UNIVERSITY OF BERGEN

Is a warmer Arctic a more contaminated Arctic?

Atlantification of Kongsfjorden in relation to contaminant levels in seabirds

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Abstract

The Arctic, and Svalbard in particular, is experiencing a more rapid warming compared to the global average. Such Atlantification of the Arctic may change food webs in a way that can also affect the contaminant levels in top predators such as seabirds. Studies have found that the black-legged kittiwake (*Rissa tridactyla*) in Kongsfjorden, Svalbard, has changed its diet from mainly Arctic prey items towards a more mixed diet with contribution from Atlantic species since 2007. Atlantic species might function as biovectors, bringing contaminants into the Arctic from more contaminated areas. However, Arctic species might have a higher contaminant load, due their high lipid content. My thesis explored if changes observed in dietary ecology, using two different approaches, can explain variability in black-legged kittiwake exposure to organochlorine contaminants during the years 2007-2020, here represented by seven compounds including polychlorinated biphenyl (PCB) 99, PCB 153, PCB 180, *β*-hexachlorocyclohexane, hexachlorobenzene, *p,p'*-dichlorodiphenyldichloroethylene (DDE), and oxychlordane. Dietary ecology was quantified by both frequency of occurrence of diet items and groups from regurgitate samples and by stable isotope analysis. Stable nitrogen isotope values (δ^{15}N) have been established as a proxy for trophic position, and stable carbon isotope values (δ^{13}C) indicate foraging habitat and primary carbon source. Annual variation in frequency of occurrence of prey species (or groups) did not explain the variation in either δ^{13}C or δ^{15}N values. There were significant differences in the annual variations in contaminants levels, but there was no clear temporal trend for any contaminants during the study period. Model selection showed that neither diet items nor stable isotope values explained the variation in contaminant levels. Instead, the null-model, with *year* as a random effect factor, was often ranked as a strong model. However, some variables, such as trophic level, carbon source and frequency of occurrence of herring, as a representative for Atlantic species, seem to have a possible relationship to contaminant exposure. Neither the claim that Atlantic species function as biovectors, nor that Arctic species have high contaminant load due to high lipid content, is supported by this thesis, as there was no clear relationship between variation in contaminant levels and the degree of Arctic or Atlantic prey species. The Atlantification and the current climate warming might affect contaminant levels in black-legged kittiwake in Kongsfjorden in other ways not studied in this thesis, such as through reduced body condition or though changes in the physical environment.

1. Introduction

The Arctic is experiencing a more rapid warming compared to the global average, and annual mean surface temperatures (land and ocean) has been measured to change three times faster in the Arctic compared to the global average (AMAP, 2021; Descamps et al., 2017). This rapid temperature change causes pronounced declines in sea-ice, in its turn affecting many of the resident and migratory Arctic species (Box et al., 2019; Descamps et al., 2017; Descamps & Strøm, 2021; Eamer et al., 2013). Among the observed impacts are the altered distribution and abundance of species (Eamer et al., 2013) as the reduction in sea ice extent leads to loss of habitat for a variety of Arctic species (Box et al., 2019; Eamer et al., 2013; Jenssen et al., 2015; Kovacs et al., 2011). In addition to such climate stress, the Arctic is considered a sink for anthropogenic contaminants, even though remote from production and consumption areas (Burkow & Kallenborn, 2000; Dietz et al., 2019). Contaminants are transported by long-range transport in the atmosphere, by riverine input and ocean currents from lower latitude industrial source areas (AMAP, 2016; Burkow & Kallenborn, 2000), and ultimately end up in Arctic wildlife (Gabrielsen, 2007; Jenssen, 2006). Altogether, rapid environmental change is observed to cause changes in food web structure and this might also lead to changes in biotic pathways of environmental contaminants such as persistent organic pollutants (POPs; Carrie et al., 2010; Descamps et al., 2017; Kovacs et al., 2011; Macdonald et al., 2003; Noyes et al., 2009).

POPs are a diverse group of compounds with agricultural and industrial origin (Gabrielsen, 2007). POPs are legacy pollutants that have been banned or regulated decades ago and are almost ubiquitously regulated under the Stockholm Convention for POPs since 17th May 2004 (Stockholm Convention, 2022). As a result, there has been a decrease in many of these POPs in the Arctic, while at the same time they may still reach levels considered harmful for both humans and wildlife (AMAP, 2016; Dietz et al., 2019; Henriksen et al., 2001; Rigét et al., 2010). There is a large body of evidence that supports POPs to pose a threat to exposed wildlife (Dietz et al., 2019; Letcher et al., 2010), and Arctic predatory marine species are of special concern because of a combination of factors (Borgå et al., 2001; Gabrielsen, 2007). As an adaptation to seasonality and large annual fluctuations in productivity, many Arctic marine organisms have annual cycles and life strategies that involve large lipid stores (Borgå et al., 2001; Gabrielsen, 2009; Varpe, 2017). POPs are lipid soluble and can therefore enter and biomagnify through the food chain, and bioaccumulate in large amounts in long-lived species (Guzzo et al., 2014; Hop et al., 2002). As a consequence, high levels of POPs have been measured in long-lived high-trophic species, such Arctic seals (Gabrielsen, 2007), Arctic fox (*Vulpes lagopus;* Andersen et al., 2015), polar bear (*Ursus maritimus;* Gabrielsen, 2007) and Arctic seabirds, such as glaucous gulls (*Larus hyperboreus;* Gabrielsen et al., 1995), great skua (*Stercorarius skua;* Bourgeon et al., 2012) and the black-legged kittiwake (*Rissa tridactyla*; Gabrielsen, 2007).

The black-legged kittiwake (hereafter referred to as kittiwake) has been shown to function as a messenger for the ecological effects of climate change (Vihtakari et al., 2018). A shift in prey species was observed for kittiwakes in Kongsfjorden (Svalbard, Norway) from primarily Arctic species up to 2006 to an increasing contribution of Atlantic fish species. This change in diet composition was observed to align with changes in sea-ice distribution and sea surface temperatures, altogether indicating the Atlantification of this high-Arctic fjord system (Pavlova et al., 2019; Renaud et al., 2012; Vihtakari et al., 2018). Boreal fish species are expanding northwards, while Arctic species retract even further north (Fossheim et al., 2015). Other observations include a decline in Arctic zooplankton in the Barents sea region (Dalpadado et al., 2012), which has consequences for food chain lipid dynamics when less lipid-rich Atlantic species prevail (Falk-Petersen et al., 2009; Fossheim et al., 2015). Such shifts in the composition of both the zooplankton and fish community might have ecotoxicological consequences for species further up the food chain depending on them. A study from the Canadian Arctic showed that the boreal species capelin (*Mallotus villosus)* exhibited higher levels of POPs than native fish species, possibly related to capelin migration to temperate regions, alongside with potential differences in trophic position, size, lipid content and feeding habitat (Pedro et al., 2017). A study from Svalbard, comparing zooplankton from an Atlantic influenced fjord (Kongsfjorden) and an Arctic influenced fjord (Liefdefjorden), found higher levels of POPs and higher bioaccumulation factors in the Atlantic influenced fjord (Hallanger, Ruus, et al., 2011). Another study from the Canadian Arctic, showed lower trophic biomagnification of contaminants in food webs consisting of only native Arctic species, compared to food webs also hosting transient and resident species (McKinney et al., 2012). Evidently, knowledge about dietary ecology and prey occurrence in top predators is essential to provide important answers to sources of contaminant exposure and how it may be impacted by on-going climate change.

As outlined above, seabirds, and in particular the kittiwake, can be considered valuable sentinels for the ecological effects of climate change, and their dietary ecology can be studied because they often regurgitate content of their proventriculus during handling (Vihtakari et al., 2018). Still, such samples only provide a "snapshot" of an individuals' feeding habits and might not be a representative for species composition in the fjord over time (Araújo et al., 2007). The precision in species determination of prey can also be a challenge due to samples being very digested, and species consisting of softer tissue might be underrepresented because they are more easily digested (Ramos et al., 2009). Still, the regurgitate samples give specific species information, and can therefore be used as an indication of the state of the fjord in relation to potential Atlantification.

In contrast to the more traditional analysis of regurgitates, analysis of stable isotopes has become increasingly popular to better understand a species' dietary ecology. Different tissues are synthesized and replaced at different rates, and the stable isotope composition in a predator generally reflects the

composition of its diet at the time of tissue synthesis (Inger & Bearhop, 2008). Stable nitrogen isotope values (δ^{15}N) have been established as a proxy for trophic position (Bearhop et al., 2002; Hobson & Clark, 1992) assuming a stepwise trophic enrichment of the heavier ^{15}N-isotope (Hop et al., 2002). Stable carbon isotope values (δ^{13}C) indicate foraging habitat and primary carbon source, as different stable carbon isotope signatures can be observed for different primary producers (Inger & Bearhop, 2008; Kelly, 2000). Benthic and coastal habitats have higher δ^{13}C values compared to pelagic and offshore habitats (Fisk et al., 2003; McKinney et al., 2013). Moreover, δ^{13}C and δ^{15}N values can be combined to determine a species' stable isotopic niche, considered a proxy for the dietary ecological niche (Jackson et al., 2011). Stable isotope values will therefore reflect all ingested prey species ingested over a time, and therefore complement other methods such as regurgitate analysis. Still, it is challenging to interpret stable isotope values as they do not directly reveal the prey composition. Contaminant exposure in kittiwakes from Kongsfjorden has not been studied in relation to the shift observed in their diet. Combining regurgitate analysis with stable isotope analysis can be promising to study dietary ecology more holistically when investigating the effects of dietary ecology on contaminant exposure.

The aim of my thesis is to investigate if contaminant concentrations in kittiwakes from Kongsfjorden (Svalbard, Norway) can be explained by changes in their diet, previously linked to how climate change impacts in this particular fjord (Vihtakari et al., 2018). The present thesis uses a large collection of regurgitate data (2007-2020), stable carbon (δ^{13}C) and nitrogen isotope (δ^{15}N) values (2007-2009, 2011-2015, 2017-2019), and contaminant concentration data (2007-2009, 2011-2015, 2017-2020). Both the origin of and differences in lipid storage among Arctic and Atlantic prey species might have consequences for the contaminant exposure in predators. While the Atlantic species might function as biovectors, bringing contaminants into the Arctic environment (McKinney et al., 2012), the Arctic species might have a higher contaminant load, due to higher lipid content (Hop et al., 2002).

My thesis considers the following objectives: 1) Is the between-year variation in prey species, as earlier observed and related to climate change by Vihtakari et al. (2018), still occurring? Based on recent information on climate warming (AMAP, 2021), there is no reason for the ongoing Atlantification found by Vihtakari et al. (2018) to have ceased, but that there are still fluctuations between cold and warm years. 2) How does the between-year variation in prey composition relate to stable isotope values, and do the stable isotope values show between-year variations as well? 3) How do changes in diet quantified by either regurgitate data, stable isotope data, or a combination, relate to contaminant levels of major legacy contaminants in kittiwakes?

2. Materials and Methods

2.1. Sample collection

Sampling for kittiwake blood and regurgitates was conducted in Kongsfjorden, Svalbard (Figure 1) during the breeding period during 2007-2020. A total of 426 blood samples and 821 regurgitate samples were collected during these years. Adult birds were caught on their nest using a loop at the end of a long pole. Birds were weighted to the nearest 2 g with a Pesola spring balance and the length of the skull was measured using a calliper (to the nearest 0.1 mm).

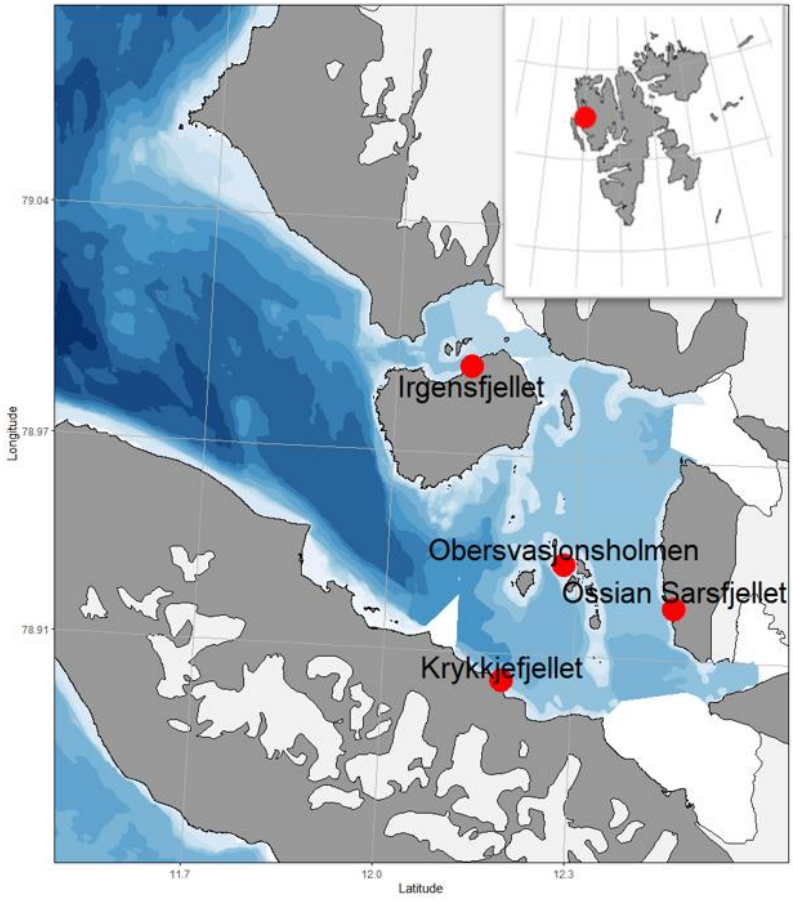

Figure 1: Overview map of Svalbard showing the location of Kongsfjorden, joined by a detailed map of Kongsfjorden showing the different black-legged kittiwake colonies at which samples were collected.

Blood samples (*n*=426) were collected during the chick-rearing period (early July to early August) during 2007-2020 at Krykkjefjellet (Figure 1), were yearly samples size varied from 23 – 65 samples. A 2.5 mL blood sample was taken from the alar vein with a heparinized syringe and a 25-gauge needle. Blood samples were kept cold during the day and erythrocyte and plasma fractions were obtained at the end of each field day by centrifugation for 10 min at 1,000 G. Samples were kept frozen at -18 ^{o}C until further analysis.

Regurgitate samples (*n*=821) were collected at different colonies in Kongsfjorden, Svalbard (Figure 1) every year during 2007-2020. Out of 821 samples, 251 were collected at Krykkjefjellet (78°53'46"N, 12°11'43"E), 275 at Irgensfjellet (78°59'37"N, 12°7'46"E), 43 at Ossian Sarsfjellet (78°56'17"N, 12°26'21"E), and 58 at Observasjonsholmen (78°56'20"N, 12°17'5"E), while 194 samples had no record of specific colony (Table Appendix.1). Regurgitate contents were collected into marked plastic bags and frozen at -18 °C. Regurgitate samples were collected from both adults (*n*=436) and chicks (*n*=131), though for 254 samples the maturity stage was not registered.

2.2. Study species and system

The black-legged kittiwake is a long lived, northern circumpolar gull species that feeds at the surface of the pelagic zone, and its opportunistic diet is composed of fish as well as planktonic invertebrates and crustaceans (Barrett, 2007; Blévin et al., 2014; Bustnes et al., 2017; Mehlum & Gabrielsen, 1993). It is a very common breeding species in the Arctic and boreal zone (Barrett, 2007). Most kittiwakes spend the winter in the West Atlantic, between Newfoundland and the Mid-Atlantic ridge, including offshore, deep water areas (Gabrielsen, 2009; SEATRACK, 2022). Kittiwakes arrive in Svalbard between March-April and leave in September. Hatching normally occurs mid-July and chicks are fed by both parents on a varied diet resulting of foraging within the fjord (Burr et al., 2016; Vihtakari et al., 2018).

Kongsfjorden is a glacial fjord located in the high-Arctic at the west coast of Spitsbergen (Figure 1). It is an open fjord without sill and is therefore strongly influenced by inflow of relatively warm and saline Atlantic water from the West Spitsbergen Current, mixed with cold and less saline Arctic and glacial melt water during summer (Cottier et al., 2005; Hop & Wiencke, 2018; Tverberg et al., 2019; Vihtakari et al., 2018). The two currents mix at the shelf-break and creates a dynamic fjord hydrography with conditions that fluctuate from year to year (Tverberg et al., 2019; Vihtakari et al., 2018). The mixture of Atlantic and Arctic water present in the fjord leads to the mixed presence of Atlantic and Arctic fauna (Hop & Wiencke, 2018; Vihtakari et al., 2018). The inner fjord basin is influenced by run-off from tidal glaciers, and this part of the fjord is considered mostly Arctic (Hop & Wiencke, 2018). The glacial fronts found in Kongsfjorden are important feeding areas for both marine mammals and seabirds, including kittiwakes (Lydersen et al., 2014). Kongsfjorden has been strongly influenced by the West Spitsbergen Current with declining sea ice cover and increasing temperatures in recent years leading to a transition of Arctic waters to a state more closely resembling that of the Atlantic (Tverberg et al., 2019; Vihtakari et al., 2018).

2.3. Molecular sexing

Molecular sexing of birds was performed at the Centre d'Etudes Biologiques de Chizé, France on erythrocytes by polymerase chain reaction amplification on part of two highly conserved genes present on sexual chromosomes. Female birds are heterogametic (ZW) while males are homogametic (ZZ), and sexing can be done by detection of the W chromosome (sequences). The method for sexing non-ratite birds developed by Fridolfsson and Ellengren (1999) is based on the detection of a constant size difference between the genes CHD1W and CHD1Z. By using highly conserved primers flanking the intron, PCR amplification and agarose electrophoresis, females can be characterised by displaying one (CHD1W) or two fragments (CHD1W and CHD1Z), while males only show one fragment (CHD1Z) clearly different in size from the female-specific CHD1W fragment.

2.4. Regurgitate analysis

Regurgitate samples were analysed at the Norwegian Polar Institute, Norway. Prey species were counted and identified to the lowest possible taxon using a stereomicroscope and identification literature (Campana, 2004; Härkönen, 1986). Fish species were determined from body morphology, though using otolith morphology when samples were too digested. Different observers have been involved in this work, and consistency in methods across observers was aimed for by following a template for reporting of observations. Due to inconsistent reporting in recent years and therefore limitations in the dataset, no statistical tests can be performed to assess between-year variation for diet data.

2.5. Stable isotope analysis

Stable isotope analysis was carried out on a total of 246 samples at the Centre d'Etudes Biologiques de Chizè, France. Samples were freeze-dried and homogenised and consisted both of erythrocytes (*n*=193) and whole blood (*n*=53). Subsamples were wrapped in tin containers and analysed for the relative abundance of stable carbon (^{13}C and ^{12}C) and nitrogen isotopes (^{15}N and ^{14}N) using a continuous flow mass spectrometer (Delta V Plus with a Conflo IV interface, Thermo Scientific, Bremen, Germany) coupled to an elemental analyser (EA Isolink, Thermo Scientific, Milan, Italy). Values are presented per convention in δ-notation (‰), representing the obtained ratios relative to the secondary isotopic reference materials Vienna PeeDee Belemnite and atmospheric N_2 for $\delta^{13}C$ and $\delta^{15}N$, respectively. Replicate measurements of internal laboratory standards (Caffeine) indicate analytical precision <0.10‰ and <0.15‰ for $\delta^{13}C$ and $\delta^{15}N$, respectively.

2.6. Persistent organic pollutant analysis

A total of 425 samples were analysed for a wide range of POPs (Table Appendix.2) at the Norwegian Institute of Air Research (NILU; Table Appendix.3). Samples consisted both of whole blood (*n*=331) and plasma (*n*=94). A detailed description of the analytical protocol can be found in Appendix.1. Samples were spiked with 20 µL of internal standard mixture (Table Appendix.4), after which deionised water saturated with ammonium sulphate and ethanol was added. Liquid-liquid extraction was carried out twice using 6 mL of cyclohexane. Supernatants were concentrated and left to dry overnight, and lipid content was determined gravimetrically on the dried samples. Sample extracts were subsequently re-dissolved in *n*-hexane and clean-up using the Zymark RapidTrace SPE Workstation. Solid phase extraction (SPE) was done using dichloromethane (DCM :*n*-hexane (1:9; *v:v*) as mobile phase and Florisil as stationary phase. After elution *iso*-octane was added as keeper and a recovery standard (^{13}C-PCB 159) was added (Table Appendix.5). Quantification of the targeted compounds was conducted using a Thermo Scientific trace 1310 gas chromatograph coupled to a Thermo Scientific TSQ9000 triple quadrupole mass spectrometer equipped with a DB-5 column (length 30 m, 0.25 µm film thickness, 0.25 mm inner diameter) with precolumn (0.53 mm) and restriction capillary column (0.18 m). Quality assurance and quality control were carried out using concurrent blank and reference material (SRM 1958 – human serum; NIST) samples (every 10th sample). The accuracy of the method ranged from 70 to 122% for all compounds, with the exception of OxC (37% accuracy caused by coelution). If a blank sample showed presence of a compound, the limit of detection (LOD) for that compound was set to three times the blank's signal. In all other cases, the LOD was set to three times the instrumental noise. The limit of quantification (LOQ) was set to three times the LOD, and their values for the targeted compounds, analysed during several batches, can be found in Table Appendix.8.

