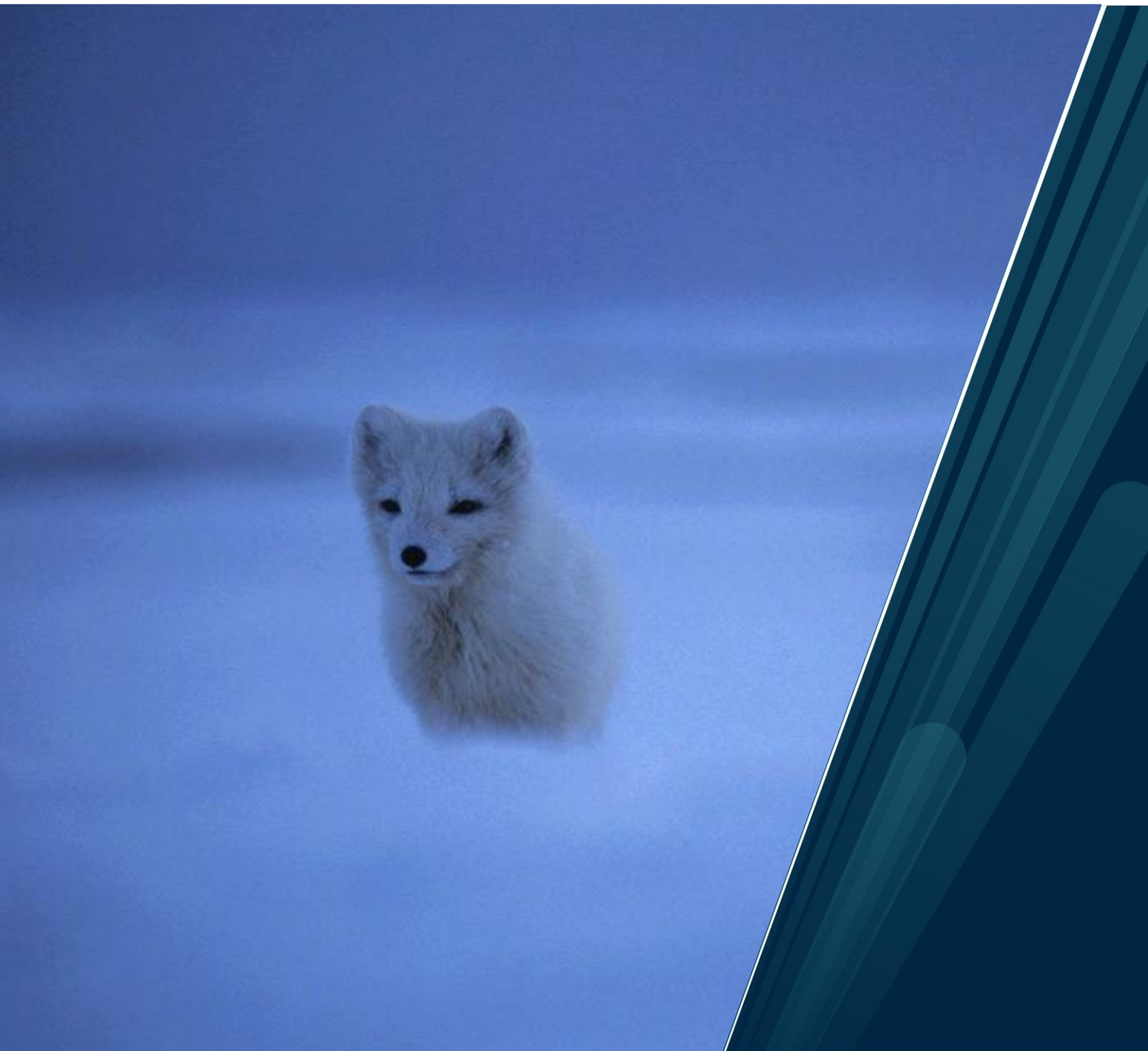UiT The Arctic University of Norway

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Temporal changes of persistent organic pollutants in Arctic foxes (*Vulpes lagopus*) from Svalbard

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Acknowledgements

Abstract

TEMPORAL CHANGES OF PERSISTENT ORGANIC POLLUTANTS IN ARCTIC FOXES (*VULPES LAGOPUS*) FROM SVALBARD

Recent evidence shows that temporal changes of persistent organic pollutants (POPs) in Arctic ecosystems are not only due to emission patterns and regulations; environment-related changes in prey availability and long-range transport may also influence concentrations of these compounds in tissues of Arctic predators. Arctic foxes (*Vulpes lagopus*) in Svalbard, Norway, scavenge and hunt opportunistically for variable prey items throughout the year. In this study, temporal trends of POPs in arctic foxes from Svalbard were updated. Organochlorinated pesticides (OCPs), polychlorinated byphenyls (PCBs) and brominated flame retardants (polybrominated diphenyl ethers [PDBEs] and hexabromocyclododecane [HBCDD]) were analysed in total of 209 liver samples from 1997 to 2019. Stable isotope values (δ^{13}C, δ^{15}N) in muscle tissue were used as proxies for feeding habits on marine versus terrestrial food and trophic position respectively. Reindeer carcasses, size of geese population and sea ice extent were used as proxies for food availability. Non-linear additive models were used to analyze temporal changes of POPs in relation to variation in feeding habits, food availability, body condition and concentrations of POPs in air from Svalbard. Measured POP changes were compared to changes corrected for temporal variation in diet and food availability, to investigate the potential effect of direct or indirect environment-related changes on POPs. All contaminants increased with higher trophic position and marine diet. All contaminants were lower in fat than lean individuals apart from BDE-47. HCB concentrations decreased when reindeer carcasses were abundant and increased with increasing sea ice extent. Changes in concentrations for contaminants showed a general decline through the whole study period, which is in accordance with the decrease of PCBs and organochlorine pesticides in Arctic biota during the last 20 to 30 years. All our estimated changes per year for adjusted and measured concentrations were similar among contaminants, with the exception for BDE-47 which showed a higher rate of decline compared to other contaminants when adjusted for covariates. HCB changes per year were lower when adjusted for covariates, with overlapping confidence intervals, meaning that changes in diet and food availability in arctic foxes could non-significantly affect the temporal changes of contaminants.

Contents

1. **Introduction**

Chemicals known as persistent organic pollutants (POPs) are characterized for their ability to remain intact for long periods of time, be distributed throughout the environment by hydrological and atmospheric processes involving soil, water and air, bioaccumulate and biomagnify, and pose a threat for wildlife and humans (AMAP, 2016). These contaminants, along with mercury, are the dominant chemicals polluting the Arctic and affecting its wildlife and human health (Mckinney et al., 2015). Various studies have examined long-term trends of POPs in apex predators, which provide important means of how these contaminants increase, decrease or stabilize in the environment in response to regulatory actions and climate change (Rigét, et al., 2019). Regulations of POPs started in 1970s and a global convention, the Stockholm Convention, established to reduce or eliminate emissions of POPs placed in 3 categories: pesticides, industrial chemicals, by-products, was ratified in 2004 (Wania and Mackay, 1993: Stockholm Convention, 2004). POPs are resistant to degradation; therefore, these chemicals may reach the Arctic from distant sites of production and use via ocean and air currents and river outflows. Most POPs are lipophilic, which facilitates their biomagnification through the lipid-rich arctic food webs and lead to high concentrations in apex predators, such as polar bears (*Ursus maritimus*) and arctic foxes (*Vulpes lagopus*) (Muir et al., 1988; Hoekstra et al., 2003; Macdonald et al., 2003). Arctic predators rely on adipose tissue as a cyclic seasonal storage of energy. Therefore, during food shortage periods, POPs become bioavailable resulting in an increased exposure to vital organs and higher risk of health issues (Helgason et al., 2013).

Temporal trends of POPs in Arctic air have shown that PCBs, DDTs and chlordanes are declining slowly, reflecting the reduction of primary emissions, while some lighter PCBs and HCB have shown increasing trends at specific locations. PBDEs have been declining in European Arctic air, whereas in the Canadian Arctic there was no evidence of a declining trend (Hung et al., 2016).

Levels and temporal trends of POPs in Arctic biota do not necessarily reflect only emission patterns but may also be influenced by the environment and feeding conditions (McKinney et al., 2013). Significant declines in PCBs and chlordanes have been found when considering body condition and diet. Higher levels of contaminants were found in lean compared to fat

arctic foxes; years with higher reindeer mortality and more available reindeer carcasses were not excluded as a possibility that could affect POP concentrations in arctic foxes (Andersen et al., 2015). Levels of PCBs have also been found to decline in polar bear plasma samples in the Barents Sea, showing higher concentrations of contaminants in lean compared to fatter individuals; body condition had a higher impact on POP concentrations than diet, except for HCB and BDE-47, where trophic position influenced concentrations (Lippold et al., 2019). Levels of THg (total mercury) have been reported to increase in female polar bear hair from the Barents Sea when correcting concentrations for stable isotope values and additionally breeding status and age (Lippold et al., 2020), similarly, liver THg levels in arctic foxes from Svalbard increased per year when concentrations were adjusted for variations in diet, sea ice extent and reindeer carcass availability (Hallanger et al., 2019).

The arctic fox inhabiting the archipelago of Svalbard relies on both the terrestrial and marine ecosystem, mainly on birds and mammals (Prestrud, 1992; Eide et al., 2005). During early spring foxes may prey on new-born ringed seal pups (*Pusa hispida*) on the sea ice (Lydersen and Gjertz, 1986), before late spring and summer there is an excess of food availability with the arrival of migrating seabirds and goose. During the winter, arctic foxes rely on carcasses of Svalbard reindeer (*Rangifer tarandus platyrhynchus*) and seals (*Phocidae*), and hunt Svalbard rock ptarmigan (*Lagopus muta hyperborea*) and food items stored during the previous season (Prestrud, 1992; Frafjord, 1993; Ehrich et al., 2015).

