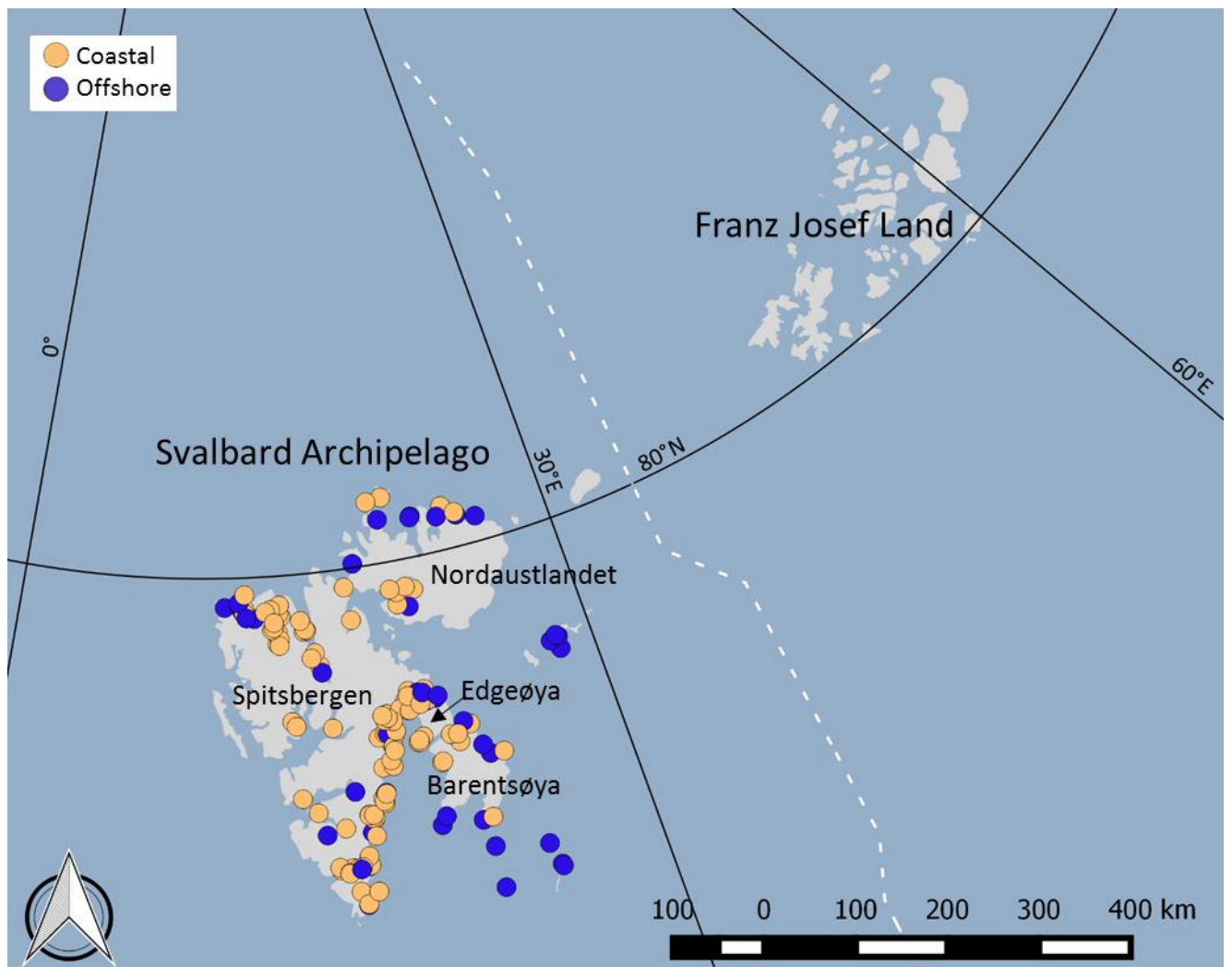


Figure S2: Capture location of female polar bears around Svalbard archipelago between March 26th and April 27th in 2000 and from 2002 to 2014. Females were categorized according to their space-use strategy, females using an offshore habitat are represented by blue dots and females using a coastal habitat are represented by yellow dots.