2.7. Data analysis

Statistical analyses were performed using the software R version 4.1.1 (R Core Team, 2021) and RStudio version 1.4.1717 (RStudio Team, 2020). Figures were made using the package *ggplot* (Wickham, 2016) and standard R plot functions. A map of Svalbard (Figure 1) was made using the R-package *PlotSvalbard* (Vihtakari, 2020). The significance level was set to $\alpha = 0.05$ for all tests.

2.7.1. Selection of study compounds

The contaminant dataset available for the present study compiled concentrations for 43 different compounds analysed for. Yet, all 43 compounds have not been targeted each year, making consistent

comparisons a challenge. The present thesis therefore focuses on seven compounds: PCB 99, PCB 153, PCB 180, *p,p'*-DDE, *β*-HCH, oxychlordane and HCB. This selection was based on the evaluation of ecotoxicological relevance (indicated by literature), potential to drive the magnitude of exposure (Figure Appendix.3), intercorrelation (Figures Apendix.1-2) and consistent detection (Tables Appendix.8-9).

To evaluate the ecotoxicological relevance of different PCB congeners different literature was consulted for weight of evidence of their frequency and toxicological impact. PCB 99, PCB 153 and PCB 180 are three of the major congeners consistently detected in humans and wildlife, especially in the Arctic (Bentzen et al., 2008; Bernhoft et al., 1997; Kucklick et al., 2002; Oskam et al., 2004). OxC is the primary metabolite of chlordane and is found to be very persistent (AMAP, 2016; Braune et al., 2019). Seabirds are able to metabolise both TC and CC, but not OxC (AMAP, 2016). From all DDT-related compounds, *p,p'*-DDE is the major metabolite and is very persistent, more so than the other metabolites, and is therefore prevalently detected as dominant organochlorine pesticide (OCP; Kelce et al., 1995). Unlike for other OCPs, HCB emissions still occur (AMAP, 2016; Andersen et al., 2015). A large proportion of time-series of HCB in biota across the North American and European Arctic over recent decades has shown a slow mean rate of decrease, while some sites show an increase in both air and biota (AMAP, 2016). Similarly, some time series show significant increasing trends for *β*-HCH (AMAP, 2016; Rigét et al., 2013). This might be explained by the chemical properties of *β*-HCH: it has a greater tendency to biomagnify in biota and is more resistant to metabolic and microbial degradation compared to both *α*-HCH and *γ*-HCH (AMAP, 2016; Li et al., 2002).

To evaluate the potential of compounds to drive the magnitude of the exposure PCB and OCP profiles were constructed, each showing the mean ± *SD* percentage of each congener or compound of the total PCB or OCP burden, respectively (Figure Appendix.3). PCB 153 is the PCB congener with the dominant contribution to the kittiwake PCB burden, followed by PCB 99 (and PCB 118) and PCB 180 for lower- and higher-chlorinated congeners, respectively. Among OCPs, OxC, *p,p*'-DDE and HCB are among the major contributors to the overall OCP burden in the kittiwakes. *β*-HCH, however, does not stand out as a large contributor compared to some other OCPs such as HeptEpox and Mirex.

To evaluate the intercorrelation between PCB congeners or OCP compounds, Pearson correlation matrix plots were evaluated (Figures Appendix.1-2). PCB 99, 153 and 180 show positive correlations with most other congeners (Figure Appendix.1) and can function as good representatives for most other PCB congeners. Still there are some PCB congeners that seems to behave distinctly different. From Figure Appendix.1. it could be argued that there are four district groups of PCB congeners that differ from each other, and PCB 99, 153 and 180 are all from the same group. Therefore, it could be argued that different PCB congeners should be included to better represent the four different groups. While the PCB congeners are all positively correlated

to each other, though not significantly, some OCPs were negatively correlated with each other (Figure Appendix.2). β-HCH, *p,p'*-DDE, HCB and OxC are positively correlated with most other OCPs. There are though some OCPs that seems to be very distinct from the others, suc as *o.p'*-DDE, CN, TN and *γ*-HCH. These might not be represented by the four OCPs chosen for this present study.

The evaluation of the detection of the targeted compounds was based on their detection frequency, calculated per year (Table Appendix.9). PCB 99, 118, 138, 153, 180, 183 and 187 has been screened for all years with a high detection frequency. HCB was screened for every year with a high detection frequency. Screening for *β*-HCH has been more consistent compared to the other HCHs, but still lacks screening for four years, and shows a low detection frequency for some years. OxC has a higher detection frequency compared to other chlordanes. *p,p'*-DDE is the one of all the DDT metabolites that has been screened for most consistently and has moreover a high detection frequency.

2.7.2. Data preparation

For the present study, contaminant, stable isotope and regurgitate observations and their morphological metadata were compiled from several datasets collected over the last decade and a quality check of the data was performed. Merging different datasets required harmonisation of lipid content, contaminant concentrations, stable isotope values and regurgitate observations.

For some contaminant samples lipid percentage could not be obtained (*n*=70). Those samples were assigned a mean value calculated from other samples obtained the same year, or, if no lipid content data was available for that same year, a mean across all other years was calculated and ascribed.

Stable isotope analysis was carried out from 2007 – 2020, except for 2010. The C:N mass ratio, indicative for lipid content, for each sample was calculated and its range (3.15 – 3.62) was not deemed to bias the obtained stable isotope values (Tartu et al., in press). Different blood components were analysed for stable isotopes, namely both erythrocytes (*n*=193) and whole blood (*n*=53). A conversion factor was therefore applied to stable isotope values obtained for whole blood according to Tartu et al. (in press):

$$\delta^{13}C_{ERY} = \delta^{13}C_{WB} \times 0.985 \qquad \text{Equation 1}$$

and

$$\delta^{15}N_{ERY} = \delta^{15}N_{WB} \times 1.004 \qquad \text{Equation 2}$$

Contaminant concentrations were excluded from statistical analysis when for a specific year they were detected in less than 50% of the sample set available (see Table Appendix.9). In other cases, when detection

was not 100% for a certain year, concentrations below LOQ were assigned a value equal to half the compound-specific LOQ value.

Contaminant analysis was performed on plasma for years 2012, 2013 and 2014, and a conversion factor was applied to these to match whole blood concentrations available for all other years. A conversion factor for each study compound was calculated from paired plasma and whole blood samples (*n*=23) collected during 2017, 2018 and 2020. The conversion factor was obtained from significant linear models according to:

$$C_{WB} = intercept + slope \times C_P + error \quad \text{Equation 3}$$

were C_{WB} = the predicted contaminant concentration in whole blood, C_P = contaminant concentration in plasma. Every model was tested for influential outliers using *outlierTest* from the package *car* (Fox & Weisberg, 2019), and those deemed influential were removed from the model (Table Appendix.11). The values used for conversion for each study compound can be found in Table Appendix.10.

Contaminant concentrations for all samples were converted from wet weight to lipid weight according to:

$$\frac{C_{ww} \times 100}{L} = C_{lw} \quad \text{Equation 4}$$

were C_{ww} = wet weight, C_{lw} = lipid weight and L = lipid percentage.

Diet items were categorized into different taxonomic groups and groups based on origin (Arctic, Atlantic, Intermediate and mesopelagic) for statistical analysis, according to Vihtakari et al. (2018), (Table 2). Frequency of occurrence (FO; %) per year was calculated as a proportion of samples containing a given diet item for both prey species found in regurgitate samples and for different groups (Figure 2). FO for all prey species were also calculated for all years combined (Table 2).

All data was checked for influential outliers using boxplots and Cleveland dotplots, and collinearity was tested with the Variance Inflation Factor (VIF) with a threshold of 3.00 (Zuur et al., 2010). Concentrations of all study compounds were *ln*-transformed to approximate a normal distribution.

2.7.3. Exploration of dietary ecological variables

Data from both stable isotope information and regurgitate samples were evaluated for temporal aspects as well as how both approaches relate to each other prior to model construction and model selection to relate diet data to contaminant data. Bird ID was registered for both contaminant and stable isotope data, and only samples containing data for both were used in the data analysis. Regurgitate data had no registration of bird

ID and only FO therefore represents the diet composition for the species in that year, rather than individual-level information.

The *SIBER* package (Jackson et al., 2011) was used to determine the stable isotope niches based on blood stable isotope values integrating the dietary niche during the chick-rearing period. This allows comparison of dietary ecological niches among years. For each year the standard ellipse area corrected for small sample sizes (SEA_C; ‰2), was calculated for the bidimensional stable isotope space outlined by $\delta^{13}C$ and $\delta^{15}N$ (Figure 6), using a 95% confidence interval for the mean of each period. Linear models were run with ANOVA to check for between-year variation in stable isotope values.

While the regurgitate samples only shows a snapshot of available prey in the fjord at the time right before the capture of the bird, stable isotope values measured in blood integrate feeding habits over 10-14 days (Boecklen et al., 2011). Therefore, relationships between stable isotope information ($\delta^{13}C$, $\delta^{15}N$ and SEA_C) and regurgitate data were analysed. The relationship between the SEA_C and FO of Arctic species (FO_{Arctic}), FO of herring (*Clupea harengus*; $FO_{Herring}$) and FO of polar cod (*Boreogadus saida*; $FO_{PolarCod}$) were investigated using linear models and ANOVA, while relationships between $\delta^{13}C$ or $\delta^{15}N$ with FO_{Arctic}, $FO_{Herring}$ or $FO_{PolarCod}$ were investigated using linear mixed effects (LME) models, with the variable *year* as a random effect factor, and ANOVA.

2.7.4. Model selection

LMEs were used to investigate the effect of dietary ecology (stable isotopes and regurgitates) on contaminant concentrations for the seven study compounds. For each compound a model selection procedure was constructed, composing different models each with a certain set of variables (Table 1), based on *a priori* hypotheses, and the variable *year* as random effect factor. Models were ranked according to Akaike's Information Criterion corrected for small sample size (AIC_C; Anderson & Burnham, 2002) using the function *aictab* from the R-package *AICcmodavg* (Mazerolle, 2020). Reporting the output of the model selection procedure has been done according to Anderson and Burnham (2002), and includes difference in AIC_C (ΔAIC_C), Akaike's weight (w) and residual squares (marginal R^2 and conditional R^2). Marginal R^2 describes the proportion of variance explained by fixed factors alone, while conditional R^2 describes the proportion of variance explained by both fixed and random factors (Nakagawa & Schielzeth, 2013). Comparing a set of candidate models, the most parsimonious model will be the one with the lowest AIC_C value, though only if the next best model has $\Delta AIC_C > 2$. ANOVA was used to investigate all models with $\Delta AIC_C < 2$. Different models were later compared to each other and in terms of how data supported the different hypotheses.

In a first hierarchy of *a priori* hypothesis testing three models were formulated based on two main groups of potential dietary ecological drivers: stable isotope analysis, regurgitate analysis, or both. This first hierarchy *a priori hypothesis* testing follows the reasoning:

1. **Stable isotope analysis:** This hypothesis assumes the dietary ecological aspects are more important drivers in contaminant levels, compared to individual prey species. Since stable isotope values reflect the diet for the last couple of days, they provide an integrated quantitative measure of both carbon source and trophic position, both shown earlier to potentially impact contaminant levels (Hop et al., 2002; McKinney et al., 2012). Moreover, stable isotope and contaminant data are linked to the same individual bird, while prey observations are not, providing the possibility for a stronger relationship between stable isotope values and contaminants data.
2. **Regurgitate analysis:** This hypothesis assumes individual prey species drive variation in contaminant levels. Arctic species are known to have high lipid content compared to sub-Arctic or Atlantic species (Borgå et al., 2001; Hallanger, Ruus, et al., 2011) and can therefore bioaccumulate more contaminants compared to less lipid-rich species (Hop et al., 2002). On the other hand, transient species have earlier been identified as biovectors for contaminants from lower latitudes into the Arctic environment (McKinney et al., 2012; Morris et al., 2016). Concentrations of contaminants are influenced by dietary intake and therefore also availability of prey with different lipid and organochlorine loads (Bustnes et al., 2017), and their diet might therefore explain contaminant exposure.
3. **Both:** This hypothesis assumes individual prey species as well as functional dietary ecological traits drive variation in plasma contaminant levels of the predator. Combining prey item information with stable isotope information provides a complementary picture of the diet for the last couple of days to explain variability in contaminant levels. They are also complimentary as stable isotope information is linked to individual birds while prey items represent the biological state of the fjord.

In a second hierarchy of *a priori* hypothesis testing the first hierarchy is dissected in more concrete combinations of the available six variables (Table 1). As such the stable isotope analysis to quantify the dietary ecology composes $\delta^{13}C$ and $\delta^{15}N$ values and the resulting two-dimensional stable isotope niche (SEA_C), while the regurgitate analysis composes FO_{Arctic}, $FO_{PolarCod}$ and $FO_{Herring}$. Considering that SEA_C is calculated from $\delta^{13}C$ and $\delta^{15}N$ values, SEA_C were never combined in the same model with either $\delta^{13}C$ or $\delta^{15}N$ due to collinearity. Since $FO_{PolarCod}$ constitutes a large proportion of FO_{Arctic}, these factors were never combined in the same model. This second hierarchy *a priori hypothesis* testing follows the reasoning:

1. **Null-model:** A null-model was included in the model selection, containing only *year* as a random effect factor, indicating that none of the dietary factors have an effect on contaminant variation, and that the variation in contaminants is better explained by other inter-annual variation.

1.1. δ^{13}C and δ^{15}N: This hypothesis assumes that the variation in contaminant exposure is not driven by specific species or functional groups but solely by the carbon source and trophic position. The stable isotope values reflect the diet for the last couple of days, and both were previously shown to be important drivers of contaminant variability (Hop et al., 2002; McKinney et al., 2012).

1.2. δ^{13}C: This hypothesis assumes that δ^{13}C values, indicative of the primary producer supporting the food chain in which the predator feeds (Araújo et al., 2007; Kelly, 2000), influence the contaminant exposure as the carbon source was previously shown to impact contaminant concentrations through a shift from benthic or nearshore and ice-associated food webs to pelagic-type food webs. (McKinney et al., 2013).

1.3. δ^{15}N: This hypothesises assumes an increase in δ^{15}N values, indicating an elevated trophic position (Bearhop et al., 2002; Hop et al., 2002), results in higher contaminant burdens as the studied compounds are prone to biomagnify in the food chain (Borgå et al., 2001; Hop et al., 2002).

1.4. SEA_C: This hypothesis assumes that the size of the stable isotope niche, a proxy for the dietary ecological niche, affects contaminant levels, assuming that increased contamination exposure results from an increased niche size. An increased niche size reflects opportunistic feeding and reflects a diet with different prey organisms.

2.1. FO_{Arctic}: Since transient species might function as biovectors for contaminants into the Arctic (Jenssen et al., 2015; Morris et al., 2016), this hypothesis assumes that years with high FO_{Arctic} will have lower levels of contaminants compared to years with low FO_{Arctic}.

2.2. $FO_{PolarCod}$: This hypothesis assumes that $FO_{PolarCod}$, representing high trophic Arctic prey species, may be an important driver of contaminant variability as the influence of lower trophic prey species included in FO_{Arctic} may not be as strong on variation in contaminant exposure in kittiwakes.

2.3. $FO_{Herring}$: This hypothesis assumes that $FO_{Herring}$, representing a high trophic Atlantic species, may be an important driver for contaminant variability, and is chosen as a representative for Atlantic species. It is hypothesised that $FO_{Herring}$ can function as a biovector, increasing contaminant concentrations (Jenssen et al., 2015; Morris et al., 2016).

3.1. δ^{13}C, δ^{15}N and FO_{Arctic}: This hypothesis assumes that the variation in contaminant exposure is not driven only by trophic position and carbon source, but also in relation to different prey types and their origin, and by taking into consideration FO_{Arctic}, these factors combined will explain variations in contaminant exposure.

3.2. **$\delta^{13}C$, $\delta^{15}N$ and $FO_{PolarCod}$:** This hypothesis assumes that the variation in contaminant exposure explained by a combination of carbon source, trophic position and $FO_{PolarCod}$, where polar cod represents a high trophic Arctic species.

3.3. **$\delta^{13}C$, $\delta^{15}N$ and $FO_{Herring}$:** This hypothesis assumes that the variation in contaminant exposure is explained by the combinations of carbon source, trophic level and $FO_{Herring}$, where herring is a representative for the Atlantic species acting as biovectors of contaminants in the Arctic.

3.4. **SEA_C and FO_{Arctic}:** This hypothesis assumes that a larger niche size with a contribution of a variety of Arctic species can influence contaminant levels. It is hypothesised that larger SEA_C will lead to higher contaminant exposure when FO_{Arctic} is low, and the diet consists of a large variety of species from outside the Arctic.

3.5. **$\delta^{13}C$ and FO_{Arctic}:** This hypothesis assumes that both carbon source and FO_{Arctic} explain the variance in contaminant exposure, as it combines information about foraging over several days ($\delta^{13}C$) linked in time to individual contaminant exposure, and it contains information about the biological state of the fjord through the prey organisms (FO_{Arctic}).

3.6. **$\delta^{15}N$ and FO_{Arctic}:** This hypothesis assumes that both trophic position and geographical origin of the prey explain the variance in contaminant exposure, as it combines information on foraging and the trophic level of prey species over several days ($\delta^{15}N$) linked in time to individual contaminant exposure, and it contains information about the biological state of the fjord through the prey organism (FO_{Arctic}).

Table 1. All candidate models, and their containing variables, used for the compound-specific model selection investigation into the dietary ecological drivers of exposure in black-legged kittiwakes in Kongsfjorden, Svalbard.

	$\delta^{13}C$	$\delta^{15}N$	SEA_C	FO_{Arctic}	$FO_{PolarCod}$	$FO_{Herring}$
Model 0						
Model 1.1.	x	x				
Model 1.2.	x					
Model 1.3.		x				
Model 1.4.			x			
Model 2.1				x		
Model 2.2.					X	
Model 2.3.						x
Model 3.1.	x	x		x		
Model 3.2.	x	x			x	
Model 3.3.	x	x				x
Model 3.4.			x	x		
Model 3.5.	x			x		
Model 3.6		x		x		

Data on $\delta^{13}C$, $\delta^{15}N$ and SEA_C were available for all years except for 2010, and therefore data from 2010 for all available variables were excluded from the analysis. Similarly, contaminant data were available for 2007-2020 except for 2016, and therefore 2016 data for all available variables were excluded from the analysis. A final sample size of *n*=224 was used for the model selection procedure for all compounds except for *β*-HCH, which because of a low frequency of detection had *n*=160.

Models were run with Maximum likelihood for model selection, but since restricted Maximum of likelihood (REML) is considered to give better estimates for the random effects than Maximum likelihood, the models for all contaminants were run with REML for final interference of the estimates.

3. Results

3.1. Between-year variation in regurgitate content

Species found in regurgitate samples varied from year to year (Figure 2; Table 2), and species were categorized into different groups as shown in Table 2. The species with the highest FO for all years combined was $FO_{PolarCod}$ (23.9%) followed by $FO_{Capelin}$ (17.0%; Table 2). Still, FO for unidentified fish (18.1%) were higher than $FO_{Capelin}$. Only six species contribute more than 5% each of the total FO for all years; polar cod (23.9%), capelin (17.0%) unidentified fish (18.1%), herring (6.6%), *Thysanoessa inermis* (16.0%) *and Themisto libellula* (5.2%). Many species, like shorthorn sculpin (*Myoxocephalus Scorpius*), threespot eelpout (*Lycodes rossi*) and *Thysanoessa longicaudata* were observed only a few times and does not contribute much to the total FO for all years combined. It is also worth mentioning that only three species are classified as Arctic species: Polar cod, *Themisto libellula* and *Limacina helicina*, while seven species are classified as Atlantic, and 17 species/items are categorized as Intermediate.

Table 2: Overview of all encountered diet items found in regurgitate samples from adult and chick kittiwakes sampled at different colonies in Kongsfjorden during the breeding seasons of 2007-2020. Frequency of occurrence (FO) calculated from pooled samples from all years for each diet item is presented. Symbol indicates grouping based on taxa related to their origin (♣ = Arctic, ♦=Atlantic, ●=intermediate, ♠= mesopelagic).