Over the last decade, several studies have shown large variation of sea ice extent in several fjords along the western coast of Spitsbergen, where the sea ice extent is reduced between 5 and 20% per decade, making fjords in the west virtually ice-free in recent winters and shifting the local climate towards more maritime conditions (Dahlke et al., 2020). Decreases in sea ice extent will likely impact arctic fox´s foraging through reduced availability of ringed seal pups, polar bear carrion and reduced access to other marine resources (Fuglei and Ims, 2008; Ims et al., 2013). Events like "rain on snow" during winter causing thick layers of ground ice and ice layers in the snow, making foraging difficult for herbivores. This leads to starvation, reduced reproduction and high mortality rates in the Svalbard reindeer population, increasing the number of reindeer carcasses available as food for the arctic fox during winter (Hansen et al., 2011; Hansen et al., 2013; Tyler and Øritsland, 1999). High number of reindeer carcasses, terrestrial food, has been associated with lower levels of POPs in arctic foxes (Andersen et al., 2015).

Migratory goose populations have increased in number in recent years in Svalbard, making them an abundant potential prey for arctic foxes. Pink-footed geese (*Anser brachyrhynchus*) population has reportedly reached ~100,000 thousand individuals as of 2015, suggesting a recent stabilization in the population due to declining survival rates, attributable to increasing hunting pressure in Denmark (Johnson et al., 2020).

The present study was designed to investigate temporal changes from 1997 to 2019 of POPs in arctic foxes from Svalbard, Norway in relation to reindeer carcass availability, goose availability, stable isotopes signatures, variation in sea ice and POPs in the air. We hypothesize that POP emission patterns and changes in feeding habits, possibly related to climate change, may affect temporal trends in the arctic fox. Arctic foxes sampled in 2013-2019 were analyzed for pollutants as part of this study and the data was combined with previously published results (Andersen et al., 2015).

2. **Material and Methods**

2.1 Sample collection

Arctic foxes were trapped in Spitsbergen, Svalbard, mainly around the Isfjorden area, Nordenskiöld Land (77-79° N, 13-18° E, Figure 1). The foxes were collected by local trappers, using baited traps during the annual harvest (Appendix 1.) between November 1st and March 15th in 1997/98 (n=12), 1998/99 (n=14), 1999/00 (n=14), 2001/02 (n=14), 2002/03 (n=14), 2003/04 (n=14), 2007/08 (n=14), 2010/11 (n=16), 2011/12 (n=14), 2012/13 (n=13), 2013/14 (n= 12), 2014/15 (n= 14), 2015/16 (n= 8), 2016/17 (n= 12), 2017/18 (n= 12) and 2018/19 (n= 12), samples from Andersen et al., 2015, previously analyzed and published (1997/2013). The foxes were weighed, sex determined and skinned at Svalbard before the frozen carcasses were transported to Tromsø where the final dissection took place. The age of the foxes was determined by counting the annuli in the cementum of a sectioned canine tooth (Grue and Jensen, 1976). When evaluating body condition, a subjective fat index based on visual inspection of the skinned carcasses was used (Prestrud and Nilssen, 1992). The index ranged from 1 to 4 (almost or close to none to extensive). Samples of skeletal muscle and liver were packed in aluminum foil and stored at -20°C until further analysis. All foxes in this

study were between 1 and 2 years old. Svalbard arctic fox vixens start reproducing at an age of 1, but at very low rates. They do not reach rates close to 100% before the age of four (Eide et al., 2012), therefore very few vixens had given birth. A total of 209 foxes were analyzed and sex, age and body condition were balanced over years.
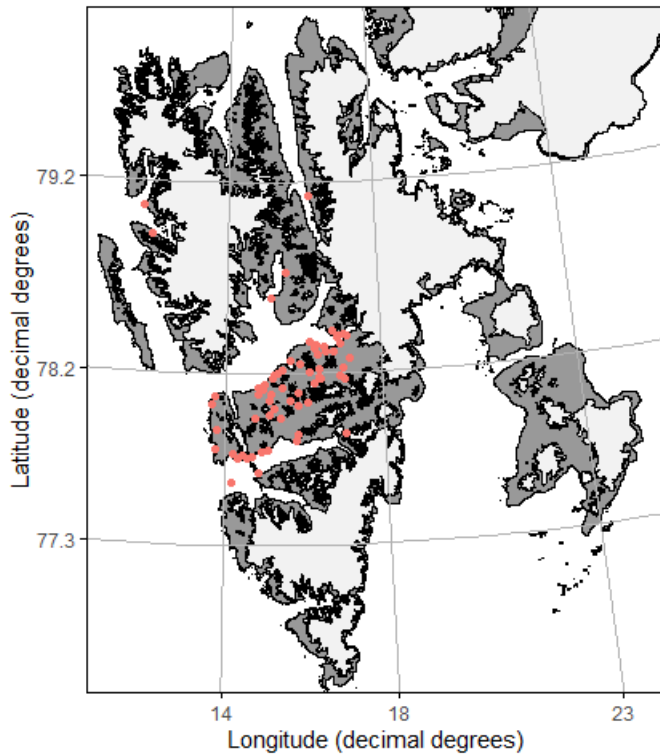
Figure 1. Map showing the study area in Nordenskiöld Land, in Spitsbergen, Svalbard; colored dots show trapping locations.

2.2 Reindeer carcass data

The number of reindeer carcasses in summer was used as an index for the availability of reindeer as terrestrial food for arctic foxes in the spring preceding the trapping season. The data was derived from a long-term monitoring survey in Adventalen (Hansen et al., 2013; Tyler and Øritsland, 1999). Due to high correlations in population density among reindeer numbers in different areas of Nordenskiöld Land (Aanes et al., 2003), we assumed that the inter annual pattern of reindeer mortality in Adventalen was a good proxy for other populations in Nordenskiöld Land.

2.3 Sea Ice data

We used data on the sea ice extent of Isfjorden as an index for availability of ringed seal pups as marine prey for arctic foxes during the spring preceding the trapping season. Average sea ice extent was calculated for each month (November-March; 1997-2019) using daily sea ice charts of Isfjorden produced by the Norwegian Ice Service (NIS) using remote sensing digital imagery (Dahlke et al., 2020). For each fox with known trapping date, we used the sum of the monthly average sea ice from first of November to the month when the fox was trapped. The sum of the monthly average sea ice included the month when the fox was trapped only if the trapping occurred after the 15th of the given month. For the foxes without known trapping date the average sea ice cover from first of November to 30th of February divided by two was used.

2.4 Goose data

Estimates for populations in spring preceding the trapping season for foxes were taken from known sites occupied by geese in Norway, these are covered by a network of trained observers who coordinate the coverage. Counts were conducted in a single day when possible, to avoid double counting. Flocks were either counted when they were leaving roost sites in the morning, when the roost was in the middle of the day, or during daytime when geese were in fields to forage. Estimates for the month of May were used based on capture-mark-resight data, estimates were expressed in thousands of birds, rounded to the nearest thousand, the May 1999 estimate was omitted since it was based on a single observation of a marked bird, see Johnson et al., 2020 for the complete and detailed methodology.

2.5 POPs in the Air

Air samples were collected at the Zeppelin station in Ny-Ålesund, Svalbard using high-volume air samplers equipped with a glass fiber filter and polyurethane foam plugs to trap particle-and-gas-phase chemicals, respectively. Weekly samples (gas + particle) phase concentrations covering 48-72 h and ~1000-1800 m^3 of air were analyzed for POPs and used to calculate yearly averages for the first calendar year of the trapping season (e.g., Jan-Dec 2018 for trapping season 2018-2019). Not all years had available air data, PCBs and BDE-47,

-153 data were available from 2001 since possible local contamination affected prior years (NILU, 2019). Detailed information on sample collection, chemical analysis, data handling and quality assurance/quality control can be found in Hung et al., (2010) and Hung et al., (2016).

2.6 Stable Isotope Analysis of δ^{13}C and δ^{15}N in arctic fox muscle

The ratio of stable isotopes of carbon (13C/12C; δ^{13}C) is used to distinguish between terrestrial and marine food items eaten, while the ratio of nitrogen stable isotopes (15N/14N; δ^{15}N) reflects the trophic position of prey/food (Fisk et al., 2001; Hop et al., 2002; Kelly, 2000). Muscle tissues were sampled from arctic foxes trapped between November and March. Stable isotope analysis is expected to reflect the autumn/winter diet, depending on when the arctic fox was caught. For detailed information about methodology on the analysis see Andersen et al., (2015).

2.7 Chemical analysis of OCPs, PCBs and PBDEs

Analysis of the liver sample contaminants was conducted at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences, Campus Oslo, Norway (MT-lab). The methodology for analyzing samples from trapping seasons prior to 2013 can be found in Andersen et al., 2015. The laboratory is accredited for testing chemicals in biological material according to the Norwegian accreditation in the requirement of the NS-EN ISO/IEC 17025 (TEST 137). The following organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) were analyzed; hexachlorobenzene (HCB), alpha-hexachlorocyclohexane (α-HCH), beta-hexachlorocyclohexane (β-HCH), gamma-hexachlorocyclohexane (γ-HCH), dichlorodiphenyltrichloroethane (p,p'-DDT), dichlorodiphenyldichloroethylene (p,p'-DDE), oxychlordane, trans- nonachlor and perchloropentacyclodecane (mirex), and PCB-28, -52, -101, -118, -138, -153 and -180, PBDEs: PBDE-47, -99, -100,, -154, -153 and -209, as well as sum of isomers α, β, and γ of HBCDD.

