NTNU

Norwegian University of
Science and Technology

Do levels of per- and polyfluorinated alkylated substances (PFASs) in snow bunting eggs increase with proximity to airports in Svalbard?

Pernille Kvernland

Environmental Toxicology and Chemistry
Submission date: May 2017
Supervisor: Bjørn Munro Jenssen, IBI
Co-supervisor: Geir W. Gabrielsen, Norsk Polar Institutt
Dorte Herzke, Norsk Institutt for Luftforskning

Norwegian University of Science and Technology
Department of Biology

NTNU

Norwegian University of
Science and Technology

IN COLLABORATION WITH

Norsk institutt for luftforskning
Norwegian Institute for Air Research

COVER PHOTO: Male snow bunting (*Plectrophenax nivalis*)
Photography by Geir W. Gabrielsen

Acknowledgments

This master thesis was written at the department of Biology, at the Norwegian University of Science and Technology (NTNU) in Trondheim, the University Centre in Svalbard (UNIS), and in collaboration with the Norwegian Polar Insitute (NPI).

Main supervisors were Bjørn M. Jenssen (NTNU), Geir W. Gabrielsen (NPI) and Dorte Herzke (Norwegian Institute for Air Research, NILU). Co-supervisors were Frode Fossøy (Norwegian Institute for Nature Research, NINA/NTNU), Bård Stokke (NINA/NTNU) and Tomasz M. Ciesielski (NTNU).

The project was funded by Avinor, The Research Council of Norway (Arctic Field Grant), Jan Christensens endowment and King Haakon the 7th educational fund for Norwegian youth.

I want to express my sincere gratitude and appreciation to my supervisors: thank you for being such enthusiastic researchers and scientists providing me with the opportunity to write a thesis based on snow buntings and their eggs in Svalbard. An additional thank you for your kind words and much appreciated encouragement. Thank you to Bård Stokke, Frode Fossøy and Arne Moksnes for helping me out during fieldwork and for teaching me how to locate snow bunting nests, and for working as living encyclopaedias, answering any question I could think of about any bird species imaginable. An additional thank you to Hanna Sofie Sørhus for keeping an eye out for snow buntings, as well as polar bears, while out in the field. Thank you to the wonderful people at NILU, Tromsø for helping me in my analyses of PFASs, Arntraut Götsch for teaching me the analytical method for extraction of PFASs and Dorte Herzke and Linda Hanssen for performing the analysis, identification and quantification of the analytes screened for.

A special thanks to Silje Marie Kristiansen for your happy spirits during our stay in Longyearbyen, Ny-Ålesund and Tromsø. Thank you for all the good memories and all the fun times we had.

Another big thank you to Frode Fossøy for helping me with the statistical analysis of my data and Tomasz M. Ciesielski for helping me with the principal component analysis.

To my family, fellow students and friends, I would like to thank you all for the wonderful times that provided some well appreciated time off from all the writing, and kept me sane. These past few years would not have been the same without you. I am forever grateful! Lastly, a very special thank you to Kristoffer, for all the patience, positivity and support through difficult times.

Thank you all for believing in me.

Trondheim, May 2017

Pernille Kvernland

Abstract

Per- and polyfluorinated alkylated substances (PFASs) are used in a variety of industrial and consumer applications, including polymer materials and in aqueous fire fighting foam (AFFFs). Research over the last 20 years has shown that these compounds are ubiquitous environmental contaminants, extremely persistent, show toxic effects and accumulate in the food web.

Pollution research in the Arctic has traditionally focused on long range transport (LRT), but local pollution from historical and present day human activities also constitute important pollution sources in the Arctic. The overall aim of the present study was to examine if the use of PFASs in AFFFs at Longyearbyen and Ny-Ålesund airport may constitute a local source of contamination and contribute to the bioaccumulation of PFASs in terrestrial biota in Svalbard. Thus, eggs of snow buntings (*Plectrophenax nivalis*) were collected along a transect from Longyearbyen airport towards Adventdalen and around the airport in Ny-Ålesund in 2016.

A total of 11 perfluorinated carboxylic acids (PFCAs), 9 perfluorinated sulfonic acids (PFSAs) and 2 fluorotelomer sulfonates (FTSs) were detected in the egg content from snow buntings. Σ_{22}PFAS concentrations increased significantly from Adventdalen towards Longyearbyen and Longyearbyen airport. Σ_{22}PFAS concentrations in Ny-Ålesund were within the same range as Longyearbyen. Overall perfluorooctane sulfonate (PFOS) comprised the majority of the contamination burden and was found at highest concentrations at all four locations. PFCAs and PFSAs with carbon chain lengths of five or less were not detected.

Linear PFOS (linPFOS) constituted 78-96.5% of total PFOS in snow bunting eggs. The linPFOS comprised 78.7% and 78.4% at Longyearbyen airport and Longyearbyen, respectively, which were within range of the PFOS electrochemical fluorination (ECF) production standard for AFFFs (76-79% linPFOS) and indicative of a local point source. Adventdalen (89.9%) and Ny-Ålesund (96.5%) had a higher linPFOS percentage indicative of LRT or exposure to old legacy PFAS sources.

The levels of PFASs in snow buntings at Longyearbyen airport were 12-90 times higher compared to the other locations. The PFAS levels at this location were above toxicity threshold described in the literature for possible sensitive bird species and are therefore of toxicological concern. Future studies should elucidate whether or not the snow bunting is a sensitive species with respect to adverse effects of PFASs.

The present study show that the eggs from snow buntings living in proximity of Longyearbyen airport are contaminated with PFAS, indicating the fire fighting training facilities at Longyearbyen airport as a point source of PFASs for the local biota that contribute to the bioaccumulation of PFASs in snow buntings residing here.

Ny-Ålesund differed from the other locations with respect to the PFSA:PFCA profile and the linPFOS:brPFOS profile, indicating a different source of exposure compared to the other locations. Further investigation for possible sources in the Ny-Ålesund area is therefore recommended.

Sammendrag

Per- og polyfluorinerte alkylere forbindelser (PFASer) brukes i en rekke ulike industri- og forbrukervarer, inkludert produksjon av polymerer og i brannslukningsskum (AFFFer). Forskning de siste 20 årene har vist at disse forbindelsene er allestedsnærværende forurensninger i miljøet, svært persistente, gifitge og at de akkumuleres oppover i næringskjeden.

Forurensningsforskning i Arktis har tradisjonelt fokusert på langdistansetransport (LRT) av forurensninger, men lokale forurensinger fra både historisk og moderne menneskelig aktivitet har vist seg å være viktige forurensningskilder i Arktis. Det overordende målet med denne studien var å undersøke om bruken av PFASer i AFFFer ved Longyearbyen og Ny-Ålesund flyplass utgjør en lokal kilde til forurensning som bidrar til bioakkumuleringen av PFASer i den terrestriske biotaen på Svalbard. Dermed ble egg fra snøspurv (*Plectrophenax nivalis*) samlet i et transekt fra Longyearbyen flyplass mot Adventdalen, og rundt flyplassen i Ny-Ålesund i 2016.

Totalt 11 perfluorinerte karboksylsyrer (PFCAer), 9 perfluorinerte svovelsyrer (PFSAer) og 2 fluorotelomer sulfonater (FTSer) ble målt i egginnholdet fra snøspurv. Σ_{22}PFAS konsentrasjoner økte signifikant fra Adventdalen mot Longyearbyen og Longyearbyen flyplass. Σ_{22}PFAS konsentrasjoner in Ny-Ålesund var i samme konsentrasjonsområde som Longyearbyen. Samlet sett utgjorde perfluorooktansulfonat (PFOS) størstedelen av forureningsbyrden og denne forbindelsen ble funnet ved høyest konsentrasjoner ved alle de fire lokasjonene undersøkt. PFCAer og PFSAer med karbonkjedelengde på fem eller mindre ble ikke detektert i noen av prøvene.

Lineær PFOS (linPFOS) utgjorde 78-96.5% av total PFOS i snøspurveggene. Longyearbyen flyplass og Longyearbyen inneholdt henholdsvis 78.7% og 78.4% linPFOS, som er innenfor den elektrokjemiske fluorinerings (ECF) produksjonsstandarden (76-79% linPFOS). Dette indikerer at Longyearbyen flyplass og Longyearbyen har en lokal PFAS kilde. Adventdalen (89.9%) og Ny-Ålesund (96.5%) hadde en høyere prosentverdi av linPFOS, som indikerer LRT eller eksponering til gamle lokale PFAS kilder.

Nivåene av PFASer i snøspurv ved Longyearbyen flyplass var 12-90 ganger høyere sammenlignet med de andre lokasjonene. PFAS nivåene ved denne lokasjonen var også over toksisitetsgrenser beskrevet i litteraturen for mulig sensitive fuglearter, og utgjør dermed en toksikologisk bekymring. Fremtidige studier burde undersøke om snøspurven er en sensitiv art eller ikke, med tanke på effektene PFASer kan ha.

Denne studien viser at egg fra snøspruv som lever i nærheten av Longyearbyen flyplass er forurenset med PFASer. Resultatene indikerer også brannslukningsstasjonene ved Longyearbyen flyplass som en mulig PFAS kilde, og en stor bidragsyter til bioakkumuleringen av PFASer i snøspruvene hekker her.

Ny-Ålesund skiller seg fra de andre lokasjonene når det kommer til både PFSA:PFCA profil og linPFOS:brPFOS profil, dette antyder en annen eksponeringskilde sammenlignet med de andre lokasjonene. Videre undersøkelser etter mulige kilder i Ny-Ålesund området er dermed anbefalt.

Abbreviations

Σ	Sum
ΣPFAS	Sum of individual perfluoroalkylated compounds
ΣFTS	Sum of individual fluorotelomer sulfonates
ΣPFCA	Sum of individual perfluoroalkylated carboxylic acids
ΣPFSA	Sum of individual perfluoroalkylated sulfonic acids
6:2 FTAB	6:2 fluorotelomer sulfonamide alkylbetaine
6:2 FTS	6:2 Fluorotelomer sulfonic acid
8:2 FTS	8:2 Fluorotelomer sulfonic acid
ADV	Adventdalen (Longyearbyen)
AFFF	Aqueous film forming foam (fire-fighting foam)
AIR	Longyearbyen airport
ANOVA	Analysis of variance
blm	Binomial linear models
BFR	Brominated flame retardants
brPFOS	Branched isomers of perfluorooctane sulfonate
ECF	Electrochemical fluorination
FTS	Fluorotelomer sulfonic acids
HPLC-MS	High-performance liquid chromatography - mass spectrometry
ISTD	Internal standard
LC	Liquid chromatogram
LD_{50}	Lethal dose 50%
linPFOS	Linear isomers of perfluorooctane sulfonate
glm	General linear models
LOAEL	Lowest observed adverse effect level
LOD	Limit of detection
LOEL	Lowest observed effect level
LOQ	Limit of quantification
LRT	Long-range transport
LYB	Longyearbyen
$mean_{Ar}$	Arithmetic mean
$mean_{Geo}$	Geometric mean
MS	Mass spectrometry
MS/MS	Tandem mass spectrometry
n	Number of observations
NÅ	Ny-Ålesund
ng	Nanogram (10^{-9}gram)
NPI	Norwegian Polar Institute
NTNU	Norwegian University of Science and Technology
PCA	Principal component analysis
PFAS	Perfluroralkylated substances
PFBA	perfluorobutaneoate
PFBS	Perfluorobutane sulfonate
PFCA	Perfluorocarboxylic acids
PFDcA	Perfluorodecanoate

PFDcS	Perfluorodecane sulfonic acid
PFDoA	Perfluorododecanoate
PFHpA	Perfluoroheptanoate
PFHpS	Perfluoroheptanesulphonic acid
PFHxA	Perfluorohexanoate
PFHxS	Perfluorohexane sulfonate
PFNA	Perfluorononanoate
PFNS	Perfluorononanesulphonic acid
PFOA	Perfluorooctaneoate
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctanesulfonamide
PFPA	Perfluoropentaneoate
PFPS	Perfluoropentanesulfonic acid
PFSA	Perfluorosulfonates
PFTeA	Perfluorotetradecanoate
PFTriA	Perfluorotridecanoic acid
PFUnA	Perfluoroundercanoate
pg	Picogram (10^{-12} gram)
PNEC	Predicted no effect concentration
POP	Persistent organic pollutants
POSF	Perfluorooctanesulfonyl fluoride
PP	Polypropylene
RiS	Research in Svalbard
rpm	Rounds per minute
RSTD	Recovery standard
SD	Standard deviation
SRM	Standard reference material
TH	Thyroid hormone
TRV	Toxicity reference value
TTR	Transthyretin
UHPLC	Ultra-high performance liquid chromatography
μL	Microliter
UNIS	University Centre in Svalbard
USEPA	United States Environmental Protection Agency
ww	Wet weight

Contents

List of Figures

List of Tables

1 Introduction

1.1 Contaminants in the Arctic

Despite the Arctic being a remote location and considered to have minor local potential point sources of contamination, the presence of chemicals that otherwise should not be found in the Arctic can only be explained by long-range transport (LRT) from lower latitudes [Macdonald et al., 2000, Burkow and Kallenborn, 2000]. Major production volumes, in addition to a chemical structure providing stability, allow these chemicals to be transported to remote areas far from where they were first produced and emitted [Muir and de Wit, 2010]. The Arctic is well connected to the other continents through the atmosphere, oceanic currents, transpolar ice packs and large Arctic rivers, which constitute possible pathways for potential contaminants, such as persistent organic pollutants (POPs), to reach the Arctic [Burkow and Kallenborn, 2000].

POPs have been widely documented in the Arctic wildlife since the 1970's, including fish, marine seabirds, terrestrial animals and humans [Barrie et al., 1992, Macdonald et al., 2000, Burkow and Kallenborn, 2000, Gabrielsen, 2007]. Most of these are pollutants that are predominantly produced and emitted in the mid-latitudes and transported to the Arctic [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].

1.2 Brominated Flame Retardants (BFRs)

Brominated flame retardants (BFRs) is a group of POPs containing brominated organic compounds used to make materials more fire resistant [Jenssen et al., 2007]. They react chemically to prevent the spread of a fire by making bromine react wih the burning material in place of oxygen, thus slowing combustion [AMAP, 2007]. BRFs tend to be environmentally stable and once released to the environment they may be subject to long-range transport. BFRs are found across the Arctic, in all environmental compartments and in key species. In addition, these chemicals have been used and produced in large volumes [Jenssen et al., 2007].

The Stockholm Convention have started banning what they refer to as so-called legacy POPs, leading the industry to replace compounds, such as BFRs with alternative flame retardant chemicals that do not contain bromine [AMAP, 2007]. The Stockholm Convention was adopted in Stocholm, Sweeden in 2001 and entered into force in 2004 (www.pops.int). The objective of the Convention is "to protect human health and the environment from persistent organic pollutants". Measures have to be taken to "reduce the total releases derived from anthropogenic sources"; the final goal is ultimate elimination, where feasible [StockholmConvention, 2002, Fiedler, 2007]. As a result of this replacement, emerging groups of POPs (often referred to as re-ermerging and/or novel POPs) are now being found in the Arctic, one of them being the per- and polyfluorinated alkylated substances (PFASs) [Giesy and Kannan, 2001, Haukås et al., 2007, Butt et al., 2010, Muir and de Wit, 2010].