Diet item	**FO** *(%)*
Fish	
♣ Polar cod (*Boreogadus saida*)	23.9
Atlantic fish	
♦ Herring (*Clupea harengus*)	6.6
♦ Capelin (*Mallotus villosus*)	17.0
♦ Atlantic cod (*Gadus morhua*)	1.4
♦ Haddock (*Melanogrammus aeglefinus*)	0.2
Other fishes	
♠ Glacier lanternfish (*Benthosema glaciale*)	1.1
● Shorthorn sculpin (*Myoxocephalus Scorpius*)	0.1
♠ White barracudina (*Arctozenus risso*)	0.9
● Daubed shanny (*Leptocliuns maculatus*)	0.6
● Snake blenny (*Lampenus lampretaeformis*)	0.2
● Rockfish (*Sebastes* spp.)	0.3
● Threespot eelpout (*Lycodes rossi*)	0.1
● Unidentified fish	18.1
Crustacea	
Krill	
● *Thysanoessa inermis*	16.
♦ *Thysanoessa longicaudata*	0.1
● Unidentified krill	0.1

♦ Northern krill (*Meganyctiphanes norvegica*)	0.1
Amphipods	
♦ *Themisto abyssorum*	0.7
♣ *Themisto libellula*	5.2
Shrimp	
• Northern prawn (*Pandalus borealis*)	1.4
♠ Cromson pasiphaeid (*Pasiphaea tarda*)	0.8
• Unidentified shrimp	0.2
• Unidentified crustacea	1.4
Other	
• Trawler waste	0.5
• Unidentified item	0.2
• Unidentified mollusca	0.1
• Chaetognath (*Parasagitta elegans*)	0.4
• Polychaetes (*Nereis* spp.)	2.4
♣ Pteropod (*Limacina helicina*)	0.2
• Chephalopods	0.1
• Fish eggs	0.1

The highest observed contribution of Atlantic fish was in 2007 (Figure 2). Capelin was the main constituent of Atlantic fish from 2007 until it was replaced by herring in 2013. The observed $FO_{PolarCod}$ ranged from around 40% for years 2011 and 2020 to low occurrence in 2015, 2018 and 2019. Fish dominated the diet composition in all years except 2010 and 2015 when krill constituted most of the diet. Nereis contributed by a small proportion (ranging around 5%) until 2015, and has contributed very little since, except in 2018. FO for both krill and Themisto shows big annual variations, but both seems to be important contributors in the diet in most years. For the years 2017 – 2020 unidentified fish constitutes a large part of the samples, ranging from 40-50%. Different personnel have analysed regurgitate samples, and it can be assumed that people with less experience in working with fish more often defines a fish as unidentified, compared to people with more experience in this area.

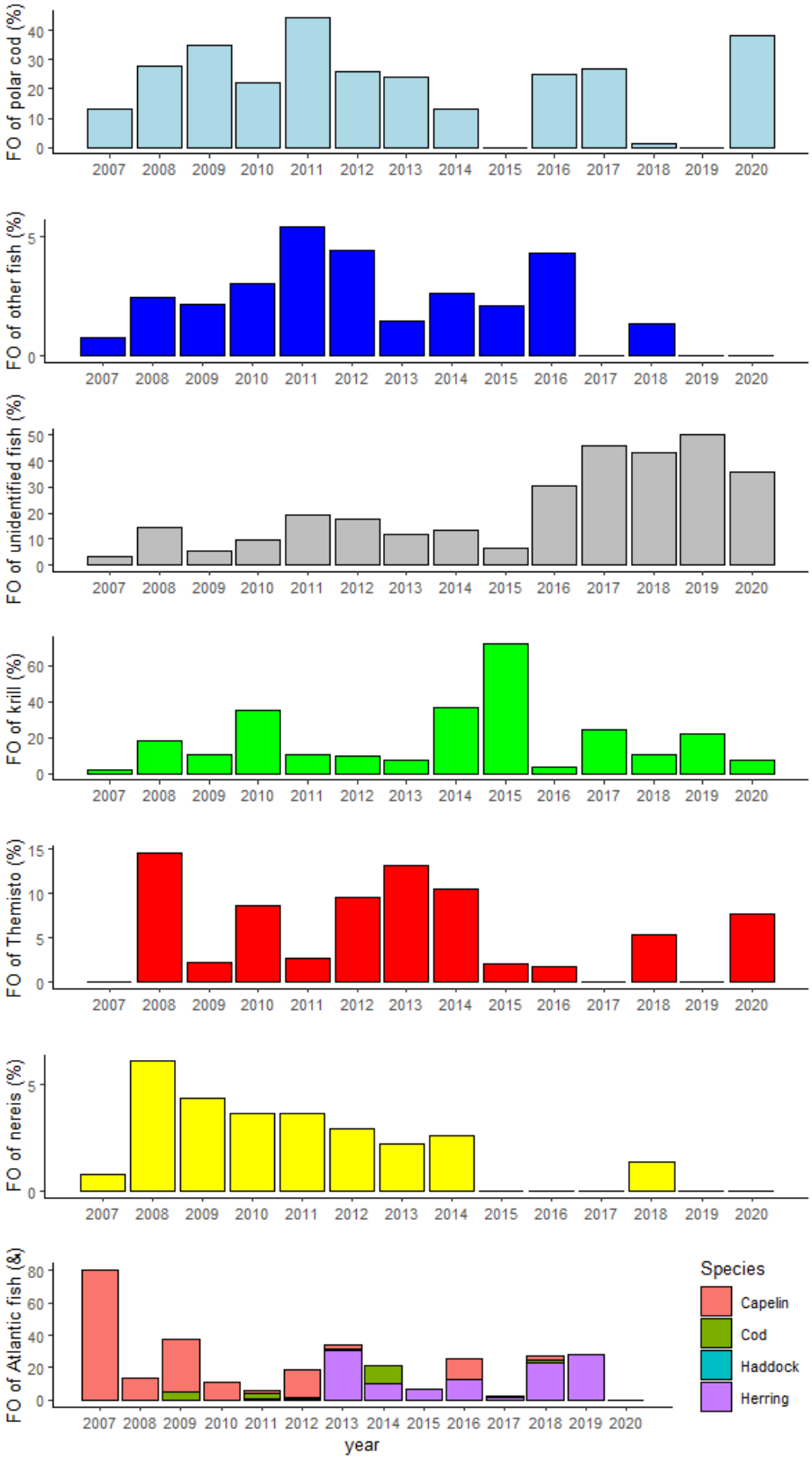

Figure 2. Frequency of occurrence of major diet items and groups from adult and chick kittiwakes from different colonies in Kongsfjorden, collected during the breeding seasons from 2007-2020. Note that each plot has different values on the y-axis.

In 2007 the Atlantic species constituted the main part of the kittiwakes' diet (Figure 3). Since 2014 the contribution of intermediate species was very high, but this might be explained by a high contribution of unidentified fish (which is categorized as intermediate; Table 2) since 2016 (Figure 2). The contribution of Arctic species was above 25% for most years, but very low in 2015 and 2018. The contribution of mesopelagic species was in general very low.

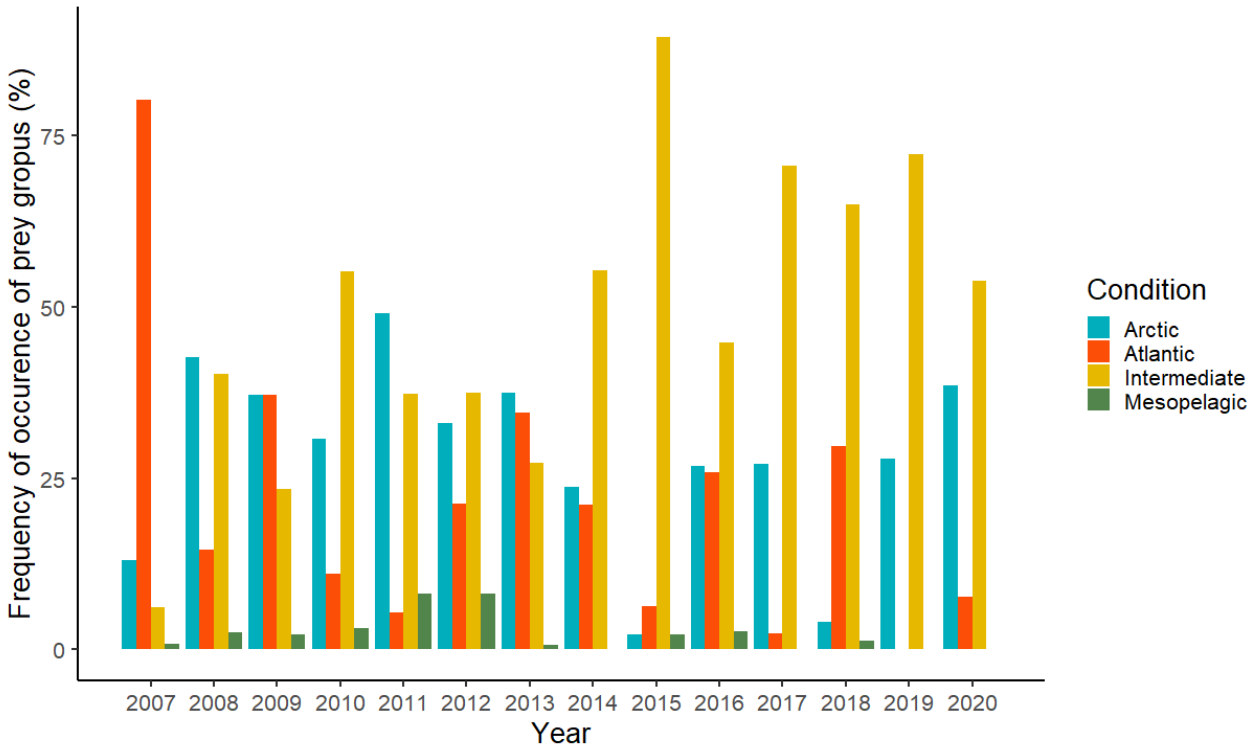

Figure 3. Frequency of occurrence of prey groups found in regurgitate samples from adult and chick kittiwakes from different colonies in Kongsfjorden, sampled during the breeding seasons from 2007-2020.

3.2. Between-year variation in stable isotope ecology

The stable isotope values for $\delta^{13}C$ and $\delta^{15}N$ varied between years ($78.89 \geq F \geq 143.70$; both $P < 0.01$;) and showed no clear pattern or trend (Figures 4 and 5, respectively). Values for $\delta^{13}C$ ranged between -21.72 and -19.66‰ (Table Appendix.19). The years 2009, 2015, 2017 and 2019 were significantly lower compared to all other years (all $P_{adj} < 0.01$), but not significantly different from each other ($0.06 \geq P_{adj} \geq 1.00$). The years 2007, 2012, 2013 and 2014 were significantly higher than all other years (all $P_{adj} < 0.01$), but not significantly different from each other ($0.22 \geq P_{adj} \geq 1.00$).

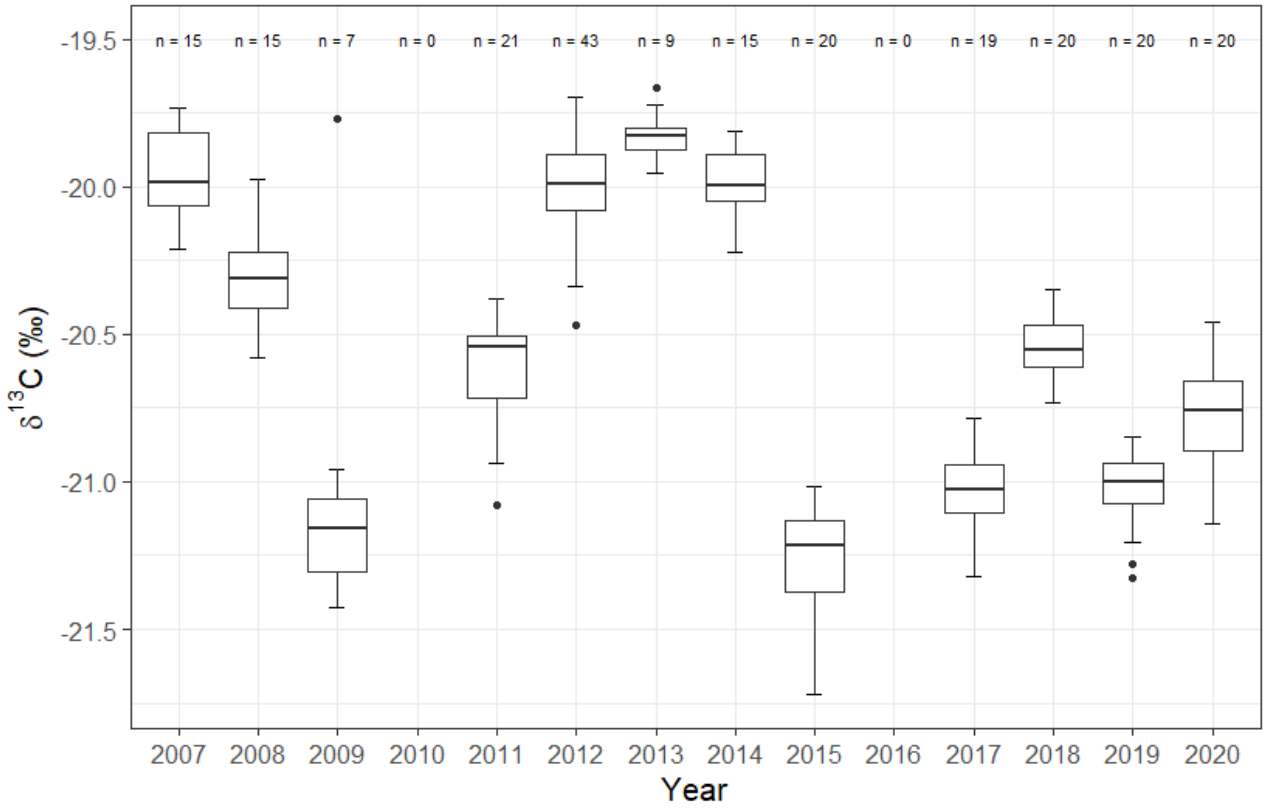

Figure 4. Between-year variation in δ^{13}C values measured in blood from adult kittiwakes during chick-rearing period in the Krykkjefjellet colony during the period from 2007-2020 (no values for 2010 and 2016). The horizontal line in the box plot indicates the median of the data, while the box is constituted by the 25 % and 75 % quartiles, and the whiskers represent 1.5 * interquartile range (IQR). n: samples size.

Values for δ^{15}N ranged between 11.05 - 15.58‰ (Table Appendix.19). The δ^{15}N values for 2020 was significantly higher than all other years ($P < 0.01$; Figure 5), while δ^{15}N values for 2013 was significantly lower than other years ($P_{adj} < 0.01$), except 2015 ($P_{adj} = 0.3$; Figure 5).

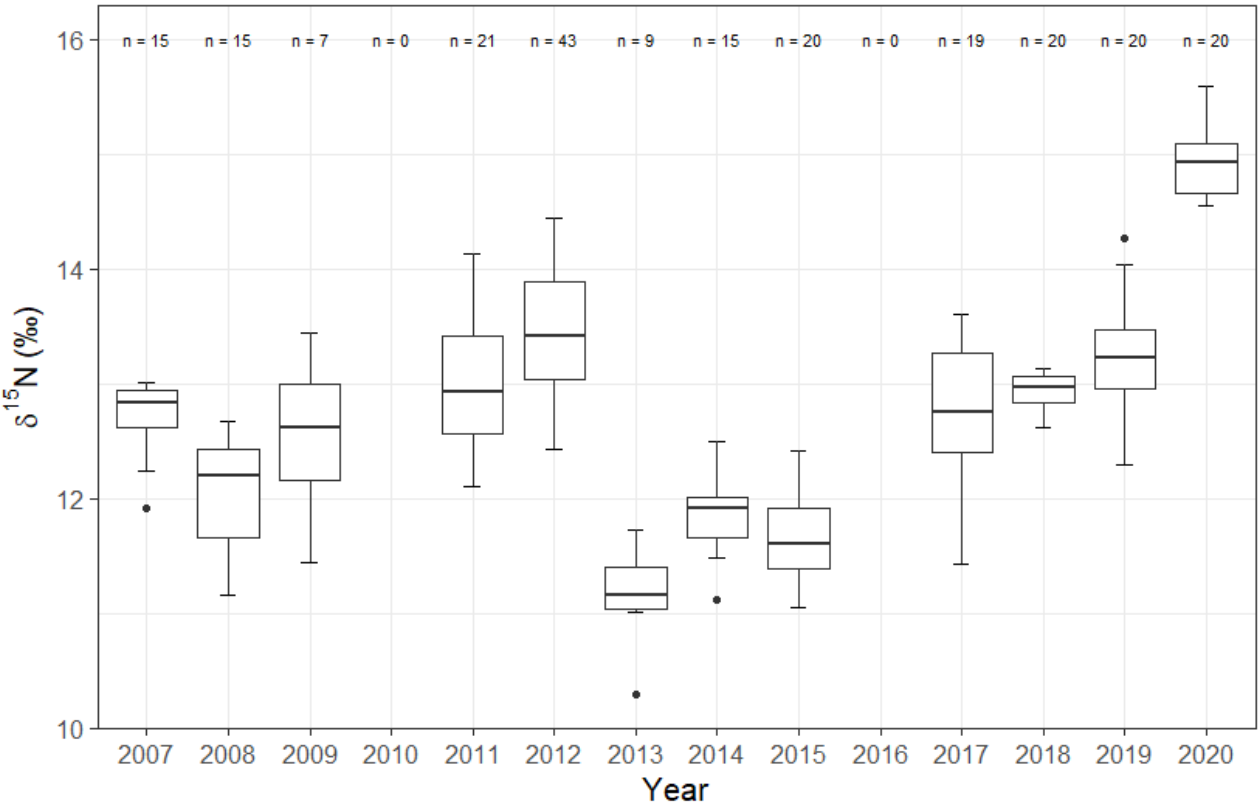

Figure 5. Between-year variation in δ^{15}N values measured in blood from adult kittiwakes during chick-rearing period in the Krykkjefjellet colony during the period from 2007-2020 (no values for 2010 and 2016). The horizontal line in the box plot indicates the median of the data, while the box is constituted by the 25 % and 75 % quartiles, and the whiskers represent 1.5 * IQR. n: samples size.

The size and location of stable isotope niches showed considerable between-year variation (Figure 6; Table 3). The stable isotope niche was largest in 2009, about ~35 larger than the smallest niche during 2018. The stable isotope niches shown in Figure 6 can be divided into three clusters. The stable isotope niches from 2009, 2011, 2015, 2017, 2018, 2019 make up one cluster, where the niches from different years also overlap. A second cluster is made up from the stable isotope niches from 2007, 2008, 2012, 2013, 2014, and these niches do not seem to overlap each other. A third and distinct cluster is made up by only the stable isotope niche from 2020. Due to limitations in the calculation of the stable isotope niches, no statistical test can be performed to assess between-year variation.