2.7.1 Lipid extraction

Extraction was based on the technique described by Brevik, (1978) and modified by Polder et al., (2008), consisting of using cyclohexane and acetone in a liquid/liquid extraction. The liver samples were cut into small pieces with a scalpel to reach the adequate weight to three decimals, 2 g (±0.200 g). Internal standards (I.S) of PCBs (PCB-29, -112, -207), and PBDEs (BDE-77, -119, -183, and 13C12-BDE-209) were added to each sample, followed by 2 mL 6% NaCl, 10 mL grade 1 water, 15 mL acetone and 20 mL of cyclohexane. Samples were then homogenized with Ultra Turrax (IKA Ultra-Turrax T25, IKA Laboratory Technology, Staufen, Germany) and then sonicated for 2 minutes with an ultrasonic homogenizer (Cole Parmer CPX 750, Vernon Hills IL, USA). After centrifugation for 10 minutes (Allegra X-12R Beckman Coulter, Fullerton, CA, USA) the top layer was collected. The second extraction was performed by adding 5 mL of acetone and 10 mL of cyclohexane to the original sample tubes, followed by sonication for 1 minute and centrifuging for 10 minutes. The extracts were collected and evaporated down to 1 mL at 40°C by a steady flow of nitrogen (purity: 99.6%; AGA AS, Oslo, Norway, pressure 0.6 bar) in a Zymark® evaporation system (TurboWap II, Zymark Corporation, Hopkinton, MA, USA). The final step consisted of transferring the concentrated extracts to 5 mL volumetric flasks and adjusting to the mentioned volume with cyclohexane.

2.7.2 Gravimetric lipid determination and acid cleanup

Lipid determination was done gravimetrically by transferring 1 mL of the sample to a pre weighed dram flask, allowing solvent to evaporate before weighing. The remaining 4 mL of extract was cleaned using sulfuric acid, H_2SO_4 (purity 96%; Fluka analytical, Sigma-Aldrich, St. Louis, USA). The extracts were evaporated using a flow of nitrogen to a final volume of 0.2 mL, then transferred to Gas Chromatography (GC) vials with glass inlets.

2.7.3 Gas Chromatography analysis of OCPs, PCBs and PBDEs

The details for the GC analyses are as described by Polder et al., 2008; Separation and detection of OCPs and PCBs were performed on a HRCG-ECD (Agilent 6890 Series) coupled to a Mass Spectrometry (MS) detector (Agilent 5975C Agilent Technologies)

operated in negative chemical ionization (NCI) mode with selected ion monitoring (SIM) and configured with a programmable temperature vaporization (PTV) injector (Agilent Technologies). The OCP compounds were separated on a DB-5 MS column (60 m, 0.25 mm i.d., 0.25 mm film thickness: J&W Scientific). The carrier gas was helium (He) at 1.3 mL/min constant flow. Detection of all brominated flame retardants except BDE-209 was performed on a HRGC (Agilent 6890 Series; Agilent Technologies) coupled to a MS detector (Agilent 5973; Agilent Technologies). Separation and identification of the compounds were performed on a Db-5 MS column (30 m, 0.25 mm i.d.; 0.25 mm film thickness; J&W Scientific). The carrier gas was Helium (He) at 1.6 mL/min constant flow. For analysis of BDE-209, samples were injected on a Gas Chromatography-Mass Spectrometry instrument (Agilent 6890 Series/5973 Network) configured with a programmable temperature vaporization (PTV) injector (Agilent Technologies), with a 10 m DB-5-MS column (J&W Scientific, Agilent Technologies). The carrier gas was Helium (He) at a 1.8 mL/min constant flow. All instruments were equipped with an auto-sampler (Agilent 7683 Series: Agilent Technologies).

The lowest level of detection (LOD) for individual compounds was determined as three times the noise level. LODs (Appendix 3) range from 0.003 ng/g to 0.049 ng/g for OCPs, 0.003 ng/g to 0.058 ng/g for PCBs and 0.005 ng/g to 0.042 ng/g for BFRs.

2.7.4 Analytical quality control

The analytical quality of the laboratory is routinely assessed by analyzing certified reference materials (CRMs) and participating in interlaboratory tests from the Arctic Monitoring and Assessment Program (AMAP) and Quasimeme. Each series consists of 17 samples, 3 blanks, one blind and 2 spiked recoveries (sheep liver), as well as the laboratory's reference material of seal blubber (MTref01). The blank samples consisted of internal standards (I.S) and solvents. GC runs include a standard repetition every 10 injections for potential drift. Recoveries (Appendix 3) were between 73 and 367 percent for OCPs, 86 and 120 percent for PCBs and 78 and 108 percent for BFRs. Results for p,p'-DDT, β-HCH and Oxychlordane were corrected based on recovery.

2.8 Data analysis

Statistical analyses included POPs detected in more than 80% of the samples, values below the LOD were replaced by half the detection limit before lipid normalization of the data. Analyses were carried out using the statistical program R, version 3.6.2 (RStudio Team, 2020). Contaminants were grouped after their chemical properties and emission histories to reduce the number of statistical tests and simplify the interpretations of the results. Only contaminants with data available for POPs in the air were considered for further analyses, ΣPCBs (PCB-118, -138, -153 and -180), ΣCHL: (*trans*-nonachlor and oxychlordane) and HCB. In addition, BDE-47 and BDE-153 were analyzed without data for POPs in the air.

The high correlation between $\delta^{13}C$ and $\delta^{15}N$ (R^2=0.73) prevented the inclusion of both explanatory variables in the same model. The variables $\delta^{13}C$, $\delta^{15}N$, average sea ice extent, number of reindeer carcasses, goose population estimates, HCB in air, ΣCHL in air and ΣPCB in air were standardized (mean=0, std. dev.=1) before the analysis (Gelman and Hill, 2007). The variable year was added as a non-linear term (spline) in the models (see below, Appendix 2). All contaminant data were log-transformed and diagnostic plots (Appendix 4) were used to determine if the model residuals met the assumptions of the statistical models applied. Constant variance and approximate normal distribution of residuals were assessed through plots of residuals against fitted values and normal quantile-quantile plots (Zuur et al., 2010).

Additive models (Wood, 2006) were used to investigate POP concentrations against year, $\delta^{13}C$, $\delta^{15}N$, reindeer mortality, sea ice coverage, geese population estimates and POPs in air. We ranked 34 candidate models using Akaike´s Information Criterion (AIC; Burnham and Anderson, 2002) for the data including POPs in air and 23 candidate models for BDE-47 and BDE-153. Model averaging was used to make inference from all models. This method produces averaged estimates of all effects of explanatory variables in the candidate list, weighted using the AIC weights (Burnham and Anderson, 2002; Lukacs et al., 2010). We considered CIs (confidence intervals) of the model averaged estimates excluding zero as showing a statistically significant effect. Model-averaged CIs have better coverage properties than post-selection CIs (Burnham and Anderson, 2002). Yearly change (%) was derived by $100*(e^{\text{estimate for year}} - 1)$ using the highest ranked model and POP concentrations against year.

Estimates were derived from linear models, i.e., the change was adjusted for biological and environmental changes and thus reflect the overall change of POPs concentrations in arctic fox food web during the whole study period. We compared the adjusted change to the non-adjusted change derived from a model that included only year as a predictor variable. These back-transformed estimates reflect changes in median concentrations (Limpert et al., 2001).

3. **Results**

PCBs was the most abundant contaminant group in arctic fox liver samples. Median concentrations (Table 1) for PCBs were the highest for trapping season 1998/99 (16327 ng/g lipid weight, CIs= 1244, 46973) and the lowest for trapping season 2018/19 (584 ng/g lipid weight, CIs= 133, 4108). Chlordanes was the second more abundant contaminant group in liver samples, having the highest median values for trapping season 1998/99 (15523 ng/g lipid weight, CIs= 1710, 36514) and the lowest concentration for trapping season 2018/19 (323 ng/g lipid weight, CIs= 53, 1852). HCBs followed as the third abundant contaminant, having the highest median concentration for trapping season 2013/14 (277 ng/g lipid weight, CIs= 37.4, 837) and the lowest median concentration for trapping season 2016/17 (29.4 ng/g lipid weight, CIs= 11.1, 442). BDE-47 concentrations were considerably lower showing the highest median concentration for trapping season 1997/98 (20.6 ng/g lipid weight, CIs= 2.86, 183) and the lowest concentration for trapping season 2016/17 (0.28 ng/g lipid weight, CIs= 0.01, 35.6). BDE-153 came up as the least abundant contaminant, with the highest median concentration for trapping season 1997/98 (7.36 ng/g lipid weight, CIs= 1.69, 29.8) and the lowest median concentration for two trapping seasons 2016/17 (0.29 ng/g lipid weight, CIs= 0.11, 2.21) and 2018/19 (0.29 ng/g lipid weight, CIs=0.11, 5.7).

Table 1. Median, Minimum and Maximum Concentrations in Liver Samples of arctic foxes Collected in Svalbard, Norway 1997-2019

	HCB			ΣCHL			ΣPCB			BDE47			BDE153		
Year	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max
1997	141	62.1	954	12436	1319	34009	12152	2590	53151	20.6	2.86	183	7.36	1.69	29.8
1998	261	50.1	962	15523	1710	36514	16327	1244	46973	14.4	3.87	70.4	4.38	3.78	34.1
1999	92.5	41.4	494	7116	1095	36940	6465	561	40034	2.96	0.62	71.9	2.53	1.13	25.3
2001	242	67	495	14808	4424	37214	12796	5135	34962	14.9	1.98	36.8	4.36	1.12	16.2
2002	48.7	10.6	381	2210	229	25574	3619	1023	18533	2.97	0.66	20.5	3.59	1.68	11.6
2003	122	8.77	317	3679	336	48726	3499	238	49667	5.79	0.97	45.6	3.8	0.75	20.9
2007	60.1	18.2	730	652	213	3735	1530	652	37119	4.32	0.86	200	1.66	0.86	49.1
2010	74.7	18.5	1030	4062	161	27970	3482	82.6	23331	2.96	0.63	65.1	1.5	0.94	7.51
2011	111	26.8	1082	1490	167	10184	2913	538	26259	3.99	0.73	9.26	1.13	0.55	5.63
2012	118	28.6	559	3066	122	21018	5694	514	34918	2.7	0.51	14.8	1.31	0.65	7.34
2013	277	37.4	837	3188	1006	52422	1557	995	28427	3.95	0.42	30.6	1.12	0.46	5.19
2014	51.2	19.5	377	5941	467	35098	2469	132	15240	1.17	0.14	4.55	0.89	0.11	4.82
2015	110	38.8	262	6966	737	41283	4084	266	11482	0.96	0.72	18.2	0.8	0.21	4.06
2016	29.4	11.1	442	478	27.5	9091	891	28.1	14605	0.28	0.1	35.6	0.29	0.11	2.21
2017	35.5	20	686	434	104	22963	994	127	12740	0.33	0.1	64.8	0.8	0.15	3.19
2018	45.9	14.8	80.2	323	53	1852	584	133	4108	0.54	0.1	2.83	0.29	0.11	5.7

*All compounds are expressed in ng/g lipid weight. SCHL= trans-nonachlor, oxychlordane; SPCB= PCB-118, -138, -153, -180. Note=

data from 1997-2013 has been published by Andersen et al., 2015.