1.3 Per- and Polyfluorinated Alkyl Substances (PFASs)

PFASs are a large group of surface-active organic compounds. Due to chemical and thermal stability, as well as their hydrophobic and lipophobic nature, they have been used for more than 50 years in a number of industrial and commercial applications [Key et al., 1997, Parsons et al., 2008, Zhao et al., 2014]. PFASs can be divided into several classes, among them the perfluoroalkyl carboxylic

acids (PFCAs), perfluoroalkyl sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Perfluorooctaneoate (PFOA), perfluorooctane sulfonate (PFOS) and 8:2 fluorotelomer sulfonic acid (8:2 FTS) are examples of PFCA, PFSA and FTS, respectively [Buck et al., 2011]. The short-chain PFSAs and PFCAs are potentially more water soluble, whereas long-chain PFSAs and PFCAs appear to bind more strongly to particles and accumulate in the food chain [Higgins and Luthy, 2006, Ahrens et al., 2010].

Perfluorinated compounds are primarily produced from two industrial synthetic routes: electrochemical fluorination (ECF) and telomerization. A mixture of structural isomers is produced by ECF, while telomerization conserves the geometry of its starting materials, which are typically linear [De Silva et al., 2009b]. ECF has been going on since the 1950's and telomerisation since the 1970's [Butt et al., 2010]. As a result of these two synthesis routes, PFASs are often found in a mixture between linear and branched isomers in the environment [Rayne et al., 2008]. The 3M Company was the major producer of perfluorooctanesulfonyl fluoride (POSF; PFOS has been suggested as the final degradation product of POSF), starting production in 1949, with the total cumulative production estimated to be approximately 96 000 tonnes in the peak years between 1970 and 2002. After 3M discontinued production in 2001, other companies began production to meet existing market demands, with an estimated 1 000 tonnes per year being produced since 2002 [Paul et al., 2008]. No estimated production volumes for the telomerization process was found. PFOS is exclusively manufactured via ECF and have at least 11 branched isomers, with a theoretical 89 isomers possible [Langlois et al., 2007, Rayne et al., 2008, Buck et al., 2011]. A ratio between the linear and branched isomers can be used to determine the source of the PFAS [De Silva and Mabury, 2004, Riddell et al., 2009]. There are also indications that the different isomers have different toxicological properties [Loveless et al., 2006].

There has been a great interest in evaluating toxicities, human exposure pathways and mode of global transport of PFASs [Kannan, 2011, Pérez et al., 2013]. Notable differences occur in the physical properties of PFASs compared with regular POPs. PFASs are lipophobic and closely associate with proteins, such as albumin (plasma) or fatty acid binding proteins (liver), rather than with lipids [Luebker et al., 2002, Han et al., 2003, Jones et al., 2003]. Therefore, PFASs will mainly accumulate in the liver, kidney and bile secretions [Butt et al., 2010]. PFSAs and PFCAs, in contrast to many chlorinated and brominated POPs, are terminal breakdown products and are not subject to biotransformation. Consequently they are only removed by excretion, and for humans the average half-life is 5.4 years for PFOS and 3.8 years for PFOA [Olsen et al., 2007, Stahl et al., 2011]. For avian species the half-life has been estimated to be 125 days for PFOS and 5.2 days for PFOA in male white leghorn chickens (*Gallus gallus*) [Yoo et al., 2009]. This long half-life in organisms will affect the potential for bioaccumulation of PFASs and thus their occurrence in the environment [Butt et al., 2010, Leat et al., 2013]. In addition, PFASs have been shown to be immunotoxic, hepatotoxic and developmentally toxic in addition to being suspected endocrine disrupting chemicals [Lau et al., 2007].

Although PFASs were commercialized over 50 years ago, they received little attention as contaminants in wildlife until Giesy and Kannan reported their presence in marine mammals, fish-eating birds, and marine and freshwater fish from around the world [Giesy and Kannan, 2001]. Kannan et al. (2004) reported the presence of PFASs in human blood from several countries [Kannan et al., 2004].

The concern over potential effects on the environment and human health of PFASs has led to the launch of several research programs on PFASs' sources, fate, transport and toxicity [Martin et al., 2004, Paul et al., 2008, Buck et al., 2011]. As previously mentioned, PFOS and related compounds were phased-out of production by their major manufacturer (3M Co.) in 2001. Furthermore, a stewardship agreement between the United States Environmental Protection Agency (USEPA) and eight leading global manufacturers, was launched to reduce 95 % of emissions of PFOA and related chemicals by 2010, and to work towards elimination by 2015. In addition to this, PFOS was also included in the Stockholm Convention on persistent organic pollutants as an Annex B substance in 2009 [Martin et al., 2004, Buck et al., 2011, Xie et al., 2013, Gewurtz et al., 2013]. However, due to lack of cost-effective alternative technology in certain applications, PFOS and related substances are still manufactured and used in China [Letcher et al., 2010, Zhang et al., 2012].

1.4 Airport Activity and AFFFs

Elevated levels of PFASs in water, soil and sediment samples around airports were identified already in the late 1990s [Cancilla et al., 1998]. The primary source of the contamination around airports is the uncontrolled historical use of PFASs (such as PFOS and 6:2 fluorotelomer sulfonic acid (6:2 FTS)) in aqueous film forming foams (AFFFs) during fire-fighting training and accidents [Awad et al., 2011, Kwadijk et al., 2014, Wang et al., 2016].

Everyday activities at airports include several different polluting activities (e.g. combustion of aviation fuels, cleaning, maintenance and repair of air crafts, de/anti-icing operations, removal of snow, weed and vegetation from the airport apron etc.). To maintain these operations, airports use a wide variety of chemicals, resulting in the environmental release of these together with run-off waters. Airports also need preparedness for accidental fires, and part of the airport therefore functions as a fire station (often located in the vicinity of the airports) [Granberg et al., 2016]. Training exercises may occur on a weekly basis or even several times per week depending on the site [Baduel et al., 2015]. These training sites have recently received extensive attention, since continuous use of fire-fighting foams has led to serious contamination of soil, ground water and watersheds with PFASs such as PFOS and PFOA released with run-off water [Granberg et al., 2016].

Hydrocarbon-fuel fires pose a serious threat to life and property, and require immediate response. To enable a quick reaction to fuel fires, effective and efficient fire-extinguishing agents are needed to prevent damage and re-ignition of the fires. AFFFs were introduced in the 1960s when the 3M Company, together with the United States Navy, developed AFFFs to extinguish hydrocarbon-fuel fires [Moody and Field, 2000, Schultz et al., 2004].

AFFF formulations are complex chemical mixtures of surfactants and other components, such as fluorosurfactants, hydrocarbon surfactants, solvents and co-solvents [Schultz et al., 2004]. The fluorochemical surfactants were mainly produced using PFOS and PFOA, but now mainly consist of 6:2 fluorotelomer sulfonamide alkylbetaine (6:2 FTAB) [Kishi and Arai, 2008, Moe et al., 2012]. Due to their surface-tension lowering properties, AFFFs containing fluorinated surfactants have superior fire-fighting capabilities compared to nonfluorinated fire extinguishing methods [Moody and Field, 1999, Place and Field, 2012]. AFFFs also have a secondary benefit of preventing re-ignition [Moody and Field, 2000]. The AFFF chemical make-up is not well known and likely varies among manufacturers, between batches and between production year [Erten-Unal et al.,

1998, Place and Field, 2012, D'Agostino and Mabury, 2013]. However, the manufacturing of PFOS increased when PFOS-based AFFFs became the product of choice for fire-fighting due to effectiveness, long shelf-life and ease of use [Paul et al., 2008].

Elevated levels of PFOS have been reported in water and biological samples, such as molluscs (*Mytilus edulis*), turtles (*Chelydra serpentina*), and wild mink (*Neovison vison*) downstream from airports with a history of fire-fighting training activities [Kärrman et al., 2011, De Solla et al., 2012, Persson et al., 2013]. Elevated levels of PFOS and perfluorohexanesulfonic acid (PFHxS) were also found in humans drinking water from a private well contaminated with AFFF from a nearby airport in Cologne, Germany [Weiss et al., 2012]. Similarly, AFFF contaminated drinking water has been suggested as one plausible factor behind increasing exposure to PFHxS in Uppsala (Sweden) residents [Glynn et al., 2012].

1.5 Local Pollution

All human activities leave traces in the environment, and most modern civilization activities result in some type of pollution. Pollution research in the Arctic has traditionally focused on the long range transport of pollutants from the industrialized middle regions to the pristine uninhabisted north [Butt et al., 2010, Hung et al., 2010]. Today it is observed that local pollution from historical and present day human activities may constitute important pollution sources in the Arctic [Granberg et al., 2016]. Considering there are several larger or smaller airports located across the Arctic, AFFF contamination might represent a local contamination source throughout the Arctic.

1.5.1 Longyearbyen

Longyearbyen (Figure 1.1) is the largest settlement and the administrative center of Svalbard. The resident population is just above 2000, but increases drastically during tourist seasons reaching over 100 000 "guest nights" in 2013, and is steadily increasing. Longyearbyen has a history of coal mining but the only active mine today is Mine 7. Today most of the garbage and waste from Longyearbyen is sent to mainland Norway for incineration. Sewage from the municipality and the airport is collected and released untreated into the Adventfjord recipient [Granberg et al., 2016]. There are two fire-fighting training areas around the airport, one old area north-east of the runway and a new active area south-east of the runway (Figure 3.1a), and the airport uses the training area in cooperation with the local fire brigade [Avinor, 2012]. The airport has been investigated regarding their use and release of contaminating chemicals with particular recent focus on PFASs. PFASs above restricted limits were detected at the old fire-fighting training site, spreading in an easterly direction (Figure 3.1a) [Granberg et al., 2016].

1.5.2 Ny-Ålesund

Ny Ålesund (Figure 1.1) was founded as a mining settlement by Kings Bay AS in 1917 and terminated as such in 1963. It is now run exclusively as an international research facility, hosting $\sim$ 25 persons in winter and $\sim$ 200 in the summer. Problematic sites in terms of contamination are the old landfill and dumping sites, the fuel storage in the settlement, the closed mining areas, the airport and the sewage system. There is currently no overview of chemical use or spills from the airport in Ny Ålesund. In 2016 the governor of Svalbard requested information from Kings Bay specifically regarding the use of firefighting foams. PFAS containing fire-fighting foams have likely been used here and perhaps in conjunction with the mines. It is, however, unclear to what extent and where this activity has taken place [Granberg et al., 2016].

Figure 1.1: A map of Spitsbergen (scale 1:1 000 000) provided by Bernt Bye (NPI).

1.6 Study Species: The snow bunting *(Plectrophenax nivalis)*

The terrestrial animal life on Svalbard is very limited due to the harsh climate. However, the birdlife is rich, especially during the summer [Axelson, 2014].

The snow bunting is a small *Emberizid* finch reaching 15-17 cm in length and weighing 30-40 g. Together with the Svalbard rock ptarmigan *(Lagopus muta hyperborea)*, the snow bunting is the only terrestrial bird species breeding regularly on Svalbard, and the only passerine bird species that regularly breed in the high Arctic environment [Smith, 1994, Hoset et al., 2009, Skøien, 2015]. The snow bunting may breed on all Arctic islands and in Greenland, Fenno-Scandinavia, Russia (east to about 60°) and the Canadian Arctic [Banks et al., 1989, Ryzhanovsky, 2015].

The Svalbard population is estimated to exist of 1 000 - 10 000 breeding pairs and believed to spend the winter in the steppes of Kazakhstan, near the Caspian Sea [Gwiazdowicz et al., 2012, Pilskog et al., 2014]. The first males usually arrive from the wintering grounds in late March to early April, 6-8 weeks before nesting and 2-4 weeks ahead of the females, in order to establish territories [Meltofte, 1983]. The breeding season starts at the end of May and the beginning of June, and lasts until end of July [Hoset et al., 2004, Fossøy et al., 2014]. However, the length of the breeding season in Spitsbergen vary considerably from one year to another depending on the climatic conditions of the year [Hoset et al., 2009]. Snow buntings make nests in open habitats (crevices, cavities and boulders) throughout the Arctic, but also breed in nooks and cavities of

buildings and other man-made constructions, such as nest boxes [Hoset et al., 2009, Smith, 1994].

The female lay 4-7 eggs which are incubated for 12-13 days [Falconer et al., 2008]. Nestlings are exclusively fed invertebrates (insects and spiders) and may fledge from the nest within 13 days after hatching [Maher, 1964, Smith, 1994]. Snow buntings usually display monogamy and are often single brooded, but may raise two broods in a single season if the year is good [Smith, 1994, Hoset et al., 2009]. Their diet changes from mainly invertebrates (insects and spiders) during summer to vegetable matter (seeds, grass, sedges and rushes) during autumn and winter, but they will feed on invertebrates if the opportunity arises [Smith, 1994].

1.6.1 Recent Studies

Recent studies have shown that local sources of POPs may play a role for levels of pollutants in biota. For instance, it has been shown that there are differences in the concentrations of POPs in eggs of snow buntings and that they differ among the four settlements in Svalbard (Longyearbyen (LYB), Ny-Ålesund (NÅ), Barenstburg and Pyramiden). Whereas levels of the "old" legacy POPs tended to be higher in Barentsburg and Pyramiden, the levels of PFASs were highest in Longyearbyen [Kristoffersen, 2012].

Due to their long migratory route across the North-Atlantic from Northern Norway to Svalbard, it is assumed that the snow buntings are income breeders, meaning they allocate energy for breeding from recently ingested resources. This in contrast to capital breeders that allocate energy for breeding from endogenous reserves [Drent and Daan, 1980]. Since snow buntings replenish their energy storages after the spring migration and allocate this energy for reproduction, and egg laying, it is likely to assume that the contaminants in their eggs mainly have a local origin [Kristoffersen et al., 2012].

Passerine birds are useful for monitoring local pollution because of their small home ranges, territories and foraging areas [Dauwe et al., 2003]. The contaminant levels in egg yolk and egg white are directly indicative of the levels that can be found in the mother birds [Drouillard and Norstrom, 2001], thus applying eggs for biomonitoring is therefore regarded as a well suited non-invasive method of sampling [Helgason et al., 2010]. Eggs of snow buntings therefore represent a good non-destructive biological matrix for investigating bioavailability and uptake of organic pollutants in Arctic terrestrial ecosystems. Eens et al. (2013) also suggest that eggs from starlings (*Sturnus vulgaris* and *Sturnus unicolor*), another passerine species, also can be used as a biomonitoring tool on a large geographical scale [Eens et al., 2013].

2 Aims

The overall aim of the present study was to examine if the use PFASs in AFFFs at the airports in Longyearbyen and Ny-Ålesund may constitute a local source of contamination and contribute to bioaccumulation of PFASs in terrestrial biota in Svalbard.

The hypothesis is that the contamination burden of PFASs will be higher in eggs from snow buntings collected from nests located closer to Longyearbyen airport (AIR) than those located in Adventdalen (ADV). However, considering that AFFFs with PFASs have not been used at Longyearbyen airport since 2005 (C.E. Ianssen, Operations manager Longyearbyen airport, Avinor; pers. comm.), it is not believed that the levels will be high enough to cause serious damage to breeding success and/or fitness. Thus, eggs of snow buntings were collected from Longyearbyen airport and westwards towards the end of the air field and eastwards towards Adventdalen (Figure 3.1a). In Ny-Ålesund (Figure 3.1b) eggs were sampled at different distances from the air field. Biometric variables (height, width, volume, eggshell thickness etc.) were recorded and concentrations of PFASs then analysed in the eggs.

Specific objectives of this study include:

- Compare the PFAS level and PFAS profile at each location to investigate if there is any pattern with respect to transportation of these suspected non-volatile compounds in the environment.
- Compare levels of PFAS detected in the snow bunting eggs with other peer-reviewed studies to investigate if levels might exceed thresholds for adverse effects on breeding success or fitness of the birds.
- Evaluate whether or not the level of PFAS contamination in eggs from snow buntings increase with proximity to airports in Svalbard where AFFFs likely and/or previously have been used.
- Compare PFAS levels in eggs from Ny-Ålesund and Longyearbyen to examine if there is any differences between a small settlement in Ny-Ålesund and the largest settlement in Svalbard (Longyearbyen).

