NTNU – Trondheim
Norwegian University of
Science and Technology

Perfluoroalkyl and Polyfluoroalkyl Substances (PFASs) Affect the Thyroid Hormone System, Body Condition, and Body Mass in Two Arctic Seabird Species

Amalie Vigdel Ask

Environmental Toxicology and Chemistry
Submission date: May 2015
Supervisor: Bjørn Munro Jenssen, IBI
Co-supervisor: Geir Wing Gabrielsen, Norsk Polarinstitutt

Norwegian University of Science and Technology
Department of Biology

NTNU – Trondheim
Norwegian University of
Science and Technology

IN COLLABORATION WITH

Cover photo: Black-legged kittiwakes. Photo: Amalie V. Ask.

Acknowledgements

First of all I need to thank all the kittiwakes and arctic skuas who, albeit somewhat reluctantly, have been part of this study.

I want to express my sincere gratitude and appreciation to my supervisors Geir Wing Gabrielsen and Bjørn Munro Jenssen. Thank you for your kind words, encouragement, and for providing me with this opportunity.

While not officially my supervisors, thank you to Olivier Chastel, Frédéric Angelier, and Sabrina Tartu for guiding this fledgling bird scientist in the art of kittiwake catching and blood sampling. And for acting as living encyclopedias, answering any question I could think of about any bird species imaginable.

A big thank you to Charline Parenteau for helping me with the thyroid hormone assays.

Thank you to the wonderful people at NILU, Tromsø for helping me in my analyses of PFASs. I must especially acknowledge Arntraut Götsch, Dorte Herzke, and Linda Hanssen for all their help.

For my last field season, "Captain" Solveig Nilsen was invaluable as my field assistant for nest checks, kittiwake capture, and blood sampling. She also navigated the sea expertly getting us safely to the colony each time despite calving glaciers.

Thank you to Sveinn Are Hanssen and Børge Moe for providing me with the arctic skua samples and for answering questions about statistics and logistics.

I also wish to thank my fellow "envitox" students for two memorable years.

Last, but certainly not least, my heartfelt gratitude for the continued support from my friends and family. Thank you to Solveig Nilsen and Åse Berg for looking over my Norwegian abstract, and particularly to Geir Vigdel and Shane Dielschneider for reading through my thesis with the eyes of hawks. And, finally, thank you Patrick for always being there.

Trondheim, May 2015

Amalie Vigdel Ask

Abstract

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are being transported into the Arctic where they are frequently detected in wildlife. These compounds are suspected thyroid hormone (TH) disruptors due to their structural similarity to triiodothyronine (T3) and thyroxine (T4), in addition to their propensity to bind to proteins. Therefore, PFASs may affect THs by competitive binding to the thyroid binding proteins in the blood. The aim of the study was to investigate the concentrations of PFASs and THs, and examine effects of PFASs on THs, body condition, and body mass in black-legged kittiwakes (*Rissa tridactyla*) and arctic skuas (*Stercorarius parasiticus*).

Blood was collected from breeding black-legged kittiwakes and arctic skuas. Black-legged kittiwakes (hereafter kittiwakes) were sampled in Kongsfjorden, Svalbard in 2013 and 2014. Arctic skuas were sampled on Brensholmen, Norway and in Kongsfjorden in 2014. The blood was analyzed for perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnDA), perfluorododecanoate (PFDoDA), perfluorotridecanoate (PFTrDA), perfluorotetradecanoate (PFTeDA), perfluorohexane sulfonate anion (PFHxS), branched and linear perfluorooctane sulfonate anion (brPFOS and linPFOS), perfluorodecane sulfonate anion (PFDS), and perfluorooctane sulfonamide (FOSA). The analyses were performed on plasma samples for the black-legged kittiwakes and on whole blood samples for the arctic skuas. PFHxA, PFHpA, PFOA, PFDS, and PFHxS were not detected in either bird species. Furthermore, total THs (TT3 and TT4) were quantified from plasma samples in both species. The resulting data was analyzed statistically to examine if there were associations between PFASs, THs, body condition (BC), and body mass.

The dominant PFASs in both kittiwakes and arctic skuas were linPFOS and PFUnDA. In both species, males generally had significantly higher concentrations of PFASs than the females. Furthermore, positive correlations between PFASs and THs were identified in both kittiwakes and arctic skuas. Male kittiwakes with high levels of PFDoDA, PFTrDA, and PFTeDA were in a better body condition than males with lower levels. Conversely, in female kittiwakes and male arctic skuas PFASs were negatively correlated to BC and body mass. The results indicate that PFASs affect the thyroid system, BC, and body mass in the two seabird species.

Sammendrag

Perfluoralkylerte stoffer (PFASer) blir transportert til Arktis hvor de ofte detekteres i arktisk fauna. Det mistenkes at forbindelsene forstyrrer tyroidhormonene (TH) på grunn av deres strukturelle likheter til trijodthyronin (T3) og thyroxin (T4), samt at PFASer binder seg til proteiner. Dermed kan PFASer påvirke tyroidhormonene ved å konkurrere om å binde til transportproteiner i blodet. Studiets formål var å undersøke PFAS- og TH-konsentrasjonene, og effekten av PFASer på TH, kroppskondisjon og kroppsvekt i krykkje (*Rissa tridactyla*) og tyvjo (*Stercorarius parasiticus*).

Det ble tatt blodprøver av hekkende krykkje og tyvjo. Blodprøvene fra krykkje ble innhentet i Kongsfjorden, Svalbard i 2013 og 2014, mens blodprøvene fra tyvjo ble innhentet på Brensholmen i Troms og i Kongsfjorden i 2014. Blodet ble analysert for perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnDA), perfluorododecanoate (PFDoDA), perfluorotridecanoate (PFTrDA), perfluorotetradecanoate (PFTeDA), perfluoroheksane sulfonat anion (PFHxS), forgreinet og lineær perfluorooctane sulfonat anion (brPFOS og linPFOS), perfluorodecane sulfonat anion (PFDS), og perfluorooctane sulfonamide (FOSA). Analysene av PFAS ble utført på plasma prøver i krykkje og på helblod prøver i tyvjo. PFHxA, PFHpA, PFOA, PFDS, og PFHxS ble ikke detektert i noen av fugleartene. Videre ble total T3 (TT3) og T4 (TT4) kvantifisert. Dataene ble statistisk analysert for å undersøke om det var sammenhenger mellom PFASer, TH, kroppskondisjon og kroppsvekt.

De dominerende PFASene i både krykkje og tyvjo var linPFOS og PFUnDA. For begge artene hadde hannene generelt høyere PFAS-konsentrasjoner enn hunnene. Videre var det positive korrelasjoner mellom PFASer og TH i både krykkje og tyvjo. Hos krykkje hadde hanner med høyere nivå av PFASer bedre kroppskondisjon enn hanner med lavere nivå. I motsetning til dette var PFAS-konsentrasjoner i krykkjehunner og tyvjohanner negativt korrelert med kroppskondisjon og kroppsvekt. Resultatene indikerer at PFASer påvirker tyroidsystemet, kroppskondisjon og kroppsvekt i begge fugleartene.

Abbreviations

ANOVA	Analysis of variance
APFO	Ammonium perfluorooctanoate
BC	Body condition
brPFOS	Branched perfluorooctane sulfonate anion
C	Carbon atom
CV	Coefficient of variance
DDT	Dichlorodiphenyltrichloroethane
DNA	Deoxyribonucleic acid
ECF	Electrochemical fluorination
EDC	Endocrine disrupting chemical
F	Fluorine atom
FASAs	Perfluoroalkane sulfonamides
FOSA	Perfluorooctane sulfonamide
G	Gauge
H	Hydrogen atom
H	Hormone (in thyroid hormone assay)
H*	Radioactive hormone (in thyroid hormone assay)
I	Iodine atom
ID	Identification
IDs	Iodothyronine deiodinases
linPFOS	Linear perfluorooctane sulfonate anion
LOD	Limit of detection
LOQ	Limit of quantification
MS	Mass spectrometry
n	Number of observations
NILU	Norwegian Institute for Air Research
NTNU	Norwegian University of Science and Technology
OSF	Octane sulfonyl fluoride
p	Significance level
PBDE	Polybrominated diphenyl ether
PCB	Polychlorinated biphenyl
PCR	Polymerase chain reaction
PFASs	Perfluoroalkyl and polyfluoroalkyl substances
PFCAs	Perfluoroalkyl carboxylic acids
PFDA	Perfluorodecanoate
PFDS	Perfluorodecane sulfonate anion
PFDoDA	Perfluorododecanoate
PFHpA	Perfluoroheptanoate
PFHxA	Perfluorohexanoate
PFHxS	Perfluorohexane sulfonate anion
PFNA	Perfluorononanoate
PFOA	Perfluorooctanoate

PFOS	Perfluorooctane sulfonate anion
PNEC	Predicted no effect concentration
PFSAs	Perfluoroalkane sulfonates
PFTeDA	Perfluorotetradecanoate
PFTrDA	Perfluorotridecanoate
PFUnDA	Perfluoroundecanoate
POPs	Persistent organic pollutants
PPAR	Peroxisome proliferator-activated receptor
r	Pearson's coefficient
R	Functional group
rpm	Rotations per minute
rT3	Reverse triiodothyronine
SD	Standard deviation
SRM	Standard reference material
sumPFOS	Sum of branched and linear perfluorooctane sulfonate anion
T2	Diiodothyronine
T3	Triiodothyronine
T4	Thyroxine
TA	Total acitivity (of radioactive hormone)
TH	Thyroid hormones
TRH	Thyrotropin releasing hormone
TSH	Thyroid stimulating hormone
TT3	Total triiodothyronine
TT4	Total thyroxine
TTR	Transthyretin
UHPLC	Ultra-high pressure liquid chromatography
UV	Ultraviolet
V	Volt

Contents

1. Introduction

1.1 Contaminants in the Arctic

The Arctic is often regarded as a pristine and unspoilt environment. However, many toxicants have been detected in the abiotic and biotic components of the environment (Lockhart et al., 1992; Hung et al., 2010; Letcher et al., 2010). Some of these originate from local sources in the Arctic, such as heavy metals and radionuclides following a mining operation (Dowdall et al., 2003; Perner et al., 2010). Others are predominantly produced in the mid-latitudes and transported into the Arctic via two main pathways: atmospheric long range transport and oceanic currents (Butt et al., 2010; Hung et al., 2010). To a lesser extent, migrating animals can also introduce toxicants into the arctic environment (Blais et al., 2005). Persistent organic pollutants (POPs) are a class of toxicants that are commonly detected in the Arctic (Butt et al., 2010; Letcher et al., 2010). The so-called legacy POPs are banned according to the Stockholm Convention (www.pops.int), and include dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCBs) as arguably the best known examples. There are, however, emerging groups of POPs being found in the Arctic, one of them being the perfluoroalkyl and polyfluoroalkyl substances (PFASs) (Giesy and Kannan, 2001; Haukås et al., 2007; Butt et al., 2010; Muir and de Wit, 2010). It is worthwhile to note here that the term "emerging" refers to the fact that while PFASs have been produced for over 50 years (Buck et al., 2011), it is only recently that they have come under scientific scrutiny.

1.2 Perfluoroalkyl and Polyfluoroalkyl Substances (PFASs)

PFASs are a group of synthetically produced chemicals which follow the basic formula of $F(CF_2)_n$-R, where R designates the functional group. The aliphatic alkyl chain varies in length, typically from C4 to C16, and is hydrophobic. Perfluoroalkyl substances are those compounds where all the hydrogen atoms have been replaced by fluorine atoms, except for on functional groups. In contrast, polyfluoroalkyl species do not have a complete fluorine substitution (Buck et al., 2011). The functional group is hydrophilic, but can have a neutral, positive, or negative charge. PFASs are further divided into several classes, among them the perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonates (PFSAs). Arguably the two best known PFASs are perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), which are examples of a PFCA and a PFSA, respectively (Buck et al., 2011).

Due to the C-F bond, which is one of the strongest covalent bonds known, PFASs are highly resistant to chemical and biological degradation (Key et al., 1997; Parsons et al., 2008). Several PFASs are synthesized for use in manufacturing and industry (Butt et al., 2010). They are used as surfactants and repellents because of their resistance to degradation, and are often found in fire-fighting foam (Moody and Field, 2000; Butt et al., 2010).

PFASs are produced by two main processes: telomerization and electrochemical fluorination (ECF). In telomerization, a telogen and a taxogen react with each other. The telogen is a perfluoroalkyl iodide, typically pentafluoroethyl, and a common taxogen is tetrafluoroethylene. The product of the reaction is a mixture of perfluoroalkyl iodides with longer perfluorinated chains. Normally this mixture is subjected to a second process in which it is reacted with ethylene. Interestingly, if both the telogen and taxogen are linear compounds the resultant mixture consists of only linear perfluoroalkyl chains (Lehmler, 2005; Buck et al., 2011).

In the ECF method an organic raw material, commonly octane sulfonyl fluoride (OSF), is subjected to electrolysis in anhydrous hydrofluoric acid. The result of which is a complete replacement of the H-atoms on the OSF with F-atoms. This is a free-radical process, which causes breakages and rearrangements of the carbon chain, leading to a mixture of linear and branched products. The ratio of linear to branched isomers is variable and depends on how the ECF process is controlled (Lehmler, 2005; Buck et al., 2011).

PFASs may have both linear and branched isomers and are found as a mixture of the two in environmental matrices (Rayne et al., 2008). PFOS, for instance, have at least 11 branched isomers, with 89 isomers possible in theory (Langlois et al., 2007; Rayne et al., 2008). Although the mixture of linear and branched isomers may be problematic in quantification, the ratio between linear and branched isomers may be used to determine the source of the PFAS (De Silva and Mabury, 2004; Riddell et al., 2009). Furthermore, there are indications that the linear and branched isomer have different toxicological properties (Loveless et al., 2006).

As stated above, PFASs are primarily produced in mid-latitudinal countries and are thought to be transported to the Arctic via atmospheric transport (Stock et al., 2007; Young et al., 2007) and oceanic currents (Prevedouros et al., 2006; Yamashita et al., 2008), either as the final degradation product or as a precursor molecule (Butt et al., 2010). PFASs are bioaccumulative and can be biomagnified in food chains (Kannan et al., 2005; Haukås et al., 2007; Houde et al., 2011), and are consequently often detected in arctic wildlife such as plankton, fish, seals, whales, polar bears and seabirds (Tomy et al., 2004; Letcher et al., 2010; Rotander et al., 2012).

The PFASs are absorbed through oral, dermal, and respiratory pathways, with oral absorption being the most efficient (Stahl et al., 2011). Once absorbed, the PFASs preferentially bind to serum albumin in the plasma (Han et al., 2003; Jones et al., 2003; Verreault et al., 2005), but also to β-lipoprotein and fatty acid binding proteins in the liver (Luebker et al., 2002). Due to PFASs' stability, they resist the biotransformation pathways in the organism, and consequently they are only removed by excretion (Stahl et al., 2011). PFASs have been shown to have immunotoxicity, hepatotoxicity, and developmental toxicity, and they are believed to be endocrine disrupting chemicals (Lau et al., 2007).

1.3 Endocrine Disrupting Chemicals

Endocrine disrupting chemicals (EDCs) have the ability to interfere with the synthesis, secretion, transport, binding, action, or elimination of hormones (Crisp et al., 1997). EDCs encompass a wide group of chemicals; for instance, more than 150 synthetic chemicals are known to interfere with the production, transport and metabolism of the thyroid hormone (Howdeshell, 2002), including organohalogen toxicants. There are indications that PFASs are endocrine disruptors and affect thyroid hormones (Oakes et al., 2005; Weiss et al., 2009; Jensen and Leffers, 2008; Cassone et al., 2012; Joensen et al., 2013).

1.4 Thyroid Hormones (THs)

Thyroid hormones (THs) are regulated through the hypothalamic-pituitary-thyroid axis. The hypothalamus secretes thyrotropin releasing hormone (TRH) which acts on the pituitary gland which in turn secretes thyroid stimulating hormone (TSH). TSH travel to the thyroid gland where it binds to receptors on the cell surface and trigger an increase in iodide uptake into the cell via the sodium/iodide symporter (Diamanti-Kandarakis et al., 2009). There, through a series of steps, the final products thyroxine (T4) and triiodothyronine (T3) are produced. Once they are released into the blood stream, the majority of the hormones bind to transthyretin (TTR) and albumin (Ishihara et al., 2003) and are transported to the target tissue where transporters on the cell surface transport the hormones into the cell. Inside the cell THs binds to the thyroid receptor which is a ligand-activated transcription factor belonging to the nuclear hormone receptor superfamily (Yen et al., 2006). T3 is the active form of the hormone, and so T4 is converted to T3 inside the cell by iodothyronine deiodinases (Ishihara et al., 2003; Diamanti-Kandarakis et al., 2009). To conclude the axis, T3 and T4 provide a negative feedback effect on the hypothalamus and pituitary, thus regulating the concentration of thyroid hormone in the body (Zoeller, 2003).

Thyroid hormones are required for proper brain development, cell differentiation, controlling metabolism, growth, body weight, thermoregulation, and reproduction, as well as hatching and molting in birds (Verreault et al., 2004; Verreault et al., 2007a; Diamanti-Kandarakis et al., 2009).

In the Arctic, wildlife face challenges regarding the low ambient temperatures and demanding climate. Even for migratory species who are only in the Arctic during the summer season, the conditions are characterized by low ambient temperatures and frequently inclement weather. Migratory species, such as seabirds, arrive in the Arctic during late spring/early summer to reproduce, before returning to their wintering areas in the autumn (Anker-Nilssen, 2000). Reproduction is a demanding event (Bryant, 1997; Welcker et al., 2013), which, when combined with the challenges of the surrounding environmental conditions, necessitates a proper thyroid function. As mentioned above, thyroid hormones are involved in metabolism, thermoregulation, reproduction and hatching, amongst other processes. If the thyroid system is disrupted, it may impact the species' ability to reproduce and thus affect population levels.

Two arctic seabird species, the black-legged kittiwake (*Rissa tridactyla*) and arctic skua (*Stercorarius parasiticus*), are experiencing population declines (Jones et al., 2008; Anker-Nilssen, 2010; Anker-Nilssen et al., 2013). The reasons for this are still unknown, and further research is required to understand the declines.

1.5 Black-legged Kittiwakes

Black-legged kittiwakes (hereafter kittiwakes) are small gulls with a circumpolar distribution (Anker-Nilssen, 2000). They are surface feeders and their diet consists mainly of fish, such as polar cod (*Boreogadus saida*) and capelin (*Millotus villosus*), but also includes invertebrates (Mehlum and Gabrielsen, 1993; Dahl et al., 2003). They will also feed on offal from fishing vessels (Anker-Nilssen, 2000).

The global breeding distribution of kittiwakes ranges from Canada, Greenland, United Kingdom, Faroe Islands, Iceland, Norway, Svalbard, and Russia (Anker-Nilssen, 2000). It is a numerous gull species, with the population on Svalbard believed to have been increasing over the last century (Anker-Nilssen, 2000), but a decline in breeding populations has recently been reported (Anker-Nilssen, 2010; Anker-Nilssen et al., 2013). The reasons for this are, as of yet, unknown, but it has been speculated that it may be linked to reduced food availability and nest predation (Anker-Nilssen et al., 2013).

Persistent organic pollutants are detectable in the kittiwakes' blood and tissues, and among them PFASs (Borgå et al., 2001; Fisk et al., 2001; Tomy et al., 2004; Letcher et al., 2010; Nøst et al., 2012). Due to PFASs being a fairly novel class of POPs, there has not been extensive studies on how they affect THs in kittiwakes, but one study did report a positive association between PFASs and THs (Nøst et al., 2012). The declining kittiwake population warrants further investigation to examine if PFASs may affect the levels of THs and thus exacerbate the decline.

1.6 Arctic Skuas

Arctic skuas, or parasitic jaegers as they are known in North America, are also circumpolar seabirds. They are kleptoparasitic and frequently harass other seabirds, such as kittiwakes, until they regurgitate and drop their food which the arctic skua recovers and eats. This is especially common for arctic skuas breeding close to a major seabird colony (Furness, 1996). In addition, arctic skuas feed on small mammals and birds, eggs, insects, berries, and carrion (Furness, 1996; Anker-Nilssen, 2000). Subsequently, this means that arctic skuas will likely be positioned at a higher trophic level compared to the kittiwakes depending on the extent of the food coming from kleptoparasitism.

Arctic skuas breed in the northern hemisphere with a notable presence in Scotland, Shetland, Iceland, Northern Norway, and on Svalbard. The southern limit of the breeding range appears to be around Scotland (O'Donald, 1983). The breeding population of arctic skuas in the

United Kingdom is in a considerable decline (Jones et al., 2008). This is believed to be partly due to a collapse in local lesser sandeel (*Ammodytes marinus*) populations, but great skuas (*Stercorarius skua*) are also displacing the arctic skuas from their territories (O'Donald, 1983; Dawson et al., 2011).

Arctic skuas are larger than kittiwakes and there is a reverse sexual dimorphism in the arctic skua with females being larger than the males (Catry et al., 1999).

There are large knowledge gaps in the scientific literature regarding toxicant levels in arctic skuas. Its relative, the great skua, which can often be found breeding in the same areas as the arctic skua (O'Donald, 1983) has been shown to have very high body burdens of POPs and polybrominated diphenyl ethers (PBDEs) (Bourgeon et al., 2012), and PFASs have been detected in its plasma and eggs (Leat et al., 2013). It is important to note, however, that great skuas feed at a higher trophic level than arctic skuas. Indeed, the great skuas are known to predate on arctic skua fledglings and juveniles (Jones et al., 2008). And as POPs are bioaccumulative, it is expected that organisms feeding at a higher trophic level will have greater concentrations of the toxicants.

Due to the decline in population levels in the United Kingdom, it is important to examine the potential impact of PFASs on the thyroid hormone system in the arctic skua. Furthermore, given the fact that there is a lack of literature on this species with regards to toxicant levels, this study will help to partially bridge that knowledge gap.

1.7 Aim of Study

The aim of this study was to examine the concentrations of PFASs and thyroid hormones in kittiwakes breeding in Kongsfjorden, Svalbard and in arctic skuas breeding on Brensholmen, Norway and in Kongsfjorden, Svalbard. Furthermore, the study aimed to investigate whether PFASs exert any adverse effects by disrupting the thyroid system or by affecting the body condition and body mass of the seabirds.

