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BIO-3950 Master's Thesis in Biology, June 2021

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Preface

The study on glaucous gulls on Svalbard was approved by the Governor of Svalbard and the Norwegian Animal Research Authority (www.fdu.no). The RiS ID of the glaucous gull project is 5267. Handling and sampling of the birds were conducted in accordance with current regulations of the Norwegian Animal Welfare Act. This study includes data collected from glaucous gulls from year 2015 until 2019.

Fieldwork was funded by the Research Council of Norway and Svalbard Science Forum (SSF) through the Arctic field grant. Contaminant and stable isotope analysis were funded by the Norwegian Polar Institute.

Abstract

The glaucous gull *Larus hyperboreus* in avian predator and scavenger occupying a high position in the Arctic marine food web. The species is therefore exposed to high levels of biomagnifying contaminants thought to cause adverse health effects. Nevertheless, exposure to these anthropogenic persistent organic pollutants can vary both temporally and spatially. Namely, when comparing two glaucous gull populations in the Svalbard archipelago, organohalogenated contaminants (OHCs) have been detected in higher levels on Bjørnøya compared to Kongsfjorden. The greater OHC concentrations in Bjørnøya glaucous gulls were hypothesized to result from this population occupying a higher trophic level. The aim of this study was to compare body burdens of OHCs between both glaucous gull populations from Bjørnøya and Kongsfjorden and investigate the influence of their dietary ecology and biological variables (body condition index and sex) on OHC levels. To do so, blood from 112 adult glaucous gulls were sampled during the breeding seasons of 2015 until 2019 in Bjørnøya and Kongsfjorden. We measured plasma concentrations of OHCs including polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and perfluorinated alkylated substances (PFASs). Feather stable isotopes of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) were analyzed to indicate trophic level and feeding habitat, respectively. Body burdens of OHCs differed significantly between the two glaucous gull populations for all 5 sampling years. Overall, the population at Bjørnøya had plasma concentrations of OHCs 5.3 times higher than the Kongsfjorden population, with mean concentrations over all five years of 1217 ± 591 ng/g wet weight (*ww*) and 229 ± 176 ng/g *ww*, respectively. The most quantitatively abundant contaminants found in the glaucous gulls were ΣPCBs and ΣPFASs in Bjørnøya and ∑PCBs in Kongsfjorden. No differences in relative trophic position between both breeding populations were reported although Kongsfjorden individuals varied significantly in $\delta^{15}N$ across sampling years, while Bjørnøya individuals remained stable over time. In addition, we highlighted no relationship between plasma OHCs and feather $\delta^{15}N$ (trophic position) although the lack of association might be due to both tissues (i.e blood and feathers) reflecting different time scales. Only ΣPFSAs and ΣPFCAs showed a positive relationship with $\delta^{13}C$, suggesting that feeding habitat, more than trophic position, might explain variance in contaminant exposure.

Abbreviations

AIC	Akaike's Information Criterion
ANOVA	Analysis of variance
BCI	Body condition index
BFR	Brominated flame retardant
DDT	Dichlorodiphenyltrichloroethane
GC	Gas chromatograph
LMM	Linear mixed effects model
LOD	Limit of detection
LOQ	Limit of quantification
MS	Mass spectrometer
NCI	Negative chemical ionization
OC	Organochlorine
OCP	Organochlorine pesticide
OHC	Organohalogenated contaminant
PCA	Principal component analysis
PCB	Polychlorinated biphenyl
PFAS	Poly- and perfluoroalkyl substances
PFCA	Perfluoroalkyl carboxylate
PFSA	Perfluoroalkyl sulfonate
POP	Persistent organic pollutant
RBC	Red blood cells
RSTD	Recovery standard
SD	Standard deviation
SRM	Standard reference material
VIF	Variance inflation factor
ww	Wet weight
^{13}C	Stable isotope of carbon
^{15}N	Stable isotope of nitrogen
$\delta^{13}C$	Delta C-13 – ratio of stable carbon isotopes, ^{13}C: ^{12}C
$\delta^{15}N$	Delta N-15 – ratio of stable nitrogen isotopes, ^{15}N: ^{14}N

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1 Introduction

1.1 Organohalogenated contaminants (OHCs) in the Arctic

The Arctic is a pristine area characterized by low air and water temperatures, large seasonal fluctuations in solar radiation, sea ice cover and a lack of nutrients (AMAP, 2002). Despite minimal local emission and production of anthropogenic contaminants in the Arctic, high levels of organohalogenated contaminants (OHCs) have been found in Arctic biota for decades (Letcher et al., 2010; Dietz et al., 2019). OHCs produced in southerly latitudes can reach the remote Arctic due to their persistence and semi-volatility (Hao et al., 2020). Transport pathways include atmospheric long-range transport, ocean and river currents, transpolar ice packs and migrating biota (Barrie et al., 1992; AMAP, 2002).

OHCs consist of carbon-based substances that contain at least one halogen component, such as chlorine, bromine or fluorine (Harrad, 2010), and can be used/found in a range of industrial compounds and bi-products, pesticides and consumer products. Based on their chemical properties OHCs can be divided into (1) organochlorines (OCs) such as polychlorinated biphenyls (PCBs) and organochlorinated pesticides (OCPs), (2) organobrominated compounds such as brominated flame retardants (BFRs), and (3) per- and polyfluoroalkyl substances (PFASs) (Letcher et al., 2010). The initial 12 contaminants (referred to as legacy contaminants) banned or restricted under the Stockholm Convention in 2004 are organochlorinated compounds (Fiedler, 2008). This global treaty aims to protect human health and the environment from persistent organic pollutants (POPs) and are currently regulating 35 groups of chemicals (http://chm.pops.int). Environmental monitoring of the 12 legacy contaminants has shown a general decreasing trend in Arctic air and biota (Dietz et al., 2019). However, due to their persistent nature, the chemicals may retain in environmental compartments (Wang et al., 2020). PFASs have recently become a global environmental concern as they are persistent, have long been shown to bioaccumulate in wildlife and are widespread in the Arctic (Lucia et al., 2017).

Historically, OCs were widely used after World War II and during the industrial revolution that followed because of their benefits in industry and as pest control in agriculture (Ritter et al., 1995). The best-known pollutants are dichlorodiphenyltrichloroethane (DDT) and PCBs. DDT was initially used as a pesticide in agriculture and vector control with the intention to kill

mosquitoes bearing malaria and lice causing typhus. When environmental concerns were raised because of its harmful effects on wild birds, DDT was restricted and banned in many countries in the early 1970s (Ritter et al., 1995; Harrad, 2010). However, DDT is still being produced as a vector control in some parts of the world (Stockholm Convention, 2019). PCBs, which are a mixture of chlorinated hydrocarbons, were also banned in the late 1970s. They were used for industrial purposes such as dielectrics in transformers and capacitors, paint additives and as heat exchange fluids (Bartlett et al., 2019). Several compounds classified as PFASs have also been listed to be regulated by the Stockholm Convention. They have been widely used in industrial and commercial applications (e.g., cosmetics, fire-fighting foams, household products, medical services and textiles) because of their chemical and thermal stability (Wang et al., 2017). The broad application range of PFASs have resulted in their wide presence in environments such as the Arctic (Bartlett et al., 2019).

In addition to being persistent, many OHCs have lipophilic properties allowing them to accumulate within the lipids of an organism (Walker, 1990; Borgå et al., 2004; AMAP, 2016). Lipids are important in the Arctic for energy storage and insulation in a cold climate with annual fluctuations in food availability (Borgå et al., 2001). Arctic organisms therefore have a high capacity to bioaccumulate lipid associated contaminants. Because of efficient energy transfer in Arctic food webs, the concentration of contaminants tends to increase with each trophic level in a process called biomagnification (Borgå et al., 2004). Organisms exhibiting high trophic levels are especially at risk of elevated concentrations of contaminants. The highest concentrations of OHCs have been detected in Arctic top predators such as the glaucous gull *Larus hyperboreus* (Verreault et al., 2010), great skua *Stercorarius skua* (Leat et al., 2019), arctic fox *Alopex lagopus* (Bolton et al., 2017) and polar bears *Urus maritimus* (Dominique et al., 2020). OHCs have been linked to a wide variety of health problems in animals and humans, including behavior and development abnormalities, effects on reproduction and immune systems, disruption of endocrine systems and birth defects (Letcher et al., 2010; Dietz et al., 2019). The long-term effects of chronic exposure to OHCs may pose serious hazards to populations, especially in long-lived species (Erikstad and Strøm, 2012). Arctic wildlife is generally not exposed to a single compound but rather a complex mixture of OHCs which may pose additive, synergistic and/or antagonistic effect combinations (Sagerup et al., 2009; Letcher et al., 2010) and may complicate the evaluation of effects of contaminants in field studies.

1.2 The role of diet in OHC accumulation

The bioaccumulation and metabolism of contaminants are dependent on a wide range of biological factors such as physiology (biotransformation and elimination), foraging behavior, breeding behavior and migration routes (Borgå et al., 2004; Leat et al., 2013). Diet constitutes one of the most important exposure routes for contaminant accumulation in Arctic top predators (AMAP, 1998; Borgå et al., 2004). For instance, Bustnes et al. (2000) found that glaucous gulls feeding on other seabirds species had higher contaminant levels compared to those with a higher intake of fish. Similar relationships between diet and contaminant exposure have also been found in great skuas (Leat et al., 2011; Leat et al., 2019) and polar bears (Tartu et al., 2017; Lippold et al., 2019). Mallory et al. (2019) also found that male Arctic terns decrease in contaminant concentrations when returning to the Arctic from the Antarctic to breed, highlighting the importance of changing diet and migration strategies in contaminant exposure.

In addition to trophic level, it is also believed that the source of organic matter and feeding habitat are important factors influencing contaminant levels (Lavoie et al., 2010). Stable isotopes of nitrogen and carbon provide useful tools to respectively assess trophic relationships and the contribution of different carbon sources to the diet (Bolton et al., 2017). The stable isotope ratio of heavier to lighter nitrogen ($^{15}N/^{14}N$, $\delta^{15}N$) is enriched with each trophic level in the food web while the stable isotope ratio of carbon ($^{13}C/^{12}C$, $\delta^{13}C$), which vary little along the food chain, reflects the primary productivity found in an area (Hobson and Clark, 1992; Campbell et al., 2000). Usually, terrestrial organisms are depleted in carbon compared to marine organisms, and benthic organisms are enriched compared to pelagic organisms (Elliott et al., 2009; Lavoie et al., 2010). Hence, differences in $\delta^{13}C$ values can indicate different foraging habitats in the marine environment (Hovinen et al., 2019). Comparing stable isotope ratios of nitrogen and carbon across populations of the same species can enable to assess the role of diet and foraging on their contaminant load.

1.3 Avian species as sentinels in the Arctic

Seabirds are suitable as sentinel species for environmental monitoring of marine pollution as many species occupy high trophic levels and are therefore susceptible to bioaccumulation of biomagnifying contaminants (Furness and Camphuysen, 1997). To be useful, a sentinel species must be sensitive to contaminant exposure and the response must be predictable and measurable

(Burger and Gochfeld, 2001). The glaucous gull *Larus hyperboreus* has long been recognized as a sentinel of OHC exposure in the Arctic (Verreault et al., 2010). It is a generalist predator and opportunistic scavenger bird with a circumpolar high Arctic distribution (Anker-Nilssen et al., 2000). Exposure to OHCs through the diet of glaucous gulls in combination with their restricted capacity to biotransform/metabolize contaminants (Henriksen et al., 2000), make them susceptible to bioaccumulate high levels of these compounds (Bustnes et al., 2003b). Namely, OHC contaminants have been recorded in glaucous gulls since the early 1970s, and previous studies reported high concentrations of organochlorines such as PCBs and dichlorodiphenyldichloroethylene (DDE) (Bourne and Bogan, 1972). The increasing occurrence of dead and dying glaucous gulls on Bjørnøya in the late 1980s sparked the assumption that chlorinated contaminants contributed to the increased mortality (Gabrielsen et al., 1995). In 1980 the population of glaucous gulls on Bjørnøya was estimated to be about 2000 breeding pairs. A new total estimate of the breeding population was made in 2006 and reported approximately 650 breeding pairs, which represented a decline of about 60% (Erikstad and Strøm, 2012) and led the species to be categorized as nearly threatened on the Svalbard Red list (Henriksen and Hilmo, 2015). In 2019, the population monitoring on Bjørnøya estimated the number of breeding pairs to be 309, a further decline of 52% since 2006 (Norsk Polarinstitutt, 2021). Physiological stress caused by high contaminant levels has been suggested to be an important factor in the dramatic population decline on Bjørnøya in combination with other natural or anthropogenic stressors (Verreault et al., 2010). Contrasting population trends have been reported in a glaucous gull population breeding in Kongsfjorden, Svalbard. In 2012 the breeding population in Kongsfjorden was estimated to consist of 25 breeding pairs (Descamps et al., 2012) versus 37 in 2019 corresponding to a population increase of approximately 32 %. (unpublished data, S. Descamps).

Glaucous gulls can feed at several or specific trophic levels and intraspecific variations in OHC concentrations may be explained by their diet variability (Bustnes et al., 2000; Sagerup et al., 2002; Verreault et al., 2010). Accordingly, previous studies have shown 5 to 30 times higher concentration of OHCs in glaucous gulls breeding in Bjørnøya compared to Kongsfjorden (Løseth, 2014; Melnes et al., 2017). With contrasting OHC exposure and population trends, the latter populations of glaucous gulls are highly relevant to investigate the influence of diet specialization on contaminant burdens.

Aim of study

The purpose of this study was to estimate body burdens of OHCs in breeding populations of glaucous gull from Bjørnøya and Kongsfjorden. We predicted greater levels of persistent chlorinated and per- and polyfluoroalkyl substances in the Bjørnøya population compared to the population residing in Kongsfjorden based on the hypothesis that Bjørnøya birds occupy a higher trophic position. In order to test this hypothesis, our study investigated the influence of biological parameters (sex and BCI) and dietary ecology ($\delta^{15}N$ and $\delta^{13}C$) on plasma OHC levels across both colonies over a 5 year period.

2 Materials and Methods

2.1 Study species

Glaucous gulls are one of the largest gull species breeding in the Arctic and nests along the coasts and on the open tundra in colonies or dispersed, usually situated in close distance to other seabird colonies (Erikstad and Strøm, 2012; Petersen et al., 2015). The birds breeding close to other bird colonies are often specialized on preying upon chicks, eggs and other adult seabird species but they can also utilize other food sources such as fish, mollusks, echinoderms, crustaceans, insects, carcasses, refuse and offal (Bakken and Tertitski, 2000; Erikstad and Strøm, 2012). On Svalbard, most glaucous gulls leave the archipelago in September-October and migrate to wintering grounds dispersed through the North-East Atlantic and return to their breeding colonies in March and April (Strøm, 2006).

Their apex position in the food web, long-lived nature, large size and their residence in the Arctic are some of the characteristics that make glaucous gulls valuable sentinel species (Burger and Gochfeld, 2004).

2.2 Sampling locations

Field samples from glaucous gulls were collected at two Arctic locations, Bjørnøya (Bear Island; 74º21.038'N 19º05.840'E) and Kongsfjorden (78º55'N 11º55'E), Svalbard, during the incubation periods (June/July) from 2015 until 2019 (Figure 1). During the summer season, these two locations are characterized by continuous daylight, low ambient temperatures, and occasionally strong winds and precipitation.

Kongsfjorden is a fjord on the northwest coast of Spitsbergen, Svalbard, influenced by Atlantic and Arctic water masses and glacial inputs. It is characterized by the surrounding mountains and by its bird cliffs and small islets (Hop et al., 2002). The study area in Kongsfjorden consisted of the islets Lovéenøyane, Breøyane, Prinz Heinrichøya, Dietrichholmen and Miedteholmen (Figure 1b).

Bjørnøya is a small, isolated island situated in the southwestern part of the Barents Sea. The landscape of the island is divided by two distinct forms, in the north and west the landscape is flat plain covered with lakes, while the smaller south and eastern parts consist of mountains. The bird cliffs reaching 400 m a.s.l are located at the south end of the island. Bjørnøya is located

in the midst of the most productive part of the Barents Sea and attracts a large number of seabirds during the breeding season (Thuesen and Barr, 2021), approximately half a million pairs of seabird breed on the south part of the island (Strøm, 2006). The samples were collected from breeding glaucous gulls located on the south-east coast of Bjørnøya, between Kapp Kolthoff and Kvalrossbukta (Figure 1c).