3 Materials and Methods

3.1 Sampling Areas

Fieldwork was conducted in Longyearbyen (78°N, 15°E) and Ny-Ålesund (78°N, 11°E). Longyearbyen was divided into three sub-locations, depending on distance from the main area of focus; Longyearbyen airport (AIR; from Longyearbyen harbour to the west end of the fence surrounding the air strip), Longyearbyen (LYB; Longyearbyen harbour to the end of Isfjorden) and Adventdalen (ADV; end of Isfjorden to Mine 6), which is considered to be a reference area. Eggs were sampled in Kongsfjorden (Ny-Ålesund) where the use of AFFFs are unknown but assumed, making NÅ a possible pristine area based on known PFAS contamination sources.

3.2 Field Procedures

The sampling was performed during nesting season in June 2016, from 03.06-11.06 and 21.06-29.06 in Longyearbyen and 12.06-20.06 in Ny-Ålesund. The fieldwork in Longyearbyen was coordinated with the activity of another NTNU-project on snow buntings, registered in the Research in Svalbard (RiS) database (RiS-ID 2273). The eggs were all sampled from natural nest sites (not nest boxes).

Nests were located by visual observation of the behaviour of male and female snow buntings. To minimize disturbance of reproduction success, only one egg was obtained from nests with ≥ 3 eggs (late egg laying period). As a result, the female is less likely to abandon the nest due to human disturbance [Bolduc and Guillemette, 2003]. Upon discovery of the nest, the eggs were checked for viability by controlling if they were warm, and (if appropriate), a floatation test was conducted to determine stage of development [van Paassen et al., 1984].

After sampling, the nests were checked regularly, as described by Falconer et al. (2008), every two or three days to keep updated information about the nest condition and to determine clutch size as well as clutch initiation [Falconer et al., 2008]. The nests were also checked to make sure the nest was not abandoned after sampling or predated by Arctic foxes. Number of hatchlings and/or fledglings was also observed. Where possible, non-viable eggs still in the nest after chicks had fledged, were sampled for comparison and statistical purposes. In total, 10 non-viable eggs were collected, the majority from ADV (n=8) and some from Longyearbyen airport (n=1) and Longyearbyen (n=1).

3.2.1 Sample Collection

The egg to be sampled was chosen at random. The sample was then wrapped in acetone-rinsed aluminium foil and placed in a zip lock bag marked with its respective sample number, nest identification, GPS-position, clutch size at time of sampling and date of sampling. It was then placed in a storage box with cotton. If possible, the eggs were cooled in a refrigerator prior to being frozen (-18°C), and the eggs were measured (height and width) in frozen condition to prevent eggs from cracking. Egg volume was then calculated using Hoyt's equation:

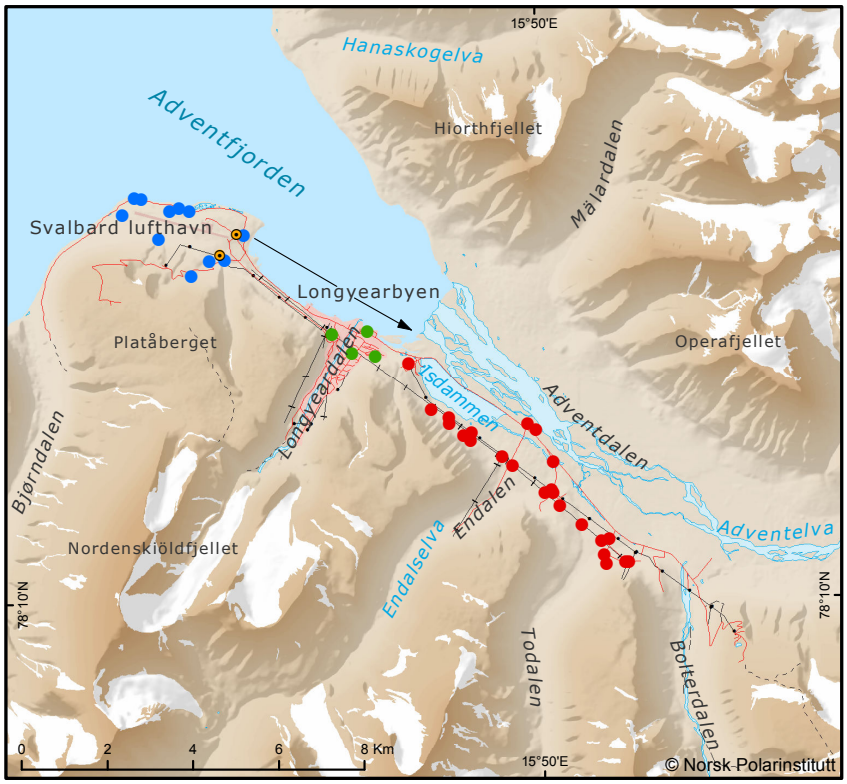
$$Volume = 0.51 \cdot LB^2 \tag{1}$$

Where L is the lenght and B is the breadth (width; maximum diameter) [Romanoff and Romanoff, 1949, Hoyt, 1979].

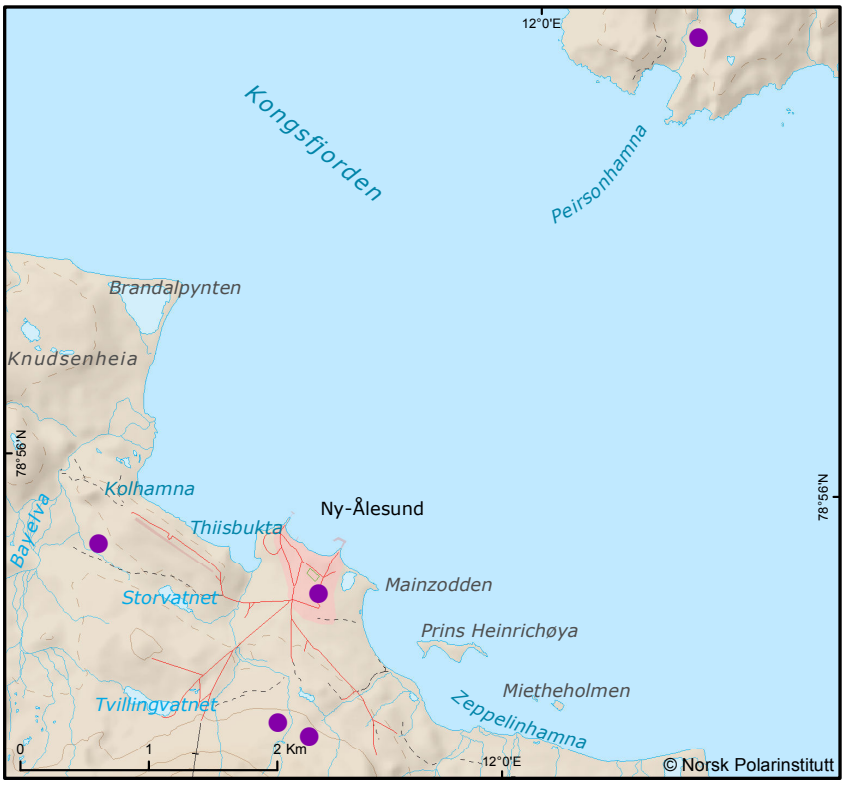
In total, 45 eggs were sampled (Figure 3.1), constituting both viable and non-viable eggs collected, from 43 different nests. For the two duplicate samples the values were averaged, resulting in a total sample size of 43 (Appendix, Figure A.1 and A.2). The duplicates were tested for differences in correlation of PFAS concentrations between eggs in the same nest (Appendix, Figure A.3 to A.4). One duplicate sample consisted of one viable and one non-viable egg (sample 6 and 43 - "new sample 6"), while the other duplicate consisted of two non-viable eggs collected (sample 38 and 41 - "new sample 38").

The samples were transported from Svalbard to Tromsø in frozen condition and kept at -18°C until further analysis.

The study on snow bunting eggs and the conduction of fieldwork was approved by the Governor of Svalbard (reference number 16/00595-2). The project is also registered in the RiS database (RiS-ID 10447).



(a) Longyearbyen (scale 1:150 000)



(b) Ny-Ålesund (scale 1:50 000)

Figure 3.1: An overview of the location of the nests where eggs from snow buntings (*Plectrophenax nivalis*) were sampled in **(a)** Longyearbyen and **(b)** Ny-Ålesund in 2016. **(a)** also include the two fire training areas (orange and black marks), the old station north-east for the runway and the new station south-east of the runway. Arrow denotes how PFASs spread in an easterly direction from Longyearbyen airport towards Adventdalen. Different colors annotate samples collected within the different locations; Longyearbyen airport (blue), Longyearbyen (green), Adventdalen (red) and Ny-Ålesund (purple). The majority of samples were collected in Longyearbyen (n=40) and a few samples collected from Ny-Ålesund (n=5). Maps provided by Bernt Bye (NPI).

3.3 Chemical Analysis

Sample preparation and analysis of PFASs was conducted at the Norwegian Institute for Air Research (NILU) in Tromsø, Norway. All samples were analysed during August 2016 under supervision of chemists with long experience and expertise. Analysis, identification and quantification of PFAS compounds were performed by highly experienced chemists from NILU.

The samples were analysed for 11 PFCAs, 9 PFSAs and 2 FTSs (22 different PFAS compounds in total). Rawdata for each sample can be seen in the Appendix (Table A.1). Specifics for each compound analysed is listed in Table 3.1, together with their chemical name, abbreviation, chemical structure, molecular weight and CAS-registry number.

Table 3.1: A list of the per- and polyfluorinated alkylated substances (PFASs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*) caught in Longyearbyen, Svalbard and Ny-Ålesund, Svalbard in 2016. The PFASs analysed for are divided into perfluoroalkyl carboxylic acids (PFCAs), perfluorinated sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs).

Group	Chemical Name	Abbreviation	Chemical Structure	Molecular Weight	CAS-ID Number
PFCA	Perfluorobutaneoate	PFBA	$C_4HF_7O_2$	214.0	375-22-4
	Perfluoropentaneoate	PFPA	$C_5HF_9O_2$	264.1	2706-90-3
	Perfluorohexanoate	PFHxA	$C_6HF_{11}O_2$	314.1	307-24-4
	Perfluoroheptanoate	PFHpA	$C_7HF_{13}O_2$	364.1	375-85-9
	Perfluorooctaneoate	PFOA	$C_8HF_{15}O_2$	414.1	335-67-1
	Perfluorononanoate	PFNA	$C_9HF_{17}O_2$	464.1	375-95-1
	Perfluorodecanoate	PFDcA	$C_{10}HF_{19}O_2$	514.1	335-76-2
	Perfluoroundercanoate	PFUnA	$C_{11}HF_{21}O_2$	564.1	2058-94-8
	Perfluorododecanoate	PFDoA	$C_{12}HF_{23}O_2$	614.1	307-55-1
	Perfluorotridecanoic acid	PFTriA	$C_{13}HF_{25}O_2$	664.1	72629-94-8
	Perfluorotetradecanoate	PFTeA	$C_{14}HF_{27}O_2$	714.1	376-06-7
PFSA	Perfluorobutane sulfonate	PFBS	$C_4HF_9O_3S$	300.1	375-73-5
	Perfluoropentanesulfonic acid	PFPS	$C_5HF_{11}O_3S$	350.1	220-310-2
	Perfluorohexane sulfonate	PFHxS	$C_6HF_{13}O_3S$	400.1	432-50-7
	Perfluoroheptanesulphonic acid	PFHpS	$C_7HF_{15}O_3S$	450.1	375-92-8
	Perfluorooctanesulfonamide	PFOSA	$C_8H_2F_{17}NO_2S$	499.1	754-91-6
	Perfluorooctane sulfonate	linPFOS	$C_8HF_{17}O_3S$	500.1	1763-23-1
	Perfluorooctane sulfonate	brPFOS	$C_8HF_{17}O_3S$	500.1	1763-23-1
	Perfluorononanesulphonic acid	PFNS	$C_9HF_{19}O_3S$	550.1	375-95-1
	Perfluorodecane sulfonic acid	PFDcS	$C_{10}HF_{21}O_3S$	600.1	335-77-3
FTS	6:2 Fluorotelomer sulfonic acid	6:2 FTS	$C_8H_5F_{13}O$	364.1	647-42-7
	8:2 Fluorotelomer sulfonic acid	8:2 FTS	$C_{10}H_5F_{17}O$	464.1	678-39-7

3.3.1 Sample Preparation

In order to achieve good extraction efficiency and representativeness, some matrices need pretreatment prior to the extraction procedure. Solid samples, such as animal tissues, need to be homogenized. In the beginning of the sample preparation, and prior to the extraction, internal standards are added to the samples to correct for the loss of analyte during sample preparation. For extraction of surfactants, such as PFASs, sonication is utilized. The samples are then placed in an ultrasonic bath where the ultrasonic waves disrupt the cell membranes and thereby facilitate extraction. Clean-up of the samples is then performed in order to remove fat and other interfering

compounds. For samples with expected low concentrations (as was expected in this study) the extract is up-concentrated, the solvents evaporated, to be able to detect the analytes. The samples are then transferred to vials and recovery standards added and the samples run on an analysing instrument. Blank samples and reference samples are often used to perform quality control and quality assessments of the analysed samples.

3.3.2 Homogenization

The eggs were allowed to thaw in room temperature over night prior to sample preparation. The eggshell was carefully removed, the developmental stage of the foetus determined and the egg content homogenized. If the egg content was all egg (i.e undeveloped embryo) or not well-developed (small embryo), the content was homogenized using a spatula. Homogenization of developed chicks were performed with a scalpel. The sample volume was insufficient to use an Ultra-Turrax or immersion blender.

Eggshell was kept for measuring eggshell thickness using a micrometer. The eggshell was allowed to dry prior to measurements, and (if possible) all measurements were performed on the widest part of the egg. The inner eggshell membrane was not removed.

3.3.3 Procedure

The egg samples were analysed for PFASs following a method developed by Powley et al. (2005) and modified by Hanssen et al. (2013) [Powley et al., 2005, Hanssen et al., 2013].

Extraction
Homogenate (0.47-2.71 gram (g)) was transferred to polypropylene (PP) tubes (50 milliliter (mL)) and spiked with internal standard (20 microliter (μL); PFAS ISTD, 0.5 nanogram (ng) / μL, 29.06.16) and methanol (4 mL) before vortexing. The samples were extracted using an ultrasonic bath (Ultrasonic Cleaner, 3 x 10 min) and vortexted between each cycle. For sedimentation, the samples were centrifuged (2000 rounds per minute (rpm), 5 min).

Cleaning
Supernatant was transferred to new PP tubes (10 mL) and up-concentrated (1 mL) using RapidVap (Rapid Vap; Labconco Corp., Kansas City, MO, USA). Eppendorf tubes (1.7 mL) were then prepared with ENVI-Carb 120/400 (25 mg $\pm$ 1 mg) (Supelco, PN, USA). The up-concentrated supernatant extract (0.8 mL) was added to the eppendorf tubes along with glacial acetic acid (50 μL) and vortexed. Next, the samples were sentrifuged (10 000 rpm, 10 min), the supernatant (0.5 mL) added to an glass vial (1.5 mL) along with a reference standard (20 μL; 3,7-dimethyl PFOA; br-PFDA) and vortexed once more. The final extract (100 μL) was then transferred to an LC-vial with insert, and stored cold until the instrumental analysis.

