

# UNEXPECTED INCREASES OF PERSISTENT ORGANIC POLLUTANT AND MERCURY LEVELS IN EAST GREENLAND POLAR BEARS (UNEXPECTED)

Technical Report from DCE - Danish Centre for Environment and Energy No. 214

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## Data sheet

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Abstract:	Climate change has taking place in the Arctic in recent years with significant consequences for the amount of sea ice and species occurrence and distribution including food availability. In this study, we account for the polar bears' food selection, likely influenced by the changes in sea ice, using quantitative fatty acid signature analysis (QFASA) and fatty acid specific stable carbon isotopes ( $\delta^{13}$ C-FA) to assess the nutritional basis of East Greenland polar bears over the last three decades. This shows that East Greenland polar bears mainly feed on ringed seals (44.9%), migrating subarctic harp seals (27.3%) and hooded seals (22.9%) and relatively rarely prey on other species such as bearded seals (4.9%), narwhals (1.0%) or walruses (0.06%). The time trend analysis reflects a clear correlation between contaminant concentrations and changes in food availability, i.e. organohalogen compounds and mercury increased with increasing consumption of harp and hooded seals, while per- and polyfluorinated alkylated substances (PFAS) decreased. Overall this indicates that changes in prey specieas, related to climate change, can lead to an increase in the some contaminant loads of Arctic top predators and thus potentially the East Greenlandic Inuit population living from subsistence harvest.
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## Sammenfatning

Hurtige klimatiske forandringer/opvarmninger finder sted i Arktis i disse år med betydelige fald i mængden af havis hvilket har betydning for hvilke arter, der forekommer i disse områder. Det er udfordrende at vurdere økosystemændringer i fjerntliggende polære miljøer. I denne undersøgelse har vi redegjort for isbjørnenes fødegrundlag som følge af ændringerne i havisen. Kvantitative fedtsyresignaturanalyse (QFASA) og fedtsyrespecifik stabile carbon isotoper (δ<sup>13</sup>C-FA) blev brugt til at vurdere fødegrundlaget for østgrønlandske isbjørne (Ursus maritimus) over de sidste tre artier. QFASA-genererede fødeestimater viste, at de Østgrønlandske isbjørne hovedsageligt spiser ringsæler (44,9 ± 18,27%), migrerende subarktiske grønlandssæler (27,3 ± 9,6%) og klapmydser ( $22,9 \pm 12,4\%$ ) og relativt sjældent spiser arter som remmesæler  $(4,9 \pm 8,9\%)$ , narhvaler  $(1,0 \pm 2,2\%)$  eller hvalrosser  $(0,06 \pm 0,06\%)$ . Analyserne af fedtsyre sammensætningen viser at fødegrundlaget er forskelligt blandt køn og aldersgrupper, hvor adulte hanisbjørne afveg mest i forhold til unge og voksne hunbjørne. Andelen af ringsæler var relativt stabil omkring 50% men begyndte at falde efter år 2000. I år 2016, det sidste år med FA-data, var andelen af ringsæler faldet til mindre end 25%. Den modsatte tendens blev observeret for klapmydser, der var i størrelsesorden 15% indtil 2000, hvorefter de steg til mere end 40% i 2016. Grønlandssælerne viste en stigning fra 15% til mere end 25% fra 1983 til 1994, hvorefter deres andel i kosten forblev relativt stabil indtil 2016. Ingen klare tendenser kunne dokumenteres for den procentuelle andel af remmesæler, hvalrosser og narhvaler. Tidstrend analysen viste en klar sammenhæng i øget koncentrationer af organohalogen forbindelser (OHC) og kviksølvbelastningen og et fald i koncentrationen af per- og polyfluorerede alkyl forbindelser (PFAS) knyttet til de beskrevne ændringer i fødegrundlaget, og i visse år var disse forhold ret markante (som eksempelvis i 1997 og 2013). De faldende  $\delta^{13}$ C-FA-værdier understøttede skift fra mere kystnære og is associerede byttedyr til mere offshore pelagisk associerede byttedyr, i overensstemmelse med fødeestimaterne. Øget klapmyds indtag og nedsat indtagelse af ringsæler fandt sted i år, hvor det Nordatlantiske Oscillations Indeks (NAOI) var lavere. Således var perioder med varmere temperaturer og mindre havis forbundet med indtagelse af flere subarktiske og færre arktiske sæler. Disse ændringer i den relative tilgængelighed eller fordeling af arktiske og subarktiske sæler/havpattedyr kan således have sundhedsmæssige konsekvenser for de østgrønlandske isbjørne. Eksempelvis resulterede den klimarelaterede ændring i fødegrundlaget konsekvent en højere eksponering over tid af "Sentinel" (indikator) OHC'er og PFAS'er, da de subarktiske sæler har højere koncentrationer af disse miljøfremmede stoffer end arktiske sæler. Undersøgelsen indikerer at klimaændringerne vil medføre en øgning i forureningsbelastningen af arktiske toprovdyr og dermed potentielt også af den østgrønlandske Inuit befolkning.

## Summary

Rapid climate changes are occurring in the Arctic, with substantial repercussions for arctic ecosystems which are all challenging to assess due to the remote polar environments. In the present study we were successful in monitoring the combined effects from climate change on the diet and the consecutive pollution-mediated biological health effects in EG polar bears. Quantitative fatty acid signature analysis (QFASA) and fatty acid-specific stable carbon isotope (δ13C-FA) signatures were used to assess diets of East Greenland polar bears (Ursus maritimus) over the past three decades. QFASA-generated diet estimates indicated that, on average across all years and groups, EG bears mainly consumed arctic ringed seals (44.9±18.27%), migratory subarctic harp seals (27.3±9.6%) and hooded seals (22.9±12.4%) and rarely, if ever, bearded seals (4.9±8.9%), narwhals (1.0±2.2%) and walruses (0.06±0.06%). We observed differences in this pattern among sex and age groups. Adult male polar bears deviated the most relatively to the juvenile and the adult female bears. The proportion of ringed seals was relatively stable around 50% prior to year 2000 after which it started to decline. The last year with FA data is 2016 where the ringed seal dietary percentage had declined to less than 25%. The opposite trend was observed for the hooded seals, which were in the magnitude of 15% up to 2000 after which it increased to more than 40% by 2016. Harp seals showed an increase from 15% to more than 25% from 1983 to 1994, after which their proportion in the diet remained relatively stable until 2016. No clear trends were observed for the proportions of bearded seals, walruses and narwhals. A clear relation in increased organohalogen compound (OHC) and mercury loads and a decline in per- and polyfluoroalkyl substance (PFAS) concentrations was linked to these diet shifts and for some years these relationships were quite pronounced such as in 1997 and 2013. As previously documented, declining *δ*13C-FA values supported shifts from more nearshore and ice-associated prey to more offshore pelagic associated prey, consistent with the diet estimates. Increased hooded and decreased ringed seal consumption occurred during years when the North Atlantic Oscillation index was lower. Thus, periods with warmer temperatures and less sea ice were associated with more subarctic (hooded seal and harp seal) and less arctic ringed seal species consumption. These changes in the relative abundance, accessibility, or distribution of arctic and subarctic marine mammals may have health consequences for East Greenland polar bears. For example, the diet change resulted in consistently slower temporal declines in adipose levels of legacy OHCs and PFASs, as the subarctic seals have higher contaminant burdens than arctic seals. Overall, considerable changes are occurring in the East Greenland marine ecosystem, with consequences for contaminant dynamics in arctic top predators and potentially also in subsistence societies.

## 1 Introduction

The Arctic is undergoing significant warming and ecosystem changes including rapid loss of sea-ice decline and food web dynamics (AMAP 2017; Perovich and Richter-Menge 2009). The loss of sea ice is high in East Greenland (EG) being around 9.8%/decade and is thereby among the most rapid in the Arctic (Perovich and Richter-Menge 2009). As part of this, EG polar bears are shifting diet with hooded and harp seals accounting for a steadily higher proportion than ringed seals do (McKinney et al. 2013). The EG subpopulation of polar bears is within the convergent ice ecoregion and therefore tends to have year-round access to sea ice (Amstrup et al. 2008). This provides the bears with access to marine mammal prey. Overall polar bears are predicted to be reduced by 50% by 2050 and become extinct within by 2100 due to the disappearance of the sea ice (Amstrup et al. 2008).

The EG polar bear subpopulation has access to multiple marine mammals as prey species including ringed seals (Pusa hispida), bearded seals (Erignathus barbatus), walruses (Odobenus rosmarus) and narwhals (Monodon monoceros) as well as seasonal access to harp seals (Pagophilus groenlandica) and hooded seals (Crystophora cristata; Laidre et al. 2008, McKinney et al. 2013). Harp and hooded seals migrate northward to their breeding and moulting patches around the same latitude or a bit north of Scoresby Sound (Ittoqqortoormiit), where they congregate in large numbers. The solid fast ice off EG has however, shrunk in extent over the last few decades, resulting in the ice edge closing up to land. This means that e.g. the hibernating polar bear females with their cubs wintering in their snow dens on land have easier access to these breeding and moulting patches in the spring at approximately the same time as the yearly quota of polar bears are hunted. The loss of sea ice is likely to have an effect on the access for the polar bear to the ice associated seals. In The Greenland Sea the ice loss rate have been around 9.8%/ which is one of the most rapid declines in the Arctic (Perovich & Richter-Menge 2009). Tracking data from central East Greenland polar bears indicated that bear during from spring until mid-late summer ranged further offshore in the pack ice corresponding with the harp and hooded seal whelping and molting areas (Wiig et al. 2003; Laidre et al. 2013). An increased number of the more temperate harp and hooded seals is likely to occur in a warming arctic, coinciding with northward retractions of ringed seals and their preferred preys (Laidre et al. 2008). Such shift in prey availability to polar bears has the potential of increasing contaminant concentration as well as new diseases from subarctic regions (Kleivane et al. 2000; McKinney et al. 2012).

Fatty acid (FA) analysis as a novel dietary tracer approach has been applied to archived polar bear adipose tissue sampled from 1983-today (McKinney et al. 2013). The quantitative fatty acid signature analysis (QFASA) compares FA in prey with those in polar bears and thus allows for diet estimates (Iverson et al. 2004).