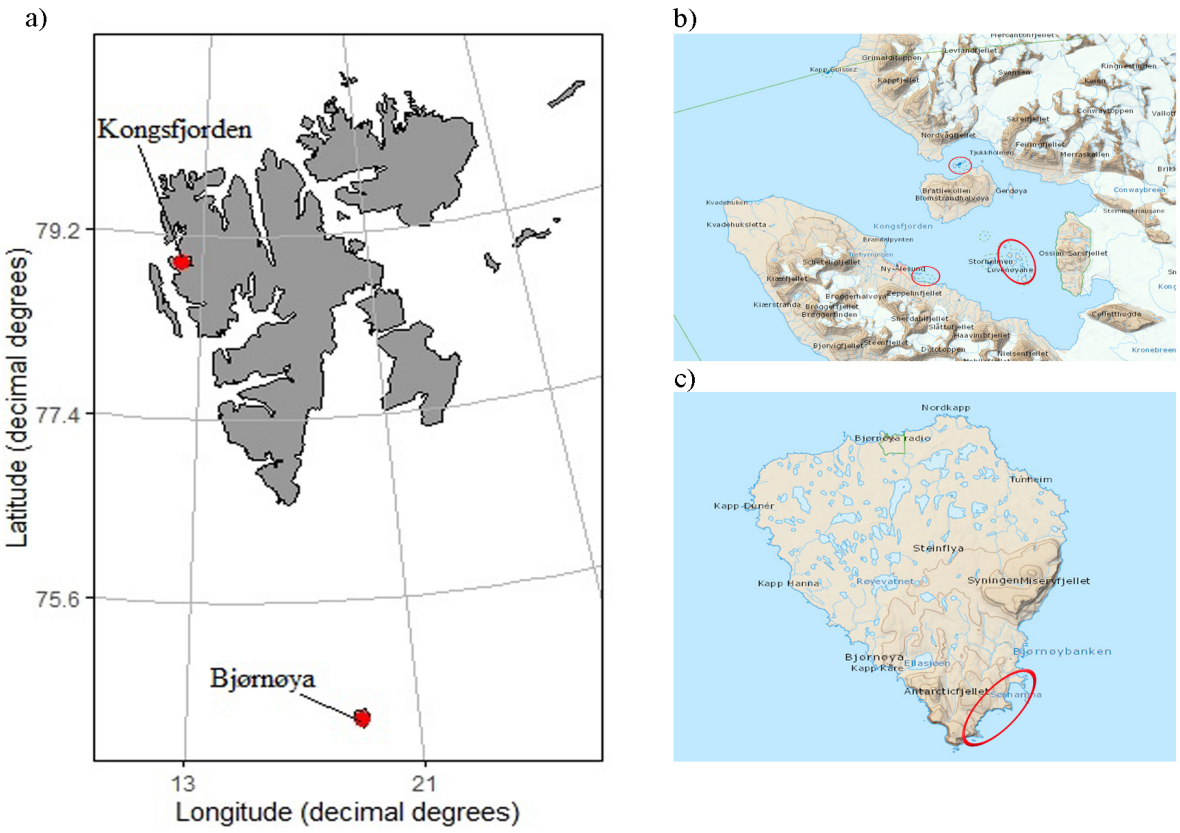

Figure 1: a) Map of the Svalbard archipelago with red dots representing the two sampling locations Bjørnøya and Kongsfjorden. b) Overview of Kongsfjorden with sampling areas highlighted with red circles (Lovenøyane, Breøyane, Prinz Heinrichøya, Dietrichholmen and Miedteholmen). c) Overview of Bjørnøya with sampling area highlighted with a red circle. Map: Norwegian Polar Institute

2.3 Field procedures

Breeding glaucous gulls were caught with automatic triggered nest traps (Bustnes et al., 2001a), a net canon (Super talon) or hand snares. The nest trap consists of a nylon thread that is set up along the edges of the nest, connected to a cord retracting mechanism. A radio transmitter is used to release the trap when the bird is laying on the nest, tightening the nylon thread around the nesting bird´s legs and trapping it. Eggs were collected from the nest and replaced with

dummy eggs during capture with the nest trap, while the eggs were kept warm in a heated bag for protection throughout the procedure. When caught, blood samples were extracted from the brachial wing vein using a heparinized syringe. Body feathers were collected from the back and chest area for analysis of stable isotopes. Morphometric data were recorded for each individual, including body mass, scull length, wing length, bill length, gonys height and tarsus length. Sex was determined from morphological measures. Captured birds were equipped with numbered steel rings from the Norwegian Ringing Centre, Stavanger Museum, as well as a three-letter coded plastic ring for simple identification as part of the annual seabird monitoring program SEAPOP (www.seapop.no). After measurements, the birds were released, and their eggs returned to the nest. The blood samples were kept cool and dark in the field until returned to the lab facilities in Ny-Ålesund or the field camp in Bjørnøya, where they were centrifuged and separated into plasma and red blood cells (RBC), before frozen at -20°C awaiting contaminant analysis.

Over the 5 sampling years, a total of 96 blood samples from 49 individuals were collected from Bjørnøya, of which 33 were recaptured 2 to 4 times. In Kongsfjorden, a total of 84 blood samples were collected from 63 individuals, of which 18 were recaptured 2 to 3 times.

2.4 Contaminant analysis of organochlorines (OCs) and per- and polyfluoroalkyl substances (PFAS)

Analysis of organochlorines (OCs) and poly- and perfluoroalkyl substances (PFAS) in blood plasma (n=180) of the glaucous gull were performed at the laboratory of The Norwegian Institute for Air Research (NILU) in Tromsø, Norway. For all compounds, the internal standard method was applied. Plasma samples were analyzed for the PCB congeners, chlorinated pesticides and PFASs listed in Table 1.

Table 1: List of the 52 chlorinated and fluorinated contaminants analyzed in blood plasma from glaucous gulls breeding in Bjørnøya and Kongsfjorden sampled during the breeding seasons of 2015 until 2019. Contaminants excluded from statistical analysis are marked with X.

			LOD or <45%	
Group	**Acronym**	**Analyte**	**Bjørnøya**	**Kongsfjorden**
HCB	HCB	**Hexachlorobenzene**		
HCH		**Hexachlorocyclohexane**		
	a-HCH	1α,2α,3β,4α,5β,6β-hexachlorocyclohexane	X	X
	b-HCH	1α,2β,3α,4β,5α,6β-hexachlorocyclohexane		
	g-HCH	1α,2α,3β,4α,5α,6β-hexachlorocyclohexane	X	X
CHLs		**Chlordane**		
	trans-chlordane	*trans*-chlordane	X	
	cis-chlrodane	*cis*-chlrodane	X	X
	oxy-chlordane	*oxy*-chlordane		
	trans-nonachlor	*trans*-nonachlor		
	cis-nonachlor	*cis*-nonachlor		
	Mirex			
DDTs		**Dichlorodiphenyldichloroethane**		
	o,p'-DDT	*o,p'*-dichloro-α,α-diphenyl-β,β,β-trichloroethane	X	X
	p,p'-DDT	*p,p'*-dichloro-α,α-diphenyl-β,β,β-trichloroethane		X
	o,p'-DDD	*o,p'*-dichloro-diphenyl-dichloroethylene	X	X
	p,p'-DDD	*p,p'*-dichloro-diphenyl-dichloroethylene	X	X
	o,p'-DDE	*o,p'*-dichloro-diphenyl-dichloroethane	X	X
	p,p'-DDE	*p,p'*-dichloro-diphenyl-dichloroethane		
PCBs		**Polychlorinated biphenyl**		
	PCB 28	2,4,4'-Trichlorobiphenyl		
	PCB 52	2,2',5,5'-Tetrachlorobiphenyl		
	PCB 99	2,2',4,4',5-Pentachlorobiphenyl		
	PCB 101	2,2'4,5,5'-Pentachlorobiphenyl		
	PCB 105	2,3,3',4,4'-Pentachlorobiphenyl		
	PCB 118	2,3'4,4',5-Pentachlorobiphenyl		
	PCB 138	2,2',3,4,4',5'-Hexachlorobiphenyl		
	PCB 153	2,2'4,4',5,5'-Hexachlorobiphenyl		
	PCB 180	2,2',3,4,4',5,5'-Heptachlorobiphenyl		
	PCB 183	2,2',3,4,4',5',6-Heptachlorobiphenyl		
	PCB 187	2,2',3,4',5,5',6-Heptachlorobiphenyl		
	PCB 194	2,2',3,3',4,4',5,5'-Octachlorobiphenyl		
PFSAs		**Perfluorosulfonate**		
	PFOSA	Perfluorooctanesulfonamide	X	X
	PFBS	perfluorobutane sulfonate	X	X
	PFHxS	Perfluorohexane sulfonate		
	PFHpS	Perfluoroheptane sulfonate		X
	PFOS	Perfluorooctane sulfonate		
	PFDcS	Perfluorodecanesulfonate		X

	PFPS	Perfluoropentanesulfonate	X	X
	PFNS	Perfluorononanesulfonate		X
	brPFOS	branched PFOS isomer		
	6:2FTS	6:2 fluorotelomer sulfonate	X	X
	8:2FTS	8:2 fluorotelomer sulfonate		X
PFCAs		**Perflorocarboxylate**		
	PFHxA	Perfluorohexanoate	X	X
	PFHpA	Perfluoroheptanoate	X	X
	PFDoA	Perfluorododecanoate		
	PFTeA	Perfluorotretradecanoate		
	PFPA	Perfluoropentanoate	X	X
	PFUnA	Perfluoroundecanoate		
	PFTriA	Perfluorotridecanoate		
	PFDcA	Perfluorodecanoate		
	PFOA	Perfluorooctaoate		
	PFNA	Perfluorononanoate		

2.4.1 Determination of OCs

Extraction and clean up

Plasma samples of the glaucous gulls were thawed at room temperature before 1g (± 0.1 g) sample was transferred to a 15 mL glass vial and spiked with 60 µL of internal standard solution (^{13}C, DDT I, PCB I and PEST I, 25 pg/µL) and thoroughly vortexed. To enhance phase separation and ensure denaturation of proteins, 2 mL deionized water saturated with ammonium sulfate and 2 mL ethanol were added to the samples. After mixing, the samples were extracted twice by adding 6 mL n-hexane and vortexed. Phase separation was allowed for 15 min before the hexane phase was transferred to a weighed 15 mL glass tube. The combined extracts were up-concentrated to 0.2 mL using a RapidVap (Labconco corporation, Kansas City, MO, USA). For the lipid determination the sample was carefully evaporated to dryness using a gentle stream of nitrogen gas, weighted and subsequently re-dissolved in 0.5 mL hexane. The lipid content of the sample was determined by subtracting the weight of the empty 15 mL vial from the weight of the dried sample.

Clean-up by a Florisil column (150-250 µm; Merck, Darmstadt, Germany, heated at 450°C for 8 hours) was conducted for biological matrix removal. After clean-up, a small amount of isooctane was added to all samples and further up-concentrated to 0.2 mL using RapidVap. Samples were transferred to gas chromatography (GC) vials with insert, and a N_2-evaporation

unit (N_2 purity of 99.995%, quality 5.0, Hydrogas, Porsgrunn, Norway) was used to up-concentrate to approximately 30 µL. 40 µL of ^{13}C PCB-159 (213 pg/µL) were added to all samples as a recovery standard prior to analysis by gas chromatograph-mass spectrometry (GC/MS). The samples were kept cool (4°C) until instrumental analysis.

Instrumental analysis

The organochlorine contaminants were analyzed as described in detail by Herzke et al. (2009) and Sonne et al. (2010). Congeners of PCBs, DDT and their metabolites, HCB, HCHs, chlordane and Mirex were analyzed by an Agilent 7890 gas chromatograph (GC) equipped with a triple-quadruple mass-spectrometer, Quattro Micro GC (Water Corporation, Manchester UK) and operated in electron impact (EI) mode. For all organochlorine pesticides (OCPs) except DDT an Agilent 7890A gas chromatograph was used in analysis equipped with a 5975C mass spectrometer operated in negative chemical ionization (NCI) mode (Agilent Technology, Böblingen, Germany). The EI ion-source temperature was set to 250°C and the NCI to 160°C, while the transfer line was kept at 280°C.

A DB-5MS column (30m; 0.25 µm film thickness, 0.25 mm inner diameter; J & W Scientific Folsom, USA) were used for separation with helium (6.0 quality; Hydrogas, Porsgrunn, Norway) as carrier gas at a flow rate of 1 mL/min. To enhance the separation the temperature program was set to 70°C for 2 min, followed by a temperature increase of 15°C/min to 180°C, before a final temperature incline of 5°C/min to 280°C where the temperature was held for 10 min.

2.4.2 Determination of PFAS

Extraction and Clean up

Plasma samples were analyzed for PFASs following a method previously described by Powley et al. (2005) and modified for plasma and blood by Hanssen et al. (2013). Frozen plasma samples were thawed at room temperature, and a volume of 0.2 mL was transferred into an Eppendorf-centrifuge tube and spiked with 20 µL of internal standard mixture (0.5 ng/µL ^{13}C PFAS mix). Following the addition of 1 mL methanol the samples were thoroughly vortexed and put in three consecutive 10 min ultrasonic treatment baths where extraction itself was achieved. In between ultrasonic treatment the tubes were vortexed, denaturizing the proteins.

When extraction of PFASs into the methanol was completed, the tubes were centrifuged for 10 minutes at 10 000 rpm to enhance phase separation.

After centrifugation the methanol supernatant was added to new Eppendorf tubes prepared with 25 mg ENVI-Carb 120/400 (Supelco 57210-U, Bellafonte, PA, USA) in 50 µL glacial acetic acid (Merck, Darmstadt, Germany) and vortexed thoroughly. After additional centrifugation for 10 minutes at 10 000 rpm, 0.5 mL of the supernatant was transferred to glass vials (2 mL) and 20 µL of recovery standard (0,1 ng/µL 3,7-diMeo-PFOA) was added. Samples were kept cool (4°C) until instrumental analysis.

Prior to analysis, an adequate of 50 µL of the samples was added to an autosampler vial with insert along with 50 µL of 2 mM NH_4OAc (Sigma-Aldrich, St. Louis, MO, USA) in water before vortexed.

Instrumental analysis

The samples were analyzed using an ultra-high pressure liquid chromatography triple-quadrupole mass spectrometry (UHPLC-MS/MS) as outlined by Hanssen et al. (2013). Analysis was performed on a Thermo Scientific quaternary Accela 1250 pump (Thermo Fisher Scientific Inc., Waltham, Ma, USA) along with a PAL Sample Manager (Thermo Fisher Scientific Inc., Waltham, MA, USA) which was coupled to a Thermo Scientific Vantage MS/MS (Vantage TSQ) (Thermo Fisher Scientific Inc., Waltham, MA, USA). The samples (10 µL) were injected on a Waters Acquity UPLC HSS 3 T column (2.1 x 100 mm, 1.8 µm) (Waters Corporation, Milford, MA, USA) equipped with a Waters Van guard HSS T3 guard column (2.1 x 5mm, 1.8 µm) (Waters Corporation, Milford, MA, USA). Separation of the samples was achieved by using the mobile phases of 2 mM NH_4OAc in 90:10 methanol/water and 2 mM NH_4OAc in methanol.

2.4.3 Quantification and Quality Assurance

Following the instrumental analysis, quantification of all compounds was conducted according to the internal standard method provided by NILU. Standards with known concentrations of ^{12}C and ^{13}C – labelled equivalents representing all groups of contaminants were analyzed. The peak of the native and labelled standards in the mass chromatogram produced a standard curve used to calculate concentrations in the sample extracts using Equation 1.

Equation 1:

$$C_{sample} = Rf(C_{std} \times Area_{sample})/Area_{std}$$

Where C_{sample} = the unknown concentration of the sample, C_{std} = the known concentration of the standard, A_{sample} = the known area under the curve from the GC/MS chromatogram of the sample, and A_{std} = the known area of the internal standard under the GC/MS curve. Rf represents the response factor from the areas and concentrations of the ^{12}C and ^{13}C labelled equivalents acquired in the standard chromatogram. Concentrations of the compounds were calculated in wet weight (*ww*), they were expressed in pg/ml or pg/g but further converted to ng/g in order to compare the concentrations.

For validation of repeatability and quality control of the method used in OCs and PFASs analysis, one blank was prepared for each batch of more than 10 and 20 samples, respectively. The analytical method was validated using a standard reference material (SRM, 1957 Human Serum, National Institute of Standards and Technology, Gaithersburg, MD, USA). The blank and the SRM samples underwent the same treatment as the glaucous gull plasma samples. The reference samples were within the given limit of accuracy and the blanks were below the limit of acceptable contamination determined by the laboratory. The average value of the blank signals was used to calculate the limit of detection (LOD) plus three times the corresponding standard deviation (SD). All solvents used in analysis were purchased from Merck-Schuchardt (Hohenbrunn, Germany) and of Supersolv® grade.

2.5 Stable isotope analysis

All isotope analyses were conducted at the Littoral Environnement et Sociétés laboratory (LIENSs) in La Rochelle, France. Feathers were cleansed of surface lipids and contaminants in a solution of 2:1 chloroform:methanol and then rinsed twice in a methanol solution. After being dried at 45°C for 48 hours the feathers were cut into small pieces to produce a fine powder. The feathers were accurately weighted between 0.1 and 0.4 mg. Nitrogen and carbon stable isotopes were measured using a continuous flow mass spectrometer (Thermo Scientific Delta V Advantage) coupled to an elemental analyzer (Thermo Scientific Flash EA 1112). Results are expressed in δ notation in ppt (‰) relative to Vienna PeeDee Belemnite (VPDB) (Lucia et al., 2016; Lucia et al., 2017). Replicate measurements of internal laboratory standards (acetanilide) were used for every 10 samples to check accuracy and indicated measurement errors <0.15‰

for both $\delta^{13}C$ and $\delta^{15}N$ values. USGS-61 and USGS-62 reference materials were also analysed for calibration. The stable isotope values are given as mean ± SD (‰). Body feathers were not available from year 2018 in Bjørnøya.

2.6 Statistical analysis

Statistical analyzes were performed in the statistical software R (R version 4.0.3 and Rstudio version 1.4.1103). The significance level was set to p=0.05 for all tests.

Contaminants were excluded from statistical analysis if they were detected in less than 45% of the samples in each location. Values below LOD (limit of detection) were assigned half the limit of LOD. Because of variance in contaminant burdens between the two locations, different compounds were excluded in each location. To reduce the number of variables, contaminants were tested for correlation using Spearman's rank correlation tests and grouped thereafter for the statistical analysis (Appendix D). DDTs and its metabolites and other chlorinated pesticides were grouped as organochlorine pesticides (OCPs). PCB congeners correlated well with each other (r: 0.59 – 0.98, $p < 0.001$), except for PCB-52 ($r < 0.2$, $p > 0.01$) and PCB-101 ($r < 0.1$, $p > 0.01$), but because of low concentrations they were summed together as one group. Fluorinated compounds were separated in two groups, sulfonates (PFSAs) and carboxylates (PFCAs). This resulted in a total of 4 groups of compounds: ΣOCPs, ΣPCBs, ΣPFSAs and ΣPFCAs. Correlation matrices can be found in Appendix D. Variables were tested for normality by using Shapiro-Wilk's W-tests ($p > 0.05$). To reduce variance heterogeneity and skewness, all contaminant groups were logarithmically transformed as a general rule.

A body condition index (BCI) was estimated using a principal component analysis (PCA) with a single size measure calculated from total head length, wing length and gonys height. The calculation was done separately for sexes as glaucous gulls are sexually dimorphic (Sagerup et al., 2009). Linear regression with the first principal component (PC1) against body weight as the dependent variable was used to obtain standardized residuals defined as the individual BCI (Sagerup et al., 2009; Melnes et al., 2017).

Lipids (%) were not discussed further or accounted for in statistical analysis as there were no significant difference between locations ($t_{124}=0.18$, $p> 0.05$). Mean ± standard deviation (SD), median and range of lipids can be found in Appendix A.

Linear mixed-effects models (LMM) in R-package lme4 were used to analyze the effect of location, year, sex, BCI and dietary ecology ($\delta^{13}C$ and $\delta^{15}N$) on ∑OCPs, ∑PCBs, ∑PFSAs and ∑PFCAs. Individual ID was set as a random effect variable to account for repeated measurements over the years. Interactions between predictors were investigated and included in the model selection if significant. Predictor variables that were highly correlated were not included in the same models ($\delta^{13}C$ and $\delta^{15}N$, $r= 0.57$, $p< 0.05$) to avoid multicollinearity. The statistical tool variance inflation factor (VIF) was used to assess dependence or multicollinearity between predictor variables. Variables with a VIF < 3 were considered to not be collinear with other variables (Zuur et al., 2010). Model averaging based on Akaike's Information Criterion (AIC) was used to make inference of all candidate models using R-package *MuMIn.* All models were ranked according to AIC and AIC weight, the best model having the lowest AICc (second-order AIC). Diagnostic residual plots were used for the highest ranked models to investigate if the distribution of the model residuals met the model assumptions.