3.3.4 Instrumental Analysis

Quantification of the different PFASs was performed by ultra-high pressure liquid chromatography triplequadropole mass spectrometry (UHPLC-MS/MS) The instrumental analysis was performed on a Thermo Scientific quaternary Accela 1250 pump (Thermo Fisher Scientific Inc., Waltham, MA, USA) with a PAL sample manager (Thermo Fisher Scientific Inc., Waltham, MA, USA) coupled to a Thermo Scientific Vantage MS/MS (Vantage TSQ) (Thermo Fisher Scientific Inc., Waltham, MA, USA); The sample (10 μL) was injected on a Waters Acquity UPLC HSS 3 T

column (2.1 x 100 mm, 1.8 μm) (Waters Corporation, Milford, MA, USA) equipped with a Waters Van guard HSS T3 guard column (2.1 x 5 mm, 1.8 μm) (Waters Corporation, Milford, MA, USA). Separation was achieved using 2 mM NH_4OAc in 90:10 methanol/water and 2 mM NH_4OAc in methanol as the mobile phases [Hanssen et al., 2013].

3.3.5 Quantification

Quantification was conducted using the LCQuan software from Thermo Scientific (Version 2.6) (Thermo Fisher Scientific Inc., Waltham, MA, USA). A standard curve was calculated based on known concentrations of ^{12}C and ^{13}C labelled PFASs, and equation 2 was used in the quantification.

$$C_{sample} = \frac{R_f(C_{Std} * Area_{Sample})}{Area_{Std}} \tag{2}$$

C_{sample} is the concentration of the sample while C_{std} is the concentration of the standard. $Area_{sample}$ is the area under the curve of the sample obtained from the chromatography while $Area_{std}$ is the area under the curve of the standard obtained from the chromatography. R_f is the response factor from the areas and concentrations of the ^{12}C and ^{13}C labelled equivalents in the chromatography.

3.3.6 Quality Assurance

For every batch, usually every 5-10 samples, one tube with a blank and one with a standard reference material (SRM; Pike-perch sample QM 03-2, Quasimeme) were included in the analysis for quality control of the method and validation of repeatability. The blank contained all reagents without the egg content, and the SRM tube contained a reference with known concentrations of PFASs. Both the blank and the SRM underwent the exact same treatment as the tubes with samples. This, together with an internal and recovery standard, formed the basis for the quality assurance. The internal standard contained known concentrations of ^{13}C labelled PFASs and was added at the very beginning of the extraction, with the assumption of an equal loss of the internal standard as the ^{12}C PFAS in the sample through the extraction and clean-up process. 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 acceptable range and the blank samples below the limit of acceptable contamination determined by the laboratory. Limit of detection (LOD) was set to threefold the instrumental noise and limit of quantification (LOQ) was set to 3 x LOD.

3.4 Statistical Analysis

Statistical analysis was performed using Excel (Microsoft Office 365 ProPlus, Version 1612) for Windows and the free statistical software R (version 3.3.2, R Development Core Team, Vienna, Austria, 2016) [R Core Team, 2016]. When using R the following packages were used: ggplot [Wickham, 2009], vegan [Oksanen et al., 2017] and plyr [Wickham, 2011].

Variables that were not normal distributed were log-transformed for approximating normality. A statistical significance level was set to $p \leq 0.05$. When testing differences between locations the data were tested using one-way analysis of variance (ANOVA) and Tukey's post hoc test. Preliminary binomial (blm) and general linear (glm) models were used to examine possible significance between PFAS concentration and the different biometric variables.

For calculations of mean (arithmetic and geometric), standard deviation (SD), median and max- and min-value, presented in tables, values below LOD were set to LOD/2. No individuals were excluded due to deviating results in the analyses (outliers), as individuals with deviating results may contain important information for biological effect studies. Concentrations of contaminants are presented and analysed in wet weight (ww) concentrations, since wet weight has been shown to be more relevant for exploring biological effects.

Different contaminant ratios were examined in order to evaluate any possible differences in the composition of the PFAS patterns between the different locations. Thus, profile between the two contaminant groups PFSAs and PFCAs were examined as well as the isomer profile between linear PFOS (linPFOS) and branched PFOS (brPFOS).

3.4.1 Data Treatment of Samples <LOD

PFASs detected in $\geq 40\%$ of the samples were included in the statistical analysis. This eliminated the following compounds: PFOSA, PFPS, PFBS, PFHpS, PFNS, PFDcS, PFBA, PFPA, PFHpA and 6:2 FTS from the statistical analysis (Appendix, Table A.4). For compounds detected in $\geq 40\%$ of the samples, individual samples with compounds quantified to be under LOD were assigned LOD/2 in order to avoid missing values in the dataset. At least one sample contained a value below LOD for all compounds analysed for. All samples measured $<$ LOQ were treated equally as other samples. LOD and LOQ for the individual compounds are presented in the Appendix (Table A.2).

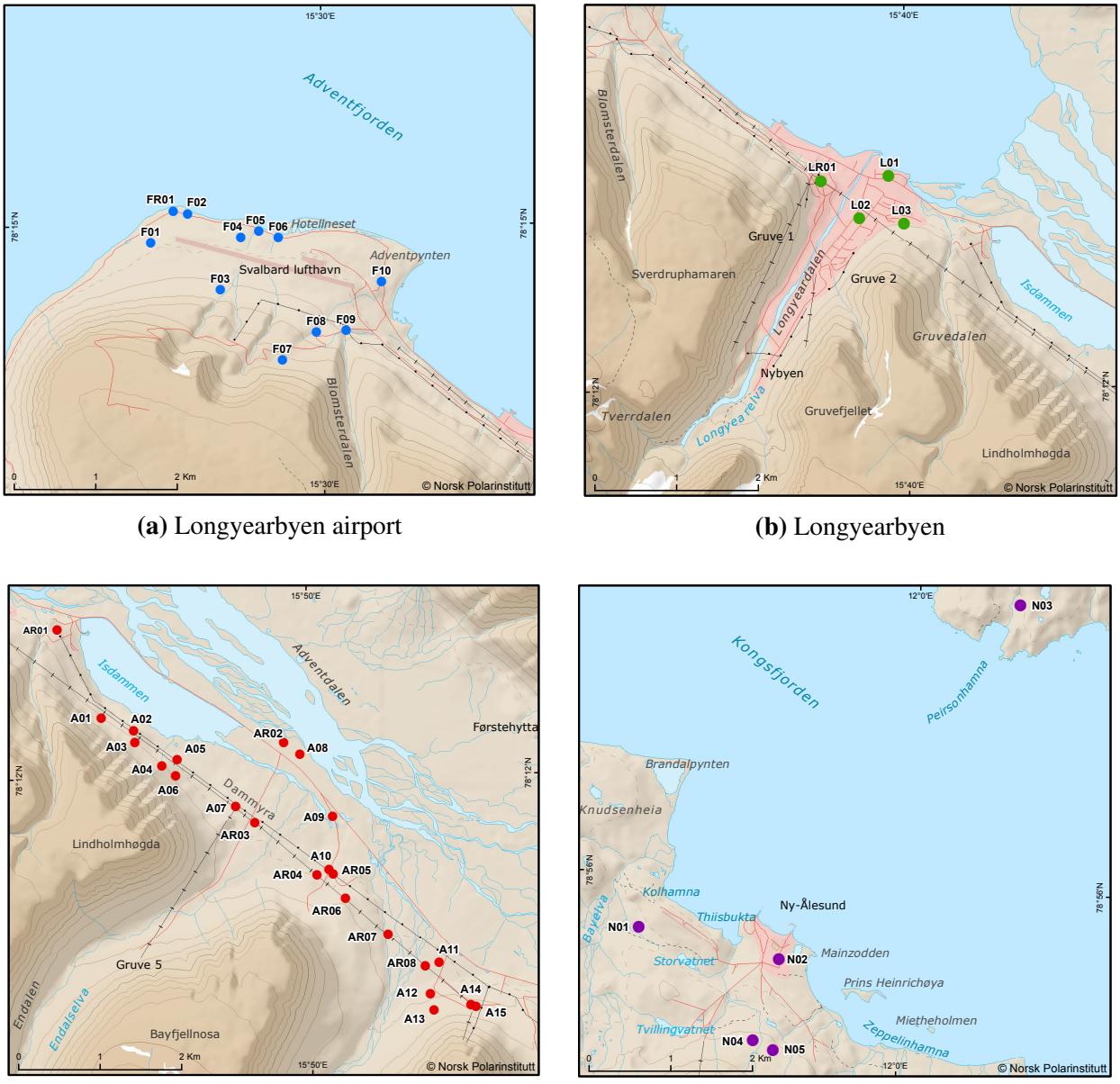
3.4.2 Principal Component Analysis

Principal component analysis (PCA) was conducted for visualization of grouping and correlation among variables. Based on eigenvalues > 1, the two most significant principal components (PC1 and PC2) were used as x and y axis, respectively, in the PCA plots. PCA reduces the number of descriptive variables to a smaller set which can account for the systematic variation in the data set [Kemsley, 1996]. Loading plots were used to interpret the relation among the variables. Variables with a loading value close to zero indicate little influence on the variation in the X matrix. Variables grouped close to each other are positively correlated, while variables positioned on the opposite of the origo have negative correlation [Melnes, 2014].

The samples from Gruve 6 (Mine 6) were included in this analysis (even though the location was not significantly different from the rest of Adventdalen and thus merged with the Adventdalen location in the former analyses) because the PCA showed some interesting differences between the two locations.

4 Results

A total of 11 eggs were collected at Longyearbyen airport (Figure 4.1a), 4 eggs collected in Longyearbyen (Figure 4.1b), 23 eggs collected from Adventdalen (Figure 4.1c) and 5 eggs collected in Ny-Ålesund (Figure 4.1d).



(a) Longyearbyen airport

(b) Longyearbyen

(c) Adventdalen

(d) Ny-Ålesund (Kongsfjorden)

Figure 4.1: An overview of sampled eggs collected from snow buntings *(Plectrophenax nivalis)* for each individual location at Svalbard in 2016, **(a)** Longyearbyen airport (n=11), **(b)** Longyearbyen (n=4), **(c)** Adventdalen (n=23) and **(d)** Ny-Ålesund (n=5). Scale 1:50 000. Maps provided by Bernt Bye (NPI). Samples with an "R" are non-viable eggs collected.

All 43 samples analysed were plotted in a histogram for visualization of how many compounds at each location were detected at concentrations above LOD (Appendix, Figure A.5 to A.8).

4.1 Biometric Variables

Total number of eggs collected for both viable and non-viable eggs at each location can be seen in Table 4.1. Non-viable eggs were collected after sampling of viable eggs were complete, and the sampling of non-viable eggs did not exceed the sampling limit of a total of 50 eggs granted for sampling by the Governor of Svalbard.

Table 4.1: Table illustrating number of eggs sampled from snow buntings (*Plectrophenax nivalis*) at each of the four locations. Table include both viable and non-viable eggs sampled along with total number of eggs sampled from each location. Samples were collected in June 2016 at Longyearbyen airport (AIR), Longyearbyen (LYB), Adventdalen (ADV) and Ny-Ålesund (NÅ).

Location	*Viable*	*Non-viable*	*Total*
AIR	10	1	11
LYB	3	1	4
ADV	15	8	23
NÅ	5	0	5
Total	33	10	43

Biometric variables were measured for all eggs (Table 4.2). On average the eggs sampled had a height of 22.24 $\pm$ 1.00 mm (arithmetic mean [mean$_{Ar}$] $\pm$ SD; n = 42), width of 16.32 $\pm$ 0.48 mm (n = 42), a volume of 3.02 $\pm$ 0.21 ml (n = 42) and an eggshell thickness of 0.1136 $\pm$ 0.0019 mm (n = 40). The earliest egg laid was 22nd May 2016 (n = 37). The average number of eggs in the nests sampled (clutch size) were 5.41 $\pm$ 1.17 (n = 40), with an average of 3.79 $\pm$ 1.73 hatchlings (n = 31) and 1.54 $\pm$ 1.87 fledglings (n = 16) (Table 4.2). There was a relatively small SD for the different variables. Some missing values exists for number of hatchlings in Longyearbyen and NÅ as well as for number of fledglings at Longyearbyen airport, Longyearbyen and NÅ (Table 4.2).

Preliminary blm tested in relation to biometric variables indicated significantly lower eggshell thickness in relation to increasing PFAS concentrations in eggs for the following compounds: PFHxS (p = 0.043), PFHxA (p = 0.011) and 8:2 FTS (p = 0.030). However, glm showed no significant difference in eggshell thickness. No significant differences were detected between any of the biometric variables between the locations. The individual biometric measurements are presented in the Appendix (Table A.3).

Table 4.2: Summary of biometric variables measured for eggs sampled from snow buntings (*Plectrophenax nivalis*) in Longyearbyen, Svalbard and Ny-Ålesund (NÅ, n = 5, Svalbard in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). Data is summarized as mean with standard deviation (SD), median and range for height, width, volume and eggshell thickness, all measured in mm, in addition to clutch size, number of hatchlings and number of fledglings. Hyphen indicate missing values.

Biometric Variable		*ADV (n=23)*	*AIR (n=11)*	*LYB (n=4)*	*NÅ (n=5)*	*Overall (n=43)*
Height (mm)	Mean ± SD	22.21 ± 1.12	22.62 ± 0.77	21.82 ± 0.75	22.11 ± 0.62	22.24 ± 1.00
	Median	22.26	22.91	21.77	22.37	22.38
	Range	20.43-24.11	20.20-23.73	20.84-22.90	20.90-22.66	20.20-24.11
	n	22	11	4	5	42
Width (mm)	Mean ± SD	16.38 ± 0.51	16.32 ± 0.43	16.23 ± 0.43	16.16 ± 0.46	16.32 ± 0.48
	Median	16.37	16.26	16.32	15.95	16.32
	Range	15.51-17.19	15.57-17.01	15.55-16.73	15.63-16.73	15.48-17.19
	n	22	11	4	5	42
Volume (ml)	Mean ± SD	3.04 ± 0.23	3.07 ± 0.19	2.93 ± 0.17	2.95 ± 0.16	3.02 ± 0.21
	Median	3.11	3.00	3.00	2.90	3.01
	Range	2.61-3.34	2.69-3.38	2.65-3.08	2.76-3.22	2.61-3.38
	n	22	11	4	5	42
Eggshell Thickness (mm)	Mean ± SD	0.1137 ± 0.0017	0.1138 ± 0.0018	0.1125 ± 0.0024	0.1127 ± 0.0022	0.1136 ± 0.0019
	Median	0.1147	0.1147	0.1125	0.1138	0.1147
	Range	0.1100-0.1149	0.1100-0.1149	0.1101-0.1149	0.1100-0.1149	0.1100-0.1149
	n	21	11	4	5	41
Clutch size (#)	Mean ± SD	5.4 ± 0.92	5.9 ± 0.67	4.3 ± 2.36	4.5 ± 1.12	5.41 ± 1.17
	Median	5.5	6	6	4.5	6
	Range	4-7	5-7	1-6	3-6	1-7
	n	21	11	3	4	39
Hatchlings (#)	Mean ± SD	4.3 ± 1.4	3.2 ± 2.0	-	-	3.79 ± 1.73
	Median	5	4	-	-	4
	Range	3-6	0-6	-	-	0-6
	n	17	10	0	0	27
Fledglings (#)	Mean ± SD	3.12 ± 1.45	-	-	-	1.54 ± 1.87
	Median	3	-	-	-	0.00
	Range	0-5	-	-	-	0-5
	n	18	0	0	0	18

4.2 PFAS Levels

Out of the 22 different PFASs screened for, all PFASs were detected in at least one of the 43 samples collected. Longyearbyen airport was the only location where all 22 PFASs were detected in at least one sample, followed by Ny-Ålesund, Longyearbyen and Adventdalen (16, 15 and 13 different PFASs detected, respectively). Longyearbyen airport was also the location where the highest values of sum (Σ) $_{22}$PFAS was observed, with mean$_{Ar}$ = 21.23 ng/g wet weight (ww), geometric mean (mean$_{Geo}$) = 0.26 ng/g ww, median = 0.21 ng/g ww, SD = ± 172.62 ng/g ww, range = 0.01-2539.57 ng/g ww, n = 11) followed by Longyearbyen (mean$_{Ar}$ = 1.29 ng/g ww, mean$_{Geo}$ = 0.15 ng/g ww, median = 0.07 ng/g ww, SD = ± 3.14 ng/g ww, range = 0.02-21.59 ng/g ww, N = 4), Ny-Ålesund (mean$_{Ar}$ = 1.72 ng/g ww, mean$_{Geo}$ = 0.07 ng/g ww, median = 0.05 ng/g ww, SD = ± 9.44 ng/g ww, range = 0.01-88.21 ng/g ww, n = 5) and Adventdalen (mean$_{Ar}$ = 0.23 ng/g ww, mean$_{Geo}$ = 0.05 ng/g ww, median = 0.05 ng/g ww, SD = ± 0.63 ng/g ww, range = 0.01-10.32 ng/g ww, n = 23). Mean PFAS concentrations, median and range for each individual PFAS and number of samples (n) above LOD can be found in the Appendix (Table A.4).