Concentrations of POPs/OHCs in EG polar bears have steadily decreased over the period 1983-2000 after which the decline stopped and in some cases the concentrations have even increased again (Dietz et al. 2008, 2013a, 2018a; Rigét et al. 2018). The observed increases are largely due to shifts in prey contributing to potential negative subclinical health effects (Letcher et al. 2010; Sonne 2010; Dietz et al. 2018b, 2019). We therefore applied QFASA to the EG polar bears and investigated the contaminant loads in bearded, hooded, and harp seal as well as narwhal adipose tissue in addition to the regularly monitored ringed seals to provide an extensive temporal study of diet changes in an Arctic top predator. Estimates of polar bear estimated intake of  $\Sigma$ PCBs based on their diet estimates from the QFASA as well as the modelled  $\Sigma$ PCB loads of the prey items back in time were related to temporal changes in polychlorinated biphenyl ( $\Sigma$ PCB) concentrations in the polar bears during the same period. As such, we aimed to predict the combined effects from climate change and pollution-mediated biological health effects in EG polar bears.

## 2 Materials and methods

#### 2.1 Sample collection

Bearded, harp and hood seal as well as narwhal samples were collected from subsistence Inuit harvest in Hurry Fjord and Gasefjord (70°33'N 26°11'W), west of Scoresby Sound in East Greenland (EG) during August – September 2015. Polar bear and ringed seal samples were collected from 1983-2016 and 1986-2016, respectively, in the Scoresby Sound region of East Greenland between 69° to 74° N as part of the native subsistence hunt. Ringed seal were sampled biannually whereas polar bears samples were taken annually as part of the Danish AMAP CORE programme. After sampling and during shipment, samples were kept frozen. At the AU Specimen Bank samples are stored at -20 °C until chemical analysis.

#### 2.2 Age determination

Ages were determined by counting annual growth layer groups in the cementum of the lower right I3 tooth using established methods for the polar bears and using the canine of the lower jaw for seals (Dietz et al. 2004). The bears were classified as follows: adult males  $\geq$  6 years of age, adult females  $\geq$  5 years, and subadults consisting of all other bears (Rosing-Asvid et al. 2002). The seals were classified as follows: adult males  $\geq$  5 years of age, adult females  $\geq$  4 years, and subadults consisting of all other seals. For seals, information on the length of the animals as well as the size of sexual organs were also taken into consideration. The grouping into subadult and adult narwhals was based on the development of sexual organs and recorded zoological body length, and for males the length of the tusk as well. Subadults of both sexes were pooled for statistical analyses of OHC levels (Dietz et al. 2004, 2013a,b, 2018a).

#### 2.3 Chemical analysis for OHCs and PFAS

Organohalogen compound (OHC) concentrations in polar bear subcutaneous adipose were previously reported in two recent temporal trend studies of legacy and emerging OHCs (Dietz et al. 2013a,b) and for ringed seals by Vorkamp et al. (2011), from which details on the analytical methods can be obtained. Perfluorinated alkyl substances (PFAS) data from liver tissues were obtained from Bossi et al. (2005a, 2005b), Dietz et al. (2008) and Rigét et al. (2012) where analytical methods are likewise described in detail. Data on OHCs and PFOS up to 2017 were added using the same methods and labs.

#### 2.4 Chemical analysis for mercury

Mercury (Hg) analyses were performed using either direct combustion techniques (Leco AMA-254 and later Milestone DMA-80) or via acid digestion and subsequent detection using FIMS (PerkinElmer FIMS-100) or ICP-MS (Agilent 7500 and 7900). Details of these analyses are described elsewhere (Dietz et al. 2000; Søndergaard et al. 2015; Sonne et al., 2019). All Hg analyses were performed at the accredited trace element laboratory at Department of Bioscience, Aarhus University, Denmark.

#### 2.5 Chemical analysis for fatty acids

Polar bear and prey adipose fatty acid (FA) analysis proceeded as detailed elsewhere (Thiemann et al. 2008a; McKinney et al. 2013). To avoid potentially oxidized outer tissues we subsampled the innermost tissue of the available samples. Lipids were extracted twice using a chloroform:methanol:water mixture (8:4:3), v:v, containing butylated hydroxytoluene as antioxidant, at a volume:weight ratio of 20:1 solvent:sample. FAs were derivatized to FA methyl esters (FAMEs) using methanol containing 0.5 N H2SO4. FAME extracts (50 mg mL-1 total FAME) were analyzed in duplicate on a Varian CP-3800 gas chromatograph with flame ionization detection (GC-FID) using a DB-23 column (Agilent Technologies, Palo Alto, CA, USA). FAs were quantified as mass percent of total FAME. Quantitative FA signature analysis (QFASA), was used in this study which statistically compares predator FA signatures to those of potential prey to generate diet estimates, and and has been used to quantify subpopulation differences in diets of Greenland and Canadian arctic and subarctic polar bears from which the analythical details can be obtained (Iverson et al., 2004, 2006; Thiemann et al., 2008a; McKinney et al., 2013).

#### 2.6 Chemical analysis for stable isotopes

The composition of stable nitrogen and carbon isotopes was measured at the Center for Permafrost (University of Copenhagen, Denmark). Briefly, a representative homogenized subsample was wrapped into a tin combustion cup, and the ratios for stable carbon and nitrogen isotopes were measured by continuous flow using an elemental analyzer (CE 1110, Thermo Electron, Milan, Italy) coupled to a mass spectrometer (Finnigan MAT Delta PLUS, Thermo Scientific, Bremen, Germany). The stable isotope ratios were calculated against the international standards Vienna PeeDee Belemnite (vPDB) and atmospheric N2 (AIR) respectively and are conventionally expressed as  $\delta(\infty)$ . Reference samples, i.e. atropine, were included for the positive evaluation of analytical performance. The instrument was calibrated by employing pure gases of CO2 and N2 against the certified reference material of sucrose and (NH4)2SO4 provided by the International Atomic Energy Agency (IAEA, Vienna, Austria). The analytical precision was maintained at <0.1‰ SD.

### 2.7 Data analysis

Graphics and statistics used R version 2.4.1 and Microsoft Excel. Data on contaminant concentrations were not available for each species or year during the 35-year period spanning (1983-2017) available for the polar bears. Ringed seal data were available from some years between 1986 and 2017 whereas the data from harp seals, hooded seals, bearded seals, walruses and narwhals all originated from 2015 (this study). In order to estimate the polar bear exposure over time the temporal trend data for these species were estimated based on the temporal trends of the ringed seals and the relative concentration ratios from 2015.

## 3 Results and discussion

The temporal trends of contaminant loads in top predators such as the polar bear are a result of global emission sources and long-range transport to the Arctic (AMAP 2018; Riget et al. 2019). In addition, composition of prey occupying different trophic levels plays a role in the contaminant exposure as well. In the present study the food of the individual polar bears were determined from the FA composition in the individual bears over time and contaminant load of the main prey items was determined.

#### 3.1 Polar bear food composition over time

The overall food composition over time for all age and sex groups were calculated using a moving average over 7 years. This temporal trend analysis shows that the proportion of ringed seals was relatively stable around 50% and started to decline after 2000 (Fig. 1a). In year 2016, the last year with FA data, the ringed seal percentage had declined to less than 25%. The opposite trend was observed for the hooded seals, which were in the magnitude of 15% until 2000 after which they increased to more than 40% by 2016. Harp seals showed an increase from 15% to more than 25% from 1983 to 1994, after which their proportion in the diet remained relatively stable until 2016. No clear trends were observed for the proportions of bearded seals, walruses and narwhals. Bearded seal was seldom consumed (4.9%) and narwhal (1.0%) and walrus (0.05%) were rarely, if ever, consumed. Actual averages per year are shown for all species and for all bears in Appendix 1, and sex and age details of the three most important seal species are shown in Appendix 2. Even for the averages among all groups the numbers may vary considerably between years, with e.g. ringed seal constituting as much as 89.7% in 1984 and as little as 13.0% in 2015.

The pattern of juvenile bears showed more or less the same pattern as the overall one. This could be expected, since juvenile bears represent approximately 51% of all of the bear samples whereas adult females represented 18% and adult males 31%, respectively (Fig. 1b). The adult females likewise showed a similar pattern but with a somehow lower amount of ringed seals in the mid-1990s (Fig. 1c). On the other hand, the adult males showed a rather different pattern as the proportion of ringed seals in males never exceeded 40% (Fig. 1d). In addition, harp seals more or less followed the same pattern as the ringed seals for the adult male polar bears, with a rather constant proportion around 40% up until year 2000, after which a rather similar decline was observed. The proportion of hooded seals for all groups was around or below 20% up until year 2000, after which a dramatic increase was observed in some cases reaching more than 40% of the diet. Some of the adult males actually had as much as 90-100% hooded seals in the latest analysed years (2011-2015; Appendix 1 & 2).



Figure 1. Temporal trends of the food composition (7-year moving average) for a) all, b) juvenile, c) adult female, and d) adult male EG polar bears from 1983 to 2016 based on QFASA.

Overall means between the three age/sex groups is shown in Fig. 2. This information confirms the somehow larger proportion of hooded seals in the adult males and the corresponding lower proportion of ringed seals. In addition, the low proportion of narwhal and walruses are obvious but few individuals may have quite high proportions with up to 50% of narwhals and 25% of walrus (Appendix 2).



**Figure 2**. Box plots on the overall proportions of preferred food intake a) bearded seal, b) harp seal, c) hooded seal, d) ringed seal, e) narwhal and f) walrus in the juvenile, adult female and adult male EG polar bears. Boxes contain the median and represent 25% and 75% quantiles while whiskers show 5 and 95% confidence intervals.

#### 3.2 OHC concentrations in polar bears over time

Temporal trends of contaminant loads in EG polar bears has been reported in a number of studies (e.g. Dietz et al. 2013a,b, 2018; McKinney et al. 2013; Rigét et al. 2016, 2018). The trends for juvenile, adult females and adult males are shown in Figs. 3-5. Temporal trend statistics were recently updated by Rigét et al. (2018), and results for the polar bears and ringed seals are provided in Table 1.