Table 2. Averaged Estimates derived from additive models with 95% Confidence Intervals (In Brackets) Explaining the log-Transformed concentrations of POPs (ng/g Lipid Weight) in arctic foxes from Svalbard, Norway, by Feeding Habits (δ^{13}C and δ^{15}N), Fatindex, Carcasses, Cumulative ice, Geese, POPs air.

Response	(Intercept)	δ^{13}C	δ^{15}N	Fatindex	Carcasses	Cumulative ice	Geese	POPs air
log(HCB)	5.2 (4.87, 5.54)	**0.42 (0.30, 0.54)**	**0.37 (0.25, 0.50)**	**-0.22 (-0.34, -0.10)**	**-0.14 (-0.27, -0.01)**	**0.25 (0.07, 0.43)**	-0.6 (-2.49, 1.29)	-0.25 (-0.68, 0.17)
log(BDE153)	1.04 (0.65, 1.44)	**0.25 (0.11, 0.39)**	**0.20 (0.06, 0.35)**	**-0.29 (-0.43, -0.15)**	0.00 (-0.12, 0.12)	0.02 (-0.12, 0.15)	0.31 (-0.01, 0.64)	NA
log(ΣPCB)	8.39 (7.79, 9.00)	**0.67 (0.49, 0.86)**	**0.57 (0.37, 0.77)**	**-0.28 (-0.46, -0.09)**	-0.07 (-0.23, 0.10)	-0.00 (-0.19, 0.19)	0.70 (-0.16, 1.57)	-0.01 (-0.26, 0.24)
log(BDE47)	0.89 (0.46, 1.32)	**0.70 (0.51, 0.90)**	**0.75 (0.55, 0.94)**	0.04 (-0.16, 0.23)	-0.14 (-0.31, 0.04)	0.10 (-0.08, 0.29)	**0.60 (0.13, 1.06)**	NA
log(ΣCHL)	8.81 (8.35, 9.27)	**0.54 (0.37, 0.70)**	**0.37 (0.20, 0.55)**	**-0.35 (-0.52, -0.18)**	-0.13 (-0.31, 0.06)	0.17 (-0.07, 0.41)	1.57 (-1.17, 4.32)	0.05 (-0.15, 0.25)

*ΣPCB= -118, -138, -153, -180; ΣCHL= Oxychlordane, Trans-nonachlor; NA= Non Available; values in bold are significant

The best models for ΣPCB, HCB, ΣCHL and BDE-153 included $\delta^{13}C$ and fat index as predictors in addition to sea ice extent, reindeer carcass availability, POPs in the air and goose availability, whereas the model for BDE-47 included goose availability and $\delta^{15}N$. Model averaged estimates (Table 2) showed that contaminant concentrations in arctic fox liver increased significantly with increasing trophic position (i.e., $\delta^{15}N$) and increasing values from $\delta^{13}C$ (ratio between $^{12}C/^{13}C$ determining diet on being terrestrial or marine), suggesting a marine diet. The increase of BDE-47 with increasing trophic position was greater than the increase of BDE-153 with increasing trophic position. Apart from BDE-47, all contaminants were lower in fat than lean individuals.

The availability of reindeer carcasses was negatively related to HCB concentrations in arctic foxes. Also, BDE-47 and ΣCHL concentrations tended to decrease non-significantly with reindeer carcasses availability. HCB concentrations increased significantly with increasing sea ice extent; it is worth to note that ΣCHL tended to increase non-significantly when the sea ice extent increased. BDE-47 concentrations increased significantly with increasing goose availability. The effect of POPs in the air had no effect on HCB, ΣCHL and ΣPCB concentrations, no estimates were available for BDE-47 and -153.

Changes in POPs concentrations over time (Figure 2) were estimated using the best models from our candidate model list (Appendix 2) and compared to non-adjusted concentrations. Concentrations of BDE-47 corrected for $\delta^{15}N$ and goose availability decreased linearly over the study period -17.7%/year (95% CIs= [-22.8, -12.3]), being our highest decrease rate per year within all the contaminants. Measured concentrations showed an overall decline for the study period with a decrease of -11.6%/year (-13.9, -9.33).

Changes in concentrations for BDE-153 showed a linear decrease for both the adjusted (i.e., $\delta^{13}C$ and fat index) and the measured concentrations, with -10.9%/year (-12.3, -9.43) and -11.8%/year (-13.3, -10.2), respectively. The overall decline for both adjusted and measured concentrations for HCB was the lesser of all the contaminants, showing a decrease of -2.21%/year (-4.00, -0.39) and -4.27%/year (-6.16, -2.35), respectively.
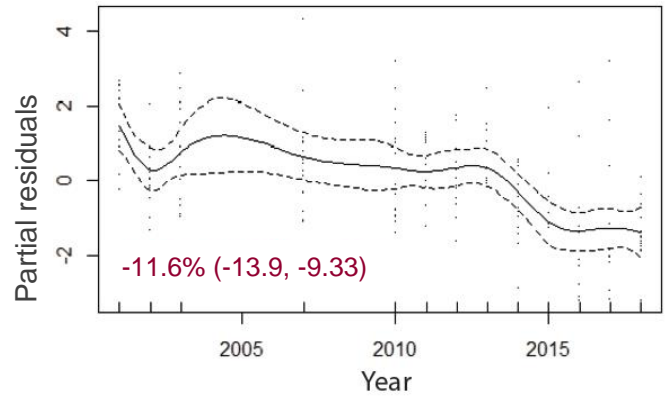
The adjusted concentrations for chlordanes showed an overall decline of -12.9%/year (-18.8, -6.52). Similarly, measured concentrations showed an overall decline of -11.1%/year (-13.5, -8.55). PCBs showed a linear decrease for adjusted concentrations -14.5%/year (-19.3, -9.39),