The overall detection percentage for each individual PFAS varied from 2% to 98% detection (Appendix, Table A.5). Overall, 5 compounds were detected in > 80% of the samples collected (PFNA (98%) > PFDcA (95%) > PFOS (93%) > PFUnA (88%) > PFTriA (81%)). An additional 7 compounds were detected in range of 40-80% (PFTeA (70%) > PFOA (65%) > PFDoA (63%) > brPFOS (49%) > PFHxS (47%) > PFHxA (40%) = 8:2 FTS (40%)). The remaining 9

compounds were detected in $< 40\%$ of the samples: (PFOSA (23%) $>$ PFHpS (16%) $>$ PFNS (7%) = PFDcS (7%) $>$ PFPS (5%) = PFBA (5%) = PFHpA (5%) $>$ PFBS (2%) = PFPA (2%) = 6:2 FTS (2%)).

The proportion of compounds detected in $> 80\%$ for the samples collected at each location were highest at Longyearbyen airport (11 compounds; PFHxS, brPFOS, PFOS, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA and 8:2 FTS), followed by Longyearbyen (7 compounds; PFOSA, PFOS, PFOA, PFNA, PFDcA, PFUnA and PFTriA), Ny-Ålesund (5 compounds; PFOS, PFNA, PFDcA, PFUnA and PFTriA) and Adventdalen (4 compounds; PFOS, PFNA, PFDcA and PFUnA). PFOS, PFNA, PFDcA and PFUnA were all detected in $> 80\%$ of the samples at all locations examined.

No compound was detected in every sample collected when evaluating the whole dataset, but location-specific compound detection show certain compounds were detected in 100% of the samples collected at that specific location. At Longyearbyen airport 8 different compounds had 100% detection (PFNA, PFDcA, PFUnA, PFTriA, PFTeA, brPFOS, PFOS and 8:2 FTS), the same applied for 7 compounds in Longyearbyen (PFOA, PFNA, PFDcA, PFUnA, PFTriA, PFOSA and PFOS) and 1 compound in Ny-Ålesund (PFOS) and Adventdalen (PFNA).

Certain compounds were detected at only one or two out of the four locations examined. This was the case for PFBA (AIR/LYB), PFPA (AIR) and PFHpA (AIR/LYB) (PFCAs, Figure 4.2), PFBS (AIR), PFPS (AIR), PFHpS (AIR/NÅ) and PFNS (AIR/NÅ) (PFSAs, Figure 4.3) and 6:2 FTS (AIR) (FTSs, Figure 4.4), making any statistical analysis difficult, or even impossible. However, there are compounds with high detection percentage in several locations, making statistical analysis possible. This apply for PFHxA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA and PFTeA (PFCAs, Figure 4.2), PFHxS, brPFOS and PFOS (PFSAs, Figure 4.3) as well as for 8:2 FTS (FTSs, Figure 4.4).

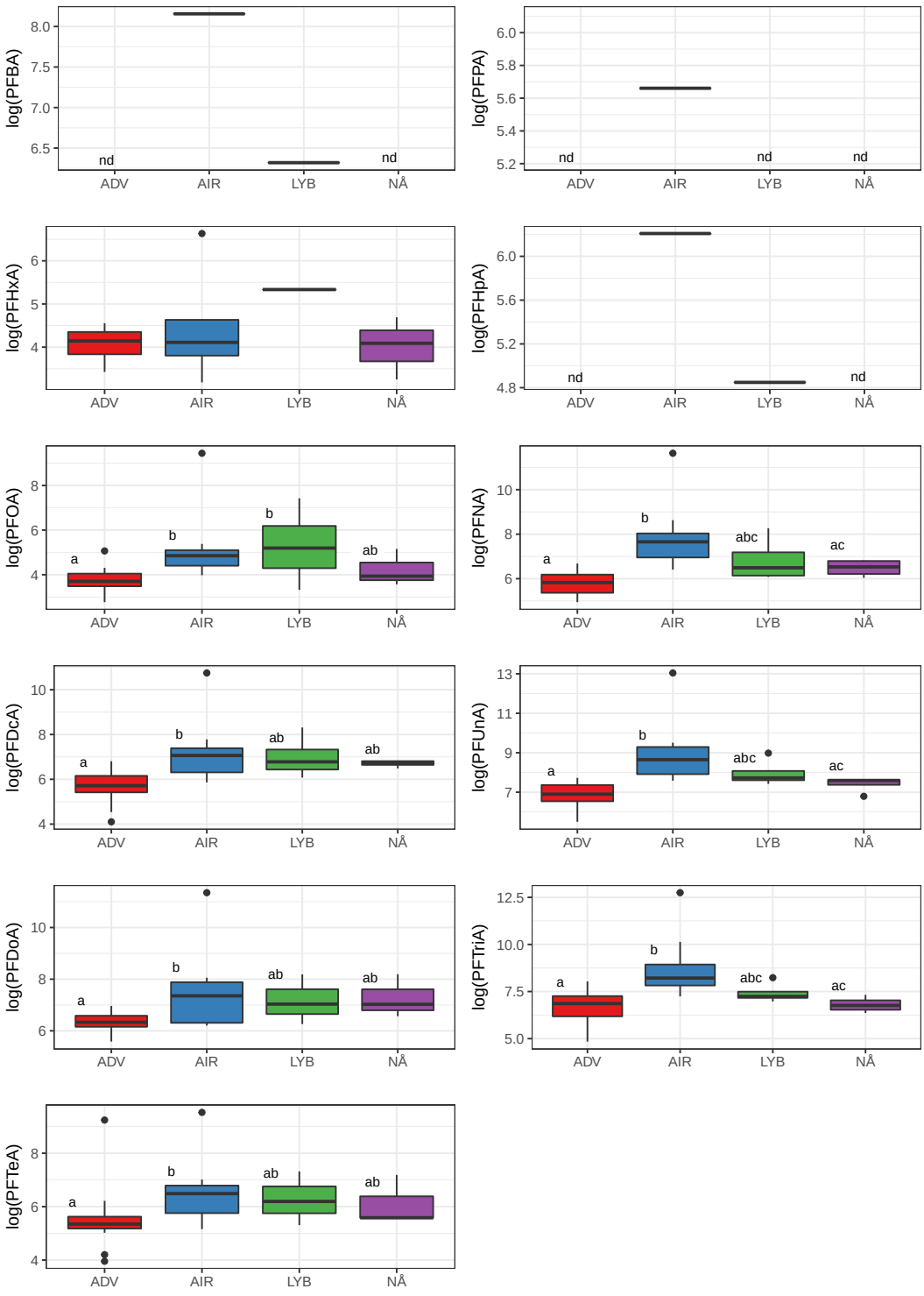


Figure 4.2: Boxplot showing the log-transformed concentrations (ng/g ww) of the egg content of the 11 different perfluoro carboxylic acids (PFCAs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected at Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) (Longyearbyen) and in Ny-Ålesund (NÅ; n=5), at Svalbard in 2016. Non-detects were excluded from the analysis. Filled dots show outliers. Differing letters (a, b, c) annotates significant differences between locations.

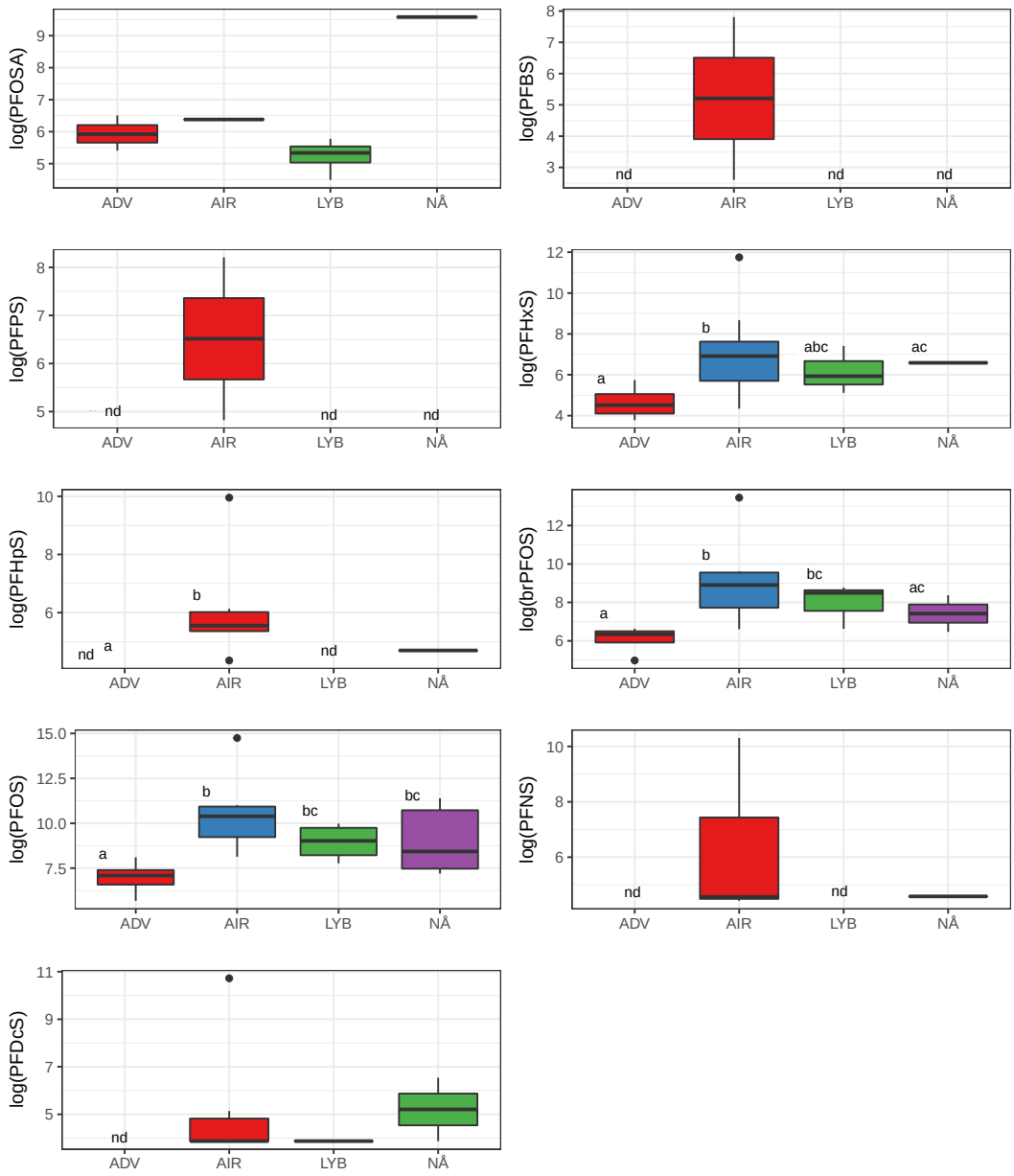


Figure 4.3: Boxplot showing the log-transformed concentrations (ng/g ww) of the egg content of the 9 different perfluorinated sulfonic acids (PFSAs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected at Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) (Longyearbyen) and in Ny-Ålesund (NÅ; n=5), at Svalbard in 2016. Non-detects were excluded from the analysis. Filled dots show outliers. Differing letters (a, b, c) annotates significant differences between locations.

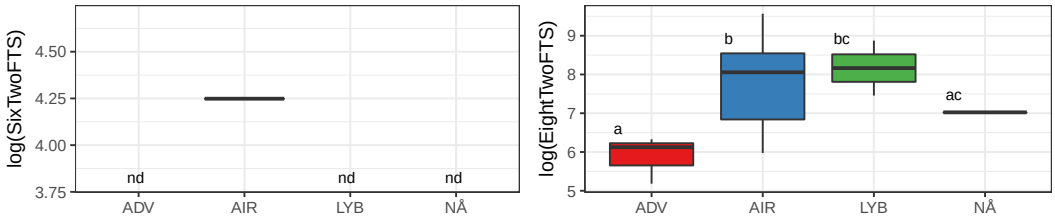


Figure 4.4: Boxplot showing the log-transformed concentrations (ng/g ww) of the egg content of the 2 different fluorotelomer sulfonates (FTSs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected at Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) (Longyearbyen) and in Ny-Ålesund (NÅ; n=5), at Svalbard in 2016. Non-detects were excluded from the analysis. Filled dots show outliers. Differing letters (a, b, c) annotates significant differences between locations with a significance level set to $p < 0.05$.

As shown in Figure 4.2, 4.3 and 4.4, Longyearbyen airport consistently had higher levels of the majority of compounds compared to Adventdalen. This was significant for PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA and PFTeA (PFSAs), PFHxS, PFHpS, linPFOS and brPFOS (PFCAs) and 8:2 FTS (FTSs) (Table 4.3). The same trend was apparent for Longyearbyen compared to Adventdalen, however there are fewer differences. The compounds that were significantly higher in Longyearbyen compared to Adventdalen were PFOA (PFCA), linPFOS, brPFOS (PFSA) and 8:2 FTS (FTS). Ny-Ålesund had tendencies to be significantly lower than Longyearbyen airport, this applied for PFNA, PFUnA, PFTriA (PFSAs), PFHxS, brPFOS (PFCAs) and 8:2 FTS (FTSs). There were no significant differences in contaminant level in Ny-Ålesund compared to Longyearbyen and Adventdalen, with the exception of linPFOS being significantly higher in Ny-Ålesund compared to Adventdalen. The levels at Ny-Ålesund were often in range with or somewhat lower than Longyearbyen, with the exception of PFOSA and PFHxS, however, these values were driven by a detection of these compounds in one sample and can therefore not be compared.

Outliers existed for all locations. However, there was one outlier that was particularly noticeable. In one sample, collected at Longyearbyen airport, there were extremely high levels of PFASs detected. This can consistently be seen in the boxplots (Figure 4.2, 4.3 and 4.4).

Table 4.3: Table showing significance level (0.001 - 1.00) found for concentration levels between each location where eggs from snow buntings (*Plectrophenax nivalis*) were sampled. Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). The PFASs analysed for were divided into perfluororalkyl carboxylic acids (PFCAs), perfluoro sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Boldface indicate samples with a significant difference detected ($p \leq 0.05$), while values close to significance are emphasized. The symbol +/- indicate higher (+) or lower (-) concentration in the former location in the columns.