3 Results

3.1 Biological characteristics

The mean, SD, median and sample size of biometric variables are presented in Appendix A (Table A1-2). The mean body weight of male glaucous gulls was significantly higher than in females in both locations ($p< 0.001$). Across all sampling years, males and females from Kongsfjorden had a mean body weight of 1736 ± 117 g and 1410 ± 105 g, respectively. At Bjørnøya, the mean body weight of males and females was 1770 ± 107 g and 1436 ± 104 g, respectively. The calculated mean BCI for males and females did not differ significantly between sexes or locations (sex: $t_{105}= 0.22$, $p= 0.83$; location: $t_{127}= 0.18$, $p= 0.86$). The mean and standard deviation of each sex is 0 ± 1. Mean BCI in Bjørnøya decreased progressively between 2015 to 2019 ($p< 0.01$), while in Kongsfjorden BCI fluctuated with an increase from 2016 to 2017 ($p< 0.05$) followed by a slight decrease between 2017 to 2019 ($p< 0.05$) (Figure 2).

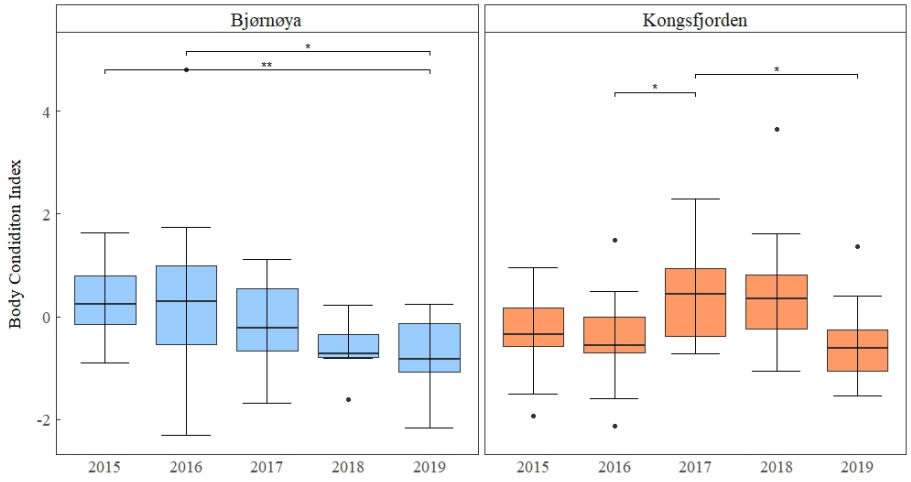

Figure 2: Body condition index (BCI) of glaucous gulls from Bjørnøya and Kongsfjorden sampled during the breeding seasons of 2015 until 2019. Top and bottom of boxes show first and third quantiles, the whiskers represent minimum and maximum values. The horizontal line inside the box represents the median. Asterisks (*) indicate significant differences between years according to t-test (*, $p< 0.05$, **, $p< 0.01$).

3.2 Contaminant concentrations and patterns

Among all the compounds analyzed, a total of 34 compounds were detected in >45% of the samples in Bjørnøya and 30 compounds in Kongsfjorden. The mean, standard deviation, median and range of the contaminant concentrations are listed in Appendix C and are given in ng/g wet weight (*ww*). The concentration of ΣOHCs differed significantly between populations throughout the sampling period (t_{114}= -15.49, $p< 0.001$), being 5.3 times higher in Bjørnøya compared to Kongsfjorden with a mean concentration of 229 ± 176 ng/g *ww* in Kongsfjorden and 1217 ± 591 ng/g *ww* at Bjørnøya. Plasma contaminant concentrations were on average 87% higher in males than in females in Kongsfjorden and 36% higher in Bjørnøya. In Kongsfjorden males had significantly higher plasma concentrations of ΣOCP, ΣPCB and ΣPFCA (ΣOCP: t_{25}=3.06, $p< 0.01$; ΣPCB: t_{30}=2.87, $p< 0.01$; ΣPFCAs: t_{39}=2.92, $p< 0.01$) compared to females. Males from Bjørnøya exhibited significantly greater plasma concentrations of ΣOCP, ΣPFCA and ΣPFSA (ΣOCP: t_{88}=3.03, $p< 0.01$; ΣPFCA: t_{91}=3.37, $p< 0.01$; ΣPFSA: t_{91}=3.69, $p< 0.001$) compared to females.

The overall highest concentrations of all contaminant groups (ΣOCPs, ΣPCBs, ΣPFCAs and ΣPFSAs) were found in Bjørnøya for all the 5 sampling years. Concentrations of ΣPCBs was the overall highest of all OHCs measured in Kongsfjorden, with a percentage contribution of 65 ± 7 % across all years followed by ΣOCPs (21 ± 7%) and ΣPFASs (13 ± 11%). For Bjørnøya,

ΣPCBs and ΣPFASs contributed equally high (PCBs: 43 ± 12%; PFASs: 44 ± 14%), followed by ΣOCPs (17 ± 4%) (Figure 3). For both locations, the hexachlorinated PCB-153 was the predominant PCB congener (Kongsfjorden 35% of ΣPCBs, Bjørnøya 31% of ΣPCBs) followed by PCB-138, -180, -118. The predominant ΣOCPs compound was *p,p'*-DDE, with a contribution of 44% to ΣOCPs in Kongsfjorden and 74% in Bjørnøya followed by *oxy*-chlordane. Of the analysed PFASs, the predominant compound was PFOS (Kongsfjorden 46% of ∑PFASs, Bjørnøya 56% of ∑PFASs). The contaminant pattern of each group to the total concentration of OHCs and concentrations of each compound group per location are illustrated in Figure 3.

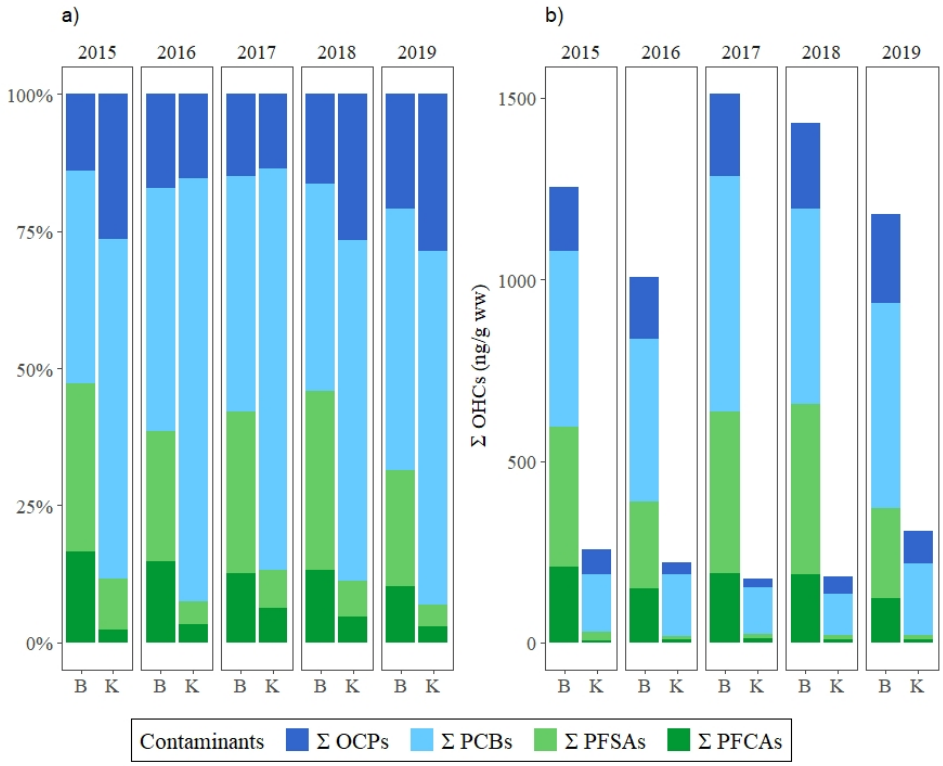

Figure 3: a) The contaminant pattern (%) of each contaminant group to the total OHC burden in glaucous gulls breeding in Bjørnøya (B) and Kongsfjorden (K) during the breeding seasons of 2015 until 2019. b) Mean concentrations (ng/g *ww*) of OHCs in glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding season in 2015 until 2019. Colors indicating the contribution of each contaminant group.

Concentrations of plasma ∑OCPs had a stable trend across sampling years in Bjørnøya, in contrast to a U-shaped concentration trend according to sampling year in Kongsfjorden. Namely, OCP concentrations in Kongsfjorden decreased between 2015 and 2017 ($p < 0.05$), being lowest in 2017, before increasing again from 2017 to 2019 ($p < 0.001$) (Figure 4). ∑PCBs

remained stable in both locations across sampling years. Variations in ∑PFSAs across years were detected in Bjørnøya as there was an increase in concentrations between 2016 and 2017 ($p < 0.001$) before decreasing from 2017 to 2019 ($p < 0.001$). Concentrations of ∑PFCAs decreased in Bjørnøya between 2015 to 2019 ($p < 0.001$) while in Kongsfjorden concentrations slightly increased between 2015 to 2017 ($p < 0.05$) (Figure 4).

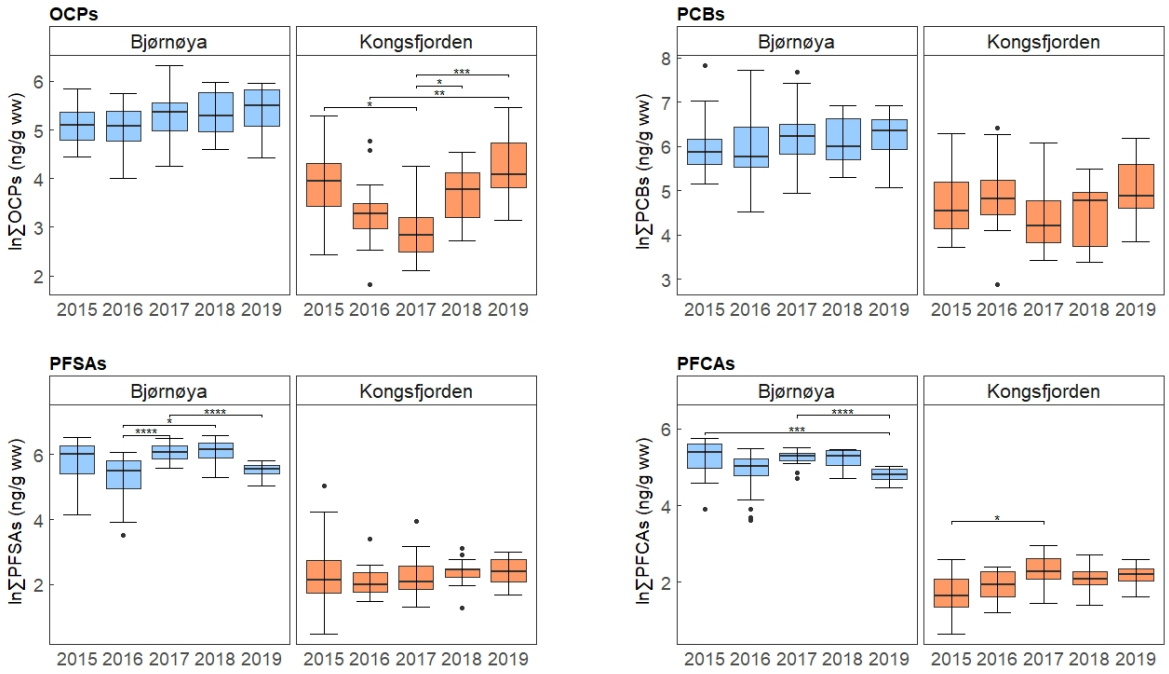

Figure 4: Trends of OCPs, PCBs, PFSAs and PFCAs (ng/g *ww*) in blood plasma of glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. Top and bottom of boxes show first and third quantiles, the whiskers represent minimum and maximum values. The horizontal line inside the box represents the median. Asterisks (*) indicate significant differences between years according to t-test (*, $p < 0.05$, **, $p < 0.01$, ***, $p < 0.001$).

3.3 Stable isotope signatures

Across all years the $\delta^{15}N$ ranged from 12.11‰ to 18.39‰ for glaucous gulls in Kongsfjorden and from 11.62‰ to 17.89‰ in Bjørnøya. $\delta^{13}C$ ranged from -20.05‰ to -16.05‰ in Kongsfjorden and -20.57‰ to -17.61‰ in Bjørnøya. Glaucous gull males from Bjørnøya exhibited slightly greater $\delta^{15}N$ signatures than males from Kongsfjorden ($t_{45} = 2.79$, $p < 0.01$), while there was no significant difference between females from both locations ($t_{70} = 1.34$, $p > 0.05$). For $\delta^{13}C$ there was no significant difference between locations for either sex (males: $t_{28} = -1.69$, $p > 0.05$; females: $t_{66} = -2.03$, $p = 0.05$). Males were enriched in $\delta^{15}N$ (Kongsfjorden: $t_{52} =$

2.6, p< 0.05; Bjørnøya: t_{55}= 3.20, p< 0.01) and $\delta^{13}C$ (Kongsfjorden: t_{36}= 3.24, p< 0.01; Bjørnøya: t_{55}= 4.02, p< 0.001) compared to females in both locations. The $\delta^{15}N$ and $\delta^{13}C$ values in male and female glaucous gull body feathers from Kongsfjorden and Bjørnøya are illustrated in Figure 5.

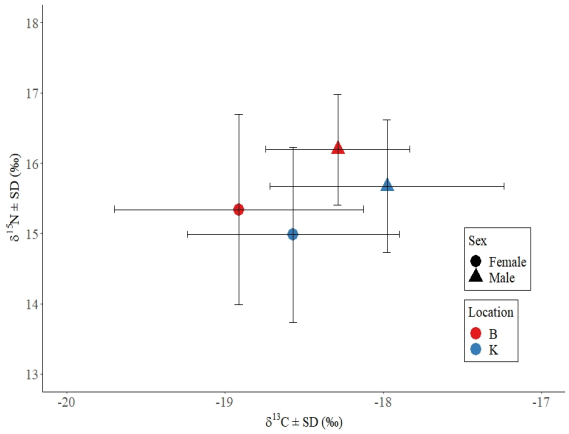

Figure 5: Bi plot with stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopic signatures (mean ± SD, ‰) in body feathers of female and male glaucous gulls breeding in Bjørnøya (B) and Kongsfjorden (K) in 2015 until 2019. Error bars represent the standard deviation (SD) of trophic position on the y-axis and carbon source on the x-axis.

Throughout the sampling years stable isotope ratios of nitrogen remained stable in Bjørnøya ($F_{3.40}$= 0.21, p= 0.89), while in Kongsfjorden $\delta^{15}N$ varied significantly between years ($F_{4.59}$= 10.15, p< 0.001) being higher in 2015 and lower in 2016 compared to the other years (Figure 6). Kongsfjorden had significantly lower $\delta^{15}N$ values in 2016 and 2017 compared to Bjørnøya (2016: t_{47}= -5.13, p< 0.001; 2017: t_{30}=-2.63, p= 0.01). Stable isotope ratios of carbon did not significantly vary across sampling years in either location (Bjørnøya: $F_{3.42}$= 2.40, p= 0.082; Kongsfjorden: $F_{4.65}$= 1.77, p= 0.146). Average (mean ± SD) stable isotopic signatures for each sampling year are presented in Figure 6 and in Appendix E.

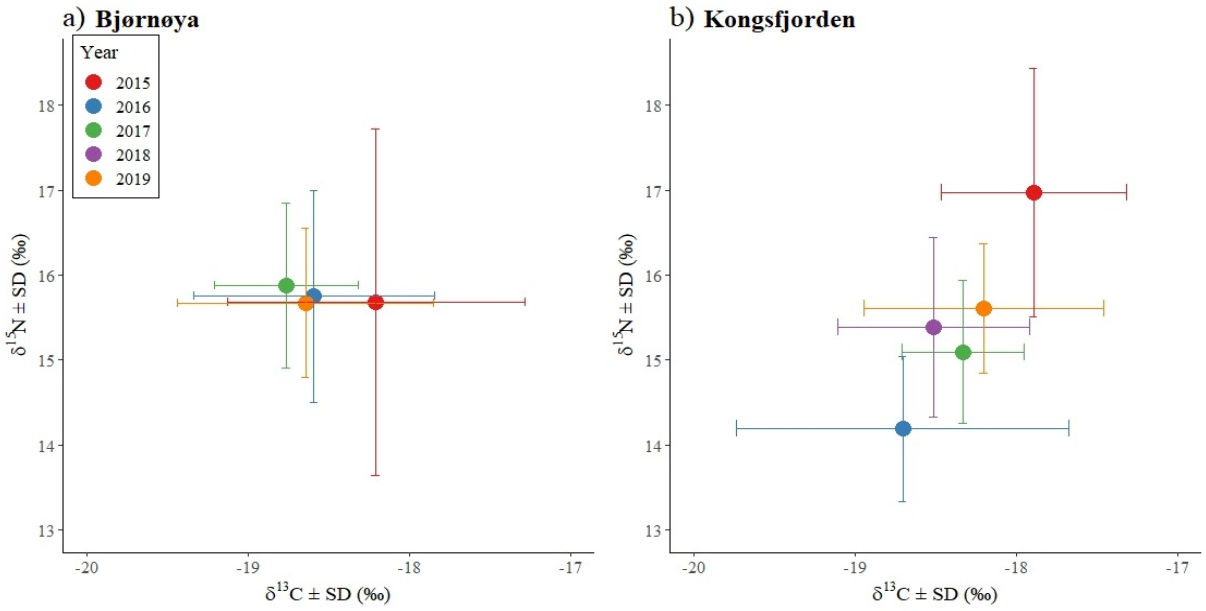

Figure 6: Bi plot of mean ± standard deviation (SD) for stable isotope ratios of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) in feathers from glaucous gulls breeding in a) Bjørnøya and b) Kongsfjorden in 2015 to 2019. Dots and their respective colors represent mean values for each sampling year. Error bars represent the SD of trophic position on the y-axis and carbon source on the x-axis.

3.4 Drivers of OHCs

Model selection for all contaminant groups can be found in Appendix G. The best-fitting models based on AIC values all included location and sex as strong predictors of variation in ∑OCPs, ∑PCBs, ∑PFCAs and ∑PFSAs. BCI, sampling year and $\delta^{13}C$ were also important drivers of contaminant concentrations, receiving a high support in some of the selected models (ΔAICc < 2, Table G). The best model predicting variation in ∑OCPs included BCI and the interaction between sampling year and location, explaining a high proportion of variability (R^2m=0.74, R^2c=0.86). The interaction term between location and sampling year indicated that the locations did not follow the same trend in OCP concentrations over sampling years (Table 2, Figure 4). OCP concentrations also decreased with increasing BCI in Bjørnøya. The best fit model explaining variations in ∑PCBs (R^2m=0.51, R^2c=0.79) included only BCI in addition to location and sex and was also negatively related to PCB concentrations in Bjørnøya. Further, the model including $\delta^{13}C$ and the interaction between location and sampling year was the best in predicting plasma ∑PFSA concentrations (R^2m= 0.94, R^2c= 0.95). Concentrations of PFSAs increased with increasing proportion of carbon (Figure 7) in both locations. The top model explaining variations in ∑PFCAs (R^2m= 0.92, R^2c= 0.94) included sampling year and the interaction between location and $\delta^{13}C$. There was an increase in concentration with increased

$\delta^{13}C$ in Bjørnøya, but no relationship was detected in Kongsfjorden (Least Square Means: Kongsfjorden: 2.13 ± 0.06, Bjørnøya: 5.00 ± 0.05) (Figure 7; Table 2). Correlations between all contaminant groups and stable isotopes are presented in Appendix F.