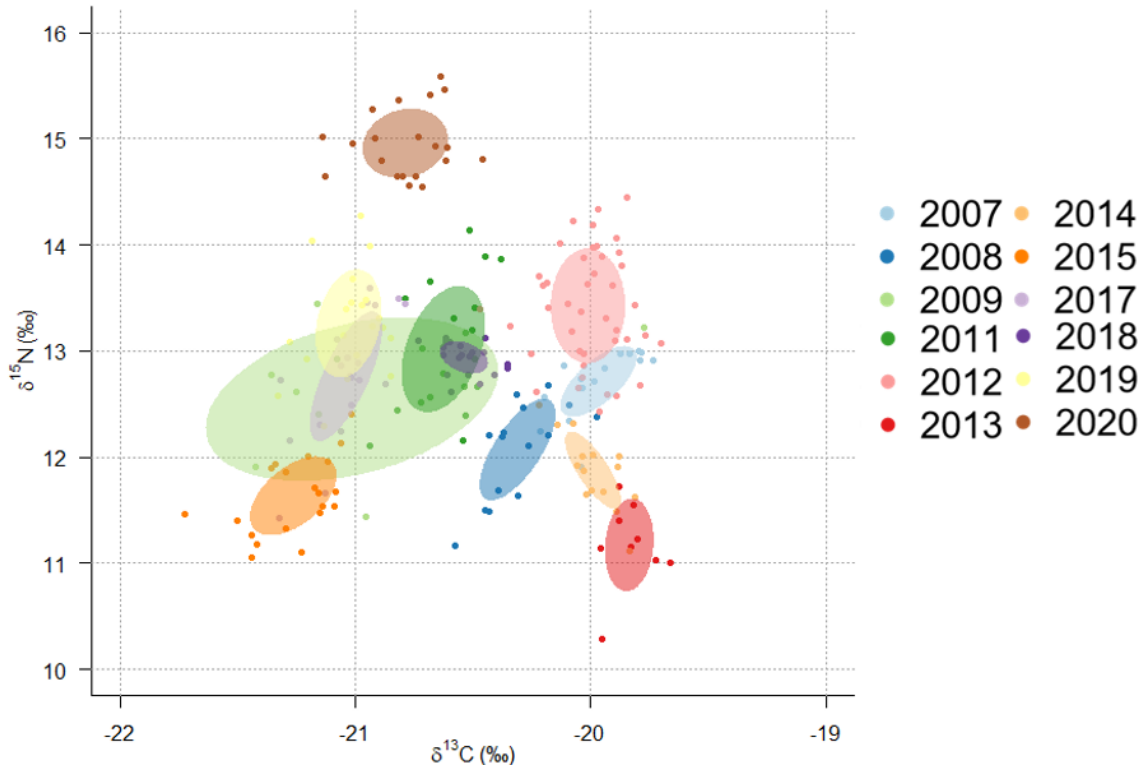

Figure 6. Standard ellipse area (SEA_C) showing stable isotope niches based on $\delta^{13}C$ and $\delta^{15}N$ values from blood from adult kittiwakes sampled during the chick-rearing stage from 2007-2009, 2011-2015, and 2017-2020 at the Krykkjefjellet colony.

Table 3. Yearly size of SEA_C for stable isotope niches shown in Figure 6, calculated from $\delta^{15}N$ and $\delta^{13}C$ values from blood sampled from adult kittiwakes during chick-rearing period at the Krykkjefjellet colony.

Years	SEA_C
2007	0.12
2008	0.16
2009	1.41
2010	-
2011	0.31
2012	0.27
2013	0.14
2014	0.09
2015	0.18
2016	-
2017	0.21
2018	0.04
2019	0.22
2020	0.18

3.3. Relationship between variation in stable isotope ecology and variation in regurgitate content

No significant relationships were found between $\delta^{15}N$ and FO_{Arctic}, $FO_{Herring}$ or $FO_{PolarCod}$ ($0.46 \geq F \geq 1.11$, $0.30 \geq P \geq 0.49$) or between $\delta^{13}C$ and FO_{Arctic}, $FO_{Herring}$ or $FO_{PolarCod}$ ($0.07 \geq F \geq 0.23$, $0.63 \geq P \geq 0.79$). Though not significant, $\delta^{13}C$ showed a positive relationship with FO_{Arctic}, $FO_{Herring}$ and $FO_{PolarCod}$, while $\delta^{15}N$ showed a negative relationship with $FO_{Herring}$ and a positive relationship with both FO_{Arctic} and $FO_{PolarCod}$ (Figure 7).

The relationships between SEA_C and FO_{Arctic}, $FO_{Herring}$ and $FO_{PolarCod}$ were all significant ($P < 0.01$, $16.70 \geq F \geq 30.57$). SEA_C and both FO_{Arctic} and $FO_{PolarCod}$ showed a slightly positive relationship (both slope = 0.01) while SEA_C and $FO_{Herring}$ showed a slightly negative relationship (slope = -0.01; Figure 7).

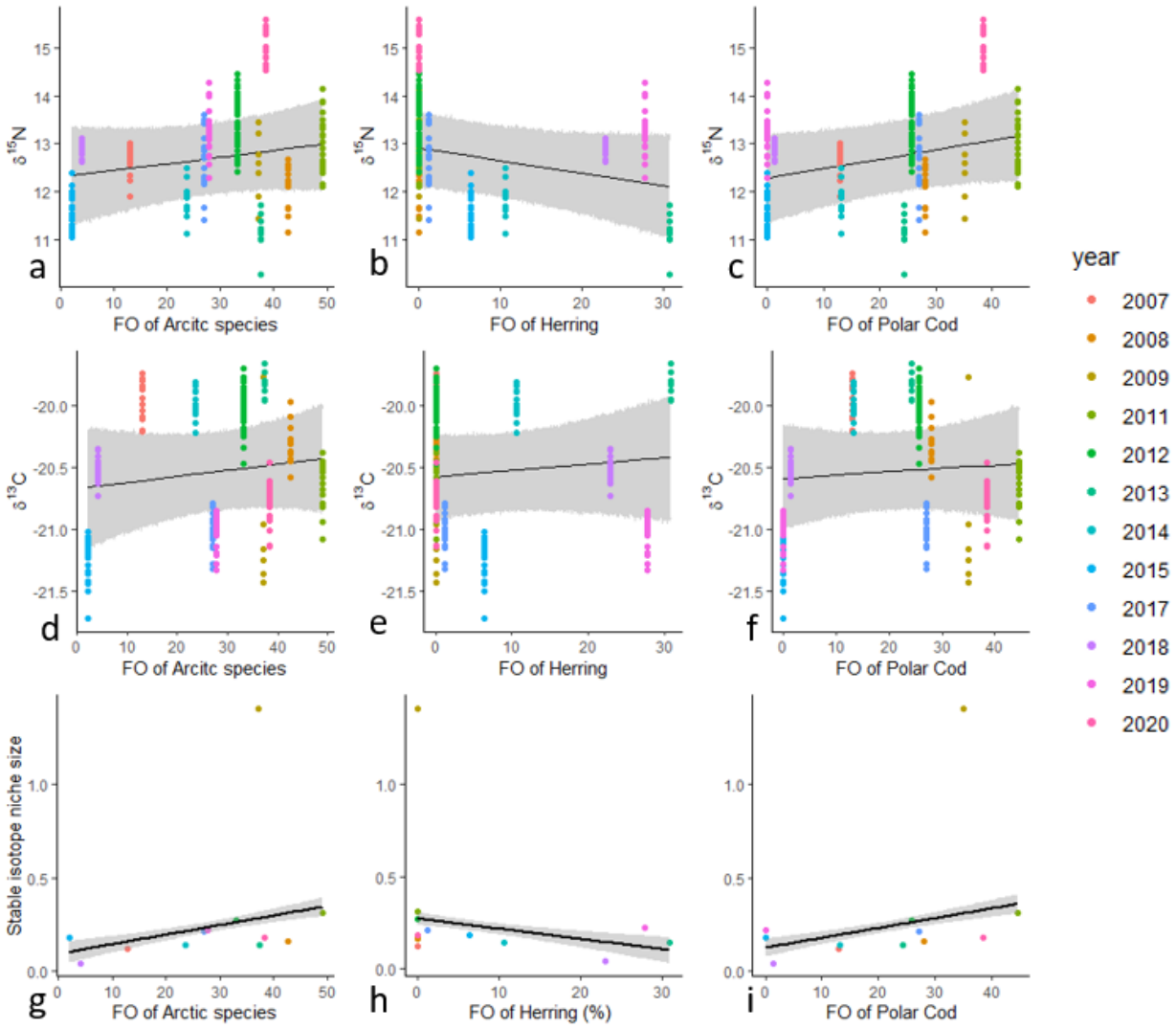

Figure 7. Relationships between FO_{Arctic}, $FO_{Herring}$ and $FO_{PolarCod}$ from regurgitate samples from chick and adult kittiwakes from different colonies in Kongsfjorden during the breeding season in relation to $\delta^{15}N$ (a, b, c), $\delta^{13}C$ (d, e, f) and stable isotope niche size (g, h, i) for blood sampled from adult kittiwakes during the chick-rearing period in Kongsfjorden. All samples were collected from 2007-2009, 2011-2015 and 2017-2020.

3.4. Between-year variation in contaminants

Concentrations of all contaminants showed between-year variation with no clear trend ($P < 0.01$; $16.72 \geq F \geq 37.17$; Figure 8). Concentrations for 2013 and 2014 were significantly lower compared to contaminants from other years ($P < 0.03$), except for HCB (2007, $0.57 \geq P \geq 0.81$) and β-HCH (2011, P=0.99). Concentrations for these two years, in addition to 2012, were analysed on plasma and converted to concentrations in whole blood. The year 2019 had high variance in contaminant levels.

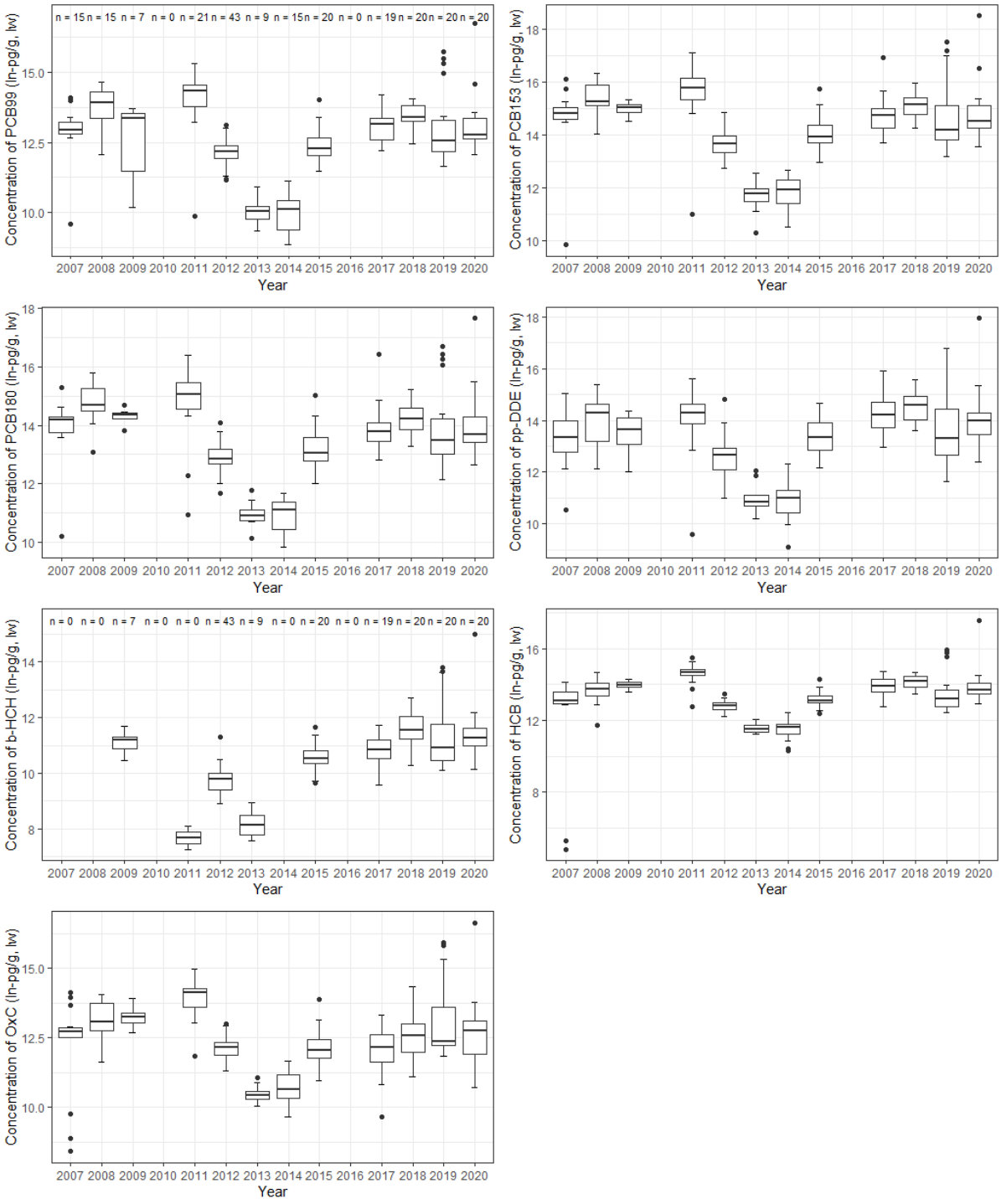

Figure 8. Between-year variation in concentrations of the studies compounds, measured in adult kittiwake blood sampled during the chick-rearing period from 2007-2009, 2011-2015 and 2017-2020 in the Krykkjefjellet colony. Note that the y-axis for each contaminant vary in scales. n: sample size, is the same for all contaminants as shown for PCB 99, except for *β*-HCH. The horizontal line in the box plot indicates the median of the data, while the box is constituted by the 25 % and 75 % quartiles, and the whiskers represent 1.5 * IQR.

3.5. Results from model selection

All contaminants had five or more candidate models identified as parsimonious (with $\Delta AIC_C < 2$), resulting that no model stood out as a clear best fit for the observed variation in concentration for any of the seven studied contaminants (Table 4). For three out of seven contaminants, i.e., *p,p'*-DDE, HCB and OxC, the null model was identified as the one with lowest AIC_C. Two out of seven contaminants, both PCB congeners, gave model 2.3, containing only $FO_{Herring}$ as the one with the lowest AIC_C. In general, only models including either only stable isotope values or only FO values seem to be present low AIC_C values, with some exceptions. There is no obvious trend for one model showing up more often than others, except maybe model 2.3 with $FO_{Herring}$ as explanatory factor, which shows up among the four models with lowest AIC_C for all contaminants except for *β*-HCH. The marginal R^2 values, describing the proportion of variance explained by the fixed factors alone, is in general very low, while the conditional R^2 values, that describes the proportion of variance explained by both the fixed and random factors, is in general high. This points towards that stable isotope values and prey item information does not really seem to contribute to explain the variation shown in contaminant levels, but that this variation is related to factors not included in this study but represented by the null-model with *year* as random effect factor.

Table 4. Most parsimonious models ($\Delta AIC_C < 2.00$) explaining concentration variability for the studied compounds measured in kittiwake blood sampled at the Krykkjefjellet colony during 2007-2009, 2011-2015 and 2017-2020. *Year* was included as a random effect for all models. Models are sorted by AIC_C, the most parsimonious model having the lowest value, and the difference is indicated by ΔAIC_C. *w* shows the Akaike's weight (Anderson & Burnham, 2002), the Marginal R^2 shows the proportion of variance explained by the fixed factors alone while the condition R^2 shows the proportion of variance explained by both the fixed and the random factors. See Table Appendix.12-18 for all models.

	Model	Factor	*P*	*F*	Slope	ΔAIC_C	*w*	Marginal R^2	Conditional R^2
PCB99	Model 1.2	$\delta^{13}C$	0.13	2.37	-0.45	0.00	0.18	0.03	0.65
	Model 0		<0.01	1119	12,53	0.35	0.15	0.00	0.68
	Model 2.3	$FO_{Herring}$	0.20	1.85	-0.04	0.39	0.14	0.10	0.68
	Model 1.1	$\delta^{13}C$	0.11	2.46	-0.49	1.77	0.07	0.04	0.64
	Model 1.1	$\delta^{15}N$	0.61	0.26	0.06				
	Model 3.3	$\delta^{13}C$	0.11	2.57	-0.47	1.82	0.07	0.12	0.64
	Model 3.3	$\delta^{15}N$	0.59	0.29	0.05				
	Model 3.3	$FO_{Herring}$	0.21	1.81	-0.04				
	Model 3.5	$\delta^{13}C$	0.11	2.59	-0.48	1.91	0.07	0.03	0.70
	Model 3.5	FO_{Arctic}	0.67	0.19	0.01				
PCB153	Model 2.3	$FO_{Herring}$	0.16	2.26	-0.04	0.00	0.21	0.11	0.66
	Model 0		<0.01	1593.58	14.27	0.38	0.17	0.00	0.66
	Model 1.3	$\delta^{15}N$	0.25	1.33	0.14	1.06	0.12	0.01	0.64

	Model 2.2	$FO_{PolarCod}$	0.49	0.47	0.02	1.90	0.08	0.03	0.66
	Model 1.4	SEA_C	0.51	0.45	0.76	1.92	0.08	0.01	0.66
PCB180	Model 2.3	$FO_{Herring}$	0.15	2.38	-0.05	0.00	0.18	0.12	0.67
	Model 1.3	$\delta^{15}N$	0.14	2.25	0.18	0.26	0.16	0.02	0.65
	Model 0		<0.01	1274.26	13.49	0.49	0.14	0.00	0.67
	Model 2.2	$FO_{PolarCod}$	0.44	0.60	0.02	1.87	0.07	0.04	0.67
	Model 1.4	SEA_C	0.47	0.56	0.84	1.91	0.06	0.02	0.67
***p,p'*-DDE**	Model 0		<0.01	1496.51	13.27	0.00	0.17	0.00	0.57
	Model 2.3	$FO_{Herring}$	0.31	1.12	-0.03	0.79	0.11	0.05	0.57
	Model 1.3	$\delta^{15}N$	0.26	1.25	0.15	0.90	0.11	0.01	0.54
	Model 1.1	$\delta^{13}C$	0.15	1.51	-0.46	0.96	0.10	0.06	0.49
	Model 1.1	$\delta^{15}N$	0.15	2.07	0.19				
	Model 1.2	$\delta^{13}C$	0.32	1	-0.32	1.02	0.10	0.02	0.53
	Model 2.2	$FO_{PolarCod}$	0.74	0.12	0.01	1.96	0.06	0.01	0.57
	Model 3.3	$\delta^{13}C$	0.20	1.38	-0.42	1.96	0.06	0.01	0.57
	Model 3.3	$\delta^{15}N$	0.21	1.96	0.17				
	Model 3.3	$FO_{Herring}$	0.35	0.95	-0.03				
β-HCH	Model 2.1	FO_{Arctic}	0.11	2.6	-0.05	0.00	0.20	0.19	0.76
	Model 0		<0.01	480.90	10.31	0.75	0.14	0.00	0.77
	Model 3.4	SEA_C	0.37	0.14	1.14	0.89	0.13	0.24	0.76
	Model 3.4	FO_{Arctic}	0.07	3.31	- 0.06				
	Model 2.2	$FO_{PolarCod}$	0.24	1.67	- 0.04	0.92	0.13	0.13	0.77
	Model 3.6	$\delta^{15}N$	0.80	0.01	0.03	1.97	0.07	0.19	0.75
	Model 3.6	FO_{Arctic}	0.10	2.7	-0.05				
HCB	Model 0		<0.01	2073.98	13.26	0.00	0.15	0.00	0.48
	Model 1.3	$\delta^{15}N$	0.17	1.87	0.18	0.15	0.14	0.02	0.45
	Model 2.2	$FO_{PolarCod}$	0.34	0.91	0.02	1.02	0.09	0.04	0.48
	Model 2.3	$FO_{Herring}$	0.36	0.92	-0.02	1.02	0.09	0.04	0.48
	Model 1.4	SEA_C	0.41	0.75	0.76	1.22	0.08	0.02	0.47
	Model 1.1	$\delta^{13}C$	0.38	0.48	-0.27	1.33	0.08	0.05	0.39
	Model 1.1	$\delta^{15}N$	0.11	2.58	0.21				
	Model 2.1	FO_{Arctic}	0.56	0.34	0.01	1.67	0.07	0.02	0.48
	Model 1.2	$\delta^{13}C$	0.78	0.08	-0.09	1.91	0.06	0.00	0.45
	Model 3.6	$\delta^{15}N$	0.21	1.73	0.17	1.95	0.06	0.04	0.45
	Model 3.6	FO_{Arctic}	0.62	0.25	0.01				
OxC	Model 0		<0.01	1870.23	12.37	0.00	0.16	0.00	0.50
	Model 2.3	$FO_{Herring}$	0.22	1.71	-0.03	0.18	0.15	0.07	0.50
	Model 1.4	SEA_C	0.28	1.32	0.96	0.59	0.12	0.03	0.48
	Model 2.2	$FO_{PolarCod}$	0.33	0.95	0.02	0.98	0.10	0.05	0.50
	Model 2.1	FO_{Arctic}	0.35	0.89	0.02	1.04	0.10	0.04	0.50
	Model 1.3	$\delta^{15}N$	0.48	0.51	0.09	1.50	0.08	0.01	0.48
	Model 1.2	$\delta^{13}C$	0.83	0.05	-0.07	1.98	0.06	0.00	0.49

4. Discussion

The aim of my thesis was to investigate if variation in contaminant concentrations in kittiwakes from Kongsfjorden, Svalbard, could be explained by changes in their diet, earlier observed to be related to ongoing climate change (Vihtakari et al., 2018). Between-year variation in prey species is still occurring, showing variations between Arctic and Atlantic diet items. There was no correlation between the variation in stable isotope values ($\delta^{13}C$ and $\delta^{15}N$) at the individual level and variation in annual population level estimated of the frequency of occurrence in diet items. The changes in diet, quantified by either regurgitate data, stable isotope data, or a combination of these, did not explain the observed variation in contaminant levels in kittiwakes, although significant between-year variation can be observed for contaminant concentrations. The model selection showed that the null-model often was among the best ranked models. The influence by between-year variation might point towards effects from factors not included in the model selection but represented by the null-model, with *year* as random factor. Still, the models pointed to some explanatory variables that occurred in the better models for several of the pollutants, in particular trophic position and $FO_{Herring}$.It is possible that there is a relationship between these factors and contaminant levels, in combination with other factors not included here.