the overall decline for measured concentrations over the study period was -11.2%/year (CIs=-13.4, -8.95).
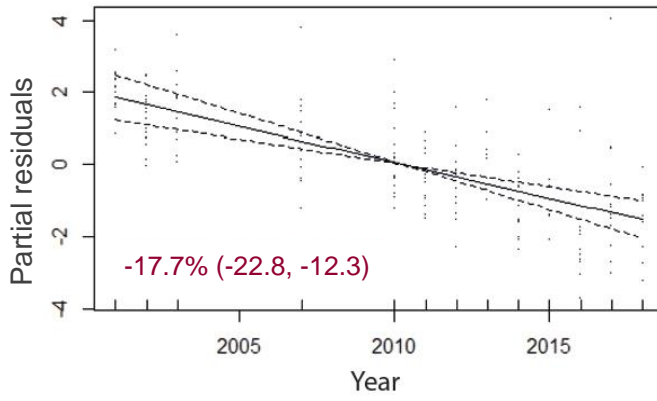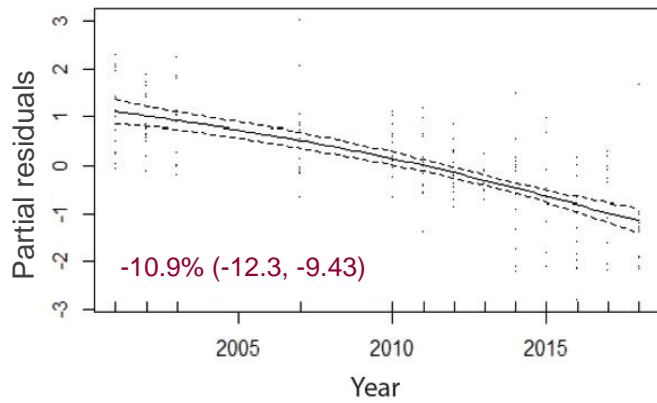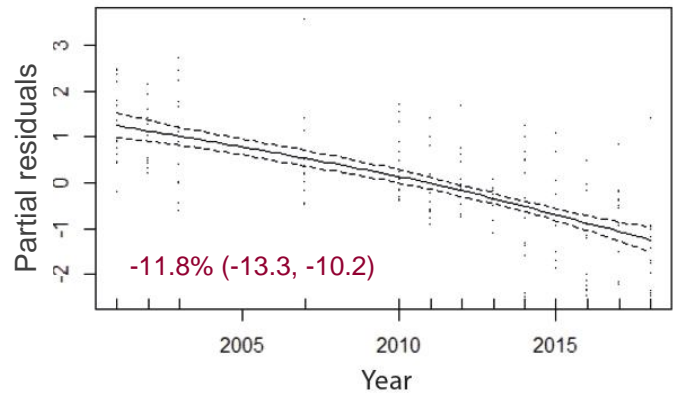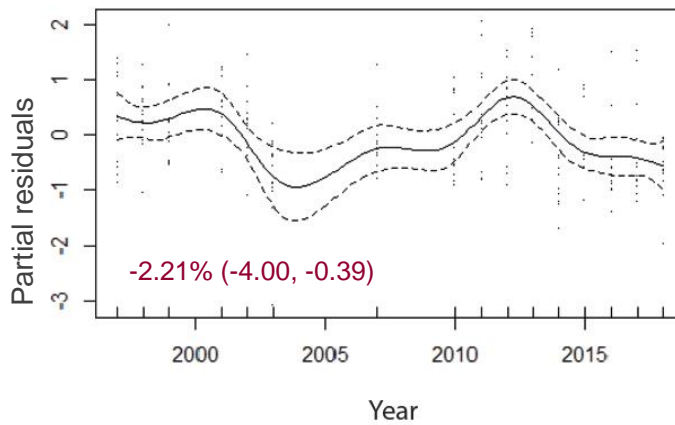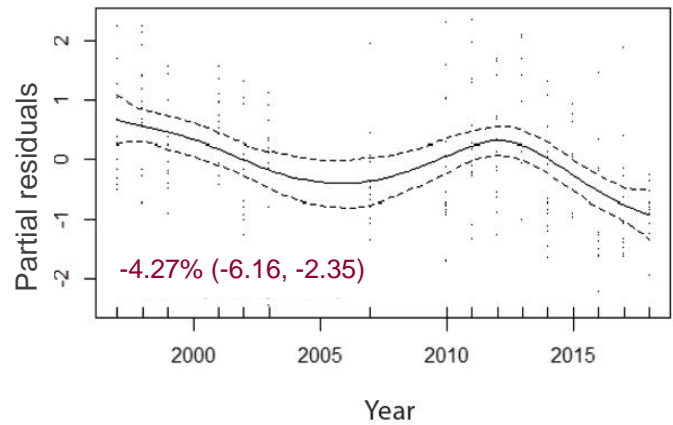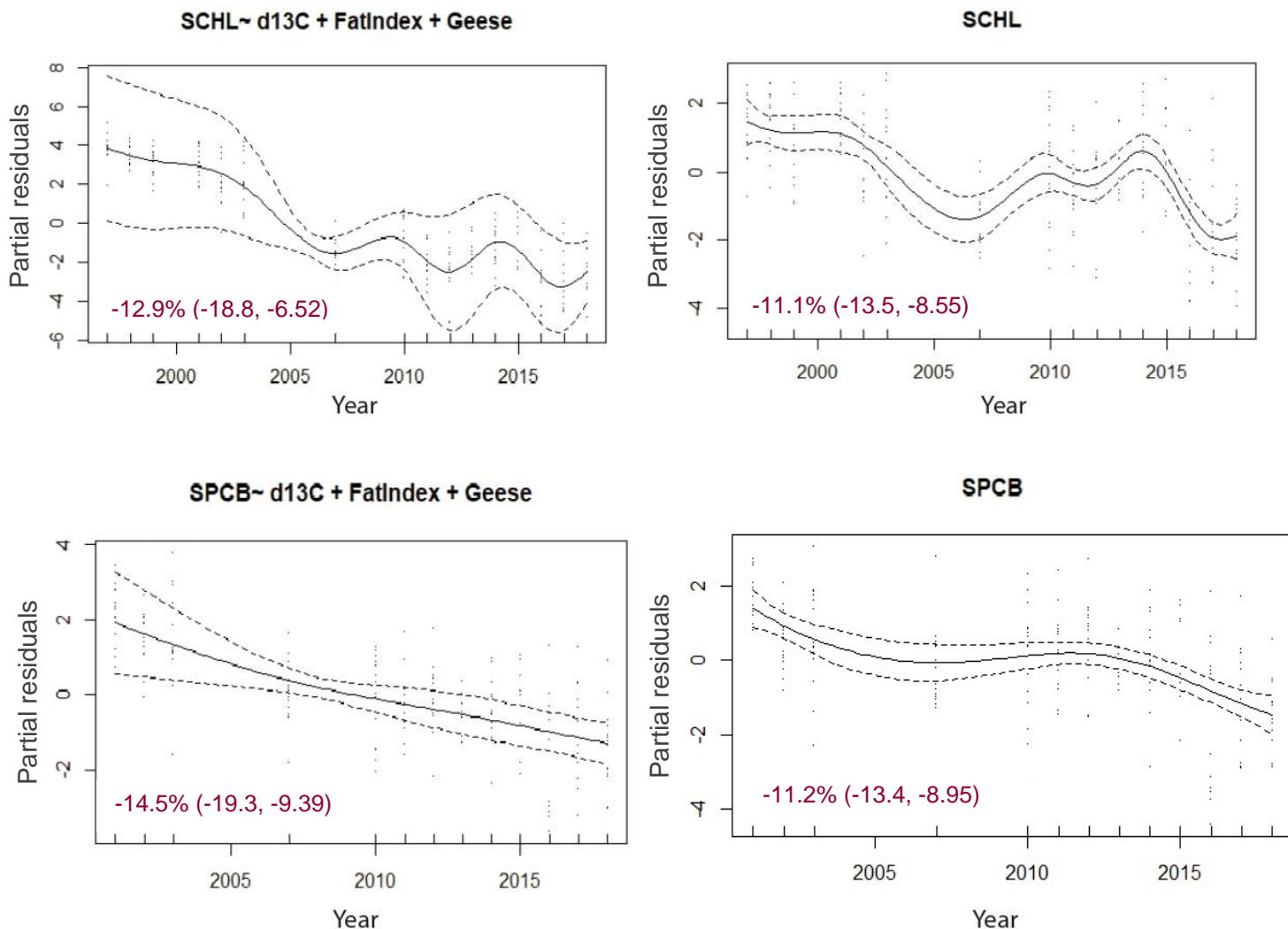
Figure 2. Temporal changes in concentrations of BDE-47, BDE-153, HCB, ∑CHL and ∑PCBs in arctic fox liver from Svalbard, from 1997 to 2019. Changes in contaminant concentrations were calculated using additive models adjusted for variables (left) and compared to measured concentrations (right). The *y*-axes show partial residuals of the highest ranked additive models. Yearly change (%) and confidence intervals were derived from linear models using the best ranked model and POPs concentrations against year.

4. **Discussion**

Previous studies have indicated that Arctic wildlife has reached levels of POPs exceeding the threshold for adverse effects (Dietz et al., 2019), therefore comparing levels in liver from threshold levels with the individuals in this study could indicate the association between those levels and potential health effects. Pedersen et al., 2015 presented a review for potential health effects in arctic foxes with only legacy POPs since there is no information on effect of new POPs in arctic foxes.

Median concentrations (Table 2) showed that ΣPCB for all seasons (584-16327 ng/g lipid weight) and ΣCHL (323-15523 ng/g lipid weight) were the contaminants with the highest concentrations among the others. As reported by Pedersen et al., 2015, levels in liver (ΣPCB= 2065 ng/g lipid weight) associated with adverse health effects from domesticated arctic foxes were lower than our highest median concentrations, being 12% from our highest concentration.

An increase in liver, kidney and thyroid gland lesions has been reported when exposed to above threshold concentrations of these contaminants, however, only kidney lesions have showed a significant correlation with high exposure to OCs (organohalogenated compounds). The last trapping season from this study showed a decrease in median concentrations for ΣPCB (584 ng/g lipid weight), being the lowest of all the study years in Svalbard. It is worth to mention that concentrations of ΣPCB in arctic foxes were above the threshold for most years.

Changes in concentrations of contaminants through the whole study period suggest that the risk for adverse health effects for arctic foxes might decrease in the future due to decreasing concentrations found in liver, however, differences might be present between wild and domesticated foxes. It could be possible that wild foxes might be less susceptible to contaminant exposure due to adaptation to increasing levels over time (Pedersen et al., 2015).

When addressing the relationships between the contaminants levels and covariates we found an increase of contaminants in foxes eating food with a higher trophic position and intake of marine diet, consistent with those reported previously by Fuglei et al., 2007 and Andersen et al., 2015 from Svalbard foxes and Bolton et al., 2017 from Pribilof Islands foxes in Alaska.

The increase of BDE-47 was greater than BDE-153 with increasing trophic position, suggesting that foxes feeding from prey with higher trophic position have higher concentrations of this contaminant.

Concentrations of all contaminants except BDE-47 decreased with fat index (i.e., from 1-4, lean to fat), consistent with previous results from Wang-Andersen et al., 1993 and Fuglei et al., 2007. During periods of starvation lipophilic contaminants may become bioavailable due to adipose tissue remobilization and increase the input of pollutants in vital organs such as liver (Fuglei et al., 2007, Helgason et al., 2013).

Concentrations for HCB decreased when reindeer carcasses were abundant, suggesting that years with high reindeer mortality could have an impact on the concentrations of this contaminant in arctic foxes from Svalbard, as also noted by Andersen et al., 2015, who analyzed the same arctic fox data for the time period of 1997 to 2014. Since populations of Svalbard reindeer are constrained due to rain-on-snow events, which encapsulate forage plants making them inaccessible (Peeters et al., 2019), we hypothesize that HCB concentrations could decrease in the future in arctic fox liver since Svalbard reindeers do not show high levels of such contaminants, as reported by Melien, 2014.

Concentrations of HCB in arctic foxes showed a significant increasing relationship with sea ice extent. Also, ΣCHL tended to increase with increasing sea ice extent. Previously reported relationships were not significant between sea ice extent and HCB concentrations in arctic foxes, however, β-HCH was reported to have a positive relationship with increasing sea ice extent (Andersen et al., 2015).

The Isfjorden area has had two periods with relatively low sea ice cover, 2006 and 2012 (Muckenhuber et al., 2016), concluding that since 2006 there is a general trend for less extensive sea ice, therefore we could expect that chances of survival rate in ringed seal pups decline with decadent conditions of proper cover on the surface of the sea ice, however, a recent study concludes that demographic parameters for ringed seals have remained unchanged despite the environmental changes and due to the possibility of young seals moving to the west fjords from east, where they concentrate in small areas of available sea ice (Andersen et al., 2021).

