Temporal trends of persistent organic pollutants in Barents Sea polar bears (*Ursus maritimus*) in relation to changes in feeding habits and body condition

*Anna Lippold,[†,‡] Sophie Bourgeon,[‡] Jon Aars,[†] Magnus Andersen,[†] Anuschka Polder,[§] Jan Ludvig Lyche,[§] Jenny Bytingsvik,[∥,⊥] Bjørn Munro Jenssen,[⊥] Andrew E. Derocher,[#] Jeffrey M. Welker,[∇,○,△] and Heli Routti,[†]**

[†] Norwegian Polar Institute, Tromsø, Norway

[‡] The Arctic University of Norway (UiT), Tromsø, Norway

[§] Norwegian University of Life Sciences (NMBU), Oslo, Norway

[∥] Akvaplan-niva AS, Tromsø, Norway

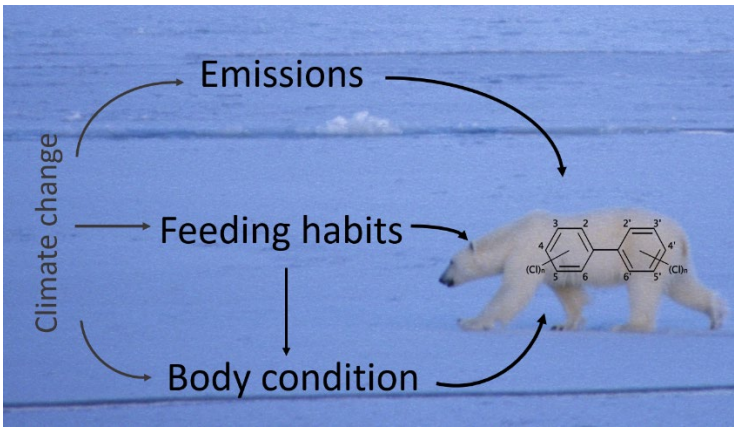
[⊥] Norwegian University of Science and Technology (NTNU) Trondheim, Norway

[#] University of Alberta, Edmonton, Canada

[∇] University of Alaska Anchorage (UAA), Anchorage, USA

[○] University of Oulu, Finland

△ University of the Arctic



18

ABSTRACT

Temporal trends of persistent organic pollutants (POPs: PCBs, OH-PCBs, *p,p'*-DDE, HCB, β-HCH, oxychlordane, BDE-47 and 153) in relation to changes in feeding habits and body condition in adult female polar bears (*Ursus maritimus*) from the Barents Sea subpopulation were examined over 20 years (1997-2017). All 306 samples were collected in the spring (April). Both stable isotope values of nitrogen (δ^{15}N) and carbon (δ^{13}C) from red blood cells declined over time, with a steeper trend for δ^{13}C between 2012 and 2017, indicating a decreasing intake of marine and high trophic level prey items. Body condition, based on morphometric measurements, had a non-significant decreasing tendency between 1997 and 2005, and increased significantly between 2005 and 2017. Plasma concentrations of BDE-153 and β-HCH did not significantly change over time, whereas concentrations of Σ_4PCB, Σ_5OH-PCB, BDE-47 and oxychlordane declined linearly. Concentrations of *p,p'*-DDE and HCB, however, declined until 2012 and 2009, respectively, and increased thereafter. Changes in feeding habits and body condition did not significantly affect POP trends. The study indicates that changes in diet and body condition were not the primary driver of POPs in polar bears, but were controlled in large part by primary and/or secondary emissions of POPs.

INTRODUCTION

Although the Arctic is barely industrialized and inhabited by less than one percent of the world's population,[1] its wildlife is exposed to high levels of long-range transported environmental contaminants. Persistent organic pollutants (POPs) are the dominant contaminants in the Arctic, and have chemical and biological properties that may affect wildlife and human health. However, few studies have the opportunity to examine longer-term trends in apex species that may reflect the cumulative biogeochemistry of POPs in the Arctic.

POPs are relatively resistant to degradation[2, 3] and reach the Arctic from distant sites of production and use via air and ocean currents as well as river outflows.[4] Owing to their lipophilic character POPs accumulate in biota and biomagnify through the food web, leading to high concentrations in apex species such as polar bears (*Ursus maritimus*).[5-8] Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are quantitatively the most abundant compounds in polar bear adipose tissue, whereas polybrominated diphenyl ethers (PBDEs) are found at lower concentrations.[9, 10] Among lipophilic POPs and their metabolites, hydroxylated (OH-) PCBs dominate in the blood circulation.[9, 10] OH-PCBs in polar bears originate from biotransformation of accumulated PCBs rather than from dietary bioaccumulation.[11] POPs have also been associated with adverse effects on wildlife[12, 13] and humans.[14, 15] In polar bears, these effects include for example alterations of the thyroid and steroid hormone systems, vitamin A levels, the immune system, lipid metabolism, and bone density.[16-26]

PCBs and OCPs were first regulated in the 1970s by national bans,[27] followed by international regulations by the United Nations Environment Program's Stockholm Convention on restriction or elimination of POPs, which entered into force in 2004. Owing to these regulations, most POP

concentrations in the Arctic have declined since the 1990s in both the air and biota.[27, 28] However, more recently, some POP concentrations have levelled off or increased.[29-31]

Contaminant levels in biota are affected by different biological and chemical factors, in addition to the emission history.[32] For instance, female polar bears transfer contaminants to their offspring through lactation,[33, 34] and thus adult female contaminant body burdens are lower than in males,[35] vary more seasonally, and accumulate less with age.[36] Seasonal variations in food availability and consequently body condition[37] also affect contaminant concentration in polar bears, because plasma levels of lipophilic contaminants tend to be more concentrated in lean compared to fat animals.[9, 38, 39]

Polar bears from the Barents Sea are among the most polluted polar bear subpopulations within the Arctic.[40-42] Contaminants are transported to the Barents Sea area by atmospheric and oceanic currents from North America and Europe,[43, 44] and river outflows from Russia.[45] Additionally, the decline of Arctic sea ice is most distinct in this area.[46] The melting sea ice might lead to secondary emissions of POPs,[47] as well as to ecological alterations in Arctic marine food webs.[48-51]

Polar bears feed mostly on ringed seals (*Pusa hispida*), and to a lesser extent on bearded (*Erignathus barbatus*) and harp seals (*Pagophilus groenlandicus*).[52-54] However, polar bears are opportunistic and will also prey or scavenge on other marine and terrestrial species like narwhal (*Monodon monoceros*), belugas (*Delphinapterus leucas*), bowhead whales (*Balaena mysticetus*), walrus (*Odobenus rosmarus*), reindeer (*Rangifer tarandus*), and seabirds.[53, 55-58] Dietary changes associated with climate driven loss of sea ice have been related to the contaminant burden in some polar bear populations.[59-61] Body condition is associated with changes in the concentrations of lipophilic POPs in Barents Sea polar bears at a seasonal and spatial scale,[9, 62] and therefore, long-term changes in body condition are also likely to affect trends of lipophilic POPs in polar bears.

There is a lack of knowledge on temporal trends of POPs in polar bears from the Barents Sea. Henriksen et al.[38] reported declining concentrations CB-153 in polar bears sampled during the 1990s, whereas Derocher et al.[63] reported variable changes in POP concentrations between 1967 and 1993-94, and Bytingsvik et al.[64] documented declining PCB concentrations between 1998 and 2008. It was hypothesised that both emission patterns and changes in feeding habits and body condition, possibly related to climate change, affect temporal trends of lipophilic POPs in Barents Sea polar bears over the last two decades. To explore this hypothesis, plasma samples from Barents Sea polar bears from 1997 until 2017 were examined and analysed for several PCBs and OH-PCBs, OCPs and PBDEs, and stable isotope values of carbon and nitrogen (δ^{13}C and δ^{15}N) were analysed in red blood cells to determine diet trends, representing carbon source (marine vs. terrestrial) and trophic level, respectively. Non-diet-adjusted contaminant trends were then compared to trends adjusted for diet and body condition in order to examine if and how climate-associated diet changes have affected contaminant levels in Svalbard polar bears from 2000 to 2017.

MATERIALS AND METHODS

Field Sampling

Adult female polar bears were opportunistically captured each year between 25th March and 5th May in the Barents Sea area during 1997-2017 as part of a yearly monitoring program run by the Norwegian Polar Institute. The 306 samples were taken from 185 individuals, of which 54 were recaptured 2 to 8 times. The bears were immobilized with tiletamine and zolazepam hydrochloride (Zoletil Forte Vet®; Virbac, France) by remote injection from a helicopter. The blood samples were stored in the cold and dark in heparinized tubes until centrifuged (3500 rpm, 10 min, within 10 h). Both red blood cell and plasma samples were stored at -20 °C until contaminant and stable

isotope analysis. A vestigial premolar tooth was taken for age estimation,[65] except for bears earlier captured and juveniles. Polar bears in this study were not weighed before 2005, thus the mass of all individuals was estimated based on body length and axillary girth (within 8% of scale mass[66]) to avoid overestimation for a part of the individuals. The body condition index (BCI) was determined based on estimated body mass and length accordingly: BCI = (ln(body mass) – 3.07· ln(length) + 10.76) ÷ (0.17 + 0.009· ln(length)).[67]

The female polar bears were either captured alone, or with cub(s) of the year (COY) or with one year old offspring (yearling, YRL). Additional information of the biology of the bears used in this study can be found in Table S1 of the supplementary information. All of the described procedures were approved by the National Animal Research Authority (NARA), Norway.

Proxies for feeding habits

As proxies for feeding habits stable isotope values of carbon and nitrogen ($\delta^{13}C$ and $\delta^{15}N$, respectively) were determined in red blood cells (n = 289, 2000-2017). In polar bear red blood cells, estimated half-lives of $\delta^{13}C$ and $\delta^{15}N$ are 1-2 and 3-4 months, respectively[68] and thus represented carbon and nitrogen sources that could stem from the previous late winter diet or from earlier accumulated fat in bears that were fasting. Analytical procedures were previously described,[59, 62] and the quality assurance is summarized in Table S2.

Chemical Analysis of POPs

POP concentrations were determined from polar bear blood plasma (n = 306, 1997-2017). The matrix was chosen for its availability. The following contaminants were analysed (* refers to: detected in > 70% of the samples and included in the statistical analyses): PCBs (CB- 28, 52, 101, 118*, 138*, 153*, 180*); OH-PCBs (4'-OH-CB-106, 4-OH-CB-107*, 4'-OH-CB-108, 3-OH-CB-

118, 4'-OH-CB-130, 3'-OH-CB-138*, 4-OH-CB-146*, 4'-OH-CB-159*, 4'-OH-CB-172, 3'-OH-CB-180, 4-OH-CB-187*); OCPs (dichlorodiphenyldichloroethylene (*p,p'*-DDE)*, (HCB)*, hexachlorocyclohexane (HCH; α, β*, γ - isomers), oxychlordane*, trans-nonachlor, toxaphene); and PBDEs (BDE-47*, 153*). All the analyses were conducted at the Laboratory of Environmental Toxicology at The Norwegian University of Life Sciences in Oslo (NMBU), which is accredited for analysis of specific POPs in biological materials of animal origin according to the requirements of NS-EN ISO/IEC 17025 (Test 137, International Electrotechnical Commission, 2005). The extraction methods used were as previously described[69] and later modified,[70] and Gabrielsen et al.[71] described the method for the extraction of OH-metabolites. The extraction method is based on liquid/liquid extraction, and the contaminants were quantified using high resolution gas chromatography (GC, Table S3 for GC equipment). The lipid content of the samples was determined gravimetrically. To ensure quality control, samples of blind, spiked recovery, blanks, in-house controls, certified European reference materials and the AMAP Ring Test[72] were analysed with the polar bear samples.