Significantly decreasing  $\sum$  PCB concentrations have been found in ringed seals from Ittoqqortoormiit. The young polar bears show a general decrease over the period, but this decrease has a significant non-linear component.  $\Sigma PCB$ concentrations in polar bears over the past three years have been low after the concentration had been consistently increasing for a number of years. For the adult polar bears, no significant decreases have been observed. As seen from Figs. 3-5 and Rigét et al. (2018) for especially young ringed seals and partly young polar bears from Ittoqqortoormiit, the decrease in concentration had particularly occurred before 1995 to then remain fairly constant but at a relatively high level. Generally, the PCB congeners exhibit the same time trend as the sum of PCBs, however, with a tendency of a greater decrease for the groups with many chlorine atoms than for the groups with fewer chlorine atoms. Rigét et al. (2013) analyzed the time trend in ringed seals from Qegertarsuag with a statistical model that included climatic variables alongside biological variables. The conclusion was that the climate seemed to affect the time trend of POPs in ringed seals, but that the time trend corrected for the climate variables was generally in line with the above. The conclusion is that  $\sum$  PCB generally show a decreasing trend in the marine environment and that this decrease had mainly occurred in the period before 1995, after which the concentration has remained constant at a relatively high level. The increasing trend in polar bears in mid-2000, which may be due to the influence of climate change on polar bear behaviour in the form of changing food habits and migration patterns (see McKinney et al. 2013), has now been replaced by low values since 2014. All time series, except for adult polar bears males show a declining trend in dichlorodiphenyltrichloroethane ( $\Sigma$ DDT) concentrations and the trend in young polar bears can be described as a significant log-linear (exponential) decline. For the adult polar bears, there is often a large individual variation in the concentrations and this, together with the fact that the number of adults in the individual years is often small, makes the statistical power to detect a trend small (Riget et al. 2018). The annual change in  $\Sigma$ DDT concentration is calculated to be between -1.4% to -9.0%. The conclusion is that  $\Sigma$ DDT shows a significantly decreasing trend and the average annual change of  $\Sigma$ DDT is comparable to that of  $\Sigma$ PCB.

All time series show a strong significantly decreasing trend in hexachlorohexane ( $\Sigma$ HCH) concentrations. The annual change is calculated to be between -1.4 to -10.3% for the significant time series. For the polar bear time series, the non-linear trend component is also significant. This is because, after a general decrease up to 2006, concentrations increased again. The annual decrease in  $\Sigma$ HCH concentrations is generally greater than is the case with  $\Sigma$ PCBs and  $\Sigma$ DDT. The hexachlorobenzene ( $\Sigma$ HCB) concentration has significantly decreased in young ringed seals. In polar bears, the  $\Sigma$ HCB concentration shows a rolling trend throughout the period and with a significant non-linear trend component. In polar bears, the concentration seemed to be increasing in recent years, but for the young polar bears a clear decrease occurred from 2013 to today. The conclusion is that the  $\Sigma$ HCB concentration shows a decreasing trend in most time series, but it is not as consistent and marked as in the case of  $\Sigma$ PCBs,  $\Sigma$ DDTs and  $\Sigma$ HCHs.

The chlordane ( $\Sigma$ CHL) concentrations are significantly decreasing for ringed seals but not for the three polar bear time series. The trend of dieldrin concentration in polar bears shows both a significant log-linear component and a significant non-linear component in the young and adult female polar bears. In males, only the non-linear component is significant. Dieldrin has clearly increased in recent years, though with a decrease over the past three years, most notably for the young polar bears (Figs. 3-5, Table 1). Octachlorostyrene (OCS) concentration shows a marked increase in recent years, especially in the young and old male polar bears, resulting in the significant non-linear component (Figs. 3-5, Table 1). However, in 2014 to 2016, the concentration has fallen again.

Toxaphene exhibits a significant exponential decline in all time series of the seals, but toxaphene was not measured in the polar bears. The conclusion is that toxaphene, like many of the other substance groups, shows a decreasing concentration. Vorkamp et al. (2015) gives a detailed description of toxaphene in the Greenlandic marine environment. Young polar bears show an unshaped trend with an increase in the concentration in Heptachlor epoxide (HEPO) in recent years, however, with a marked decrease since 2014 and 2015, similar to several other POPs. In adult females, the concentration decreases significantly. The concentrations of mirex and photomirex have generally been at a low level in the East Greenlandic polar bears, but especially a few high values after 2010 are the cause of the significant non-linear trend components found for photomirex in the young polar bears.



Figure 3. Temporal trends of major contaminants in juvenile EG polar bears from 1983 to 2017.



Figure 4. Temporal trends of major contaminants in adult female EG polar bears from 1984 to 2016.



Figure 5. Temporal trends of major contaminants in adult male EG polar bears from 1989 to 2016.

 Table 1. Temporal trends of major contaminants in EG polar bears and ringed seals between 1983 and 2013 (extracted from Rigét et al. 2018).

Contaminant	Species	Region	Period	Years with data	Average yearly change	p for log- lineær trend	p for non log- lineær trend	The power of the time series to detect a 5% yearly change	Number of years needed to detect a 5% yearly change with 80% power
ΣΡCB	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	14	-4.40%	<0.01**	1		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1994-2016	13	-1.90%	0.11	0.71	0.71	14
	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-2.40%	<0.01**	0.03*		
	Polar bear, ad F, adipose	Ittoqqortoormiit	1984-2016	23	-2.10%	0.03*	0.57		
	Polar bear, ad M,adipose	Ittoqqortoormiit	1989-2016	26	-0.30%	0.69	0.11	0.99	17
Trichlorophenyl	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	14	-2.10%	<0.01**	0.67		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1994-2016	13	-0.30%	0.76	0.1	0.89	12
Tetrachlorophenyl	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	13	-5.40%	<0.01**	0.34		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1999-2016	13	-2.20%	<0.01*	0.13		
	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-5.20%	<0.01**	0.01*		
	Polar bear, ad F, adipose	Ittoqqortoormiit	1984-2016	23	-7.00%	<0.01**	0.09		
	Polar bear, ad M,adipose	Ittoqqortoormiit	1989-2016	22	-2.30%	<0.01**	0.03*		
Pentachlorophenyl	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	14	-4.20%	<0.01**	0.67		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1994-2016	13	-1.80%	0.06	0.99	0.88	12
	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-2.90%	<0.01**	<0.01**		
	Polar bear, ad F,adipose	Ittoqqortoormiit	1984-2016	23	-3.50%	<0.01**	0.31		
	Polar bear, ad M,adipose	Ittoqqortoormiit	1989-2016	16	-1.00%	0.14	<0.01**		
Hexachlorophenyl	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	14	-4.30%	<0.01**	0.97		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1994-2016	13	-1.30%	0.26	0.31	0.68	14
	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-2.00%	<0.01**	0.01**		
	Polar bear, ad F,adipose	Ittoqqortoormiit	1984-2016	23	-2.20%	<0.01**	0.54		
	Polar bear, ad M,adipose	Ittoqqortoormiit	1989-2016	22	<0.01%	0.99	0.01*		
Heptachlorophenyl	Ringed seal blubber ≤4 year	Ittoqqortoormiit	1986-2016	14	-5.20%	<0.01**	1		
	Ringed seal blubber >4 year	Ittoqqortoormiit	1994-2016	13	-1.40%	0.43	0.68	0.36	18
	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-2.70%	<0.01**	0.07		
	Polar bear, ad F, adipose	Ittoqqortoormiit	1984-2016	23	-2.10%	0.09	0.38	78	>20
	Polar bear, ad M, adipose	Ittoqqortoormiit	1989-2016	22	-1.20%	0.24	0.12	0.95	19
Octachlorophenyl	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-1.60%	0.03*	0.04*		
	Polar bear, ad F,adipose	Ittoqqortoormiit	1984-2016	23	-0.60%	0.68	0.65	0.61	>20
	Polar bear, ad M, adipose	Ittoqqortoormiit	1989-2016	22	1.50%	0.17	0.04*		
Nonachlorophenyl	Polar bear, juv, adipose	Ittoqqortoormiit	1983-2016	29	-0.60%	0.38	<0.01**		
	Polar bear, ad F,adipose	Ittoqqortoormiit	1984-2016	23	0.50%	0.77	0.31	0.44	>20
	Polar bear, ad M,adipose	Ittoqqortoormiit	1989-2015	21	1.30%	0.39	0.43	0.63	>20

#### 3.3 PFAS concentrations in juvenile polar bears over time

Only juvenile polar bears have been analysed for PFAS by Rigét et al. 2019. The time analyzes of the compounds PFOSA and PFOA in polar bears have been made by a regression analysis for so-called censored data (data that cannot have values below the detection limit), since half or several of the annual median values are below the detection limit (marked in red in Table 2). In these cases, it is not possible to test for a non-linear trend component.

For the polar bears, there has been a marked increase from 2015 to 2016. The coming years will show whether this increase continues. In polar bears, the compounds PFOA show a significant increase in the period from 1984 to 2016, with an average annual increase of 2.0%, while PFOSA shows a significantly decreasing trend. The compounds PFOS, PFHxS, PFNA, PFDA, PFUnA, PFDoA and PFTrA also show a general increase over the period but also a significant non-linear trend component. Several of these connections peaked markedly around 2006 and have subsequently decreased significantly (see Fig. 6).

Table 2. Temporal trend data of PFAS for EG polar bears and ringed seal (extracted from Rigét et al. 2018).

Contaminant	Species	Region	Period	Years with data	Average yearly change	p for log- lineær trend	p for non log- lineær trend
PFOS	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	0.70%	0.13	<0.01**
PFHxS	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	7.90%	<0.01**	<0.01**
PFOSA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	-5.70%	<0.01**	
PFOA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	2.30%	<0.01**	
PFNA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	5.70%	<0.01**	0.09
PFDA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	3.60%	<0.01**	<0.01**
PFUnA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	4.20%	<0.01**	0.01*
PFDoA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	2.60%	<0.01**	<0.01**
PFTrA	Polar bear, juv, liver	Ittoqqortoormiit	1984-2016	27	3.80%	<0.01**	0.03*
PFOS	Ringed seal, juv, liver	Ittoqqortoormiit	1986-2016	10	4.20%	0.02*	<0.01**
PFNA	Ringed seal, juv, liver	Ittoqqortoormiit	1986-2016	10	9.00%	<0.01**	
PFDA	Ringed seal, juv, liver	Ittoqqortoormiit	1986-2016	10	9.40%	0.11	1
PFUnA	Ringed seal, juv, liver	Ittoqqortoormiit	1986-2016	10	5.90%	<0.01**	0.02*

\*Substances marked in red are components where half or more of the yearly median values were under the detection limit.



Figure 6. Temporal trends PFAS concentrations in EG juvenile polar bears from 1984 to 2017.