Hence, we suggest that due to the unchanged demography, access to ringed seals pups could increase levels of HCB in arctic fox liver, even if the relationship estimate between increasing concentrations of contaminants and increasing sea ice extent is not too high. Although, low levels of HCB in ringed seals have been reported in a time series from 1992 to 2014 (MOSJ, 2021).

Goose availability (i.e., capture- mark and resight counts) and POPs in the air from Svalbard (i.e., air samples) have not been taken into consideration before as explanatory covariates for pollutant concentrations in arctic foxes. In this study increasing concentrations of BDE-47 were positively related to increasing availability of goose as prey. This pollutant was detected at low levels in arctic fox (Table 2).

Studies have reported contaminant concentrations in different waterfowl birds but to our knowledge none on PBDEs in pink footed geese. Previous publications have reported low pollutant concentrations (ΣPCB= 1.23 ± 0.80 ng/g w.w.) in eggs from barnacle geese (*Branta leucopsis*) from a colony in Svalbard (Hitchcock et al., 2019), still no concentrations were reported for brominated pollutants that could explain the positive relationship found in this study. Increasing concentrations of BDE-47 in waterfowl birds could be related to feeding in wintering grounds, where local contamination sources are important for PCBs, OCPs and PBDEs concentrations in eggs from great tit (*Parus major*) in Belgium, as suggested by Van den Steen et al., 2007.

No significant relationships were established between pollutants in the air and the concentrations of these in arctic fox livers. This evidence could show progress achieved as a result of national and international control measures applied before and since the establishment of the Stockholm Convention (Hung et al., 2016) and possibly the influence of other factors in POPs levels in arctic foxes besides pollutants in the air. Results from the air monitoring in Zeppelin show that most legacy POPs in air are declining or have stabilized. PCBs concentrations in air had the lowest ever observed in 2018, showing lower concentrations that those reported in other European countries with consistent low concentrations in summertime than wintertime, as well, chlordanes had the lowest mean of concentrations in the same year without any seasonality, finally HCB has been reported to increase ten years ago but currently on the decline (NILU, 2019), meaning that primary

emissions stopped, and that long term slow decline is controlled by degradation rates in secondary repositories (Stroebe et al., 2004).

Changes in concentrations for contaminants showed a general decline through the whole study period, following the decrease of PCBs and organochlorine pesticides in Arctic biota during the last 20 to 30 years (Rigét et al., 2019). These declining changes follow our expectations since all the contaminants analyzed are currently banned, though some of them were regulated not so long ago and added to the Stockholm Convention (http://chm.pops.int). However, the decrease in Arctic biota for HCB has been slow, as our estimate showed, due to emissions and releases that may continue, supported by increasing concentrations in Arctic air (Hung et al., 2016). All our estimated changes per year for adjusted and measured concentrations were similar among contaminants, with the exception for BDE-47 which showed an increased declining rate of change per year when adjusted for covariates, possibly due to low levels of this contaminant in goose and their position in the food web. Also, all our adjusted and measured estimates had confidence intervals overlapping, additionally, HCB changes per year were slightly slower when adjusted for covariates, meaning that changes in diet and food availability in arctic foxes could affect the temporal changes of contaminants, a reason that could also explain why ΣPCB concentrations decrease faster per year when adjusted for covariates.

The shift in diet towards lower trophic levels and less marine diet could significantly affect contaminant changes in future time series. As well, impact of climate change either directly (e.g., increased volatilization from both primary and secondary sources) or indirectly (e.g., change in Arctic land use and emission patterns, increase in mining/shipping in the North) (UNEP/AMAP, 2011) could determine the direction of pollutant patterns in the future.

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Appendix 1

Trap Season	n	Sex		Body condition				Age	
		F	M	1	2	3	4	1y	2y
1997-1998	12	6	6	2	4	4	2	10	2
1998-1999	14	4	10	3	4	5	2	8	6
1999-2000	14	9	5	3	3	5	3	6	8
2001-2002	14	5	9	3	4	4	3	9	5
2002-2003	14	7	7	3	5	4	2	8	6
2003-2004	14	8	6	3	3	7	1	7	7
2007-2008	14	9	5	0	2	10	2	9	5
2010-2011	16	8	8	4	4	4	4	11	5
2011-2012	14	8	6	0	7	4	3	11	3
2012-2013	13	5	8	2	5	5	1	9	4
2013-2014	12	5	7	1	3	7	1	12	0
2014-2015	14	6	8	3	5	3	3	5	9
2015-2016	8	2	6	0	4	4	0	1	7
2016-2017	12	4	8	1	5	4	2	11	1
2017-2018	12	4	8	3	3	4	2	10	2
2018-2019	12	6	6	1	2	8	1	9	3
Totals (n)	209	96	113	32	63	82	32	136	73

Appendix 1. Annual harvest, sample collection.

Note: data from 1997-2013 has been published by Andersen et al., 2015.

Appendix 2

Table. GAMM Model Selection Table Explaining the POPs Concentrations (ng/g Lipid Weight) In Liver of Arctic Foxes From Svalbard by Feeding Habits (δ^{13}C And δ^{15}N) POPs in Air, Fatindex, Carcasses, Cumulative ice and Geese.

SPCB

(Intercept)	s(Year)	SPCB_air	d13C	Fatindex	Carc	cum_ice	geese	d15N	df	logLik	AIC	delta	weight
8.30	+		0.67	-0.28			0.69		6	-248.09	510.08	0.00	0.24
8.50	+		0.68	-0.27					7	-248.31	510.76	0.68	0.17
8.32	+		0.67	-0.29	-0.06		0.69		8	-247.68	511.47	1.39	0.12
8.53	+		0.66	-0.28	-0.07				8	-247.69	511.71	1.63	0.11
8.50	+	0.02	0.67	-0.27					8	-248.04	512.38	2.30	0.08

HCB

(Intercept)	s(Year)	HCB_air	d13C	Fatindex	Carc	cum_ice	geese	d15N	df	logLik	AIC	delta	weight
5.20	+		0.42	-0.22	-0.14	0.25			14	-242.07	513.05	0.00	0.32
5.21	+		0.42	-0.22	-0.16	0.27	-0.67		15	-241.28	513.83	0.77	0.22
5.21	+	-0.26	0.41	-0.22	-0.13	0.21	NA		15	-242.05	514.12	1.06	0.19
5.21	+	-0.23	0.41	-0.22	-0.14	0.24	-0.62		16	-240.99	514.99	1.93	0.12
5.17	+		0.43	-0.21		0.27			13	-244.63	516.04	2.99	0.07

SCHL

(Intercept)	s(Year)	SCHL_air	d13C	Fatindex	Carc	cum_ice	geese	d15N	df	logLik	AIC	delta	weight
8.79	+		0.54	-0.34			1.81		13.00	-312.42	652.48	0.00	0.15
8.83	+		0.53	-0.36	-0.14				13.00	-312.74	652.67	0.20	0.13
8.81	+		0.54	-0.35	-0.13	0.16			14.00	-312.21	653.07	0.59	0.11
8.82	+		0.53	-0.35	-0.12		1.21		14.00	-311.80	653.10	0.62	0.11
8.79	+		0.55	-0.34		0.18			13.00	-313.26	653.23	0.75	0.10

BDE47

(Intercept)	s(Year)	d13C	Fatindex	Carc	cum_ice	geese	d15N	df	logLik	AIC	delta	weight
0.82	"+"					0.55	0.74	5.00	-256.76	523.53	0.00	0.15
1.01	"+"			-0.12			0.73	5.00	-256.25	524.44	0.92	0.10
1.00	"+"						0.76	4.00	-257.31	524.46	0.93	0.10
1.03	"+"	0.69		-0.17				6.00	-256.12	524.66	1.13	0.09
0.75	"+"		0.03			0.55	0.74	6.00	-256.72	525.43	1.91	0.06

BDE153

(Intercept)	s(Year)	d13C	Fatindex	Carc	cum_ice	geese	d15N	df	logLik	AIC	delta	weight
1.08	+	0.25	-0.29					5.00	-202.44	416.56	0.00	0.29
0.98	+	0.24	-0.29			0.31		6.00	-202.44	416.89	0.33	0.24
1.08	+	0.25	-0.29		0.01			6.00	-202.43	418.51	1.96	0.11
1.08	+	0.25	-0.29	0.00				6.00	-202.43	418.55	1.99	0.11
0.98	+	0.24	-0.29	0.00		0.31		7.00	-202.44	418.89	2.33	0.09

*The plus sign indicates that a smoothed variable was included in the model s(Year): smoothed factor YEAR.

* Model selection tables for BD47 and BDE153 didn't include data for POPs in air.