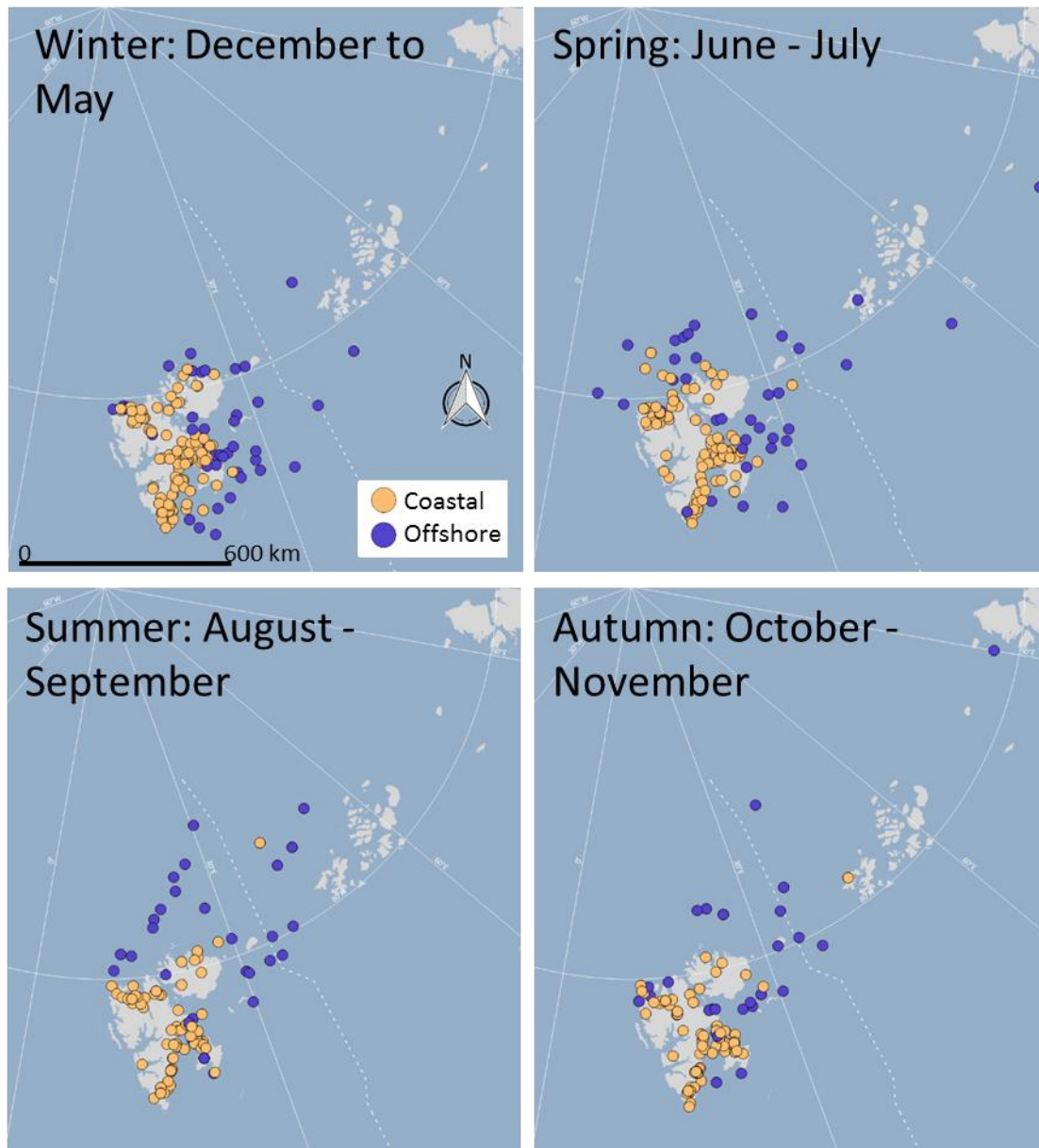


Figure S3: Seasonal habitat use of offshore (blue dots) and coastal (yellow dots) female polar bears caught in 2000-2014. Dot represent average position of the seasonal home range centroid. It has to be noticed that one coastal female adopted an offshore strategy within a given year.