Some of the data used in this study was used for other studies with different focus and the quality assurance details are available,[9, 64, 73, 74] and summarized in Table S3 in the SI. The recovery of spiked reference samples was relatively consistent for all samples except β-HCH (58-122%; Table S3), thus β-HCH concentrations were corrected for this variation.

Data Analysis

The statistical analysis was conducted using the program R version 3.4.2. Individual PCB congeners correlated well (r: 0.63-0.87, $p \leq 0.0001$), except for PCB-118 ($r < 0.1$, $p > 0.1$), and were summed based on their chemical structural similarity. The same was applied for OH-PCBs, which all correlated significantly (r: 0.21-0.66, $p < 0.002$). Although BDE-47 and BDE-153 have

148 a similar structure and correlated significantly ($r = 0.48$, $p < 0.0001$), they were not summed due
149 to their different emission histories.[75, 76] Lipophilic compounds (PCBs, OCPs and PBDEs) were
150 lipid-normalized prior statistical analysis (ng/g lw), while concentrations of OH-PCBs were
151 analysed in wet weight basis. All POPs were ln-transformed to approximate a normal distribution.
152 Concentrations below the limit of detection (LOD) were assigned 0.5*LOD (10.8% of *p,p'*-DDE,
153 5.2% of BDE-153, and 3.9% of 3'-OH-CB-138).

154 Temporal changes in contaminant concentration, feeding habits ($\delta^{13}C$ and $\delta^{15}N$) and BCI were
155 investigated using generalized additive mixed models (GAMM; R-package *mgcv*,[77] level of
156 smoothing (k) = 9, except for β-HCH: k = 4). Models for $\delta^{13}C$ had only year as non-linear term,
157 while the model for BCI and $\delta^{15}N$ additionally included breeding status (solitary females, females
158 with COY, and females with YRL) as fixed factor.[9, 62] As partial residual plots from the GAMMs
159 suggested non-linear trends over time, possible break points for the trends were determined using
160 model selection on maximum likelihood fitted linear mixed models (LMER, package *lme4*[78]) with
161 a list of eight candidate models, including models with years from 2005-2012 as potential break
162 points and one model without breakpoint (Table S4). The period 2005-2012 for potential break
163 points was chosen to avoid temporal trends < 5 years. The break point was chosen according to
164 the model with the lowest Akaike Information Criterion (AIC; R-package *MuMIn*[79]), unless the
165 simplest model (*i.e.* no break point) was within the selection of models with ΔAIC < 2 (Table
166 S4).[80] To quantify the yearly changes, the dataset was divided into two according to the selected
167 break point, unless the most parsimonious model was the one with no break point. Estimates for
168 the yearly changes were derived from linear mixed models for each data subset (LMER, package
169 *lme4*[78]). For assessment of their significance 95% confidence intervals (CIs) were used. Polar bear

ID was included as a random factor in all statistical analyses to account for the recaptured individuals.

GAMMs were then used to analyse the effect of year, feeding habits ($\delta^{13}C$ and $\delta^{15}N$), body condition (BCI), age, and breeding status on POP concentrations in polar bears. Nine candidate models were defined, with year as non-linear term, $\delta^{13}C$ and $\delta^{15}N$ values in red blood cells, BCI, breeding status and age as fixed predictor variables (Table S5). Highly correlated predictor variables (*i.e.* $\delta^{13}C$ and $\delta^{15}N$, $r = 0.85$, $p < 0.002$) were not included in the same models.[81] BCI, $\delta^{13}C$ and $\delta^{15}N$ were standardized (mean = 0, standard deviation = 1) to facilitate the comparison between effect sizes. Model averaging based on AIC was used to make inference from all candidate models and predictor variables. The models were ranked according to AIC (Table S5), which was then used to calculate AIC weight ($e^{(0.5(AICmin-AICi))}$; relative likelihood divided by the sum of all likelihoods). To make inference from all candidate models, AIC weights were further used to calculate model averaged estimates for all predictor variables,[80] and 95% CIs were used to determine whether the parameters were significantly different from 0 at the 5% confidence level.

Plots of the highest ranked GAMMs (with the lowest AICs) were used to depict temporal trends of POP concentrations in polar bear plasma. The plots from the highest ranked GAMMs illustrate trends adjusted for their respective most influential predictor variable(s) and thus reflect temporal trends of POP concentrations that polar bears were exposed to. The plots from the adjusted models were then visually compared to plots from models with only year as a predictor variable, which reflect temporal trends of POP concentration measured in polar bear plasma. Break points for the POP trends as well as quantification of yearly changes were determined as described above for diet parameters and BCI. However, as POP concentrations were ln-transformed, the annual changes (%) in the median concentration were calculated using the following formula: 100 * (e

193 $^{\text{estimate for year}}$ – 1). Covariates for adjusted trends in LMERs were included according to the highest
194 ranked GAMMs. Polar bear ID was included as a random factor for all analyses with contaminants
195 as response variables. Throughout the analyses, diagnostic plots were used to assess whether the
196 distribution of the model residuals met the model assumptions, *i.e.* constant variation of residuals
197 (Figure S1). Residual plots revealed two outliers for oxychlordane models. After exclusion of the
198 two outliers with oxychlordane below LOD, estimates for breeding status (with COY vs. solitary)
199 and age changed from non-significant (95% CI -0.15, 0.25 and -0.034, 0.0024, respectively) to
200 significant (Table 2). However, the estimates are likely more robust without the outliers.

201 RESULTS AND DISCUSSION

202 *POP concentrations*

203 Fifteen compounds were analysed and detected in ≥ 70 % of the samples and are summarized in
204 Table 1, additional concentrations are given in Table S7. CB-153 had the highest concentrations
205 (mean ranging from 789-3446 ng/g lipid weight) in polar bear plasma, followed by CB-180 (471-
206 1798 ng/g) and oxychlordane (256-1513 ng/g; Table 1 and Table S7). The other contaminants
207 followed in decreasing order: $\sum_5$OH-PCBs > HCB > *p,p'*-DDE and β-HCH > BDE-47 > BDE-153
208 (Table 1). This is in accordance with earlier studies on polar bears from the Barents Sea and other
209 areas such as Alaska and eastern Greenland.[16, 82, 83]

210 *Trends of biological variables*

211 Ratios for δ^{13}C and δ^{15}N decreased over the study, which is in accordance with Routti et al.[59] in a
212 study from 2000-2014 including trends of stable isotope ratios (δ^{13}C and δ^{15}N) partly based on the
213 same polar bears. Average values for δ^{13}C decreased in total by 0.85‰ units from 2000 to 2012
214 (95% CIs: -1.2, -0.5). Between 2012 and 2017, average values for δ^{13}C decreased an additional

1.12 ‰ (95% CIs: -1.51, -0.5). The yearly decline was thus steeper during the latter (0.28‰) than during the former period (0.08‰). The "Suess effect", *e.g.* the gradual decrease of $\delta^{13}C$ in the atmosphere due to combustion of fossil fuels, has likely very little influence on the observed $\delta^{13}C$ decrease in polar bears. Instead, the $\delta^{13}C$ decrease in polar bears was found to be over four times higher than the changes attributed to the Suess effect.[84] As carbon isotopes ($\delta^{13}C$) indicate sources of primary productivity,[85] *e.g.* marine vs. terrestrial, our results suggest a growing proportion of terrestrial food items in polar bear diet over the study, especially after 2012.

Average values for $\delta^{15}N$ decreased linearly in total by -0.98 ‰ from 2000 to 2017 (95% CIs: -1.48, -0.49; change per year: 0.061‰; Figure 1). The decline in $\delta^{15}N$ which fractionates and changes predictably between trophic levels[85] and thus reflects trophic position, indicates a shift of polar bear diet towards a lower trophic level. This is in accordance with the trend for $\delta^{13}C$, as terrestrial Arctic food chains are shorter than Arctic marine food chains and thereby a shift towards a terrestrial diet would mean a shift towards lower trophic levels.[6] As previously suggested, the change in polar bear diet is likely linked to the sea ice decline in the Barents Sea.[59] The number of days per year with optimal habitat for polar bears has decreased over time in the Barents Sea area, as has the spatial overlap of polar bears and ringed seals in summer and autumn.[86, 87] A shift towards a less marine and lower trophic level diet linked to sea ice extend has also been reported at a spatial scale for Barents Sea polar bears.[62, 73] However, a clear conclusion about a shift in diet cannot be drawn, as the depletion of stable isotope ratios could also be related to changes at the base of the food web,[88, 89] or, possibly, changes in length of the fasting period.[90]

Average BCI values (corrected for breeding status) had an estimated decreasing tendency with confidence intervals slightly crossing 0 from 1997 until 2005 (-0.03 BCI scale units/year; 95% CIs: -0.09, 0.03; Figure 1) and increased significantly thereafter (0.02 BCI scale units/year; 95%

CIs: 0.003, 0.04). The decreasing tendency in BCI between 1997 and 2005 translates to a loss of 1.3 kg/year (95% CIs: -3.52, 1.01 kg) for a bear with average body condition and length, whereas the increase in BCI since 2005 translates to a gain of 0.84 kg/year (95 % CIs: 0.12 kg, 1.56 kg). The declining tendency in BCI between 1997 and 2005 is in accordance with the results reported in a study on female polar bears from the Southern Hudson Bay subpopulation, where a significant decrease in body condition of 1.3 kg/year between 1984 and 2009 was reported.[91] Decline in available sea ice habitat has been related to decrease in body condition in the Southern Beaufort Sea subpopulation,[92, 93] whereas a 44 days increase in the number of days with reduced sea ice was not associated to any changes in body condition in polar bears from the Chukchi Sea subpopulation.[94] Unexpectedly, body condition of female polar bears from the Barents Sea has increased after 2005, although sea ice has retreated by ~ 50% since the late 1990s in the area,[95] and the length of the ice-free season has increased by over 20 weeks between 1979 and 2013.[46] These changes are also accompanied by winter sea ice retreat that is especially pronounced in the Barents Sea compared to other Arctic areas.[96] Despite the declining sea ice in the Barents Sea, polar bears are likely not lacking food as long as sea ice is present during their peak feeding period. Polar bears feed extensively from April to June when ringed seals have pups and are particularly vulnerable to predation, whereas the predation rate during the rest of the year is likely low.[97, 98] The decline of sea ice in the Barents Sea has led to high densities of ringed seals in spring in areas where sea ice is present.[99] Furthermore, due to a lack of snow, some pups are born on open ice, making them vulnerable to predation.[99] Telemetry studies suggest that ringed seals and polar bears used the same areas close to the coast of Svalbard and still have a high degree of spatial overlap during spring despite changing sea ice conditions.[87]

Relationships between biological variables and POP concentrations

All the highest ranked statistical models (GAMMs) included diet proxies ($\delta^{13}C$ or $\delta^{15}N$), and BCI or breeding status as predictors (Table S5). Model averaged estimates showed that concentration of nearly all contaminants increased with trophic level and increasing proportion of carbon from marine sources (Table 2). Furthermore, concentrations of all compounds except *p,p'*-DDE were higher in lean compared to fatter individuals (Table 2). These findings are consistent with studies on Barents Sea polar bears (using some of the same females as in this study), which focused on seasonal and spatial differences of POPs.[9, 73] Body condition index had a slightly higher impact on POP concentrations than diet, except for HCB, BDE-47 and $\sum_5$OH-PCBs, where carbon source or trophic level influenced concentrations more than BCI (Table 2). Concentrations of $\sum_4$PCB, BDE-153 and $\sum_5$OH-PCB were higher in females with COYs compared to solitary females, whereas contaminant concentrations were similar in females with yearlings compared to solitary females (Table 2). After giving birth, female polar bears nurse their cubs in the den for more than two months entirely relying on their body fat.[100, 101] This leads to weight loss (-0.36 BCI units, CIs: -0.49, -0.23 for females with COYs compared to solitary females) and to higher contaminant concentrations as bears deplete their energy stores, however, nursing females also transfer a part of the lipophilic contaminant burden to their offspring via the lipid enriched (about 20-45% [102, 103]) milk.[34] After a year, the nursing females increase their body condition, and the lipophilic compounds become less concentrated (Table 2; [34]).