In ringed seals, PFOA and PFOSA concentrations have been above the detection limit in recent years and therefore suggest increasing trends. PFNA and PFDA have also been increasing, but in recent years they have become either stable or shown a weak decline. The PFUnA has been increasing up to around 2010, but now seems to be declining. For PFOS the non-linear trend component is significant. In both areas, the concentration increased until 2006, after which the concentration dropped significantly. Since 2006, the PFOS concentration has shown a markedly decreasing trend in both polar bears and ringed seals series (Riget et al. 2019; Fig. 6). This thus indicates that the PFOS concentration has peaked as a result of the international regulation. A more detailed discussion of these time series can be found in Rigét et al. (2013).

#### 3.4 Mercury concentrations in polar bears over time

As seen from Fig. 7, the liver (Hg) concentrations varied considerably from year to year. Concentrations in juveniles, adult females and adult males seem to covary in the periods 1993, 1995-1996 and 2013, indicating that some ecologic conditions such as species and food availability may influence hepatic mercury concentrations. For some years there seems to be a linkage with the high Hg exposures and low ringed seal/high hooded seal diet (e.g. 1993), whereas 2013 only had a high percentage of hooded seals for adult females (Fig. 8; Table 3). Especially the adult females have some very high peaks being more than 4-6 fold above the averages in the adjacent years. It is also worth mentioning that observed peaks of OHCs of all three age/sex groups of polar bears likewise occurred in 1993 and in 2013 when both juvenile and adult males had high concentrations whereas females peaked one year earlier, in 2012.

As seen from Table 3 below only the juvenile bears revealed significant time trends (p<0.01; both log-linear and linear) with an increase of 2.1% per year (Rigét et al. 2018). Increases were likewise found in polar bear (>2 years of age) hair and in sculpin liver (Table 3).



Figure 7. Temporal trends of mercury in EG juvenile, adult female and adult male polar bears between 1983 to 2016.

Table 3. Temporal trend Hg data for EG polar bears and ringed (extracted from Rigét et al. 2018).

Species	Region	Period	Years with data	Average yearly change	p for log- e lineær trend	p for non log- lineær trend	The power of the time series to detect a 5% yearly change	Number of years needed to detect a 5% yearly change with 80% power
Sculpin, liver	Ittoqqortoormiit	1995-2015	10	2.10%	<0.01**	0.03*		
Ringed seal, liver, ≤4 years	Ittoqqortoormiit	1986-2016	13	3.20%	0.09	0.32	0.24	>20
Ringsæl, liver, >4 year	Ittoqqortoormiit	1986-2016	13	0.30%	0.78	0.15	0.48	17
Polar bear, liver, juv	Ittoqqortoormiit	1983-2016	25	2.10%	<0.01**	<0.01**		
Polar bear, liver, ad	Ittoqqortoormiit	1984-2016	23	0.80%	0.56	0.07	0.67	>20
Polar bear, hair, >2 year	Ittoqqortoormiit	1973-2017	24	0.40%	0.13	0.02*		



Figure 8. Temporal trends of the proportion of hooded seal as food for EG juvenile, adult female and adult male polar bears between 1983 to 2016.

#### 3.5 Correlations between the analysed contaminants

To avoid repetitive correlations of the polar bears and their exposure to the many analysed contaminants correlation matrices were calculated for a number of OHCs in EG polar bears (Fig. 9; Table 4). From the Figure and Table it is evident that  $\sum$ PCBs is significantly correlated with eight out of 11 other OHC groups ( $\sum$ DDT,  $\sum$ HCH, PECB,  $\sum$ OCS, Dieldrin,  $\sum$ CHL, HEPO and Mirex) and only  $\sum$ HCB, the brominated flame retardants hexabromocyclododecane (HBCDD) and polybrominated diphenyl ethers (PBDE) as well as PFOS did not correlate with  $\sum$ PCB. PBDEs and PFOS, showing an insignificant negative

relation with  $\sum$ PCBs, were not expected to be positively correlated with  $\sum$ PCBs and most other contaminants, as these substances have shown emission related increases up until 2004-2006 (Dietz et al. 2013a,b; Rigét et al. 2018) and PFOS is less fat-soluble. It is hence not necessary to repeat the graphics and Tables for all the OHCs especially not the ones that co-vary with  $\sum$ PCBs or other OHCs.



**Figure 9**. Correlation matrix for selected OHCs in EG polar bears. The colours indicate whether there is positive (red) or a negative (blue) correlation. The size of the circles indicates the magnitude of the correlation coefficient and crosses indicate insignificant correlations at the 5% significance level.

	PCB	DDT	HCH	НСВ	PECB	OCS	DIEL	CHL	HEPO	MIREX	HBCDD	PBDE	PFOS	HG
РСВ	1.000	0.418	0.703	0.292	0.789	0.660	0.797	0.843	0.802	0.497	-0.205	-0.191	-0.247	0.162
DDT	0.022	1.000	0.708	-0.195	0.500	0.297	0.601	0.622	0.657	-0.258	-0.610	-0.229	-0.317	-0.334
нсн	0.000	0.000	1.000	-0.036	0.897	0.545	0.765	0.817	0.839	-0.049	-0.570	-0.490	-0.646	-0.140
НСВ	0.117	0.303	0.852	1.000	0.205	0.438	0.398	0.042	0.116	0.595	0.664	0.397	-0.148	0.149
PECB	0.000	0.005	0.000	0.276	1.000	0.689	0.763	0.806	0.826	0.206	-0.291	-0.336	-0.638	0.089
ocs	0.000	0.111	0.002	0.015	0.000	1.000	0.661	0.612	0.605	0.435	0.209	0.202	-0.367	0.246
DIEL	0.000	0.000	0.000	0.029	0.000	0.000	1.000	0.853	0.875	0.245	-0.220	-0.126	-0.401	-0.160
CHL	0.000	0.002	0.000	0.854	0.000	0.002	0.000	1.000	0.980	0.080	-0.413	-0.406	-0.350	-0.023
HEPO	0.000	0.000	0.000	0.543	0.000	0.000	0.000	0.000	1.000	0.047	-0.387	-0.271	-0.439	-0.155
MIREX	0.005	0.169	0.797	0.001	0.276	0.016	0.191	0.724	0.805	1.000	0.428	0.348	0.057	0.526
HBCDD	0.326	0.001	0.003	0.000	0.158	0.316	0.291	0.070	0.056	0.033	1.000	0.777	0.172	0.193
PBDE	0.312	0.223	0.006	0.030	0.070	0.284	0.508	0.061	0.147	0.060	0.000	1.000	0.202	0.021
PFOS	0.214	0.107	0.000	0.460	0.000	0.060	0.038	0.119	0.022	0.778	0.444	0.313	1.000	-0.163
HG	0.419	0.088	0.486	0.459	0.658	0.216	0.425	0.923	0.441	0.005	0.388	0.919	0.435	1.000
P values	ns	p>0.05	*	P<0.05	**	p<0.01	***	p<0.001						

Table 4. Correlation matrix (upper right half) and p values (lower left half) for a number of OHCs in EG polar bears...

#### 3.6 Food preferences and distribution

The differences in contaminant concentration of the different polar bear food species (see section 3.7 below) are most likely a function of the preferred food of the different seal species as well geographic range and for narwhal their metabolic capabilities (see section 3.6).

#### 3.6.1 Ringed seals

The ringed seal is a tru Arctic ice-associated pinniped with a circumpolar distribution (Burns 1970, Smith 1987, Wiig et al. 1999, Lydersen et al. 2004, Laidre et al. 2008). Data on East Greenland ringed seal diet has been reported from 30 animals in Kong Oscars Fjord back in 1985 (Siegstad et al. 1998). The investigations documented that Arctic cod (Arctogadus glacialis; 60.0%) and polar cod (Boreogadus saida; 5.0%) were important species. However, also other Cottidae species (22.4%) were found together with *Themisto spp.* (4.2%). A later study by Labansen et al. (2010) was conducted on ringed seals samples in 2002-2004. This study revealed Themisto libellula as the most important species (42.2%), polar cod (34.0%) as the second most important species followed by Cottidae species (3,9%), Liparis spp. (2.9%), Stiachiadae (2.0%), Arctic cod (1.4%), and other unidentified fish (5.2%) were also important species. Limited tagging information is available on ringed seals from the east coast of Greenland. However, Kapel et al. (1998) reported on five ringed seals tagged in Kong Oscars Fjord with conventional rototags in 1985. Two of these tags were retrieved from seals shot at the entrance of Ittoggortoormiit in February 1988 and March 1991 indicating some movements between the East Greenland fjords. In connection with Environmental Impact Assessment work conducted in East Greenland, 20 satellite tags were deployed on ringed seals between Trail Island and the northern part of Dove Bay (Rosing-Asvid and Dietz 2018, Hamilton et al. 2021). The 20 tagged seals could be divided into one group that was resident, staying in or near the tagging area as long as they transmitted data and a group that swam southward, and spent the winter far from where they were tagged. Two adults (a male and a female), eight juvenile and two young of the year (YOY) stayed in the sheltered area west of Store Koldewey as long as their tags worked. These seals were all tagged in Fanger Sund, except one adult and one YOY that were tagged in Mønsted Hus. One seal tagged in Fanger Sund together with three seals from Mønsted Hus and the two from Mount Norris Fjord swam southward far from the tagging area (one all the way to Newfoundland - about 3,500 km from where it was tagged). The two seals tagged at Lille Koldewey both seemed to be associated with their tagging area (east of Store Koldewey), one of them, however, made long strays both north, south and east out on the shelf. The tagging location seems to have a strong influence on the likelihood of being resident or migratory (Rosing-Asvid and Dietz 2018, Hamilton et al. 2021).

#### 3.6.2 Harp seals

The harp seals have a similar range as the hooded seal within the North Atlantic where their high-density whelping patches are located along the pack ice off the the Greenland Sea (Wiig and Lie 1984, Laidre et al. 2008). Main diet of harp seals along the East Greenland coastline is unknown. The food along the west coast of Greenland up to Upernavik consists mainly of capelin (*Mallotus villosus*), supplemented with krill and various small fish. North of Upernavik, harp seals mostly eat polar cod and the amphipod Themisto libellula. On the fishing banks in the open water areas, the relatively few collected stomachs of harp seal suggest that the sand lance (*Ammodytes spp.*) found there are a very important source of food.

#### 3.6.3 Bearded seals

Bearded seals are like the ringed seal widely distributed throughout the circumpolar Arctic, but mainly over the relatively shallower waters of the continental shelf often in association with polynyas, shore leads and moving pack ice. The main diet of the bearded seal consist of benthic food like bristle worms, snails, crabs, sea cucumbers and sea anemones are of importance (Lowry et al. 1980, Laidre et al. 2008). However, in addition capelin, sand lance, cottidae and various flatfish. However, also bottom dwelling species such as bristle worms, snails, crabs, sea cucumbers and sea anemones are of importance.