4.1. Between-year variation in prey items from regurgitate samples

The results show variation in the occurrence of prey species from year to year (Figure 2 and 3). There did not seem to be any obvious trend or pattern of increasing or decreasing species or groups of any kind and results only showed what seemed to be between-year fluctuations. Vihtakari et al. (2018) studied regurgitate samples from Kongsfjorden, but while they used samples from 1982-2016, my thesis continued on this time-series, and used samples from 2007-2020. Vihtakari et al. (2018) found a shift from Arctic prey dominance until 2006, and after that a more mixed diet with high contributions of Atlantic species. This mixed diet continues in the recent years following after Vihtakari et al. (2018). Assuming that the current climate warming will continue, one could expect a higher contribution of Atlantic fish in Kongsfjorden in the future. Griffith et al. (2019) claim that polar species might not be able to shift their ecological geographical range northward in response to climate warming, and that Arctic species must either adapt or go extinct. This would affect the entire Arctic food web, and some of these effects are already visible. There is evidence which claims that marine mammals appear to decline in body condition due to poorer nutritional quality of Atlantic prey fish (Hamilton et al., 2017), and that the Atlantic zooplankton species, which are lower in fat content, have become increasingly dominating in the pelagic community (Huenerlage et al., 2016). However, other studies argue that previous concern regarding replacement of Arctic zooplankton may be unsupported, and that the

incoming Atlantic species may continue to support top predators in the European Arctic (Renaud et al., 2018). Even though my thesis did not find any clear trends towards an increasing contribution of Atlantic species, this change might still be happening, and might have consequences for geographical distribution, energy transfer and contaminant exposure in Arctic species, such as the kittiwake.

Unidentified fish have been a high contribution in the kittiwake diet since 2016 (Figure 2). This might mask the contribution of fish that should belong either to the Arctic or the Atlantic group. Polar cod is the only fish species defined as Arctic. The otoliths of polar cod are bigger than otoliths from both capelin and herring (Christiansen et al., 2005), and they are often present and easy to find in regurgitate samples containing polar cod. This could mean that a large proportion of unidentified fish are not Arctic but belong either to the group of Intermediate or Atlantic species. A lower proportion of unidentified fish might have led to other results.

There is a small contribution from the group defined as Mesopelagic for some years (Figure 3). The kittiwake is a surface-feeding seabird, and the explanation behind these mesopelagic species in their diet might be upwelling waters by the tidal glacier fronts that flow into the fjord (Vihtakari et al., 2018). Mesopelagic fish have been observed near the surface by glacier fronts (Vihtakari et al., 2018), and these are known feeding areas for kittiwakes (Lydersen et al., 2014). My thesis followed the grouping done by Vihtakari et al. (2018), and the four main groups for prey items were kept. In hindsight, it could be argued that it would be better to exclude the category of mesopelagic species and distribute the prey items from this group according to the three remaining groups. Still, their contribution is very small, so placing them into the other groups may not have an effect.

Frequency of occurrence was used to describe the kittiwake diet in this study, but there are better approaches that can be used to quantify the contribution from different prey items. Using frequency of occurrence can reveal presence/absence information related to prey species, but other approaches to quantify the diet give better insight into feeding ecology. Frequency of occurrence does not take into consideration prey size or prey behaviour. The importance of the same number of large fish versus the same number of smaller invertebrates is not well reflected by using frequency of occurrence. Using frequency of occurrence for prey with different spatial distribution patterns might also lead to unreliable comparisons. The presence of schooling fish in regurgitate samples indicates a rather large relative abundance of this species present in the fjord, while the presence of a non-schooling fish does not indicate anything more than just the presence of this one fish. Other methods, such as percentage of wet weight or relative abundance of prey species might give a more representative picture. Ramos et al. (2009) used biomass percentages to quantify regurgitate samples in their study, and this would have been a better approach to use in my thesis, but this was not possible, due to inconsistent reporting in recent years.

4.2. Stable isotope values in relation to diet items from regurgitate samples

Previous studies comparing the use of regurgitates and stable isotope analyses to better understand a species feeding ecology have shown that stable isotope analysis is a reliable method to determine the importance of certain exploited resources (Ramos et al., 2009; Weiser & Powell, 2011). Still, it does not provide the detailed species information achieved by the conventional dietary analysis (Ramos et al., 2009; Weiser & Powell, 2011). Ramos et al. (2009) measured stable isotope ratios from yellow-legged gulls (*Larus michahellis*) from the Mediterranean coast from four different colonies and compared these with regurgitate samples. They found that different diets from different colonies could be distinguished by using stable isotope values (δ^{13}C and δ^{15}N; Ramos et al., 2009). In my thesis, no clear relationship was found between stable isotope values and diet, either between diet groups or diet items. Still, the diet from the study of Ramos et al. (2009) varied a lot between different colonies, with contribution from both the marine and the terrestrial food chains, and meat waste from dump sites. The stable isotopic signatures from these contributions might be more dissimilar compared to the differences between Arctic and Atlantic marine species.

Results showed a significant relationship between SEA_C and FO_{Arctic}, $FO_{Herring}$ and $FO_{PolarCod}$ (Figure 7). It is surprising that the relationship between SEA_C and $FO_{Herring}$ is negative, while relationship between SEA_C and $FO_{PolarCod}$ is positive. It was unexpected, as both $FO_{Herring}$ and $FO_{PolarCod}$ represents one species, that the increase in one specific species the kittiwake diet can lead to increased stable isotope niche size. The results show that it does not seem to be a clear concept of stable isotope niche size as a proxy related to the FO of prey items.

The regurgitate samples give a "snapshot in time" of the species composition in the fjord while the stable isotope values reflect dietary assimilation over a slightly longer time. Still, the tissue turn-over rate for blood is expected to be 10-14 days (Boecklen et al., 2011). The species composition in the fjord is not expected to change drastically during the time of sampling, and while each sample represent a "snapshot", the sum of all samples represents the state of the fjord during that season. In this way, the potential time aspect of the regurgitate samples and stable isotope values are not expected to be that different. Still, the two approaches are expected to provide complementary information about the feeding ecology of kittiwakes. The regurgitate samples provide the taxonomic detail and information on the state of the fjord, while the stable isotopes takes into account all assimilated food and thereby avoids digestibility biases (Ramos et al., 2009).

The variation in δ^{13}C showed two different clusters, where 2009, 2015, 2017 and 2019 had low values, while 2007, 2012, 2013 and 2014 had high values (Figure 4). There were no obvious differences in diet items between these two clusters (Figure 2 and 3). The differences in δ^{13}C value from consumers with a benthic or near-shore carbon source is typically enriched by 5‰, compared to consumers deriving carbon from pelagic

phytoplankton (Ricca et al., 2007). The difference in $\delta^{13}C$ levels found in this study was around 2‰, which is less than expected from a shift between benthic and pelagic sources. Still, the difference observed in $\delta^{13}C$ levels might be attributed to differences in offshore and coastal feeding (Amélineau et al., 2019). Kittiwakes in Kongsfjorden feed mainly inside the fjord, but move further out of the fjord when prey abundance in the fjord is low (Vihtakari et al., 2018). This might indicate that during 2009, 2015, 2017 and 2019, which show low $\delta^{13}C$ values, kittiwakes foraged in more offshore areas, while during the years 2007, 2012, 2013 and 2014, which show higher values for $\delta^{13}C$, indicate the use of more coastal habitats.

The range found in $\delta^{15}N$ values in this present study (11.05-15.58‰) is larger, but still comparable with values found for kittiwakes (12.9-14.2‰) by Borgå et al. (2005). In my study, the years with the lowest value for $\delta^{15}N$ were 2013 and 2015 (Figure 5) and the main prey item for kittiwakes during 2015 was krill (Figure 2). Since krill occupies a low trophic position (Hop et al., 2002), low $\delta^{15}N$ values for this year were expected. Krill was also a major contributor to the kittiwake diet in 2010, but stable isotope data from this year is lacking. A comparison between 2015 and 2010 could have indicated if the low value of $\delta^{15}N$ was indeed related to the high contribution of krill. This illustrates the importance of complete time-series for comparisons between years. 2013 also had low $\delta^{15}N$ values, but the diet during this year was much more varied compared to 2015, and there was no clear indication as to why $\delta^{15}N$ values were low in 2013.

2020 was the year with the highest $\delta^{15}N$ value. The diet during this year was composed mainly of fish (Figure 2). Wassmann et al. (2006) states that polar cod, herring, and capelin, which are the main contributors of fish in the kittiwake diet, occupy the same trophic level. Since higher trophic positions are indicated by high $\delta^{15}N$ values (Amélineau et al., 2019; Ramos et al., 2009; Weiser & Powell, 2011), the high $\delta^{15}N$ value for 2020 was expected. Still, this distribution of prey items is not unique for 2020. The diet from 2020 and 2017 are quite similar, but the $\delta^{15}N$ value differs by more than 2‰. The variation in diet items cannot explain this difference in $\delta^{15}N$ value. Comparing values with studies done by Borgå et al. (2005) show that the range for kittiwakes from 2020 (14.54-15.58‰) from my thesis is more similar to the range occupied by black guillemot (*Cepphus grylle*; 13.7-15.0‰) than by kittiwakes (12.9-14.2‰). Borgå et al. (2005) points out that this higher trophic position for the black guillemot might be related to feeding on demersal or larger fish during pursuit diving. This pursuit diving behaviour is different from the surface feeding behaviour of kittiwakes and cannot explain the high $\delta^{15}N$ values for kittiwakes during 2020.

The difference between years with low (2013, 2014, 2015) and high (2020) $\delta^{15}N$ is between 3 - 4‰, which is the typical trophic enrichment for $\delta^{15}N$ in high-latitude environments according to Ricca et al. (2007). There was a difference in diet between the years 2015 and 2020, as the diet in 2015 mainly consisted of krill and the diet from 2020 mainly consisted of fish. These differences in diet relates well to the observed difference

in δ^{15}N values, and relates to the described differences in δ^{15}N observed between trophic levels (Ricca et al., 2007). Still, there is no clear difference in diet between the remaining years with low value (2013 and 2014) and the year with high value (2020) for δ^{15}N. A possible explanation to this might be related to underrepresentation of lower tropic level prey like small invertebrates from regurgitate samples, due to samples being too digested to identify these (Ramos et al., 2009).

4.3. Variation in contaminant concentrations

The results show significant between-year variation in contaminant levels (Figure 8). For all contaminants except *β*-HCH, the years 2013 and 2014 have the lowest values. These years, in addition to 2012, were analysed on plasma and the values were converted to whole blood values using a conversion factor to allow comparisons between all years. It is difficult to determine if the observed low values from 2013 and 2014 are due to analysing a different blood component, or if the contaminant levels for these years actually are lower compared to other years. The fact that values from 2012 are not as low as values from 2013 and 2014 might indicate that the conversion factor is reliable. Still, it is necessary to treat values from these years with caution. Bustnes et al. (2010) presented the same problem with contaminant concentrations from different tissues, and they resolved this by only converting values from wet weight to lipid weight. For future studies it should be investigated how different these two approaches are regarding concentrations measured in different tissue, but the best option would be to analyse for contaminants on the same tissue during the entire study period.

Comparing the levels of contaminants found in my thesis with other studies shows that results are within the range that can be expected (Goutte et al., 2015; Nordstad et al., 2012). A study done in Hornøya, Norway, showed PCB levels in kittiwakes similar to those found in Kongsfjorden (Sagerup et al., 2014). However, the kittiwakes in Kongsfjorden showed more similar values of *p,p*-DDE to the Atlantic puffin, compared to lower values found in the kittiwakes in Hornøya by Sagerup et al. (2014). The levels for HCB were higher in kittiwakes from Kongsfjorden compared to both kittiwakes and Atlantic puffins in Hornøya.

4.4. Contaminant concentrations in relation to dietary ecology

The model selection procedure showed no clear single model that could be identified as the most parsimonious for any of the studied contaminants. However, some models with certain factors showed up more often than others. The two models containing only $FO_{Herring}$ or δ^{15}N seemed to be identified more often than most other models to explain contaminant exposure. Yet, these did not show significant effect of these factors on contaminant concentrations. Comparing these models to the null-model reveals that the conditional R^2 often are the same as for then null-model (Table 4). Borgå et al. (2005) found that PCB concentrations in Arctic seabirds could not be explained directly by either carbon source or trophic position,

but only by combining these dietary parameters with other factors such as migratory pattern, age and contaminant metabolism. This aligns with the findings in my thesis. Parts of the diet could impact the contaminant concentrations in kittiwakes, but dietary information alone, either proxied with stable isotope values or regurgitate information, cannot explain the variation in contaminant levels.

In my thesis the stable isotope values and the contaminant concentrations were measured in blood from the same bird, while regurgitate samples could not be linked to any specific bird. There is a possibility that the diet would have shown a stronger relationship to contaminant levels if the diet samples also could be linked to the same bird that the blood sample for contaminant analysis was taken from.

Since there was no clear relationship between the two different approaches to dietary ecology, it could be assumed that both these approaches could affect contaminant exposure. The marginal R^2 is in general low for most models but is higher for those models combining information from both regurgitate samples and stable isotope values. Nonetheless, these combined models rarely have a very low AIC_C value, as the AIC_C approach "punishes" models for increased complexity. Combining the two different dietary ecology approaches did not seem to help in explaining the contaminant variation compared to keeping the two approaches separate.

Both $\delta^{13}C$ and $\delta^{15}N$ have previously been related to contaminant exposure (Hop et al., 2002; McKinney et al., 2012), and since both stable isotope samples and contaminant samples were obtained from the same bird, stable isotope values were expected to have an effect on contaminant levels. It was unexpected that none of them, either in combination or separate, seemed to be important drivers for the variation in contaminant exposure according to the model selection (Table 4). Still, they show up among the models with the lowest AIC_C values, but none could be identified as the one single parsimonious model for any contaminants.

Carbon source has previously shown to influence some lower chlorinated PCBs (McKinney et al., 2012). McKinney et al. (2012) showed that across trophic positions, benthic carbon source ($\delta^{13}C$) was associated with higher concentrations of lower chlorinated PCBs. The lowest chlorinated PCB congener included in this study (PCB 99) rated the model containing $\delta^{13}C$ with lowest AIC_C value. As this was not the case for any of the other contaminants in the study, this might point towards a more plausible relationship between $\delta^{13}C$ and the lower chlorinated PCBs, in agreement with what was found by McKinney et al. (2012).

The differences in $\delta^{13}C$ values in this study could be related to differences in offshore and coastal feeding, where higher levels of $\delta^{13}C$ are attributed to more coastal habitats (Amélineau et al., 2019). Studies on polar bears that feed mainly in offshore areas showed higher levels of contaminants compared to more coastal feeding bears (Blévin et al., 2020). This difference in contaminant level based on different feeding areas is also possible for the kittiwakes in Kongsfjorden. Even though the relationship between $\delta^{13}C$ and

contaminants is not very clear from this study, the relationship between offshore and inshore feeding in kittiwakes in relation to $\delta^{13}C$ and contaminant exposure for lower chlorinated PCBs can be a topic for future research.

It could also be expected that the model containing $\delta^{15}N$ as an explanatory factor would be an important driver for contaminant exposure, since many contaminants have been documented to biomagnify in the food chain and are often found in high levels in top predators (Bearhop et al., 2002; Hop et al., 2002). For both PCB 153, PCB 180, *p,p'*-DDE and HCB the model containing only $\delta^{15}N$ came out among the third best ranked models. Since this model showed up with a low AIC_C value for several contaminants, it can be assumed that there is a relationship between contaminant exposure and trophic level, but the connection is not as clear as shown in other studies (Fisk et al., 2001; Hop et al., 2002). Borgå et al. (2005) found that even though kittiwakes showed lower $\delta^{15}N$ values compared to black guillemot, the kittiwake had higher PCB concentrations. Borgå et al. (2005) relates this to different turnover rates of contaminants and protein and suggests that the diet from the over-wintering areas for kittiwakes in more contaminated areas or the occasionally feeding on seal blubber from carcasses after polar bear kills. These explanations for a lack of a clear relationship between trophic position and contaminant exposure are also relevant in my thesis.

For PCB 153 and PCB180, the model containing only $FO_{Herring}$ was ranked with the lowest AIC_c value. This might point towards a relationship between Atlantic species and PCB contaminant levels in the kittiwake. Atlantic fish species such as herring have been identified to function as biovectors transporting contaminants from more polluted areas into the Arctic environment (Hallanger, Warner, et al., 2011; McKinney et al., 2012; Morris et al., 2016). However, the most recent AMAP report (2022) points out that the importance of these biovectors as new sources of contaminants into the Arctic region is unknown. The previous observed shift in prey items from Arctic to more Atlantic species was identified as driver for the significantly higher PCB concentrations in polar bears (Kleivane et al., 2000). Later this was questioned by Tartu et al. (2017), who claimed that the high PCB concentrations was a result of declining sea ice and therefore reduced feeding opportunities and consequently declining body condition, rather than changes in diet composition. Still, if the incoming Atlantic species have poorer nutritional value compared to Arctic species, this might lead to declining body condition in Arctic species (Gabrielsen, 2009; Hamilton et al., 2017). Declining body condition might lead to remobilization of contaminants from adipose tissue which elevates contaminant levels in the blood (Bustnes et al., 2017). In this manner the incoming species do not function directly as biovectors, but this indirectly has consequences for contaminant levels in Arctic wildlife. My study did not take into consideration the aspect of body condition, regarding incoming species and nutritional value in relation to contaminant exposure, because this was outside of the scope of this thesis. The nutritional value of incoming

Atlantic species could be topic for future research, and body condition could be included as a factor to explain contaminant variability in kittiwakes in future studies.

$FO_{PolarCod}$ seemed to be a more important driver for contaminant exposure compared to FO_{Arctic}. The difference between these two factors is that FO_{Arctic} includes all Arctic species, also those on lower trophic levels, while $FO_{PolarCod}$ only includes this one species which has a high trophic position. Maybe this can also be related to the findings presented earlier by Borgå et al. (2005), that the diet is an important factor in contaminant exposure, in addition to other factors. $FO_{PolarCod}$ takes into consideration both trophic position and origin of species (Arctic or Atlantic) but includes these in the same factor. This leads to a lower AIC_C value than either combining these (FO_{Arctic} and $\delta^{15}N$) or just using the FO_{Arctic} that does not take trophic position into account.

4.5. Strengths, weaknesses, and future studies

Vihtakari et al. (2018) state that there was a shift in Kongsfjorden in 2006 from Arctic prey dominance in kittiwakes, to a more mixed diet with high contribution of Atlantic species. Since contaminant data for my study only go back to 2007, it is possible that the time range of this study did not capture the shift observed from an Arctic to a more mixed diet, and each year during the whole time period for 2007-2020 is composed of this mixed diet. An accompanying change in contaminant levels may therefore have taken place before 2007 and therefore not captured by my study. The time series lack data for two out of 14 years (2010 and 2016). Still, the total of twelve years should be sufficient for the between-year comparisons done in this study.

Another drawback is the problem with contaminant analyses being conducted on different tissue during the study period. The years analysed on plasma showed low contaminant values and makes it hard to trust these data, even though they were converted to whole blood concentrations. There is a possibility that these differences in concentration might have impacted the model selection. For future research I recommend to analyse for contaminants on the same tissue to allow for as trustworthy comparisons as possible.

There is a large contribution of unidentified fish in the regurgitate samples from 2016 - 2020. These items are therefore placed in the category of intermediate items, even though they might belong in either the Arctic or the Atlantic group. Ideally there would be fewer unidentified fish, but this is hard to accomplish when otoliths are not found and when samples are very digested. Results from regurgitate samples are biased towards prey types that are more resistant to digestion, and probably underestimate the importance of small and soft prey items (Ramos et al., 2009). Future genetic methods that can be used for species identification might help overcome this problem.

Bustnes et al. (2017) showed that there are large differences in contaminant levels during the different stages of the breeding period. My study used contaminant concentrations measured in blood samples taken during chick-rearing period. This makes for trustworthy comparisons for between-year variations in contaminant concentrations, knowing that differences in concentrations are not related to sampling during different stages in the breeding period. Samples were taken at different times during the chick-rearing period, but ideally, all blood samples would have been taken at same number of days after hatching.