It is hypothesized that PFAS concentrations will be detected in the two species, and that they will be associated with altered thyroid hormone levels. Furthermore, it is expected that high levels of PFASs will have an effect on the body condition and body mass of the two species.

2. Materials and Methods

2.1 Sampling Areas

The black-legged kittiwakes (*Rissa tridactyla*) were all sampled from one colony ("Krykkjefjellet" located 6 km south-east of Ny-Ålesund, 78°55'00'N, 11°56'00'E) in Kongsfjorden, Svalbard during the chick-rearing period of their breeding cycle. The sampling periods were between 03.07 - 06.07 in 2013 and 14.07 - 20.07 in 2014. Each year 15 male and 15 female kittiwakes were sampled in the colony resulting in 60 samples.

The breeding arctic skuas (*Stercorarius parasiticus*) were sampled on Brensholmen (36 km from Tromsø, 69°35'20'N, 18°02'29'E), Norway and at different nest sites around Kongsfjorden, Svalbard. On Brensholmen they were sampled on 10.06 and 12.06 in 2014, and in Kongsfjorden they were sampled between 27.06 - 15.07 in 2014. Five individuals were sampled on Brensholmen and 11 individuals were sampled in Kongsfjorden.

During summer the weather conditions in Kongsfjorden are characterized by continuous daylight, low ambient temperatures (below 5-7 °C), and occasional precipitation.
On Brensholmen in June, the conditions are characterized by continuous daylight, temperatures averaging between 10-20 °C, and occasional rain showers (Norwegian Meteorological Institute).

Figure 1: A map of Kongsfjorden, Svalbard (Norwegian Polar Institute).

2.2 Field Procedures

Breeding kittiwakes were caught on the nest using a telescopic rod with a nylon noose attached at the end. The noose was placed around the neck of the bird and tightened before the bird was lifted off the nest. Measurements were taken of the skull and beak length (hereafter

referred to as skull length), wing length, tarsus length, and body mass. A maximum of 2.5 mL of blood was collected in total from the brachial vein using a heparinized needle and syringe (25G needle with 0.5mL and 2.5 mL syringe, Terumo, Hatagaya, Tokyo, Japan). The blood was stored on ice during field work and centrifuged (6000 rpm, 10 minutes) upon returning to the laboratory in Ny-Ålesund. After centrifugation the plasma was transferred into another Eppendorf tube and frozen at -20 °C. The remaining red blood cells were kept in the original 1.5mL Eppendorf tubes and frozen at -20 °C to be used for sex determination and possible further analysis in other projects. At the end of the field season all samples were kept frozen and shipped to laboratories in Tromsø, Norway; Trondheim, Norway; and Chizé, France.

The arctic skuas were caught either on their nests using a nest trap or by the use of a cannon net during the incubation period. Measurements were taken of the skull length, wing length, tarsus length, and body mass. Using a heparinized needle and syringe (25G needle and 2.5 mL syringe, Terumo, Hatagaya, Tokyo, Japan), 2 mL of blood was taken from the brachial vein. The blood was stored on ice while in the field and centrifuged at 6000 rpm for 10 minutes after returning to laboratory facilities in Ny-Ålesund and Tromsø, depending on where the bird was caught. The plasma, remaining red blood cells, and whole blood was frozen at -20 °C, and kept frozen until laboratory analyses commenced.

The study on kittiwakes and arctic skuas on Svalbard was approved by the Governor of Svalbard and the Norwegian Animal Research Authority (www.fdu.no). The study on arctic skuas on Brensholmen was approved by the County Governor of Troms. The RiS IDs of the kittiwake project and arctic skuas project were 6596 and 3608, respectively. All handling and sampling of the birds were in accordance with the regulations of the Norwegian Animal Welfare Act.

2.3 Analyses of Perfluoroalkyl and Polyfluoroalkyl Substances

All the PFAS analyses were performed at the Norwegian Institute for Air Research (NILU) in Tromsø, Norway. The samples of kittiwakes from 2013 were analyzed in February 2014 and the samples of kittiwakes and arctic skuas from 2014 were analyzed in August 2014. All the kittiwake samples were plasma samples while the arctic skua samples were whole blood samples.

The samples were analyzed for the PFCAs; perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDA), perfluoroundecanoate (PFUnDA), perfluorododecanoate (PFDoDA), perfluorotridecanoate (PFTrDA), perfluorotetradecanoate (PFTeDA), the PFSAs; perfluorohexane sulfonate anion (PFHxS), branched and linear perfluorooctane sulfonate anion (brPFOS and linPFOS), perfluorodecane sulfonate anion (PFDS), and finally for the perfluoroalkane sulfonate (FASA) perfluorooctane sulfonamide (FOSA). The PFAS analytes are listed in Table 1 together with their chemical structures and CAS registry numbers. In

addition, the sum of branched and linear PFOS (abbreviated sumPFOS) was calculated based on the quantified concentrations.

Table 1: A list of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) analyzed for in breeding black-legged kittiwakes (*Rissa tridactyla*) and arctic skuas (*Stercorarius parasiticus*) caught in Kongsfjorden, Svalbard and Brensholmen, Norway 2013 and 2014. The PFASs analyzed are divided into perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkane sulfonates (PFSAs), and perfluoroalkane sulfonamides (FASAs).

Group	Abbreviation	Analyte	Chemical structure	CAS Registry Number
PFCAs	PFHxA	Perfluorohexanoate	$C_5F_{11}COO^-$	92612-52-7
	PFHpA	Perfluoroheptanoate	$C_6F_{13}COO^-$	120885-29-2
	PFOA	Perfluorooctanoate	$C_7F_{15}COO^-$	45285-51-6
	PFNA	Perfluorononanoate	$C_8F_{17}COO^-$	72007-68-2
	PFDA	Perfluorodecanoate	$C_9F_{19}COO^-$	73829-36-4
	PFUnDA	Perfluoroundecanoate	$C_{10}F_{21}COO^-$	196859-54-8
	PFDoDA	Perfluorododecanoate	$C_{11}F_{23}COO^-$	171978-95-3
	PFTrDA	Perfluorotridecanoate	$C_{12}F_{25}COO^-$	862374-87-6
	PFTeDA	Perfluorotetradecanoate	$C_{13}F_{27}COO^-$	365971-87-5
PFSAs	PFHxS	Perfluorohexane sulfonate anion	$C_6F_{13}SO_3^-$	108427-53-8
	brPFOS	Branched perfluorooctane sulfonate anion	$C_8F_{17}SO_3^-$	45298-90-6
	linPFOS	Linear perfluorooctane sulfonate anion	$C_8F_{17}SO_3^-$	45298-90-6
	PFDS	Perfluorodecane sulfonate anion	$C_{10}F_{21}SO_3^-$	126105-34-8
FASAs	FOSA	Perfluorooctane sulfonamide	$C_8F_{17}SO_2NH_2$	754-91-6

2.3.1 Extraction and Clean-up

The blood samples were analyzed for PFASs following a method developed by Powley et al. (2005) and modified for plasma and blood by Hanssen et al. (2013). A total of 200 µL of plasma (400 µL if whole blood was used) was pipetted into an Eppendorf centrifuge tube. Then 20 µL of the internal standard (0.1 ng/µL ^{13}C PFAS mix) was added to each sample. A complete list of the ^{13}C PFAS mix is included in Appendix A.

Following the addition of 1 mL of methanol (LiChrosolv, Merck , Darmstadt, Germany) the tubes were thoroughly vortexed. The extraction itself was achieved through placing the tubes into an ultrasonic bath 3 times 10 minutes. In between the ultrasonic treatment, the tubes were vortexed, denaturizing the proteins. After the extraction of PFASs into the methanol was completed, the tubes were centrifuged for 5 minutes at 10000 rpm. The supernatant was transferred to an Eppendorf tube containing 25 mg ENVI-Carb 120/400 (Supelco 57210-U, Bellefonte, PA, USA) and 50 µL glacial acetic acid (Merck, Darmstadt, Germany). After the transfer the tube was vortexed before centrifugation at 10000 rpm for 10 minutes. Exactly 0.5 mL of the supernatant was transferred to a glass vial and 20 µL of the recovery standard (0.1 ng/µL 3,7-diMeO-PFOA) was added.

Prior to analysis, 50 µL of the solution was added to an autosampler vial along with 50 µL of 2 mM NH_4OAc in water.

2.3.2 Instrumental Analysis

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

2.3.3 Quantification and Quality Assurance

Following the instrumental analysis, the samples were quantified using LCQuan software (version 2.6, Thermo Fisher Scientific Inc., Waltham, MA, USA).
A standard curve was calculated based on known concentration of ^{12}C and ^{13}C labeled PFASs, and equation 1 below was used in the quantification.

$$C_{sample} = \frac{Rf(C_{std} * Area_{sample})}{Area_{std}} \qquad \text{(Equation 1)}$$

C_{sample} is the concentration of the sample, C_{std} is the concentration of the standard, $Area_{sample}$ is the area under the curve of the sample obtained from the chromatography, and $Area_{std}$ is the area under the curve of the standard obtained from the chromatography. Finally, Rf is the response factor determined from the areas and concentrations of the ^{12}C and ^{13}C labeled equivalents in the chromatography.

For every batch of 10 samples, one tube with a blank and one with a standard reference material (SRM 1957, National Institute of Standards and Technology, Gaithersburg, MD, USA) were included. The blank contained all reagents save the plasma or whole blood and the SRM tube contained 200 µL of a reference with known concentrations of PFASs. Both the blank and SRM underwent the exact same treatment as the tubes with samples. This, together with an internal and a recovery standard, formed the basis of the quality assurance. The internal standard contained known concentrations of ^{13}C labeled PFASs. This standard was added at the very beginning of the extraction, with the assumption of an equal loss of the internal standard and the ^{12}C PFAS in the sample through the extraction and clean-up processes. The recovery standard was added just before running the samples on the instrument for the purpose of determining the performance of the analytical method used. The reference samples were within the acceptable range and the blank samples below the limit of acceptable contamination determined by the laboratory .

2.4 Analyses of Thyroid Hormones

The hormone assays, total triiodothyronine (TT3) and total thyroxine (TT4), were performed at the Centre d'Etudes Biologiques de Chizé, France.

The method for determining TT3 and TT4 is the same, and is a radioimmunoassay (Chastel et al., 2003). The principles behind it is the competitive binding between a known concentration of hormone containing radiolabeled ^{125}I (denoted H*, L-3,5,3'-^{125}I-Triiodothyronine and L-^{125}I-Thyroxine, PerkinElmer Inc., Waltham, MA, USA) and the unknown concentration of the sample hormone (denoted H) for binding sites on an antibody. To the H* solution, 8-anilino-1-naphthalenesulfonic acid ammonium salt (Sigma-Aldrich, St. Louis, MO, USA) was added to prevent THs from binding to the thyroxin binding globulin.

The assay ran over three consecutive days. On the first day plasma was pipetted into labeled polypropylene tubes in duplicates. A series of known concentrations of the thyroid hormone in question was included to create a calibration curve (human triiodothyronine and thyroxine, Sigma-Aldrich, St. Louis, MO, USA). There were two tubes for measuring the total activity (TA) of H* (following H* addition, the tubes were capped to avoid cross-contamination), three tubes for measuring non-specific binding where all reagents were added except for sample plasma and the first antibody (whole antiserum from rabbit, Sigma-Aldrich, St. Louis, MO, USA). Three tubes were included to measure the maximum binding of H*. No sample plasma was added to the tube so there was no competition for binding sites for H*. In addition, several reference materials were run along with the samples for quality control; see below.

The samples and reference materials were treated identically, and 10 - 25 µL (the volume was recorded) of sample/reference material was pipetted into the tubes in duplicates. Then the first antibody and H* was added. All the tubes were covered with aluminium foil before being vortexed and stored at 4 °C for at least 24 hours to allow sufficient time for the reaction to reach equilibrium.

On the second day, the second antibody (sheep against rabbit antibody, Sigma-Aldrich, St. Louis, MO, USA) was added to all the tubes except for TA. This antibody bound the first one, and aided immunoprecipitation. The tubes were subsequently vortexed and stored overnight at 4 °C.

On the third and final day, 1 mL barbital buffer (sodium 5,5-diethylbarbiturate and barbital, Sigma-Aldrich, St. Louis, MO, USA) with PEG 6000 (polyethylene 6000, Sigma-Aldrich, St. Louis, MO, USA), which further aided the precipitation, was added to all tubes except TA. The tubes were centrifuged for 45 minutes at 4700 - 4800 rpm at 18-20 °C. They were then decanted and left to dry for at least 60 minutes before being placed in the gamma counter (2470 Automatic Gamma Counter Wizard^2, PerkinElmer Inc., Waltham, MA, USA) to be analyzed.

The reference materials were goat serum, canary serum, L1, L2, and L3 (Liquicheck immunology control), deer serum, and human serum. All reference materials were supplied by Bio-Rad (Bio-Rad Laboratories Inc., Hercules, CA, USA). For quality control, the reference materials were used and were within the acceptable range determined by the laboratory. Furthermore, since every sample was run in duplicates, only samples with a coefficient of variance (CV) less than 15% were used in the statistics.

For kittiwakes, the mean CV calculated for the TT4 assays was 3.82% and 3.34%. Mean CV for the TT3 assays was 4.76% and 4.50%. The mean inter-assay variation for TT3 was 8.98% and 11.4% for TT4. For arctic skuas, the mean intra-assay CV of the duplicates was 3.32% for TT3 and 4.24% for TT4, while the mean inter-assay CV was 8.98% for TT3 and 11.4% for TT4.

2.5 Sex Determination

Sex determination by polymerase chain reaction (PCR) and gel electrophoresis were performed on kittiwake samples which previously had not been molecularly sexed. Upon completion of the electrophoresis female kittiwakes' DNA will yield two bands and the males' one band. This occurs because the female is heterogametic (ZW) and the male homogametic (ZZ) (Griffiths et al., 1998).

DNA was extracted from whole blood which was dissolved in alcohol prior to the extraction. A variable portion of blood (2-4 µL) was pipetted into an Eppendorf tube containing 200 µL 5% Chelex (Bio-Rad Laboratories Inc., Hercules, CA, USA), a chelating agent. The tubes were subsequently incubated twice; first at 56 °C for 20 minutes and then at 96 °C for exactly 8 minutes. The samples were centrifuged for 3 minutes at 12000 rpm and 20 µL of the supernatant was pipetted into a clean tube. Then 8 µL of PCR stock (in the proportion of 0.05 µL Taq, 1.95 µL autoclaved H_2O, 0.40 µL dNTP Mix, 0.60 µL MgCl, 1.00 µL 10X, 1.00 µL 10 µM primer 2718, 1.00 µL 10 µM primer 2550, 2.00 µL Q-solution, Invitrogen, Thermo Fisher Scientific Inc., Waltham, MA, USA) was added to the wells of the PCR plate. To each well 2.0 µL DNA was added. The temperature cycle used was 94 °C for 30 seconds, 46 °C for 45 seconds, 70 °C for 45 seconds for a total of 35 cycles. Finally the temperature was maintained at 70 °C for 10 minutes followed by 4 °C indefinitely using the GeneAmp PCR System 9700 (Thermo Fisher Scientific Inc., Waltham, MA, USA). Following PCR the products were pipetted into a prepared 1% agarose gel containing ethidium bromide. The gel ran at 75 V for 45-50 minutes after which it was placed underneath an ultraviolet light for the bands to be read (Figure 2).

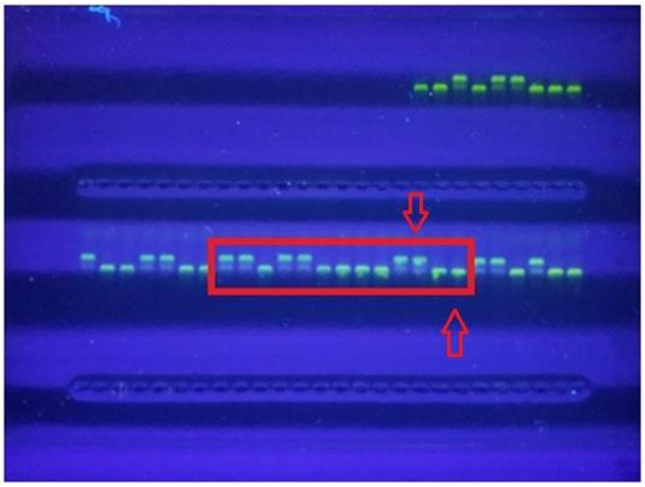

Figure 2: A photograph of the 1% agarose gel with DNA products extracted from the red blood cells of black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard in 2013 and 2014. The DNA was amplified using PCR. Two bands indicates a female (marked by the downward-pointing arrow) and one band indicates a male (marked by the upward-pointing arrow). Bands within the rectangle represent 13 of the individuals included in this study.

Most of the arctic skuas had been caught and sampled in previous years and thus their sexes had been determined molecularly. However, for two individuals (caught in 2014) their sexes had not been confirmed by molecular sexing, and their sexes were determined biometrically since females are larger than the males (Catry et al., 1999). When a pair of breeding arctic skuas were caught, the larger of the two was assumed to be the female and the smaller one the male.

2.6 Statistical Analyses

From a total of 60 kittiwakes caught, four individuals were sampled in both years. All of the four repeated individuals were females and for each individual all its observations from one year were deleted. To avoid introducing bias into the dataset, the year from which the observations were excluded was selected at random. It was important to have only independent data as some of the statistical methods employed assume that the data is independent.

Furthermore, one male and one female kittiwake were excluded from the TT4 data sets due to too high variance (>15%). This means that the final sample size for male kittiwakes is 30 individuals for biometric variables and PFAS concentrations, and 29 individuals for TT4 levels. For female kittiwakes the final sample size is 26 individuals for biological variables and all PFASs except brPFOS (where $n = 14$), and 25 individuals for TT4 levels.

For arctic skuas, one male was excluded from the TT3 data set due to too high variance (>15%). As a result the sample size is nine for biological variables, PFAS and TT4 concentrations, and eight for TT3 concentrations.

As for the female arctic skuas, two individuals were excluded from both the TT3 and TT4 data sets due to too high variances (>15%), and the final sample size is therefore five for THs and seven for biological variables and PFASs.

Statistical analyses were performed using SPSS (version 21.0, IBM, SPSS Inc., Chicago, IL, USA) and SigmaPlot (version 12.0, Cranes Software, Chicago, IL, USA).

A statistical significance level of $p < 0.05$ was set. When testing for sex, year, and location differences, data that passed Shapiro-Wilk's test for normality and Levene's test for homogeneity were tested using one-way ANOVA (analysis of variance) and Tukey's post hoc test. If the data failed one or both of the aforementioned tests, the data was tested using Kruskal-Wallis one-way analysis of variance on ranks (hereafter Kruskal-Wallis) and Dunn's post hoc test.

2.6.1 Data Below the Limit of Detection

A minimum of 70% of the individuals had to have PFAS concentrations above the instrument's limit of detection (LOD) in order for the PFAS to be included in the statistical analyses. This eliminated PFHxA, PFHpA, PFOA, PFHxS, and PFDS in both kittiwakes and arctic skuas. In kittiwakes FOSA was below the LOD for all samples. The substance brPFOS was excluded in the kittiwake females from 2013 and all the arctic skuas. In arctic skuas PFTeDA was also rejected. If less than 30% of the individuals had concentrations below LOD, those that were below LOD were replaced with a randomly selected concentration between 0 and the LOD of the PFAS. The LOD was < 0.05 ng/mL for all compounds, except FOSA where it was < 0.10 ng/mL. The limit of quantification (LOQ) was set to be 3 times LOD.

2.6.2 Body Condition

The body condition (BC) of each individual bird was determined by using the biometric variable - length of skull, wing, or tarsus - that was most correlated to body mass. In both male and female kittiwakes body mass was most strongly correlated to the length of tarsus. This was also the case for male arctic skuas, but in female arctic skuas it was the skull length which was most closely associated with body mass. The standardized residuals of linear regression plots of either tarsus or skull length as the independent variable against body mass as the dependent variable was determined to be the body condition.

2.6.3 Year and Location Differences in Perfluoroalkyl and Polyfluoroalkyl Substance and Thyroid Hormone Concentrations

The kittiwake datasets were tested for differences between the two years (2013 and 2014). The arctic skua datasets were tested for differences between the two locations (Brensholmen

and Kongsfjorden). These tests were performed to examine if the data from the two years and locations could be pooled for the respective species.

2.6.4 Correlation Matrices and Linear Regression

Correlation matrices were calculated for the finalized datasets using PFAS concentrations in pmol/mL instead of ng/mL. That is preferable to ng/mL as it may be the amount of PFAS that determines the effect and not the total mass of the substance. However, correlation matrices based on PFAS concentrations in ng/mL yielded the same results. Pearson's coefficient, *r*, was used as the coefficient of correlation along with *p* for significant results.

Based on the results from the correlation matrices, statistically significant correlations were examined using linear regression. Due to some low sample sizes the adjusted R^2 is reported rather than the R^2 as the adjusted R^2 takes into account sample size (Leach and Henson, 2007; Ivarsson et al., 2013). The adjustment formula employed by SPSS is the following:

$$Adjusted\ R^2 = 1 - \left(\frac{n-1}{n-k-1}\right)(1 - R^2) \qquad \text{(Equation 2)}$$

Where *n* is the sample size and *k* is the number of predictor variables.

In order to test the assumption of heteroscedasticity of the data in linear regression, three plots were calculated and examined visually for each model. Specifically, the plots calculated were a histogram of the standardized residuals of the dependent variable, a normal P-P plot of the standardized residuals with the observed cumulative against the expected cumulative probability, and finally a scatterplot of the regression standardized predicted value against the regression standardized residual.

2.6.5 Confounding Factors

To determine if the percentage of fat in the plasma and whole blood (hereafter percentage of fat in blood), body condition, or body mass, were confounding variables in the linear regression, a multiple regression was performed where the suspected confounding variables were included. If the R value changed by less than 10% when the suspected confounding variable was removed, it was deemed to not be a significant confounding variable.