#### 3.6.4 Hooded seals

The hooded seals have a similar range as the harp seal within the North Atlantic where their high-density whelping patches are located along the pack ice off the the Greenland Sea, as well as off the coast of Newfoundland, the Gulf of St. Lawrence and the Davis Strait (Wiig and Lie 1984, Laidre et al. 2008). The hooded seal consumes a large part of its food in the areas of open water and precisely what it eats is not known. However, in the coastal areas, hooded seal often eats larger fish including Greenland halibut (*Reinhardtius hippoglossoides*), deepwater redfish (*Sebastes marinus*) and Atlantic cod (*Gadus morrhua*), as well as squids. Studies of hooded seals' FA composition show that Atlantic argentine (*Argentina silus*), a large shoal fish living in deep water in most of the hooded seals distribution area, is also an important food item (Rosing-Asvid 2010). Hooded seals breeding off the Scoresby Sound Area spend the winter in the North Atlantic and may range close to the Norwegian coast and as far south as the Shetland Island closer to the emissions from the temperate European contaminant sources (Vacquie-Garcia et al. 2017)

#### 3.6.5 Narwhals

Narwhals live in the dense pack ice during the winter follows the ice edge during spring and summers in ice free waters (Dietz et al. 2008). According to Heide-Jørgensen et al. (1994) narwhals prey primarily on Arctic cod (64%) followed by polar cod (15%), unidentified gadids (19%) and invertebrates (2%). However, these percentages may vary dependant on where the samples are obtained (e.g. ice edge, ice cracks or open water; Finley & Gibb 1982). During winter for example, narwhals feed on Greenland halibut (Heide Jørgensen & Laidre 2006). The narwhals around Scoresby Sound use their summers within the inner fjords of the sound. However, during winter when the fjord freezes up the narwhals migrate to the fjord and spend the winter in the pack ice offshore on the latitude of the Blosseville Coast (Heide Jørgensen et al. 2015).

#### 3.7 Metabolic capabilities

Ancient convergent losses of the Paraoxonase 1 gene yield potential risks for modern marine mammals as these are particularly vulnerable to adverse health effects from organophosphorus pesticide pollution because of a functional loss of the primary mammalian metabolic defense mechanism (Meyer et al. 2018). This finding is just one example of an evolutionary deficiency that puts marine mammals at increased risk for modern-day pollution. Toothed whales, which originated in the mid-Eocene from herbivorous artiodactyls (cloven-hooved land mammal), also show a reduced metabolic ability to eliminate persistent environmental pollutants such as biomagnifiable  $\sum PCBs$  compared with carnivorous predators such as polar bears, seals, walruses, and humans. Because they lack the ability to filter these chemicals, extreme concentrations of  $\sum$ PCBs and mercury have been found in high trophic-feeding cetaceans, including killer whales (*Orcinus orca*). Moreover, toothed whales lack the keratinous pollutant sequestration routes, such as hair, that relieve carnivorous marine mammals from their contaminant burden including mercury. Given the vulnerability of marine mammals, global regulation and remediation of harmful marine pollutants, including organophosphorus compounds and  $\sum$ PCBs, should be urgently prioritized by regulatory bodies, such as the United Nations Environment Programme, the Stockholm Convention, and the U.S. Environmental Protection Agency. Failing to protect these cetaceans could lead to pollution-mediated population collapses and an irreversible loss of biodiversity and ecosystem services.

## 3.8 Contaminant loads in ringed seals relative to other polar bear prey species

#### 3.8.1 OHCs

Samples from the EG polar bear prey species were analysed for contaminant concentrations at Aarhus University and at Environment and Climate Change Canada, Carleton University, Canada.  $\Sigma$ PCB is the dominating contaminant in polar bears as well as in the food items of the polar bears, which is true and even more expressed for the toxic effects of these contaminants (Dietz et al. 2015, 2018a,b). According to Dietz et al. (2015, 2018a,b), SPCBs constitute the major risk for effects on the reproduction, the immune system as well as carcinogenic effects in polar bears. **SPCB** loads in the formerly dominating food item, the ringed seal, was lower than in the other potential food items of the polar bear from the Scoresby Sound area collected in 2015, including harp seals (1.6 fold), bearded seals (1.8 fold), hooded seals (7.2 fold) and narwhals (11.2 fold; Fig. 10; Table 5 for  $\Sigma$  PCBs only). The higher concentrations in these species are linked to what these seal and whale species consume, but also to the seasonal distribution and the metabolic capabilities of cetaceans vs pinnipeds. A similar pattern was observed for the other contaminant groups including  $\Sigma$ DDT  $\Sigma$ CHL,  $\Sigma$ HCH and PBDE (Fig. 11).



Figure 10. Mean ∑PCB concentrations in EG polar bear food items.

Table 5. ∑PCB concentrations (ng/g l.w) of EG polar bear food items.

Species	Mean	SD	Median	Min	Max	Reference
Ringed seal adult 2014	293	111	264	160	563	Present study
Ringed seal juv 2014	513	115	534	360	654	Present study
Ringed seal average 2014	403	113	399	160	654	Present study
Bearded seal 2015	653	523	564	107	1,652	Present study
Harp seal 2015	732	1,313	315	139	4,440	Present study
Hooded seal juvenile 2015	1,935	1,015	1,556	937	3,269	Present study
Hooded seal adult female 2008	3,840	1,220	3,740	1,270	5,830	Villanger et al. 2013
Hooded seal average	2,888	1,015	2,648	937	5,830	Present study; Villanger et al. 2013
Narwhal 2015	4,507	4,151	3,734	382	19,035	Present study



Figure 11. Median concentrations of  $\Sigma$ PCB,  $\Sigma$ DDT  $\Sigma$ CHL,  $\Sigma$ HCH and PBDE concentrations (ng/g l.w) in EG polar bear food items.

#### 3.8.2 PFASs

To reduce the number of PFAS comparisons we merged the species comparisons into  $\Sigma$ 5PFSAs (sum of perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluoroheptane sulfonate (PFHpS), perfluorooctane sulfonate (PFOS) and perfluorodecane sulfonate (PFDS)), and  $\Sigma$ 6PFCAs (sum perfluorononanoate (PFNA), perfluorodecanoate (PFDA), of perfluoroundecanoate (PFUnA), perfluorododecanoate (PFDoA), perfluorotridecanoate (PFTrA), and perfluorotetradecanoate (PFTeA)). A bit surprisingly, the ringed seals had higher PFSA (1.9-3.2 fold) and PFCA (1.1-2.8 fold) concentrations in their livers compared to the bearded, harp and hooded seals as well as the narwhals (Fig. 12; Table 6). As the difference between the ringed seals and the alternative species were opposite to the OHCs and mercury a decline in the years for the PFAS should be expected, where high concentrations were found OHCs and Hg namely in 1993 and in 2013. Such declines were observed in 2013 for PFNA, PFUnA and PFHxS whereas a year delay in such declines were observed for PFOS, PFTrA, PFDoA and PFOSA, which could indicate a delayed metabolism and longer half time of these substances.



Figure 12. Median and ranges of PFAS concentrations in EG polar bear food items.

PFAS, Species	n	Mean	SD	Median	Min	Max	Reference
∑5PFSAs							
Ringed seal 2014	20	51.8	13.5	53.1	22.4	85.2	AMAP Core Programme
Bearded seal 2015	7	20.8	5.2	22.7	13.5	27.0	Present study
Harp seal 2015	11.0	27.7	11.3	25.6	17.2	49.1	Present study
Hooded seal 2015	5.0	16.4	6.9	14.3	8.4	26.9	Present study
Narwhal 2015	24.0	20.8	20.9	13.0	4.4	75.7	Present study
∑6PFCAs							
Ringed seal 2014	20	31.0	7.7	31.6	16.1	48.5	AMAP Core Programme
Bearded seal 2015	7	11.0	3.3	10.3	7.1	16.6	Present study
Harp seal 2015	11.0	20.5	6.1	20.6	11.4	30.5	Present study
Hooded seal 2015	5.0	21.7	15.8	16.8	10.3	49.3	Present study
Narwhal 2015	24.0	27.4	22.0	18.3	10.2	89.2	Present study

Table 6. PFAS concentrations in livers of EG polar bear food items (ng/g w.w.)..

#### 3.8.3 Hg

As for the Hg loads of the possible prey items of the EG polar bears the patterns were different than for the OHCs. Ringed seals were higher than both bearded seals (2.1 fold) and harp seals (3.6 fold; Fig. 13; Table 7). However, the hooded seals (5.4 fold) were considerably higher than the ringed seals and the narwhals (1.6 fold) were slightly higher than the ringed seals. The years with 4-8 times higher Hg loads in the polar bear livers (see section 3.3) are likely to be linked to higher consumptions of hooded seals and narwhals (Fig. 13; Table 7).



Figure 13. Median and ranges of Hg concentrations in EG polar bear food items.

Table. 7. Hg concentrations in EG polar bear food items (ng/g l.w.).

Species	n	Mean	SD	Median	Min	Max	Reference
Ringed seal 2014	20	20.0	9.1	18.7	6.6	37.5	AMAP Core Programme
Bearded seal 2015	7	9.3	8.5	6.3	3.7	28.1	Present study
Harp seal 2015	11.0	5.5	8.3	3.1	0.7	30.2	Present study
Hooded seal 2015	5.0	108.5	116.0	72.8	34.5	313.8	Present study
Narwhal 2015	24.0	31.9	29.9	28.2	1.2	120.9	Present study

#### 3.9 Modelling polar bear intake relative to observed contaminant loads

We have modelled the  $\sum$ PCB intake from the proportion of the average food items estimated from the fatty acids from the juvenile polar bears. More scenarios could have been presented but with 10 POPs, 10 PFCs and 12 metals within three age groups, a total of 72 plots and 72 Tables could have been presented, which was considered beyond the scope of this report. A quite clear relationship was observed between the calculated average  $\sum$ PCB intake and the actually measured  $\sum$ PCB loads in the adipose of the juvenile polar bears (Fig. 14). The extremely high body burden of the juvenile polar bears from 2013 could however, not be explained by the average species composition and their respective average concentrations alone. One variable that cannot be readily explained with the used method is the age and sex composition of the observed species composition of the food items. If a larger proportion of adult male seals or whales had been consumed in 2013 then the exposure would be much higher than if only juvenile or yearling individuals had been eaten.