The blood samples were also taken from the same colony during the entire study period (Krykkjefjellet), which rules out any hidden effect that could arise from different sampling colonies. Regurgitate samples on the other hand were collected from different colonies in Kongsfjorden. This gives a bigger sample size compared to if only samples from Krykkjefjellet were used. Still, samples from several colonies makes for greater uncertainties that the samples are representative for the birds in the colony in focus. Bertrand et al. (2021) showed that the feeding areas for kittiwakes in Kongfjorden during the summer of 2017 were overlapping to a large extent for the colonies Krykkjefjellet, Ossian Sarsfjellet, and Observasjonsholmen, while feeding areas for kittiwakes from Irgensfjellet differed the most. Still, (Bertrand et al., 2021) used data from only one year, and the feeding area for kittiwakes might not be the same every year. Even though the regurgitate samples represent the available prey in the fjord, there might be local differences within the fjord and between the feeding areas for kittiwakes. To include all the regurgitate samples from all colonies is good to increase sample size, another alternative would be to just use samples from the colony in focus, that also could be traced back to the individual bird, so that both blood samples, stable isotope samples and regurgitate samples could be linked to the same individual.

As mentioned above, Atlantic fish species migrating to the Arctic have been identified as biovectors, bringing contaminants from polluted areas into the Arctic (Hallanger, Warner, et al., 2011; McKinney et al., 2012; Morris et al., 2016). The kittiwake is a migratory species itself and spends winter the West Atlantic (SEATRACK, 2022). A possible explanation to the lack of correlation between the Atlantic prey and contaminant exposure could be that the contribution of Atlantic prey species in the Arctic is not very different from the prey composition for the bird outside its breeding grounds, and therefore does not have a big effect on contaminant levels. It is possible that the effect of Atlantic species in the Arctic region can have a greater influence on Arctic endemic species.

In addition to diet, the degree of remobilization of contaminants from adipose tissue contributes to determining the concentrations of circulating contaminants (Bustnes et al., 2017). As mentioned above, this could be a consequence of poorer nutritional values from Atlantic prey (Hamilton et al., 2017), but it could also be related to the period of sampling. A study by Bustnes et al. (2017) compared contaminant exposure

in kittiwakes at different times in the breeding season. They found that for some contaminants the concentration was 2.5 times higher in the chick-rearing period compared to pre-breeding (Bustnes et al., 2017). This increase occurred concurrently with reduction in body mass, and was probably a result of remobilization of contaminants from adipose tissue (Bustnes et al., 2017). The kittiwakes body condition decreased 14.8 % and 8.4 % for females and males, respectively, during the first part of chick-rearing period (Moe et al., 2002). During this period contaminants from adipose tissue would increasingly remobilize and therefore increase blood concentrations as breeding progressed (Bustnes et al., 2017). Blood samples from my thesis were collected during chick-rearing period. Perhaps the remobilization of contaminants from adipose tissue masks the possible effect of diet during this period.

The variation in $\delta^{13}C$ values from this study might be related to differences in offshore and inshore feeding for kittiwakes in Kongsfjorden. A further investigation of this relationship, and also in relation to the lower chlorinated PCBs is a topic for future research. Body condition should also be included as an explanatory variable in future studies.

Many factors could contribute to contaminant exposure in kittiwakes, and among these are also environmental factors. It is known that contaminants are transported into the Arctic by both water currents and air currents (AMAP, 2016; Burkow & Kallenborn, 2000; Dietz et al., 2019). Environmental conditions have previously been shown to be related to contaminant levels in seabirds, such as demonstrated by the study of Bustnes et al. (2010) showing a correlation between the Arctic oscillation and contaminant exposure in glaucous gulls. Environmental factors such as temperature and sea ice extent are probably good proxies for Atlantification processes and should be included in future research on contaminant levels.

5. Conclusions

This study showed that diet is not a major driving factor in contamination levels in kittiwakes from Kongsfjorden. The null-model was often ranked as a strong model, pointing towards that interannual variabilities not included in this study is related to the significant between-year variations in contaminant levels. Still, some factors from my thesis, such as trophic level, carbon source and frequency of occurrence of herring, as a representative for Atlantic species, seem to be related to variation in contaminant levels. Other factors not included in this study, such as environmental factors and body condition should be included in future research, and more research is needed to predict if a warmer Arctic is a more contaminated Arctic.

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Appendix

Table of content, appendix

Appendix.1. Detailed analytical protocol for the persistent organic pollutant analysis.

All blood and plasma samples (*n*=425) were analysed for a variety of OHCs at the Norwegian Institute of Air Research (NILU) in Tromsø, Norway.

The analysis for PCB congeners targeted PCB 28,52, 101, 105, 114, 118, 123, 138, 153, 156, 157, 167, 180, 189 and 194. The targeted OCPs included HCB, *alpha-*, *beta-*, and *gamma*-hexachlorocyclohexane (*α-, β-, γ-*HCH), oxychlordane (Oxy-CD), *trans-* and *cis-* chlordane (t-CD and c-CD), *trans-* and *cis*-nonachlor (t-NC and c-NC), mirex, *p,p'*-DDT, *o,p'*-DDT *p,p'*-DDE, *o,p'*-DDE, *p,p'*-DDD and *o,p'*-DDD.

Samples were homogenized by gentle vortexing at 3,000 rpm for 10 seconds, then ~1 mL of sample was transferred to a pre-weighed 15 mL glass tube and weighed again. Samples were spiked with 20 µL of an internal standard mixture (POPs mix Krykkje, 25.02.21, table Appendix.4) and vortexed. 2 mL deionised water saturated with ammonium sulphate and 2 mL of ethanol was added and then vortexed. Samples were extracted twice with 6 mL of cyclohexane (total extraction with 12 mL),then shaken for 15 minutes on a shaking table and centrifuged for 5 minutes with 2000 rpm for phase separation, and supernatants were combined and concentrated to ~0,2 ml using RapidVap. Samples were covered lightly with aluminium foil and left to dry overnight. The glass tubes were weighted and lipid content was determined gravimetrically. Samples were re-dissolved in 0.5 mL of n-hexane.

Solid phase extraction clean-up was performed using the Zymark RapidTrace SPE Workstation. The RapidTrace Workstation loads each sample onto a separate SPE column. The columns were packed with ~1 g of florisil with one frit at the bottom and one at the top of the column. Frits and columns were cleaned with dichloromethane (DCM)before use. The mobile phase solvent was DCM :n-hexane (1:9, *v:v*), and florisil (0.15-0.25 mm mesh size, burned at 450^{O}C for 8 h) was chosen as the stationary phase to retain unwanted polar compounds from the matrix in the SPE column. Eluents were collected in 15 mL glass tubes. *Iso*-octane was added as keeper, and the samples were evaporated to 0.2 mL using a RapidVap. Extracts were transferred to gas chromatography (GC) vials with insert, and glass tubes rinsed with *n*-hexane. The extract volume was reduced to ~30 µL using Genevac ™ miVac Centrifugal Concentrator and 10 µL of recovery standard (^{13}C-PCB 159 (21.7 pg µL^{-1})) added. The vials were stored at 5^{o}C until gas chromatography mass spectrometer (GC-MS) analysis. For details on chemicals and instruments see table 5.1 and 5.2 in the Appendix.

Quantification of the targeted compounds was conducted using a Thermo scientific trace 1310 GC equipped with a Thermo scientific TSQ9000 triple quadrupole mass spectrometer, equipped with a DB-5MS column (length 30 m, 0.25 µm film thickness, 0.25 mm inner diameter) with precolumn (0.53 mm) and restriction capillary column (0.18 m). Helium was used as carrier gas with a flow rate at 1 mL min^{-1}. The initial temperature was set to 70^{o}C for 2 min, then increased with 15^{o}C min^{-1} until 180^{o}C, followed by an increase

of 5^{o}C min^{-1} up to 280^{O}Cwhich was then kept for 10 min. The analytes were quantified by using the ratio of the analyte and the internal standard responses. A standard curve was made from the peak areas of the labelled standards in the mass chromatogram. The LCQuan softwear package from Thermo Scientific (Version 2.6) was used for quantification.

Glassware used for analysis was washed, rinsed with acetone and cyclohexane and burned for 8 h in 450^{O}C. Glassware that had been in contact with biological material was treated with virkon before following normal washing procedure.

To validate the quality of the analysis one blank and one reference material (SRM®1958 - human serum from NIST) was concurrently analysed every 10th sample. The accuracy of the method ranged from 70 to 122% for all compounds, with the exception of oxychlordane (37% accuracy caused by coelution). If a blank showed presence of a compound, the limit of detection (LOD) for that compound was set to three times the blank signal. In all other cases, the LOD was set to three times the instrumental noise. LODs for the different compounds can be found in Table Appendix.8 .

Table Appendix.1. Yearly regurgitate sample size per colony and maturity. -: no sample taken.

	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020	Total
Krykkjefjellet	16	23	2	17	10	14	31	26	43	31	15	-	2	21	251
Irgensfjellet	38	25	3	66	45	56	-	-	-	23	19	-	-	-	275
Observasjonsholmen	-	-	5	-	-	-	-	-	-	11	23	19	-	-	58
Ossian Sarsfjellet	-	-	-	-	-	-	-	-	-	18	15	8	-	2	43
Unknown colony	6	5	55	18	5	7	42	-	-	9	10	23	14	-	194
Adult	55	40	63	72	39	45	37	9	15	33	-	-	15	13	436
Chick	2	12	2	28	12	23	7	0	-	21	13	1	-	10	131
Unknown maturity	3	1	-	1	9	9	29	17	28	38	69	49	1	-	254

Table Appendix.2. Targeted contaminants and their abbreviations.

Official name	Abbreviation
2,4,4,'-Trichlorobiphenyl	PCB 28
2,4',5-Trichlorobiphenyl	PCB 31
2',3,4-Trichlorobiphenyl	PCB 33
2,4,4'-Trichlorobiphenyl	PCB 37
2,2'4,4'-Tetrachlorobiphenyl	PCB 47/49
2,2'5,5'-Tetrachlorobiphenyl	PCB 52
2,2',4,4',5-Pentachlorobiphenyl	PCB 99
2,2',4,5,5'-Pentachlorobiphenyl	BCB 101
2,3,3',4,4'-Pentachlorobiphenyl	PCB 105
2,3',4,4',5-Pentachlorobyphenyl	PCB 118
2,3',4,4',5'-Pentachlorobyphenyl	PCB 123
2,2',3,4,4',5'-Hexachlorobiphenyl	PCB 138
2,2',3,4,5,5'-Hexachlorobyphenyl	PCB 141
2,2',3,4',5',6-Hexachlorobiphenyl	PCB 149
2,2',4,4',5,5'-Hexachlorobiphenyl	PCB 153
2,3,3',4,4',5-Hexachorobiphenyl	PCB 156
2,3,3',4,4',5'-Hexachlorobiphenyl	PCB 157
2,3',4,4',5,5'-Hexachlorobiphenyl	PCB 167
2,2',3,4,4',5,5'-Heptachlorobyphenyl	PCB 180
2,2',3,4,4',5',6-Heptachlorobiphenyl	PCB 183
2,2',3,4',5,5',6-Heptachlorobiphenyl	PCB 187

2,3,3',4,4',5,5'-Heptachlorobiphenyl	PCB 189
2,2',3,3',4,4',5,5'-Octachlorobiphenyl	PCB 194
Hexachlorobenzene	HCB
α-1,2,3,4,5,6-Hexachlorocyclohexane	*α*-HCH
β-1,2,3,4,5,6-Hexachlorocyclohaxane	*β*-HCH
γ-1,2,3,4,5,6 Hexachlorocyclohexane	γ -HCH
Heptachlor	Hept
Heplachlor epoxide	HeptEpox
Oxychlordane	OxC
Trans-chlordane	TC
Cis-chlordane	CC
Trans-nonachlor	TN
Cis-nonachlor	CN
Mirex	Mirex
o,p'-dichlorodiphenyltrichloroethane	*o,p'*-DDT
p,p'- dichlorodiphenyltrichloroethane	*p,p'*-DDT
p,p'-Dichlorodiphenyldichloroethylene	*o,p'*-DDD
p,p'-Dichlorodiphenyldichloroethylene	*p,p'*-DDD
o,p'-dichlorodiphenyldichloroethane	*o,p'*-DDE
p,p'-dichlorodiphenyldichloroethane	*p,p'*-DDE

Table Appendix.3. Yearly sample size of different blood (fractions) used for stable isotope and persistent organic pollutant analysis. Duplicate samples from POPs analysis from 2017,2018 and 2020 were used to make conversion factor from concentrations measured in plasma to whole blood.

	Stable isotope analysis		Persistent organic pollutant analysis	
	Erythrocytes	**Whole blood**	**Whole blood**	**Plasma**
2007	15		53	
2008	15		46	
2009		7	51	
2010			58	
2011	21		24	
2012	43			44
2013	9			23
2014	3	12		27
2015	8	12	20	
2016		22		
2017	19		19	12
2018	20		20	7
2019	20		20	
2020	20		20	5

Table Appendix.4. Internal standard mixture composition.

<table>
<tr><th>Amount (µL)</th><th>Substance</th></tr>
<tr><td>540</td><td>PESTI (06.19)

<table><tr><th>Compound</th><th>Concentration (pg µL^{-1})</th></tr><tr><td>^{13}C-<i>trans</i>-nonachlor (1.03.19)</td><td>73.9</td></tr><tr><td>^{13}C-<i>cis</i>-nonachlor (1.04.17)</td><td>51.3</td></tr><tr><td>^{13}C-<i>tans</i>-chlordane (1.04.17)</td><td>50.9</td></tr><tr><td>^{13}C-<i>cis</i>-chlordane (1.03.19)</td><td>74.6</td></tr><tr><td>^{13}C-oxychlordane (50.16)</td><td>516</td></tr><tr><td>^{13}C-heptachlor epoxid (06.19)</td><td>1003</td></tr><tr><td>^{13}C-heptachlor (06.19)</td><td>1212</td></tr><tr><td>^{13}C-dieldrin (06.19)</td><td>1002</td></tr><tr><td>^{13}C-mirex (06.19)</td><td>261</td></tr><tr><td>^{13}C-endosulfan I (1.03.19)</td><td>99.0</td></tr><tr><td>^{13}C-endosulfan II (1.03.19)</td><td>99.4</td></tr><tr><td>^{13}C-Endosulfan Sulfate (1.03.19)</td><td>74.3</td></tr><tr><td>d^{14}-Trifluralin (di-n-propyl) (1.03.19)</td><td>74.7</td></tr><tr><td>^{13}C-endrin (50.16)</td><td>981</td></tr><tr><td>^{13}C-aldrin (06.19)</td><td>1009</td></tr><tr><td>^{13}C-isodrin (06.19)</td><td>1973</td></tr></table></td></tr>
<tr><td>540</td><td>DDT I (17.20)

<table><tr><th>Compound</th><th>Concentration (pg µL^{-1})</i></th></tr><tr><td>^{13}C-<i>alpha</i>-HCH (16.20)</td><td>1000</td></tr><tr><td>^{13}C-<i>beta</i>-HCH (16.20)</td><td>200</td></tr><tr><td>^{13}C-<i>gamma</i>-HCH (16.20)</td><td>1005</td></tr><tr><td>^{13}C-<i>delta</i>-HCH (38.18)</td><td>1000</td></tr><tr><td>^{13}C-<i>p,p'.</i>-DDE</td><td>324</td></tr><tr><td>^{13}C-<i>o,p.</i>'-DDD (38.18)</td><td>318</td></tr><tr><td>^{13}C p,p.'-DDT (38.18)</td><td>318</td></tr></table></td></tr>
<tr><td>540</td><td>PCB I

<table><tr><th></th><th>Compound</th><th>Concentration (pg µL^{-1})</i></th></tr><tr><td>13C components</td><td>^{13}C-PePCB (1.15.18)</td><td>92.1</td></tr><tr><td></td><td>^{13}C-HCB (1.15.18)</td><td>92.8</td></tr><tr><td>13C PCB-mix (15.18)</td><td>^{13}C PCB 28</td><td>237</td></tr><tr><td></td><td>^{13}C PCB 52</td><td>239</td></tr><tr><td></td><td>^{13}C PCB 101</td><td>236</td></tr><tr><td></td><td>^{13}C PCB 105</td><td>239</td></tr><tr><td></td><td>^{13}C PCB 114</td><td>237</td></tr><tr><td></td><td>^{13}C PCB 118</td><td>236</td></tr><tr><td></td><td>^{13}C PCB 123</td><td>242</td></tr><tr><td></td><td>^{13}C PCB 138</td><td>237</td></tr><tr><td></td><td>^{13}C PCB 153</td><td>238</td></tr></table></td></tr>
</table>

	^{13}C PCB 156	236
	^{13}C PCB 157	236
	^{13}C PCB 167	237
	^{13}C PCB 180	238
	^{13}C PCB 189	237
	^{13}C PCB 209	237
3780	*iso*-octane	

Table Appendix.5. Recovery standard composition.

Amount *(µL)*	Compound
70	13C PCB -159
9930	*iso*-octane

Table Appendix.6. Details of chemicals used in contaminant analysis.

Chemical	Purity (quality)	Supplier	CAS-number
Acetone	SupraSolv®	Merck	67-64-1
Ammonium Sulfate $(NH_4)_2SO_4$	Emsure ®	Merck	7783-20-2
Cyclohexane	Suprasolv ®	Merck	110-82-7
Dichlormethane	Suprasolv ®	Merck	75-09-2
Ethanol absolutt alkohol prima ren	99,9 %	Antibac	
Florisil		Merck	1343-88-0
Isooctane	Supra Solv ®	Merck	540-84-1
Metanol	Suprasolv ®	Merck	67-56-1
n-hexane	≥99.0% PESTINORM		
Helium	6.0 quality	Yara Praxair AS, Norway	

Table Appendix.7. Details on equipment used in contaminant analysis.

Usage/description	Equipment	Supplier
Scale for weighing	PG802	Mettler Toldeo, Colombus, Ohio, USA
Analytical scale for weighing	Sartorius BP211D Dual Range analytical Balance	Mettler Toldeo, Colombus, Ohio, USA
Centrifuge	Eppendrof ® Centrifuge 5702	Merck
Vortexer	WIZARD IR Infrared Vortex Mixer	VWR international, Radnor, Pennsylvania, USA
Horizontal shaker	IKA ® HS 501 Horizontal shaker	
To dissolve ammonium sulfate in water	Ultrasonic Cleaner USC – THD	VWR international, Radnor, Pennsylvania, USA
Volume reduction 12 ml → 0,2 ml	RapidVap RapidVap ® Vacuum Dry Evaporation System (Labconco, Kansas City, USA).	Labconco, Kansas City, USA
Volume reduction 250 µL → 30 µL	miVac Quattro Centrifugal Concentrators	Genevac ™, Ipswich, UK
Glass pipettes	Disposable glass Pasteur pipettes (230 mm)	VWR, international, Radnor, Pennsylvania, USA
Multipipette	Multipipette ® E3	
Finnpipette	Finnpipette ™ F2 Variable Volume Pipettes	Thermo Fisher Scientific
Burning of florisil	Carbolite ™ CWF Chamber Furnace	Carbolite, Parsons Lane, Hope Hope Valley, England
Cleaning routine of glass equiptment	Laboratory High Temperature Oven – LHT5/120	Carbolite, Parsons Lane, Hope Hope Valley, England
Solid phase extraction clean up	Zymark RapidTrace SPE Workstation	Caliper Life Sciences, Mountain in View, USA

Gas chromatograph	Thermo scientific trace 1310 gas chromatograf	Thermo Fisher Scientific Inc., Waltham, MA, USA
Mass spectrometer	Thermo scientific TSQ9000 triple quadrupole mass spectrometer, USA	Thermo Fisher Scientific Inc., Waltham, MA, USA
Frits	Frits: Glass fiber	Affinisep, Petit Couronne, France
Columns for solid phase extraction clean up	AttractSPE^{TM}HLB	Affinisep, Petit Couronne, France
VirKon tablets for cleaning glasswear in contact with biological samples	Rely+On TM Virkon TM Tablets	Lanxess

Table Appendix.8. LOQs (*pg g^{-1}, ww*) for all targeted compounds. -: the LOQ value was not reported; n/a: the compound was not targeted in this analytical batch.