3. Results

3.1 Year and Location Differences in Perfluoroalkyl and Polyfluoroalkyl Substance and Thyroid Hormone Concentrations

For TT3 a statistically significant difference was found when comparing levels between female kittiwakes caught in 2013 and 2014. The female birds sampled in 2014 had significantly higher TT3 concentrations than the female birds sampled in 2013 (ANOVA and Tukey's post hoc test, $p = 0.003$). No significant differences were found between TT4 concentrations in male and female kittiwakes from 2013 compared to kittiwakes sampled in 2014. In arctic skuas, there were no significant differences between individuals sampled on Brensholmen and in Kongsfjorden for either TT3 (ANOVA, $p = 0.097$) or TT4 (ANOVA, $p = 0.095$).

For kittiwakes, PFNA levels were significantly higher in males and females sampled in 2013 than 2014 (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$). Levels of PFDoDA, PFTrDA, and PFTeDA were significantly higher in males and females sampled in 2014 than in 2013 (ANOVA and Tukey's post hoc test, $p < 0.001$, $p < 0.001$ for PFDoDA, PFTrDA, Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$ for PFTeDA).

In arctic skuas only PFNA was significantly higher in birds sampled in Kongsfjorden compared to individuals sampled on Brensholmen (Kruskal-Wallis and Dunn's post hoc test, $p = 0.020$).

Although there were some differences in concentrations between years and locations, the PFASs' mechanisms of action will not differ. Thus, the data sets were pooled to obtain a larger sample size and variation in PFAS and TH concentrations which in turn granted a greater statistical power for the linear regression.

3.2 Black-Legged Kittiwakes

3.2.1 Biological Variables

The mean, standard deviation (SD), median, range, sample size of body condition (BC), and other biometric variables are presented in Table 2. The individual biometric data is located in Appendix B.

Except for BC, the other variables were all significantly higher in male than female kittiwakes (skull: Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$; wing: ANOVA and Tukey's post hoc test, $p = 0.001$; tarsus: Kruskal-Wallis and Dunn's post hoc test, $p = 0.048$; body mass: ANOVA and Tukey's post hoc test, $p < 0.001$).

Table 2: Summary of biometric data for male and female black-legged kittiwakes (*Rissa tridactyla*) sampled in Kongsfjorden, Svalbard, in 2013 and 2014. Data is summarized as mean ± standard deviation (SD), median, range, *n* denotes the sample size, and body condition is abbreviated BC.
*Variables are significantly higher in male than female kittiwakes (skull: Kruskal-Wallis and Dunn's, $p < 0.001$; wing: ANOVA and Tukey's, $p = 0.001$; tarsus: Kruskal-Wallis and Dunn's, $p = 0.048$; mass: ANOVA and Tukey's, $p < 0.001$).

	Males				Females			
	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
Skull (mm)*	94.0 ± 1.98	93.7	91.6 - 98.6	30	88.8 ± 2.11	88.5	85.6 - 94.4	26
Wing (mm)*	318 ± 6.59	320	306 - 335	30	313 ± 6.46	312	298 - 322	26
Tarsus (mm)*	34.9 ± 1.46	34.9	30.6 - 37.6	30	34.1 ± 1.51	34.3	29.6 - 35.9	26
Mass (g)*	434 ± 34.8	433	345 - 495	30	394 ± 24.0	398	340 - 435	26
BC	0.00 ± 0.983	-0.115	-2.38 - 1.61	30	0.00 ± 0.980	0.231	-2.86 - 1.37	26

3.2.2 Perfluoroalkyl and Polyfluoroalkyl Substance Concentrations

A summary of the PFAS levels in kittiwakes are shown in Table 3. The summarized and individual PFAS concentrations in ng/mL are located in Appendix B.

There were significantly higher levels in males than females for linPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), sumPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), PFUnDA (ANOVA and Tukey's post hoc test, $p = 0.025$), PFDoDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.045$), PFTrDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.003$), and PFTeDA (ANOVA and Tukey's post hoc test, $p = 0.004$).

Table 3: A summary of the concentrations (pmol/mL plasma) of perfluoroalkyl and polyfluoroalkyl substances (PFASs) detected in male and female black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. The data is presented as mean ± standard deviation (SD), median, and range. *n* denotes the sample size.
*Male kittiwakes had significantly higher levels than females of linPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), sumPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), PFUnDA (ANOVA and Tukey's post hoc test, $p = 0.025$), PFDoDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.045$), PFTrDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.003$), and PFTeDA (ANOVA and Tukey's post hoc test, $p = 0.004$).

	Males				Females			
PFAS (pmol/mL)	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
brPFOS	0.360 ± 0.376	0.226	0.0200 - 1.34	30	0.197 ± 0.0969	0.182	0.0400 - 0.328	14
linPFOS*	14.7 ± 7.16	12.9	4.94 - 38.7	30	8.68 ± 6.29	7.20	1.87 - 29.2	26
sumPFOS*	15.0 ± 7.45	13.0	4.99 - 39.9	30	8.94 ± 6.51	7.40	1.87 - 30.0	26
PFNA	3.30 ± 2.36	2.62	0.812 - 12.1	30	2.69 ± 1.61	2.21	0.895 - 7.20	26
PFDA	2.51 ± 1.01	2.34	0.764 - 5.16	30	2.15 ± 0.853	2.08	0.701 - 4.05	26
PFUnDA*	12.3 ± 4.29	10.9	6.15 - 21.6	30	9.97 ± 3.09	9.72	4.38 - 16.8	26
PFDoDA*	2.58 ± 1.16	2.66	0.699 - 4.81	30	1.89 ± 0.856	1.98	0.258 - 3.36	26
PFTrDA*	7.92 ± 2.73	7.80	3.35 - 12.7	30	5.52 ± 2.10	5.80	1.80 - 9.12	26
PFTeDA*	0.923 ± 0.555	0.914	0.118 - 2.29	30	0.520 ± 0.408	0.584	0.0140 - 1.412	26

Figure 3 shows the relative concentration of PFASs in males and females, where the mean concentration of the respective PFASs in males were divided by the corresponding mean value in females. Although not statistically significant for all PFASs (see Table 3), it appears that male kittiwakes had a consistently higher burden than females of all detected PFASs.

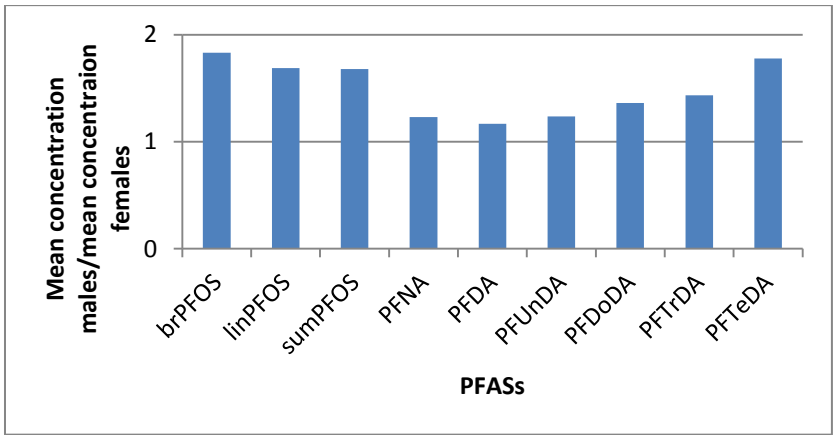

Figure 3: The proportions of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) between male and female black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. The mean concentration detected in male kittiwakes for respective PFASs was divided by the mean concentration in female kittiwakes. As the bar is above 1 for all compounds, it suggests that the PFAS burden in males was larger than the PFAS burden for females.

The contribution of the respective PFASs in pmol/mL plasma on the total PFAS load was also examined for male and female kittiwakes (Figure 4). All PFASs except sumPFOS were included as it is sum of branched and linear PFOS.

The most obvious difference is that the most abundant PFAS in males was linPFOS compared to PFUnDA in females. Although it was only in females that the levels of PFUnDA were significantly higher than those of linPFOS (Kruskal-Wallis and Dunn's post hoc test, $p = 0.002$). In male kittiwakes there were no significant difference between concentrations of linPFOS and PFUnDA (Kruskal-Wallis, $p = 0.906$). In males PFDoDA was present at a higher level than PFDA whereas in females it was PFDA which is more abundant than PFDoDA. However, the differences in concentration between PFDA and PFDoDA was not statistically significant in either sex (males; Kruskal-Wallis, $p = 0.132$, and females; ANOVA, $p = 0.673$).

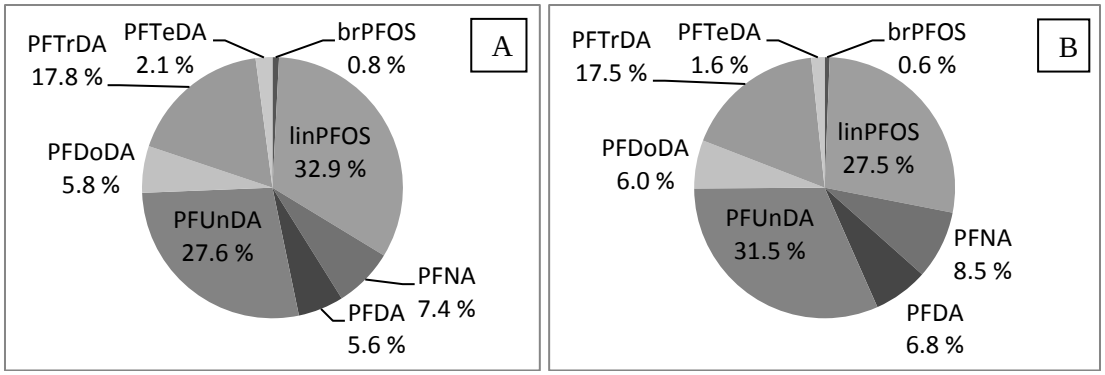

Figure 4: The relative abundance of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in male (chart A) and female (chart B) black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. Pie charts are based on PFAS concentrations in pmol/mL plasma. The concentration of PFUnDA was significantly higher than the concentration of linPFOS in females (Kruskal-Wallis and Dunn's post hoc test, $p = 0.002$), but there was no significant difference in levels of linPFOS and PFUnDA in male kittiwakes. Aside from that, the proportions of the PFAS load tended to be the same for both sexes.

3.2.3 Thyroid Hormones

The mean, SD, median, range, and sample size of TT3 and TT4 concentrations are presented in Table 4. For individual thyroid hormone measurements refer to Appendix B.
No significant differences were detected between TT3 or TT4 concentrations in male and female kittiwakes (Kruskal-Wallis, $p = 0.565$; ANOVA, $p = 0.839$, respectively).

Table 4: Summary of the mean ± standard deviation (SD), median, and range in concentrations of total triiodothyronine (TT3) and total thyroxine (TT4) in black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. *n* denotes the sample size. No significant differences in concentrations between males and females were detected for either hormone.

	Males				Females			
	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
TT3 (ng/mL)	2.78 ± 0.713	2.68	1.75 - 4.59	30	2.96 ± 1.08	3.21	1.12 - 4.49	26
TT4 (ng/mL)	36.45 ± 11.2	36.89	18.61 - 70.05	29	37.18 ± 14.9	34.39	9.53 - 79.83	25

3.2.4 Linear Regression

3.2.4.1 Correlation Matrices

Correlation matrices were calculated using the Pearson correlation coefficient, *r*, and linear regressions were performed on the results they yielded (Table 5 for associations with thyroid hormones and Table 6 for associations with body condition and mass). Neither BC nor body mass were significantly associated with the thyroid hormones.

Table 5: Results from the correlation matrix for perfluoroalkyl and polyfluoroalkyl substances (PFASs) and total triiodothyronine (TT3) and total thyroxine (TT4) in black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. *r* is the Pearson correlation coefficient and *p* is the significance level. Only significant associations are shown.

	Males		Females			
	TT4		TT4		TT3	
PFAS	*r*	*p*	*r*	*p*	*r*	*p*
brPFOS	-	-	0.565	0.035	-	-
linPFOS	0.554	0.002	-	-	-	-
sumPFOS	0.549	0.002	-	-	-	-
PFDA	0.473	0.009	-	-	-	-
PFUnDA	0.421	0.023	-	-	-	-
PFDoDA	-	-	-	-	0.496	0.01
PFTrDA	-	-	-	-	0.527	0.006
PFTeDA	-	-	-	-	0.479	0.013

Table 6: Results from the correlation matrix for perfluoroalkyl and polyfluoroalkyl substances (PFASs), body condition (BC), and body mass in black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. *r* is the Pearson correlation coefficient and *p* is the significance level. Only significant associations are shown.

	Males				Females			
	BC		Body mass		BC		Body mass	
PFAS	*r*	*p*	*r*	*p*	*r*	*p*	*r*	*p*
linPFOS	-	-	-	-	-0.586	0.002	-0.513	0.007
sumPFOS	-	-	-	-	-0.580	0.002	-0.506	0.008
PFNA	-	-	-	-	-0.643	<0.001	-0.527	0.006
PFDA	-	-	-	-	-0.434	0.027	-0.415	0.035
PFDoDA	0.520	0.003	0.530	0.003	-	-	-	-
PFTrDA	0.594	0.001	0.583	0.001	-	-	-	-
PFTeDA	0.668	<0.001	0.656	<0.001	-	-	-	-

3.2.4.2 Confounding Factors

For all relationships tested, the percentage of fat in the blood only had an effect on the association between TT4 and brPFOS in female kittiwakes. For all other associations, the changes in the R value were below 10%, and thus the effects of BC, body mass, and percentage of fat in the blood were deemed insignificant.

3.2.4.3 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances and Thyroid Hormones

In male kittiwakes only TT4 were found to be correlated with PFASs (Figure 5). Concentrations of TT4 were significantly correlated to concentrations of linPFOS (R: 0.554, $p = 0.002$), sumPFOS (R: 0.549, $p = 0.002$), PFDA (R: 0.473, $p = 0.009$), and PFUnDA (R: 0.421, $p = 0.023$).

In female kittiwakes the only PFAS correlated with TT4 was brPFOS (R: 0.565, $p = 0.035$). However, TT3 was correlated with PFDoDA (R: 0.496, $p = 0.010$), PFTrDA (R: 0.527, $p = 0.006$), and PFTeDA (R: 0.479, $p = 0.013$) (Figure 6).

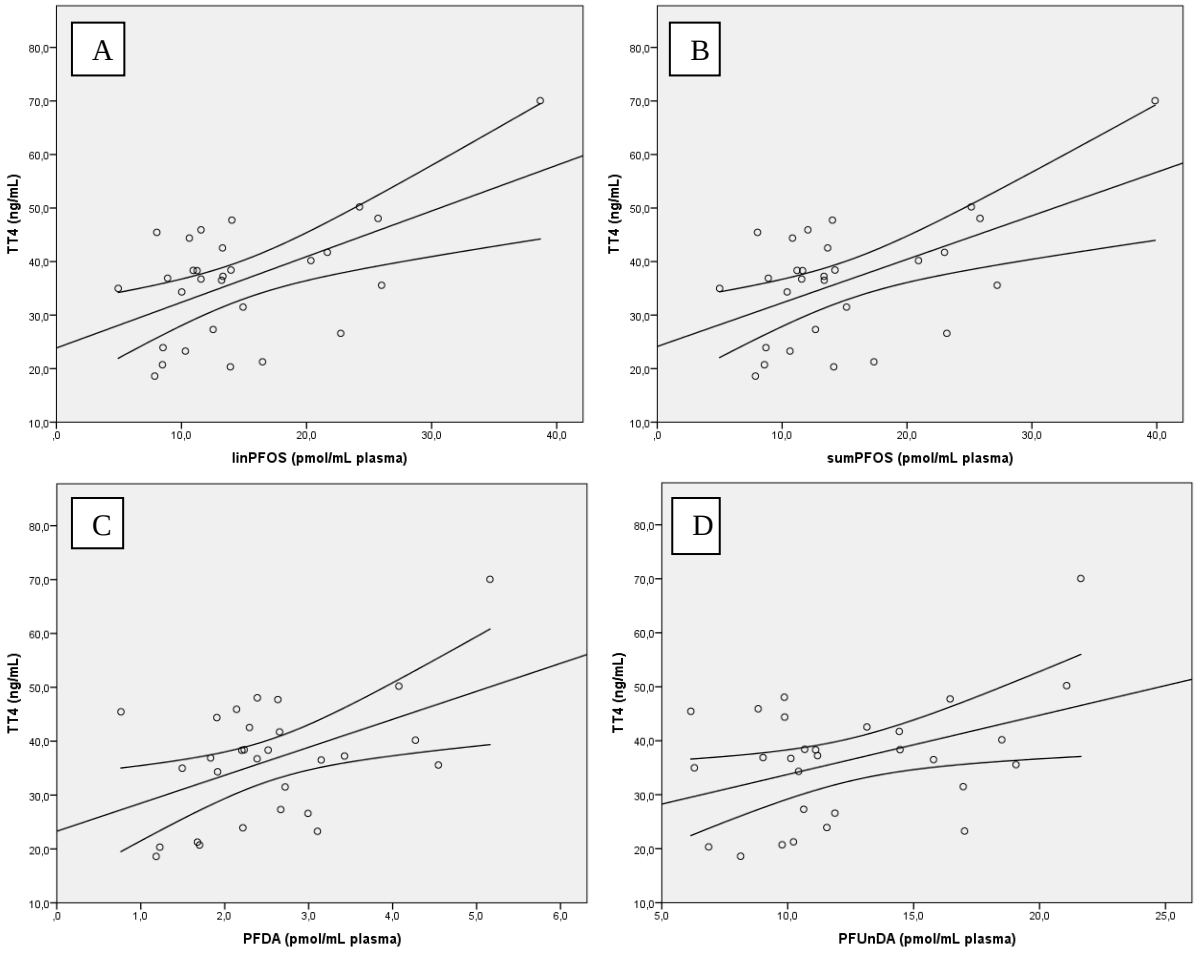

Figure 5: Linear regression (± 95% confidence interval) of total thyroxine (TT4, ng/mL plasma) against perfluoroalkyl and polyfluoroalkyl substances (PFASs, pmol/mL plasma) in male black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. $n = 29$.
A: Adjusted R^2: 0.218, $p = 0.002$. B: Adjusted R^2: 0.276, $p = 0.002$. C: Adjusted R^2: 0.195, $p = 0.009$.
D: Adjusted R^2: 0.146, $p = 0.023$.

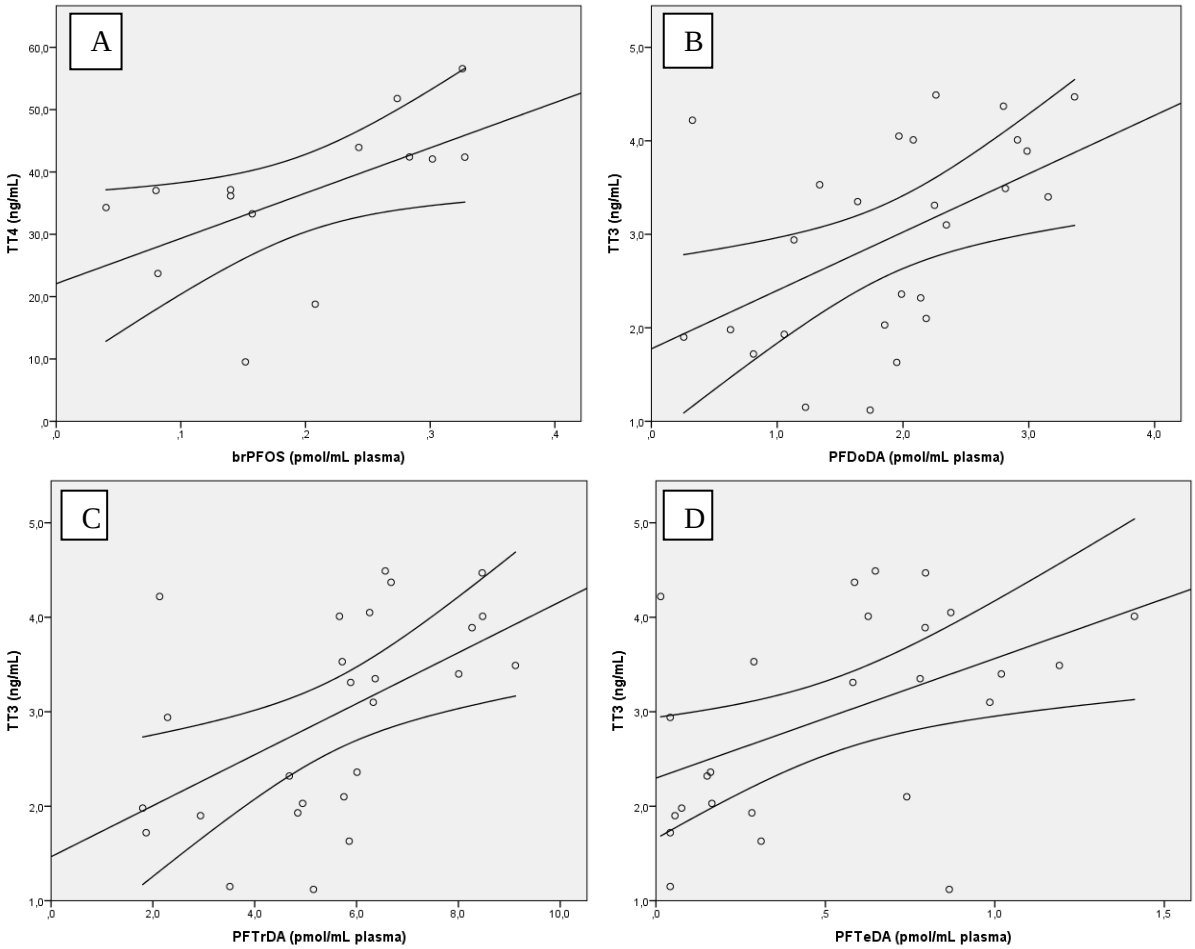

Figure 6: Linear regression (± 95% confidence interval) of total thyroxine (TT4, ng/mL plasma) against perfluoroalkyl and polyfluoroalkyl substances (PFASs, pmol/mL plasma) in female black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. n = 14 for brPFOS and n = 26 for PFDoDA, PFTrDA, and PFTeDA.
A: Adjusted R^2: 0.263, p = 0.035. B: Adjusted R^2: 0.215, p = 0.010. C: Adjusted R^2: 0.247, p = 0.006.
D: Adjusted R^2: 0.197, p = 0.013.