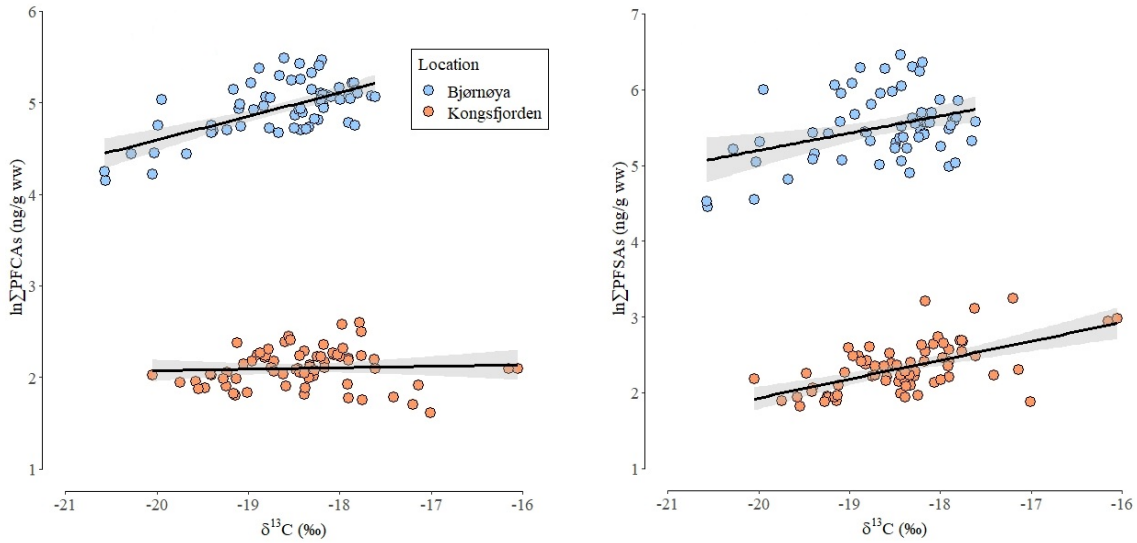

Figure 7: Linear regression models (LMER) of total PFSA and PFCA concentrations in relation to $\delta^{13}C$ (‰) with 95% confidence intervals from samples of glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding season from 2015 until 2019. The y-axis shows partial residuals from the highest ranked model controlled for the effects of location (Bjørnøya or Kongsfjorden), sampling year (2015-2019), body condition index (BCI) and sex (female or male) (Appendix G).

Table 2: Linear regression parameter estimates from model selection with 95% confidence intervals (CI) explaining variations in ln-transformed OHC (ng/g *ww*) concentrations from glaucous gulls breeding in Bjørnøya and Kongsfjorden from 2015 until 2019. Predictors included $\delta^{15}N$ and $\delta^{13}C$ values in feathers, location (Bjørnøya or Kongsfjorden), sampling year (2015-2019), body condition and sex (female or male). Significance was set to 0.05 and if the 95% confidence interval did not span 0, there was a significant relationship between response and predictor.

Parameter	Estimate	CI (95%)	p-value
ln∑OCPs			
Intercept	4.788	4.396 - 5.179	< 0.001
Location K	-1.115	-1.661 - (-0.569)	**< 0.001**
Year 2016	0.150	-0.240 - 0.537	0.373
Year 2017	0.263	-0.164 - 0.690	0.208
Year 2018	0.032	-0.427 - 0.493	0.850
Year2019	0.458	-0.005 - 0.921	0.045
Male	0.375	0.161 - 0.585	**0.001**
BCI	-0.102	-0.193 - (-0.013)	0.025
Location K: 2016	-0.653	-1.241 - (-0.065)	**0.039**
Location K: 2017	-0.932	-1.555 - (-0.306)	**0.006**
Location K: 2019	-0.101	-0.744 - 0.542	0.769
ln∑PCBs			
Intercept	5.867	5.621 - 6.113	< 0.001
Location K	-1.250	-1.526 - (-0.973)	**< 0.001**
Male	0.378	0.096 - 0.659	**0.011**
BCI	-0.216	-0.313 - (-0.109)	**< 0.001**
ln∑PFSAs			
Intercept	5.170	4.768 - 5.538	< 0.001
Location K	-2.544	-3.071 - (-1.932)	**< 0.001**
Year 2016	-0.018	-0.431 - 0.365	0.929
Year 2017	0.794	0.363 - 1.230	**<0.001**
Year 2018	-0.296	-0.818 - 0.159	0.211
Year 2019	0.263	-0.182 - 0.716	0.266
$\delta^{13}C$	0.121	0.027 – 0.209	0.012
Male	0.308	0.122 - 0.524	**0.004**
Location K: 2016	-0.561	-1.182 - 0.030	0.066
Location K: 2017	-1.244	-1.920 - (-0.629)	**< 0.001**
Location K: 2018	-0.665	-1.370 – (-0.038)	0.046
ln∑PFCAs			
Intercept	4.717	4.499 - 4.934	< 0.001
Location K	-2.868	-3.008 - (-2.729)	**< 0.001**
Year 2016	0.032	-0.188 - 0.253	0.784
Year 2017	0.423	0.192 - 0.655	**< 0.001**
Year 2018	0.166	-0.113 - 0.448	0.259
Year 2019	0.103	-0.135 - 0.343	0.411
$\delta^{13}C$	0.146	0.050 - 0.243	**0.004**
Male	0.268	0.124 - 0.413	**< 0.001**
Location K: $\delta^{13}C$	-0.183	-0.308 - (-0.058)	**0.006**

4 Discussion

Previous studies have reported organohalogenated contaminant (OHC) concentrations in glaucous gulls from Bjørnøya (Bustnes et al., 2003b; Verreault et al., 2005; Verreault et al., 2018) and Kongsfjorden (Verreault et al., 2007; Melnes et al., 2017; Sebastiano et al., 2020) separately but none have directly compared the two populations despite striking differences. We showed that glaucous gulls from Bjørnøya exhibited 5.3 times higher body burdens of OHCs compared to Kongsfjorden gulls. This study examined 5 years of contaminant and stable isotope data to investigate if differences in OHC plasma concentrations were related to the dietary ecology of the two populations. Our data collected on breeding birds showed high plasma OHC concentrations but weak associations with feather stable isotope ratios of nitrogen, indicating that the birds' trophic position during the non-breeding season (indicated by feather $\delta^{15}N$) is a weak driver of variations in plasma levels of OHCs during the breeding season (Table 2). Variations in contaminant occurrence were related to $\delta^{13}C$ only for PFSAs and PFCAs when biological variables sex and body condition index (BCI) were accounted for in the model (Figure 7).

4.1 Contaminant levels and patterns

In this study, concentrations of OHCs were significantly higher in Bjørnøya compared to Kongsfjorden and were consistently higher in all 5 sampling years (Figure 3). Contaminant patterns were dominated by PCBs and OCPs in Kongsfjorden and PCBs and PFASs in Bjørnøya. ∑PCBs (sum of 12 congeners) represented a high proportion of the total contaminant load in all sampling years. Many marine avian top predators such as great skuas (Leat et al., 2019), great black-backed gulls *Larus marinus* (Bustnes et al., 2005) and glaucous gulls (Verreault et al., 2010) have been reported to have high levels of PCBs due to their high persistence in the environment. In Kongsfjorden, plasma ∑PCB levels were comparable to previously reported levels (Melnes et al., 2017). Over our study period (2015-2019), Bjørnøya exhibited three times higher mean concentrations of ∑PCBs compared to Kongsfjorden and higher than previously reported on Bjørnøya in 2001 (Verreault et al., 2004) (Table C). This increase is unexpected as PCB time-trends have shown decreasing trends in Arctic biota (AMAP, 2016; Dietz et al., 2019). The reason for the increase is unknown but could be related to climate change and decreased ice cover resulting in changes in dietary composition and feeding habits in glaucous gulls and their prey (McKinney et al., 2013). Within each population,

concentrations did not show temporal variations over the 5 year sampling period (Figure 4). PCB-153 was the major PCB congener found in this study followed by PCB-138 and PCB-180, respectively. PCB-153 is thought to be one of the most persistent congeners found in the biota and has been reported in high concentrations in several studies on glaucous gulls (Gabrielsen et al., 1995; Bustnes et al., 2004; Sagerup et al., 2009; Melnes et al., 2017).

The mean ∑OCPs (all sampling years) were slightly higher in Kongsfjorden than what has been previously reported (Melnes et al., 2017). However, this can be explained by temporal variations in OCPs whose concentrations varied across sampling years being higher in 2015 and 2019 compared to the years 2016 - 2018 (Figure 4). The yearly variations in plasma organochlorine pesticides may be explained by annual diet variability (Bustnes et al., 2000). As observed for ∑PCBs, Bjørnøya gulls exhibited four times higher levels of ∑OCPs than glaucous gulls from Kongsfjorden. In the study by Verreault et al. (2007), the levels of OCPs reported in glaucous gulls from Bjørnøya were higher than levels found in the current study. Decreasing concentrations of OCPs in Bjørnøya is consistent with the general observed trend of decreasing "legacy" contaminants (Dietz et al., 2019). The major organochlorine pesticide detected was the DDT metabolite *p,p′*-DDE, which is commonly detected in high concentrations in Arctic biota (Borgå et al., 2007). Kongsfjorden gulls exhibited higher mean *p,p′*-DDE levels than reported by Melnes et al. (2017). According to air concentrations monitored at the Zeppelin station in Ny-Ålesund, there has been an increased proportion in aged sources of DDT (*p,p′*-DDE) from 1994 to 2017 (Bohlin-Nizzetto et al., 2019; Wong et al., 2021). Also, the prey items of seabirds have been found to be abundant with *p,p′*-DDE, as DDT is metabolised by many levels of the food chain. Accumulation in the glaucous gull may therefore be a combination of uptake from their diet and from metabolising DDT (Borgå et al., 2001).

The most striking difference in OHC concentrations between Kongsfjorden and Bjørnøya was observed for PFASs (Figure 3). Glaucous gulls from Bjørnøya exhibited more than 20 times higher concentration in ∑PFASs (∑PFSAs and ∑PFCAs) compared to Kongsfjorden. Few studies have investigated PFAS concentrations in glaucous gulls at Bjørnøya, the first and only one conducted in 2004 by Verreault et al. (2005). They revealed high concentrations of sulfonates (PFSAs) and carboxylates (PFCAs) in liver, egg, brain and plasma samples, PFOS being the predominant compound. In the present study, PFOS was also the dominating PFAS compound in both colonies and had the highest concentration of all screened contaminants in

Bjørnøya. The levels that we measured were twice higher than those reported by Verreault et al. (2005). In contrast, Kongsfjorden had lower or similar PFAS levels compared to earlier studies (Melnes et al., 2017; Sebastiano et al., 2020). High levels were also found for PFUnA, brPFOS, PFTriA, PFDcA and PFNA, which is in accordance with patterns detected in plasma and eggs of glaucous gulls and other Arctic seabird species (Verreault et al., 2005; Miljeteig et al., 2009; Lucia et al., 2015; Melnes et al., 2017; Sebastiano et al., 2020).

The contribution of PFASs to the total contaminants load across all years was equal to PCBs in Bjørnøya (Figure 3), suggesting the increasing prevalence of PFASs in Arctic wildlife. PFOS and its related compounds have been regulated by the Stockholm Convention since 2009 and are listed under Annex B (restricted) after being detected in the abiotic environment, biota, food items and in humans (Muir et al., 2019). In studies of Arctic wildlife, an increasing trend of PFOS was detected until the mid-2000s when it presumably hit its peak, followed by a general decreasing trend the following years (Rigét et al., 2013). However, PFOS still represents an environmental problem due to its potential for long-range transport, high persistence in the environment, high potential for bioaccumulation and its potential to cause toxic effects (Schiavone et al., 2009; Butt et al., 2010).

Pharmacokinetic studies suggested both PFSAs and PFCAs to be easily absorbed from oral exposure and poorly metabolized (Harrad, 2010). The rate of elimination generally decreases with increasing length of the fluorinated carbon chain as long-chained PFSAs and PFCAs are more easily bioaccumulated than short-chained compounds (Giesy and Kannan, 2002; Tomy et al., 2004; Buck et al., 2011). However, the molecular size might limit the ability to bioaccumulate when the chain is longer than 12 carbons (Martin et al., 2003). Following regulations of PFASs, there has been a change in industrial practices where long-chained PFASs compounds have been replaced by either short-chained homologues or other fluorinated and non-fluorinated compounds. The environmental levels of these compounds are therefore expected to increase in the coming years (Yeung et al., 2017).

Overall, the high OHC levels reported in our study might affect the health of glaucous gulls and might cause negative effects at the population level. Namely, negative effects on survival have been related to increased OC exposure in glaucous gulls from Bjørnøya (Erikstad et al., 2013). Other studies have also related OHCs to increased absence from nest, reduced embryo quality,

lowered condition of chicks and increased corticosterone levels which might suppress parental care (Bustnes et al., 2001a; Bustnes et al., 2003b; Verboven et al., 2009; Verboven et al., 2010).

4.2 Drivers of OHC variations

4.2.1 Trophic position and carbon source

Body feathers were selected to investigate the dietary ecology of the two populations of glaucous gull. Feathers are irrigated by the blood flow and its circulating isotopic values during feather growth. When feather growth stops, the blood vessels will atrophy and become disconnected from the circulatory system of the bird (Burger and Gochfeld, 1992), therefore reflecting the isotopic values during feather growth. Based on the knowledge of the feather molt sequence of glaucous gulls, isotope values of body feathers sampled during the breeding period correspond to the winter period between January and March (Verreault et al., 2018). As plasma was the matrix used to investigate concentrations of OHCs, plasma OHCs and feather stable isotopes reflect different time scales as plasma display faster turnover rates, providing information between 2 weeks to one month prior to sampling (Hovinen et al., 2019).

With each trophic level a tissue enrichment in $\delta^{15}N$ of 3-4‰ can be expected for a consumer (Hobson and Clark, 1992; Kelly, 2000; Sagerup et al., 2002) while $\delta^{13}C$ shows limited trophic enrichment (Kelly, 2000). Within each colony, $\delta^{15}N$ in body feathers varied by up to 6.3 ‰ between individuals for all sampling years combined, corresponding to about two trophic levels between the highest and lowest measured values. Yet, stable isotope ratio analysis showed no significant difference between the two colonies expect for $\delta^{15}N$ in males where Bjørnøya exhibited higher values. However, due to large inter-individual variations within each colony it is likely that individuals from both locations had similar dietary composition ($\delta^{13}C$) and fed at similar trophic levels ($\delta^{15}N$) during the non-breeding season (Figure 5). Results from the present study showed that Kongsfjorden exhibited a larger degree of dietary variations across years compared to Bjørnøya (Figure 6). Namely, in Kongsfjorden glaucous gulls, the $\delta^{15}N$ values were between 6 – 15% lower in 2016 and 7-18% higher in 2015 compared to the other years. The observed annual changes may indicate changes in the trophic niche which can result from changing foraging strategies and/or food availability (Hovinen et al., 2019). A 19-year time series of black-legged kittiwake diet in Kongsfjorden reported decreasing contribution of Arctic species and increased contribution of Atlantic fishes, most likely related to recent climate change, indicating a shift towards a more variable diet (Vihtakari et al., 2018). The glaucous

gull is an opportunistic feeder, which means that dietary source may considerably vary among individual birds and years, what may play a role in the accumulation of contaminants (Sebastiano et al., 2020).

Previous studies on glaucous gulls have reported associations between feeding ecology and contaminants. Bustnes et al. (2000) investigated differences in organochlorine concentrations in two colonies of glaucous gulls breeding at Bjørnøya in relation to their diet. They found evidence that the colony situated in the seabird cliff had a higher intake of seabird eggs and exhibited higher levels of OCs compared to the colony situated at sea level which had a higher intake of fish, suggesting that the birds feeding at a higher trophic level exhibited higher pollutant burdens. Sagerup et al. (2002) also found positive correlations between $\delta^{15}N$ and OCs in glaucous gulls from Bjørnøya indicating that trophic level explained some of the observed variations in OC concentrations. In this study, no association was found between $\delta^{15}N$ and concentrations of ∑PCBs and ∑PFCAs (Appendix G). A positive correlation was found for ∑OCPs and ∑PFSAs with $\delta^{15}N$ (Figure E). However, when controlling for biological variables (sex and BCI) the $\delta^{15}N$ did not explain much variance in these contaminant groups (Appendix G). This may indicate that trophic level was not an important driver of OHCs in this study.

∑PFSAs were positively related to feather $\delta^{13}C$ whereas ∑PFCAs were only associated to $\delta^{13}C$ on Bjørnøya (Figure 7), suggesting that foraging habitat was generally a better predictor of PFAS concentrations in glaucous gulls than trophic level. Previous studies have usually either found negative (Leat et al., 2019) or no relationships between $\delta^{13}C$ and OHCs in seabird tissues (Ruus et al., 2002; Sagerup et al., 2002; Leat et al., 2013). However, in coastal breeding eagles *Haliaeetus leucocephalus*, Elliott et al. (2009) found increasing concentrations of PCBs and DDE with increasing $\delta^{13}C$ levels. Values of $\delta^{13}C$ are higher (i.e, less negative) in areas with high primary productivity, like in coastal areas, while there are lower (i.e, more negative) offshore (Hobson and Clark, 1992). Benthic habitats are also associated with increased $\delta^{13}C$, suggesting that glaucous gulls feeding in coastal/benthic environments might be more contaminated than pelagic feeding glaucous gulls.

4.2.2 Biological variables sex and BCI

Studies of glaucous gulls have shown that males have higher circulating concentrations of organochlorines than females after clutch completion (Bustnes et al., 2003a; Melnes et al.,

2017). This was also the case in the present study and sex was selected in all top models explaining variations in OHCs (Table 2). Sex differences in organochlorine levels have been attributed to egg laying and diet specialization in arctic seabirds (Norstrom et al., 1986). Females have the ability to deposit lipid associated contaminants to the egg yolk during egg formation, thereby eliminating some of their burdens (Verreault et al., 2006b). In the present study, female glaucous gulls exhibited lower levels of lipid soluble compounds supporting this hypothesis, except for PCBs in Bjørnøya. During the incubation period, females may utilize lipids from their body reserves to maintain body condition and associated energetic costs, which may lead to remobilization of PCBs (Bustnes et al., 2010) and possibly be an explanation for this finding. Studies have also found sex differences in the amphipathic PFASs compounds among arctic wildlife. Melnes et al. (2017) found concentrations of PFASs to be twice as high in female compared to male glaucous gulls, in contrast Blévin et al. (2017) found higher concentrations of PFASs in male black-legged kittiwakes compared to females. In the present study males in Bjørnøya exhibited higher concentrations of PFASs compared to females. In biological tissues, the concentration of PFOS increases with lipid content, suggesting that it is likely affected by similar biological processes as lipophilic compounds (Hitchcock et al., 2019).