Group	Abbreviation	AIR-ADV	LYB-ADV	NÅ-ADV	LYB-AIR	NÅ-AIR	NÅ-LYB
PFCA	PFBA	0.518 (+)	0.312 (+)	1.000 (-)	0.873 (+)	0.784 (-)	0.499 (-)
	PFPA	0.377 (+)	1.000 (-)	1.000 (-)	0.739 (-)	0.689 (-)	1.000 (+)
	PFHxA	0.800 (+)	0.994 (+)	0.888 (+)	0.897 (-)	**0.004 (+)**	0.988 (+)
	PFHpA	0.505 (+)	0.348 (+)	1.000 (-)	0.905 (+)	0.776 (-)	0.534 (-)
	PFOA	**0.009 (+)**	**0.017 (+)**	0.933 (+)	0.862 (+)	0.312 (-)	0.175 (-)
	PFNA	< **0.001 (+)**	0.318 (+)	0.987 (-)	0.377 (-)	**0.003 (-)**	0.363 (-)
	PFDcA	**0.004 (+)**	0.147 (+)	0.968 (+)	0.991 (-)	0.176 (-)	0.492 (-)
	PFUnA	< **0.001 (+)**	0.217 (+)	0.987 (+)	0.777 (-)	**0.048 (-)**	0.543 (-)
	PFDoA	**0.007 (+)**	0.475 (+)	0.757 (+)	0.838 (-)	0.472 (-)	0.970 (-)
	PFTriA	< **0.001 (+)**	0.155 (+)	0.861 (+)	0.639 (-)	**0.045 (-)**	0.654 (-)
	PFTeA	**0.006 (+)**	0.650 (+)	0.956 (+)	0.669 (-)	0.229 (-)	0.941 (-)
PFSA	PFBS	0.377 (+)	1.000 (+)	1.000 (+)	0.739 (-)	0.689 (-)	1.000 (+)
	PFPS	0.146 (+)	1.000 (+)	1.000 (+)	0.524 (-)	0.456 (-)	1.000 (+)
	PFHxS	< **0.001 (+)**	*0.089 (+)*	0.987 (+)	0.478 (-)	**0.004 (-)**	0.331 (-)
	PFHpS	**0.004 (+)**	1.000 (-)	0.984 (+)	0.119 (-)	0.157 (-)	0.993 (+)
	PFOSA	0.974 (-)	*0.077(+)*	0.700 (+)	*0.064* (+)	0.579 (+)	0.624 (-)
	linPFOS	< **0.001 (+)**	**0.026 (+)**	**0.009 (+)**	0.426 (-)	0.423 (-)	1.000 (+)
	brPFOS	< **0.001 (+)**	**0.009 (+)**	0.496 (+)	0.151 (-)	< **0.001 (-)**	0.356 (-)
	PFNS	0.224 (+)	1.000 (-)	0.991 (+)	0.615 (-)	0.719 (-)	0.996 (+)
	PFDcS	0.284 (+)	1.000 (-)	0.766 (+)	0.670 (-)	0.986 (-)	0.891 (+)
FTS	6:2 FTS	0.377 (+)	1.000 (+)	1.000 (+)	0.739 (-)	0.689 (-)	1.000 (+)
	8:2 FTS	< **0.001 (+)**	**0.024 (+)**	0.918 (+)	*0.053 (-)*	< **0.001 (-)**	0.227 (-)

The contaminant group that were the most abundant in eggs of snow buntings was the Σ_9PFSAs followed by Σ_{11}PFCAs and Σ_2FTSs for all locations except Adventdalen, where Σ_{11}PFCAs were most prominent (Table 4.4, 4.5 and 4.6). However, the ratio between the PFSAs and PFCAs varied between locations, this is discussed later on (Figure 4.6a).

When comparing the average burden of each of the PFAS groups examined (Table 4.7) it appeared that snow buntings living in proximity to Longyearbyen airport experienced the highest ΣPFAS burden of all locations examined. It can also be seen that Longyearbyen and Ny-Ålesund, as settlements, had similar contamination burden while Adventdalen, which was the reference area, had the lowest overall burden.

Table 4.4: Table showing mean (arithmetic and geometric) concentrations (ng/g ww), in addition to standard deviation (SD), median, range and number of samples (n) >limit of detection (LOD) for the 11 compounds of perfluorinated carboxylic acids (PFCAs) out of 22 per- and polyfluorinated alkylated substances (PFASs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). For significant differences, see Figure 4.2.

Location		PFBA	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDcA	PFUnA	PFDoA	PFTriA	PFTeA
AIR	$Mean_{Ar}$	0.33	0.04	0.10	0.06	1.24	12.27	5.22	47.56	8.92	36.81	1.77
	$Mean_{Geo}$	0.02	0.01	0.03	0.02	0.11	2.53	1.32	7.34	1.13	6.09	0.65
	SD	± 1.00	± 0.08	± 0.21	± 0.14	± 3.60	± 31.92	± 13.06	± 131.08	± 24.03	± 97.19	± 3.80
	Median	0.01	0.01	0.01	0.01	0.01	2.11	1.17	5.70	1.49	3.70	0.66
	Range	0.01-3.49	0.01-0.29	0.01-0.76	0.01-0.50	0.01-12.62	0.61-113.11	0.35-46.48	1.97-461.90	0.01-84.84	1.39-343.43	0.17-13.74
	n >LOD	1	1	5	1	9	11	11	11	10	11	11
LYB	$Mean_{Ar}$	0.15	0.01	0.006	0.04	0.53	1.43	1.58	3.54	1.31	1.90	0.55
	$Mean_{Geo}$	0.03	0.01	0.02	0.020	0.20	0.93	1.08	2.87	0.41	1.67	0.21
	SD	± 0.24	± 0	± 0.09	± 0.05	± 0.67	± 1.43	± 1.46	± 2.56	± 1.37	± 1.09	± 0.58
	Median	0.01	0.01	0.01	0.01	0.21	0.70	0.90	2.26	0.83	1.39	0.35
	Range	0.01-0.56	0.01	0.01-0.21	0.01-0.13	0.03-1.68	0.44-3.89	0.44-4.07	1.68-7.96	0.01-3.59	1.06-3.77	0.01-1.51
	n >LOD	1	0	1	1	4	4	4	4	3	4	3
ADV	$Mean_{Ar}$	0.01	0.01	0.03	0.01	0.03	0.39	0.36	0.96	0.29	0.69	0.57
	$Mean_{Geo}$	0.01	0.01	0.02	0.01	0.02	0.34	0.26	0.47	0.08	0.19	0.07
	SD	± 0	± 0	± 0.03	± 0	± 0.03	± 0.19	± 0.24	± 0.67	± 0.33	± 0.78	± 2.08
	Median	0.01	0.01	0.01	0.01	0.02	0.34	0.28	0.92	0.01	0.36	0.07
	Range	0.01	0.01	0.01-0.10	0.01	0.01-0.16	0.14-0.80	0.01-0.90	0.01-2.27	0.01-1.06	0.01-3.09	0.01-10.32
	n >LOD	0	0	8	0	12	23	22	19	11	15	13
NÅ	$Mean_{Ar}$	0.01	0.01	0.04	0.01	0.06	0.55	0.66	1.38	1.09	0.77	0.38
	$Mean_{Geo}$	0.01	0.01	0.03	0.01	0.03	0.29	0.34	0.63	0.22	0.39	0.11
	SD	± 0	± 0	± 0.04	± 0	± 0.06	± 0.33	± 0.34	± 0.81	± 1.33	± 0.50	± 0.49
	Median	0.01	0.01	0.03	0.01	0.04	0.53	0.81	1.94	0.71	0.73	0.26
	Range	0.01	0.01	0.01-0.11	0.01	0.01-0.17	0.01-0.91	0.01-0.91	0.01-2.04	0.01-3.60	0.01-1.52	0.01-1.32
	n >LOD	0	0	3	0	3	4	4	4	3	4	3

Table 4.5: Table showing mean (arithmetic and geometric) concentrations (ng/g ww), in addition to standard deviation (SD), median, range and number of samples (n) >limit of detection (LOD) for the 9 compounds of perfluoro sulfonic acids (PFSA) out of 22 per- and polyfluorinated alkylated substances (PFASs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). For significant differences, see Figure 4.3.

Location		PFOSA	PFBS	PFPS	PFHxS	PFHpS	brPFOS	linPFOS	PFNS	PFDcS
AIR	$Mean_{Ar}$	0.07	0.24	0.36	12.64	2.05	69.27	256.68	2.77	4.18
	$Mean_{Geo}$	0.02	0.02	0.03	0.76	0.17	6.71	28.70	0.10	0.10
	SD	± 0.17	± 0.70	± 01.05	± 36.00	± 6.00	± 197.62	± 722.21	± 8.58	± 13.03
	Median	0.01	0.01	0.01	0.69	0.08	7.39	32.37	0.05	0.05
	Range	0.01-0.59	0.01-2.46	0.01-3.67	0.01-126.38	0.05-21.00	0.74-693.96	3.41-2539.57	0.05-29.97	0.05-45.39
	n >LOD	1	1	2	10	6	11	11	2	2
LYB	$Mean_{Ar}$	0.21	0.01	0.01	0.55	0.05	3.04	11.01	0.05	0.05
	$Mean_{Geo}$	0.19	0.01	0.01	0.19	0.05	1.03	7.67	0.05	0.05
	SD	± 0.08	± 0	± 0	± 0.65	± 0	± 2.71	± 7.97	± 0	± 0
	Median	0.21	0.01	0.01	0.27	0.05	2.82	10.03	0.05	0.05
	Range	0.09-0.32	0.01	0.01	0.01-1.65	0.05	0.05-6.47	2.36-21.59	0.05	0.05
	n >LOD	4	0	0	3	0	3	4	0	0
ADV	$Mean_{Ar}$	0.08	0.01	0.01	0.04	0.05	0.13	1.15	0.05	0.05
	$Mean_{Geo}$	0.02	0.01	0.01	0.02	0.05	0.07	0.73	0.05	0.05
	SD	± 0.17	± 0	± 0	± 0.07	± 0	± 0.20	± 0.83	± 0	± 0
	Median	0.01	0.01	0.01	0.01	0.05	0.05	0.92	0.05	0.05
	Range	0.01-0.67	0.01	0.01	0.01-0.31	0.05	0.05-0.76	0.05-3.31	0.05	0.05
	n >LOD	4	0	0	6	0	4	20	0	0
NÅ	$Mean_{Ar}$	2.91	0.01	0.01	0.16	0.06	1.02	28.25	0.06	0.18
	$Mean_{Geo}$	0.05	0.01	0.01	0.03	0.06	0.20	8.45	0.06	0.08
	SD	± 5.79	± 0	± 0	± 0.28	± 0.02	± 1.66	± 34.28	± 0.02	± 0.26
	Median	0.01	0.01	0.01	0.01	0.05	0.05	1.34	0.05	0.05
	Range	0.01-14.49	0.01	0.01	0.01-0.72	0.05-0.11	0.05-4.31	1.34-88.21	0.05-0.10	0.05-0.69
	n >LOD	1	0	0	1	1	2	5	1	1

Table 4.6: Table showing mean (arithmetic and geometric) concentrations (ng/g ww), in addition to standard deviation (SD), median, range and number of samples (n) >limit of detection (LOD) for the 2 fluorotelomer sulfonates (FTS) out of 22 per- and polyfluorinated alkylated substances (PFASs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). For significant differences, see Figure 4.4.

Location		*6:2 FTS*	*8:2 FTS*
AIR	$Mean_{Ar}$	0.02	4.46
	$Mean_{Geo}$	0.02	2.50
	SD	± 0.02	± 4.59
	Median	0.01	3.15
	Range	0.01-0.07	0.39-14.27
	n >LOD	1	11
LYB	$Mean_{Ar}$	0.01	2.24
	$Mean_{Geo}$	0.01	0.41
	SD	± 0	± 2.91
	Median	0.01	0.89
	Range	0.01	0.05-7.15
	n >LOD	0	3
ADV	$Mean_{Ar}$	0.01	0.09
	$Mean_{Geo}$	0.01	0.06
	SD	± 0	± 0.13
	Median	0.01	0.05
	Range	0.01	0.05-0.56
	n >LOD	0	3
NÅ	$Mean_{Ar}$	0.01	0.26
	$Mean_{Geo}$	0.01	0.09
	SD	± 0	± 0.43
	Median	0.01	0.05
	Range	0.01	0.05-1.12
	n >LOD	0	1

Table 4.7: Table showing mean (arithmetic and geometric) burden (ng/g ww), standard deviation (SD), median, range and number of samples (n) for each group of per- and polyfluorinated alyklated substances (PFASs) and the total Σ_{22}PFAS burden in eggs from snow buntings (*Plectrophenax nivalis*) at each location. The three groups are perfluorinated carboxylic acids (PFCAs), perfluorinated sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sublocations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23).Significant differences between locations are annotated with letters (a, b) with a significance level set to $p < 0.05$.

Location		Σ_{11}*PFCA*	Σ_{9}*PFSA*	Σ_{2}*FTS*	Σ_{22}*PFAS*
AIR	$Mean_{Ar}$	114.31	348.24	4.48 ^{a}	467.03
	$Mean_{Geo}$	20.43	37.45	2.54	65.16
	SD	305.99	985.14	4.58	1293.64
	Median	15.63	42.56	3.16	65.31
	Range	4.85-1081.15	4.35-3462.34	0.41-14.28	12.39-4556.15
	n	11	11	11	11
LYB	$Mean_{Ar}$	11.11	14.98	2.26 ^{b}	28.34
	$Mean_{Geo}$	8.27	10.32	0.47	20.79
	SD	9.45	11.18	2.91	19.98
	Median	6.47	13.08	0.90	24.25
	Range	4.12-27.36	3.62-30.12	0.06-7.16	9.06-55.81
	n	4	4	4	4
ADV	$Mean_{Ar}$	3.35	1.57	0.11 ^{b}	5.02
	$Mean_{Geo}$	2.49	1.25	0.08	4.16
	SD	2.46	1.01	0.13	3.09
	Median	2.85	1.47	0.06	3.91
	Range	4.12-27.36	3.62-30.12	0.06-7.16	0.97-12.86
	n	23	23	23	23
NÅ	$Mean_{Ar}$	4.97	32.66	0.28 ^{ab}	37.91
	$Mean_{Geo}$	2.85	15.13	0.11	25.76
	SD	2.48	32.92	0.43	31.11
	Median	5.61	16.06	0.06	21.72
	Range	0.13-6.92	2.00-88.46	0.06-1.14	8.99-88.65
	n	5	5	5	5

4.3 PFAS Patterns

4.3.1 Contamination Profiles

The contribution of the individual PFASs (ng/g ww) on the total PFAS load was examined for all locations (Figure 4.5). For Longyearbyen airport, Longyearbyen and Ny-Ålesund 5 different PFASs were dominating the contamination pattern, while in Adventdalen 7 different PFASs dominated. The contaminant pattern did not indicate any major distribution difference of compounds between locations.

All locations appeared to have the largest contamination burden resulting from PFOS. For Ny-Ålesund (74.5%), Longyearbyen airport (54.7%) and Longyearbyen (38.8%) PFOS clearly dominated the contamination profile. For Adventdalen (22.9%) PFOS was also the dominating compound, but less pronounced compared to the other locations. PFUnA was generally the second most dominating compound in the contamination profile. This applied for all locations, with the highest levels found in Adventdalen (19.0%) followed by Longyearbyen (12.5%), Longyearbyen airport (10.3%) and Ny-Ålesund (3.7%).

Together, PFOS and PFUnA constitute a contamination burden of 65% at Longyearbyen airport, 51.3% in Longyearbyen, 41.9% in ADV and as much as 78.2% in NÅ. These two contaminants were clearly the dominating PFASs at all locations, while a range of different PFASs played a minor role to the remaining overall contaminant burden, and their relative contribution differed between locations.

PFTriA and brPFOS was additionally detected in 3 out of the 4 locations; PFTriA was detected at Longyearbyen airport, Longyearbyen and Adventdalen, but not in Ny-Ålesund, and brPFOS was detected at Longyearbyen airport, Longyearbyen and Ny-Ålesund but not in Adventdalen.