3.2.4.4 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances and Body Condition

Body condition was found to have a positive correlation with PFDoDA (R: 0.520, p = 0.003), PFTrDA (R: 0.594, p = 0.001), and PFTeDA (R: 0.668, p < 0.001) in male kittiwakes (Figure 7). The BC in female kittiwakes, however, was negatively correlated to linPFOS (R: -0.586, p = 0.002), sumPFOS (R: -0.580, p = 0.002), PFNA (R: -0.643, p < 0.001), and PFDA (R: -0.434, p = 0.027) (Figure 8).

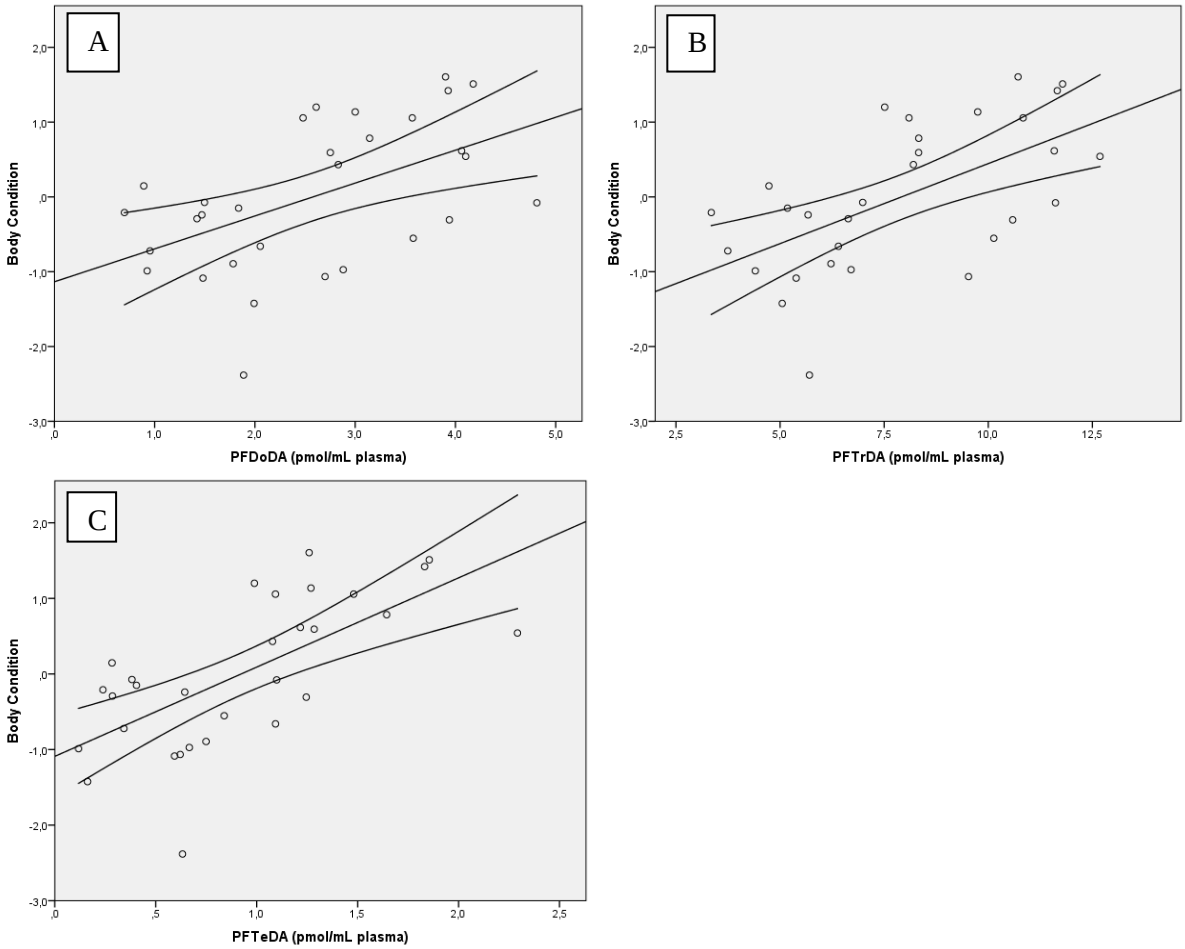

Figure 7: Linear regression (± 95% confidence interval) with body condition against perfluoroalkyl and polyfluoroalkyl substances (PFASs) in male black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. $n = 30$.
A: Adjusted R^2: 0.244, $p = 0.003$. B: Adjusted R^2: 0.330, $p = 0.001$. C: Adjusted R^2: 0.426, $p < 0.001$.

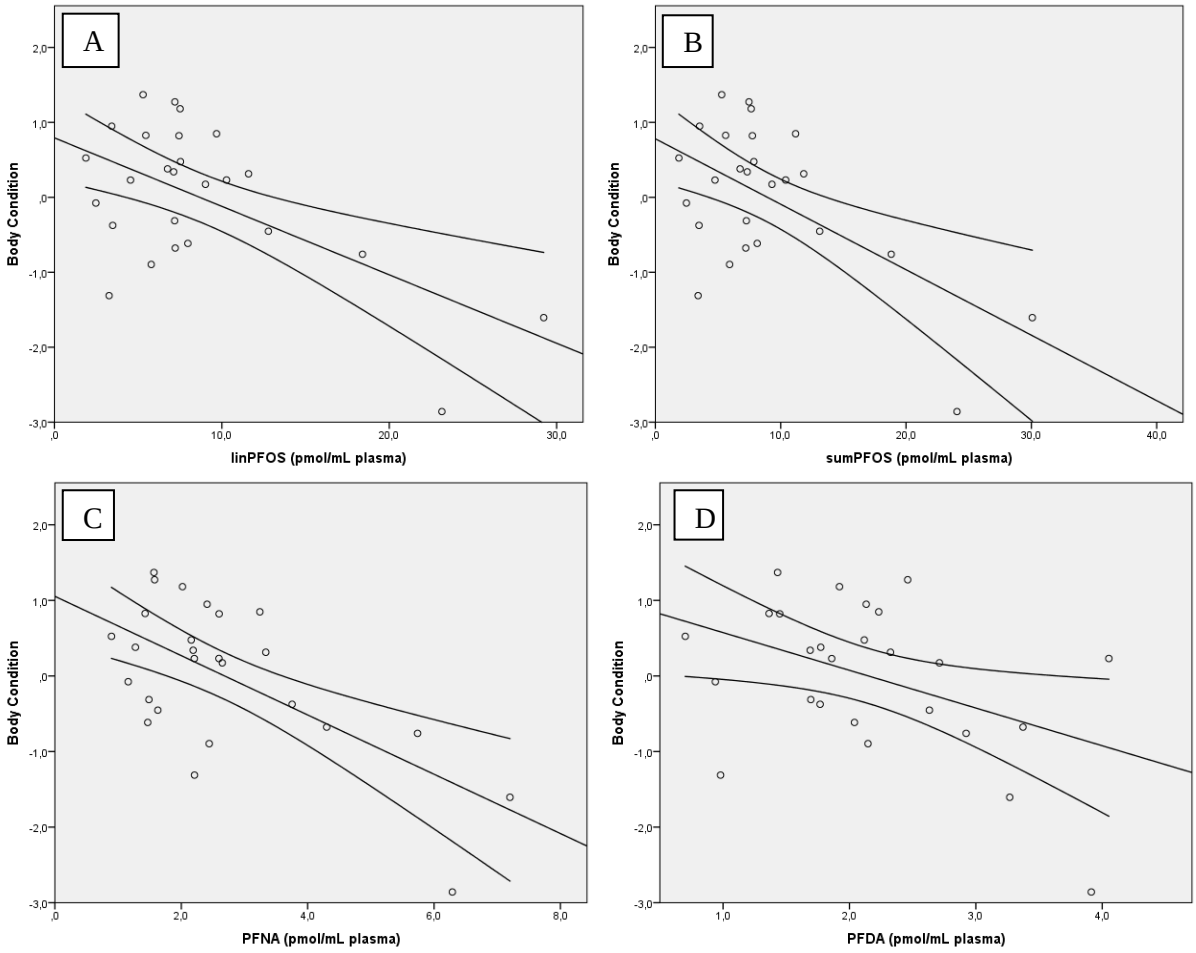

Figure 8: Linear regression (± 95% confidence interval) with body condition against perfluoroalkyl and polyfluoroalkyl substances (PFASs) in female black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard, in 2013 and 2014. $n = 26$.
A: Adjusted R^2: 0.316, $p = 0.002$. B: Adjusted R^2: 0.309, $p = 0.002$. C: Adjusted R^2: 0.388, $p < 0.001$.
D: Adjusted R^2: 0.155, $p = 0.027$.

3.3 Arctic Skuas

3.3.1 Biological Variables

The mean, SD, median, range, sample size of BC, and other biometric variables are presented in Table 7. The individual biometric data is located in Appendix C.

Body mass was the only biometric variable with a significant difference between male and female arctic skuas with females being heavier than males (ANOVA and Tukey's post hoc test, $p < 0.001$).

Table 7: Summary of biometric data for male and female arctic skuas (*Stercorarius parasiticus*) sampled on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. Data is summarized as mean ± standard deviation (SD), median, range, *n* denotes the sample size, and body condition is abbreviated BC.
*Females weighed significantly more than males (ANOVA and Tukey's post hoc test, $p < 0.001$).

	Males				Females			
	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
Skull (mm)	77.2 ± 1.44	76.8	75.1 - 80.0	9	77.8 ± 3.25	77.7	73.5 - 81.6	7
Wing (mm)	335 ± 6.91	338	325 - 344	9	342 ± 10.1	342	331 - 359	7
Tarsus (mm)	45.4 ± 1.38	45.2	43.9 - 47.7	9	45.7 ± 1.28	45.6	43.4 - 47.2	7
Mass (g)*	427 ± 34.6	425	375 - 472	9	500 ± 27.1	499	445 - 527	7
BC	0.00 ± 0.935	-0.204	-1.07 - 1.99	9	0.00 ± 0.913	-0.0196	-1.46 - 1.08	7

3.3.2 Perfluoroalkyl and Polyfluoroalkyl Substance Concentrations

A summary of the PFAS levels in arctic skuas are shown in Table 8. The summarized and individual PFAS concentrations in ng/mL are located in Appendix C.

Male arctic skuas had significantly higher levels than females of linPFOS, sumPFOS, PFDA, PFUnDA, PFDoDA, and PFTrDA (ANOVA and Tukey's post hoc test, $p = 0.008$, 0.010, 0.005, 0.008, 0.010, and 0.005 respectively). It was only PFNA and FOSA which had similar concentrations in male and female arctic skuas.

Table 8: A summary of the concentrations (pmol/mL whole blood) of perfluoroalkyl and polyfluoroalkyl substances (PFASs) detected in male and female arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. The data is presented as mean ± standard deviation (SD), median, and range. *n* denotes the sample size.
*Male arctic skuas had significantly higher levels than females of linPFOS, sumPFOS, PFDA, PFUnDA, PFDoDA, and PFTrDA (ANOVA and Tukey's post hoc test, $p = 0.008$, 0.010, 0.005, 0.008, 0.010, and 0.005 respectively).

	Males				Females			
PFAS (pmol/ml)	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
linPFOS*	20.0 ± 7.78	20.4	8.59 - 34.2	9	9.90 ± 4.44	11.1	3.57 - 15.6	7
sumPFOS*	20.8 ± 8.49	21.7	8.59 - 34.9	9	10.2 ± 4.67	11.4	3.57 - 16.2	7
PFNA	1.99 ± 2.87	1.21	0.0431 - 9.39	9	1.28 ± 0.806	1.66	0.0216 - 2.03	7
PFDA*	2.91 ± 0.787	2.72	2.01 - 4.57	9	1.63 ± 0.752	1.54	0.702 - 2.77	7
PFUnDA*	12.4 ± 3.40	12.4	7.86 - 17.5	9	7.30 ± 3.02	7.62	2.49 - 11.0	7
PFDoDA*	2.63 ± 0.725	2.65	1.23 - 3.28	9	1.30 ± 0.698	1.21	0.289 - 2.26	7
PFTrDA*	5.24 ± 1.81	5.56	2.85 - 8.08	9	2.64 ± 1.06	2.76	0.877 - 3.98	7
FOSA	2.90 ± 2.09	2.48	0.100 - 5.56	9	1.33 ± 0.431	1.15	0.948 - 2.16	7

Figure 9 shows the relative concentration in males and females, where the mean concentration of the respective PFASs in males were divided by the corresponding mean value in females.

Although not statistically significant for all PFASs (see Table 8), it appears that male arctic skuas had a consistently higher burden than females of all detected PFASs.

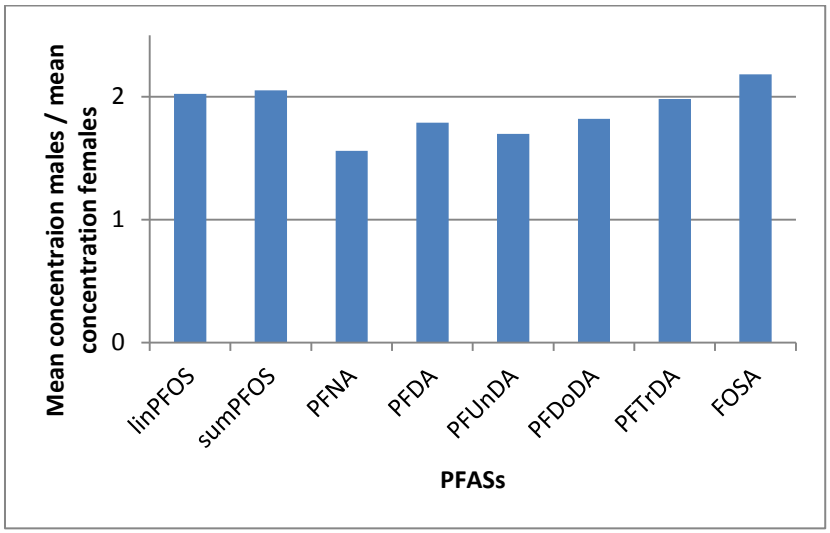

Figure 9: The proportions of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) between male and female arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. The mean concentration detected in male arctic skuas for respective PFASs was divided by the mean concentration in female arctic skuas. As the bar is above 1 for all compounds, it suggests that the PFAS burden in males was larger than the PFAS burden for females.

The contribution of the respective PFASs in pmol/mL whole blood on the total PFAS load was also examined for male and female arctic skuas (Figure 10). All PFASs except for sumPFOS were included as it is sum of branched and linear PFOS.

For both males and females, linPFOS was more abundant than PFUnDA. However, it was only significantly higher in males (Kruskal-Wallis and Dunn's post hoc test, $p = 0.024$). In males PFDA and FOSA were equally abundant, whereas in females PFDA was slightly more abundant than FOSA, although it was not significant (ANOVA, $p = 0.380$).

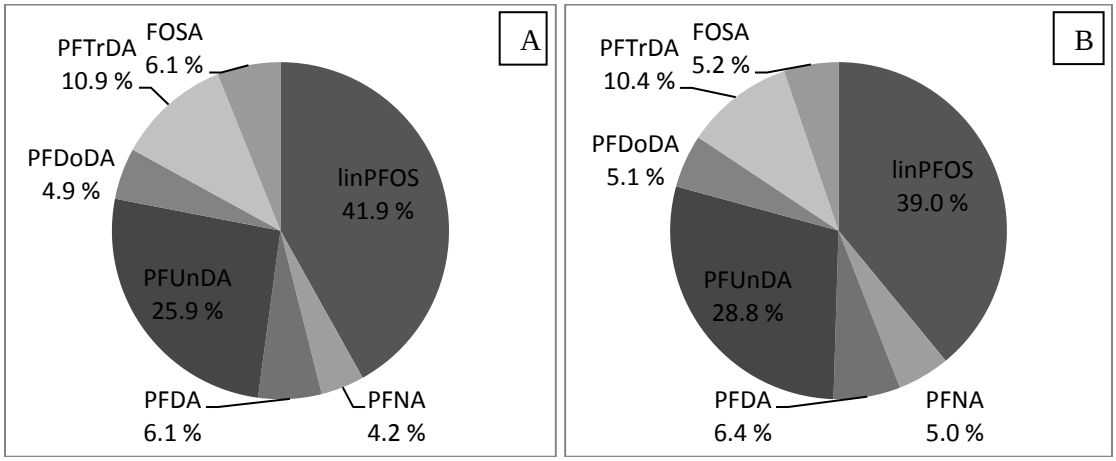

Figure 10: The relative abundance of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in male (chart A) and female (chart B) arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. Pie charts are based on PFAS concentrations in pmol/mL whole blood. Females had significantly higher levels of linPFOS than PFUnDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.024$), while in males there was no significant difference between linPFOS and PFUnDA. Aside from that, the proportions of the PFAS load tended to be similar in both sexes.

3.3.3 Thyroid Hormones

The mean, SD, median, range, and sample size of TT3 and TT4 concentrations are presented in Table 9. For individual thyroid hormone measurements refer to Appendix C.

There was no significant difference in TT3 concentration between male and female arctic skuas (ANOVA, $p = 0.114$). Nor were there significant differences in TT4 levels between males and females (ANOVA, $p = 0.095$).

Table 9: Summary of the mean ± standard deviation (SD), median, and range in concentrations of total triiodothyronine (TT3) and total thyroxine (TT4) in arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. *n* denotes the sample size. No significant differences in hormone levels were detected.

	Males				Females			
	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
TT3 (ng/mL)	3.22 ± 0.884	3.16	2.18 - 4.99	8	2.42 ± 0.687	2.36	1.61 - 3.20	5
TT4 (ng/mL)	46.65 ± 16.9	41.28	25.02 - 80.18	9	30.59 ± 13.8	31.94	11.21 - 46.68	5

3.3.4 Linear Regression

3.3.4.1 Correlation Matrices

Correlation matrices were calculated using the Pearson correlation coefficient, *r*, and linear regressions were performed on the results they yielded (Table 10 for associations with thyroid hormones and Table 11 for associations with body mass). All PFAS concentrations are in pmol/mL whole blood. Neither BC nor body mass were significantly associated with the thyroid hormones.

Table 10: Results from the correlation matrix for perfluoroalkyl and polyfluoroalkyl substances (PFASs) and total thyroxine (TT4) in arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. *r* is the Pearson correlation coefficient and *p* is the significance level. Only significant associations are shown.

	Males		Females	
	TT4		TT4	
PFAS	*r*	*p*	*r*	*p*
linPFOS	-	-	0.905	0.035
sumPFOS	-	-	0.901	0.037
PFUnDA	0.793	0.011	-	-
PFDoDA	0.846	0.004	-	-
FOSA	0.675	0.046	-	-

Table 11: Results from the correlation matrix for perfluoroalkyl and polyfluoroalkyl substances (PFASs) and body mass in male arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. *r* is the Pearson correlation coefficient and *p* is the significance level. Only significant associations are shown.

	Males	
	Body mass	
PFAS	*r*	*p*
PFDA	-0.710	0.032
PFUnDA	-0.687	0.041
PFTrDA	-0.731	0.025
FOSA	-0.734	0.024

3.3.4.2 Confounding Factors

The percentage of fat in the blood was only obtainable for female arctic skuas, and so only the effects of BC and body mass were tested in the models for male arctic skuas. They had no significant effects on the linear regressions. For female arctic skuas, the percentage of fat in the blood, BC, and body mass were tested. All changes in the R values were below 10% and thus deemed insignificant.

3.3.4.3 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances and Thyroid Hormones

Only TT4 had any significant associations with PFASs in both sexes. In male arctic skuas, TT4 was positively associated with PFUnDA (R: 0.793, $p = 0.011$), PFDoDA (R: 0.846, $p = 0.004$), and FOSA (R: 0.675, $p = 0.046$) (Figure 11). Whereas only linPFOS (R: 0.905, $p = 0.035$) and sumPFOS (R: 0.901, $p = 0.037$) were positively associated with TT4 in female arctic skuas (Figure 12).

3.3.4.4 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances and Body Mass

Significant associations between body mass and PFASs were only found in male arctic skuas (Figure 13). Body mass was negatively associated with PFDA (R: -0.710, $p = 0.032$), PFUnDA (R: -0.687, $p = 0.041$), PFTrDA (R: -0.731, $p = 0.025$), and FOSA (R: -0.734, $p = 0.024$).