Figure 14. Polar bear intake of juvenile polar bears relative to juvenile polar bear adipose concentrations of ∑PCBs (ng g-1 lw).

In order to test the effects of the variable prey selection on the  $\sum$ PCB exposure correlations between the calculated  $\sum$ PCB intake from the consumed prey and the analysed adipose  $\sum$ PCB loads concentrations were conducted for the individual bears groups over time as well as all the bears (Table 8; Fig. 15). As seen from the Table both juveniles, adult females, adult male and all the bears showed a significant correlations indicating a strong linkage between the climate related changes in the food intake and the actual exposure.

**Table. 8.** Correlations between the modelled intakes of  $\sum PCB$  based on the on QFASA profiles and the analysed adipose  $\sum PCB$  concentrations in the monitored EG polar bear between 1983 and 2016 (ng/g l.w.).

Group	DF	Corr. Coef.: r	P value	Significance level
Juvenile polar bears	166	0.575	4.66615E-16	***
Adult female polar bears	60	0.498	4.41778E-05	***
Adult male polar bears	106	0.196	0.04319	*
All	334	0.436	5.61554E-17	***



Estimated intake ∑PCB ng/g l.w.

**Figure 15.** Correlations plot and regression lines between the modelled  $\sum$ PCB intakes based on the on QFASA profiles and the analysed adipose  $\sum$ PCB concentrations in the monitored EG polar bear between 1983 and 2016 (ng/g l.w.) For correlations and significance levels see Table 8.

## 3.10 Polar bear contaminant loads relative to emissions and climate parameters

Both emissions and climate change are determining the contaminant load in the polar bears. So far, both parameters have not been modelled for Arctic species such as ringed seals and polar bears, which would involve complicated multivariate analyses. McKinney et al. (2013) previously documented that increased hooded seal and decreased ringed seal consumption occurred during years when the North Atlantic Oscillation index (NAO) was lower. Thus, periods with warmer temperatures and less sea ice were associated with more subarctic (hooded seals and harp seals) and less arctic seal species (ringed seals) consumption. McKinney et al. (2013) likewise concluded that diet change resulted in consistently slower temporal declines in adipose levels of legacy persistent organic pollutants of polar bears, as the subarctic seals have higher contaminant burdens than arctic seals.

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# Appendices to UNEXPECTED AU Technical report, August 2021

Unexpected Increases of Persistent Organic Pollutant and Mercury levels in East Greenland Polar Bears (UNEXPECTED)

Report to DANCEA

August 2021

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Year	Ringed seal	Harp seal	Hooded seal	Bearded seal	Narwhal	Walrus
1983	32.0	. 0.0	17.9	50.1	0.0	0.0
1984	89.7	7.8	0.0	2.5	0.0	0.0
1986	39.4	30.6	26.0	4.1	0.0	0.0
1987	47.9	26.3	22.9	2.3	0.6	0.0
1988	50.8	38.8	8.5	1.9	0.0	0.0
1989	76.3	16.5	4.2	3.0	0.0	0.0
1990	44.3	27.5	18.9	7.2	2.1	0.1
1991	77.5	14.8	0.0	7.7	0.0	0.0
1992	45.3	33.8	15.0	5.2	0.6	0.1
1993	27.4	36.9	30.6	3.3	1.8	0.0
1994	60.8	18.2	14.1	6.8	0.0	0.1
1995	15.9	43.9	30.2	8.4	1.7	0.0
1996	48.4	33.2	15.1	3.1	0.0	0.1
1999	56.8	28.2	9.3	5.7	0.0	0.0
2000	56.1	22.1	16.1	5.6	0.0	0.1
2001	47.0	29.4	18.9	4.6	0.0	0.0
2003	64.6	13.2	11.9	10.3	0.0	0.0
2004	58.4	30.0	7.3	4.2	0.0	0.1
2006	34.0	23.3	38.7	3.5	0.5	0.0
2007	35.5	27.3	30.9	6.3	0.0	0.0
2008	45.0	23.0	24.8	2.8	4.3	0.0
2009	29.2	30.9	34.0	3.1	2.8	0.0
2010	43.0	25.4	26.2	5.4	0.0	0.0
2011	33.2	32.2	31.2	3.3	0.0	0.2
2012	46.7	17.6	31.5	0.7	3.5	0.0
2013	19.9	32.5	34.3	2.7	10.6	0.0
2014	33.1	33.4	31.1	2.2	0.0	0.2
2015	13.0	30.2	52.3	4.5	0.0	0.1
Averag	44.9	27.3	21.8	4.9	1.0	0.05
SD	18.19	9.62	12.44	8.90	2.22	0.06

Calculated percent proportion of the polar bear food over time estimated from the fatty acid composition in the adipose tissue

	Ad E			Ad M			luvenile			Total Mean of ringed seal	Total Mean of barn seal	Total Mean of booded seal
	Mean of	Mean of	Mean of	Mean of	Mean of	Mean of	Mean of	Mean of	Mean of	Ingedised	narpiscar	nooucuiscui
Year	ringed seal	harp seal	hooded.seal	ringed.seal	harp.seal	hooded.seal	ringed.seal	harp.seal	hooded.seal			
1983							32.0	.0.0	17.9	32.0	0.0	17.9
1984	93.4	6.3	0.0				86.9	9.0	0.1	89.7	7.8	0.0
1986	52.3	27.7	17.2				35.1	31.6	28.9	39.4	30.6	26.0
1987							47.9	26.3	22.9	47.9	26.3	22.9
1988							50.8	38.8	8.5	50.8	38.8	8.5
1989				58.7	26.9	10.3	83.4	12.4	1.7	76.3	16.5	4.2
1990	31.8	25.3	32.5	24.5	48.0	11.8	67.8	14.2	14.2	44.3	27.5	18.9
1991	88.7	0.0	0.0				73.8	19.7	0.0	77.5	14.8	0.0
1992	36.3	30.0	23.1	37.7	37.0	19.7	54.3	32.6	8.8	45.3	33.8	15.0
1993	20.7	19.8	52.0	14.1	41.2	40.5	32.5	41.1	21.8	27.4	36.9	30.6
1994	95.8	1.4	0.0	58.0	19.7	11.3	57.4	19.7	17.8	60.8	18.2	14.1
1995	28.8	40.9	15.2	5.8	61.1	26.3	17.7	35.9	35.9	15.9	43.9	30.2
1996				3.1	71.5	21.8	63.5	20.5	12.9	48.4	33.2	15.1
1999	56.0	36.9	3.1	45.4	44.2	3.1	60.7	19.4	14.0	56.8	28.2	9.3
2000	54.5	18.9	22.6	46.4	23.4	21.8	61.3	23.1	10.3	56.1	22.1	16.1
2001	68.7	14.8	12.3	15.9	45.3	33.5	50.7	29.0	15.9	47.0	29.4	18.9
2003	71.3	5.6	11.2	61.8	25.4	5.1	63.6	9.0	16.2	64.6	13.2	11.9
2004	91.8	0.0	0.0	38.8	49.0	8.9	74.5	13.7	7.2	58.4	30.0	7.3
2006	82.3	17.7	0.0	22.8	29.5	40.6	27.8	17.8	52.8	34.0	23.3	38.7
2007	60.5	16.8	19.8	30.7	22.8	37.3	10.3	54.3	31.6	35.5	27.3	30.9
2008	53.5	7.4	37.4	38.7	30.0	21.2	63.9	21.0	12.4	45.0	23.0	24.8
2009	0.0	34.7	57.8	28.2	31.4	34.2	49.4	25.6	21.3	29.2	30.9	34.0
2010	49.3	14.7	30.0	12.5	37.1	49.2	56.2	26.9	9.4	43.0	25.4	26.2
2011				0.0	11.5	88.5	41.5	37.3	16.8	33.2	32.2	31.2
2012	86.7	9.5	2.5	68.7	23.1	7.9	17.4	17.5	57.3	46.7	17.6	31.5
2013	0.0	27.8	67.5	23.5	40.4	33.2	21.3	25.8	27.0	19.9	32.5	34.3
2014				25.7	18.6	53.8	40.6	48.2	8.4	33.1	33.4	31.1
2015	29.5	41.3	26.0	8.8	22.3	63.1	8.9	32.5	54.5	13.0	30.2	52.3
Grand												
Total	53.6	20.4	20.4	31.0	34.2	28.2	50.2	25.7	18.5	44.9	27.3	21.8

Food composition of the major three seal species per age/sex group and overall polar bears from Ittoqqortoormiit over time estimated from the fatty acid composition in the adipose tissue.

#### Chemical analysis for OHCs (PPM lw).

				lipids (%)			$\Sigma_{61}$ PCBs (ng g <sup>-1</sup> lw)				
		n	mean ± SD	median	min - max	п	mean ± SD	median	min - max		
ringed seal	all	20	92.20 ± 3.05	91.67	86.36 - 98.98	20	406.76 ± 149.84	384.41	205.01 - 676.64		
	newborn subadult	14	92.75 ± 3.42	91.91	86.36 - 98.98	14	351.07 ± 130.97	318.67	205.01 - 654.55		
	adult - f adult - m	6	90.91 ± 1.43	90.84	89.05 - 93.02	6	536.71 ± 108.46	555.18	392.02 - 676.64		
bearded seal	all	6	83.36 ± 3.41	85.13	77.06 - 85.80	6	436.95 ± 298.87	410.16	71.54 - 980.80		
	newborn subadult adult - f adult - m	6	83.36 ± 3.41	85.13	77.06 - 85.80	6	436.95 ± 298.87	410.16	71.54 - 980.80		
harp seal	all	9	84.70 ± 21.36	86.86	34.30 - 116.60	9	292.92 ± 139.18	259.16	117.90 - 522.37		
	newborn subadult adult - f adult - m	4 5	74.67 ± 27.05 92.73 ± 13.50	86.17 87.35	34.30 - 92.03 83.58 - 116.60	4 5	307.65 ± 210.74 281.14 ± 71.03	295.17 259.16	117.90 - 522.37 221.29 - 395.51		
hooded seal	all	5	76.66 ± 10.10	78.70	64.79 - 89.31	5	1,118.58 ± 586.15	1,053.82	495.66 - 2,024.43		
	newborn subadult adult - f adult - m	5	76.66 ± 10.10	78.70	64.79 - 89.31	5	1,118.58 ± 586.15	1,053.82	495.66 - 2,024.43		
narwhal	all	19	67.16 ± 15.67	66.52	38.30 - 93.33	19	1,646.21 ± 798.66	1,901.94	114.85 - 2,792.29		
	newborn subadult adult - f adult - m	2 11 5 1	$85.18 \pm 11.53$ $65.41 \pm 15.04$ $67.83 \pm 15.29$	85.18 66.52 59.40 46.99	77.03 - 93.33 38.30 - 93.20 54.80 - 91.39	2 11 5 1	1,639.78 ± 1,396.09 1,989.70 ± 552.68 866.90 ± 729.08	1,639.78 2,053.63 821.77 1,777.34	652.60 - 2,626.96 966.14 - 2,792.29 114.85 - 2,049.88		

### Table 4

Chemical analysis for PFAS analyses.	