Temporal trends of POPs

Levels of $\sum_4$PCB and $\sum_5$OH-PCBs in polar bear plasma (not adjusted for biological variables) declined from 1997 to 2017 by 5 and 6% per year, respectively (see Figure 2 for LMER trend estimates and 95% CIs). As OH-PCBs originate by a large degree from biotransformation of PCBs

283 in polar bears,[11] their trends are expected to follow the PCB trend. PCB concentrations have
284 declined since the early 1990s as shown for CB-153, which declined in plasma samples from
285 Barents Sea polar bears from 1990 to 1998.[38] When the trend of $\sum_4$PCB was corrected for carbon
286 source and BCI, the declining trend tended to level off (Figure 2), whereas the adjustments did not
287 change the trend for $\sum_5$OH-PCB. However, break point analyses did not suggest any significant
288 change in the $\sum_4$PCB trend (Table S6). PCB and OH-PCB concentrations in Arctic foxes (*Vulpes*
289 *lagopus*) from Svalbard also declined from 1997 to 2013.[104, 105] About half of 347 analysed PCB
290 trends declined in the Arctic biota, whereas the remaining time series showed no trend or the trend
291 was non-linear.[31, 28] However, PCB concentrations in East Greenland polar bears were found to
292 increase by 31% between 2008 and 2013.[106] In air, the decline of PCB at three Arctic stations
293 including Svalbard, slowed down in recent years.[30] PCB concentrations generally display a less
294 pronounced decline after 2000 in both biotic and abiotic matrices.[30, 31] This might be due to
295 climate-change driven secondary emissions,[50, 107] while ongoing emission from inadvertent
296 production or poorly disposed PCB containing products can also not be excluded.[108-110]

297 The four OCPs analysed had different temporal trend patterns. Model (GAMM) results indicated
298 that concentrations of *p,p'*-DDE declined by 6% per year before 2012, and increased thereafter by
299 21% per year until 2017 (Figure 2). The decline was slightly steeper when the trend was corrected
300 for its best model covariates (Figure 2), however with widely overlapping 95% CIs. The decline
301 of *p,p'*-DDE is consistent with studies on Arctic foxes from Svalbard sampled between 1997-
302 2012.[104] Also, ΣDDT concentrations declined in East Greenland polar bears between 1983 and
303 2008 and increased thereafter until 2011.[106] Most time series in Arctic biota starting before 2000
304 reported declining trends or no trend for *p,p'*-DDE.[28, 31] Although it was suggested that *p,p'*-DDE
305 concentrations in air are more regulated by transport from direct sources than by secondary

306 emissions, the increase of *p,p'*-DDE from 2012 to 2017 might possibly be related to *e.g.* boreal
307 forest fires that released previously stored DDE.[111]

308 Concentrations of HCB declined by 6% per year before 2009, and increased thereafter by 8% per
309 year until 2017 in the present study (Figure 2). Correcting the trend for its best model covariates
310 (Figure 2) moderately affected it, however with widely overlapping 95% CIs. The decline of HCB
311 concentrations is not consistent with studies on Arctic foxes from Svalbard, as concentrations in
312 the Arctic fox food web were stable from 1997-2012.[104] However, non-linear trends were not
313 investigated in the Arctic fox study. HCB increased over the last decade in air samples from
314 Svalbard and Iceland, as well as in black guillemot (*Cepphus grylle*) eggs and male polar bears
315 from East Greenland.[29, 30] HCB has a long atmospheric lifetime and high vapour pressure,[111-113]
316 however, its atmospheric concentrations correlated only weakly or not at all with ambient
317 temperature and sea ice cover at several Arctic stations.[111, 114] The weak correlation of HCB with
318 ambient temperature and sea ice cover suggests that HCB concentrations in air are more influenced
319 by primary than secondary emissions. HCB is still emitted, as it can be formed as a by-product
320 under the production of chlorinated chemicals and incomplete combustion processes.[111, 115]
321 Although Bossi et al.[111] argued that HCB concentrations are primarily driven by primary
322 emissions, it is likely that the increasing trend after 2009 observed in the present study is also
323 affected by secondary emission, *i.e.* re-emission from the retreat of sea ice and increased
324 volatilization by increasing atmospheric temperatures.

325 Concentrations of β-HCH were stable over the study period, also when corrected for trophic level
326 and BCI. Similarly, β-HCH concentrations in Arctic foxes were stable during 1997-2013 in
327 Svalbard.[104] More than half of the temporal trend studies including β-HCH showed non-significant
328 or non-linear trends in biota, additionally, both increasing and decreasing trends were found.[28, 31]

For instance, β-HCH concentrations decreased in East Greenland polar bears between 1983 and 2006[106] and in belugas from the Eastern Beaufort Sea between 2005 and 2015;[116] and increased later in the East Greenland polar bear subpopulation between 2006 and 2013.[106] The dominant transport pathway of β-HCH to the Arctic operates via the ocean,[117] which could have led to continuous re-emissions from melting ice into the ocean.[118, 119] The high loss of sea ice in the Barents Sea[95, 96] could explain the relatively stable levels of β-HCH in the present study.

Concentrations of oxychlordane declined non-linearly by 7% per year over the study (Figure 2). LMERs did not suggest a significant break point (see Table S6) and correcting for trophic level and BCI affected the trend moderately (Figure 2). The decline of oxychlordane is consistent with trends in Arctic foxes from Svalbard.[104] However, about two thirds of the 20 time series of oxychlordane reported for Arctic biota showed no trends or non-linear trends, and about one third non-linearly decreasing trends in Arctic biota.[28, 31]

BDE-47 decreased by 3% per year, and correcting the trend for trophic level and breeding status did not significantly affect it. Interestingly, about 30 % of BDE-47 trend studies reviewed by Riget et al.[31] in Arctic wildlife reported increasing trends, and only about 10% reported declining trends. BDE-47 is quantitatively the major component in commercial penta-BDE, which has been the most used commercial PBDE mixture.[75, 76] The decline of BDE-47 is consistent with the regulation of the penta-BDE mixture, which started in the early 2000s by the European Union and the U.S.,[120, 121] and even earlier on a national level.[122] Tetra-and penta-BDE were added to the Stockholm Convention in 2009.[75, 76]

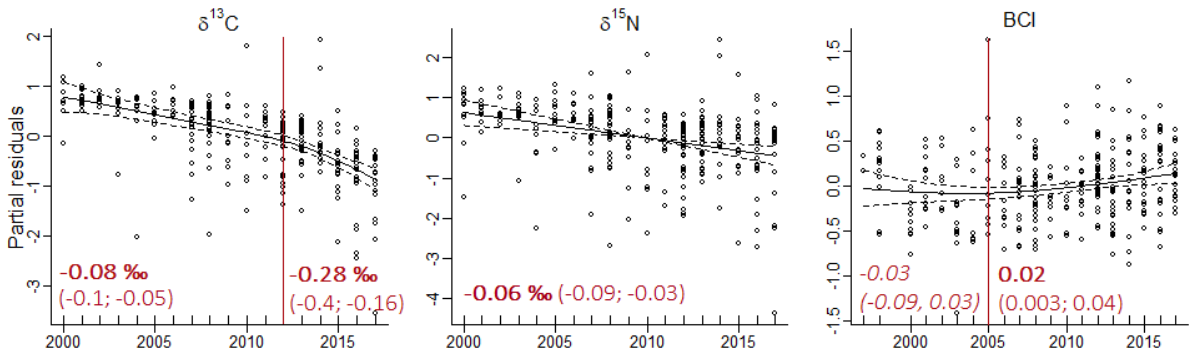
BDE-153 concentrations, in contrast, remained stable over the study in the polar bear plasma, while BDE-153 concentrations adjusted for the variation in trophic level and body condition increased by about 3% per year. Dietz et al.[123] reported an increasing trend of BDE-153

concentrations in East Greenland polar bears between 1983 and 2010. BDE-153 is only contained in commercial PBDE mixtures as a minor component, but its presence in the environment can also result from debromination of BDE-209.[124, 125] The commercial deca-BDE mixture contains about 98% of BDE-209 and was produced at much higher quantities than the penta- and octa-BDE mixtures, and predominately used in the Americas and Asia.[126] The commercial deca-BDE mixture was added to the Stockholm Convention in 2017.[127]

Temporal trends of PBDEs in Arctic biota vary spatially. Houde et al.[128] reported increasing trends of PBDEs in Canadian ringed seals before 2008, and a decline thereafter. Concentrations of summed PBDEs in East Greenland polar bears and Canadian belugas were stable between 1991-2007 and 1997-2013, respectively.[129, 130] Dietz et al.[123] reported increasing trends of summed PBDEs in East Greenland polar bears between 1983 and 2010. The discrepancy between studies on PBDE time trends may be related to the spatial variation in production and use. For instance, the majority (> 97%) of the world's total penta-BDE was used in North America, where it also was used longer than in Europe.[75]

In conclusion, POP concentrations in Svalbard polar bears have generally been declining from 1997 until 2017. However, concentrations of *p,p'*-DDE and HCB increased during the second half of the present study, while BDE-153 increased slightly over the study (the latter only when adjusted for the variation in trophic level and BCI). The increases may be related to climate-change driven secondary emissions,[50, 107] and/or potential ongoing primary emission or application.[108, 109, 131] The shift in diet towards lower trophic level and less marine food items did not significantly affect contaminant trends in the present study, yet this could be expected if the diet shift of polar bears becomes more distinct. Contaminant trends might become more difficult to predict in the context of ongoing climate change, as impacts are expected to be far-reaching in respect to ecology