Appendix 3

2013 POPs	LOD (ng/g)	Recovery percentage	2014 POPs	LOD (ng/g)	Recovery percentage	2020 POPs	LOD (ng/g)	Recovery percentage
HCB	0.125	113	HCB	0.035	124	HCB	0.003	73
β-HCH	0.489	118	β-HCH	0.130	111	β-HCH**	0.010	163
Oxyclordane	0.188	88	Oxyclordane	0.064	71	Oxychlordane	0.012	131
cis-chlordane*	1.861	115	cis-chlordane**	0.058	99			
trans-nonachlor	0.202	89	trans-nonachlor	0.058	74	trans-nonachlor	0.003	96
Mirex	0.192	119	Mirex	0.040	112	Mirex	0.017	106
p,p'-DDE	0.349	93	p,p'-DDE	0.092	100	p,p'-DDE	0.049	137
p,p'-DDT*	4.490	106	p,p'-DDT*	0.088	35	p,p'-DDT**	0.028	367
PCB-28*	2.724	120	PCB-28**	0.156	122	PCB-28	0.006	97
PCB-52*	2.085	128	PCB-52*	0.068	127	PCB-52	0.058	92
PCB-101*	3.439	110	PCB-101*	0.174	117	PCB-101	0.012	120
PCB-118	0.292	125	PCB-118	0.120	115	PCB-118	0.004	98
PCB-138	0.220	116	PCB-138	0.089	114	PCB-138	0.004	101
PCB-153	0.300	116	PCB-153	0.126	120	PCB-153	0.004	86
PCB-180	0.186	107	PCB-180	0.074	107	PCB-180	0.003	93
BDE-47	0.050	107	BDE-47	0.035	99	BDE-47	0.005	80
BDE-99	0.050	124	BDE-99	0.030	101	BDE-99	0.007	95
BDE-100	0.050	127	BDE-100	0.030	103	BDE-100	0.007	90
BDE-153	0.050	110	BDE-153	0.035	98	BDE-153	0.005	85
BDE-154**		115	BDE-154*		97	BDE-154	0.006	81
BDE-209	3.25**	112	BDE-209**	0.500	108	BDE-209	0.042	108
HBCDD	0.500	100	HBCDD***	0.400	322	HBCDD (hexabromcyclo dodecan)	0.143	78

2013: *Due to interferences these analytes could not be quantified.

**For BDE-209 a detection limit equal to the average blank +2* standard deviation has been used, instead of 3* noise.

2014: *Due to interferences these analytes could not be quantified.

**In the series analyzed in 2013, PCB-28 and cis-chlordane could not be analyzed due to interference. Conditions were better for tests this year.

***HBCDD is corrected for recovery percentage.

**For BDE-209 a detection limit equal to the average blank +2* standard deviation has been used, instead of 3* noise.

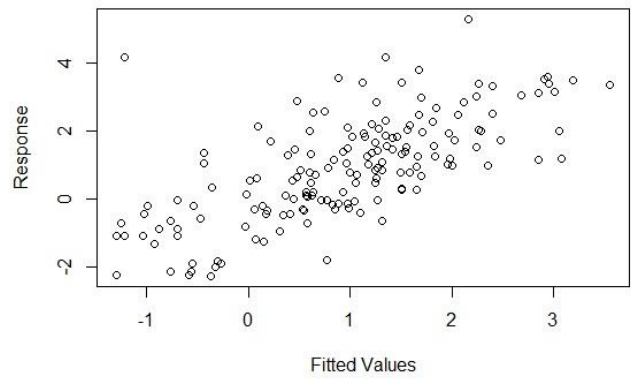2020: **Corrected for recovery percentage.

Appendix 4

BDE-47

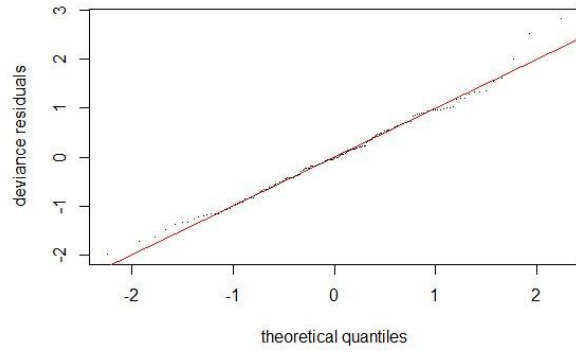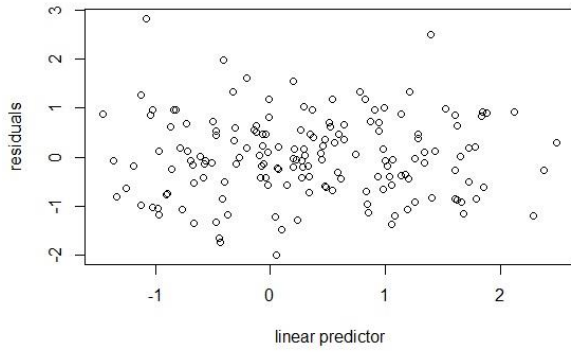**Resids vs. linear pred.**
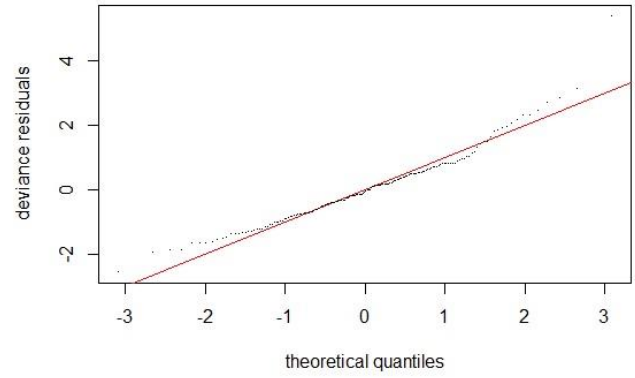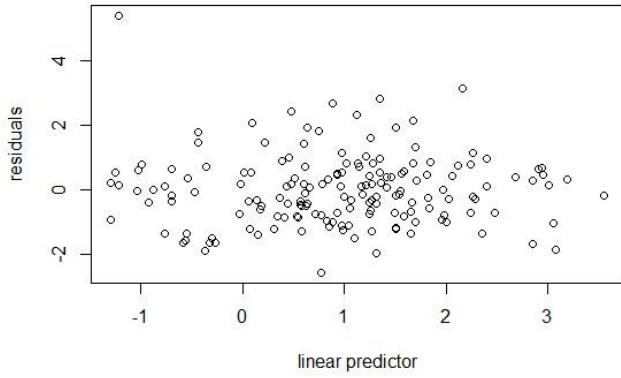
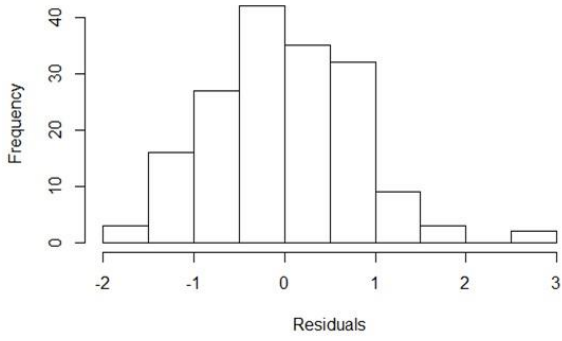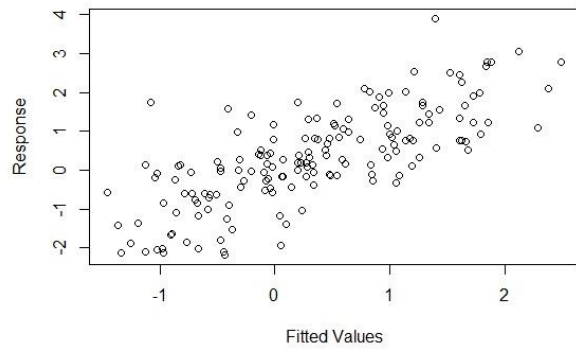**Histogram of residuals**
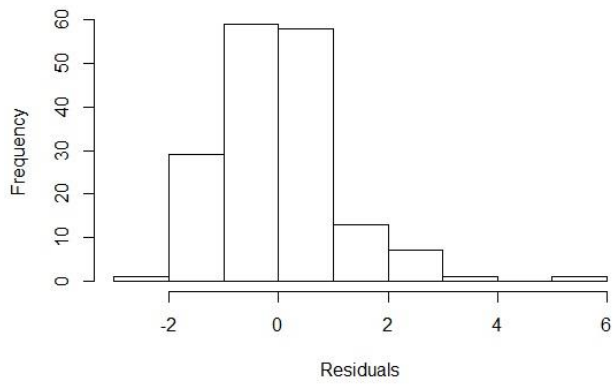
Response vs. Fitted Values

Appendix 4 continued

BDE-153

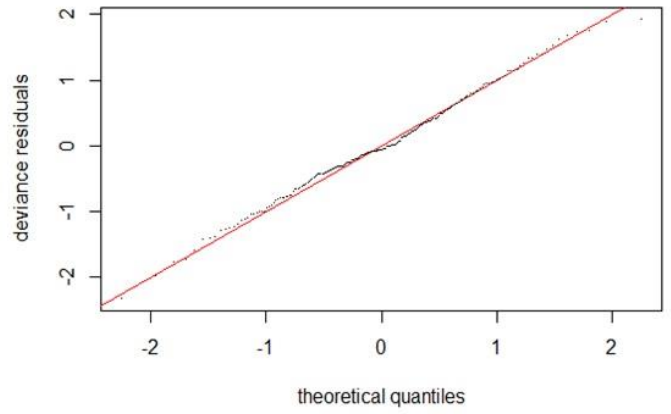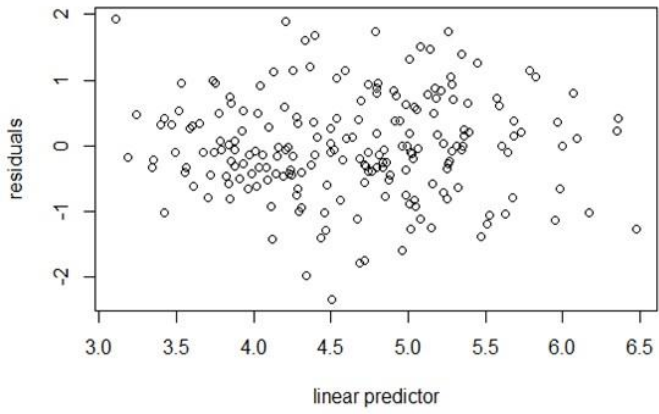**Resids vs. linear pred.**