			Σ5Ρ	FSAs (ng g <sup>-1</sup> ww)	)		Σ₄PFCAs (ng g <sup>-1</sup> ww)				
		n	mean ± SD	median	min - max	п	mean ± SD	median	min - max		
ringed seal	all	20	51.84 ± 13.53	53.06	22.43 - 85.23	20	31.01 ± 7.65	31.63	16.05 - 48.48		
	newborn subadult adult - f	14	54.56 ± 13.33	54.39	36.17 - 85.23	14	29.3 ± 6.75	30.17	16.05 - 40.05		
	adult - m	6	45.51 ± 12.85	51.43	22.43 - 55.15	6	34.99 ± 8.77	35.75	23.49 - 48.48		
bearded seal	all	6	20.77 ± 5.23	22.66	13.53 - 26.96	6	10.99 ± 3.28	10.26	7.07 - 16.61		
	newborn subadult adult - f adult - m	6	20.77 ± 5.23	22.66	13.53 - 26.96	6	10.99 ± 3.28	10.26	7.07 - 16.61		
harp seal	all	8	27.72 ± 11.26	25.60	17.15 - 49.09	8	20.46 ± 6.07	20.58	11.39 - 30.53		
	newborn subadult adult - f adult - m	3 5	27.92 ± 11.11 27.59 ± 12.65	26.32 24.88	17.70 - 39.75 17.15 - 49.09	3 5	15.73 ± 3.99 23.30 ± 5.44	16.56 23.17	11.39 - 19.23 15.54 - 30.53		
hooded seal	all	5	$16.39 \pm 6.9$	14.33	8.40 - 26.86	5	21.7 ± 15.75	16.80	10.32 - 49.25		
	newborn subadult adult - f adult - m	5	16.39 ± 6.90	14.33	8.40 - 26.86	5	21.70 ± 15.75	16.80	10.32 - 49.25		
narwhal	all	17	20.79 ± 20.9	13.03	4.39 - 75.67	17	27.40 ± 21.96	18.30	10.16 - 89.20		
	newborn subadult adult - f adult - m	1 10 5 1	29.60 ± 23.52 5.67 ± 1.69	13.03 17.03 4.72 16.11	7.29 - 75.67 4.39 - 8.24	1 10 5 1	36.90 ± 24.59 11.91 ± 1.17	16.88 25.18 12.00 20.31	16.93 - 89.20 10.16 - 13.11		

#### Chemical analysis for mercury.

				dry weight (%)			Hg (µg g <sup>-1</sup> dw)		
			n mean ± SD	median	min - max	n	mean ± SD	median	min - max
ringed seal	liver	all	20 28.69 ± 1.16	28.71	26.86 - 31.19	20	$18.95 \pm 9.10$	18.65	6.57 - 37.51
		newborn subadult	14 28.72 ± 1.26	28.86	26.86 - 31.19	14	18.57 ± 8.64	17.63	6.57 - 36.54
		adult - f adult - m	6 28.60 ± 0.99	28.38	27.27 - 29.79	6	19.84 ± 10.93	20.14	8.53 - 37.51
bearded seal	muscle	all	7 27.86 ± 4.78	28.46	20.15 - 33.6	7	$0.68 \pm 0.26$	0.54	0.39 - 1.10
		newborn subadult adult - f adult - m	6 27.76 ± 5.23	28.49 28.46	20.15 - 33.6	6 1	0.66 ± 0.28	0.53 0.80	0.39 - 1.10
	liver	all	7 29.13 ± 1.74	29.96	27.02 - 31.63	7	9.34 ± 8.45	6.31	3.71 - 28.18
		newborn subadult adult - f adult - m	6 29.39 ± 1.76	29.98 27.61	27.02 - 31.63	6 1	9.73 ± 9.19	6.18 7.04	3.71 - 28.18
	kidney	all	7 23.09 ± 1.02	23.18	21.38 - 24.34	7	$5.72 \pm 4.90$	3.60	3.03 - 16.51
		newborn subadult adult - f adult - m	6 23.38 ± 0.75	23.22 21.38	22.33 - 24.34	6 1	6.13 ± 5.24	3.79 3.27	3.03 - 16.51
harp seal	muscle	all	11 32.86 ± 3.75	32.30	27.39 - 41.95	11	$0.53 \pm 0.17$	0.52	0.29 - 0.86
		newborn subadult adult - f adult - m	5 33.55 ± 1.64 5 30.35 ± 1.83	33.19 31.22	32.30 - 36.32 27.39 - 31.91	5 5	$0.59 \pm 0.16$ $0.47 \pm 0.19$	0.52 0.42	0.48 - 0.86 0.29 - 0.77
	liver	all	11 28.8 ± 3.00	29.76	20.55 - 31.42	11	5.54 ± 8.33	3.10	0.68 - 30.23
		newborn subadult adult - f adult - m	5 30.08 ± 0.99 5 27.39 ± 4.10	30.06 28.82	28.70 - 31.42 20.55 - 30.45	5 5	$3.95 \pm 1.56$ $2.18 \pm 1.24$	3.32 2.68	3.00 - 6.70 0.68 - 3.64
	kidney	all	11 23.24 ± 1.75	22.99	20.49 - 26.16	11	$2.15 \pm 0.89$	1.88	0.84 - 3.44
		newborn subadult adult - f adult - m	5 24.40 ± 1.64 5 22.64 ± 0.95	24.47 22.78	22.50 - 26.16 21.28 - 23.64	5 5	$2.42 \pm 0.90$ $1.63 \pm 0.56$	2.49 1.84	1.47 - 3.44 0.84 - 2.28
hooded seal	muscle	all	5 31.79 ± 1.16	31.62	30.72 - 33.77	5	$0.91 \pm 0.29$	0.88	0.62 - 1.36
		newborn subadult adult - f adult - m	5 31.79 ± 1.16	31.62	30.72 - 33.77	5	0.91 ± 0.29	0.88	0.62 - 1.36
	liver	all	5 35.19 ± 6.17	32.05	30.08 - 44.52	5	108.53 ± 116.01	72.79	34.45 - 313.79
		newborn subadult adult - f adult - m	5 35.19 ± 6.17	32.05	30.08 - 44.52	5	108.53 ± 116.01	72.79	34.45 - 313.79
	kidney	all	5 22.38 ± 1.47	22.91	20.53 - 23.86	5	17.35 ± 10.81	11.23	9.08 - 34.7
		newborn subadult adult - f adult - m	5 22.38 ± 1.47	22.91	20.53 - 23.86	5	17.35 ± 10.81	11.23	9.08 - 34.7
narwhal	blubber	all	24 92.54 ± 5.05	93.98	77.62 - 96.82	3	$0.17 \pm 0.07$	0.14	0.13 - 0.25
		newborn subadult adult - f adult - m	2 93.41 ± 2.60 14 92.01 ± 5.59 7 94.86 ± 1.77 1	93.41 93.98 95.77 82.03	91.57 - 95.25 77.62 - 96.82 92.82 - 96.75	1 2	0.20 ± 0.08	0.13 0.20	0.14 - 0.25
	muscle	all	24 25.53 ± 1.62	25.40	21.26 - 29.21	24	5.58 ± 15.10	2.72	0.81 - 76.33
		newborn subadult adult - f adult - m	$\begin{array}{cccc} 2 & 26.04 \pm 1.75 \\ 14 & 25.86 \pm 1.63 \\ 7 & 24.70 \pm 1.64 \\ 1 \end{array}$	26.04 25.33 25.32 25.71	24.80 - 27.28 23.49 - 29.21 21.26 - 26.18	2 14 7 1	$2.16 \pm 1.90$ $2.17 \pm 0.96$ $13.77 \pm 27.59$	2.16 2.32 3.56 2.99	0.81 - 3.5 0.94 - 3.7 2.65 - 76.33
	liver	all	24 25.88 ± 2.92	25.77	18.51 - 34.16	24	31.87 ± 29.89	28.15	1.22 - 120.90
		newborn subadult adult - f adult - m	$\begin{array}{cccc} 2 & 26.68 \pm 1.33 \\ 14 & 25.07 \pm 2.32 \\ 7 & 27.52 \pm 3.90 \\ 1 \end{array}$	26.68 25.38 25.91 24.08	25.74 - 27.62 18.51 - 27.41 22.19 - 34.16	2 14 7 1	$38.59 \pm 51.99$ $25.90 \pm 33.00$ $42.38 \pm 19.61$	38.59 12.64 45.39 28.29	1.83 - 75.35 2.26 - 120.90 1.22 - 59.13
	kidney	all	24 21.56 ± 2.36	21.99	13.13 - 25.62	24	8.21 ± 5.36	7.18	1.07 - 25.17
		newborn subadult adult - f adult - m	$\begin{array}{ccc} 2 & 20.48 \pm 1.24 \\ 14 & 21.71 \pm 2.89 \\ 7 & 21.57 \pm 1.61 \\ 1 \end{array}$	20.48 22.04 22.14 21.47	19.60 - 21.35 13.13 - 25.62 18.59 - 23.28	2 14 7 1	6.67 ± 7.92 6.97 ± 6.10 10.26 ± 1.86	6.67 6.04 10.80 14.39	1.07 - 12.27 1.95 - 25.17 7.16 - 12.34
	skin	all	24 23.49 ± 1.66	23.66	20.79 - 28.11	24	$2.13 \pm 0.96$	2.39	0.44 - 3.83
		newborn subadult adult - f adult - m	$\begin{array}{cccc} 2 & 25.01 \pm 1.75 \\ 14 & 23.42 \pm 1.65 \\ 7 & 23.09 \pm 1.76 \\ 1 \end{array}$	25.01 23.40 23.67 24.29	23.77 - 26.25 21.13 - 28.11 20.79 - 25.24	2 14 7 1	$2.03 \pm 2.16$ $1.74 \pm 0.81$ $2.82 \pm 0.58$	2.03 1.98 2.77 2.96	0.50 - 3.56 0.44 - 2.62 2.07 - 3.83

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#### Chemical analysis for stable isotopes.