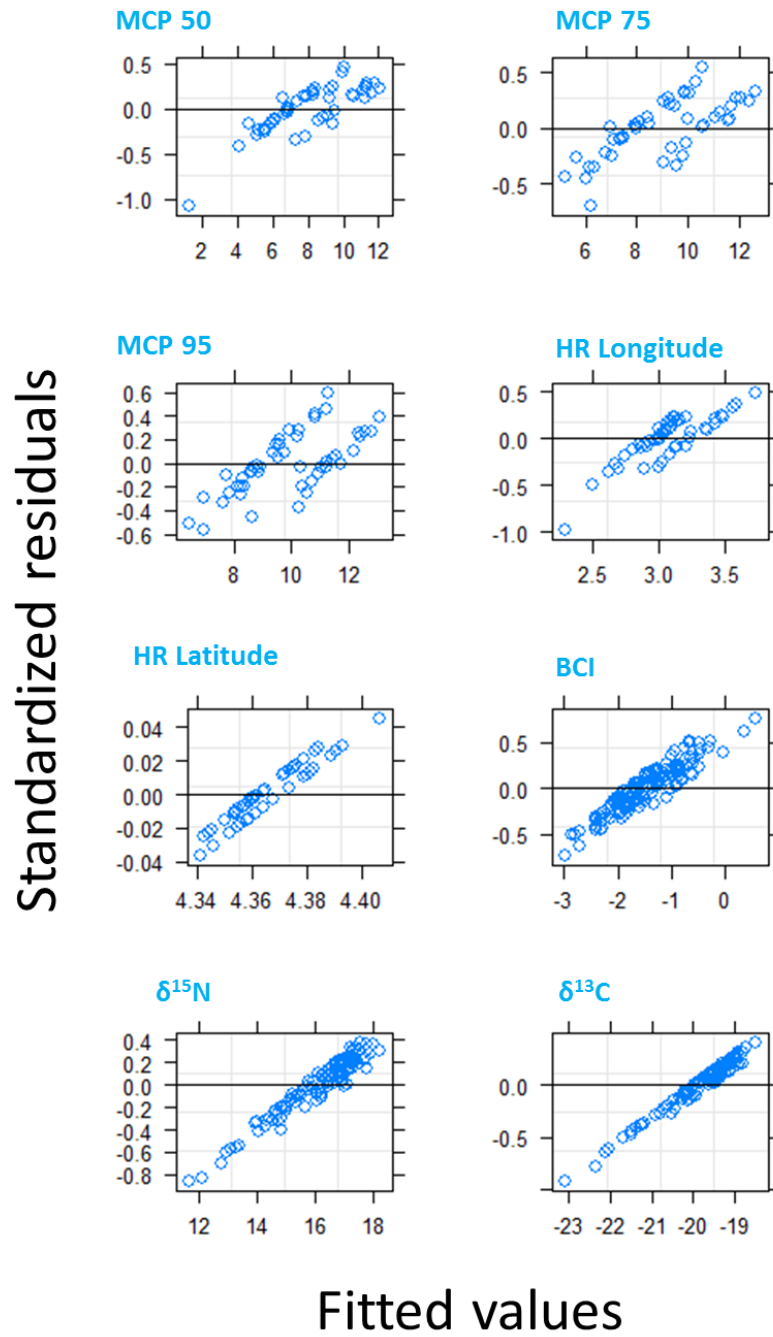


Figure S4: Response vs. fitted values for the mixed models used to assess the effect of space-use strategy (coastal or offshore) on space-use components (average annual home ranges, average annual longitudinal and latitudinal position), body condition (BCI) and diet proxies ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) in female polar bears from the Barents Sea (2000-2014).