We reported higher feather nitrogen stable isotope ratios in males compared to female glaucous gulls (Figure 5, Appendix E). Glaucous gulls are sexually dimorphic (Sagerup et al., 2002) with males being larger than females, and the observed sex differences in $\delta^{15}N$ might be a result of males capturing larger and/or higher trophic preys than females (Verreault et al., 2006a). This finding corresponds well with the observed higher levels of contaminants detected in males and supports the assumption of sex-specific diet specialization.

BCI was also a strong predictor of ∑OCPs and ∑PCBs as plasma concentrations were higher in lean compared to fatter individuals (Table 2). A similar finding was reported by Helberg et al. (2005) in great black-backed gull from northern Norway where negative associations were found between body condition and OHCs in female birds. Several other studies have reported relationships between BCI and contaminant burdens. Sagerup et al. (2000) found a negative correlation between BCI and several OCs in glaucous gulls. Henriksen et al. (1998) found a doubling of OHCs with a 100g decrease in body mass in black-legged kittiwakes *Rissa tridactyl*. However, a previous study on glaucous gulls from Bjørnøya indicated relatively stable PCB-153 levels despite fluctuations in body mass (Bustnes et al., 2001b). An association

between ∑PFASs and BCI were not found in this study. Aas et al. (2014) found that concentrations of PFOS and other PFAS compounds were not affected by body condition in any of the analyzed tissues in arctic fox. Similar findings were reported in studies of sea otters *Enhydra lutris nereis* (Kannan et al., 2006) and harbor seals *Phoca vitulina* (Van de Vijver et al., 2003). The discrepancies reported on the influence of body condition on pollutant loads highlight the importance of assessing the annual condition of animals in time trend analyzes and other studies on OHCs.

4.3 Considerations about the study design

The aim of this study was to estimate body burdens of OHCs in breeding populations of glaucous gull from Bjørnøya and Kongsfjorden and to test the hypothesis that Bjørnøya occupies a higher trophic level. It is challenging to highlight a relationship between contaminant levels and dietary ecology inferred from two different tissues reflecting different time periods. Blood stable isotopes during the breeding season reflect the dietary ecology of glaucous gulls when they have access to seabird eggs and chicks and might therefore exhibit a higher trophic level compared to the non-breeding season (Bustnes et al., 2000; Sagerup et al., 2002). Blood stable isotope ratios of carbon and nitrogen were initially planned for the present study but could not be included at the given time for this master thesis due to time constraints. However, preliminary results showed that blood stable isotopes from sampling year 2017 did not correlate with feather stable isotopes (r: -0.03 – (-0.27), $p > 0.05$), indicating that 1) the birds may have variations in foraging strategy during the breeding and non-breeding season and 2) feather stable isotopes might not be a good proxy for feeding ecology during the breeding season. The mean relative trophic positions in blood and feathers from Bjørnøya in 2017 were similar during the breeding and non-breeding seasons (blood $\delta^{15}N$: 15.3 ± 0.3‰, feather $\delta^{15}N$: 15.9 ± 1.0‰), while Kongsfjorden exhibited somewhat lower $\delta^{15}N$ during the breeding season compared to the non-breeding season (blood $\delta^{15}N$: 13.8 ± 0.7‰, feather $\delta^{15}N$: 15.1 ± 0.8‰). Further investigation of blood stable isotopes would be needed to determine diet specialization across populations during the breeding season.

According to SEATRACK (interactive map application, www.seapop.no), both glaucous gull populations migrate to the same areas in the Barents Sea, northern parts of Norway and Iceland during the non-breeding season. This might indicate that the observed contrasting contaminant

burdens may rather be related to the conditions at their breeding area. Leat et al. (2019) concluded that the large-scale spatial variation of OHCs in great skuas are most likely dominated by the local diet and trophic position rather than wintering area and long-range transport of contaminants. Elevated concentrations were also found in the colony of great skuas breeding in Bjørnøya compared to the other colonies in this study. As the diet of great skua and the glaucous gulls overlap, they might similarly accumulate contaminants. Hence, further studies on the diet of glaucous gulls may give a better understanding of the differences between Bjørnøya and Kongsfjorden. The Kongsfjorden breeding population is located near large colonies of common eider ducks *Somateria mollissima* and barnacle geese *Branta leucopsis* suggesting that the diet of glaucous gulls may be influenced by the eggs and chicks from these benthic and herbivore/grazing species (Strøm, 2006; Wold et al., 2011), whereas in Bjørnøya large colonies of pelagic black-legged kittiwakes and brünnichs guillemot *Uria lomvia* are found (Bustnes et al., 2000). The trophic level of their preys (benthic vs pelagic) could potentially explain the contrasting contaminant levels found between the glaucous gull populations.

To determine the dietary ecology of the glaucous gull, and to be able to compare it across populations, it would be advantageous to sample prey species and compare their isotopic signatures with those from glaucous gulls as has been previously done in other studies (Ceia et al., 2014; Ehrich et al., 2014; Leal et al., 2017). Variations in isotope values at the base of the food web translate to all trophic levels and might be incorrectly attributed to changes in diet or climate when only measuring the isotopic signature in the organism of interest. The glaucous gulls were sampled at two different locations and are opportunistic feeders. Accounting for baseline values is therefore particularly important. Failure to account for differences in baseline can result in erroneous interpretations and a reduced signal to noise ratio, as a consumer's trophic level depends on the proportion of the diet each source constitutes (Casey and Post, 2011). Further research is therefore required to determine individual diet specialization and baseline isotopic values in the monitoring area, as this would give a better understanding of how feeding ecology might influence OHC concentrations. It would also be beneficial to use tissues reflecting the same time period when investigating relationships among variables dependent on the sampling time.

5 Conclusion and future studies

In this study, a significant difference in levels of OHCs were found between two populations of glaucous gulls breeding in Bjørnøya and Kongsfjorden. Bjørnøya gulls exhibited 5.3 times higher concentrations of OHCs (sampling year 2015 to 2019) compared to gulls from Kongsfjorden.

The total contaminant pattern in Kongsfjorden was dominated by the chlorinated PCBs, while Bjørnøya was dominated by PCBs and the fluorinated PFASs. PFAS concentrations in Bjørnøya were higher than what has previously been reported in glaucous gulls, demonstrating the importance of continued monitoring of these more recent anthropogenic compounds. Significant temporal variations were found for OCPs in Kongsfjorden and PFASs in Bjørnøya, reflecting annual diet variability.

There was no difference in relative trophic position between the colonies breeding in Bjørnøya and Kongsfjorden. Annual variations in $\delta^{15}N$ were found for glaucous gulls breeding in Kongsfjorden indicating changes in foraging strategy or food availability. No associations between $\delta^{15}N$ and OHC concentrations were found when accounting for the biological parameters sex and BCI. However, $\delta^{13}C$ was an important driver of PFASs concentrations, indicating that foraging habitat affected contaminant concentrations in the glaucous gulls in this study. Our results should be interpreted with caution as the feather stable isotope signatures and blood contaminant levels reflect different time periods.

Further studies should include more predictor variables explaining variations in OHCs when investigating concentration differences between populations. For instance, clutch size and breeding effort would provide additional information on inter-colony differences as reproductive performance in birds varies in relation to prey availability and might be a good indication of individual fitness (Pierotti and Annett, 1990; Sydeman et al., 2001). Contaminant trends may also be more difficult to predict in the future because of the ongoing climate change which might impact ecology, biology and the distribution of contaminants in the Arctic (Mckinney et al., 2015). Increasing temperatures lead to changes in the Arctic ecosystem because of decreased sea ice cover, less snow and altered nutrient availability. This could lead to redistributed ice stored contaminants and changes in food webs, migration patterns, body condition and reproduction (Mckinney et al., 2015; Dietz et al., 2019). Finally,

the rapidly changing Arctic ecosystem due to climate change might add additional stress to species already living in challenging environments, therefore these multiple stressors should be accounted for.

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7 Appendix

7.1 Appendix A: Biometric measurements

Table A1: Mean ± standard deviation (SD), median, range and number (N) of morphological measurements and lipids (%) in male glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. BCI are scaled values for each sex locations combined, when not separating for location and sampling year the mean ± SD is 0 ± 1. The asterisks (*) indicate significant differences between locations according to t-test (* p< 0.05).

Males									
		Bjørnøya				**Kongsfjorden**			
	Year	N	Mean ± SD	Median	Range	N	Mean ± SD	Median	Range
Lipid (%)	2015	13	0.97 ± 0.30	0.98	0.20 - 1.44	4	0.93 ± 0.18	0.87	0.80 - 1.19
	2016	17	1.43 ± 1.48	1.15	0.61 - 7.12	4	0.99 ± 0.14	1.01	0.80 - 1.13
	2017	8	0.53 ± 0.16	0.51	0.35 - 0.77	5	0.81 ± 0.20	0.88	0.46 - 0.96
	2018	3	0.65 ± 0.11	0.62	0.55 - 0.77	3	1.39 ± 0.16	1.45	1.21 - 1.50
	2019	5	1.38 ± 0.18	1.43	1.10 - 1.59	7	1.30 ± 0.19	1.31	1.04 - 1.55
Body weight (g)	2015	13	1795 ± 92	1775	1635 - 1965	5	1708 ± 144	1730	1530 - 1850
	2016	16	1730 ± 112	1730	1490 - 1910	4	1728 ± 146	1700	1580 - 1930
	2017	8	1780 ± 110	1780	1650 - 1920	5	1758 ± 111	1770	1630 - 1880
	2018	3	1770 ± 76	1770	1720 - 1870	3	1773 ± 65	1770	1710 - 1840
	2019	5	1750 ± 150	1750	1520 - 1880	7	1730 ± 128	1700	1590 - 1930
Total head length (mm)	2015	13	149.4 ± 4.3	147.5	142.7 - 156.2	5	151.4 ± 3.4	152.0	146.0 - 155.0
	2016	17	151.6 ± 3.8	152.9	143.6 - 157.0	4	150.2 ± 4.9	150.5	144.0 - 156.0
	2017	8	154.3 ± 3.1	153.9	150.1 - 159.0	5	151.2 ± 2.5	152.0	147.0 - 153.0
	2018	3	157.0 ± 3.1	157.8	153.6 - 159.7	3	150.0 ± 0.0	150.0	150.0 - 150.0
	2019	4	158.6 ± 3.0	159.8	154.3 - 160.7	7	151.9 ± 2.1	152.0	149.0 - 155.5
Tarsus length (mm)	2015	13	76.2 ± 3.2	76.4	71.1 - 81.5	5	72.8 ± 4.6	74.7	66.7 - 78.0
	2016	17	75.2 ± 2.8	75.4	71.0 - 81.5	4	76.6 ± 3.5	77.2	71.8 - 80.0
	2017	8	76.3 ± 2.5	76.1	73.4 - 81.5	5	75.9 ± 2.6	75.0	73.2 - 79.4
	2018	3	77.3 ± 3.9	78.3	73.0 - 80.5	3	76.0 ± 3.0	75.3	73.4 - 79.3
	2019	5	76.5 ± 4.4	74.9	72.4 - 81.6	7	74.8 ± 2.8	75.6	70.0 - 78.3
Bill lenght (mm) *	2015	13	64.1 ± 3.5	65.0	57.8 - 70.5	5	62.2 ± 1.4	62.6	60.2 - 64.0
	2016	17	64.2 ± 3.3	64.1	57.9 - 70.2	4	65.2 ± 2.9	65.2	61.9 - 68.3
	2017	8	66.5 ± 1.9	66.6	63.7 - 69.1	5	65.2 ± 3.6	66.1	59.6 - 69.1
	2018	3	67.4 ± 2.3	66.5	65.6 - 70.0	3	62.8 ± 0.4	62.6	62.5 - 63.2
	2019	5	66.5 ± 3.1	66.1	62.0 - 69.7	7	62.6 ± 0.8	62.9	61.4 - 63.6
Gonis height (mm)	2015	13	23.0 ± 1.1	22.8	21.2 - 24.8	5	22.8 ± 0.8	23.0	21.5 - 23.6
	2016	17	23.0 ± 0.8	23.2	21.5 - 24.5	4	22.6 ± 0.5	22.7	21.9 - 23.1
	2017	8	23.5 ± 1.1	23.5	21.8 - 25.3	5	22.3 ± 0.3	22.4	21.9 - 22.5
	2018	3	23.9 ± 0.8	24.1	23.1 - 24.6	3	22.5 ± 0.9	22.5	21.6 - 23.4
	2019	5	23.7 ± 1.1	24.0	22.2 - 25.1	7	24.3 ± 2.6	23.1	22.6 - 29.9
Wing length (mm)	2015	13	486.4 ± 7.2	487.0	474.0 - 498.0	5	484.4 ± 6.2	484.0	477.0 - 492.0

	2016	17	483.8 ± 6.1	485.0	474.0 - 495.0	4	486.8 ± 18.0	490.0	462.0 - 505.0
	2017	8	485.1 ± 6.8	485.0	478.0 - 495.0	5	471.0 ± 5.0	472.0	463.0 - 476.0
	2018	3	491.0 ± 4.6	492.0	486.0 - 495.0	3	484.7 ± 10.3	482.0	476.0 - 496.0
	2019	5	484.6 ± 8.2	282.0	472.0 - 494.0	7	484.0 ± 5.9	485.0	473.0 - 491.0
BCI	2015	13	0.46 ± 0.67	0.55	-0.58 - 1.56	5	-0.41 ± 1.30	-0.34	-1.93 - 0.96
	2016	16	-0.03 ± 1.04	-0.03	-2.30 - 1.74	4	-0.13 ± 1.08	-0.63	-0.74 - 1.49
	2017	8	-0.06 ± 0.96	0.07	-1.45 - 1.12	5	0.50 ± 1.10	0.54	-0.71 - 1.73
	2018	3	-0.39 ± 0.53	-0.68	-0.72 - 0.23	3	0.43 ± 0.92	0.35	-0.44 - 1.38
	2019	4	-0.64 ± 1.14	-0.31	-2.17 - 0.24	7	-0.55 ± 0.95	-0.61	-1.54 - 1.37

Table A2: Mean ± standard deviation (SD), median, range and number (N) of morphological measurements in female glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. BCI are scaled values for each sex locations combined, when not separating for location and sampling year the mean ± SD is 0 ± 1. The asterisks (*) indicate significant differences between locations according to t-test (* $p < 0.05$, ** $p < 0.01$).

Females

		Bjørnøya				**Kongsfjorden**			
	Year	N	Mean ± SD	Median	Range	N	Mean ± SD	Median	Range
Lipid (%)	2015	13	1.01 ± 0.15	1.07	0.71 - 1.20	10	0.80 ± 0.14	0.85	0.48 - 0.92
	2016	15	1.11 ± 0.63	1.03	0.30 - 3.02	15	0.96 ± 0.22	0.93	0.66 - 1.51
	2017	8	0.58 ± 0.23	0.62	0.11 - 0.87	16	0.78 ± 0.13	0.80	0.51 - 1.10
	2018	4	0.37 ± 0.19	0.39	0.14 - 0.55	10	1.41 ± 0.20	1.40	1.19 - 1.82
	2019	9	1.35 ± 0.18	1.38	1.06 - 1.55	7	1.21 ± 0.07	1.20	1.09 - 1.29
Body weight (g)	2015	13	1440 ± 100	1425	1265 - 1625	12	1400 ± 48	1385	1330 - 1480
	2016	15	1482 ± 135	1490	1300 - 1850	15	1360 ± 95	1350	1150 - 1500
	2017	8	1434 ± 88	1440	1260 - 1550	16	1447 ± 95	1455	1290 - 1655
	2018	4	1409 ± 70	1395	1340 - 1505	10	1497 ± 135	1485	1290 - 1770
	2019	9	1382 ± 35	1380	1350 - 1460	7	1351 ± 74	1360	1270 - 1490
Total head length (mm) **	2015	13	136.2 ± 3.6	136.2	129.4 - 142.3	12	137.1 ± 1.7	137.5	134.0 - 139.0
	2016	14	137.9 ± 3.8	136.8	130.3 - 142.6	15	136.5 ± 3.5	137.0	129.0 - 141.0
	2017	8	141.2 ± 4.2	140.9	134.7 - 148.4	16	136.6 ± 3.4	136.5	131.1 - 143.0
	2018	4	143.7 ± 2.7	143.7	140.9 - 146.8	10	137.6 ± 3.1	138.0	132.0 - 143.0
	2019	9	140.9 ± 2.3	140.9	138.6 - 146.2	7	136.6 ± 2.7	137.0	133.5 - 141.0
Tarsus length (mm)	2015	13	70.7 ± 2.4	70.8	65.8 - 75.2	10	66.7 ± 5.3	68.0	57.1 - 73.1
	2016	15	68.6 ± 2.6	67.5	66.1 - 74.2	15	70.4 ± 2.1	70.3	67.8 - 74.6
	2017	8	70.8 ± 2.3	71.1	65.8 - 73.1	16	70.1 ± 1.9	69.5	67.0 - 74.7
	2018	4	70.6 ± 0.5	70.7	70.0 - 71.2	10	72.4 ± 2.9	71.8	68.7 - 77.6
	2019	9	70.3 ± 2.2	69.7	67.5 - 73.8	7	69.1 ± 2.9	69.7	65.1 - 73.5
Bill lenght (mm)	2015	13	57.3 ± 1.7	57.2	54.7 - 60.0	12	56.7 ± 1.8	56.9	53.5 - 60.3
	2016	15	57.0 ± 1.9	57.4	52.86 - 59.5	15	57.2 ± 2.2	57.5	51.1 - 59.6
	2017	8	57.8 ± 2.2	58.6	53.9 - 60.3	16	58.0 ± 2.2	58.3	53.6 - 61.8
	2018	4	59.1 ± 1.1	58.9	58.2 - 60.5	10	58.5 ± 1.7	59.0	55.0 - 60.6

	2019	9	57.5 ± 1.5	57.9	55.3 - 59.5	7	57.0 ± 1.8	57.0	54.5 - 59.3
Gonis height (mm) *	2015	13	20.9 ± 1.2	20.9	19.4 - 24.1	12	20.5 ± 0.7	20.6	19.4 - 21.8
	2016	15	20.7 ± 0.4	20.8	19.6 - 21.2	15	20.8 ± 0.7	20.6	19.8 - 22.2
	2017	8	21.3 ± 0.5	21.4	20.4 - 21.8	16	20.5 ± 0.9	20.3	19.0 - 21.9
	2018	4	21.7 ± 0.5	21.8	21.0 - 22.0	10	21.3 ± 1.0	21.3	19.8 - 22.6
	2019	9	21.3 ± 0.8	21.4	19.9 - 22.7	7	20.2 ± 1.0	20.3	18.9 - 21.6
Wing length (mm)	2015	13	459.4 ± 7.8	459.0	442.0 - 474.0	12	462.8 ± 13.6	462.0	445.0 - 485.0
	2016	15	457.5 ± 7.0	458.0	446.0 - 474.0	15	456.3 ± 8.3	457.0	442.0 - 473.0
	2017	8	459.9 ± 3.8	459.5	454.0 - 466.0	16	458.2 ± 10.4	454.0	444.0 - 480.0
	2018	4	463.5 ± 4.4	461.5	461.0 - 470.0	10	466.3 ± 7.5	463.5	458.0 - 480.0
	2019	9	458.1 ± 7.0	458.0	445.0 - 471.0	7	456.3 ± 3.6	456.0	450.0 - 462.0
BCI	2015	13	0.24 ± 0.77	-0.01	-1.90 - 1.63	12	-0.20 ± 0.38	-0.22	-0.61 - 0.40
	2016	15	0.66 ± 1.45	0.64	-1.37 – 4.80	15	-0.53 ± 0.75	-0.43	-2.14 - 0.50
	2017	8	-0.22 ± 0.73	-0.22	-1.68 - 0.66	16	0.42 ± 0.86	0.33	-0.67 - 2.29
	2018	4	-0.80 ± 0.66	-0.79	-1.61 - 0.01	10	0.53 ± 1.32	0.25	-1.05 - 3.64
	2019	9	-0.72 ± 0.42	-0.84	-1.17 - 0.08	7	-0.49 ± 0.65	-0.61	-1.21 - 0.41

7.2 Appendix B: Limits of detection for OHCs

Table B: Limits of detection (LOD, pg/g ww) and detection rate (DR, %) for OHCs analyzed in plasma of glaucous gulls breeding in Bjørnøya and Kongsfjorden, Svalbard during the breeding season of 2015 until 2019.