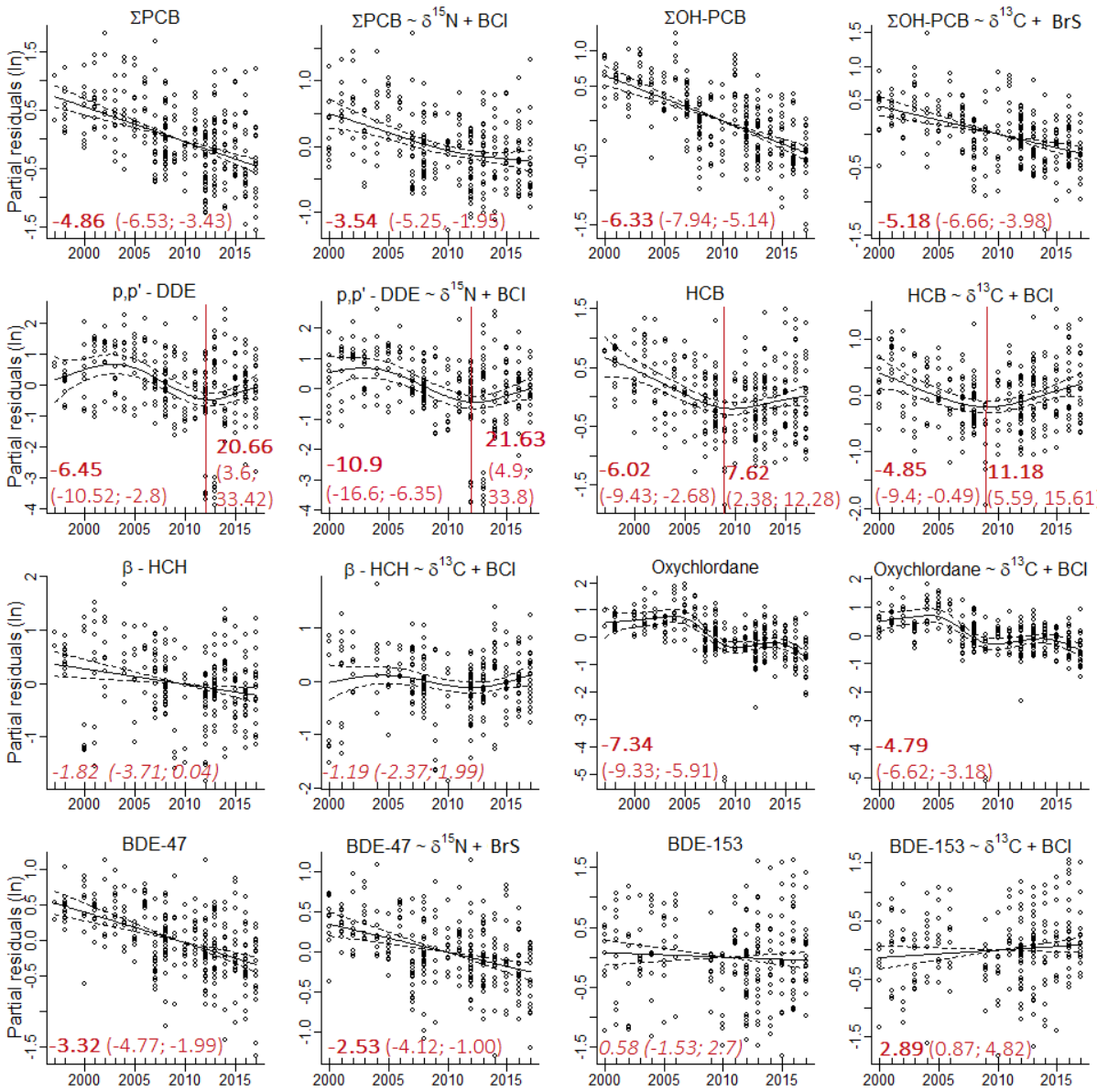
(*e.g*. changes in food webs or migration patterns), biology (*e.g.* changes in body condition or reproduction), or the distribution in abiotic compartments (*e.g.* contaminant pathways, distribution or storage).



379

380 **Figure 1**. Trends of $\delta^{13}C$, $\delta^{15}N$ and body condition (BCI) of Barents Sea polar bears from 1997/
381 2000 until 2017. Ratios for $\delta^{13}C$ and $\delta^{15}N$ represent carbon source (high values: marine diet, low
382 values: terrestrial diet), and trophic level, respectively, in polar bear winter diet. Ratios for $\delta^{15}N$
383 were significantly influenced by breeding status and therefor corrected for it.[62] BCI indicates the
384 "fatness" of the bears (corrected for breeding status; arbitrary scale without units). The Y-axis of
385 all plots show partials residuals (the actual values for stable isotope ratios and BCI can be found
386 in the SI, Table S1). Trends are shown in ‰ for diet proxies and as scale units for BCI for the
387 given time period, with 95% CI (derived from lme), and indicate change per year. Trends in italics
388 are not significant.

389



390

391 **Figure 2**. Temporal trends of Σ4PCB, Σ5OH-PCBs OCPs, and PBDEs in adult female polar bears
392 from The Barents Sea area, 1997(2000)-2017. Left column: non adjusted trends; right column:
393 adjusted for biological variables (BCI: body condition index; BrS: breeding status). The trend
394 estimates (% change per year) are derived from linear mixed models (lmer) and given with 95%
395 confidence intervals. The y-axes show partial residuals of the highest ranked GAMM (Table S5),

i.e. the effects of year have been controlled for the variables included in the highest ranked model for the given compound. Trends in bold are significant, trends in italics are not.

399 **Table 1**. Median, minimum and maximum concentrations contaminants in plasma samples of
400 female polar bears collected in the Barents Sea area between 1997 and 2017. All compounds are
401 expressed in ng/g lipid weight except for Σ_5OH-PCB (ng/g wet weight). No samples were taken in
402 1999. n.a.: not analysed. Σ_4PCB: CB-118, 138, 153, 180; Σ_5OH-PCB: 4-OH-CB-107, 3'-OH-CB-
403 138, 4-OH-CB-146, 4'-OH-CB-159, 4-OH-CB-187.

Year	n	Lipid %	Σ_4PCB	Σ_5OH-PCB	*p,p'*-DDE	HCB	β-HCH	OxyCHL	BDE-47	BDE-153
1997	2	0.9	5661	n.a.	42	203	42	1087	17	n.a.
		0.7; 1.1	3082; 8240		40; 45	201;	35; 49	727; 1447	14; 20	
1998	13	1.1	3208	n.a.	24	168	28	740	20	n.a.
		0.9; 1.3	2315; 10188		7; 44	62; 283	18; 45	545; 1589	11; 49	
2000	10	1.1	3746	156	24	104	4	977	18	3.82
		0.6; 1.4	1736; 11199	35; 251	6; 226	36; 346	2; 61	447; 1775	3.10; 42	0.66; 7.68
2001	11	1	5066	151	83	258	23	858	21	4.11
		0.6; 1.6	2700; 14453	96; 210	5; 119	128;	2; 86	503; 3468	8.78; 28	0.74; 10
2002	9	1	5422	113	79	92	21	1259	17	3.65
		0.7; 1.5	2274; 22175	66; 230	8; 143	40; 460	12; 56	716; 3039	8.03; 44	0.71; 12
2003	11	1.3	3333	151	35	84	n.a.	689	21	2.48
		1; 1.6	1654; 5930	90; 289	8; 127	28; 292		345; 1034	14; 33	2.01; 8.65
2004	10	1	4185	143	58	126	24	1198	16	3.13
		0.5; 1.6	1500; 14461	40; 213	6; 287	44; 219	10; 136	458; 3879	6.77; 37	0.59; 9.85
2005	10	1.2	3948	113	59	114	15	1513	13	3.78
		0.7; 1.4	2101; 14166	84; 198	5; 130	35; 301	3; 51	343; 3621	6.25; 26	0.81; 8.58
2006	10	1.1	4564	196	52	111	30	1307	28	5.30
		0.8; 1.3	2141; 9267	73; 524	8; 257	18; 233	11; 53	250; 2726	19; 42	0.88; 10
2007	19	1.5	1778	105	22	78	21	405	7.41	n.a.
		0.8; 2.1	914; 21535	34; 192	4; 130	24; 229	7; 51	207; 1710	3.67; 18	
2008	31	1.3	1887	73	21	73	14	514	11	n.a.
		0.8; 1.6	743; 9003	15; 130	4; 228	33; 339	6; 42	172; 2155	1.30; 34	
2009	10	1.2	2059	71	20	37	3	295	13	2.59
		0.8; 1.7	1060; 6760	27; 141	5; 134	15; 109	2; 38	1; 956	6.47; 57	0.85; 7.06
2010	10	1.2	1924	64	7	56	13	432	9.11	1.82
		0.8; 1.5	777; 2855	26; 141	4; 74	27; 204	3; 30	245; 794	6.32; 21	0.63; 3.12
2011	13	1.3	3461	159	20	123	n.a.	385	16	4.20
		0.9; 1.6	1539; 7978	63; 290	7; 263	46; 324		282; 1552	6.73; 25	2.05; 11
2012	33	1.2	1426	74	14	59	18	351	10	2.19
		0.8; 1.7	513; 3910	29; 149	0; 103	21; 206	4; 40	21; 953	2.73; 51	0.56; 9.09
2013	29	1.2	2239	66	25	111	26	467	12	3.91
		0.8; 2	930; 12068	32; 262	0; 182	31; 603	11; 95	172; 1859	2.74; 31	0.56; 20
2014	16	1.2	2296	51	6	90	22	477	8.90	2.37
		0.5; 1.6	603; 12087	10; 171	4; 474	21; 219	3; 91	101; 1232	1.25; 29	0.62; 18
2015	17	1.3	2410	63	20	104	15	461	10	3.03
		0.9; 1.5	871; 9208	12; 178	5; 80	24; 566	6; 54	173; 960	2.18; 55	0.33; 17
2016	23	1.2	1394	45	17	87	13	313	8.03	3.68
		0.8; 1.6	558; 12772	24; 184	1; 153	29; 352	4; 53	90; 1195	2.57; 29	0.74; 21
2017	19	1.3	1508	46	16	69	14	256	8.23	2.71
		1.1; 1.3	310; 9512	6; 147	0; 85	19; 294	4; 75	42; 1394	1.24; 22	0.73; 13

Table 2. GAMM-derived model-averaged estimates with 95 % confidence intervals (in brackets) explaining the ln-transformed concentrations of POPs (ng/g lipid weight, and ln/g wet weight for Σ_5OH-PCBs) in female polar bears from the Barents Sea, Norway, by feeding habits (δ^{13}C and δ^{15}N), body condition index (BCI), and breeding status (YRL: with yearlings, COY: with cubs of the year). Age was also included in the models (years; range: 7-19). Values for diet proxies and BCI have been standardized to attain comparability. Σ_4PCB: CB-118, 138, 153, 180; Σ_5OH-PCB: 4-OH-CB-107, 3'-OH-CB-138, 4-OH-CB-146, 4'-OH-CB-159, 4-OH-CB-187.

response	(intercept)	δ^{15}N red blood cells	δ^{13}C red blood cells	BCI	breeding status: YRL	breeding status: COY	age
ln(Σ_4PCB)	7.82 (7.74, 7.91)	0.11 (0.03, 0.19)	0.12 (0.03, 0.20)	-0.3 (-0.37, -0.22)	-0.25 (-0.47, -0.03)	0.24 (0.07, 0.41)	-0.02 (-0.04, -0.003)
ln(Σ_5OH-PCB)	4.29 (4.20, 4.37)	0.20 (0.13, 0.26)	0.24 (0.17, 0.31)	-0.04 (-0.10, 0.02)	-0.01 (-0.15, 0.17)	0.31 (0.19, 0.43)	0.00 (-0.02, 0.01)
ln(*p,p'*-DDE)	2.36 (-2.11, 6.82)	0.16 (-0.01, 0.33)	0.11 (-0.08, 0.29)	0.29 (0.08, 0.38)	-0.01 (-0.44, 0.42)	-0.56 (-0.89, -0.22)	-0.01 (-0.04, 0.03)
ln(HCB)	4.56 (4.31, 4.81)	0.08 (-0.01, 0.16)	0.14 (0.05, 0.24)	-0.09 (-0.16, -0.009)	-0.15 (-0.37, 0.07)	0.07 (-0.1, 0.24)	-0.02 (-0.03, -0.002)
ln(β-HCH)	3.17 (3.06, 3.3)	0.15 (0.05, 0.25)	0.13 (-0.02, 0.24)	-0.28 (-0.38, -0.19)	-0.40 (-0.68, -0.12)	0.1 (-0.12, 0.31)	-0.03 (-0.05, -0.01)
ln(OxyCHL)	6.18 (6.08, 6.28)	0.12 (0.03, 0.22)	0.11 (-0.001, 0.21)	-0.25 (-0.3, -0.16)	-0.32 (-0.58, -0.06)	0.05 (-0.15, 0.25)	-0.02 (-0.03, 0.002)
ln(BDE-47)	0.36 (-2.19, 2.91)	0.2 (0.12, 0.27)	0.19 (0.11, 0.27)	-0.09 (-0.15, 0.02)	-0.07 (-0.26, 0.11)	0.13 (-0.01, 0.27)	-0.01 (-0.03, 0.004)
ln(BDE-153)	1.1 (0.99, 1.19)	0.11 (0.009, 0.21)	0.1 (-0.03, 0.18)	-0.34 (-0.4, -0.25)	-0.17 (-0.45, 0.11)	0.41 (0.19, 0.62)	-0.01 (-0.03, 0.007)

ASSOCIATED CONTENT

Supporting Information.