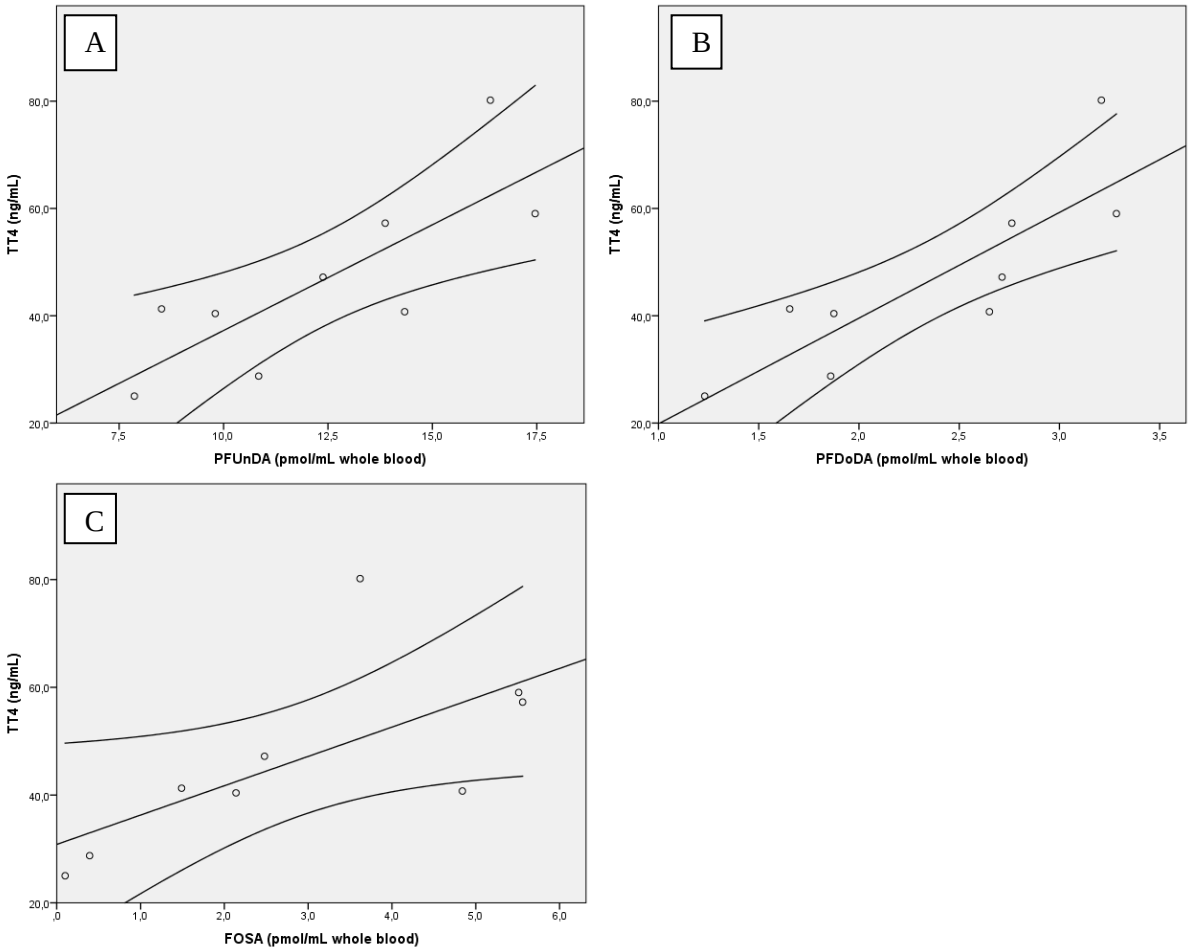

Figure 11: Linear regression (± 95% confidence interval) with total thyroxine (TT4, ng/mL plasma) against perfluoroalkyl and polyfluoroalkyl substances (PFASs) in male arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. n = 9.
A: Adjusted R^2: 0.575, p = 0.011. B: Adjusted R^2: 0.675, p = 0.004. C: Adjusted R^2: 0.377, p = 0.046.

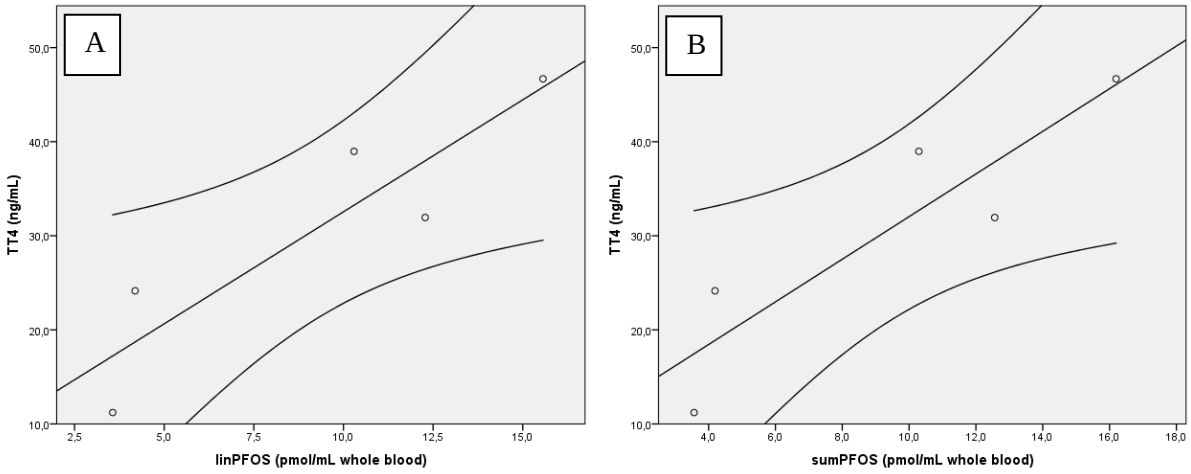

Figure 12: Linear regression (± 95% confidence interval) with total thyroxine (TT4, ng/mL plasma) against perfluoroalkyl and polyfluoroalkyl substances (PFASs) female arctic skuas (*Stercorarius parasiticus*), caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. n = 5.
A: Adjusted R^2: 0.758, p = 0.035. B: Adjusted R^2: 0.748, p = 0.037.

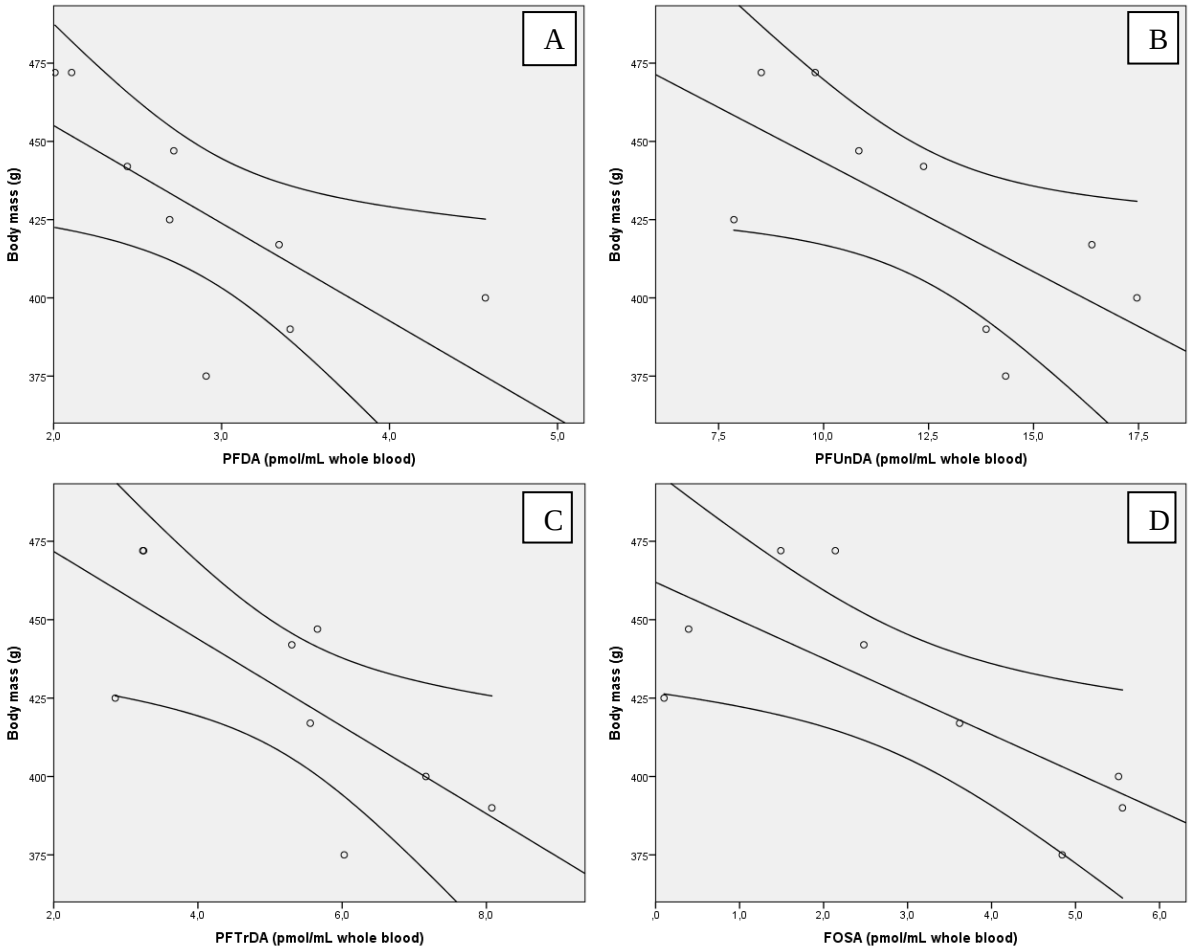

Figure 13: Linear regression (± 95% confidence interval) with body mass against perfluoroalkyl and polyfluoroalkyl substances (PFASs) in male arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. $n = 9$.
A: Adjusted R^2: 0.433, $p = 0.032$. B: Adjusted R^2: 0.396, $p = 0.041$. C: Adjusted R^2: 0.467, $p = 0.025$.
D: Adjusted R^2: 0.474, $p = 0.024$.

3.4 Comparison between Black-Legged Kittiwakes and Arctic Skuas

The differences in PFAS concentrations between kittiwakes and arctic skuas do not lend themselves to a direct comparison due to the two different sample matrices used in this study (plasma and whole blood). But when visually comparing the mean and standard deviation of PFAS concentrations summarized in Table 12, male arctic skuas appear to have had a higher concentration of linPFOS and sumPFOS than male kittiwakes. Both male and female kittiwakes seem to have had higher levels of PFNA and PFTrDA than male and female arctic skuas. As brPFOS and PFTeDA were excluded for arctic skuas and FOSA was not detected in any kittiwakes, these three compounds were not included in Table 12.

The TH concentrations are in ng/mL plasma for both species, and male arctic skuas had significantly higher levels of TT4 (ANOVA and Tukey's post hoc test, $p = 0.042$) than male kittiwakes (Table 12).

In male kittiwakes correlations between PFASs and TT4 were detected, while in female kittiwakes there were correlations between PFASs and both THs (Figures 5 and 6). Only TT4 was found to be correlated to PFASs in arctic skuas (Figure 11 and 12). The correlations were positive in both species.

Male kittiwakes had positive correlations between PFASs and BC (Figure 7), whereas in both female kittiwakes and male arctic skuas the associations between PFASs and BC and body mass were negative (Figures 8 and 13).

Table 12: A comparison between thyroid hormones (total triiodothyronine (TT3) and total thyroxine (TT4), ng/mL plasma) and perfluoroalkyl and polyfluoroalkyl substances (PFASs, pmol/mL plasma and whole blood) in black-legged kittiwakes (*Rissa tridactyla*, PFAS concentrations in plasma) and arctic skuas (*Stercorarius parasiticus*, PFAS concentrations in whole blood). The black-legged kittiwakes were caught in Kongsfjorden, Svalbard, in 2013 and 2014. The arctic skuas were caught on Brensholmen, Norway and in Kongsfjorden, Svalbard, in 2014. The mean ± standard deviation (SD) concentrations of analytes are given for the kittiwakes and arctic skuas, and the *p*-value of the comparisons between concentrations.
* Male arctic skuas had significantly higher concentrations of TT4 than male kittiwakes (ANOVA and Tukey's post hoc test, $p = 0.042$).

	Male			Female		
Analyte	Kittiwake Mean ± SD	Arctic Skua Mean ± SD	*p*- value	Kittiwake Mean ± SD	Arctic Skua Mean ± SD	*p*- value
TT3	2.78 ± 0.713	3.22 ± 0.884	0.155	2.96 ± 1.08	2.42 ± 0.687	0.293
TT4*	36.45 ± 11.2	46.65 ± 16.9	0.042	37.18 ± 14.9	30.59 ± 13.8	0.369
linPFOS	14.7 ± 7.16	20.0 ± 7.78	-	8.68 ± 6.29	9.90 ± 4.44	-
sumPFOS	15.0 ± 7.45	20.8 ± 8.49	-	8.94 ± 6.51	10.2 ± 4.67	-
PFNA	3.30 ± 2.36	1.99 ± 2.87	-	2.69 ± 1.61	1.28 ± 0.806	-
PFDA	2.51 ± 1.01	2.91 ± 0.787	-	2.15 ± 0.853	1.63 ± 0.752	-
PFUnDA	12.3 ± 4.29	12.4 ± 3.40	-	9.97 ± 3.09	7.30 ± 3.02	-
PFDoDA	2.58 ± 1.16	2.63 ± 0.725	-	1.89 ± 0.856	1.30 ± 0.698	-
PFTrDA	7.92 ± 2.73	5.24 ± 1.81	-	5.52 ± 2.10	2.64 ± 1.06	-

4. Discussion

The current study found that PFASs were positively correlated to TT4 in both sexes for kittiwakes and arctic skuas. It was only in female kittiwakes where positive correlations between PFASs and TT3 were detected. In kittiwakes PFASs were correlated to both BC and body mass with positive correlations in males and negative correlations in females. There were also negative correlations between PFASs and body mass in male arctic skuas, but no significant correlations were detected between PFASs and BC. Concentrations of PFASs in female arctic skuas had no significant correlations with either BC or body mass.

4.1 Perfluoroalkyl and Polyfluoroalkyl Substance Concentrations and Abundance

One goal of the present study was to investigate the concentrations of PFASs in kittiwakes and arctic skuas. In male kittiwakes linPFOS and PFUnDA were the dominant PFASs, whereas in female kittiwakes PFUnDA was the most abundant compound followed by linPFOS (Figure 4).

A study on kittiwake fledglings found linPFOS to be the dominant PFAS in line with the result in the current work, however, the differences in sex were not considered in the fledglings (Nøst et al., 2012). Tartu et al. (2014) found PFTrDA to be the dominant PFAS detected in 20 kittiwakes caught in 2012 from the same colony ("Krykkjefjellet, Kongsfjorden, Ny-Ålesund) as the present study. Furthermore, the concentrations of PFTrDA in male and female kittiwakes reported by Tartu et al. (2014) were higher than those detected in the present study. For the other compounds, the levels were comparable to the levels detected in kittiwakes in the present study.

Tomy et al. (2004) detected PFOS in liver tissue from kittiwakes caught in Canada with slightly higher concentrations than detected in the present study for kittiwakes. While it is not clear when those particular kittiwakes were sampled, it must necessarily have been in or prior to 2004. The PFOS phase-out happened in 2001 (3M, 2000), and the higher concentration reported by Tomy et al. (2004) may be due to the recent sampling after cessation of PFOS synthesis by its major producer while the kittiwakes in the present study were sampled in 2013 and 2014. This is supported by a temporal study on ringed seals (*Phoca hispida*) and polar bears (*Ursus maritimus*) from Greenland which found that PFOS concentrations started decreasing around 2006 (Rigét et al., 2013). However, a study on guillemot (*Uria aalge*) eggs from the Baltic Sea found a decline in PFOS levels prior to the phase-out (Holmström et al., 2005). This suggests the difference in PFOS levels between the kittiwakes sampled in Canada and the kittiwakes in the current study may either be a result of geographical variations in levels, or a difference in diet (Braune et al., 2002; Braune et al., 2005; Mallory and Braune, 2012).

In both male and female arctic skuas the dominant compounds were linPFOS followed by PFUnDA, however there was only a significant difference in levels of linPFOS and PFUnDA for male arctic skuas (Figure 10).

The scientific literature on toxicant levels in arctic skua is scarce, and no studies on PFAS levels seem to have been published. Concentrations of PFASs were reported in plasma from great skuas sampled in 2008 on Shetland (Leat et al., 2012). The concentrations of PFASs were elevated in great skua plasma compared to the concentrations in whole blood reported for arctic skua herein. This was particularly notable for PFOS where great skuas had a four-fold elevation in concentration compared to the PFOS burden in arctic skuas. However, PFOS was measured in plasma for the great skuas and in whole blood for the arctic skuas in the present study. A study by Hanssen et al. (2013) investigated concentrations of selected PFASs (PFOA, PFHxS, PFOS, PFNA, PFUnDA, and FOSA) in whole blood and plasma samples drawn from humans. They found that, of the PFASs studied, only FOSA was detected at a higher concentration in whole blood compared to plasma. The other compounds all had greater concentrations in plasma than in whole blood. This is in accordance with the results of other studies (Ehresman et al., 2007; Martin et al., 2009).

Thus, part of the reason why great skuas displayed higher PFAS concentrations than arctic skuas may be due to the expected difference in concentration between plasma and whole blood. Indeed, FOSA was the only compound where the arctic skuas had a higher concentration than the great skuas consistent with the study by Hanssen et al. (2013). Furthermore, great skuas feed at a higher trophic level than arctic skuas, and is thus expected to bioaccumulate more PFASs than arctic skuas. PFCAs with ≤ 7 fluorinated carbons are not bioaccumulative, and PFSAs are more bioaccumulative than PFCAs of the same chain length (Conder et al., 2008). This compares well with the differences seen between the great skuas and arctic skuas as PFOS is considered to be the most bioaccumulative PFAS and is the compound which had the greatest difference in concentration between the two species. Concentrations of PFNA (8 C) and PFDA (9 C) were only slightly higher in great skuas than in arctic skuas, and they are just above the threshold for bioaccumulation reported in Conder et al. (2008). This may explain the fact that the difference in these compounds between the great skuas and arctic skuas is not as high as might have been expected.

Tao et al., (2006) analyzed whole blood from the south polar skua (*Stercorarius maccormicki*) for PFASs. Only PFOS was detected, at 0.88 ng/mL whole blood. It is not stated whether this was for male or female south polar skuas, and it should be noted that only three individuals were sampled. This concentration is six- and twelve-fold lower than what was detected in female and male arctic skuas in this study, respectively. The south polar skuas were sampled in 1998 - 1999 in Antarctica. As the PFOS phase-out by the main producer, 3M, was not initiated until 2001 (3M, 2000), the difference in PFOS concentration is likely due to the fact that most of the PFAS manufacturing and use has occurred in the northern hemisphere. Indeed, a study by Zhao et al. (2012) analyzed PFASs in sea-water from the Atlantic ocean, Greenland sea and Southern ocean in 2009 - 2010. The concentration of ΣPFAS was highest in the Atlantic ocean (260 pg/L), followed by the Greenland sea (140 pg/L), and lowest in the Southern Ocean (30 pg/L). Therefore, the low concentration detected in south polar skua is likely to be a result of the lower levels of PFAS contamination in the antarctic environment.

There have been indications that there are no differences in PFAS concentrations between the sexes in arctic wildlife (Verreault et al., 2005; Butt et al., 2007a; Butt et al., 2007b; Haukås et al., 2007). This contradicts the results in the present study where sex differences were found for most of the detected PFASs in both kittiwakes and arctic skuas. Some other studies have also reported sex differences for PFASs in great skuas (Leat et al., 2012) and in lesser black-backed gulls (*Larus fuscus*) (Bustnes et al., 2008). These studies found higher concentrations of PFASs in males compared to females consistent with the current study. Both arctic skuas and great skuas have reverse sexual dimorphism where the female is larger than the male. Great skua females are able to bring down larger prey than the males (Leat et al., 2012), and so it is unlikely that diet is the reason for the observed sex differences. Maternal deposition of PFASs into the eggs is a more probable explanation. Several avian studies have reported detecting PFASs in eggs (Verreault et al., 2005; Verreault et al., 2007b; Löfstrand et al., 2008; Miljeteig et al., 2009; Miljeteig and Gabrielsen, 2010). An experimental study by Newsted et al. (2007) on mallards (*Anas platyrhynchos*) and northern bobwhite quail (*Colinus virginianus*) exposed to PFOS found that it was associated with proteins in the egg yolk, particularly very low-density lipoprotein. PFASs, in general, bind to proteins, and have been labeled proteinophilic (Han et al., 2003). The observed sex differences reported in the present study are, thus, likely a result of the female bird being able to deposit some of her PFAS burden into the eggs.

A study on branched and linear ammonium perfluorooctanoate (APFO) indicates that branched and linear PFAS isomers have different toxicological properties (Loveless et al., 2006). Branched PFOS was excluded for arctic skuas in the present study as it was below the limit of detection in more than 30% of the samples. For kittiwakes linPFOS was positively correlated to TT4 in male kittiwakes while in female kittiwakes it was brPFOS which had a positive correlation to TT4. Furthermore, linPFOS had a negative correlation to BC in female kittiwakes, but brPFOS was not significantly correlated to BC in either male or female kittiwakes. The different effects might be due to a pronounced difference in concentration between the two isomers with linPFOS being detected at greater levels than brPFOS in kittiwakes (Figure 4). However, in the study by Loveless et al. (2006) equal doses of branched and linear isomers were administered to the experimental animals, and a difference in toxicity was still observed. Thus, there are indications of branched and linear PFOS isomers having different toxicity in the present study, with brPFOS seemingly less potent than linPFOS.

Newsted et al., (2005) calculated avian toxicity reference values for PFOS based on experiments with northern bobwhite quail. The predicted no effect concentration (PNEC) of PFOS in serum of a trophic level 4 avian predator is 2.4 µg/mL for males and 0.15 µg/mL for females. The highest detected PFOS concentration in the current study was 10.4 ng/mL, or 0.0104 µg/mL, in male arctic skuas (Appendix C). This is two orders of magnitude lower than the PNEC. Female birds are more sensitive (Newsted et al., 2005), and female arctic skuas had a PFOS burden of 5.08 ng/mL, or 0.00508 µg/mL. This is 30 times lower than the PNEC. It should be noted that the arctic skua concentrations of PFOS are from whole blood so it is possible that the concentration in serum would be slightly higher. However, the observed concentrations in the current study are still well below the PNECs calculated by Newsted et

al. (2005), and it is unlikely that the concentrations of PFOS and other PFASs in themselves pose an overt toxicological risk.

It is important to keep in mind that species differences may occur. This might be due to a difference in biotransformation capacity (Fisk et al., 2001; Buckman et al., 2004), and this, in turn, could affect the biotransformation of precursor molecules, such as FOSA, into their final degradation products. As such, this may explain the difference in PFAS profiles found in seabirds. Feeding ecology is another reason for potential differences in PFAS concentrations between, and even within, species (Bustnes et al., 2000).

Finally, when comparing PFAS concentrations detected in different tissues (e.g. plasma, whole blood, liver, eggs), it is with the understanding that PFASs are not evenly distributed in the body of the organism. Martin et al. (2009) reported that PFAS concentrations in rainbow trout (*Oncorhynchus mykiss*) were greatest in the blood followed by the liver. This is supported by a study in glaucous gulls (Verreault et al., 2005) and harbour seals (*Phoca vitulina*) (Ahrens et al., 2009). Verreault et al. (2005) reported similar levels of PFASs in liver and egg samples from glaucous gulls. Thus, when comparing PFAS concentrations between kittiwakes and arctic skuas in Table 12, it is important to keep in mind that the PFAS concentrations in kittiwakes are from plasma samples and in arctic skuas they are from whole blood samples. As discussed previously, PFAS concentrations are expected to be lower in whole blood samples compared to plasma samples, with the exception of FOSA (Hanssen et al., 2013). Therefore, the PFNA and PFTrDA concentrations in kittiwakes and arctic skuas may in reality be comparable.