LOD (pg/g ww)

	Bjørnøya		**Kongsfjorden**				
Compound	DR (%)	2015-2019	DR (%)	2015	2016-2017	2018	2019
HCB	100	0.12	100	0.31	407.54	50.89	50.90
α-HCH	0	49.74	0	46.15	48.53	62.34	62.34
β-HCH	100	66.42	99	68.00	64.80	93.54	93.54
γ-HCH	3	21.97	0	13.99	29.68	58.86	58.86
Heptachlor	-	-	0	46.45	-	-	-
c-chlordane	31	17.74	20	4.77	17.31	3.72	3.72
t-chlordane	0	9.51	49	3.20	9.28	10.09	10.09
oxy-chlordane	100	11.61	100	25.16	283.84	44.20	44.20
c-nonachlor	100	2.981	100	2.49	2.91	20.12	20.12
t-nonachlor	100	8.94	100	3.76	8.72	10.28	10.28
mirex	100	22.69	100	24.30	22.13	25.97	25.97
o,p'-DDT	-	-	15	238.37	42.93	36.22	36.22
p,p'-DDT	53	44.00	22	337.21	43.90	38.71	38.71
o,p'-DDD	-	-	0	76.18	41.95	13.30	13.30
p,p'-DDD	-	-	0	68.15	42.91	14.22	14.22
o,p'-DDE	-	-	1	46.80	42.91	8.71	8.71
p,p'-DDE	100	14.56	96	65.48	123.9	230.49	230.49

PCB-28	99	31.73	100	16.15	95.21	34.81	34.81
PCB-52	75	21.06	74	21.24	20.55	33.00	33.00
PCB-99	100	18.38	100	16.71	17.93	45.94	45.94
PCB-101	60	24.08	57	19.41	23.49	52.69	52.69
PCB-105	100	28.11	100	19.57	27.42	50.10	50.10
PCB-118	100	21.18	100	18.49	20.66	44.68	44.68
PCB-138	100	20.83	100	60.51	20.32	43.34	43.34
PCB-153	100	17.63	100	24.14	122.39	37.85	37.85
PCB-180	100	75.18	100	55.96	73.34	129.00	129.00
PCB-183	100	59.69	99	43.81	58.24	106.22	106.22
PCB-187	100	73.25	100	51.09	71.46	123.78	123.78
PCB-194	100	10.23	100	208.65	97.56	57.38	57.38
PFOSA	0	20.00	0	100.00	24.39	20.00	24.39
PFBS	0	50.00	0	37.48	48.78	50.00	48.78
PFPS	0	50.00	0	-	24.39	50.00	24.39
PFHxS	99	20.00	97	10.00	55.12	20.00	24.39
PFHpS	98	20.00	33	-	24.39	20.00	122.8
brPFOS	100	135.45	75	246.38	24.39	135.45	92.08
PFOS	100	794.30	100	153.40	24.39	794.3	1212.09
PFNS	68	50.00	0	-	24.39	50.00	24.39
PFDcS	97	75.00	23	153.40	48.78	75.00	48.78
PFBA	0	50.00	0	-	117.07	50.00	117.07
PFPA	3	111.67	10	-	108.95	50.00	108.95
PFHxA	0	25.00	0	6.77	24.39	25.00	66.4
PFHpA	29	42.43	0	6.77	41.39	25.00	69.84
PFOA	99	130.30	84	12.83	127.12	130.30	166.22
PFNA	100	50.00	100	14.49	0.00	50.00	19.51
PFDcA	100	29.88	100	-	29.15	29.88	52.59
PFUnA	100	57.64	99	25.58	56.24	57.64	34.15
PFDoA	100	43.31	97	20.08	42.26	43.31	45.58
PFTriA	100	82.95	100	14.87	44.78	82.95	48.78
PFTeA	100	50.00	77	10.16	48.78	50.00	84.05
6:2FTS	0	50.00	0	-	24.39	50.00	24.39
8:2 FTS	57	50.00	0	-	-	50.00	-

7.3 Appendix C: OHC concentrations

Table C: Mean ± standard deviation (SD), median and range of concentrations (ng/g *ww*) of OHCs measured in plasma from glaucous gulls breeding in Bjørnøya (n = 95) and Kongsfjorden (n = 84) during the breeding seasons of 2015 until 2019. The sums of OCPs, PCBs, PFSAs, PFCAs, PFASs and total OHCs are included and denoted with Σ.

		Bjørnøya				**Kongsfjorden**			
	Year	N	Mean ± SD	Median	Range	N	Mean ± SD	Median	Range
HCB	2015	26	13.67 ± 2.88	12.82	7.75 - 19.56	14	4.97 ± 1.10	4.78	3.70 - 7.58
	2016	32	13.39 ± 1.50	13.65	8.22 - 15.70	19	9.28 ± 3.88	8.82	2.69 - 19.72
	2017	16	16.21 ± 7.69	13.9	11.39 - 42.02	21	7.60 ±3.66	6.55	3.60 - 15.83
	2018	7	15.76 ± 7.09	13.31	11.87 - 31.75	13	3.20 ± 0.47	3.22	2.61 - 4.01
	2019	14	13.09 ± 0.78	13.40	11.54 - 13.96	14	3.39 ± 0.42	3.45	2.54 - 3.98
β-HCH	2015	26	1.13 ± 0.62	0.97	0.35 - 3.22	14	0.46 ± 0.29	0.33	0.22 - 1.11
	2016	32	1.24 ± 0.67	1.02	0.40 - 2.96	18	0.53 ± 0.29	0.43	0.25 - 1.27
	2017	16	1.38 ± 0.87	1.20	0.40 - 3.41	21	0.40 ± 0.28	0.33	0.13 - 1.29
	2018	7	1.52 ± 0.72	1.35	0.72 - 2.80	13	0.34 ± 0.17	0.38	0.14 - 0.67
	2019	14	1.56 ± 0.70	1.60	0.43 - 2.79	14	0.44 ± 0.17	0.43	0.17 - 0.84
Oxy-CD	2015	26	16.86 ± 9.65	14.66	5.82 - 49.28	14	10.00 ± 9.49	5.79	3.03 - 36.78
	2016	32	16.25 ± 8.92	13.43	3.48 - 41.55	19	11.15 ±14.49	6.37	1.13 - 61.50
	2017	16	20.47 ± 15.57	16.58	4.66 - 65.15	21	6.13 ± 5.62	3.54	1.54 - 23.67
	2018	7	18.24 ± 11.23	12.90	7.30 - 35.60	13	4.89 ± 2.84	3.56	1.72 - 10.23
	2019	14	20.80 ± 9.20	20.52	6.40 - 36.60	14	11.79 ± 10.32	6.25	2.99 - 30.30
t-NC	2015	26	1.01 ± 0.60	0.83	0.22 - 2.36	14	0.68 ± 0.28	0.66	0.29 - 1.24
	2016	32	0.97 ± 0.54	0.86	0.11 - 2.14	19	1.01 ± 1.27	0.66	0.36 - 6.08
	2017	16	1.29 ± 1.03	0.99	0.25 - 4.04	21	0.65 ±0.43	0.63	0.08 - 2.19
	2018	7	1.19 ± 1.45	0.41	0.14 - 4.23	13	0.69 ± 0.37	0.49	0.34 - 1.53
	2019	14	1.07 ± 0.84	0.85	0.08 - 3.36	14	0.66 ± 0.54	0.60	0.19 - 2.23
c-NC	2015	26	1.72 ± 0.86	1.73	0.56 - 3.72	14	0.67 ± 0.30	0.74	0.13 - 1.15
	2016	32	1.57 ± 0.70	1.39	0.67 - 3.12	19	0.62 ± 0.30	0.55	0.16 - 0.36
	2017	16	1.92 ± 1.16	1.77	0.92 - 5.77	21	0.51 ± 0.21	0.49	0.18 - 0.89
	2018	7	1.99 ± 0.63	1.84	1.00 - 2.90	13	0.75 ± 0.21	0.80	0.37 - 1.08
	2019	14	1.94 ± 0.96	1.87	0.43 - 3.91	14	0.76 ± 0.48	0.75	0.16 - 1.83
t_CD	2015	-	-	-	-		-	-	-
	2016	-	-	-	-	19	0.43 ± 0.21	0.41	0.15 - 1.08
	2017	-	-	-	-	21	0.35 ± 0.19	0.31	0.10 - 0.91
	2018	-	-	-	-		-	-	-
	2019	-	-	-	-		-	-	-
Mirex	2015	26	10.06 ± 5.12	9.01	4.59 - 26.32	14	4.45 ± 3.99	2.75	1.13 - 15.06
	2016	32	10.47 ± 5.76	8.34	3.05 - 30.18	19	4.21 ± 3.76	3.07	0.39 - 17.08
	2017	16	14.39 ± 8.52	13.68	4.38 - 36.81	21	3.38 ± 3.71	1.95	0.62 - 12.84
	2018	7	14.50 ± 7.91	11.64	5.96 - 25.88	13	3.35 ± 2.02	3.43	0.87 - 6.94
	2019	14	15.06 ± 5.97	17.33	4.69 - 23.00	14	5.29 ± 3.17	4.14	1.87 - 10.92

p,p'-DDT	2015	16	0.19 ± 0.11	0.16	0.08 - 0.51	-	-	-	-
	2016	14	0.30 ± 0.15	0.27	0.13 - 0.68	-	-	-	-
	2017	6	0.25 ± 0.23	0.16	0.11 - 0.70	-	-	-	-
	2018	5	0.13 ± 0.04	0.12	0.09 - 0.20	-	-	-	-
	2019	9	0.29 ± 0.13	0.25	0.08 - 0.47	-	-	-	-
p,p'-DDE	2015	26	131.39 ± 51.18	121.26	59.07 - 247.29	13	49.43 ± 43.65	33.47	13.37 - 151.31
	2016	32	128.72 ± 55.09	118.77	37.36 - 247.94	18	6.93 ± 7.13	4.84	1.05 - 31.15
	2017	16	170.76 ± 100.09	158.14	46.78 - 432.17	20	4.77 ± 4.20	3.04	1.23 - 15.49
	2018	7	181.31 ± 102.72	143.00	70.92 - 320.50	13	35.24 ± 21.98	31.03	9.15 - 72.25
	2019	14	192.28 ± 91.07	197.20	58.79 - 317.38	14	65.18 ± 51.29	44.84	15.13 - 194.91
∑OCPs	2015	26	175.9 ± 66.05	162.80	85.60 - 344.30	14	67.11 ± 57.66	51.29	11.46 - 196.69
	2016	32	172.73 ± 69.40	161.57	55.33 - 312.98	19	33.77 ± 28.04	26.62	6.18 - 118.06
	2017	16	226.51 ± 124.09	215.88	69.75 - 554.11	21	23.57 ± 17.20	17.05	8.25 - 70.25
	2018	7	234.60 ± 121.28	199.10	100.20 - 393.40	13	48.46 ± 27.43	43.98	15.25 - 94.37
	2019	14	245.98 ± 106.12	247.84	83.27 - 386.91	14	87.51 ± 63.64	58.87	23.09 - 234.18
PCB 28	2015	26	1.61 ± 1.18	1.31	0.42 - 6.56	14	0.53 ± 0.26	0.45	0.28 - 1.22
	2016	31	1.72 ± 0.97	1.42	0.42 - 5.21	19	0.35 ± 0.12	0.35	0.18 - 0.72
	2017	16	2.16 ± 1.55	1.73	0.68 - 5.90	21	0.29 ± 0.14	0.27	0.12 - 0.61
	2018	7	1.89 ± 0.91	1.93	0.89 - 3.54	13	0.19 ± 0.09	0.20	0.06 - 0.32
	2019	14	2.43 ± 1.16	2.67	0.75 - 3.89	14	0.28 ± 0.12	0.25	0.11 - 0.53
PCB 52	2015	20	0.33 ± 0.20	0.27	0.10 - 0.79	13	0.23 ± 0.16	0.20	0.05 - 0.61
	2016	16	0.31 ± 0.20	0.27	0.08 - 0.75	16	0.29 ± 0.19	0.27	0.07 - 0.81
	2017	10	0.42 ± 0.27	0.32	0.23 - 1.13	18	0.27 ± 0.17	0.25	0.05 - 0.55
	2018	7	0.31 ± 0.07	0.32	0.19 - 0.40	8	0.28 ± 0.16	0.20	0.07 - 0.50
	2019	12	0.40 ± 0.23	0.37	0.08 - 0.82	5	0.17 ± 0.10	0.13	0.07 - 0.29
PCB 99	2015	26	19.23 ± 16.73	14.12	6.38 - 89.86	14	7.26 ± 6.14	4.97	2.45 - 21.18
	2016	32	18.17 ± 13.50	13.25	3.9 - 76.10	19	9.12 ± 8.59	6.44	1.15 - 37.93
	2017	16	19.97 ± 16.36	15.87	5.01 - 62.84	21	6.16 ± 5.18	4.05	1.47 - 17.61
	2018	7	17.57 ± 9.15	14.99	7.83 - 31.07	13	5.37 ± 3.27	5.03	1.64 - 11.94
	2019	14	20.23 ± 9.35	18.13	6.45 - 35.31	14	10.27 ± 8.21	6.92	2.61 - 32.45
PCB101	2015	25	1.43 ± 0.73	1.19	0.52 - 3.16	11	0.56 ± 0.41	0.54	0.08 - 1.55
	2016	16	1.01 ± 0.52	0.86	0.27 - 1.74	12	1.01 ± 0.53	0.86	0.31 - 1.96
	2017	6	0.91 ± 0.53	0.78	0.49 - 1.90	13	1.04 ± 0.69	0.81	0.06 - 2.51
	2018	3	0.53 ± 0.15	0.55	0.37 - 0.67	7	0.75 ± 0.51	0.72	0.18 - 1.51
	2019	7	1.04 ± 0.48	1.04	0.32 - 1.83	3	0.52 ± 0.20	0.43	0.39 - 0.75
PCB 105	2015	26	13.37 ± 7.59	11.75	4.91 - 40.83	14	3.26 ± 2.43	2.34	1.32 - 10.57
	2016	32	11.87 ± 6.04	9.34	3.14 - 31.73	19	3.59 ± 2.61	2.90	0.57 - 12.68
	2017	16	13.70 ± 10.08	10.98	3.56 - 39.97	21	2.59 ± 1.98	2.05	0.83 - 8.13
	2018	7	10.34 ± 4.54	9.55	5.13 - 17.71	13	2.59 ± 1.98	2.05	0.83 - 8.13
	2019	14	11.83 ± 5.81	13.02	3.72 - 19.86	14	3.46 ± 2.10	2.39	1.25 - 6.79
PCB 118	2015	26	35.90 ± 25.64	29.16	13.59 - 138.06	14	12.93 ± 10.86	8.26	4.56 - 44.66
	2016	32	30.71 ± 19.89	23.12	6.57 - 113.54	19	14.25 ± 11.25	10.17	1.70 - 54.08
	2017	16	45.23 ± 35.62	34.50	11.21 - 144.94	21	10.13 ± 8.68	6.31	2.91 - 32.57
	2018	7	38.96 ± 21.68	30.42	15.74 - 71.38	13	9.60 ± 5.62	9.87	3.22 - 21.49
	2019	14	43.05 ± 19.78	44.64	12.54 - 69.84	14	17.18 ± 12.31	11.09	4.75 - 43.15