	2007	2008	2009	2010	2011	2012	2013	2014	2015	2017	2018	2019	2020
Batch nr.	1	2	3	4	5	6	7	8	9	9	9	9	9
HCB	1.2	-	216	423	1534.5	-	1536	9	357	357	357	357	357
***α*-HCH**	52.8	33.3	36.3	147.9	14.7	108	65.7	1236	24.6	24.6	24.6	24.6	24.6
***β*-HCH**	82.5	57	33.3	628.5	13.8	255	132.9	2175	4.5	4.5	4.5	4.5	4.5
***γ*-HCH**	28.2	23.4	29.1	110.7	27.6	57	33.6	354	470.4	470.4	470.4	470.4	470.4
Hept	118.5	104.4	191.7	221.4	n/a	n/a	n/a	888	n/a	n/a	n/a	n/a	n/a
HeptEpox	15	36	2490	737.7	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
OxC	43.5	735	735	253.5	591.6	-	217.2	408	27	27	27	27	27
TC	5.7	n/a	142.8	48.3	28.2	21	11.46	165	4.8	4.8	4.8	4.8	4.8
CC	11.7	10.8	182.1	94.2	68.7	17.85	13.35	255.9	4.8	4.8	4.8	4.8	4.8
TN	7.2	6.9	333.3	46.2	5.4	18	8.13	221.1	42	42	42	42	42
CN	5.1	3.6	181.5	32.4	27.6	-	20.67	133.2	15	15	15	15	15
Mirex	65.1	57	n/a	2400	n/a	-	28.68	1779	40.2	40.2	40.2	40.2	40.2
***o,p'*-DDT**	285	n/a	n/a	104.1	n/a	58.8	0	219.6	30	30	30	30	30
***p,p'*-DDT**	424.2	1227	n/a	354	n/a	57.6	0	174.6	-	-	-	-	-
***o,p'*-DDD**	243.3	n/a	n/a	n/a	n/a	29.79	92.4	88.5	15	15	15	15	15
***p,p'*-DDD**	n/a	n/a	n/a	n/a	n/a	28.05	99.9	83.1	-	-	-	-	-
***o,p'*-DDE**	274.2	n/a	n/a	6.6	n/a	115.2	58.5	283.2	15	15	15	15	15
***p,p'*-DDE**	366.9	807	204	-	63	-	82.2	201.6	15	15	15	15	15
PCB 28	38.1	624	n/a	78.3	282.9	-	45.3	51.3	180	180	180	180	180
PCB 31	223.5	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 33	60	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 37	197.4	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 47/49	173.4	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 52	52.2	284.7	n/a	19.5	63.3	222.6	93.6	255.9	390	390	390	390	390
PCB 99	140.4	945	160.5	257.1	82.2	-	150	330	309	309	309	309	309
PCB 101	161.7	1266	91.2	47.4	177.6	309	174.9	405	366	366	366	366	366
PCB 105	145.2	n/a	15	1122	195	414	224.1	546	309	309	309	309	309
PCB 118	152.4	1329	-	1401	328.8	-	177	426	318	318	318	318	318
PCB 123	139.2	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 128	239.1	n/a	n/a	50.4	83.7	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 138/163	204.9	1299	-	-	322.8	-	109.5	747	318	318	318	318	318
PCB 141	145.8	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 149	200.1	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 153	181.2	-	-	-	-	-	91.5	663	339	339	339	339	339
PCB 156	174.9	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 157	261.9	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 167	244.5	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 170	276.6	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a

PCB 180	259.5	738	-	56.7	276.3	-	105	849	312	312	312	312	312
PCB 183	212.7	627	45	55.2	34.8	-	83.1	720	240	240	240	240	240
PCB 187	241.8	780	53.1	50.4	34.8	-	95.4	873	273	273	273	273	273
PCB 189	199.8	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
PCB 194	245.1	n/a	6	49.5	114	498	633	819	279	279	279	279	279

Table Appendix.9. Detection frequency (*%*) for each year for each contaminant.

	2007	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**	**2015**	**2017**	**2018**	**2019**	**2020**
Total number of samples	53.0	46.0	51.0	57.0	25.0	44.0	23.0	27.0	20.0	31.0	27.0	20.0	25.0
Compounds													
PCB 28	84.9	0.0	0.0	66.7	24.0		100.0	74.1	5.0	6.5	51.9	10.0	20.0
PCB 31	1.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 33	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 37	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 47/49	41.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 52	0.0	0.0	0.0	0.0	4.0	0.0	0.0	0.0	0.0	0.0	3.7	0.0	0.0
PCB 99	**96.2**	**67.4**	**80.4**	**89.5**	**92.0**	**100.0**	**100.0**	**100.0**	**90.0**	**100.0**	**100.0**	**100.0**	**100.0**
PCB 101	0.0	0.0	33.3	0.0	4.0	0.0	0.0	0.0	0.0	0.0	3.7	0.0	0.0
PCB 105	90.6	0.0	96.1	0.0	92.0	97.7	91.3	25.9	0.0	3.2	29.6	0.0	12.0
PCB 118	96.2	78.3	100.0	15.8	92.0	100.0	100.0	96.3	100.0	100.0	100.0	100.0	100.0
PCB 123	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 128	50.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 138	96.2	93.5	100.0	100.0	92.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
PCB 141	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 149	1.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 153	**96.2**	**100.0**	**100.0**	**100.0**	**92.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**
PCB 157	15.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 167	43.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 170	86.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 180	**96.2**	**95.7**	**98.0**	**98.2**	**92.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**
PCB 183	90.6	73.9	96.1	96.5	92.0	100.0	100.0	44.4	55.0	80.6	100.0	95.0	96.0
PCB 187	92.5	78.3	96.1	100.0	92.0	100.0	100.0	63.0	90.0	61.3	100.0	100.0	96.0
PCB 189	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PCB 194	41.5	0.0	90.2	57.9	88.0	97.7	8.7	3.7	0.0	3.2	7.4	0.0	4.0
HCB	**96.2**	**100.0**	**98.0**	**96.5**	**88.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**
α-HCH	0.0	0.0	43.1	0.0	0.0	0.0	0.0	74.1	0.0	0.0	0.0	0.0	0.0
β-HCH	**0.0**	**0.0**	**94.1**	**5.3**	**0.0**	**56.8**	**60.7**	**0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**
γ-HCH	0.0	0.0	96.1	8.8	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Heptachlor	0.0	0.0	0.0	3.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Heptachlor epoxide	86.8	93.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Oxy-chlordane	**88.7**	**69.6**	**52.9**	**68.4**	**92.0**	**100.0**	**100.0**	**96.3**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**
Trans-chlordane	54.7	0.0	17.6	42.1	92.0	0.0	0.0	18.5	100.0	100.0	100.0	100.0	100.0
Cis-chlordane	7.5	8.7	0.0	0.0	0.0	9.1	4.3	0.0	20.0	54.8	74.1	30.0	48.0
Trans-nonachlor	30.2	50.0	0.0	14.0	92.0	95.5	95.7	11.1	90.0	71.0	96.3	100.0	100.0
Cis-nonachlor	83.0	67.4	0.0	14.0	68.0	100.0	95.7	22.2	95.0	93.5	100.0	100.0	100.0

Mirex	64.2	69.6	0.0	0.0	0.0	0.0	100.0	7.4	100.0	100.0	100.0	100.0	100.0
o,p'-DDT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	5.0	4.0
p,p'-DDT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
o,p'-DDD	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
p,p'-DDD	0.0	0.0	0.0	0.0	0.0	2.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0
o,p'-DDE	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
***p,p'*-DDE**	**94.3**	**82.6**	**78.4**	**100.0**	**96.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**	**100.0**

Table Appendix.10. Compound specific conversion factors for contaminants measured in plasma, obtained from paired samples to convert to blood concentrations.

	Intercept	Slope	Error
PCB99	-54.25	0.45	0.020
PCB153	520.20	0.33	0.010
PCB180	194.00	0.31	0.006
p,p'-DDE	-68.48	0.54	0.042
β-HCH	10.39	0.59	0.019
OxC	269.05	0.38	0.120
HCB	144.84	0.63	0.032

Table Appendix.11. Value for significant outliers removed from data for making conversion factors for contaminant concentrations based on paired samples of whole blood and plasma.

Contaminant	Value in blood samples (*pg g^{-1}, ww*)	*P*-value
PCB99	3202.06	0.001
PCB153	10502.65	0.001
PCB153	16370.24	0.001
PCB153	42372.09	0.001
PCB180	14942.14	0.001
PCB180	6712.42	0.001
PCB180	4619.11	0.001
p,p'-DDE	-	
β-HCH	880.18	0.001
β-HCH	642.30	0.001
β-HCH	457.87	0.001
β-HCH	928.17	0.001
β-HCH	254.16	0.001
β-HCH	360.05	0.001
OxC	4674.45	0.001
OxC	2512.92	0.001
OxC	4205.89	0.001
HCB	6685.24	0.001

Table Appendix.12. Results of model selection for candidate models explaining **PCB 99** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	***w***	**Marginal R^2**	**Conditional R^2**
Model 1.2	0.00	0.18	0.03	0.65
Model 0	0.35	0.15	0.00	0.68
Model 2.3	0.39	0.14	0.10	0.68
Model 1.1	1.77	0.07	0.04	0.64
Model 3.3	1.82	0.07	0.12	0.64
Model 3.5	1.91	0.07	0.03	0.70
Model 2.2	2.03	0.06	0.02	0.68
Model 2.1	2.33	0.05	0.02	0.68
Model 1.3	2.40	0.05	0.00	0.68
Model 1.4	2.41	0.05	0.00	0.68
Model 3.2	2.84	0.03	0.07	0.63
Model 3.1	3.71	0.03	0.04	0.64
Model 3.6	4.41	0.02	0.01	0.68
Model 3.4	4.42	0.02	0.01	0.68

Table Appendix.13. Results of model selection for candidate models explaining **PCB 153** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 2.3	0.00	0.21	0.11	0.66
Model 0	0.38	0.17	0.00	0.66
Model 1.3	1.06	0.12	0.01	0.64
Model 2.2	1.90	0.08	0.03	0.66
Model 1.4	1.92	0.08	0.01	0.66
Model 2.1	2.33	0.06	0.01	0.66
Model 1.2	2.44	0.06	0.00	0.66
Model 3.3	2.90	0.05	0.12	0.63
Model 3.6	3.07	0.04	0.02	0.64
Model 1.1	3.10	0.04	0.01	0.63
Model 3.4	3.99	0.03	0.02	0.66
Model 3.5	4.41	0.02	0.00	0.66
Model 3.2	4.76	0.02	0.04	0.63
Model 3.1	5.12	0.02	0.02	0.63

Table Appendix.14. Results of model selection for candidate models explaining **PCB 180** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc. The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 2.3	0.00	0.18	0.12	0.67
Model 1.3	0.26	0.16	0.02	0.65
Model 0	0.49	0.14	0.00	0.67
Model 2.2	1.87	0.07	0.04	0.67
Model 1.4	1.91	0.06	0.02	0.67
Model 3.3	2.10	0.06	0.04	0.67
Model 1.2	2.14	0.06	0.00	0.70
Model 3.6	2.20	0.05	0.03	0.65
Model 1.1	2.30	0.06	0.02	0.66
Model 2.1	2.34	0.06	0.01	0.67
Model 3.2	3.91	0.03	0.06	0.67
Model 3.4	3.94	0.02	0.02	0.67
Model 3.5	4.06	0.02	0.02	0.70
Model 3.1	4.27	0.02	0.03	0.66

Table Appendix.15. Results of model selection for candidate models explaining ***p,p'*-DDE** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 0	0.00	0.17	0.00	0.57
Model 2.3	0.79	0.11	0.05	0.57
Model 1.3	0.90	0.11	0.01	0.54
Model 1.1	0.96	0.10	0.06	0.49
Model 1.2	1.02	0.10	0.02	0.53
Model 2.2	1.96	0.06	0.01	0.57
Model 3.3	1.96	0.06	0.01	0.57
Model 1.4	2.01	0.06	0.00	0.57
Model 2.1	2.04	0.06	0.00	0.57
Model 3.6	2.91	0.04	0.01	0.54
Model 3.2	2.99	0.04	0.07	0.49
Model 3.1	3.02	0.04	0.06	0.49
Model 3.5	3.10	0.04	0.02	0.53
Model 3.4	4.04	0.02	0.00	0.57

Table Appendix.16. Results of model selection for candidate models explaining ***β*-HCH** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 2.1	0.00	0.20	0.19	0.76
Model 0	0.75	0.14	0.00	0.77
Model 3.4	0.89	0.13	0.24	0.76
Model 2.2	0.92	0.13	0.13	0.77
Model 3.6	1.97	0.07	0.19	0.75
Model 3.5	2.07	0.07	0.18	0.77
Model 1.4	2.71	0.05	0.01	0.77
Model 1.2	2.80	0.05	0.00	0.78
Model 2.3	2.84	0.05	0.00	0.77
Model 1.3	2.85	0.05	0.00	0.77
Model 3.1	4.11	0.03	0.18	0.76
Model 1.1	4.93	0.02	0.00	0.78
Model 3.2	5.09	0.02	0.13	0.76
Model 3.3	7.07	0.01	0.00	0.78

Table Appendix.17. Results of model selection for candidate models explaining **HCB** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 0	0.00	0.15	0.00	0.48
Model 1.3	0.15	0.14	0.02	0.45
Model 2.2	1.02	0.09	0.04	0.48
Model 2.3	1.02	0.09	0.04	0.48
Model 1.4	1.22	0.08	0.02	0.47
Model 1.1	1.33	0.08	0.05	0.39
Model 2.1	1.67	0.07	0.02	0.48
Model 1.2	1.91	0.06	0.00	0.45
Model 3.6	1.95	0.06	0.04	0.45
Model 3.2	2.27	0.05	0.11	0.39
Model 3.3	2.67	0.04	0.08	0.39
Model 3.1	2.91	0.04	0.08	0.38
Model 3.4	3.16	0.03	0.02	0.47
Model 3.5	3.48	0.03	0.02	0.44

Table Appendix.18. Results of model selection for candidate models explaining **OxC** concentrations from kittiwakes from Kongsfjorden in 2007-2020. Year is included as a random effect for all models. The models are sorted by AICc, the most fitting models having the lowest value, and the difference is indicated by ΔAICc The Akaike's weight is shown by *w*. Models are categorized in three groups, where nr. 1 includes only stable isotopes as explanatory factors, model nr. 2 includes both stable isotope information and FO diet data and nr. 3 includes only FO diet data. For more information on explanatory variables in the models, see Table 3 in materials and methods.

Model	**ΔAIC_C**	**w**	**Marginal R^2**	**Conditional R^2**
Model 0	0.00	0.16	0.00	0.50
Model 2.3	0.18	0.15	0.07	0.50
Model 1.4	0.59	0.12	0.03	0.48
Model 2.2	0.98	0.10	0.05	0.50
Model 2.1	1.04	0.10	0.04	0.50
Model 1.3	1.50	0.08	0.01	0.48
Model 1.2	1.98	0.06	0.00	0.49
Model 3.4	2.17	0.05	0.06	0.49
Model 3.6	2.62	0.04	0.05	0.48
Model 3.5	2.87	0.04	0.05	0.47
Model 1.1	3.30	0.03	0.01	0.45
Model 3.3	3.65	0.03	0.08	0.46
Model 3.1	4.19	0.02	0.07	0.45
Model 3.2	4.30	0.02	0.06	0.45

Table Appendix.19. *Mean* ± standard deviation (SD), median and range of $\delta^{13}C$, $\delta^{15}N$ and lipid levels (%) from kittiwakes in Kongsfjorden during chick rearing period.

	Year	n	Mean	± SD	Median	Range
$\delta^{13}C$	2007	15	- 19.96	0.16	-19.99	-20.21, -19.73
	2008	15	-20.81	0.15	- 20.3125	-20.58, -19.97
	2009	7	-21.012	0.57	- 21.16	- 21.42, -19.77
	2011	21	- 20.62	0.17	- 20.54	-21.08, -20.38
	2012	43	- 20.01	0.16	- 19.99	- 20.56, -19.70
	2013	9	- 19.83	0.96	- 19.83	- 19.96, -19.66
	2014	15	- 19.99	0.11	- 19.99	- 20.22, -19.81
	2015	20	- 21.26	0.18	- 21.22	- 21.72, -21.02
	2017	19	-21.04	0.15	- 21.03	- 21.32, - 20.79
	2018	20	- 20.54	0.10	- 20.55	- 20.73, -20.35
	2019	20	- 21.03	0.14	- 21.00	- 21.32, - 20.85
	2020	20	- 20.79	0.18	- 20.76	- 21.14, -20.46
$\delta^{15}N$	2007	15	12.71	0.32	12.84	11.91, 13.00
	2008	15	12.07	0.46	12.2	11.16, 12.67
	2009	7	12.54	0.70	12.61	11.43, 13.44
	2011	21	13.01	0.58	12.92	12.11, 14.13
	2012	43	13.42	0.54	13.41	12.43, 14.44
	2013	9	11.17	0.41	11.15	10.28, 11.72
	2014	15	11.87	0.35	11.91	11.12, 12.49
	2015	20	11.63	0.36	11.6	11.05, 12.41
	2017	19	12.75	0.61	12.75	11.42, 13.60
	2018	20	12.97	0.15	12.96	12.82, 13.13
	2019	20	13.25	0.50	13.23	12.29, 14.27
	2020	20	14.95	0.32	14.92	14.54, 15.58
% lipid	2007	15	0.29	0.11	0.3	0.12, 0.48
	2008	15	0.26	0.26	0.17	0.03, 1.10
	2009	7	0.3	0	0.3	0.30, 0.30
	2011	21	0.18	0.09	0.17	0.08, 0.49
	2012	43	1.00	0.18	0.97	0.55, 1.56
	2013	9	2.63	0.36	2.57	2.13, 3.25
	2014	15	2.34	1.41	2.11	0.95, 5.35
	2015	20	0.29	0.08	0.28	0.28, 0.62
	2017	19	0.17	0.03	0.16	0.14, 0.27
	2018	20	0.28	0	0.28	0.28, 0.28
	2019	20	0.36	0.18	0.38	0.03, 0.63
	2020	20	0.26	0.06	0.28	0.01, 0.28

Table Appendix.20. *Mean* ± standard deviation (SD), median and range of PCB99, PCB153, PCB180, *p,p*'-DDE, bHCH, HCB and OxC (pg g^{-1}, lw) measured in blood from adult kittiwakes from Krykkjefjellet, Svalbard, during chick rearing period.

	year	n	mean	+-SD	median	Range
PCB 99	2007	15	512520.5	327338.8	410581.2	14518.78, 1302950.28
	2008	15	1170942	669062.2	1089600	175455.9, 226847.4
	2009	7	488185.7	361061.9	622066.7	26750.0,900366.7
	2011	21	1681741	1010816	1649394	19129.59, 4381010.75
	2012	43	473848.5	205104.5	441102.6	170377.4, 1111477.8
	2013	9	62875.85	30814.66	55355.5	29603.42, 126600.89
	2014	15	63837.58	41259.92	67441.86	18098.28, 159827.37
	2015	20	304868.3	259066.6	217669.8	94705.78, 1225506.43
	2017	19	527417.3	364151.6	444317	67672.33, 1466798.94
	2018	20	731292.3	313600	654360.9	251351.6, 1278648.2
	2019	20	1217405	1981422	287509.8	115327, 6741333
	2020	20	1401531	4040285	349589.5	175047.7, 18475828.8
PCB 153	2007	15	3300905	2320195	2727298	18744.62, 9804536.72
	2008	15	5648512	3175378	4271800	1226744, 12550100
	2009	7	3338448	856647	3370500	2025833, 4584833
	2011	21	8699790	5917298	7264239	59715.67, 26956139.78
	2012	43	953295.9	462879.5	858608.5	344510.9, 2783717.8
	2013	9	138238	77846.63	130328.7	29175.34, 280815.76
	2014	15	156242.5	89709.41	149621.2	36890.81, 315457.92
	2015	20	1677456	1525234	1110618	421995.3, 6923445.6
	2017	19	3593081	4762096	2485361	892361.4, 22397969.2
	2018	20	3954838	1975063	3798894	1556187, 8655678
	2019	20	7030694	11518965	1479408	525354.1, 40260470.4
	2020	20	8318671	23974021	2052077	781919.1, 109332249.7
PCB 180	2007	15	3300905	2320195	2727298	26851.66, 4381610.17
	2008	15	5648512	3175478	4271800	1226744, 12550100
	2009	7	3338448	856647	3370500	2025833, 454833
	2011	21	8699790	5917298	7264239	59715.7, 26956139.7
	2012	43	953295	462879	858608	344510.9, 2783717.8
	2013	9	138238	77846	130328	29175, 280815
	2014	15	156243	89709	149621	36890, 315457
	2015	20	1677456	1525234	1110618	421995, 6923445
	2017	19	3593081	4762096	2485361	892361, 22397969
	2018	20	3954838	1975063	3798894	1556187, 8655678
	2019	20	7030694	11518965	1479408	525354, 40260470
	2020	20	8318671	23974021	2052077	781919, 109332249
p,p'-DDE	2007	15	936558.9	947396.1	623609.9	37964.3, 3448939.1
	2008	15	1681896	1325055	1631876	186489.7, 4782942.9
	2009	7	935723.8	586444.8	843700	164533.3, 1749066.7
	2011	21	2073635	1626993	1604124	14661.29, 6115625