4.2 Thyroid Hormone Concentrations

There were no differences in thyroid hormone concentrations between males and females for either kittiwakes or arctic skuas. The only detected difference was that male arctic skuas had significantly higher levels of TT4 than male kittiwakes (Table 12). Neither BC nor mass were significantly correlated with the thyroid hormones in either bird species.

Elliot et al. (2013) measured TT3 and TT4 in breeding kittiwakes from Alaska in 2010. The reported concentration of TT3 is similar to the concentrations reported for kittiwakes and arctic skuas in the present study. The levels of TT4 were also comparable between the kittiwakes from Alaska and the female kittiwakes and arctic skuas in this study, but the TT4 concentration in male kittiwakes and arctic skuas are higher than what is reported in Elliot et al. (2013).

Concentrations of TT3 were examined in kittiwakes from Kongsfjorden in 2001 and 2010 by Welcker et al. (2013). The kittiwakes caught in 2001 had considerably lower TT3 levels than the kittiwakes and arctic skuas of this study. In 2010, however, the TT3 concentrations were similar to those reported for kittiwakes and arctic skuas in the present study.

Thyroid hormones were investigated in fledgling kittiwakes by Nøst et al. (2012). The reported concentration for TT3 is similar to the concentration detected in the kittiwakes and arctic skuas of the present study. The levels of TT4 in fledgling kittiwakes were notably lower than for the breeding kittiwakes and arctic skuas in the current study. The levels of TT3 reported herein for female kittiwakes and arctic skuas also corresponded fairly well with TT3 levels detected in female kittiwakes sampled in 2005 (Rønning et al., 2008).

Care must be taken when comparing thyroid hormone concentrations, as they are likely to change depending on season and geographical location (Burger and Denver, 2002).
Even comparing TH concentrations within a species could be inappropriate, and especially so for comparing concentrations between kittiwakes and arctic skuas. Nevertheless, as scientific literature on arctic skuas is sparse, no reported TH concentrations were found to compare against the TH results for arctic skuas presented in the current study.

4.3 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances and Thyroid Hormones

When testing if percentage of fat in the blood, BC, and body mass were confounding factors in the models of the associations between PFASs and THs, the association between brPFOS and TT4 in female kittiwakes appeared to be confounded by the blood fat percentage. However, theory indicates that brPFOS associates with proteins in the blood, not lipid content (Han et al., 2003; Jones et al., 2003). Thus the percentage of fat in the blood is disregarded as a confounding variable for the brPFOS and TT4 model in female kittiwakes.

In the present study male kittiwakes had positive correlations between their levels of TT4 and levels of linPFOS, sumPFOS, PFDA, and PFUnDA (Figure 5). Concentration of TT4 was only positively correlated to brPFOS in female kittiwakes (Figure 6), but levels of TT3 were positively correlated to levels of PFDoDA, PFTrDA, and PFTeDA in female kittiwakes (Figure 6). In arctic skuas, only TT4 was significantly correlated to PFAS concentrations. In male arctic skuas TT4 was positively correlated to PFUnDA, PFDoDA, FOSA (Figure 11) whereas TT4 was positively correlated to linPFOS and sumPFOS in females (Figure 12).

The scientific literature on the effects of PFASs on the thyroid hormone system in free-ranging seabirds is scarce, but Nøst et al. (2012) reported a positive association between TT4 and PFHpS, brPFOS, linPFOS, and PFNA in kittiwake fledglings. The study also investigated associations in northern fulmar fledglings where TT4 was found to have a positive association with PFHpS, brPFOS, and PFNA. Furthermore, a study on adult northern fulmars in Canada found a positive correlation between plasma levels of TT3 and concentrations of ΣPFCAs in liver tissue (Braune et al., 2011). Although comparing concentrations detected in different compartments may yield a tenuous correlation, it should be mentioned that Dauwe et al. (2007) found PFAS concentrations detected in liver to be strongly correlated with concentrations detected in the blood ($N = 16$, $r = 0.80$, $p < 0.001$) of great tits (*Parus major*). Thus, there are several indications that PFASs are positively associated with levels of TT3

and TT4 in seabirds. This is in contrast to the results in other studies that report negative correlations between PFASs and THs in rats and mice (Thibodeaux et al., 2003; Chang et al., 2008), monkeys (Seacat et al., 2002), chicken embryos (Cassone et al., 2012), and humans (Wang et al., 2014; Berg et al., 2015).

PFASs may interfere with the thyroid hormone system through multiple pathways (Zoeller et al., 2007; Boas et al., 2012; Webster et al., 2014). Firstly, it has been proposed that PFASs increase T4 excretion by up-regulating hepatic transporters which, in turn, increases uptake of T4 into the liver (Yu et al., 2011). There are also indications that PFASs increase glucuronidation and subsequent elimination of T4 in the liver by up-regulating the hepatic enzyme, UGT1A1 (Chang et al., 2008; Yu et al. 2009). However, these two mechanisms of action do not appear to be prominent in the current work as PFASs have an exclusively positive correlation with TT4 (Table 5 and 10). If more TT4 was being cleared from the kittiwakes and arctic skuas, a negative correlation would be expected.

Secondly, PFASs may interfere with iodothyronine deiodinases (IDs). The IDs are enzymes which convert T4 into T3 and convert T4 and T3 into reverse T3 (rT3) and diiodothyronine (T2) which are inactive metabolites (Jarque and Piña, 2014). A toxicogenomic study conducted by Wei et al. (2008) found that rare minnows (*Gobiocypris rarus*) exposed to PFOA had a significant down-regulation of hepatic ID type 2. Conversely, a study by Shi et al. (2009) reported significant up-regulation of ID type 1 in zebrafish (*Danio rerio*) larvae following PFOS exposure.

Another disruptive effect of PFASs is displacing THs from binding proteins. Weiss et al. (2009) examined the TTR binding potencies of PFASs in humans. An interesting result was that for PFASs with a fluorinated carbon chain between 4 to 8 carbons, TTR binding potencies were significantly higher for PFSAs than for PFCAs. Furthermore, the more fluorinated the chain, the higher the binding potency. However, a study by Ucán-Marín et al. (2009) found that avian TTR is more similar to crocodilian TTR than human TTR, and so caution is prudent when comparing TTR binding potencies between different species. Additionally, albumin is a more important transport protein than TTR for avian THs (McNabb, 2007), and so competitive binding of PFASs to TTR is of less importance in birds than humans.

Lastly, PFASs could act directly on the thyroid gland itself. Coperchini et al. (2015) conducted an *in vitro* study where the effects of PFOA and PFOS on thyroid cells were evaluated. They observed decreased cell proliferation and increased cell death at the highest concentration. Furthermore, the substances were found to enter the cell, most likely by gradient-based diffusion. The concentration used was not environmentally relevant, but a low-dose, chronic exposure might result in cytotoxicity. Indeed, in a study by Hu et al. (2003) the ability of PFOS to alter the membrane permeability of fish leukocytes was demonstrated.

Many of the studies observing toxic effects on the thyroid system do so at PFAS concentrations notably higher than those found in the environment. Nevertheless, it does give

an indication of the toxicity of PFASs at high, acute doses which may be indicative of effects from lower, but chronic, doses.

4.4 Associations between Perfluoroalkyl and Polyfluoroalkyl Substances, Body Condition, and Body Mass

Body condition for male kittiwakes in the present study was positively correlated to levels of PFDoDA, PFTrDA, and PFTeDA (Figure 7). A similar result was reported by Tartu et al., (2014) where PFNA was positively correlated to BC in male kittiwakes.

The mechanism of action behind the ability of PFASs to increase body mass and improve body condition in male kittiwakes may be related to the structural resemblance of PFASs to fatty acids. Indeed, a study by Vanden Heuvel et al. (2006) demonstrated the ability of PFOA and PFOS to activate the nuclear receptor peroxisome proliferator-activated receptor (PPAR). PPAR is involved in lipid storage and homeostasis, as well as up-regulation of fatty acid transport protein in the liver (Escher and Wahli, 2000; Li and Glass, 2004; Feige et al., 2006). Furthermore, a study by Guruge et al. (2006) on male rats found that PFOA induced genes involved in metabolism and transport of fatty acids and lipids, in addition to genes involved in growth.

On the other hand, female kittiwakes with higher levels of linPFOS, sumPFOS, PFNA, and PFDA had a poorer BC than females with lower levels of those PFASs (Figure 8). This is similar to the observation in male arctic skuas where individuals with higher concentrations of PFDA, PFUnDA, PFTrDA, and FOSA had a lower body mass than those individuals with lower concentrations (Figure 13). Negative relationships between PFASs and mass have been reported in rats, mice, and rabbits (Case et al., 2001; Thibodeaux et al., 2003), monkeys (Seacat et al., 2002), and chicken embryos (Cassone et al., 2012).

It is perplexing that male kittiwakes show an improvement in BC with higher levels of certain PFASs while the BC in female kittiwakes deteriorate with increasing concentrations of other PFASs. This apparent inconsistency may be related to a sex difference. Wei et al. (2008) noted that genes involved in β-oxidation of fatty acids in the mitochondria were down-regulated in female rare minnows, but unchanged in male rare minnows exposed to PFOA. Thus, it is possible that PFASs will behave differently in male and female kittiwakes. Another possibility for the discrepancy of PFASs being positively correlated to BC in male kittiwakes and PFASs being negatively correlated to female kittiwakes, are the individual compounds themselves. Body condition in male kittiwakes is linked to PFDoDA, PFTrDA, and PFTeDA, compounds with 11, 12, and 13 fluorinated carbons, respectively. Conversely, in female kittiwakes BC is correlated to PFOS, PFNA, and PFDA, compounds with 8, 8, and 9 fluorinated carbons, respectively. As the length of the fluorinated carbon chain is known to have an impact on bioaccumulation and protein binding (Conder et al., 2008; Weiss et al., 2009), for instance, it is possible that the difference observed in males and females is due to the longer chain length in males. Indeed, there were significantly higher levels of PFDoDA,

PFTrDA, and PFTeDA in male compared to female kittiwakes (Table 4). However, in male arctic skuas there was a negative relationship between levels of PFTrDA and body mass. This would suggest that sex and the fluorinated carbon chain length do not affect whether the correlation is positive or negative, although care must be taken when comparing different species. Finally, although Tartu et al. (2014) observed a positive correlation between PFNA and BC in male kittiwakes, PFNA was not correlated to BC in male kittiwakes in the present study. The compound was, however, negatively correlated to BC in female kittiwakes.

Altogether, the contradicting results highlight the need to further investigate effect of PFASs on body condition and body mass.

4.5 Future Perspectives

The results in the current study report a sex difference in PFAS levels and toxicity. As there are conflicting results reported in the scientific literature with regards to the sex differences, it would be prudent to include sex as a factor in future research on the effects of PFASs on wildlife.

The current work analyzed plasma samples from kittiwakes and arctic skuas for total T3 and T4. As PFASs have a structural resemblance to THs and bind to proteins in the blood, they may disrupt the thyroid system by displacing THs from binding sites on proteins. The free hormone is then available for excretion. Therefore, examining the ratio between free and total hormone may be insightful, and future studies should incorporate free T3 and T4 into the experimental design.

In addition, several studies assessing the effects of PFAS burden on THs in humans reported a positive correlation between PFOS and TSH (Wang et al., 2013; Webster et al., 2014; Berg et al., 2015). TSH is a sensitive biomarker and intimately linked to levels of T3 and T4 (Zoeller et al., 2007; Glinoer and Spencer, 2010). Thus, analyzing for TSH in future studies investigating thyroid disruption could help discern more about the effect of PFASs on the thyroid system in arctic seabirds.

As previously discussed, the literature on the effects of PFASs on BC and body mass is contradictive. More experimental research is needed to elucidate the mechanism(s) of action, in addition to field studies on arctic seabirds.

Finally, the present study is, to the author's best knowledge, the first to report concentrations of PFASs and THs in the arctic skua. Moreover, it is the first study to assess the associations between PFASs and THs, and PFASs and body mass in arctic skuas. The declining arctic skua populations in the United Kingdom add urgency to investigating levels of possible harmful anthropogenic toxicants and their effects in this species.

5. Conclusion

Perfluoroalkylated substances were detected in kittiwakes and arctic skuas, in accordance with the hypothesis. In both species the males had generally higher levels of PFASs than the females. As hypothesized there were positive correlations between PFASs and THs in both kittiwakes and arctic skuas, although the experimental design did not allow for a causal relationship to be determined. Similarly, there were correlations between PFASs and BC, and PFASs and body mass in kittiwakes and arctic skuas, respectively. In male kittiwakes, PFASs were positively correlated to BC. Conversely, in female kittiwakes and male arctic skuas, negative correlations were identified between PFASs and BC as well as PFASs and body mass. Neither body condition nor body mass in female arctic skuas were correlated to their body burden of PFASs. The mechanisms of how PFASs exert their effects upon the thyroid hormone system, body condition, and body mass are unknown, and future research efforts should be directed towards elucidating this.

References

3M. Phase-out plan for POSF-based products. *U.S. Environmental Protection Agency*, (2000), Docket ID OPPT-2002-0043.

Ahrens L., Siebert U., and Ebinghaus R. Total body burden and tissue distribution of polyfluorinated compounds in harbour seals (*Phoca vitulina*) from the German Bight. *Marine Pollution Bulletin*, 58 (2009), 520-525.

Anker-Nilssen T., Bakken V., Strøm H., Golovkin A.N., Bianki V.V., Tatarinkova I.P. The Status of Marine Birds Breeding in the Barents Sea Region. *Norwegian Polar Institute Research Report*, 113 (2000).

Anker-Nilssen T. (ed). Seabirds in Norway 2009 - Results from the SEAPOP Programme. *SEAPOP Short Report*, (2010), 12 pp.

Anker-Nilssen T., Strøm H. (ed.), Barret R.T., Descamps S., Erikstad K-E., Fauchald P., Lorentsen S-H., Moe B., Systad G.H. Sjøfugl i Norge 2012. Resultater fra SEAPOP-programmet. *SEAPOP Short Report*, 1 (2013), 13 pp.

Berg V., Nøst T.H., Hansen S., Elverland A., Veyhe A-S., Jorde R., Odland J.Ø., Sandanger T.M. Assessing the relationship between perfluoroalkyl substances, thyroid hormones and binding proteins in pregnant women; a longitudinal mixed effects approach. *Environment International*, 77 (2015), 63-69.

Blais J.M., Kimpe L.E., McMahon D., Keatley B.E., Mallory M.L., Douglas M.S.V. and Smol J.P. Arctic Seabirds Transport Marine-Derived Contaminants. *Science*, 309 (2005), 445.

Boas M., Feldt-Rasmussen U., and Main K.M. Thyroid effects of endocrine disrupting chemicals. *Molecular and Cellular Endocrinology*, 355 (2012), 240-248.

Borgå K., Gabrielsen G.W. and Skaare J.U. Biomagnification of organochlorines along a Barents Sea food chain. *Environmental Pollution*, 113 (2001), 187-198.

Bourgeon S., Leat E.H.K., Magnusdóttir E., Fisk A.T., Furness R.W., Strøm H., Hanssen S.A., Petersen Æ., Olafsdóttir K., Borgå K., Gabrielsen G.W., and Bustnes J.O. Individual variation in biomarkers of health: Influence of persistent organic pollutants in Great skuas (*Stercorarius skua*) breeding at different geographical locations. *Environmental Research*, 118 (2012), 31-39.

Braune B.M., Donaldson G.M., and Hobson K.A. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences. *Environmental Pollution*, 117 (2002), 133-145.

Braune B.M., Outridge P.M., Fisk A.T., Muir D.C.G., Helm P.A., Hobbs K., Hoekstra P.F., Kuzyk Z.A., Kwan M., Letcher R.K., Lockhart W.L., Norstrom R.J., Stern G.A., and Stirling I. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: An overview of spatial and temporal trends. *Science of The Total Environment*, 351-352 (2005), 4-56.

Braune B.M., Trudeau S., Jeffrey D.A., and Mallory M.L. Biomarker responses associated with halogenated organic contaminants in northern fulmars (*Fulmarus glacialis*) breeding in the Canadian Arctic. *Environmental Pollution*, 159 (2011), 2891-2898.

Bryant D.M. Energy expenditure in wild birds. *Proceedings of the Nutrition Society*, 56 (1997), 1025-1039.

Buck R.C., Franklin J., Berger U., Conder J.M., Cousins I.T., de Voogt P., Jensen A.A., Kannan K., Mabury S.A., and van Leeuwen S.P.J. Perfluoroalkyl and Polyfluoroalkyl Substances in the Environment: Terminology, Classification, and Origins. *Integrated Environmental Assessment and Management*, 7 (2011), 513-541.

Buckman A.H., Norstrom R.J., Hobson K.A., Karnovsky N.J., Duffe J. and Fisk A.T. Organochlorine contaminants in seven species of Arctic seabirds from northern Baffin Bay. *Environmental Pollution*, 128 (2004), 327-338.

Burger M.F., and Denver R.J. Plasma thyroid hormone concentrations in a wintering passerine bird: their relationship to geographic variation, environmental factors, metabolic rate, and body fat. *Physiological and Biochemical Zoology*, 75 (2002), 187-199.

Bustnes J.O., Erikstad K.E., Bakken V., Mehlum F., Skaare J.U. Feeding Ecology and the Concentration of Organochlorines in Glaucous Gulls. *Ecotoxicology*, 9 (2000), 179-186.

Bustnes J.O., Borgå K., Erikstad K.E., Lorentsen S-H., Herzke D. Perfluorinated, Brominated, and Chlorinated Contaminants in a Population of Lesser Black-Backed Gulls (*Larus fuscus*). *Environmental Toxicology and Chemistry*, 27 (2008), 1383-1392.

Butt C.M., Mabury S.A., Muir D.C.G., and Braune B.M. Prevalence of Long-Chained Perfluorinated Carboxylates in Seabirds from the Canadian Arctic between 1975 and 2004. *Environmental Science and Technology*, 41 (2007a), 3521-3528.

Butt C.M., Muir D.C.G., Stirling I., Kwan M., and Mabury S.A. Rapid Response of Arctic Ringed Seals to Changes in Perfluoroalkyl Production. *Environmental Science and Technology*, 41 (2007b), 42-49.

Butt C.M., Berger U., Bossi R. and Tomy G.T. Levels and trends of poly- and perfluorinated compounds in the arctic environment. *Science of The Total Environment*, 408 (2010), 2936-2965.

Case M.T., York R.G., and Christian M.S. Rat and rabbit oral developmental toxicology studies with two perfluorinated compounds. *International Journal of Toxicology*, 20 (2001), 101-109.

Cassone C.G., Vongphachan V., Chiu S., Williams K.L., Letcher R.J., Pelletier E., Crump D., and Kennedy S.W. *In Ovo* Effects of Perfluorohexane Sulfonate and Perfluorohexanoate on Pipping Success, Development, mRNA Expression, and Thyroid Hormone Levels in Chicken Embryos. *Toxicological Sciences*, 127 (2012), 216-224.

Catry P., Phillips R.A., and Furness R.W. Evolution of Reversed Sexual Size Dimorphism in Skuas and Jaegers. *The Auk*, 116 (1999), 158-168.

Chang S-C., Thibodeaux J.R., Eastvold M.L., Ehresman D.J., Bjork J.A., Froelich J.W., Lau C., Singh R.J., Wallace K.B., and Butenhoff J.L. Thyroid hormone status and pituitary function in adult rats given oral doses of perfluorooctanesulfonate (PFOS). *Toxicology*, 243 (2008), 330-339.

Chastel O., Lacroix A., and Kersten M. Pre-breeding energy requirements: thyroid hormone, metabolism and the timing of reproduction in the house sparrows *Passer domesticus*. *Journal of Avian Biology*, 34 (2003), 298-306.

Conder J.M., Hoke R.A., de Wolf W., Russell M.H., and Buck R.C. Are PFCAs Bioaccumulative? A Critical Review and Comparison with Regulatory Criteria and Persistent Lipophilic Compounds. *Environmental Science and Technology*, 42 (2008), 995-1003.

Coperchini F., Pignatti P., Lacerenza S., Negri S., Sideri R., Testoni C., de Martinis L., Cottica D., Magri F., Imbriani M., Rotondi M., and Chiovato L. Exposure to perfluorinated compounds: in vitro study on thyroid cells. *Environmental Science and Pollution Research*, 22 (2015), 2287-2294.

Crisp T.M., Clegg E.D. and Cooper R.L. Special Report on Environmental Endocrine Disruption: An Effects Assessment and Analysis. EPA/630/R-96/012, 1997.

Dahl T.M., Falk-Petersen S., Gabrielsen G.W., Sargent J.R., Hop H. and Millar R.M. Lipids and stable isotopes in common eider, black-legged kittiwake and northern fulmar: a trophic study from an Arctic fjord. *Marine Ecology Progress Series*, 256 (2003), 257-269.

Dauwe T., Van de Vijver K., De Coen W., and Eens M. PFOS levels in the blood and liver of a small insectivorous songbird near a fluorochemical plant. *Environment International*, 33 (2007), 357-361.

Dawson N.M., MacLeod C.D., Smith M., and Ratcliffe N. Interactions with Great Skuas *Stercorarius skua* as a factor in the long-term decline of an Arctic Skua *Stercorarius parasiticus* population. *International Journal of Avian Science*, 153 (2011), 143-153.

De Silva A.O., and Mabury S.A. Isolating Isomers of Perfluorocarboxylates in Polar Bears (*Ursus maritimus*) from Two Geographical Locations. *Envionmental Science and Technology*, 38 (2004), 6538-6545.

Diamanti-Kandarakis E., Bourguignon J., Giudice L.C., Hauser R., Prins G.S., Soto A.M., Zoeller R.T. and Gore A.C. Endocrine-Disrupting Chemicals: An Endocrine Society Scientific Statement. *Endocrine Reviews*, 30 (2009), 293-342.

Dowdall M., Gerland S. and Lind B. Gamma-emitting natural and anthropogenic radionuclides in the terrestrial environment of Kongsfjord, Svalbard. *Science of The Total Environment*, 305 (2003), 229-240.