PCB 138	2015	26	117.39 ± 110.46	88.88	40.95 - 593.03	14	30.26 ± 27.36	18.73	8.31 - 94.75
	2016	32	106.22 ± 85.36	78.03	22.85 - 490.55	19	40.29 ± 35.43	30.37	4.78 - 149.20
	2017	16	156.45 ± 128.86	125.43	35.05 - 499.60	21	28.92 ± 27.72	17.27	7.54 - 91.64
	2018	7	119.05 ± 65.60	92.08	49.12 - 215.99	13	24.77 ± 15.64	25.38	6.33 - 54.40
	2019	14	131.22 ± 64.06	117.18	42.49 - 235.57	14	44.02 ± 34.70	30.68	10.88 - 140.05
PCB 153	2015	26	146.27 ± 149.18	105.51	48.96 - 788.70	14	56.09 ± 54.41	32.87	13.65 - 198.17
	2016	32	142.23 ± 130.59	94.66	27.78 - 751.15	19	61.34 ± 58.80	43.01	5.39 - 237.90
	2017	16	212.59 ± 183.65	158.01	41.39 - 725.15	21	45.20 ± 45.85	24.33	9.45 - 160.30
	2018	7	169.75 ± 113.50	117.07	61.65 - 354.76	13	40.56 ± 27.05	41.34	10.93 - 84.78
	2019	14	177.08 ± 83.88	186.95	46.71 - 318.32	14	72.71 ± 58.44	46.76	15.60 - 187.14
PCB 180	2015	26	74.22 ± 84.19	49.59	23.49 - 446.22	14	32.72 ± 33.31	17.10	6.72 - 121.39
	2016	32	66.55 ± 70.47	46.72	12.87 - 409.96	19	27.50 ± 27.94	18.83	2.28 - 119.83
	2017	16	91.71 ± 80.81	66.82	17.45 - 322.06	21	22.43 ± 25.62	11.01	3.20 - 90.76
	2018	7	81.47 ± 61.28	51.20	24.45 - 181.64	13	19.32 ± 14.12	16.78	4.15 - 42.89
	2019	14	78.23 ± 39.12	82.76	19.15 - 142.58	14	34.02 ± 28.87	20.67	7.04 - 95.58
PCB 183	2015	26	12.56 ± 14.12	8.36	3.84 - 75.38	14	4.53 ± 4.41	2.61	1.08 - 15.93
	2016	32	10.82 ± 11.19	7.19	2.04 - 64.96	18	4.68 ± 4.14	3.39	1.43 - 16.40
	2017	16	19.00 ± 16.45	14.52	3.74 - 65.19	21	3.61 ± 3.93	1.85	0.66 - 12.95
	2018	7	15.65 ± 10.38	12.10	5.56 - 29.96	13	2.97 ± 2.06	3.02	0.69 - 6.51
	2019	14	16.23 ± 7.91	17.43	4.21 - 29.52	14	5.08 ± 3.97	3.36	1.18 - 12.83
PCB 187	2015	26	29.34 ± 24.08	22.44	9.82 - 127.94	14	5.52 ± 5.35	3.82	1.36 - 21.94
	2016	32	25.41 ± 19.29	18.64	5.58 - 109.54	19	6.06 ± 4.61	4.69	0.97 - 20.97
	2017	16	41.81 ± 31.98	34.10	11.34 - 128.51	21	5.25 ± 4.93	3.25	1.26 - 17.11
	2018	7	42.20 ± 23.14	45.40	16.13 - 82.84	13	4.93 ± 3.22	4.29	1.20 - 11.77
	2019	14	42.60 ± 21.34	38.15	13.08 - 76.51	14	6.64 ± 4.46	5.43	2.26 - 20.80
PCB 194	2015	26	10.49 ± 10.06	7.42	3.11 - 53.90	14	4.56 ± 4.71	2.11	0.82 - 15.42
	2016	32	8.88 ± 9.90	5.53	1.74 - 58.28	19	3.29 ± 3.49	2.52	0.25 - 15.72
	2017	16	12.52 ± 10.82	10.16	2.59 - 43.67	21	2.88 ± 3.44	1.48	0.33 - 13.28
	2018	7	13.02 ± 9.06	7.99	4.54 - 26.61	13	2.26 ± 1.76	1.79	0.41 - 5.71
	2019	14	11.89 ± 5.96	13.41	2.84 - 21.24	14	4.05 ± 3.35	2.49	1.11 - 10.96
∑PCBs	2015	26	462.00 ± 441.28	341.5	163.7 - 2363.6	14	158.33 ± 145.99	94.44	41.32 - 536.35
	2016	32	423.23 ± 364.7	302.15	87.03 - 2111.01	19	171.11 ± 152.20	123.73	18.00 - 606.15
	2017	16	615.80 ± 514.23	486.40	133.3 - 2037.8	21	128.33 ± 126.63	67.08	30.85 - 437.31
	2018	7	510.40 ± 313.06	382.50	191.8 - 952.4	13	112.71 ± 73.69	117.50	29.85 - 242.39
	2019	14	535.60 ± 252.77	548.40	152.9 - 953.2	14	197.88 ± 149.90	131.55	47.08 - 489.16
PFHxS	2015	26	1.63 ± 0.56	1.70	0.54 - 2.55	15	1.11 ± 1.73	0.34	0.01 - 5.78
	2016	32	0.96 ± 0.35	0.92	0.42 - 1.93	19	0.49 ± 0.37	0.38	0.12 - 1.75
	2017	16	0.90 ± 0.21	0.89	0.61 - 1.38	17	0.54 ± 0.53	0.35	0.06 - 2.06
	2018	7	1.34 ± 0.48	1.44	0.64 - 2.04	12	0.71 ± 0.38	0.61	0.26 - 1.51
	2019	13	0.81 ± 0.28	0.80	0.34 - 1.17	14	0.72 ± 0.40	0.62	0.41 - 1.83
PFHpS	2015	26	1.75 ± 0.77	1.92	0.30 - 2.95	-	-	-	-
	2016	32	0.90 ± 0.49	0.97	0.03 - 1.68	-	-	-	-
	2017	16	2.01 ± 0.61	2.03	1.17 - 3.09	-	-	-	-
	2018	7	2.52 ± 1.02	2.26	1.17 - 4.06	-	-	-	-
	2019	12	1.12 ± 0.36	1.25	0.65 - 1.88	-	-	-	-

PFOS	2015	26	330.10 ± 152.77	346.82	54.26 - 575.02	15	18.68 ± 29.83	7.94	1.39 - 118.93
	2016	32	206.62 ± 97.69	209.98	28.81 - 372.29	19	7.69 ± 4.06	6.56	3.66 - 21.59
	2017	16	391.20 ± 106.44	383.10	234.30 - 583.80	18	10.57 ± 8.90	7.52	3.54 - 42.31
	2018	7	413.70 ± 148.84	423.20	182.40 - 622.40	12	9.12 ± 3.84	8.99	2.59 - 17.68
	2019	14	217.70 ± 49.61	224.30	134.30 - 289.10	14	9.19 ± 3.86	8.52	4.01 - 15.04
PFDcS	2015	25	0.84 ± 0.46	0.89	0.24 - 2.17	-	-	-	-
	2016	30	0.54 ± 0.27	0.50	0.15 - 1.13	-	-	-	-
	2017	16	0.92 ± 0.29	0.98	0.35 - 1.28	-	-	-	-
	2018	7	0.88 ± 0.40	0.89	0.30 - 1.36	-	-	-	-
	2019	14	0.53 ± 0.23	0.52	0.21 - 1.12	-	-	-	-
PFNS	2015	17	0.76 ± 0.26	0.67	0.48 - 1.45	-	-	-	-
	2016	21	0.56 ± 0.08	0.55	0.47 - 0.73	-	-	-	-
	2017	16	0.65 ± 0.12	0.64	0.48 - 0.92	-	-	-	-
	2018	5	0.65 ± 0.09	0.68	0.54 - 0.74	-	-	-	-
	2019	6	0.57 ± 0.11	0.54	0.49 - 0.80	-	-	-	-
brPFOS	2015	26	52.02 ± 29.30	55.98	7.30 - 102.87	7	7.56 ± 11.83	0.65	0.27 - 30.19
	2016	32	28.79 ± 15.58	29.73	3.73 - 56.20	15	1.27 ± 1.67	0.81	0.32 - 7.10
	2017	16	50.46 ± 17.85	47.54	28.69 - 87.57	10	2.10 ± 2.60	0.88	0.23 - 8.08
	2018	7	50.14 ± 28.39	42.91	13.76 - 101.20	12	1.99 ± 1.02	1.85	0.52 - 3.88
	2019	14	29.46 ± 6.97	29.84	18.67 - 42.33	14	2.05 ± 0.92	1.82	0.85 - 3.87
8:2 FTS	2015	15	0.27 ± 0.17	0.22	0.08 - 0.66	-	-	-	-
	2016	23	0.16 ± 0.11	0.12	0.02 - 0.38	-	-	-	-
	2017	10	0.11 ± 0.06	0.08	0.05 - 0.22	-	-	-	-
	2018	2	0.11 ± 0.03	0.11	0.09 - 0.14	-	-	-	-
	2019	4	0.06 ± 0.02	0.06	0.05 - 0.09	-	-	-	-
∑PFSAs	2015	26	387.00 ± 182.93	408.40	63.50 - 685.30	15	23.32 ± 39.94	8.28	1.44 - 154.90
	2016	32	238.25 ± 113.70	240.26	33.34 - 429.49	19	9.19 ± 5.79	7.36	4.37 - 30.44
	2017	16	446.20 ± 124.50	434.00	265.80 - 668.80	18	12.25 ± 11.28	8.09	3.60 - 52.45
	2018	7	469.10 ± 178.30	471.60	198.40 - 731.30	12	11.76 ± 4.93	11.49	3.53 - 22.76
	2019	14	249.70 ± 56.60	255.20	154.70 - 334.90	14	11.96 ± 5.02	10.89	5.29 - 20.23
PFDoA	2015	26	15.16 ± 5.03	15.32	4.03 - 24.17	13	0.41 ± 0.34	0.33	0.06 - 1.25
	2016	32	12.22 ± 4.67	12.42	2.96 - 20.14	19	0.58 ± 0.25	0.54	0.20 - 1.03
	2017	16	14.81 ± 3.06	15.46	8.48 - 19.15	18	0.96 ± 0.46	0.85	0.32 - 1.87
	2018	7	13.45 ± 3.28	14.90	8.55 - 16.44	12	0.63 ± 0.31	0.53	0.27 - 1.26
	2019	14	9.19 ± 1.72	9.27	6.18 - 11.41	14	0.65 ± 0.22	0.60	0.30 - 1.05
PFTeA	2015	26	3.03 ± 1.24	2.82	0.98 - 7.59	7	0.16 ± 0.06	0.15	0.08 - 0.23
	2016	32	2.60 ± 1.14	2.39	0.89 - 5.73	15	0.33 ± 0.15	0.33	0.08 - 0.67
	2017	16	2.73 ± 0.84	2.91	1.08 - 4.32	12	0.44 ± 0.23	0.33	0.16 - 0.84
	2018	7	2.38 ± 0.66	2.50	1.32 - 3.11	12	0.40 ± 0.23	0.33	0.11 - 0.82
	2019	14	1.87 ± 0.46	1.78	1.11 - 2.85	14	0.35 ± 0.15	0.30	0.17 - 0.62
PFUnA	2015	26	102.10 ± 37.56	110.59	25.63 - 159.25	15	2.30 ± 1.33	1.99	0.68 - 6.20
	2016	32	77.82 ± 31.39	80.01	16.19 - 134.39	19	2.14 ± 0.93	1.04	0.78 - 4.89
	2017	16	100.42 ±19.04	104.91	59.94 - 128.29	18	4.67 ± 2.36	4.00	1.51 - 8.41
	2018	7	93.82 ± 24.28	99.90	59.55 - 116.56	11	2.46 ± 0.75	2.25	1.41 - 4.28
	2019	14	62.68 ± 11.96	61.94	41.75 - 78.68	14	2.06 ± 0.52	2.15	1.26 - 3.13

PFTriA	2015	26	29.84 ± 10.90	27.00	8.24 - 56.10	15	1.35 ± 1.07	1.29	0.08 - 3.55
	2016	32	24.69 ± 10.29	24.41	7.48 - 50.65	19	2.07 ± 0.93	2.14	0.53 - 3.89
	2017	16	27.33 ± 6.68	28.39	13.79 - 39.28	18	2.58 ± 1.07	2.17	1.18 - 5.40
	2018	7	25.09 ± 6.75	26.95	15.34 - 33.10	12	2.27 ± 1.06	1.93	1.05 - 4.02
	2019	14	16.85 ± 3.10	16.46	11.61 - 21.41	14	3.23 ± 1.01	3.37	1.54 - 4.75
PFDcA	2015	26	28.40 ± 11.41	31.22	6.34 - 43.15	15	0.58 ± 0.30	0.54	0.16 - 1.29
	2016	32	18.51 ± 7.70	18.66	3.95 - 32.70	19	0.64 ± 0.28	0.56	0.26 - 1.18
	2017	16	26.07 ± 5.55	25.88	16.04 - 38.97	18	1.02 ± 0.48	0.80	0.33 - 1.77
	2018	7	27.96 ± 8.19	29.99	14.66 - 35.18	12	0.78 ± 0.24	0.75	0.39 - 1.26
	2019	14	16.52 ± 2.90	17.25	11.41 - 20.44	14	0.72 ± 0.23	0.66	0.37 - 1.20
PFOA	2015	26	1.24 ± 0.57	1.33	0.14 - 2.31	8	0.20 ± 0.24	0.10	0.02 - 0.77
	2016	31	0.47 ± 0.24	0.49	0.12 - 1.02	18	0.17 ± 0.12	0.14	0.03 - 0.52
	2017	16	0.39 ± 0.15	0.41	0.19 - 0.64	17	0.12 ± 0.10	0.08	0.02 - 0.33
	2018	7	0.55 ± 0.19	0.46	0.41 - 0.94	11	0.34 ± 0.17	0.26	0.17 - 0.68
	2019	14	0.40 ± 0.19	0.32	0.15 - 0.79	11	0.34 ± 0.11	0.34	0.18 - 0.58
PFNA	2015	26	27.64 ± 11.94	29.12	4.45 - 49.34	15	1.30 ± 0.55	1.31	0.39 - 2.05
	2016	32	13.72 ± 6.37	13.66	3.96 - 29.83	19	1.41 ± 0.67	1.33	0.54 - 3.38
	2017	16	18.50 ± 4.87	17.09	11.83 - 31.57	18	1.30 ± 0.51	1.35	0.48 - 2.45
	2018	7	24.62 ± 10.23	24.47	9.67 - 39.22	12	1.97 ± 0.84	2.06	0.80 - 3.08
	2019	14	13.61 ± 2.81	13.77	8.74 - 17.85	14	1.73 ± 0.56	1.50	1.07 - 2.81
∑PFCAs	2015	26	207.40 ± 74.73	219.44	49.81 - 313.90	15	6.06 ± 2.90	5.23	1.92 - 13.23
	2016	32	150.01 ± 58.89	153.00	37.17 - 241.95	19	7.26 ± 2.64	6.94	3.36 - 11.01
	2017	16	190.20 ± 35.46	197.50	111.90 - 235.0	18	10.94 ± 4.41	9.83	4.20 - 19.44
	2018	7	187.90 ± 51.72	197.90	109.50 - 235.00	12	8.63 ± 3.20	8.12	4.08 - 15.02
	2019	14	121.10 ± 21.08	121.60	86.70 - 150.00	14	9.00 ± 2.27	9.14	5.09 - 13.41
∑PFASs	2015	26	594.40 ± 254.21	633.00	113.30 - 984.30	15	29.37 ± 39.32	16.80	3.36 - 158.00
	2016	32	388.26 ± 166.76	395.20	70.51 - 633.81	19	16.45 ± 7.50	14.33	7.73 - 41. 09
	2017	16	636.50 ± 146.78	625.10	377.80 - 905.60	18	23.18 ± 13.25	20.26	11.26 - 66.07
	2018	7	656.9 ± 226.85	667.10	308.00 - 961.70	12	20.39 ± 7.77	19.61	7.90 - 37.78
	2019	14	370.80 ± 73.67	382.70	242.00 - 461.70	14	20.96 ± 6.72	20.91	10.38 - 31.30
∑OHCs	2015	26	1232.30 ± 609.43	1187.80	433.70 - 3431.60	14	255.95 ± 200.40	201.07	84.74 - 752.36
	2016	32	984.20 ± 482.42	911.40	261.00 - 2918.10	19	221.32 ± 183.17	165.17	31.92 - 726.27
	2017	16	1478.70 ± 667.85	1449.50	597.10 - 3329.90	18	189.68 ± 151.50	137.14	69.96 - 540.00
	2018	7	1402.00 ± 627.28	1187.30	690.60 - 2289.50	12	181.56 ± 104.57	186.02	58.78 - 358.24
	2019	14	1152.40 ± 389.77	1226.40	593.50 - 1706.80	14	306.35 ± 216.25	213.16	80.55 - 750.68

7.4 Appendix D: Correlation matrices

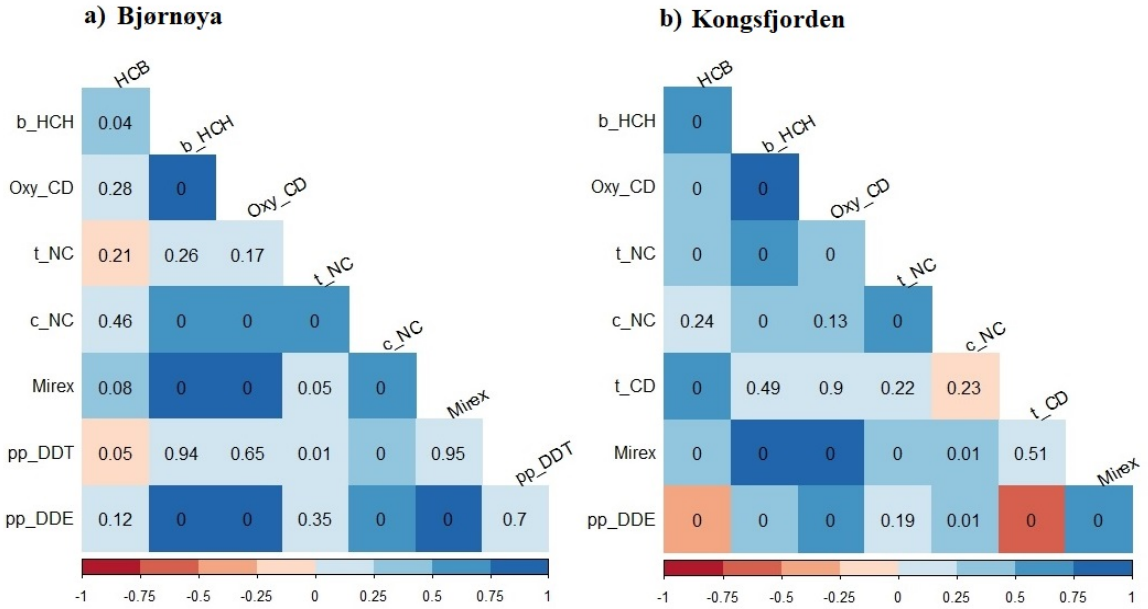

Figure D1: Spearman's ranks correlation matrix for organochlorine pesticides (OCPs) in glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. Colour gradient indicate degree of correlation where increasing distance from 0 (red or blue) means stronger correlation. P-values are presented with numbers, significance level is set at 0.05.