	2012	43	399801.8	435540.8	317142.8	59728.97, 2686656.97
	2013	9	72355.85	49636.85	50856.97	26877.05, 169569.54
	2014	15	72459.48	57894.95	59778.9	9037.9, 221632.8
	2015	20	783772.1	537138.5	626341.8	192774.1, 2378743.2
	2017	19	1930047	1730543	1472941	418686, 8081903
	2018	20	2472930	1515784	2205805	810960.9, 5860392.9
	2019	20	2616680	4682207	588440.6	114501.5, 19226238.5
	2020	20	4354559	13704813	1181961	242532.1, 62430689.4
β-HCH	2007	0				
	2008	0				
	2009	7	71423.81	27845.11	72166.67	35100, 120033.3
	2011	0				
	2012	43	19528.76	12188.05	17442.05	7231.96, 80495.41
	2013	9	3809.48	1870.65	3375.29	1934.72, 7649.24
	2014	0				
	2015	20	45443.16	25219.08	37323.41	15403.7, 115309.7
	2017	19	56483.69	26631.6	52026.47	14134.34, 122700.16
	2018	20	129987.5	88123.76	103269.5	28993.56, 333771,75
	2019	20	202492.7	309735.5	54515.55	24308.63, 977778.71
	2020	20	243537.1	707458.8	77537.1	25137.31, 3244675.10
HCB	2007	15	630577.8	436047.6	492630.9	124.20, 1371978.53
	2008	15	1023481	619020.7	914363.1	121932.8, 2314674.8
	2009	7	1189952	297466	1166000	781300, 1643533
	2011	21	2485818	1136658	2423309	357107, 5308362
	2012	43	371862.9	110850	362923.1	202380.8, 729409
	2013	9	105895.6	29030.89	97548.09	78024, 169396
	2014	15	118184	68309.9	110778.9	29827.9, 251631.7
	2015	20	606519.7	328475.1	490827.7	237159.6, 1642670.2
	2017	19	1216058	555732.6	1111276	350667.9, 2446302.8
	2018	20	1493030	509369	1459513	706602.9, 2404020.7
	2019	20	1793212	2670539	536064	253351, 8116286
	2020	20	3071546	9370891	901784	411222.6, 42851694.6
OxC	2007	15	430599.6	403729	336448.9	4502.19, 1395072
	2008	15	626474.8	372097.4	476825.4	112626.4, 1273480
	2009	7	604757.1	254269.2	575866.7	320900, 1108733
	2011	21	1344686	705094	1378635	137676.5, 3204330.0
	2012	43	196955.6	82181.43	188921.9	81421.43, 449558.19
	2013	9	37108.11	13096.68	33525.34	23346.8, 63172.5
	2014	15	53306.65	32014.38	41304.26	15558.57, 117376.92
	2015	20	247389.1	227656.2	174052.1	58616.9, 1062417.5
	2017	19	238286.9	173329.1	188742.9	15798.3, 604037.6
	2018	20	444215.6	460044.2	289129	66490.54, 1680938.88
	2019	20	1402491	2447897	237657.5	136804.4, 8284487.2
	2020	20	1173488	3733112	342568.1	44816.52, 17003216.82

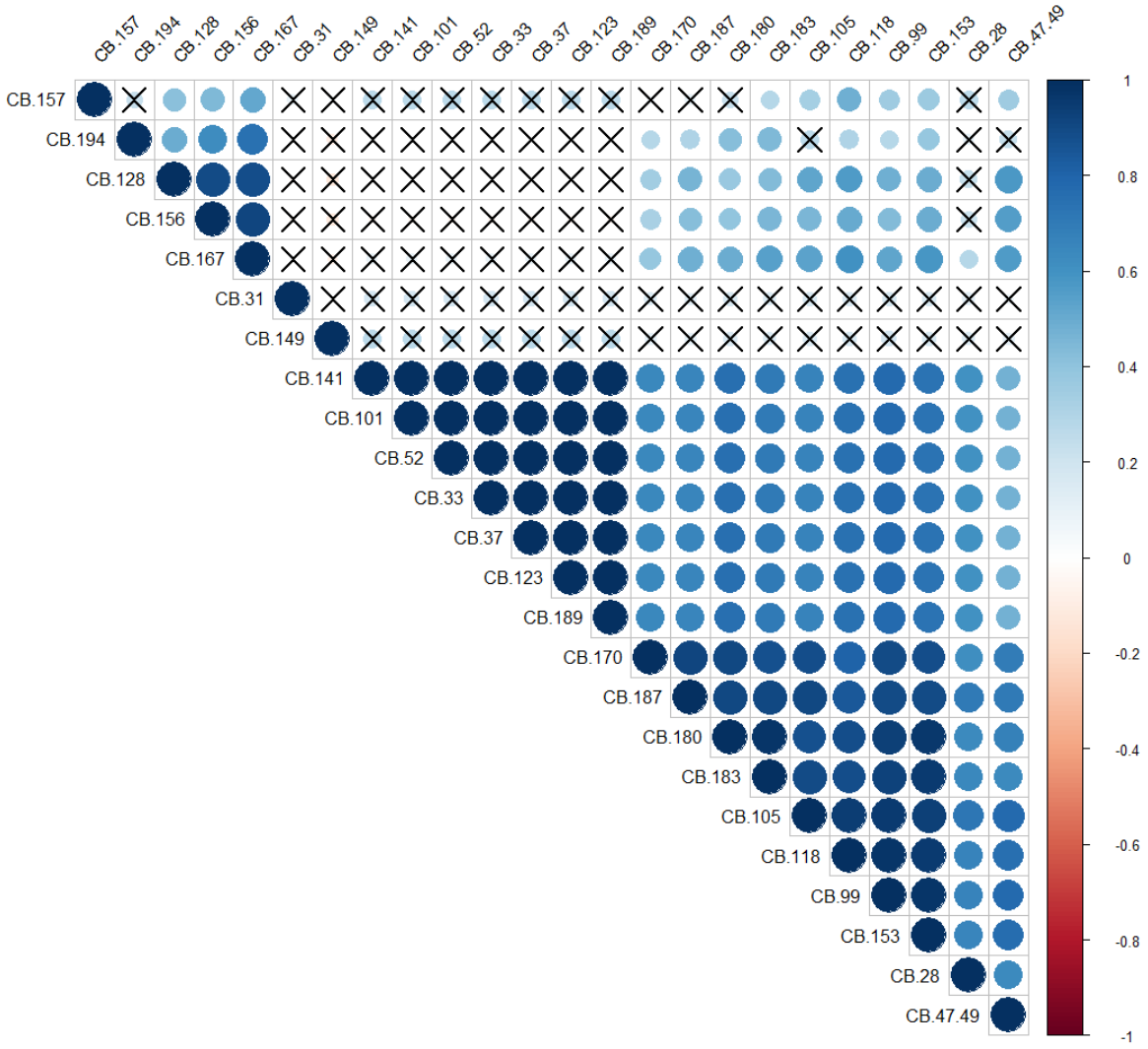

Figure Appendix.1. Correlation matrix for PCB congener concentrations (lipid weight) measured in blood from adult kittiwakes from 2007-2009, 2011-2015, and 2017-2020 during chick-rearing period in Krykkjefjellet, Kongsfjorden. Blue indicates a positive correlation according to the colour-scale. X indicates that the correlation is not significant. PCB congeners are ranked with hierarchal clustering.

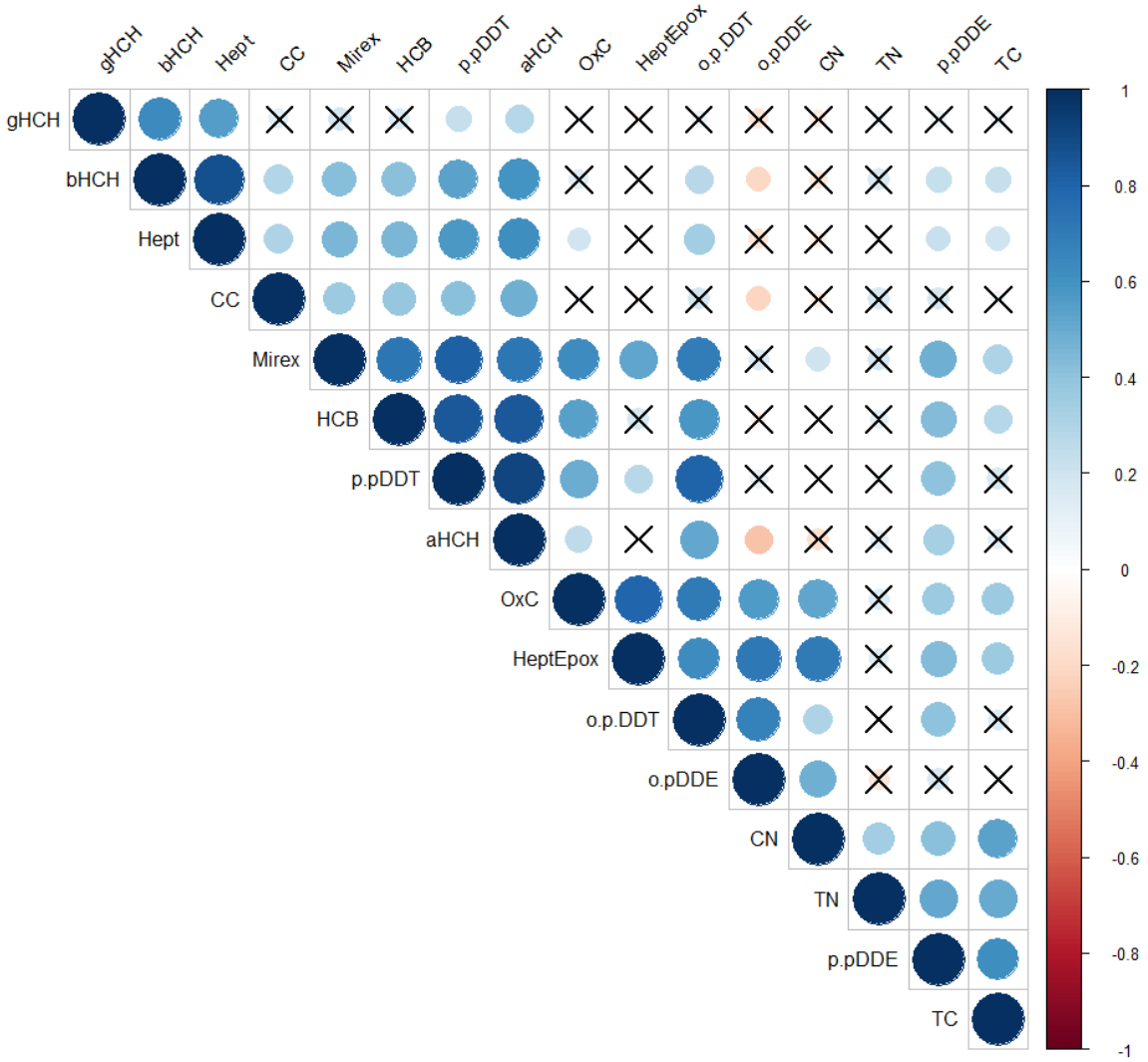

Figure Appendix.2. Correlation matrix for OCP compound concentrations (lipid weight) measured in blood from adult kittiwakes from 2007-2009, 2011-2015, and 2017-2020 during chick-rearing period in Krykkjefjellet, Kongsfjorden. *p,p'*-DDD and *o,p'*-DDD were removed not analysed due to very low detection for many years (Table Appendix.8). Blue indicates a positive correlation and red indicates a negative correlation according to the colour-scale. X indicates that the correlation is not significant. OCP compounds are ranked with hierarchal clustering.

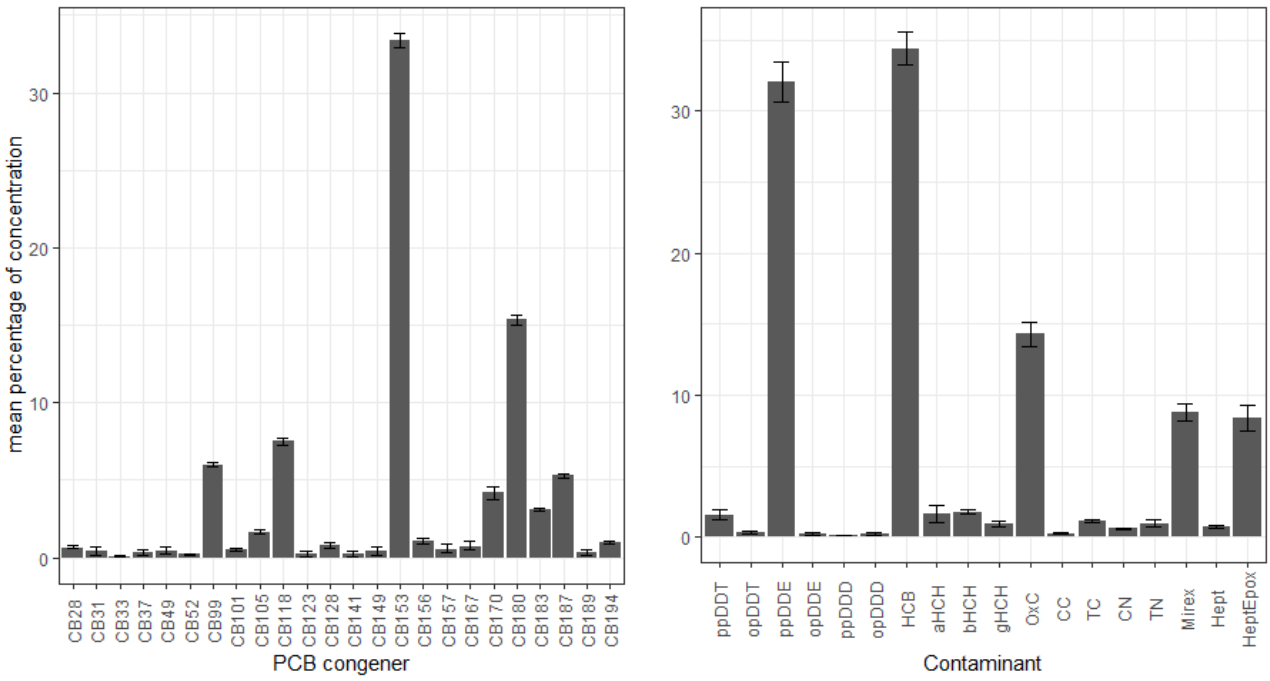

Figure Appendix.3. Compound profiles for PCB congeners (left) and OCP compounds (right) measured in blood from adult kittiwakes from 2007-2009, 2011-2015, and 2017-2020 during chick-rearing period in Krykkjefjellet, Kongsfjorden Each profile depicts the mean percentage for the contribution of the concentration of every PCB congener or OCP compound to the total PCB or OCP burden. Error bars indicate standard deviation for the contribution of the concentration for every compound.

Appendix B

Table Appendix.B.1: Content in «PEST KV. STD 5 (06.19) gl 1 av 1», diluted in isooctane.

	Component	Concentration
PEST I (06.19)	^{13}C-*trans*-nanochlor (1.03.19)	5.92
	^{13}C-*cis*-nonaChlor (1.04.17)	4.11
	^{13}C-*trans*-chlordane (1.04.17)	4.07
	^{13}C-*cis*-chlordane (1.03.19	5.98
	^{13}C-Oxychlordane (50.16)	41.3
	^{13}C-Heptachlor epoxid (06.19)	80.3
	^{13}C-HeptaChlor (06.19)	97.0
	^{13}C-Dieldrin (06.19)	80.2
	^{13}C-Mirex (06.19)	20.9
	^{13}C-Endosulfan I (1.03.19)	7.92
	^{13}C-Endosulfan II (1.03.19)	7.96
	^{13}C-Endosulfan Sulfate (1.03.19)	5.95
	D14-Trifluralin (di-n-propyl)	5.98
	^{13}C-Endrin (50.16)	78.6
	^{13}C-Aldrin (06.19)	80.8
	^{13}C-Isoddrin (06.19)	158
12C Pest Mix (1.39.17)	^{12}C-Dieldrin	39.9
	^{12}C-Aldrin	39.9
	^{12}C-Endrin	26.6
	^{12}C-Mirex	26.6
	^{12}C-Isodrin	15.9
	^{12}C-Trifluralin	15.9
	^{12}C-*trans*-chlordene	39.9
	^{12}C-*α*-Chlordane	10.6
	^{12}C-*γ*-Chlordane	10.6
	^{12}C-Oxychlordane	15.9
	^{12}C-*trans*-nonachlor	10.6
	^{12}C-*cis*-nonachlor	10.6
	^{12}C-heptachlor	15.9
	^{12}C-Heptachlor epoxide	26.6
	^{12}C-Heptachlorendoepoxide	26.6
	^{12}C-Endosulfan sulphate	10.6
12C Endosulfan MIX (44.17)	Endosulfan I (44.17)	12.3
	Endosulfan II (46.17)	11.6
PG (1.24.18)	1,2,3,4 TCN	10.7

Table Appendix.B2. Content in «DDT kv std 1 (39.18) gl 1 av 1». Diluted in isooctane.

	Component	Concentration
DDT i (17.20)	^{13}C-*α*-HCH (16.20)	20.0
	^{13}C-*β*-HCH (16.20)	3.99
	^{13}C-*γ*-HCH (16.20)	20.1
	^{13}C-*δ*-HCH (38.18)	20.0
	^{13}C-*p,p'*-DDE (38.18)	6.46
	^{13}C-*o,p'*-DDD (38.18)	6.34
	^{13}C-*p,p'*-DDT (38.18)	6.35
12C Pest Mix (1.19.19)	^{12}C-*α*-HCH	15.6
	^{12}C-*β*-HCH	4.17
	^{12}C-*γ*-HCH	10.4
	^{12}C-*o,p'*-DDE	5.21
	^{12}C-*p,p'*-DDE	5.21
	^{12}C-*o,p'*-DDD	5.21
	^{12}C-*p,p'*-DDD	5.21
	^{12}C-*o,p'*-DDT	5.21
	^{12}C-*p,p'*-DDT	5.21
12C d-HCH (2.39.18)	δ-HVH (1-16-13)	15.3
PG (1.24.18)	1,2,3,4 TCN	13.3

Table Appendix.B.3. Content in "PCB KV.STD 2 (15.18) gl 1 av 1". Diluted in isooctane.

	Component	**Concentrations** (*pg/µL*)
PCB I (15.18)	^{13}C PeCB (1.15.18)	7.71
	^{13}C HCB (1.15.18)	7.67
	^{13}C PCB 28	18.5
	^{13}C PCB 52	18.7
	^{13}C PCB 101	18.5
	^{13}C PCB 105	18.7
	^{13}C PCB 114	18.5
	^{13}C PCB 118	18.4
	^{13}C PCB 123	18.9
	^{13}C PCB 138	18.5
	^{13}C PCB 153	18.6
	^{13}C PCB 156	18.6
	^{13}C PCB 157	18.4
	^{13}C PCB 167	18.6
	^{13}C PCB 180	18.6
	^{13}C PCB 189	18.5
	^{13}C PCB 209	18.5
12C PCB mix (15.18)	^{12}C PCB 18	7.5
	^{12}C PCB 28	7.5
	^{12}C PCB 31	7.5
	^{12}C PCB 33	7.5
	^{12}C PCB 37	7.5
	^{12}C PCB 47	7.5
	^{12}C PCB 52	7.5
	^{12}C PCB 66	7.5
	^{12}C PCB 74	7.5
	^{12}C PCB 99	7.5
	^{12}C PCB 101	7.5
	^{12}C PCB 105	7.5
	^{12}C PCB 114	7.5
	^{12}C PCB 118	7.5
	^{12}C PCB 122	7.5
	^{12}C PCB 123	7.5
	^{12}C PCB 128	7.5
	^{12}C PCB 138	7.5
	^{12}C PCB 141	7.5
	^{12}C PCB 149	7.5
	^{12}C PCB 153	7.5
	^{12}C PCB 156	7.5
	^{12}C PCB 157	7.5
	^{12}C PCB167	7.5
	^{12}C PCB 170	7.5
	^{12}C PCB 180	7.5
	^{12}C PCB 183	7.5

	^{12}C PCB 187	7.5
	^{12}C PCB 189	7.5
	^{12}C PCB 194	7.5
	^{12}C PCB 206	7.5
	^{12}C PCB 209	7.5
Single components	^{12}C HCB (1.28.09)	7.8
	^{12}C PeCB (1.28.09)	7.8
	PG (1.36.13) 1,2,3,4 TCN	67.3