Ehresman D.J., Froelich J.W., Olsen G.W., Chang S-C., and Butenhoff J.L. Comparison of human whole blood, plasma, and serum matrices for the determination of perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA), and other fluorochemicals. *Environmental Research*, 103 (2007), 176-184.

Elliott K.H., Welcker J., Gaston A.J., Hatch S.A., Palace V., Hare J.F., Speakman J.R., and Anderson W.G. Thyroid hormones correlate with resting metabolic rate, not daily energy expenditure, in two charadriiform seabirds. *Biology Open*, 2 (2013), 580-586.

Escher P., and Wahli W. Peroxisome proliferator-activated receptors: insight into multiple cellular functions. *Mutation Research/Fundamental and Molecular Mechanisms of Mutagenesis*, 448 (2000), 121-138.

Fisk A.T., Moisey J., Hobson K.A., Karnovsky N.J., and Norstrom R.J. Chlordane components and metabolites in seven species of Arctic seabirds from the Northwater Polynya: relationships with stable isotopes of nitrogen and enantiomeric fractions of chiral components. *Environmental Pollution*,113 (2001), 225-238.

Feige J.N., Gelman L., Michalik L., Desvergne B., and Wahli W. From molecular action to physiological outputs: Peroxisome proliferator-activated receptors are nuclear receptors at the crossroads of key cellular functions. *Progress in Lipid Research*, 45 (2006), 120-159.

Furness, R.W. Family Stercorariidae (Skuas). Pp 556-572 in: del Hoyo J., Elliott A. and Sargatal J. eds. (1996). *Handbook of the Birds of the World*. Vol 3. Hoatzin to Auks. Lynx edition, Barcelona.

Giesy J.P. and Kannan K. Global Distribution of Perfluorooctane Sulfonate in Wildlife. *Environmental Science and Technology*, 35 (2001), 1339-1342.

Glinoer D., and Spencer C.A. Serum TSH determinations in pregnancy: how, when and why? *Nature Reviews Endocrinology*, 6 (2010), 526-529.

Griffiths R., Double M.C., Orr K., and Dawson R.J.G. A DNA test to sex most birds. *Molecular Ecology*, 7 (1998), 1071-1075.

Guruge K.S., Yeung L.W.Y., Yamanaka N., Miyazaki S., Lam P.K.S., Giesy J.P., Jones P.D., and Yamashita N. Gene Expression Profiles in Rat Liver Treated With Perfluorooctanoic Acid (PFOA). *Toxicological Sciences*, 89 (2006), 93-107.

Han X., Snow T.A., Kemper R.A., and Jepson G.W. Binding of Perfluorooctanoic Acid to Rat and Human Plasma Proteins. *Chemical Research in Toxicology*, 16 (2003), 775-781.

Hanssen L., Dudarev A.A., Huber S., Odland J.Ø., Nieboer E., and Sandanger T.M. Partition of perfluoroalkyl substances (PFASs) in whole blood and plasma, assessed in maternal and umbilical cord samples from inhabitants of arctic Russia and Uzbekistan. *Science of the Total Environment*, 447 (2013), 430-437.

Haukås M., Berger U., Hop H., Gulliksen B., and Gabrielsen G.W. Bioaccumulation of per- and polyfluorinated alkyl substances (PFAS) in selected species from the Barents Sea food web. *Environmental Pollution*, 148 (2007), 360-371.

Holmström K.E., Järnberg U., and Bignert A. Temporal Trends of PFOS and PFOA in Guillemot Egss from the Baltic Sea, 1968-2003. *Environmental Science and Technology*, 39 (2005), 80-84.

Houde M., De Silva A.O., Muir D.C.G., and Letcher R. J. Monitoring of Perfluorinated Compounds in Aquatic Biota: An Updated Review. *Environmental Science and Technology*, 45 (2011), 7962-7973.

Howdeshell, K.L. A model of the development of the brain as a construct of the thyroid system. *Environmental Health Perspectives*, 110, suppl 3 (2002), 337-348.

Hu W.Y., Jones P.D., DeCoen W., King L., Fraker P., Newsted J., and Giesy J.P. Alterations in cell membrane properties cased by perfluorinated compounds. *Comparative Biochemistry and Physiology Part C: Toxicology and Pharmacology*, 135 (2003), 77-88.

Hung H., Kallenborn R., Breivik K., Su Y., Brorstrom-Lunden E., Olafsdottir K., Thorlacius J.M., Leppanen S., Bossi R., Skov H., Manoe S., Patton G.W., Stern G., Sverko E. and Fellin P. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Program (AMAP): 1993-2006. *Science of The Total Environment*, 408 (2010), 2854-2873.

Ishihara A., Nishiyama N., Sugiyama S. and Yamauchi K. The effect of endocrine disrupting chemicals on thyroid hormone binding to Japanese quail transthyretin and thyroid hormone receptor. *General and Comparative Endocrinology*, 134 (2003), 36-43.

Ivarsson A., Andersen M.B., Johnson U., and Lindwall M. To adjust or not adjust: Nonparametric effect sizes, confidence intervals, and real-world meaning. *Psychology of Sport and Exercise*, 14 (2013), 97-102.

Jarque S., and Piña B. Deiodinases and thyroid metabolism disruption in teleost fish. *Environmental Research*, 135 (2014), 361-375.

Jensen A.A. and Leffers H. Emerging endocrine disruptors: perfluoroalkylated substances. *International Journal of Andrology*, 31 (2008), 161-169.

Joensen U.N., Veyrand B., Antignac J., Jensen M.B., Petersen J.H., Marchand P., Skakkebæk N.E., Andersson A., Le Bizec B. and Jørgensen N. PFOS (perfluorooctanesulfonate) in serum is negatively associated with testosterone levels, but not with semen quality, in healthy men. *Human Reproduction*, 28 (2013), 599-608.

Jones P.D., Hu W., De Coen W., Newsted J.L. and Giesy J.P. Binding of perfluorinated fatty acids to serum proteins. *Environmental Toxicology and Chemistry*, 11 (2003), 2639-2649.

Jones T., Smith C., Williams E., and Ramsay A. Breeding performance and diet of Great Skuas *Stercorarius* skua and Parasitic Jaegers (Arctic Skuas) *S. parasiticus* on the west coast of Scotland. *Bird Study*, 55 (2008), 257-266.

Kannan K., Tao L., Sinclair E., Pastva S.D., Jude D.J. and Giesy J.P. Perfluorinated Compounds in Aquatic Organisms at Various Trophic Levels in a Great Lakes Food Chain. *Archives Environmental Contamination and Toxicology*, 48 (2005), 559-566.

Key B.D., Howell R.D. and Criddle C.S. Fluorinated Organics in the Biosphere. *Environmental Science and Technology*, 31 (1997), 2445-2454.

Langlois I., Berger U., Zencak Z., and Oehme M. Mass spectral studies of perfluorooctane sulfonate derivatives separated by high-resolution gas chromatography. *Rapid Communication in Mass Spectrometry*, 21 (2007), 3547-3553.

Lau C., Anitole K., Hodes C., Lai D., Pfahles-Hutchens A. and Seed J. Perfluoroalkyl Acids: A Review of Monitoring and Toxicological Findings. *Toxicological Sciences*, 99 (2007), 336-394.

Leach L.F., and Henson R.K. The Use and Impact of Adjusted R^2 Effects in Published Regression Research. *Multiple Linear Regression Viewpoints*, 33 (2007), 1-11.

Leat E.H.K., Bourgeon S., Eze J.I., Muir D.C.G., Williamson M., Bustnes J.O., Furness R.O., and Borgå K. Perfluoroalkyl substances in eggs and plasma of an avian top predator, Great skua (*Stercorarius skua*), in the north Atlantic. *Environmental Toxicology and Chemistry*, 32 (2013), 569-576.

Lehmler H.J. Synthesis of environmentally relevant fluorinated surfactants - a review. *Chemosphere*, 58 (2005), 1471-1496.

Letcher R.J., Bustnes J.O., Dietz R., Jenssen B.M., Joergensen E.H., Sonne C., Verreault J., Vijayan M. M. and Gabrielsen G.W. Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish. *Science of The Total Environment*, 408 (2010), 2995-3043.

Li A.C., and Glass C.K. PPAR- and LXR-dependent pathways controlling lipid metabolism and the development of atherosclerosis. *Journal of Lipid Research*, 45 (2004), 2161-2173.

Lockhart W.L., Wagemann R., Tracey B., Sutherland D. and Thomas D.J. Presence and implications of chemical contaminants in the freshwaters of the Canadian Arctic. *Science of The Total Environment*, 122 (1992), 165-243.

Loveless S.E., Finlay C., Everds N.E., Frame S.R., Gillies P.J., O'Connor J.C., Powley C.R., and Kennedy G.L. Comparative responses of rats and mice exposed to linear/branched, linear, or branched ammonium perfluorooctanoate (APFO). *Toxicology*, 220 (2006), 203-217.

Löfstrand K., Jörundsdóttir H., Tomy G., Svavarsson J., Weihe P., Nygård T., and Bergman Å. Spatial trends of polyfluorinated compounds in guillemot (*Uria aalge*) eggs from North-Western Europe. *Chemosphere*, 72 (2008), 1475-1480.

Luebker D.J., Hansen K.J., Bass N.M., Butenhoff J.L., and Seacat A.M. Interactions of fluorochemicals with rat liver fatty acid-binding protein. *Toxicology*, 176 (2002), 175-185.

Mallory M.L. and Braune B.M. Tracking contaminants in seabirds of Arctic Canada: Temporal and spatial insights. *Marine Pollution Bulletin*, 64 (2012), 1475-1484.

Martin J.W., Mabury S.A., Solomon K.R., and Muir D.C.G. Bioconcentration and tissue distribution of perfluorinated acids in rainbow trout (*Oncorhynchus mykiss*). *Environmental Toxicology and Chemistry*, 22 (2009), 196-204.

McNabb, A.F.M. The Hypothalamic-Pituitary-Thyroid (HPT) Axis in Birds and Its Role in Bird Development and Reproduction. *Critical Reviews in Toxicology*, 37 (2007), 163-193.

Mehlum F. and Gabrielsen G.W. The diet of high-arctic seabirds in coastal and ice-covered, pelagic areas near the Svalbard archipelago. *Polar Research*, 12 (1993), 1-20.

Miljeteig C., Strøm H., Gavrilo M.V., Volkov A., Jenssen B.M., and Gabrielsen G.W. High Levels of Contaminants in Ivory Gull *Pagophila eburnea* Eggs from the Russian and Norwegian Arctic. *Environmental Science and Technology*, 43 (2009), 5521-5528.

Miljeteig C., and Gabrielsen G.W. Contaminants in Brünnich's guillemots from Kongsfjorden and Bjørnøya in the period from 1993 to 2007. *Brief Report Series*, Norwegian Polar Institute (2010).

Moody C.A. and Field J.A. Perfluorinated Surfactants and the Environmental Implications of Their Use in Fire-Fighting Foams. *Environmental Science and Technology*, 34 (2000), 3864-3870.

Muir D.C.G and de Wit C.A. Trends of legacy and new persistent organic pollutants in the circumpolar arctic: Overview, conclusions and recommendations. *Science of The Total Environment*, 408 (2010), 3044-3051.

Newsted J.L., Jones P.D., Coady K.K, and Giesy J.P. Avian Toxicity Reference Values for Perfluorooctane Sulfonate. *Environmental Science and Technology*, 39 (2005), 9357-9362.

Newsted J.L., Coady K.K., Beach S.A., Butenhoff J.L., Gallagher S., and Giesy J.P. Effects of perfluorooctane sulfonate in mallard and northern bobwhite quail exposed chronically via the diet. *Environmental Toxicology and Pharmacology*, 23 (2007), 1-9.

Norwegian Meteorological Institute, http://met.no/Klima/Klimastatistikk/Vanlig_var/Trondelag_og_Nord-Norge/Tromso_Troms/, retrieved 23.04.2015.

Nøst T.H., Helgason L.B., Harju M., Heimstad E.S., Gabrielsen G.W. and Jenssen B.M. Halogenated organic contaminants and their correlations with circulating thyroid hormones in developing Arctic seabirds. *Science of the Total Environment*, 414 (2012), 248-256.

Oakes K.D., Sibley P.K., Martin J.W., Maclean D.D., Solomon K.R., Mabury S.A. and Van Der Kraak G.J. Short-term exposure of fish to perfluorooctane sulfonate: Acute effects on fatty acyl-CoA oxidase activity, oxidative stress, and circulating sex steroids. *Environmental Toxicology and Chemistry*, 24 (2005), 1172-1181.

O'Donald P. The Arctic Skua. *Cambridge University Press*, Cambridge, (1983).

Parsons J.R., Sáez M., Dolfing J. and de Voogt P. Biodegradation of Perfluorinated Compounds. *Reviews of Environmental Contamination and Toxicology*, 196 (2008), 53-71.

Perner K., Leipe Th., Kuijpers A., Mikkelsen N., Andersen T.J. and Harff J. Contamination of arctic Fjord sediments by Pb-Zn mining at Maarmorilik in central West Greenland. *Marine Pollution Bulletin*, 60 (2010), 1065-1073.

Powley C.R., George S.W., Ryan T.W., and Buck R.C. Matrix Effect-Free Analytical Methods for Determination of Perfluorinated Carboxylic Acids in Environmental Matrixes. *Analytical Chemistry*, 77 (2005), 6353-6358.

Prevedouros K., Cousins I.T., Buck R.C. and Korzeniowski S.H. Sources, Fate and Transport of Perfluorocarboxylates. *Environmental Science and Technology*, 40 (2006), 32-44.

Rayne S., Forest K., and Friesen K.J. Congener-specific numbering systems for the environmentally relevant C_4 through C_8 perfluorinated homologue groups of alkyl sulfonates, carboxylates, telomer alcohols, olefins, and acids, and their derivatives. *Journal of Environmental Science and Health, Part A: Toxic/Hazardous Substances and Environmental Engineering*, 43 (2008), 1391-1401.

Riddell N., Arsenault G., Benskin J.P., Chittim B., Martin J.W., McAlees A., and McCrindle R. Branched Perfluorooctane Sulfonate Isomer Quantification and Characterization in Blood Serum Samples by HPLC/ESI-MS(MS). *Environmental Science and Technology*, 43 (2009), 7902-7908.

Rigét F., Bossi R., Sonne C., Vorkamp K., and Dietz R. Trends of perfluorochemicals in Greendland ringed seals and polar bears: Indications of shifts to decreasing trends. *Chemosphere*, 93 (2013), 1607-1614.

Rotander A., Karrman A., van Bavel B., Polder A., Riget F., Audonsson G.A., Vikingsson G., Gabrielsen G.W., Bloch D. and Dam M. Increasing levels of long-chain perfluorocarboxylic acids (PFCAs) in Arctic and North Atlantic marine mammals, 1984-2009. *Chemosphere*, 86 (2012), 278-285.

Rønning B., Moe B., Chastel O., Broggi J., Langset M., and Bech C. Metabolic adjustments in breeding female kittiwakes (*Rissa tridactyla*) include changes in kidney metabolic intensity. *Journal of Comparative Physiology B: Biochemical, Systemic, and Environmental Physiology*, 178 (2008), 779-784.

Seacat A.M., Thomford P.J., Hansen K.J., Olsen G.W., Case M.T., and Butenhoff J.L. Subchronic Toxicity Studies on Perfluorooctanesulfonate Potassium Salt in Cynomolgus Monkeys. *Toxicological Sciences*, 68 (2002), 249-264.

Shi X., Liu G., Wu G., and Zhou B. Waterborne exposure to PFOS causes disruption of the hypothalamus-pituitary-thyroid axis in zebrafish larvae. *Chemosphere*, 77 (2009), 1010-1018.

Stahl T., Mattern D. and Brunn H. Toxicology of perfluorinated compounds. *Environmental Sciences Europe*, 23 (2011), 1-52.

Stock N.L., Furdui V.I., Muir D.C.G. and Mabury S.A. Perfluoroalkyl Contaminants in the Canadian Arctic: Evidence of Atmospheric Transport and Local Contamination. *Environmental Science and Technology*, 41 (2007), 3529-3536.

Tao L., Kannan K., Kajiwara N., Costa M.M., Fillmann G., Takahashi S., and Tanabe S. Perfluorooctanesulfonate and Related Fluorochemicals in Albatrosses, Elephant Seals, Penguins, and Polar Skuas from the Southern Ocean. *Environmental Science and Technology*, 40 (2006), 7642-7648.

Tartu S., Gabrielsen G.W., Blévin P., Ellis H., Bustnes J.O., Herzke D., and Chastel O. Endocrine and Fitness Correlates of Long-Chain Perfluorinated Carboxylates Exposure in Arctic Breeding Black-Legged Kittiwakes. *Environmental Science and Technology*, 48 (2014), 13504-13510.

Thibodeaux J.R., Hanson R.G., Rogers J.M., Grey B.E., Barbee B.D., Richards J.H., Butenhoff J.L., Stevenson L.A., and Lau C. Exposure to Perfluorooctane Sulfonate during Pregnancy in Rat and Mouse. I: Maternal and Prenatal Evaluations. *Toxicological Sciences*, 74 (2003), 369-381.

Tomy G.T., Budakowski W., Halldorson T., Helm P.A., Stern G.A., Friesen K., Pepper K., Tittlemier S.A. and Fisk A.T. Fluorinated Organic Compounds in an Eastern Arctic Marine Food Web. *Environmental Science and Technology*, 38 (2004), 6475-648.

Ucán-Marín F., Arukwe A., Mortensen A., Gabrielsen G.W., Fox G.A., and Letcher R.J. Recombinant Transthyretin Purification and Competitive Binding with Organohalogen Compounds in Two Gull Species (*Larus argentatus* and *Larus hyperboreus*). *Toxicological Sciences*, 107 (2009), 440-450.

Vanden Heuvel J.P., Thompson J.T., Frame S.R., and Gillies P.J. Differential Activation of Nuclear Receptors by Perfluorinated Fatty Acid Analogs and Natural Fatty Acids: A comparison of Human, Mouse, and Rat Peroxisome Proliferator-Activated Receptor-α, -β, and -γ, Liver X Receptor-β, and Retinoid X Receptor-α. *Toxicological Sciences*, 92 (2006), 476-489.

Verreault J., Skaare J.U., Jenssen B.M. and Gabrielsen G.W. Effects of Organochlorine Contaminants on Thyroid Hormone Levels in Arctic Breeding Glaucous Gulls, Larus hyperboreus. *Environmental Health Perspectives*, 112 (2004), 532-537.

Verreault J., Houde M., Gabrielsen G.W., Berger U., Haukås M., Letcher R.J., and Muir D.C.G. Perfluorinated Alkyl Substances in Plasma, Liver, Brain, and Eggs of Glaucous Gulls (*Larus hyperboreus*) from the Norwegian Arctic. *Environmental Science and Technology*, 39 (2005), 7439-7445.

Verreault J., Bech C., Letcher R.J., Ropstad E., Dahl E. and Gabrielsen G.W. Organohalogen contamination in breeding glaucous gulls from the Norwegian Arctic: Associations with basal metabolism and circulating thyroid hormones. *Environmental Pollution*, 145 (2007a),138-145.

Verreault J., Berger U., and Gabrielsen G.W. Trends of Perfluorinated Alkyl Substances in Herring Gull Eggs from Two Coastal Colonies in Northern Norway: 1983-2003. *Environmental Science and Technology*, 41 (2007b), 6671-6677.

Wang Y., Starling A.P., Haug L.S., Eggesbo M., Becher G., Thomsen C., Travlos G., King D., Hoppin J.A., Rogan W.J., and Longnecker M.P. Association between perfluoroalkyl substances and thyroid stimulating hormone among pregnant women: a cross-sectional study. *Environmental Health*, 12 (2013), 76.

Wang Y., Rogan W.J., Chen P-C., Lien G-W., Chen H-Y., Tseng Y-C., Longnecker M.P., and Wang S-L. Association between Maternal Serum Perfluoroalkyl Substances during Pregnancy and Maternal and Cord Thyroid Hormones: Taiwan Maternal and Infant Cohort Study. *Environmental Health Perspectives*, 122 (2014), 529-534.

Webster G.M., Venners S.A., Mattman A., and Martin J.W. Associations between Perfluoroalkyl acids (PFASs) and maternal thyroid hormones in early pregnancy: A population-based cohort study. *Environmental Research*, 133 (2014), 338-347.

Wei Y., Liu Y., Wang J., Tao Y., and Dai J. Toxicogenomic analysis of the hepatic effects of perfluorooctanoic acid on rare minnows (*Gobiocypris rarus*). *Toxicology and Applied Pharmacology*, 226 (2008), 285-297.

Weiss J.M., Andersson P.L., Lamoree M.H., Leonards P.E.G., van Leeuwen S.P.J., and Hamers T. Competitive Binding of Poly- and Perfluorinated Compounds to the Thyroid Hormone Transport Protein Transthyretin. *Toxicological Sciences*, 109 (2009), 206-216.

Welcker J., Chastel O., Gabrielsen G.W., Guillaumin J., Kitaysky A.S., Speakman J.R., Tremblay Y., and Bech C. Thyroid Hormones Correlate with Basal Metabolic Rate but Not Field Metabolic Rate in a Wild Bird Species. *PLoS ONE*, 8 (2013), e56229.

Yamashita N., Taniyasu S., Petrick G., Wei S., Gamo T., Lam P.K.S. and Kannan K. Perfluorinated acids as novel chemical tracers of global circulation of ocean waters. *Chemosphere*, 70 (2008), 1247-1255.

Yen P.M., Ando S., Feng X., Liu Y., Maruvada P. and Xia X. Thyroid hormone action at the cellular, genomic and target gene levels. *Molecular and Cellular Endocrinology*, 246 (2006), 121-127.

Young C.J., Furdui V.I., Franklin J., Koerner R.M., Muir D.C.G. and Mabury S.A. Perfluorinated Acids in Arctic Snow: New Evidence for Atmospheric Formation. *Environmental Science and Technology*, 41 (2007), 3455-3461.

Yu W-G., Liu W., J Y-H. Effects of Perfluorooctane Sulfonate on Rat Thyroid Hormone Biosynthesis and Metabolism. *Environmental Toxicology and Chemistry*, 28 (2009), 990-996.