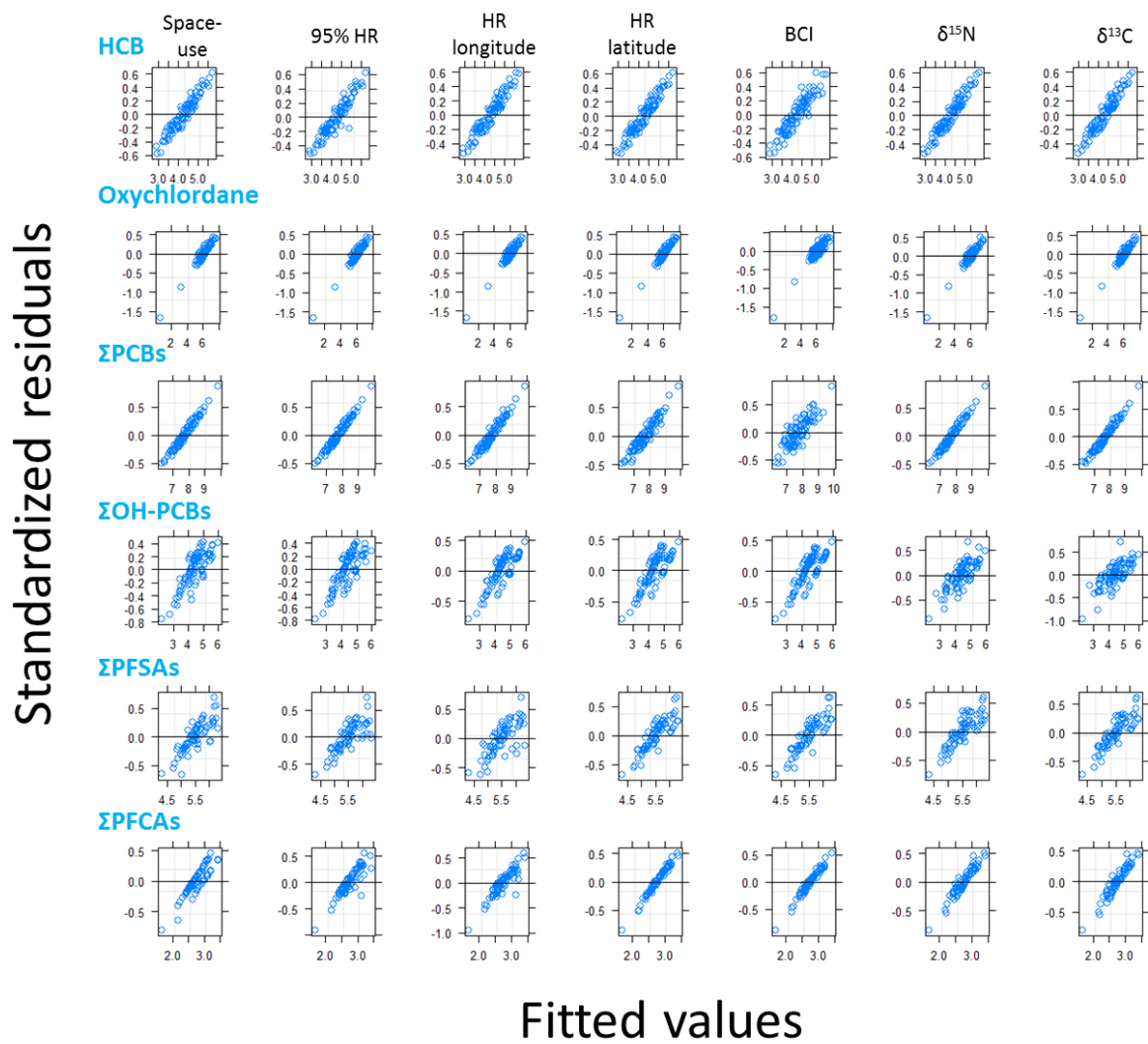


Figure S5: Response vs. fitted values for seven of the candidate models used to explain ln-transformed concentrations of pollutants according to space-use strategy (coastal or offshore), space-use components (average annual home ranges, average annual longitudinal and latitudinal position), body condition (BCI) and diet proxies ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) in female polar bears from the Barents Sea (2000-2014).