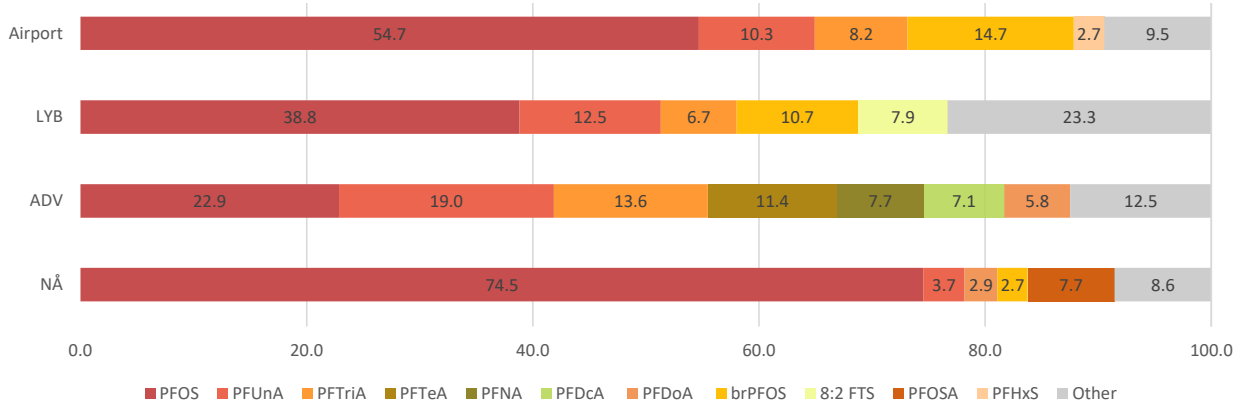
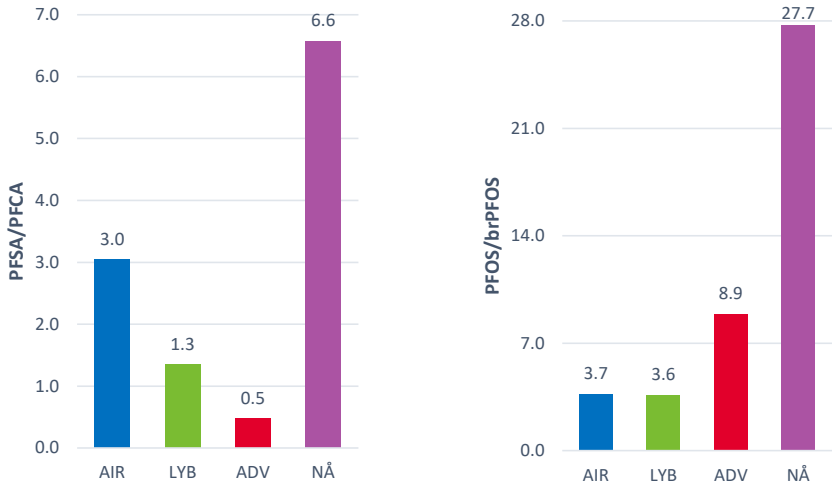


Figure 4.5: Stacked barplot showing the relative abundance (% of total contaminant load) of the 5-7 most pronounced perfluoroalkyl and polyfluoroalkyl substances (PFASs) in eggs from snow buntings (*Plectrophenax nivalis*) sampled at each location; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) and Ny-Ålesund (NÅ; n=5), Svalbard in 2016. The contaminant profile is based on concentrations in ng/g ww, where 100% is Σ_{22}PFAS.

4.3.2 Contaminant Ratios



(a) PFSA:PFCA **(b)** linPFOS:brPFOS

Figure 4.6: Two figures showing the relationship between **(a)** perfluror sulfonic acids (PFSAs) : perfluroro carboxylic acids (PFCAs) (PFSA:PFCA) and **(b)** linear PFOS : branched PFOS (linPFOS:brPFOS) for all samples of eggs collected from snow buntings (*Plectrophenax nivalis*) at the four different locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) and Ny-Ålesund (NÅ; n=5). All samples were collected in 2016.

It appeared that all locations had a PFSA:PFCA profile dominated by the PFSAs (all values are above 1:1) except for Adventdalen, where the relationship was 0.5 (i.e. dominated by PFCAs) (Figure 4.6a). The PFSA:PFCA ratio decreased with increasing distance from Longyearbyen airport (3.1) to Longyearbyen (1.3) and Adventdalen (0.5). Ny-Ålesund, however, had a high ratio of PFSA:PFCA (6.6), more than twice that of what was found at Longyearbyen airport (3.1), and more than five fold higher than the ratio found in LYB (1.3).

The relationship between linPFOS and brPFOS at all locations were dominated by linPFOS (Figure 4.6b). At Longyearbyen airport and Longyearbyen the relationship was relatively low, with values at 3.7 and 3.6, respectively. The linPFOS levels in Adventdalen (8.9) was intermediate while at Ny-Ålesund (27.7) the relationship was more than threefold higher than Adventdalen and more than sevenfold higher than the levels found at Longyearbyen airport.

When considering the percentage of PFSAs in relation to total PFSA and PFCA (Table 4.8), Adventdalen was the only location with less than 50% PFSAs (32.0%) while Longyearbyen airport (75.3%), Longyearbyen (57.4%) and Ny-Ålesund (86.8%) all had a percentage above 50%. The percentage of linPFOS in relation to total PFOS (Table 4.8) was dominated by linear PFOS; constituting $>$ 78% at all locations (AIR 78.7%, LYB 78.4%, ADV 89.9% and NÅ 96.5%).

Table 4.8: Table illustrating the percentage (%) of perfluorinated sulfonic acids (PFSAs) relative to the total PFSA and perfluoro carboxylic acids (PFCAs). Additionally, % of linear PFOS (linPFOS) relative to total PFOS is shown. Numbers are based on levels found in eggs from snow buntings (*Plectrophenax nivalis*) sampled at Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4), Adventdalen (ADV; n=23) and Ny-Ålesund (NÅ; n=5), Svalbard in 2016.

Location	*PFSA (%)*	*linPFOS (%)*
AIR	75.3	78.7
LYB	57.4	78.4
ADV	32.0	89.9
NÅ	86.8	96.5

4.4 Relationships between PFAS concetrations and biometric variables

The PCA including the different PFASs (R2X=0.801, Q2=0.660) resulted in 2 principal components with eigenvalues > 1, explaining 71.10% and 9.04%, respectively, of the variation between the compounds.

The PCA indicated relationships among the different PFAS groups (Figure 4.7). All compounds were located close to each other along the PC1 axis indicating a positive correlation between the compounds and PC1. The PFCAs were also located close to each other along PC2, as was the case for the PFSAs and the FTSs. However, the PFSAs and FTSs were located opposite of the PFCAs along PC2, indicating negative relationships between these groups.

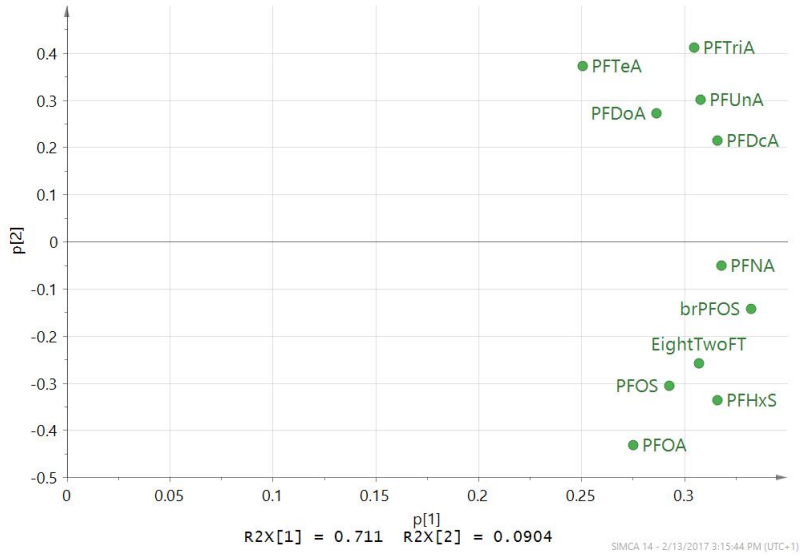


Figure 4.7: Principal component analysis loading plot with selected PFASs found in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23).

The samples from Gruve 6 (Mine 6) were included in the score plot analysis (Figure 4.8), even though the location was not significantly different from the rest of Adventdalen. However, the PCA indicated the samples collected around Mine 6 to have a different PFAS composition compared to the rest of the samples collected in Adventdalen. The score plot also indicated a contamination burden dominated by the PFCAs at Mine 6, as seen by the samples being clustered in the upper part of the PC2 plot. This is consistent with the low PFSA percentage found for ADV (33.6%). The remaining locations did not appear to show any distinct pattern regarding their PFAS distribution.

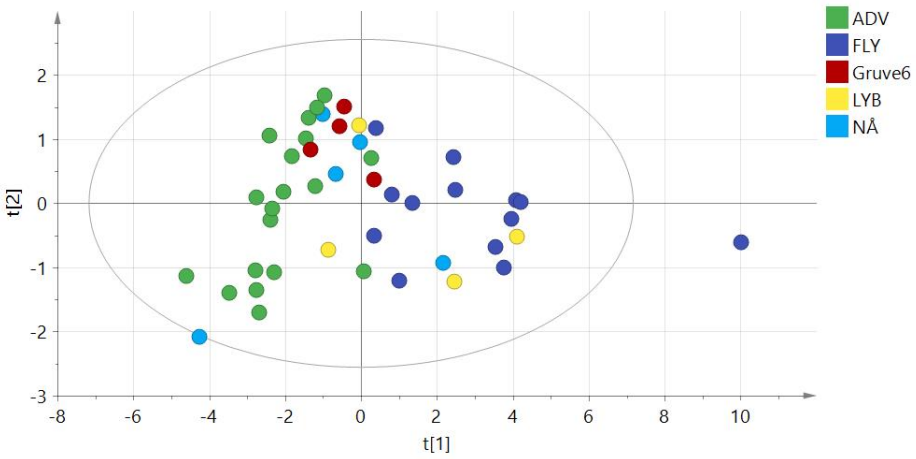


Figure 4.8: Principal component score plot based on the selected PFASs in relation to the location where the sample was collected. Samples are eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). Mine 6 (Gruve 6, n=4), which was fused with Adventdalen, is distinguished in the PCA plot.

A PCA including the biometric variables and the different PFASs (R2X=0.582, Q2=0.331) resulted in 2 principal components with eigenvalues > 1, explaining 46.00 % and 12.20 %, respectively, of the variation between the biometric variables and the different PFASs (Figure 4.9).

The plot indicated a positive relationship along PC1 between the PFASs as a group of contaminants. Furthermore, the PCA also indicated negative relationship along PC1 between the PFASs and all the measured biological variables, with eggshell thickness and number of hatchlings as having the possible strongest negative correlations. From the PCA it can also be seen that first egg laid and volume of the egg appear to be negatively correlated with the concentrations of the PFASs in the eggs.

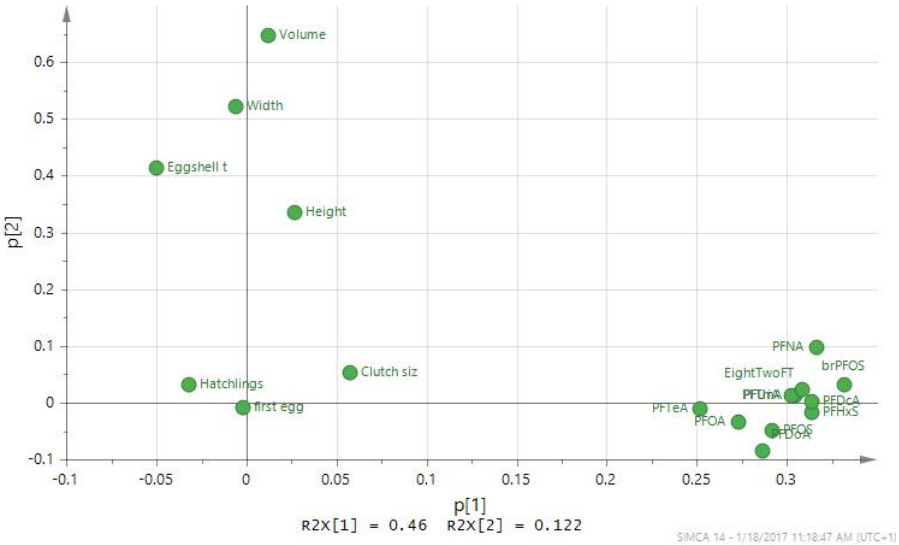


Figure 4.9: Principal component analysis loading plot with all biometric variables (volume, width, eggshell thickness, height, hatchlings, clutch size and first egg laid) and all PFASs in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23).

The score plot (Figure 4.10) showed that the different locations were more or less grouped together, with Longyearbyen airport and Longyearbyen mainly found on the right side of the loading plot, while Adventdalen and Ny-Ålesund were primarily located on the left side. This was consistent with the contamination burden at Longyearbyen airport and Longyearbyen being higher than what was found at Adventdalen and Ny-Ålesund.

The score plot also indicated that eggshell thickness, width, height and volume were larger in Adventdalen than Longyearbyen airport (considering they are opposite along PC1). Although there were, as previously mentioned, no significant differences among the locations with respect to the biometric variables.

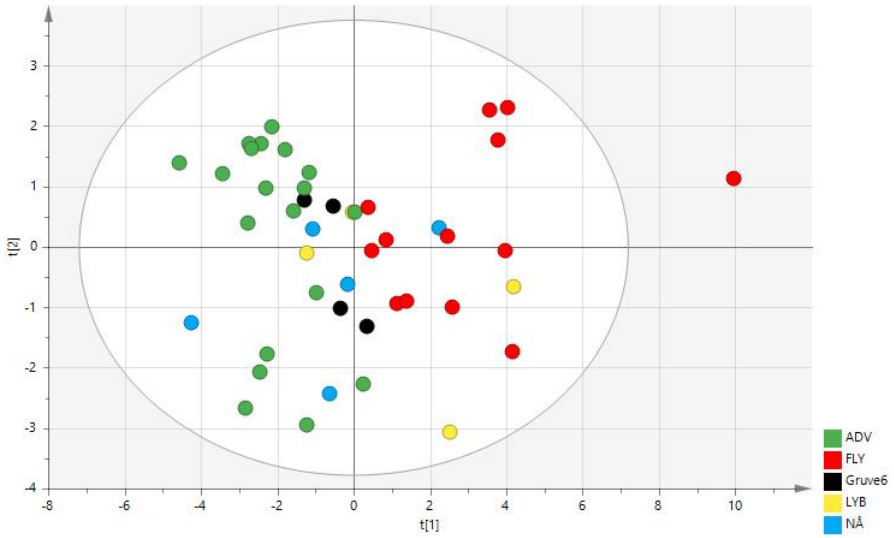


Figure 4.10: Principal component score plot based on the different locations and the levels of PFASs found in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). Mine 6 (Gruve 6, n=4), which was fused with Adventdalen, is distinguished in the PCA plot.

5 Discussion

5.1 Biometric Variables

No significant differences were found between locations with respect to the biometric variables. However, preliminary statistics with blm suggested eggshell thinning in samples where the PFAS concentrations were significantly higher. However, when proceeding with glm to further examine this case, it appeared that eggshell thickness did not differ significantly among the locations. To my knowledge, no records of eggshell thickness have been reported previously for the snow bunting. Eggshell thinning in relation to PFASs in marine top predators, such as ivory gull (*Pagophila eburnea*) and yellow legged gull (*Larus michahellis*) have previously been examined, however no association between PFAS levels and eggshell thickness were found for these species [Miljeteig et al., 2012, Vicente et al., 2012]. There were no differences in the other biometric variables (i.e height, width, volume) between the locations.