The following files are available free of charge. Biological information on the study animals, details on quality assurance for stable isotope and chemical analyses, LMER model selection tables for breakpoints of temporal trends, GAMM selection explaining POP concentration, concentrations of single PCB and OH-PCB congeners, and diagnostic residual plots of GAMMs explaining POP concentrations (PDF).

AUTHOR INFORMATION

Corresponding Author

* Heli Routti: heli.routti@npolar.no; tel. +4777750541

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Funding Sources

The study was funded by the Norwegian Polar Institute, the Ministry of Climate and Environment, the Norwegian Environment Agency and the Research Council of Norway. Additional funding for fieldwork was provided by the Center for Ice, Climate and Environment (ICE) at the Norwegian Polar Institute and the World Wildlife Fund (WWF).

ACKNOWLEDGMENT

The authors thank Rupert Krapp (NPI, Longyearbyen), the helicopter (Airlift) and Lance crews for logistic help and Øystein Wiig for his help in the field. Further acknowledgment is addressed to Erik Gilmore, Mahin Karimi, Randi Grønnestad, Katharina Løken, Mariska Hoorweg, Astrid Kolind Riemer, and Vidar Torget for contributing to the chemical analysis, while Elisabeth Lie is acknowledged for her technical advises at the Laboratory of Environmental Toxicology. Anette Wold and Martin Kristiansen provided technical help in sample preparation, and Matthew Rogers, Annalis Brownlee in mass spectrometry analyses for stable isotopes. Sabrina Tartu helped organizing the data set. Kjetil Sagerup and Dorothee Ehrich provided helpful comments on the study.

REFERENCES

1. Emelyanova, A., Population projections of the Arctic by levels of education. *International Institute for Applied Systems Analysis, Austria* **2017**, (WP-17-022),
2. Jones, K. C.; Voogt, P. d., Persistent organic pollutants (POPs): state of the science. *Environ. Pollut.* **1999,** *100*, (1-3), 209-221. 10.1016/S0269-7491(99)00098-6
3. Sinkkonen, S.; Paasivirta, J., Degradation half-life times of PCDDs, PCDFs and PCBs for environmental fate modeling. *Chemosphere* **2000,** *40*, (9-11), 943-949. 10.1016/S0045-6535(99)00337-9
4. Macdonald, R. W.; Harner, T.; Fyfe, J.; Loeng, H.; Weingartner, T. *AMAP Assessment 2002: The Influence of Global Change on Contaminant Pathways to, within, and from the Arctic.*; Oslo, Norway, 2003; p xii+65.
5. Hoekstra, P. F.; O'Hara, T. M.; Fisk, A. T.; Borgå, K.; Solomon, K. R.; Muir, D. C. G., Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort–Chukchi Seas. *Environ. Pollut.* **2003,** *124*, (3), 509-522. 10.1016/s0269-7491(02)00482-7
6. Kelly, B. C.; Ikonomou, M. G.; Blair, J. D.; Morin, A. E.; Gobas, F. A. P. C., Food Web–Specific Biomagnification of Persistent Organic Pollutants. *Science* **2007,** *317*, (5835), 236-239. 10.1126/science.1138275
7. Muir, D.; Norstrom, R. J.; Simon, M., Organochlorine Contaminants in Arctic Marine Food Chains: Accumulation of Specific Polychlorinated Biphenyls and Chlordane-Related Compounds. *Environ. Sci. Technol.* **1988,** *22*, (9), 1071-1079. 10.1021/es00174a012
8. Sørmo, E. G.; Jenssen, B. M.; Lie, E.; Skaare, J. U., Brominated Flame Retardants in Aquatic Organisms from the North Sea in Comparision with Biota from the High Arctic Marine Environment. *ET&C* **2009,** *28*, (10), 2082-2090. 10.1897/08-452.S1

9. Tartu, S.; Bourgeon, S.; Aars, J.; Andersen, M.; Polder, A.; Thiemann, G. W.; Welker, J. M.; Routti, H., Sea ice-associated decline in body condition leads to increased concentrations of lipophilic pollutants in polar bears (Ursus maritimus) from Svalbard, Norway. *Sci. Total Environ.* **2017,** *576*, 409-419. 10.1016/j.scitotenv.2016.10.132
10. Letcher, R. J.; Morris, A. D.; Dyck, M.; Sverko, E.; Reiner, E. J.; Blair, D. A. D.; Chu, S. G.; Shen, L., Legacy and new halogenated persistent organic pollutants in polar bears from a contamination hotspot in the Arctic, Hudson Bay Canada. *Sci. Total Environ.* **2018,** *610-611*, 121-136. 10.1016/j.scitotenv.2017.08.035
11. Letcher, R. J.; Gebbink, W. A.; Sonne, C.; Born, E. W.; McKinney, M. A.; Dietz, R., Bioaccumulation and biotransformation of brominated and chlorinated contaminants and their metabolites in ringed seals (Pusa hispida) and polar bears (Ursus maritimus) from East Greenland. *Environ. Int.* **2009,** *35*, (8), 1118-24. 10.1016/j.envint.2009.07.006
12. AMAP *Biological Effects of Contaminants on Arctic Wildlife and Fish*; Tromsø, Norway, 2018; p vii+84.
13. Routti, H.; Jenssen, B. M.; Tartu, S., Ecotoxicologic Stress in Arctic Marine Mammals, With Particular Focus on Polar Bears. In *Marine Mammal Ecotoxicology - Impacts of Multiple Stressors on Population Health*, Fossi, C. M.; Panti, C., Eds. Elsevier: 2018; pp 345-380.
14. Kim, K. H.; Kabir, E.; Jahan, S. A., Exposure to pesticides and the associated human health effects. *Sci. Total Environ.* **2017,** *575*, 525-535. 10.1016/j.scitotenv.2016.09.009
15. van den Berg, M.; Kypke, K.; Kotz, A.; Tritscher, A.; Lee, S. Y.; Magulova, K.; Fiedler, H.; Malisch, R., WHO/UNEP global surveys of PCDDs, PCDFs, PCBs and DDTs in human milk and benefit-risk evaluation of breastfeeding. *Arch. Toxicol.* **2017,** *91*, (1), 83-96. 10.1007/s00204-016-1802-z
16. Braathen, M.; Derocher, A. E.; Wiig, Ø.; Sørmo, E. G.; Lie, E.; Skaare, J. U.; Jenssen, B. M., Relationships between PCBs and Thyroid Hormones and Retinol in Female and Male Polar Bears. *Environ. Health Perspect.* **2004,** *112*, (8), 826-833. 10.1289/ehp.6809
17. Skaare, J. U.; Bernhoft, A.; Wiig, O.; Norum, K. R.; Haug, E.; Eide, D. M.; Derocher, A. E., Relationships between plasma levels of organochlorines, retinol and thyroid hormones from polar bears (Ursus maritimus) at Svalbard. *J. Toxicol. Environ. Health A.* **2001,** *62*, (4), 227-41. 10.1080/009841001459397
18. Bourgeon, S.; Riemer, A. K.; Tartu, S.; Aars, J.; Polder, A.; Jenssen, B. M.; Routti, H., Potentiation of ecological factors on the disruption of thyroid hormones by organo-halogenated contaminants in female polar bears (Ursus maritimus) from the Barents Sea. *Environ. Res.* **2017,** *158*, 94-104. 10.1016/j.envres.2017.05.034
19. Oskam, I. C.; Ropstad, E.; Dahl, E.; Lie, E.; Derocher, A. E.; Wiig, O.; Larsen, S.; Wiger, R.; Skaare, J. U., Organochlorines affect the major androgenic hormone, testosterone, in male polar bears (Ursus maritimus) at Svalbard. *J. Toxicol. Environ. Health A.* **2003,** *66*, (22), 2119-39. 10.1080/15287390390211342
20. Bernhoft, A.; Skaare, J. U., Possible Immunotoxic Effects of Organochlorines in Polar Bears (Ursus Maritimus) at Svalbard. *J. Toxicol. Environ. Health* **2000,** *59*, (7), 561-574. 10.1080/009841000156682
21. Sonne, C.; Dietz, R.; Born, E. W.; Riget, F. F.; Kirkegaard, M.; Hyldstrup, L.; Letcher, R. J.; Muir, D. C. G., Is Bone Mineral Composition Disrupted by Organochlorines in East Greenland Polar Bears (Ursus maritimus)? *Environ. Health Perspect.* **2004,** *112*, (17), 1711-1716. 10.1289/ehp.7293

22. Daugaard-Petersen, T.; Langebaek, R.; Riget, F. F.; Letcher, R. J.; Hyldstrup, L.; Jensen, J. B.; Bechshoft, T.; Wiig, O.; Jenssen, B. M.; Pertoldi, C.; Lorenzen, E. D.; Dietz, R.; Sonne, C., Persistent organic pollutants, skull size and bone density of polar bears (Ursus maritimus) from East Greenland 1892-2015 and Svalbard 1964-2004. *Environ. Res.* **2018,** *162*, 74-80. 10.1016/j.envres.2017.12.009
23. Sonne, C.; Dyck, M.; Riget, F. F.; Beck Jensen, J. E.; Hyldstrup, L.; Letcher, R. J.; Gustavson, K.; Gilbert, M. T.; Dietz, R., Penile density and globally used chemicals in Canadian and Greenland polar bears. *Environ. Res.* **2015,** *137*, 287-91. 10.1016/j.envres.2014.12.026
24. Tartu, S.; Lille-Langoy, R.; Storseth, T. R.; Bourgeon, S.; Brunsvik, A.; Aars, J.; Goksoyr, A.; Jenssen, B. M.; Polder, A.; Thiemann, G. W.; Torget, V.; Routti, H., Multiple-stressor effects in an apex predator: combined influence of pollutants and sea ice decline on lipid metabolism in polar bears. *Scientific reports* **2017,** *7*, (1), 16487. 10.1038/s41598-017-16820-5
25. Ciesielski, T. M.; Hansen, I. T.; Bytingsvik, J.; Hansen, M.; Lie, E.; Aars, J.; Jenssen, B. M.; Styrishave, B., Relationships between POPs, biometrics and circulating steroids in male polar bears (Ursus maritimus) from Svalbard. *Environ. Pollut.* **2017,** *230*, 598-608. 10.1016/j.envpol.2017.06.095
26. Gustavson, L.; Ciesielski, T. M.; Bytingsvik, J.; Styrishave, B.; Hansen, M.; Lie, E.; Aars, J.; Jenssen, B. M., Hydroxylated polychlorinated biphenyls decrease circulating steroids in female polar bears (Ursus maritimus). *Environ. Res.* **2015,** *138*, 191-201. 10.1016/j.envres.2015.02.011
27. Li, Y. F.; Macdonald, R. W., Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. *Sci Total Environ* **2005,** *342*, (1-3), 87-106. 10.1016/j.scitotenv.2004.12.027
28. AMAP *Temporal Trends in Persistent Organic Pollutants in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway (www.amap.no), 2015; p vi+71pp.
29. Riget, F.; Vorkamp, K.; Bossi, R.; Sonne, C.; Letcher, R. J.; Dietz, R., Twenty years of monitoring of persistent organic pollutants in Greenland biota. A review. *Environ. Pollut.* **2016,** *217*, 114-23. 10.1016/j.envpol.2015.11.006
30. Hung, H.; Katsoyiannis, A. A.; Brorstrom-Lunden, E.; Olafsdottir, K.; Aas, W.; Breivik, K.; Bohlin-Nizzetto, P.; Sigurdsson, A.; Hakola, H.; Bossi, R.; Skov, H.; Sverko, E.; Barresi, E.; Fellin, P.; Wilson, S., Temporal trends of Persistent Organic Pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP). *Environ. Pollut.* **2016,** *217*, 52-61. 10.1016/j.envpol.2016.01.079
31. Rigét, F.; Bignert, A.; Braune, B.; Dam, M.; Dietz, R.; Evans, M.; Green, N.; Gunnlaugsdóttir, H.; Hoydal, K. S.; Kucklick, J. R.; Letcher, R. J.; Muir, D.; Schuur, S.; Sonne, C.; Stern, G.; Tomy, G.; Vorkamp, K.; Wilson, S., Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota. *Sci. Total Environ.* **2019,** *649*, 10.1016/j.scitotenv.2018.08.268
32. Borgå, K.; Fisk, A. T.; Hoekstra, P. F.; Muir, D. C. G., Biological and Chemical Factors of Importance in the Bioaccumulation and Trophic Transfer of Persistent Organochlorine Contaminants in Arctic Marine Food Webs. *ET&C* **2004,** *23*, (10), 2367-2385. 10.1897/03-518
33. Bernhoft, A.; Wiig, O.; Skaare, J. U., Organochlorines in polar bears (ursus maritimus) at Svalbard. *Environ. Pollut.* **1997,** *95*, (2), 159-175. 10.1016/S0269-7491(96)00122-4