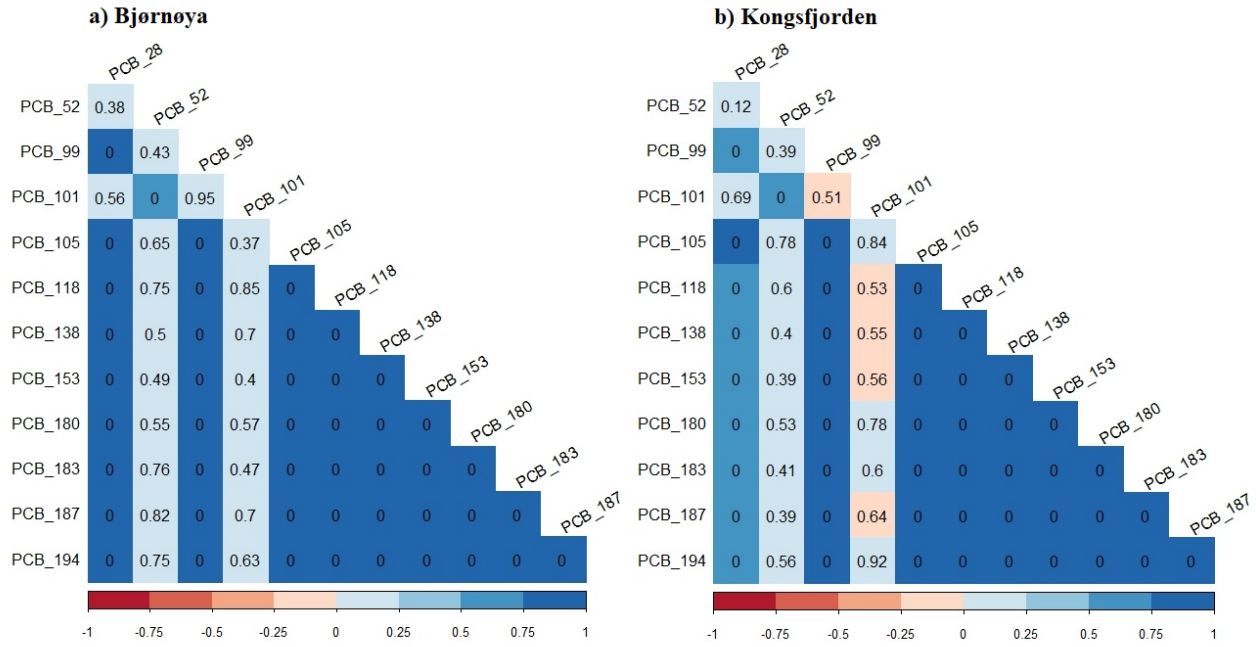

Figure D2: Spearman's ranks correlation matrix for plychlorinated biphenyls (PCBs) in glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. Colour gradient indicate degree of correlation where increasing distance from 0 (red or blue) means stronger correlation. P-values are presented with numbers, significance level is set at 0.05.

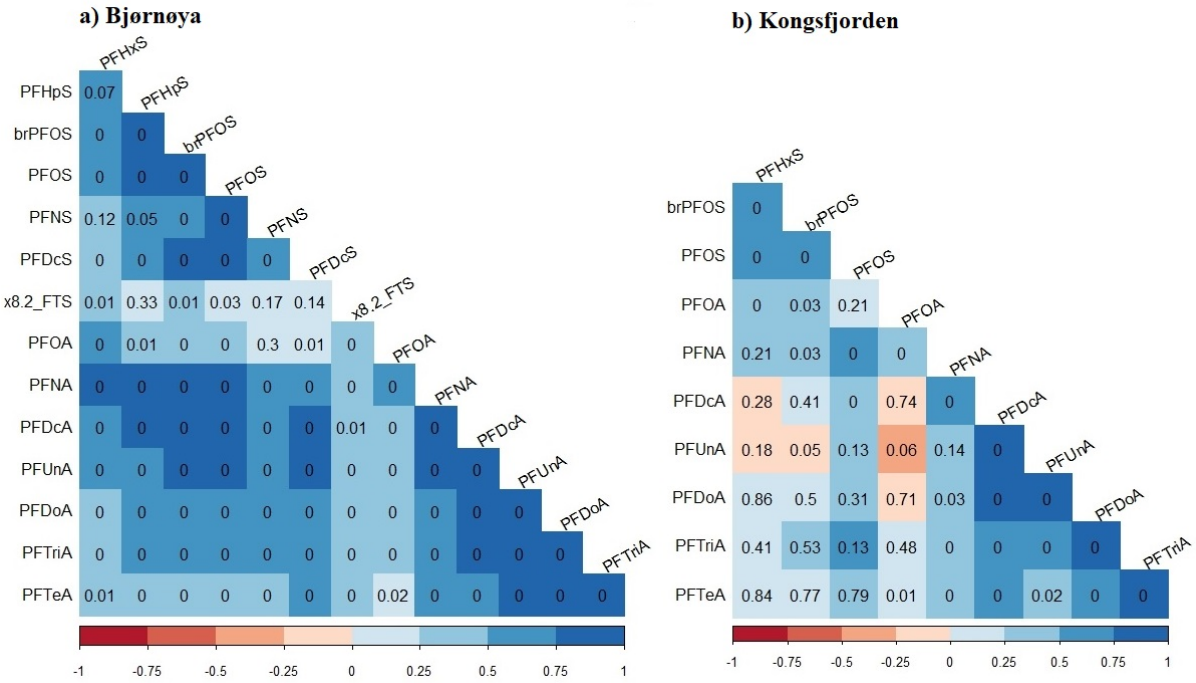

Figure D3: Spearman's ranks correlation matrix for perfluoroalkyl and polyfluoroalkyl substances (PFASs) in glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. Colour gradient indicate degree of correlation where increasing distance from 0 (red or blue) means stronger correlation. P-values are presented with numbers, significance level is set at 0.05.

7.5 Appendix E: Stable isotopes

Table E: Body feather isotopic signatures (mean ± SD, ‰) in male and female glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. Asterisks (*) indicate significant differences between locations according to t-test (*, $p < 0.05$).

			Isotopic Signatures			
Males						
	Bjørnøya			Kongsfjorden		
Year	N	δ13C	δ15N	N	δ13C	δ15N
2015	3	-17.90 ± 0.27	16.28 ± 0.16	3	-17.80 ± 0.31	17.33 ± 1.37
2016	16	-18.33 ± 0.53	16.13 ± 0.99	4	-18.09 ± 1.36	15.07 ± 0.44
2017	8	-18.53 ± 0.29	16.32 ±0.66	5	-18.09 ± 0.36	15.48 ±0.53
2018	-	-	-	3	-18.00 ± 0.09	15.58 ± 0.37
2019	5	-17.99 ± 0.16	16.14 ± 0.52	7	-17.90 ± 0.91	15.48 ± 0.66
Total	32	-18.29 ± 0.46	16.19 ± 0.79*	22	-17.98 ± 0.74	15.67 ± 0.94*
Females						
	Bjørnøya			Kongsfjorden		
Year	N	δ13C	δ15N	N	δ13C	δ15N
2015	3	-18.52 ± 1.33	15.07 ±3.04	4	-17.96 ± 0.76	16.70 ± 1.68
2016	15	-18.89 ± 0.87	15.30 ± 1.41	15	-18.87 ± 0.91	13.96 ± 0.79
2017	8	-18.99 ± 0.47	15.43 ± 1.07	16	-18.41 ± 0.36	14.97 ± 0.90
2018	-	-	-	10	-18.67 ± 0.60	15.33 ± 1.20
2019	9	-19.00 ± 0.78	15.41 ± 0.96	7	-18.50 ± 0.41	15.73 ± 0.89
Total	35	-18.91 ± 0.79*	15.34 ± 1.35	52	-18.57 ± 0.67*	14.98 ± 1.25

7.6 Appendix F: Relationship between OHCs and stable isotopes

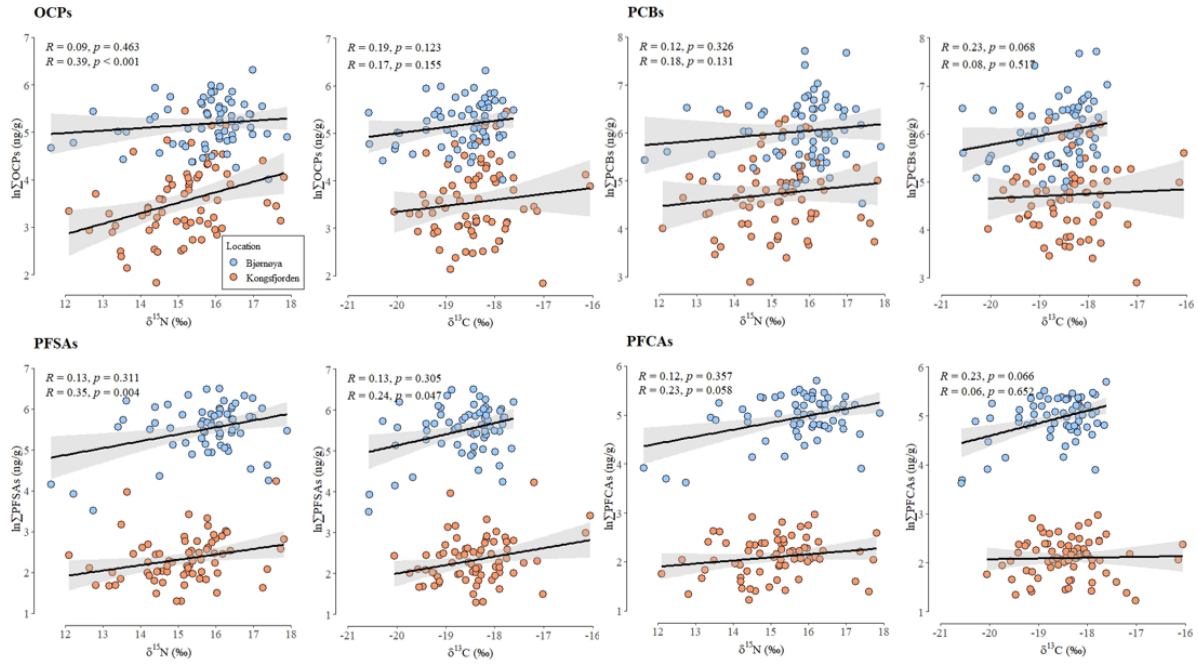

Figure F: Relationships between plasma OHC concentrations and body feather stable isotope ratios with 95% confidence intervals measured in glaucous gulls breeding in Bjørnøya and Kongsfjorden during the breeding seasons of 2015 until 2019. The correlation estimates are derived from Spearman's rank correlation tests.

7.7 Appendix G: Model selection OHCs

Table G: Model selection table for linear mixed effects models (LMM) explaining concentrations of ln transformed OHCs in glaucous gulls breeding in Bjørnøya and Kongsfjorden according to ΔAIC and AIC weights. Degrees of freedom (df) and marginal and conditional R^2 are reported. Continuous predictor variables were standardized prior to analysis. Predictors included $\delta^{15}N$ and $\delta^{13}C$ values in feathers, location (Bjørnøya or Kongsfjorden), sampling years (2015-2019), body condition and sex (female or male). Models with AIC < 2 and lowest K were chosen as the best models highlighted in bold.

Intercept	Location	Year	Location:Year	Sex	BCI	$\delta^{15}N$	$\delta^{15}N$:Location	$\delta^{13}C$	$\delta^{13}C$:Location	K	AICc	ΔAIC	weight	R^2m	R^2c
ln∑OCPs															
4.79	**+**	**+**	**+**	**+**	**-0.11**					**13**	**222.48**	**0.00**	**0.49**	**0.74**	**0.86**
4.79	+	+	+	+	-0.11			-0.01		14	224.96	2.48	0.14		
4.82	+	+	+	+	-0.11	-0.04	+			15	224.97	2.49	0.14		
4.75	+	+	+	+						12	224.99	2.51	0.14		
4.74	+	+	+	+				0.01		13	227.42	4.93	0.04		
4.76	+	+	+	+		-0.01	+			14	228.12	5.64	0.03		
4.95	+	+	+		-0.12					12	231.31	8.83	0.01		
4.96	+	+	+		-0.12	0.01	+			14	232.89	10.41	0.00		
4.92	+	+	+		-0.11			0.04		13	232.94	10.46	0.00		
4.90	+	+	+							11	234.96	12.48	0.00		
4.87	+	+	+					0.06		12	235.67	13.19	0.00		
4.89	+	+	+			0.05	+			13	236.47	13.98	0.00		
5.01	+			+	-0.16	-0.06	+			8	240.76	18.28	0.00		
5.15	+	+								8	246.41	23.93	0.00		
4.97	+			+	-0.13					6	248.73	26.25	0.00		
5.00	+			+		-0.01	+			7	248.86	26.38	0.00		
5.19	+				-0.17	-0.01	+			7	249.86	27.38	0.00		
4.97	+			+	-0.14			-0.01		7	250.93	28.45	0.00		
4.97	+			+						5	253.31	30.83	0.00		
4.97	+			+				0.01		6	255.49	33.01	0.00		
5.17	+					0.05	+			6	258.56	36.07	0.00		
5.14	+					0.16				5	260.19	37.71	0.00		
5.18	+				-0.15					5	260.61	38.13	0.00		
5.19	+				-0.14			0.06		6	261.75	39.27	0.00		
5.18	+									4	266.35	43.87	0.00		
5.20	+							0.08		5	266.43	43.95	0.00		
5.20	+							0.08	+	6	268.62	46.14	0.00		
4.22										3	362.84	140.36	0.00		
ln∑PCBs															
5.87	**+**			**+**	**-0.21**					**6**	**278.16**	**0.00**	**0.49**	**0.51**	**0.79**

5.87	+			+	-0.21	0.01				7	280.34	2.18	0.16			
5.87	+			+	-0.21			-0.00		7	280.38	2.22	0.16			
6.04	+				-0.22					5	282.76	4.60	0.05			
5.66	+	+		+	-0.18					10	282.89	4.73	0.05			
6.03	+				-0.22	0.05				6	284.27	6.11	0.02			
6.05	+				-0.22			0.04		6	284.45	6.29	0.02			
5.64	+	+		+	-0.18	0.03				11	285.10	6.94	0.02			
5.66	+	+		+	-0.18			0.01		11	285.25	7.09	0.01			
5.82	+	+			-0.19					9	286.80	8.64	0.01			
5.76	+	+			-0.18	0.06				10	288.14	9.98	0.00			
5.80	+	+			-0.18			0.05		10	288.43	10.27	0.00			
5.86	+			+						5	290.20	12.04	0.00			
5.61	+	+		+						9	290.88	12.72	0.00			
5.87	+			+		0.03				6	292.16	14.00	0.00			
5.87	+			+				0.01		6	292.35	14.19	0.00			
5.58	+	+		+		0.04				10	292.85	14.69	0.00			
5.61	+	+		+				0.02		10	293.12	14.96	0.00			
5.76	+	+								8	295.22	17.06	0.00			
6.04	+									4	295.66	17.50	0.00			
5.68	+	+				0.08				9	295.92	17.76	0.00			
5.73	+	+						0.07		9	296.22	18.06	0.00			
6.02	+					0.08				5	296.38	18.22	0.00			
6.05	+							0.07		5	296.54	18.38	0.00			
6.06	+							0.11	+	6	298.32	20.16	0.00			
6.03	+					0.06	+			6	298.51	20.35	0.00			
5.75	+	+	+							11	298.68	20.52	0.00			
5.29										3	354.32	76.16	0.00			
ln∑PFSAs																
5.17	**+**	**+**	**+**	**+**				**0.12**		**13**	**204.48**	**0.00**	**0.58**	**0.92**	**0.94**	
5.17	+	+	+	+	0.01			0.12		14	206.95	2.47	0.17			
5.20	+	+	+	+		0.09				13	208.00	3.52	0.10			
5.20	+	+	+	+						12	208.93	4.46	0.06			
5.20	+	+	+	+	-0.00	0.09				14	210.49	6.01	0.03			
5.30	+	+	+					0.17		12	210.74	6.26	0.03			
5.20	+	+	+	+	-0.01					13	211.36	6.88	0.02			
5.30	+	+	+		0.01			0.17		13	213.18	8.70	0.01			
5.36	+	+	+			0.15				12	216.43	11.95	0.00			
5.36	+	+	+		-0.01	0.14				13	218.86	14.38	0.00			
5.42	+	+	+							11	222.03	17.55	0.00			
5.43	+	+	+		-0.02					12	224.33	19.85	0.00			
5.33	+			+		0.14				6	225.38	20.90	0.00			
5.33	+			+	-0.02	0.14				7	227.43	22.95	0.00			
5.38	+			+				0.11		6	228.04	23.56	0.00			
5.69	+	+								8	229.34	24.86	0.00			
5.38	+			+	-0.01			0.11		7	230.21	25.73	0.00			
5.32	+			+						5	230.27	25.79	0.00			
5.47	+					0.18				5	232.25	27.77	0.00			
5.33	+			+	-0.02					6	232.26	27.78	0.00			
5.48	+				-0.03	0.18				6	234.08	29.60	0.00			
5.47	+					0.20	+			6	234.39	29.91	0.00			

5.54	+						0.16		5	234.59	30.11	0.00		
5.54	+			-0.01			0.16		6	236.68	32.20	0.00		
5.54	+						0.18	+	6	236.70	32.22	0.00		
5.52	+								4	242.61	38.13	0.00		
5.52	+			-0.03					5	244.28	39.80	0.00		
3.60									3	454.67	250.19	0.00		
ln∑PFCAs														
4.72	**+**	**+**		**+**			**0.15**	**+**	**11**	**121.30**	**0.00**	**0.52**	**0.94**	**0.95**
4.66	+	+		+		0.08			10	123.42	2.12	0.18		
4.72	+	+		+	-0.01		0.14	+	12	123.65	2.35	0.16		
4.68	+	+		+	-0.02	0.08			11	125.34	4.04	0.07		
4.73	+	+		+					9	126.32	5.01	0.04		
4.74	+	+		+	-0.02				10	128.04	6.74	0.02		
4.82	+	+					0.20	+	10	131.84	10.54	0.00		
4.75	+	+				0.12			9	133.42	12.12	0.00		
4.82	+	+			-0.01		0.19	+	11	134.18	12.88	0.00		
4.77	+	+			-0.02	0.12			10	135.26	13.95	0.00		
4.81	+			+		0.08			6	137.65	16.34	0.00		
4.85	+			+			0.13	+	7	139.12	17.82	0.00		
4.81	+			+	-0.01	0.08			7	139.82	18.51	0.00		
4.81	+			+					5	140.37	19.06	0.00		
4.85	+			+	0.00		0.13	+	8	141.37	20.07	0.00		
4.88	+	+							8	141.64	20.33	0.00		
4.81	+			+	-0.01				6	142.41	21.11	0.00		
4.89	+	+			-0.03				9	143.14	21.83	0.00		
4.92	+					0.12			5	145.86	24.56	0.00		
4.93	+	+	+						11	146.27	24.97	0.00		
4.91	+					0.16	+		6	146.69	25.39	0.00		
4.98	+						0.18	+	6	147.74	26.44	0.00		
4.92	+				-0.01	0.12			6	147.92	26.61	0.00		
4.98	+				0.00		0.18	+	7	149.96	28.66	0.00		
4.97	+						0.08		5	150.32	29.02	0.00		
4.96	+								4	153.20	31.90	0.00		
4.96	+				-0.02				5	154.97	33.66	0.00		
3.23									3	400.51	279.21	0.00