				δ <sup>13</sup> C (‰)				δ <sup>15</sup>	N (‰)	
		n	mean ± SD	median	min - max	n	mean ± SD		median	min - max
ringed seal	all	20	-20.54 ± 0.38	-20.50	-21.5619.91	20	+14.19 ± 0.83		+14.30	+12248 - +15.63
	newborn subadult adult - f	6	$-20.62 \pm 0.41$	-20.52	-21.5620.03	6	+14.19 ± 0.79	•	+14.22	+12.88 - +15.63
	adult - m	14	$-20.35 \pm 0.24$	-20.44	-20.5719.91	14	$+14.19 \pm 1.00$		+14.40	+12.24 - +14.95
earded seal	all	7	$-21.18 \pm 1.02$	-21.10	-22.3719.31	7	+14.76 ± 1.60	•	+15.01	+11.73 - +16.49
	newborn subadult adult - f adult - m	6 1	-21.26 ± 1.09	-21.35 -20.69	-22.3719.31	6 1	+14.90 ± 1.70	;	+15.20 +13.94	+11.73 - +16.49
arp seal	all	11	-20.71 ± 1.58	-20.25	-25.0219.67	11	+12.36 ± 1.57		+12.62	+9.50 - +14.81
	newborn subadult adult - f adult - m	5 5	$-20.46 \pm 0.96$ $-20.10 \pm 0.36$	-20.02 -20.25	-22.1419.83 -20.5419.67	5 <b>*</b> 5	+14.81 +12.99 ± 1.10	;	+14.81 +13.43	+14.81 - +14.81 +11.25 - +14.06
ooded seal	all	5	-19.87 ± 1.01	-19.58	-21.6119.03	5	+13.51 ± 0.89		+13.46	+12.38 - +14.79
	newborn subadult adult - f adult - m	5	-19.87 ± 1.01	-19.58	-21.6119.03	5	+13.51 ± 0.89		+13.46	+12.38 - +14.79
arwhal	all	24	$-20.57 \pm 0.40$	-20.51	-22.0620.11	24	+14.36 ± 0.52		+14.32	+13.50 - +15.95
	newborn subadult adult - f	2 14 7	-20.56 ± 0.17 -20.54 ± 0.27 -20.60 ± 0.66	-20.56 -20.55 -20.38	-20.6820.44 -21.1520.18 -22.0620.11	2 14 7	+15.33 ± 0.87 +14.41 ± 0.25 +14.01 ± 0.54		+15.33 +14.38 +13.85	+14.72 - +15.95 +14.03 - +14.91 +13.50 - +15.10

Chemical analysis for mercury and other elements from bearded, harp and hooded seals from 2015 from the Scoresby Sound area.

			<b>Bearded seal</b> 2015: <i>n</i> =7				<b>Harp seal</b> 2015: <i>n</i> =11			<b>Hooded seal</b> 2015: <i>n</i> =5			
			mean ± SD	median	min - max	mean ± SD	median	min - max	mean ± SD	median	min - max		
δ <sup>13</sup> C	muscle	‰	-21.18 ± 1.02	-21.10	-22.3719.31	-20.71 ± 1.58	-20.25	-25.0219.67	-19.87 ± 1.01	-19.58	-21.6119.03		
δ <sup>15</sup> N	muscle	‰	+14.76 ± 1.60	+15.01	+11.73 - +16.49	+12.36 ± 1.57	+12.62	+9.50 - +14.81	+13.51 ± 0.89	+13.46	+12.38 - +14.79		
Cd	muscle	µg g <sup>-1</sup> dw	$0.15 \pm 0.26$	0.03	0.01 - 0.73	$0.13 \pm 0.12$	0.14	0.01 - 0.38	0.29 ± 0.18	0.25	0.16 - 0.61		
	liver	µg g <sup>-1</sup> ww	$1.79 \pm 1.65$	1.11	0.87 - 5.47	$4.85 \pm 3.87$	4.25	0.35 - 10.56	15.31 ± 6.04	16.09	7.04 - 22.82		
	kidney	µg g <sup>-1</sup> ww	$8.41 \pm 11.98$	2.68	1.98 - 34.18	$18.21 \pm 12.58$	16.48	1.26 - 38.28	87.97 ± 37.18	103.76	21.78 - 110.36		
Zn	muscle	µg g <sup>-1</sup> dw	91.09 ± 40.7	93.18	33.29 - 167.20	68.93 ± 21.96	73.76	29.73 - 103.53	80.39 ± 43.89	58.52	51.40 - 156.48		
	liver	µg g <sup>-1</sup> ww	42.95 ± 3.52	43.35	37.55 - 46.32	40.05 ± 15.43	43.91	17.26 - 70.76	51.29 ± 18.85	42.10	41.12 - 84.83		
	kidney	µg g <sup>-1</sup> ww	23.47 ± 4.05	22.01	19.53 - 31.28	25.28 ± 6.10	24.55	18.12 - 41.81	68.59 ± 23.28	70.63	30.63 - 90.17		
Hg	muscle	µg g <sup>-1</sup> dw	$0.68 \pm 0.26$	0.54	0.39 - 1.10	$0.53 \pm 0.17$	0.52	0.29 - 0.86	0.90 ± 0.29	0.88	0.62 - 1.36		
	liver	µg g <sup>-1</sup> ww	2.74 ± 2.55	1.82	1.17 - 8.44	$1.63 \pm 2.45$	0.93	0.14 - 8.89	36.22 ± 36.43	23.06	14.13 - 100.58		
	kidney	µg g <sup>-1</sup> ww	1.32 ± 1.13	0.87	0.68 - 3.80	$0.50 \pm 0.21$	0.44	0.19 - 0.89	3.86 ± 2.30	2.57	1.86 - 7.34		
Se	muscle	µg g <sup>-1</sup> dw	$2.73 \pm 2.41$	1.73	1.47 - 8.14	$1.23 \pm 0.31$	1.26	0.80 - 1.95	$1.32 \pm 0.27$	1.30	0.99 - 1.68		
	liver	µg g <sup>-1</sup> ww	$2.32 \pm 1.12$	1.72	1.63 - 4.72	$1.90 \pm 1.21$	1.72	0.37 - 4.74	14.52 ± 13.06	9.28	6.56 - 37.63		
	kidney	µg g <sup>-1</sup> ww	$2.86 \pm 0.55$	2.92	2.13 - 3.49	$2.56 \pm 0.89$	2.41	1.47 - 4.62	3.77 ± 1.08	3.66	2.65 - 5.53		

Chemical analysis for mercury and other elements from bearded, harp and hooded seals from 2015 from the Scoresby Sound area.

				<b>Ringed s</b> 2014: <i>n</i> =	<b>eal</b> 20		<b>Narwhal</b> 2015: <i>n</i> =24				
			mean ± SD	median	min - max	mean ± SD	median	min - max			
δ <sup>13</sup> C	muscle	‰	-20.54 ± 0.38	-20.50	-21.5619.91	-20.57 ± 0.40	-20.51	-22.0620.11			
δ <sup>15</sup> N	muscle	‰	$+14.19 \pm 0.83$	+14.30	+12248 - +15.63	+14.36 ± 0.52	+14.32	+13.50 - +15.95			
Cd	muscle liver kidney skin blubber	μg g <sup>-1</sup> dw μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww	13.02 ± 8.44	11.38	2.95 - 31.16	$11.55 \pm 52.8620.01 \pm 16.3667.77 \pm 38.940.02 \pm 0.030.08 \pm 0.13$	0.62 16.08 73.92	0.03 - 259.71 0.01 - 54.00 0.16 - 153.90 0.00 - 0.14 0.00 - 0.49			
Zn	muscle liver kidney skin blubber	μg g <sup>-1</sup> dw μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww				$89.42 \pm 24.08$ $35.21 \pm 10.35$ $45.21 \pm 8.22$ $83.58 \pm 39.63$ $2.03 \pm 0.61$	87.59 34.67 45.14	54.69 - 161.95 4.43 - 52.43 34.6 - 63.66 56.29 - 262.25 0.96 - 3.22			
Hg	muscle liver kidney skin blubber	μg g <sup>-1</sup> dw μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww	5.43 ± 2.62	5.13	2.05 - 11.18	$5.58 \pm 15.10 \\ 8.16 \pm 7.97 \\ 1.75 \pm 1.17 \\ 0.50 \pm 0.22 \\ 0.05 \pm 0.05$	2.72 6.83 1.56	0.81 - 76.33 0.42 - 32.48 0.23 - 5.54 0.10 - 0.85 0.03 - 0.23			
Se	muscle liver kidney skin blubber	μg g <sup>-1</sup> dw μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww μg g <sup>-1</sup> ww	4.03 ± 1.27	4.26	1.57 - 5.95	$2.18 \pm 5.35 \\ 4.40 \pm 3.30 \\ 2.42 \pm 0.67 \\ 5.48 \pm 1.97 \\ 0.13 \pm 0.07$	1.10 3.36 2.30	0.79 - 27.27 0.90 - 14.36 0.76 - 4.00 0.61 - 8.37 0.02 - 0.31			

UNEXPECTED INCREASES OF PERSISTENT ORGANIC POLLUTANT AND MERCURY LEVELS IN EAST GREENLAND POLAR BEARS (UNEXPECTED)

Climate change has taking place in the Arctic in recent years with significant consequences for the amount of sea ice and species occurrence and distribution including food availability. In this study, we account for the polar bears' food selection as a result of the changes in sea ice using quantitative fatty acid signature analysis (QFASA) and fatty acid specific stable carbon and isotopes ( $\delta^{13}$ C-FA) to assess the nutritional basis of East Greenland polar bears over the last three decades. This show that East Greenland polar bears mainly eat ringed seals (44.9%), migrating subarctic harp seals (27.3%) and hooded seals (22.9%) and relatively rarely eat species such as bearded seals (4.9%), narwhals (1.0%) or walruses (0.06%). The time trend analysis reflect a clear correlation between increased concentrations of organohalogen compounds (OHC) and mercury and a decrease in the concentration of per- and polyfluorinated alkyl compounds (PFAS), respectively, and changes in food availability. Overall this indicate that climate change lead to an increase in the contaminant loads of Arctic top predators and thus potentially the East Greenlandic Inuit population living from subsistence harvest.