34. Polischuk, S. C.; Norstrom, R. J.; Ramsay, M. A., Body burdens and tissue concentrations of organochlorines in polar bears (Ursus maritimus) vary during seasonal fasts. *Environ. Pollut.* **2002,** *118*, (1), 29-39. 10.1016/S0269-7491(01)00278-0
35. Norstrom, R. J.; Belikov, S. E.; Born, E. W.; Garner, G. W.; Malone, B.; Olpinski, S.; Ramsay, M. A.; Schliebe, S.; Stirling, I.; Stishov, M. S.; Taylor, M. K.; Wiig, Ø., Chlorinated Hydrocarbon Contaminants in Polar Bears from Eastern Russia, North America, Greenland, and Svalbard: Biomonitoring of Arctic Pollution. *Arch. Environ. Contam. Toxicol.* **1998,** *35*, (2), 354–367. 10.1007/s002449900387
36. Dietz, R.; Riget, F. F.; Sonne, C.; Letcher, R.; Born, E. W.; Muir, D. C., Seasonal and temporal trends in polychlorinated biphenyls and organochlorine pesticides in East Greenland polar bears (Ursus maritimus), 1990-2001. *Sci. Total Environ.* **2004,** *331*, (1-3), 107-24. 10.1016/j.scitotenv.2004.03.025
37. Stirling, I.; McEwan, E. H., The caloric value of whole ringed seals (Phoca hispida) in relation to polar bear (Ursus maritimus) ecology and hunting behavior. *Can. J. Zool.* **1975,** *53*, 1021-1027. 10.1139/z75-117
38. Henriksen, E. O.; Wiig, Ø.; Skaare, J. U.; Gabrielsen, G. W.; Derocher, A. E., Monitoring PCBs in polar bears: lessons learned from Svalbard. *J. Environ. Monit.* **2001,** *3*, (5), 493-498. 10.1039/b102683f
39. Polischuk, S. C.; Letcher, R. J.; Norstrom, R. J.; Ramsay, M. A., Preliminary results of fasting on the kinetics of organochlorines in polar bears (Ursus maritimus). *Sci. Total Environ.* **1995,** *160*, (161), 465-472. 10.1016/0048-9697(95)04380-J
40. McKinney, M. A.; Letcher, R. J.; Aars, J.; Born, E. W.; Branigan, M.; Dietz, R.; Evans, T. J.; Gabrielsen, G. W.; Peacock, E.; Sonne, C., Flame retardants and legacy contaminants in polar bears from Alaska, Canada, East Greenland and Svalbard, 2005-2008. *Environ. Int.* **2011,** *37*, (2), 365-74. 10.1016/j.envint.2010.10.008
41. Muir, D. C. G.; Backus, S.; Derocher, A. E.; Dietz, R.; Evans, T. J.; Gabrielsen, G. W.; Nagy, J.; Norstrom, R. J.; Sonne, C.; Stirling, I.; Taylor, M. K.; Letcher, R. J., Brominated flame retardants in polar bears (Ursus maritimus) from Alaska, the Canadian Arctic, East Greenland, and Svalbard. *Environ. Sci. Technol.* **2006,** *40*, (2), 449-455. 10.1021/es051707u
42. Andersen, M.; Lie, E.; Derocher, A.; Belikov, S. E.; Bernhoft, A.; Boltunov, A. N.; Garner, G. W.; Skaare, J. U.; Wiig, O., Geographic variation of PCB congeners in polar bears (Ursus maritimus) from Svalbard east to the Chukchi Sea. *Polar Biol.* **2001,** *24*, (4), 231-238. 10.1007/s003000000201
43. Hansen, B.; Larsen, K. M. H.; Hátún, H.; Kristiansen, R.; Mortensen, E.; Østerhus, S., Transport of volume, heat, and salt towards the Arctic in the Faroe Current 1993–2013. *Ocean Sci.* **2015,** *11*, (5), 743-757. 10.5194/os-11-743-2015
44. Shindell, D. T.; Chin, M.; Dentener, F.; Doherty, R. M.; Faluvegi, G.; Fiore, A. M.; Hess, P.; Koch, D. M.; MacKenzie, I. A.; Sanderson, M. G.; Schultz, M. G.; Schulz, M.; Stevenson, D. S.; Teich, H.; Textor, C.; Wild, O.; Bergmann, D. J.; Bey, I.; Bian, H.; Cuvelier, C.; Duncan, B. N.; Folberth, G.; Horowitz, L. W.; Jonson, J.; Kaminski, J. W.; Marmer, E.; Park, R.; Pringle, K. J.; Schroeder, S.; Szopa, S.; Takemura, T.; Zeng, G.; Keating, T. J.; Zuber, A., A multi-model assessment of pollution transport to the Arctic. *Atmos. Chem. Phys.* **2008,** *8*, 5353–5372. 10.5194/acp-8-5353-2008
45. Burkow, I. C.; Kallenborn, R., Sources and transport of persistent pollutants to the Arctic. *Toxicol. Lett.* **2000,** *112*, (113), 87-92. 10.1016/S0378-4274(99)00254-4

46. Laidre, K. L.; Stern, H.; Kovacs, K. M.; Lowry, L.; Moore, S. E.; Regehr, E. V.; Ferguson, S. H.; Wiig, Ø.; Boveng, P.; Angliss, R. P.; Born, E. W.; Litovka, D.; Quakenbush, L.; Lydersen, C.; Vongraven, D.; Ugarte, F., Arctic marine mammal population status, sea ice habitat loss, and conservation recommendations for the 21st century. *Conserv. Biol.* **2015,** *29*, (3), 724-737. 10.1111/cobi.12474
47. Ma, J.; Hung, H.; Tian, C.; Kallenborn, R., Revolatilization of persistent organic pollutants in the Arctic induced by climate change. *Nat. Clim. Change* **2011**, (1), 255-260. 10.1038/nclimate1167
48. Post, E.; Forchhammer, M. C.; Bret-Harte, M. S.; Callaghan, T. V.; Christensen, T. R.; Elberling, B.; Fox, A. D.; Gilg, O.; Hik, D. S.; Høye, T. T.; Ims, R. A.; Jeppesen, E.; Klein, D.; Madsen, J.; McGuire, D.; Rysgaard, S.; Schindler, D. E.; Stirling, I.; Tamstorf, M. P.; Tyler, N. J. C.; van der Wal, R.; Welker, J. M.; Wookey, P. A.; Schmidt, N. M.; Aastrup, P., Ecological Dynamics Across the Arctic Associated with Recent Climate Change. *Science* **2009,** *325*, (5946), 1355-1358. 10.1126/science.1173113
49. Wassmann, P.; Duarte, C. M.; Agustì, S.; Sejr, M. K., Footprints of climate change in the Arctic marine ecosystem. *Glob. Change Biol.* **2011,** *17*, (2), 1235-1249. 10.1111/j.1365-2486.2010.02311.x
50. Macdonald, R. W.; Harner, T.; Fyfe, J., Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. *Sci. Total Environ.* **2005,** *342*, (1-3), 5-86. 10.1016/j.scitotenv.2004.12.059
51. Kallenborn, R.; Halsall, C.; Dellong, M.; Carlsson, P., The influence of climate change on the global distribution and fate processes of anthropogenic persistent organic pollutants. *J. Environ. Monit.* **2012,** *14*, 2854–2869. 10.1039/c2em30519d
52. Derocher, A.; Wiig, Ø.; Andersen, M., Diet composition of polar bears in Svalbard and the western Barents Sea. *Polar Biol.* **2002**, (25), 448-452. 10.1007/s00300-002-0364-0
53. Bentzen, T. W.; Follmann, E. H.; Amstrup, S. C.; York, G. S.; Wooller, M. J.; O'Hara, T. M., Variation in winter diet of southern Beaufort Sea polar bears inferred from stable isotope analysis. *Can. J. Zool.* **2007,** *85*, (5), 596-608. 10.1139/z07-036
54. Thiemann, G.; Iverson, S.; Stirling, I., Polar bear diets and arctic marine food webs: Insights from fatty acid analysis. *Ecol. Monogr.* **2008,** *78*, (4), 591-613. 10.1890/07-1050.1
55. Calvert, W.; Stirling, I., Interactions between polar bears and overwintering walruses in the central canadian high Arctic. In *Bears: Their Biology and Management - A Selection of Papers from the Eighth International Conference on Bear Research and Management*, International Association for Bear Research and Management: Victoria, British Columbia, Canada, February 1989 (1990) 1990; Vol. 8, pp 351-356.
56. Derocher, A.; Wiig, O.; Bangjord, G., Predation of Svalbard reindeer by Polar bears. *Polar Biol.* **2000**, (23), 675-678. 10.1007/s003000000138
57. Smith, T. G.; Sjare, B., Predation of Belugas and Narwhals by Polar Bears in Nearshore Areas of the Canadian High Arctic. *Arctic* **1990,** *43*, (2), 99-102. 10.14430/arctic1597
58. Stempniewicz, L., The polar bear Ursus maritimus feeding in a seabird colony in Frans Josef Land *Polar Research* **1993,** *12*, (1), 33-36. 10.1111/j.1751-8369.1993.tb00420.x
59. Routti, H.; Aars, J.; Fuglei, E.; Hanssen, L.; Lone, K.; Polder, A.; Pedersen, A. O.; Tartu, S.; Welker, J. M.; Yoccoz, N. G., Emission Changes Dwarf the Influence of Feeding Habits on Temporal Trends of Per- and Polyfluoroalkyl Substances in Two Arctic Top Predators. *Environ. Sci. Technol.* **2017,** *51*, (20), 11996-12006. 10.1021/acs.est.7b03585