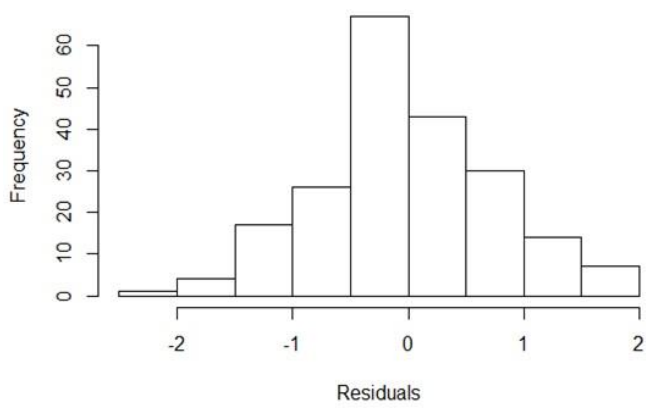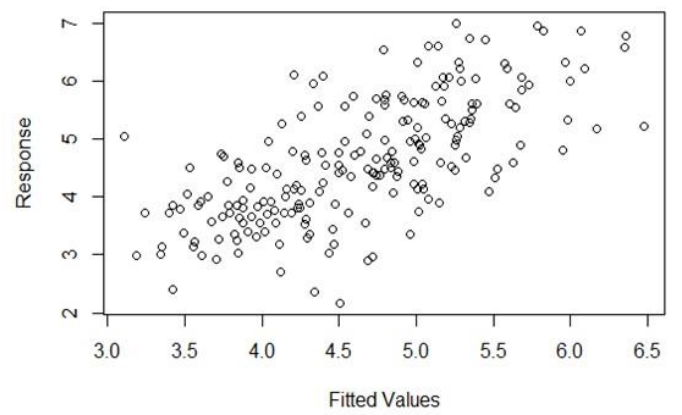**Histogram of residuals**

Response vs. Fitted Values

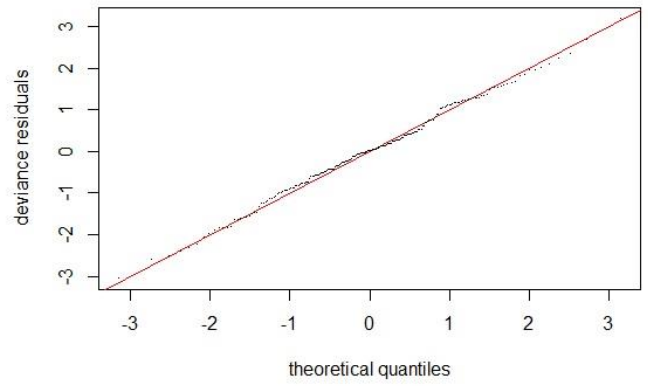Appendix 4 continued

HCB

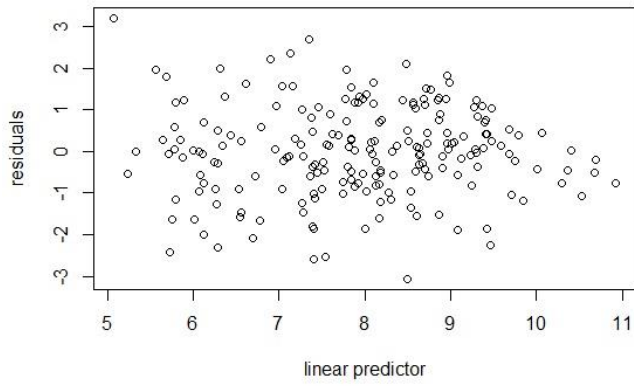**Resids vs. linear pred.**

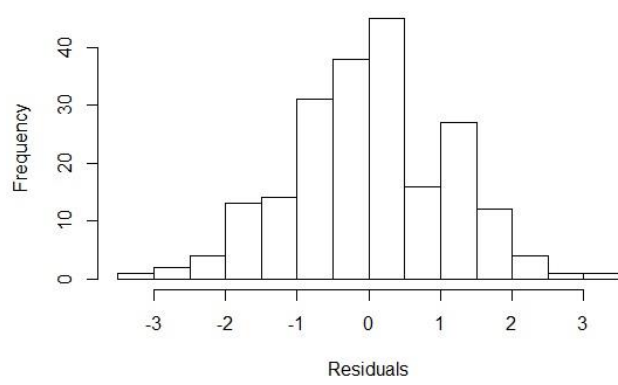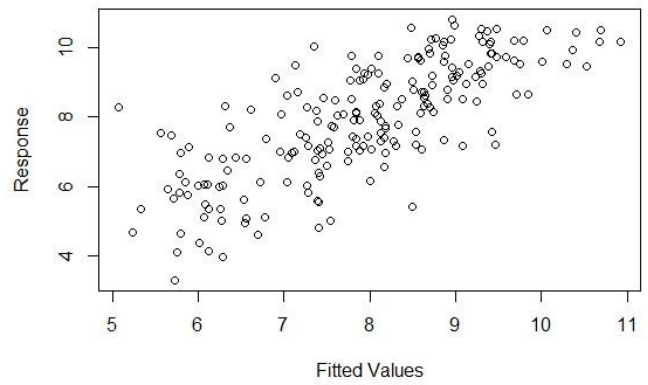**Histogram of residuals**

Response vs. Fitted Values

Appendix 4 continued

ΣCHL

Resids vs. linear pred.

Histogram of residuals
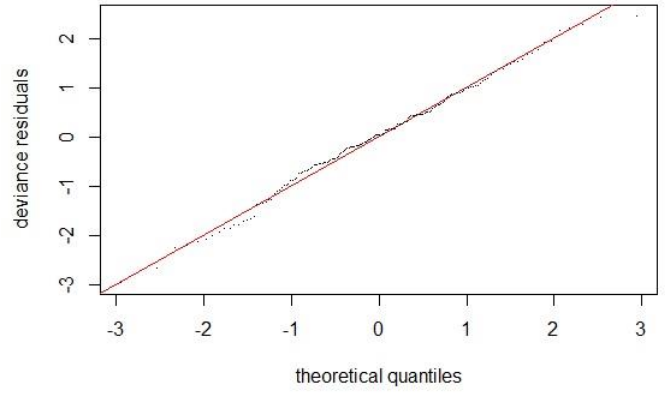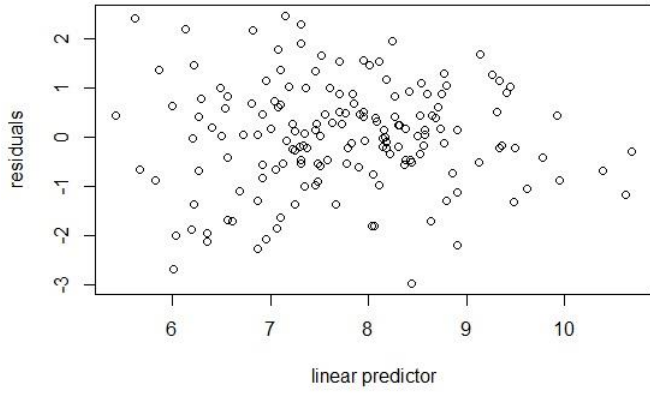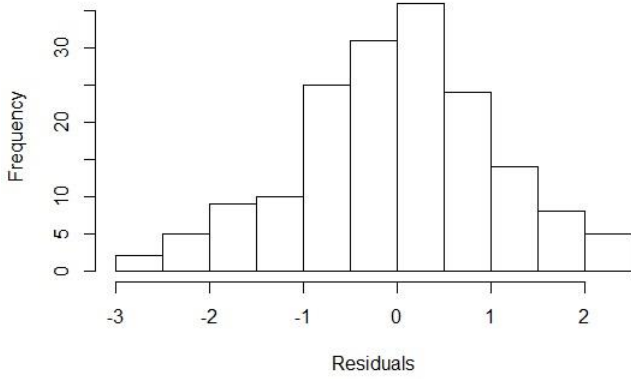

Response vs. Fitted Values
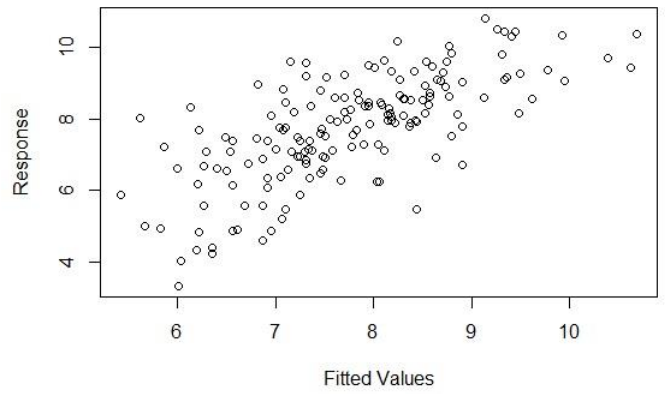
Appendix 4 continued

ΣPCB

Resids vs. linear pred.

Histogram of residuals

Response vs. Fitted Values

Appendix 5

Mean concentrations of contaminants in Arctic foxes collected in Svalbard, Norway from 1997 to 2019 in ng/g wet weight.					
Year	HCB	ΣCHL	ΣPCB	BDE47	BDE153
1997	17.1	846	1020	1.97	0.56
1998	26.8	1223	1483	1.91	0.66
1999	11.3	821	926	1.01	0.44
2001	17.3	1242	1350	1.37	0.55
2002	7.5	417	379	0.29	0.35
2003	11.1	813	787	1.13	0.52
2007	5.26	53.6	217	0.89	0.25
2010	12.1	546	379	0.77	0.17
2011	18.1	188	375	0.29	0.13
2012	14	320	581	0.29	0.14
2013	22.5	237	117	0.54	0.05
2014	6.49	589	249	0.1	0.08
2015	8.34	754	329	0.2	0.09
2016	5.91	109	193	0.32	0.04
2017	6.66	194	139	0.42	0.05
2018	2.49	30.1	52.3	0.04	0.04

*ΣPCB= PCB-118, -138, -153, -180 ΣCHL=trans-nonachlor, oxychlordane