Yu W-G., Liu W., Liu L., J Y-H. Perfluorooctane sulfonate increased hepatic expression of OAPT2 and MRP2 in rats. *Archives of toxicology*, 85 (2011), 613-621.

Zhao Z., Xie Z., Möller A., Sturm R., Tang J., Zhang G., and Ebinghaus R. Distribution and long-range transport of polyfluoroalkyl substances in the Arctic, Atlantic Ocean and Antarctic coast. *Environmental Pollution*, 170 (2012), 71-77.

Zoeller R.T. Challenges Confronting Risk Analysis of Potential Thyroid Toxicants. *Risk Analysis*, 23 (2003), 143-162.

Zoeller R.T., Tan S.W., Tyl R.W. General Background on the Hypothalamic-Pituitary-Thyroid (HPT) Axis. *Critical Reviews in Toxicology*, 37 (2007), 11-53.

Appendix A

Table A1: A list over the perfluoroalkyl and polyfluoroalkyl substances (PFASs) included in the internal standard used during the extraction and clean-up processes of PFAS analysis.

PFAS	Concentration (ng/µL)
PFBA	0.1
PFPA	0.1
PFHxA	0.1
PFHpA	0.1
PFOA	0.1
PFNA	0.1
PFDcA	0.1
PFUnDA	0.1
PFDoDA	0.1
PFTeDA	0.1
PFHxS	0.0946
PFOS	0.0956
PFOSA	0.1
PFHxPA	0.3
6:2 FTS	0.095

Appendix B

Table B1: Individual measurements of skull, wing, and tarsus length, mass, total triiodothyronine (TT3) and total thyroxine (TT4) in male (denoted "M") and female (denoted "F") black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard in 2013 and 2014.

Bird ID	Year	Metal ring	Sex	Skull (mm)	Wing (mm)	Tarsus (mm)	Mass (g)	TT3 (ng/mL)	TT4 (ng/mL)
KOC13-96	2013	6217915	M	92.4	314	34.5	395	1.93	41.72
KOC13-97	2013	6227149	M	96.8	327	36.7	450	3.86	20.33
KOC13-98	2013	6217908	M	93.6	310	36.7	435	2.5	48.06
KOC13-100	2013	6177713	M	96.5	306	33.8	390	2.89	21.27
KOC13-102	2013	6177361	M	91.9	314	30.6	405	2.66	36.73
KOC13-103	2013	6177704	M	96.6	322	35.7	405	2.13	18.61
KOC13-105	2013	6177771	M	97.3	320	36.5	395	2.5	37.25
KOC13-111	2013	6146117	M	93.8	321	35.6	430	2.2	36.89
KOC13-114	2013	NA	M	91.8	335	34.8	400	2.43	35.58
KOC13-115	2013	NA	M	93.5	325	33.7	345	2.55	27.31
KOC13-117	2013	NA	M	94.6	323	35.1	430	1.9	26.59
KOC13-120	2013	6218018	M	94.5	319	36.8	415	1.75	-
KOC13-122	2013	NA	M	93	323	33.8	420	3.29	45.45
KOC13-125	2013	6227133	M	91.6	324	34.9	415	3.57	70.05
KOC13-128	2013	6217998	M	93.8	315	33.4	400	2.81	45.92
KOC13-79	2013	6227019	F	86.8	312	33.8	410	3.53	79.83
KOC13-101	2013	6124137	F	86.3	314	31.1	355	1.93	24.53
KOC13-104	2013	6217916	F	92.9	307	34.9	405	1.15	34.39
KOC13-110	2013	6227150	F	88.4	317	34	365	4.22	60.35
KOC13-112	2013	6217901	F	90	306	35.1	340	2.32	24.61
KOC13-113	2013	NA	F	89.1	314	35.3	395	1.72	31.64
KOC13-116	2013	6124124	F	89.2	314	29.6	365	1.63	52.69
KOC13-119	2013	6218106	F	86.4	319	31.8	375	1.9	26.01
KOC13-121	2013	6124135	F	88.4	320	35.5	370	2.03	30.14
KOC13-123	2013	6227132	F	86.4	309	33.4	400	1.98	-
KOC13-124	2013	6167503	F	88.1	298	35.5	425	2.94	24.6
KOC13-127	2013	6227134	F	90.8	310	35.5	390	2.36	31.62
KOC-14-46	2014	6227017	M	95.4	323	35.7	475	3.4	42.54
KOC-14-47	2014	6146158	M	92.9	310	35.3	485	2.12	38.35
KOC-14-48	2014	6217914	M	92.2	311	34.9	455	2.69	40.18
KOC-14-49	2014	6177763	M	92.1	312	34.8	485	3.72	36.51
KOC-14-50	2014	6146196	M	91.6	324	34.3	445	2.13	38.31
KOC-14-51	2014	6217903	M	93.4	313	32	455	3.79	34.32
KOC-14-52	2014	6217913	M	92.3	322	35.5	415	4.59	34.98
KOC-14-56	2014	6223880	M	94.9	315	33.4	445	1.9	38.41
KOC-14-57	2014	6177734	M	94.3	308	36.4	470	3.15	23.93
KOC-14-58	2014	6172945	M	92.6	317	34.9	475	3.66	44.38
KOC-14-59	2014	6218112	M	94.9	325	34.7	430	2.8	50.21
KOC-14-60	2014	6223988	M	91.8	323	35.7	475	2.46	20.73
KOC-14-63	2014	6202868	M	97.6	321	34.5	450	3.32	31.5

Table B1 continued: Individual measurements of skull, wing, and tarsus length, mass, total triiodothyronine (TT3) and total thyroxine (TT4) in male (denoted "M") and female (denoted "F") black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard in 2013 and 2014.

KOC-14-65	2014	6124118	M	94.3	312	37.6	440	2.08	23.27
KOC-14-66	2014	6217918	M	98.6	318	35.9	495	2.73	47.74
KOC-14-53	2014	6202843	F	87.7	305	35.9	435	4.47	51.78
KOC-14-54	2014	6177890	F	88.1	302	34	380	3.4	33.29
KOC-14-55	2014	6223886	F	86.7	319	34.2	385	3.89	56.57
KOC-14-62	2014	6218046	F	86	305	34.4	400	3.49	42.41
KOC-14-64	2014	6177774	F	88.6	312	33.8	385	3.1	23.72
KOC-14-67	2014	6146187	F	90.1	318	35.8	415	3.31	37.02
KOC-14-68	2014	6223997	F	89.8	312	34.2	405	4.01	42.38
KOC-14-69	2014	6223983	F	88.2	311	35.2	420	4.49	42.08
KOC-14-70	2014	6177963	F	85.6	308	32.6	390	4.37	18.79
KOC-14-71	2014	6218012	F	91.1	320	34	400	4.01	43.94
KOC-14-74	2014	6146191	F	89.5	310	34.3	425	4.05	34.29
KOC-14-79	2014	6202322	F	90	321	33.2	405	1.12	9.53
KOC-14-80	2014	6223986	F	94.4	322	35.5	430	2.1	36.17
KOC-14-61	2014	6218015	F	88.9	321	34.8	380	3.35	37.15

Table B2: A summary of the concentrations (ng/ml plasma) of perfluorinated alkylated substances (PFASs) detected in male and female black-legged kittiwakes (*Rissa tridactyla*) caught in 2013 and 2014 in Kongsfjorden, Svalbard. The data is presented as mean ± standard deviation (SD), median, and range. *n* denotes the sample size.
*There were significantly higher levels in males than females for linPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), sumPFOS (Kruskal-Wallis and Dunn's post hoc test, $p < 0.001$), PFUnDA (ANOVA and Tukey's post hoc test, $p = 0.025$), PFDoDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.045$), PFTrDA (Kruskal-Wallis and Dunn's post hoc test, $p = 0.003$), and PFTeDA (ANOVA and Tukey's post hoc test, $p = 0.004$).

	Males				Females			
PFAS (ng/mL)	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
brPFOS	0.180 ± 0.188	0.113	0.01 - 0.669	30	0.0983 ± 0.0484	0.0912	0.02 - 0.164	14
linPFOS*	7.33 ± 3.58	6.43	2.47 - 19.3	30	4.34 ± 3.14	3.6	0.937 - 14.6	26
sumPFOS*	4.47 ± 3.72	6.5	2.50 - 19.9	30	4.47 ± 3.25	3.7	0.937 - 15.0	26
PFNA	1.53 ± 1.09	1.21	0.377 - 5.61	30	1.25 ± 0.745	1.02	0.415 - 3.34	26
PFDA	1.29 ± 0.519	1.2	0.392 - 2.65	30	1.11 ± 0.438	1.07	0.360 - 2.08	26
PFUnDA*	6.94 ± 2.42	6.14	3.47 - 12.2	30	5.62 ± 1.74	5.48	2.47 - 9.49	26
PFDoDA*	1.58 ± 0.713	1.63	0.429 - 2.96	30	1.16 ± 0.526	1.21	0.158 - 2.07	26
PFTrDA*	5.26 ± 1.81	5.18	2.22 - 8.42	30	3.67 ± 1.40	3.85	1.20 - 6.07	26
PFTeDA*	0.659 ± 0.397	0.653	0.0841 - 1.64	30	0.371 ± 0.291	0.417	0.01 - 1.01	26

Table B3: Individual concentrations of perfluoroalkyl and polyfluoroalkyl subtances (PFASs) in black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard in 2013 and 2014. Concentrations in brackets are under the limit of detection (LOD) and have been replaced by a randomly selected concentration between zero and the LOD.

Bird ID	brPFOS (ng/mL)	linPFOS (ng/mL)	sumPFOS (ng/mL)	PFNA (ng/mL)	PFDA (ng/mL)	PFUnDA (ng/mL)	PFDoDA (ng/mL)	PFTrDA (ng/mL)	PFTeDA (ng/mL)
KOC13-96	0.67	10.8	11.5	1.48	1.36	8.14	1.66	6.33	0.44
KOC13-97	0.11	6.95	7.07	1.67	0.63	3.87	0.55	3.14	0.20
KOC13-98	0.07	12.9	12.9	3.74	1.23	5.56	0.87	4.41	0.20
KOC13-100	0.44	8.24	8.67	0.86	0.86	5.77	0.91	3.58	0.42
KOC13-102	(0.01)	5.78	5.78	1.86	1.23	5.71	0.92	4.64	0.27
KOC13-103	(0.03)	3.93	3.93	0.94	0.61	4.58	0.57	2.92	0.08
KOC13-105	(0.01)	6.66	6.67	1.42	1.76	6.31	1.22	3.35	0.12
KOC13-111	(0.01)	4.45	4.45	1.13	0.94	5.09	0.90	3.77	0.46
KOC13-114	0.61	13.0	13.6	5.61	2.34	10.7	1.77	4.45	0.48
KOC13-115	0.07	6.26	6.33	1.23	1.37	6.00	1.16	3.79	0.45
KOC13-117	0.23	11.4	11.6	2.55	1.54	6.69	1.13	3.44	0.29
KOC13-120	0.05	5.80	5.85	1.15	0.97	4.57	1.09	4.13	0.54
KOC13-122	(0.04)	4.01	4.01	1.34	0.39	3.47	0.43	2.22	0.17
KOC13-125	0.59	19.3	19.9	3.86	2.65	12.2	2.20	6.73	0.60
KOC13-128	0.25	5.78	6.03	1.63	1.10	4.98	0.59	2.49	0.24
KOC13-79	-	4.84	5.59	1.51	1.15	4.80	0.82	3.80	0.21
KOC13-101	-	9.20	9.41	2.66	1.50	6.13	0.65	3.22	0.20
KOC13-104	-	2.27	2.38	1.02	0.96	3.56	0.75	2.33	(0.03)
KOC13-110	-	1.63	1.70	1.02	0.50	2.73	0.20	1.42	(0.01)
KOC13-112	-	11.6	12.0	2.92	2.01	9.49	1.31	3.11	0.11
KOC13-113	-	1.74	1.74	1.74	0.91	4.61	0.50	1.24	(0.03)
KOC13-116	-	5.14	5.20	1.20	2.08	8.18	1.20	3.89	0.22
KOC13-119	-	1.24	1.24	0.54	0.48	3.22	0.16	1.95	(0.04)
KOC13-121	-	14.6	15.0	3.34	1.68	7.35	1.14	3.28	0.12
KOC13-123	-	0.94	0.94	0.42	0.36	2.47	0.39	1.20	0.05
KOC13-124	-	1.71	1.76	1.12	1.10	4.46	0.70	1.52	(0.03)
KOC13-127	-	3.61	3.61	2.00	1.73	8.98	1.22	3.99	0.11
KOC-14-46	0.180	6.64	6.82	0.99	1.18	7.41	2.19	7.20	1.06
KOC-14-47	0.127	5.47	5.60	1.17	1.29	8.15	2.41	7.74	1.31
KOC-14-48	0.288	10.2	10.5	1.23	2.20	10.4	2.49	7.69	0.87
KOC-14-49	0.079	6.60	6.68	1.06	1.62	8.90	2.56	7.83	1.32
KOC-14-50	0.194	5.62	5.82	0.58	1.13	6.26	1.74	5.45	0.77
KOC-14-51	0.188	5.01	5.20	0.91	0.98	5.88	1.84	6.47	0.91
KOC-14-52	(0.04)	2.47	2.50	0.38	0.77	3.55	1.26	4.25	0.78
KOC-14-56	0.138	6.98	7.11	1.39	1.15	6.02	1.69	5.53	0.92
KOC-14-57	0.090	4.26	4.35	1.33	1.14	6.51	1.93	5.53	1.17
KOC-14-58	0.101	5.31	5.41	0.51	0.98	5.57	1.60	4.99	0.71
KOC-14-59	0.455	12.1	12.6	1.20	2.09	11.9	2.95	7.71	0.78
KOC-14-60	0.050	4.24	4.29	0.94	0.87	5.51	1.52	5.38	0.78
KOC-14-63	0.114	7.46	7.58	1.09	1.40	9.57	2.52	8.42	1.64
KOC-14-65	0.159	5.16	5.31	1.62	1.60	9.60	2.42	7.03	0.89

Table B3 continued: Individual concentrations of perfluoroalkyl and polyfluoroalkyl subtances (PFASs) in black-legged kittiwakes (*Rissa tridactyla*) caught in Kongsfjorden, Svalbard in 2013 and 2014. Concentrations in brackets are under the limit of detection (LOD) and have been replaced by a randomly selected concentration between zero and the LOD.

KOC-14-66	(0.02)	7.01	7.01	1.09	1.35	9.27	2.40	7.12	0.90
KOC-14-53	0.137	3.60	3.73	0.73	1.26	6.65	2.07	5.62	0.57
KOC-14-54	0.079	3.98	4.06	0.68	1.05	6.15	1.94	5.32	0.73
KOC-14-55	0.163	6.39	6.55	0.76	1.35	6.52	1.83	5.49	0.57
KOC-14-62	0.142	4.51	4.65	1.23	1.39	6.06	1.73	6.06	0.85
KOC-14-64	(0.04)	3.59	3.63	0.69	0.87	5.51	1.44	4.20	0.70
KOC-14-67	(0.04)	3.38	3.38	0.59	0.91	5.35	1.38	3.91	0.42
KOC-14-68	0.164	3.76	3.93	1.00	1.09	5.39	1.28	3.76	0.45
KOC-14-69	0.151	3.72	3.87	1.21	0.74	5.88	1.39	4.36	0.46
KOC-14-70	0.104	3.56	3.66	1.02	0.87	5.45	1.72	4.43	0.42
KOC-14-71	0.121	5.80	5.92	1.55	1.19	6.79	1.79	5.63	1.01
KOC-14-74	(0.02)	2.65	2.65	0.73	0.74	4.84	1.21	4.15	0.62
KOC-14-79	0.076	2.73	2.80	0.66	0.70	4.06	1.07	3.42	0.62
KOC-14-80	0.070	3.75	3.82	0.94	0.99	6.59	1.34	3.82	0.53
KOC-14-61	0.070	2.89	2.96	1.13	1.10	4.86	1.01	4.23	0.56

Appendix C

Table C1: Individual measurements of skull, wing, and tarsus length, mass, total triiodothyronine (TT3) and total thyroxine (TT4) in male (denoted "M") and female (denoted "F") arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard in 2014.

Bird ID	Year	Metal ring	Sex	Skull (mm)	Wing (mm)	Tarsus (mm)	Mass (g)	TT3 (ng/mL)	TT4 (ng/mL)
ST1-14	2014	6228571	M	78.4	344	47.3	447	3.06	28.75
ST2-14	2014	6217934	F	77.7	332	47.2	497	-	-
ST3-14	2014	6228551	F	79.5	339	45	527	3.2	46.68
ST4-14	2014	6228552	M	76.1	329	45.2	417	-	80.18
ST5-14	2014	5184807	F	81.6	359	45.3	515	2.36	38.97
ST6-14	2014	6228573	F	73.5	342	46.5	499	-	-
ST7-14	2014	6217936	M	76.3	342	47.7	472	3.53	41.28
ST8-14	2014	6217938	M	77.9	341	44.7	425	2.43	25.02
ST9-14	2014	5184808	M	76.8	325	45.9	442	4.99	47.2
ST10-14	2014	6228574	M	76.8	338	45.8	472	3.26	40.41
ST11-14	2014	6223849	F	81.3	351	46.7	491	3.01	11.21
T1-BH-14	2014	6218805	F	74	345	45.6	445	1.61	31.94
T2-BH-14	2014	6218813	M	77.4	335	43.9	375	2.18	40.75
T3-BH-14	2014	6218811	F	76.8	331	43.4	525	1.9	24.15
T4-BH-14	2014	6218808	M	75.1	327	43.9	390	2.64	57.26
T5-BH-14	2014	6218810	M	80.0	338	44.5	400	3.64	59.04

Table C2: A summary of the concentrations (ng/mL whole blood) of perfluoroalkyl and polyfluoroalkyl substances (PFASs) detected in male and female arctic skuas (*Stercorarius parasiticus*) caught in 2014 on Brensholmen, Norway and in Kongsfjorden, Svalbard. The data is presented as mean ± standard deviation (SD), median, and range. *n* denotes the sample size.
*Male arctic skuas had significantly higher levels than females of linPFOS, sumPFOS, PFDA, PFUnDA, PFDoDA, and PFTrDA (ANOVA and Tukey's post hoc test, p = 0.008, 0.010, 0.005, 0.008, 0.010, and 0.005 respectively).

	Males				Females			
PFAS (ng/mL)	Mean ± SD	Median	Range	*n*	Mean ± SD	Median	Range	*n*
linPFOS*	10.0 ± 3.89	10.2	4.29 - 17.1	9	4.95 ± 2.22	5.53	1.78 - 7.78	7
sumPFOS*	10.4 ± 4.25	10.8	4.29 - 17.5	9	5.08 ± 2.33	5.68	1.78 - 8.10	7
PFNA	0.924 ± 1.33	0.561	0.02 - 4.36	9	0.593 ± 0.374	0.768	0.01 - 0.941	7
PFDA*	1.50 ± 0.405	1.4	1.03 - 2.35	9	0.837 ± 0.387	0.79	0.361 - 1.43	7
PFUnDA*	6.98 ± 1.92	6.98	4.43 - 9.85	9	4.11 ± 1.71	4.29	1.41 - 6.21	7
PFDoDA*	1.45 ± 0.445	1.63	0.755 - 2.02	9	0.796 ± 0.429	0.742	0.177 - 1.386	7
PFTrDA*	3.48 ± 1.21	3.69	1.89 - 5.36	9	1.75 ± 0.704	1.83	0.583 - 2.64	7
FOSA	1.45 ± 1.05	1.24	0.05 - 2.78	9	0.664 ± 0.215	0.575	0.473 - 1.08	7

Table C3: Individual concentrations of perfluoroalkyl and polyfluoroalkyl subtances (PFASs) in arctic skuas (*Stercorarius parasiticus*) caught on Brensholmen, Norway and in Kongsfjorden, Svalbard in 2014. Concentrations in brackets are under the limit of detection (LOD) and have been replaced by a randomly selected concentration between zero and the LOD.

Bird ID	linPFOS (ng/mL)	sumPFOS (ng/mL)	PFNA (ng/mL)	PFDA (ng/mL)	PFUnDA (ng/mL)	PFDoDA (ng/mL)	PFTrDA (ng/mL)	FOSA (ng/mL)
ST1-14	8.56	8.56	0.755	1.40	6.11	1.14	3.76	0.196
ST2-14	5.53	5.70	0.776	0.638	3.08	0.528	1.53	0.536
ST3-14	7.78	8.10	0.768	0.790	4.29	0.742	1.92	0.473
ST4-14	7.60	7.69	0.614	1.72	9.24	1.97	3.69	1.81
ST5-14	5.15	5.15	0.941	1.06	6.21	1.14	2.64	1.08
ST6-14	6.19	6.49	0.912	1.13	5.08	1.10	2.48	0.554
ST7-14	10.2	10.8	1.15	1.08	4.80	1.02	2.15	0.744
ST8-14	12.4	15.0	4.36	1.38	4.43	0.755	1.89	0.050
ST9-14	6.06	6.06	0.454	1.25	6.98	1.67	3.52	1.24
ST10-14	4.29	4.29	0.216	1.03	5.53	1.15	2.15	1.07
ST11-14	1.78	1.78	0.132	0.455	3.03	0.499	1.30	0.833
T1-BH-14	6.14	6.28	0.610	1.43	5.70	1.39	1.83	0.597
T2-BH-14	11.1	11.1	(0.02)	1.49	8.08	1.63	4.00	2.42
T3-BH-14	2.09	2.09	(0.01)	0.361	1.41	0.177	0.58	0.575
T4-BH-14	12.7	12.8	0.182	1.75	7.82	1.70	5.36	2.77
T5-BH-14	17.1	17.5	0.561	2.35	9.85	2.02	4.75	2.75

Appendix D

Table D1: A list over the molecular weight (g/mol) of perfluoroalkyl and polyfluoroalkyl substances (PFASs) included in statistical analysis.

PFAS	Molecular weight (g/mol)
PFNA	464.0
PFDA	514.0
PFUnDA	563.9
PFDoDA	614.0
PFTrDA	664.0
PFTeDA	714.2
PFOS	500.0
FOSA	499.14