60. McKinney, M. A.; Iverson, S. J.; Fisk, A. T.; Sonne, C.; Riget, F. F.; Letcher, R. J.; Arts, M. T.; Born, E. W.; Rosing-Asvid, A.; Dietz, R., Global change effects on the long-term feeding ecology and contaminant exposures of East Greenland polar bears. *Glob. Change Biol.* **2013,** *19*, (8), 2360-72. 10.1111/gcb.12241
61. McKinney, M. A.; Peacock, E.; Letcher, R., Sea Ice-associated Diet Change Increases the Levels of Chlorinated and Brominated Contaminants in Polar Bears. *Environ. Sci. Technol.* **2009,** *43*, (12), 4334–4339. 10.1021/es900471g
62. Tartu, S.; Bourgeon, S.; Aars, J.; Andersen, M.; Ehrich, D.; Thiemann, G. W.; Welker, J. M.; Routti, H., Geographical Area and Life History Traits Influence Diet in an Arctic Marine Predator. *PLoS One* **2016,** *11*, (5), e0155980. 10.1371/journal.pone.0155980
63. Derocher, A.; Wolkers, H.; Colborn, T.; Schlabach, M.; Larsen, T. S.; Wiig, Ø., Contaminants in Svalbard polar bear samples archived since 1967 and possible population level effects. *Sci. Total Environ.* **2003,** *301*, (1-3), 163-174. 10.1016/S0048-9697(02)00303-0
64. Bytingsvik, J.; Lie, E.; Aars, J.; Derocher, A. E.; Wiig, O.; Jenssen, B. M., PCBs and OH-PCBs in polar bear mother-cub pairs: a comparative study based on plasma levels in 1998 and 2008. *Sci. Total Environ.* **2012,** *417-418*, 117-28. 10.1016/j.scitotenv.2011.12.033
65. Christensen-Dalsgaard, S. N.; Aars, J.; Andersen, M.; Lockyer, C.; Yoccoz, N. G., Accuracy and precision in estimation of age of Norwegian Arctic polar bears (Ursus maritimus) using dental cementum layers from known-age individuals. *Polar Biol.* **2009,** *33*, (5), 589-597. 10.1007/s00300-009-0734-y
66. Derocher, A. E.; Wiig, Ø., Postnatal growth in body length and mass of polar bears (Ursus maritimus) at Svalbard. *J. Zool.* **2002,** *256*, (3), 343-349. 10.1017/s0952836902000377
67. Cattet, M. R. L.; Caulkett, N. A.; Obbard, M. E.; Stenhouse, G. B., A body-condition index for ursids. *Can. J. Zool.* **2002,** *80*, (7), 1156-1161. 10.1139/z02-103
68. Rode, K. D.; Stricker, C. A.; Erlenbach, J.; Robbins, C. T.; Cherry, S. G.; Newsome, S. D.; Cutting, A.; Jensen, S.; Stenhouse, G.; Brooks, M.; Hash, A.; Nicassio, N., Isotopic Incorporation and the Effects of Fasting and Dietary Lipid Content on Isotopic Discrimination in Large Carnivorous Mammals. *Physiol. Biochem. Zool.* **2016,** *89*, (3), 182-97. 10.1086/686490
69. Brevik, E. M., Gas Chromatographic Method for the Determination of Organochlorine Pesticides in Human Milk. *Bull. Environ. Contam. Toxicol.* **1978,** *19*, (1), 281-286. 10.1007/BF01685799
70. Polder, A.; Gabrielsen, G. W.; Odland, J. O.; Savinova, T. N.; Tkachev, A.; Loken, K. B.; Skaare, J. U., Spatial and temporal changes of chlorinated pesticides, PCBs, dioxins (PCDDs/PCDFs) and brominated flame retardants in human breast milk from Northern Russia. *Sci. Total Environ.* **2008,** *391*, (1), 41-54. 10.1016/j.scitotenv.2007.10.045
71. Gabrielsen, G. W.; Villanger, G. D.; Lie, E.; Karimi, M.; Lydersen, C.; Kovacs, K. M.; Jenssen, B. M., Levels and patterns of hydroxylated polychlorinated biphenyls (OH-PCBs) and their associations with thyroid hormones in hooded seal (Cystophora cristata) mother-pup pairs. *Aquat. Toxicol.* **2011,** *105*, (3-4), 482-91. 10.1016/j.aquatox.2011.08.003
72. AMAP *Arctic Pollution 2009. Arctic Monitoring and Assessment Programme*; Oslo, 2009; p xi+83.
73. Tartu, S.; Aars, J.; Andersen, M.; Polder, A.; Bourgeon, S.; Merkel, B.; Lowther, A. D.; Bytingsvik, J.; Welker, J. M.; Derocher, A.; Jenssen, B. M.; Routti, H., Choose your poison - Space-use strategy influences pollutant exposure in Barents Sea polar bears. *Environ. Sci. Technol.* **2018**, 10.1021/acs.est.7b06137

691 74. Bytingsvik, J.; van Leeuwen, S. P.; Hamers, T.; Swart, K.; Aars, J.; Lie, E.; Nilsen, E.
692 M.; Wiig, O.; Derocher, A. E.; Jenssen, B. M., Perfluoroalkyl substances in polar bear mother-
693 cub pairs: a comparative study based on plasma levels from 1998 and 2008. *Environ. Int.* **2012,**
694 *49*, 92-99. 10.1016/j.envint.2012.08.004
695 75. Alaee, M., An overview of commercially used brominated flame retardants, their
696 applications, their use patterns in different countries/regions and possible modes of release.
697 *Environ. Int.* **2003,** *29*, (6), 683-689. 10.1016/s0160-4120(03)00121-1
698 76. AMAP, Chemicals of Emerging Arctic Concern. *Arctic Monitoring and Assessment*
699 *Programme, Oslo, Norway.* **2016**, xvi+353.
700 77. Wood, S., Package 'mgcv'. **2018**,
701 78. Bates, D.; Mächler, M.; Bolker, B.; Walker, S., Fitting Linear Mixed-Effects Models
702 Using lme4. *J. Stat. Softw.* **2015,** *67*, (1), 1-51. 10.18637/jss.v067.i01
703 79. Barton, C., MuMIn: Multi-Model Inference. R package version 1.15.6. **2016**,
704 80. Burnham, K. P.; Anderson, D. W., *Model Selection and Multimodel Inference - A*
705 *Practical Information-Theoretic Approach*. Springer Verlag: New York, 2002.
706 81. Burnham, K. P.; Anderson, D. R.; Huyvaert, K. P., AIC model selection and multimodel
707 inference in behavioral ecology: some background, observations, and comparisons. *Behav. Ecol.*
708 *Sociobiol.* **2010,** *65*, (1), 23-35. 10.1007/s00265-010-1029-6
709 82. Kucklick, J. R.; Struntz, W. D. J.; Becker, P. R.; York, G. W.; O'Hara , T. M.;
710 Bohonowych, J. E., Persistent organochlorine pollutants in ringed seals and polar bears collected
711 from northern Alaska. *Sci. Total Environ.* **2002,** *287*, (1-2), 45-59. 10.1016/S0048-
712 9697(01)00997-4
713 83. Dietz, R.; Riget, F. F.; Sonne, C.; Born, E. W.; Bechshoft, T.; McKinney, M. A.; Letcher,
714 R. J., Three decades (1983-2010) of contaminant trends in East Greenland polar bears (Ursus
715 maritimus). Part 1: legacy organochlorine contaminants. *Environ. Int.* **2013,** *59*, 485-93.
716 10.1016/j.envint.2012.09.004
717 84. Olsen, A.; Omar, A. M.; Bellerby, R. G. J.; Johannessen, T.; Ninnemann, U.; Brown, K.
718 R.; Olsson, K. A.; Olafsson, J.; Nondal, G.; Kivimäe, C.; Kringstad, S.; Neill, C.; Olafsdottir, S.,
719 Magnitude and origin of the anthropogenic CO2increase and13C Suess effect in the Nordic seas
720 since 1981. *Global Biogeochem. Cy.* **2006,** *20*, (3), n/a-n/a. 10.1029/2005gb002669
721 85. Hobson, K. A.; Schell, D. M.; Renouf, D.; Noseworthy, E., Stable carbon and nitrogen
722 isotopic fractionation between diet and tissues of captive seals: implications for dietary
723 reconstructions involving marine mammals. *Can. J. Fish. Aquat. Sci.* **1996**, (53), 528–533
724 10.1139/f95-209
725 86. Lone, K.; Merkel, B.; Lydersen, C.; Kovacs, K. M.; Aars, J., Sea ice resource selection
726 models for polar bears in the Barents Sea subpopulation. *Ecography* **2018,** *41*, (4), 567-578.
727 10.1111/ecog.03020
728 87. Hamilton, C. D.; Kovacs, K. M.; Ims, R. A.; Aars, J.; Lydersen, C., An Arctic predator-
729 prey system in flux: climate change impacts on coastal space use by polar bears and ringed seals.
730 *J. Anim. Ecol.* **2017,** *86*, (5), 1054-1064. 10.1111/1365-2656.12685
731 88. Post, D. M., Using stable isotopes to estimate trophic position: Models, Methods, and
732 Assumptions. *Ecology* **2002,** *83*, (3), 703-718. 10.1890/0012-
733 9658(2002)083[0703:USITET]2.0.CO;2
734 89. St Louis, V. L.; Derocher, A. E.; Stirling, I.; Graydon, J. A.; Lee, C.; Jocksch, E.;
735 Richardson, E.; Ghorpade, S.; Kwan, A. K.; Kirk, J. L.; Lehnherr, I.; Swanson, H. K.,
736 Differences in mercury bioaccumulation between polar bears (Ursus maritimus) from the

737 Canadian high- and sub-Arctic. *Environ. Sci. Technol.* **2011,** *45*, (14), 5922-8.
738 10.1021/es2000672
739 90. Polischuk, S. C.; Hobson, K. A.; Ramsay, M. A., Use of stable-carbon and -nitrogen
740 isotopes to assess weaning and fasting in female polar bears and their cubs. *Can. J. Zool.* **2001,**
741 *79*, (3), 499-511. 10.1139/z01-007
742 91. Obbard, M. E.; Cattet, M. R. L.; Howe, E. J.; Middel, K. R.; Newton, E. J.; Kolenosky,
743 G. B.; Abraham, K. F.; Greenwood, C. J., Trends in body condition in polar bears (Ursus
744 maritimus) from the Southern Hudson Bay subpopulation in relation to changes in sea ice. *Arctic*
745 *Science* **2016,** *2*, (1), 15-32. 10.1139/as-2015-0027
746 92. Rode, K. D.; Amstrup, S. C.; Regehr, E. V., Reduced body size and cub recruitment in
747 polar bears associated with sea ice decline. *Ecol. Appl.* **2010,** *20*, (3), 768-782. 10.1890/08-
748 1036.1
749 93. Rode, K. D.; Wilson, R. R.; Douglas, D. C.; Muhlenbruch, V.; Atwood, T. C.; Regehr, E.
750 V.; Richardson, E. S.; Pilfold, N. W.; Derocher, A. E.; Durner, G. M.; Stirling, I.; Amstrup, S.
751 C.; St Martin, M.; Pagano, A. M.; Simac, K., Spring fasting behavior in a marine apex predator
752 provides an index of ecosystem productivity. *Glob. Change Biol.* **2018,** *24*, (1), 410-423.
753 10.1111/gcb.13933
754 94. Rode, K. D.; Regehr, E. V.; Douglas, D. C.; Durner, G.; Derocher, A. E.; Thiemann, G.
755 W.; Budge, S. M., Variation in the response of an Arctic top predator experiencing habitat loss:
756 feeding and reproductive ecology of two polar bear populations. *Glob. Change Biol.* **2014,** *20*,
757 (1), 76-88. 10.1111/gcb.12339
758 95. Årthun, M.; Eldevik, T.; Smedsrud, L. H.; Skagseth, Ø.; Ingvaldsen, R. B., Quantifying
759 the Influence of Atlantic Heat on Barents Sea Ice Variability and Retreat*. *J. Clim.* **2012,** *25*,
760 (13), 4736-4743. 10.1175/jcli-d-11-00466.1
761 96. Dobricic, S.; Vignati, E.; Russo, S., Large-Scale Atmospheric Warming in Winter and the
762 Arctic Sea Ice Retreat. *J. Clim.* **2016,** *29*, 2869-2888. 10.1175/JCLI-D-15-0417.1
763 97. Atkinson, S. N.; Ramsay, M. A.; Nelson, R. A., Changes in the Body Composition of
764 Fasting Polar Bears (Ursus maritimus): The Effect of Relative Fatness on Protein Conservation.
765 *Physiol. Biochem. Zool.* **1996,** *69*, (2), 304-316. 10.2307/30164186
766 98. Messier, F.; Taylor, M. K.; Ramsay, M. A., Seasonal activity patterns of female polar
767 bears (Ursus maritimus) in the Canadian Arctic as revealed by satellite telemetry. *J. Zool.* **1992,**
768 *226*, (2), 219-229. 10.1111/j.1469-7998.1992.tb03835.x
769 99. Lydersen, C.; Vaquie-Garcia, J.; Lydersen, E.; Christensen, G. N.; Kovacs, K. M., Novel
770 terrestrial haul-out behaviour by ringed seals (Pusa hispida) in Svalbard, in association with
771 harbour seals (Phoca vitulina). *Polar Res.* **2017,** *36*, (1), 1374124.
772 10.1080/17518369.2017.1374124
773 100. Amstrup, S. C., Human Disturbances of Denning Polar Bears in Alaska. *Arctic* **1993,** *46*,
774 (3), 246-250. 10.14430/arctic1349
775 101. Ramsay, M. A.; Stirling, I., Reproductive biology and ecology of female polar bears
776 (Ursus maritimus). *J. Zool.* **1988,** *214*, (4), 601-634. 10.1111/j.1469-7998.1988.tb03762.x
777 102. Derocher, A.; Adriashek, D.; Arnould, J. P. Y., Aspects of milk composition and lactation
778 in polar bears. *Can. J. Zool.* **1993,** *71*, (3), 561-567. 10.1139/z93-077
779 103. Oehme, M.; Biseth, A.; Schlabach, M.; Wiig, Ø., Concentrations of polychlorinated
780 Dibenzo-p-Dioxins, Dibenzofurans and non-ortho substituted biphenyls in polar bear milk from
781 Svalbard (Norway). *Environ. Pollut.* **1995,** *90*, (3), 401-407. 10.1016/0269-7491(95)00007-E

104. Andersen, M. S.; Fuglei, E.; Konig, M.; Lipasti, I.; Pedersen, A. O.; Polder, A.; Yoccoz, N. G.; Routti, H., Levels and temporal trends of persistent organic pollutants (POPs) in arctic foxes (Vulpes lagopus) from Svalbard in relation to dietary habits and food availability. *Sci. Total Environ.* **2015,** *511*, 112-22. 10.1016/j.scitotenv.2014.12.039
105. Routti, H.; Andersen, M. S.; Fuglei, E.; Polder, A.; Yoccoz, N. G., Concentrations and patterns of hydroxylated polybrominated diphenyl ethers and polychlorinated biphenyls in arctic foxes (Vulpes lagopus) from Svalbard. *Environ. Pollut.* **2016,** *216*, 264-272. 10.1016/j.envpol.2016.05.056
106. Dietz, R.; Desforges, J. P.; Gustavson, K.; Riget, F. F.; Born, E. W.; Letcher, R. J.; Sonne, C., Immunologic, reproductive, and carcinogenic risk assessment from POP exposure in East Greenland polar bears (Ursus maritimus) during 1983-2013. *Environ. Int.* **2018,** *118*, 169-178. 10.1016/j.envint.2018.05.020
107. Ma, J.; Hung, H.; Macdonald, R. W., The influence of global climate change on the environmental fate of persistent organic pollutants: A review with emphasis on the Northern Hemisphere and the Arctic as a receptor. *Glob. Planet. Change* **2016,** *146*, 89-108. 10.1016/j.gloplacha.2016.09.011
108. Vorkamp, K., An overlooked environmental issue? A review of the inadvertent formation of PCB-11 and other PCB congeners and their occurrence in consumer products and in the environment. *Sci. Total Environ.* **2016,** *541*, 1463-1476. 10.1016/j.scitotenv.2015.10.019
109. Bogdal, C.; Muller, C. E.; Buser, A. M.; Wang, Z.; Scheringer, M.; Gerecke, A. C.; Schmid, P.; Zennegg, M.; Macleod, M.; Hungerbuhler, K., Emissions of polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins, and polychlorinated dibenzofurans during 2010 and 2011 in Zurich, Switzerland. *Environ. Sci. Technol.* **2014,** *48*, (1), 482-90. 10.1021/es4044352
110. Carlsson, P.; Breivik, K.; Brorstrom-Lunden, E.; Cousins, I.; Christensen, J.; Grimalt, J. O.; Halsall, C.; Kallenborn, R.; Abass, K.; Lammel, G.; Munthe, J.; MacLeod, M.; Odland, J. O.; Pawlak, J.; Rautio, A.; Reiersen, L. O.; Schlabach, M.; Stemmler, I.; Wilson, S.; Wohrnschimmel, H., Polychlorinated biphenyls (PCBs) as sentinels for the elucidation of Arctic environmental change processes: a comprehensive review combined with ArcRisk project results. *Environmental science and pollution research international* **2018**, 10.1007/s11356-018-2625-7
111. Bossi, R.; Skjoth, C. A.; Skov, H., Three years (2008-2010) of measurements of atmospheric concentrations of organochlorine pesticides (OCPs) at Station Nord, North-East Greenland. *Environ. Sci. Process. Impacts* **2013,** *15*, (12), 2213-9. 10.1039/c3em00304c
112. Pouch, A.; Zaborska, A.; Pazdro, K., The history of hexachlorobenzene accumulation in Svalbard fjords. *Environ. Monit. Assess.* **2018,** *190*, (6), 360. 10.1007/s10661-018-6722-3
113. Wania, F.; Mackay, D., Tracking the Distribution of Persistent Organic Pollutants. *Environ. Sci. Technol.* **1996,** *30*, (9), 10.1021/es962399q
114. Su, Y.; Hung, H.; Blanchard, P.; Patton, G. W.; Kallenborn, R.; Konoplev, A.; Fellin, P.; Li, H.; Geen, C.; Stern, G.; Rosenberg, B.; Barrie, L. A., Spatial and Seasonal Variations of Hexachlorocyclohexanes (HCHs) and Hexachlorobenzene (HCB) in the Arctic Atmosphere. *Environ. Sci. Technol.* **2006,** *40*, (21), 6601-6607. 10.1021/es061065q
115. Ambrus, A.; Hamilton, D. J.; Kuiper, H. A.; Racke, K. D., Significance of impurities in the safety evaluation of crop protection products (IUPAC Technical Report). *Pure Appl. Chem.* **2003,** *75*, (7), 937-973. 10.1351/pac200375070937

827 116. Nöel, M.; Loseto, L. L.; Stern, G., Legacy contaminants in the Eastern Beaufort Sea
828 beluga whales (Delphinapterus leucas): Are temporal trends reflecting regulations? *Arctic*
829 *Science* **2018,** *4*, (3), 373-387. 10.1139/as-2017-0049
830 117. Li, Y. F.; Macdonald, R. W.; Jantunen, L. M.; Harner, T.; Bidleman, T. F.; Strachan, W.
831 M. J., The transport of b-hexachlorocyclohexane to the western Arctic Ocean: a contrast to a-
832 HCH. *Sci. Total Environ.* **2002,** *291*, (1-3), 229-246. 10.1016/S0048-9697(01)01104-4
833 118. Wohrnschimmel, H.; Tay, P.; von Waldow, H.; Hung, H.; Li, Y. F.; Macleod, M.;
834 Hungerbuhler, K., Comparative assessment of the global fate of alpha- and beta-
835 hexachlorocyclohexane before and after phase-out. *Environ. Sci. Technol.* **2012,** *46*, (4), 2047-
836 54. 10.1021/es203109q
837 119. Pucko, M.; Stern, G. A.; Macdonald, R. W.; Jantunen, L. M.; Bidleman, T. F.; Wong, F.;
838 Barber, D. G.; Rysgaard, S., The delivery of organic contaminants to the Arctic food web: why
839 sea ice matters. *Sci. Total Environ.* **2015,** *506-507*, 444-52. 10.1016/j.scitotenv.2014.11.040
840 120. EPA, Polybrominated Diphenyl Ethers (PBDEs) Action Plan. *epa.gov/assesing-and-*
841 *managing-chemicals-under-tsca* **2009**,
842 121. EU, Directive 2002/95/EC of the European Parliament and of the Council of 27 January
843 2003 on the restriction of the use of certain hazardous substances in electrical and electronic
844 equipment. *Official Journal of the European Union L37* **2003,** *46*, 19-23.
845 122. Alcock, R. E.; Busby, J., Risk migration and scientific advance: the case of flame-
846 retardant compounds. *Risk Anal.* **2006,** *26*, (2), 369-81. 10.1111/j.1539-6924.2006.00739.x
847 123. Dietz, R.; Riget, F. F.; Sonne, C.; Born, E. W.; Bechshoft, T.; McKinney, M. A.;
848 Drimmie, R. J.; Muir, D. C.; Letcher, R. J., Three decades (1983-2010) of contaminant trends in
849 East Greenland polar bears (Ursus maritimus). Part 2: brominated flame retardants. *Environ. Int.*
850 **2013,** *59*, 494-500. 10.1016/j.envint.2012.09.008
851 124. Söderström, G.; Sellström, U.; De Wit, C. A.; Tysklind, M., Photolytic Debromination of
852 Decabromodiphenyl Ether (BDE-209). *Environ Sci Technol* **2004,** *38*, (1), 127-132.
853 10.1021/es034682c
854 125. Stapleton, H. M.; Brazil, B.; Holbrook, D.; Mitchelmore, C. L.; Benedict, R.;
855 Konstantinov, A.; Potter, D., In Vivo and In Vitro Debromination of Decabromodiphenyl Ether
856 (BDE 209) by Juvenile Rainbow Trout and Common Carp. *Environ. Sci. Technol.* **2006,** *40*,
857 (15), pp 4653-4658. 10.1021/es060573x
858 126. De Wit, C. A., An overview of brominated flame retardants in the environment.
859 *Chemosphere* **2002,** *46*, (5), 583-624. 10.1016/S0045-6535(01)00225-9
860 127. Convention, S., SC-8/10: Listing of decabromodiphenyl ether. In UNEP: 2017.
861 128. Houde, M.; Wang, X.; Ferguson, S. H.; Gagnon, P.; Brown, T. M.; Tanabe, S.; Kunito,
862 T.; Kwan, M.; Muir, D. C., Spatial and temporal trends of alternative flame retardants and
863 polybrominated diphenyl ethers in ringed seals (Phoca hispida) across the Canadian Arctic.
864 *Environ. Pollut.* **2017,** *223*, 266-276. 10.1016/j.envpol.2017.01.023
865 129. McKinney, M. A.; Stirling, I.; Lunn, N. J.; Peacock, E.; Letcher, R. J., The role of diet on
866 long-term concentration and pattern trends of brominated and chlorinated contaminants in
867 western Hudson Bay polar bears, 1991-2007. *Sci. Total Environ.* **2010,** *408*, (24), 6210-22.
868 10.1016/j.scitotenv.2010.08.033
869 130. Simond, A. E.; Houde, M.; Lesage, V.; Verreault, J., Temporal trends of PBDEs and
870 emerging flame retardants in belugas from the St. Lawrence Estuary (Canada) and comparisons
871 with minke whales and Canadian Arctic belugas. *Environ. Res.* **2017,** *156*, 494-504.
872 10.1016/j.envres.2017.03.058

873 131. Wang, X.; Sun, D.; Yao, T., Climate change and global cycling of persistent organic
874 pollutants: A critical review. *Sci. China Earth Sci.* **2016,** *59*, (10), 1899-1911. 10.1007/s11430-
875 016-5073-0

876