5.2 PFAS Levels and Patterns

Snow bunting eggs collected in proximity to Longyearbyen airport had the largest Σ_{22}PFAS burden of all locations, followed by Ny-Ålesund $>$ Longyearbyen $>$ Adventdalen. PFOS was the individual compound that resulted in the largest contribution to the overall PFAS burden experienced, and this applied for all locations examined.

Comparing the restults from the present study with Kristoffersen (2012), which sampled snow bunting eggs from Longyearbyen and Ny-Ålesund in 2010 and 2011, found levels (Σ_6PFAS of 16.43 ng/g ww in Longyearbyen and Σ_6PFAS 37.80 ng/g ww in NÅ) [Kristoffersen, 2012] that were in range with what was found in the present study. The levels detected in Longyearbyen (Σ_{22}PFAS of 28.34 ng/g ww) were somewhat higher than the levels found by Kristoffersen (2012). For NÅ (Σ_{22}PFAS of 37.91 ng/g ww) the levels were in range.

There were no significant differences in contaminant levels in Ny-Ålesund compared to Longyearbyen and Adventdalen, with the exception of linPFOS. Mean linPFOS levels were significantly higher in the samples from Ny-Ålesund, however, the value is driven by one sample with extremely high levels. This sample was collected at the old school in Ny-Ålesund, the same location where Kristoffersen (2012) found extremely high PFAS levels in one of her samples.

Dauwe et al. (2007) determined PFOS levels in blood (24-1625 ng/ml) and livers (553-11359 ng/g ww) of great tits (*Parus major*) sampled near a large fluorochemical plant in Antwerp, Belgium. The levels exceeded the hepatic benchmark concentration (600 ng/g ww) for the protection of avian species for all but one sample [Dauwe et al., 2007]. Another study, by Hoff et al. (2005), measured the hepatic PFOS concentrations in great tit and blue tit (*Parus caeruleus*) nestlings (86–2788 ng/g ww and 317–3322 ng/g ww for great and blue tit, respectively) from Blokkersdijk, a bird reserve in the proximity of the same fluorochemical [Hoff et al., 2005]. Levels from the present study were generally in the lower range or lower than than the PFOS levels reported in these two studies.

Haugerud (2011) collected plasma samples from glaucous gulls (*Larus hyperboreus*) breeding in Kongsfjorden (Ny-Ålesund) in 2010. She found mean Σ_6PFCA = 7.58 ng/g ww, Σ_2PFSA = 33.23 ng/g ww and Σ_8PFAS = 40.80 ng/g ww. When comparing these levels to those found in Ny-Ålesund in the present study (Σ_{11}PFCA = 4.97 ng/g ww, Σ_9PFSA = 32.66 ng/g ww and Σ_{22}PFAS

= 37.91 ng/g ww) the levels were within the same range, despite the snow bunting being a lower trophic terrestrial species and the glaucous gull being one of the largest Arctic avian apex predators [Haugerud, 2011]. Considering the Haugerud study sampled blood plasma and the present study sampled eggs, the results might not be directly comparable. A study on herring gulls (*Larus smithsonianus*) indicated that the transfer of different PFAS compounds from the female to the egg differs [Gebbink and Letcher, 2012]. However, a study by Drouillard and Nordstrom (2001) found that eggs directly reflects the composition of maternal tissues, not the diet, at the time of yolk formation [Drouillard and Norstrom, 2001]. Whether or not this apply for plasma as well is uncertain.

Verreault et al. (2005) sampled several tissues from Glaucous gulls, including eggs, from Bear Island in 2004 [Verreault et al., 2005]. They found ΣPFAS in eggs to be 41.8 ± 5.27 ng/g ww, similar to the levels found in snow bunting eggs from Ny-Ålesund (37.91 ± 31.11 ng/g ww) in the present study. Levels at Longyearbyen airport (467.03 ± 1293.64 ng/g ww), however, far exceeds the glaucous gull levels. Their reported PFOS levels (104 ± 13.2 ng/g ww) were at the time the highest levels reported so far in any Arctic seabird species. Additionally, plasma levels were within a range equivalent or slightly above what had been detected in polar bears from the same location. The present study found mean PFOS levels at Longyearbyen airport (256.68 ± 722.21) to be higher than reported to glaucous gulls reported by Verreault et al. (2005). Considering the glaucous gull is at the peak of the food chain, the high levels of Σ_{22}PFAS, including PFOS, found in the eggs from snow buntings is alarming. High PFAS levels in low trophic species might result in even higher PFAS levels in species at the top of the food chain that feed on lower trophic species, increasing the likelihood of reaching levels that potentially cause adverse effects.

The predominant PFAS in peregrine falcon (*Falco peregrinus*) eggs sampled at Greenland was PFOS (83 ng/g ww mean concentration in samples from 2006), followed by PFTriA (7.2 ng/g ww) and PFUnA (4.2 ng/g ww) [Holmström et al., 2010]. These levels are lower than those found in snow buntings in the present study (Longyerabyen airport: PFOS 256.68 ng/g ww, PFTria 36.81 ng/g ww and PFUnA 47.56 ng/g ww). Considering the snow bunting is a terrestrial low trophic species preyed upon by peregrine falcons [Vorkamp et al., 2005], these high contaminant levels are of concern. On the other hand, it could be the case that Greenland is generally less contaminated compared to Svalbard. Also, peregrine falcons do not breed at Svalbard, further complicating comparisons.

Tawny owl (*Strix aluco*) eggs sampled from Norway [Ahrens et al., 2011] were less contaminated than eggs from snow buntings collected at Longyearbyen airport regarding PFOS, PFTriA and PFUnA. PFOS was the predominant compound in tawny owl eggs with a geometric mean of 10.1 ng/g ww (28.70 ng/g ww in bunting eggs at Longyearbyen airport), followed by PFTriA at 0.36 ng/g ww (6.09 ng/g ww at Longyearbyen airport) and PFUnA at 0.19 ng/g ww (7.34 ng/g ww at Longyearbyen airport). However, when comparing the levels in tawny owl to the samples collected in Adventdalen it can be seen that the levels of PFOS and PFTriA (0.73 ng/g ww and 0.19 ng/g ww, respectively) are lower in eggs of snow buntings while concentrations of PFUnA (0.47 ng/g ww) are higher in snow bunting eggs compared to tawny owl eggs. Longyearbyen and Ny-Ålesund had higher levels of PFTriA (1.67 ng/g ww and 0.39 ng/g ww, respectively) and PFUnA (2.87 ng/g ww and 0.63 ng/g ww, respectively), but lower levels of PFOS (7.67 ng/g ww and 8.45 ng/g ww, respectively) compared to levels found in tawny owl eggs. However, it is worth mentioning that the study by Ahrens et al. (2011) evaluated temporal trends from samples collected over several years, complicating comparisons considering levels are likely to vary from one year to another.

PFAS levels found in this study were in range with the levels in eggs of snow buntings examined in a previous study from Svalbard [Kristoffersen, 2012]. However, when comparing the levels from the present study with levels found in an Arctic marine top predator, it can be seen that the eggs collected at Longyearbyen airport exceeded the levels found in glaucous gull eggs. Levels were, however, lower at the remaining locations examined herein. The PFAS levels in eggs from snow buntings collected at Longyearbyen airport were also higher than eggs sampled from Nordic terrestrial predatory birds (tawny owls and peregrine falcons).

5.2.1 Contaminant Profiles

The predominant PFAS found in snow bunting eggs was PFOS and PFUnA. In addition, PFTriA and brPFOS was found at 3 of the 4 locations examined. This is consistent with previous findings, both Holmstrom et al. (2010) and Ahrens et al. (2011) found these compounds to dominate the contamination pattern in peregrine falcon eggs (Greenland) and tawny owl eggs (Norway) [Holmström et al., 2010, Ahrens et al., 2011]. PFOS is one of the predominant anthropogenic PFASs being measured worldwide in the aquatic and the terrestrial environment, including humans [Houde et al., 2008].

The eggs collected from Longyearbyen airport were by far the most contaminated, with levels up to 12 x higher (student t.test, $p = 0.041$) than Ny-Ålesund, the location with the second highest levels (Σ_{22}PFAS 467.03 ng/g ww, 37.91 ng/g ww respectively). This is likely the result of snow buntings being exposed to a local source of PFASs at Longyearbyen airport. Levels were also almost 90 x higher (student t.test, $p = 0.045$) than the levels found in Adventdalen (Σ_{22}PFAS 5.03 ng/g ww). This could indicate high PFAS contamination around Longyeabyen airport, but also that the levels decline rapidly from Longyearbyen airport to Adventalen over a relatively short distance. The high levels found at Longyearbyen airport result in a large contribution of PFASs on the total contaminant load experienced by snow buntings, putting an additional strain on their already high levels of other legacy POPs, that have previously been found to exceed environmental guidelines for protecting wildlife [Choy et al., 2010, Kristoffersen, 2012].

The individual PFASs found to dominate the contamination profile (PFOS, PFUnA and PFTriA) in eggs from snow buntings, at all locations, are consistent with previous findings [Kristoffersen, 2012]. Additionally, a larger range of different PFASs were detected at Longyearbyen airport compared to the other locations, with Adventdalen as the location with fewest PFASs detected.

5.2.2 Contamination Ratios

The most prominent PFAS group detected in snow bunting eggs were the PFSAs (C6-C8), followed by the PFCAs (C6-C14) and FTSs (C8) respectively. This was the case for all four locations investigated.

Field biomonitoring studies by Houde et al. have provided strong evidence that PFOS, PFHxS, and C8-C12 PFCAs can bioaccumulate and biomagnify through food webs, reaching elevated concentrations in in species feeding at high trophic levels [Houde et al., 2006]. The present study showed that high concentrations can be accumulated even in low trophic birds if they live in areas with high environmental concentrations of PFASs.

It has previously been found that PFCAs and PFSAs with perfluoroalkyl chain lengths shorter than 7 and 6 carbons, respectively, have insignificant bioconcentration factors (BCFs) and low bioaccumulation factors (BAFs) in rainbow trout (*Oncorhynchus mykiss*) [Martin et al., 2013]. However, both BCFs and BAFs increased with increasing length of the perfluoroalkyl chain. BCFs increased by a factor of 8 for each additional carbon in the perfluoroalkylchain between 8 and 12 carbons, but this relationship deviated from linearity for the longest PFCA tested. Sulfonates had greater BCFs and BAFs, half-lives, and rates of uptake than the corresponding carboxylate of equal perfluoroalkyl chain length, indicating that chain length was not the only determinant of PFAS bioaccumulation potential and that the acid functional group must also be considered [Martin et al., 2013]. Despite the fact that birds and fish might not be directly comparable due to different physiology, the results from Martin et al. (2013) is in accordance with the findings from the present study, with the PFSAs found in higher concentrations than the PFCAs. It could be that the molecular properties or mechanisms of the PFASs behave similarly despite species differences.

ECF, the only manufacturing process for PFOS and its precursors, yields a mixture of 70% linear and 30% branched isomers [Houde et al., 2008]. The PFOS pattern at Longyearbyen airport (78.7%) and Longyearbyen (78.4%) were more similar to the PFOS ECF standard (76-79% linear PFOS) [Kärrman et al., 2007] than the samples from Adventdalen (89.9%) and Ny-Ålesund (96.5%), indicative of a more direct exposure to a technical mixture (local source) at Longyearbyen airport and Longyearbyen compared to Adventdalen and Ny-Ålesund. The linear PFOS isomer was in higher abundance in snow buntings breeding in Adventdalen and Ny-Ålesund compared to an ECF standard product, indicative of old legacy sources or LRT. The observed difference in isomer profile between Longyearbyen airport and Longyearbyen compared to Adventdalen and Ny-Ålesund, might result from different exposure sources, like food up-take or point source vs LRT. It is also possible that other processes could be at work that alter the PFOS isomer pattern in these biotic samples compared to the PFOS technical mixture [Chu and Letcher, 2009]. Houde et al. concluded that the differences in environmental behavior of the isomers imply that as fresh sources decline, only the linear isomer of PFOS will remain (at least in the biosphere) [Houde et al., 2008]. This could indicate that older PFAS sources have a larger fraction of linPFOS or that the source is LRT compared to local, and that a higher fraction of brPFOS suggest a newer local point source. However, Powley et al. concluded that differing pharmacokinetics complicate the use of branched to linear ratios of PFCAs in attributing their presence to a specific manufacturing process [Powley et al., 2008]. Pharmacokinetics studies have demonstrated that branched PFOA isomers are preferentially excreted, relative to linear, making quantitative source assignment impossible in biological samples [Benskin et al., 2009, De Silva et al., 2009c, De Silva et al., 2009a]. Riddel et al, however, concluded that isomer patterns should not be ignored as it may provide useful information regarding possible exposure sources [Riddell et al., 2009]. In this case, it is likely that Longyearbyen airport and Longyearbyen are close to local point sources of PFASs, while Adventdalen and Ny-Ålesund are contaminated by older PFAS sources or PFASs that have been LRT to these locations.

Overall, the most prominent PFAS group (PFSA) found in eggs of snow buntings were in agreement with previous studies where PFSAs have been found to dominate the contaminant pattern [Martin et al., 2004, Ahrens et al., 2011, Lescord et al., 2015]. Based on linPFOS:brPFOS ratio, it appear that Longyearbyen airport and Longyearbyen, with a linPFOS percentage at $\sim 78\%$, are exposed to a technical mixture from a local point source. On the other hand, Ny-Ålesund and Adventdalen appear to have a linPFOS percentage indicative of old PFAS sources or PFASs that have been long range transported.

5.3 Toxicological Implications

Among the reported effects of PFASs in different species are reproductive toxicity [Lau et al., 2003, Luebker et al., 2005], neurotoxicity [Johansson et al., 2008, Liu et al., 2010], hepatotoxicity [Miller et al., 1975, Malinverno et al., 2005], immunotoxicity [Nakayama et al., 2008, Peden-Adams et al., 2009, Grandjean et al., 2012], and effects on metabolism [Berthiaume and Wallace, 2002]. Laboratory studies have shown that PFOS toxicity result in decreased weight gain and increased liver mass in mallard (*Anas platyrhynchos*) and northern bobwhite quail (*Colinus virginianus*) [Newsted et al., 2005, Newsted et al., 2006], and higher mortality, reduced hatchability and liver histopathological changes in white leghorn chicken [Molina et al., 2006]. However, in the present study there were not found any effects on weight or size of the eggs in relation to PFAS burden.

A range of different effect-studies have been performed on different species giving varying results related to toxicity thresholds. An acute and chronic dietary exposure study of northern bobwhite quail and mallard estimated a predicted no effect concentration (PNEC) for PFOS to be 350 ng/g for reproductive endpoints such as egg production, fertility, hatchability and survival, and growth of offspring [Newsted et al., 2005]. However, concentrations in feed was not measured in the present study, making it difficult to compare levels.

Eggs of great cormorant (*Phalacrocorax carbo sinensis*), herring gull (*Larus argentatus*) and the domestic White Leghorn chicken were exposed *in ovo* (at an early or embryonic stage of development) by injection into the air sac. The lowest observed effect level (LOEL) was 900 and 1500 ng/g for PFOS and PFOA, respectively. The the lethal dose 50% (LD_{50}) was 8500 ng/g for PFOS and 2500 ng/g for PFOA while the benchmark dose (BMD_{10}; the dose on the dose-response curve where a 10% effect is seen) were 1260 ng/g and for PFOS and 1010 ng/g for PFOA [Nordén et al., 2016]. The highest $mean_{Ar}$ Σ_{22}PFAS levels found in snow buntings from the present study was 467.03 ng/g ww at Longyearbyen airport, these levels are below what is reported to have adverse effects. One egg collected at Longyearbyen airport, however, had a Σ_{22}PFAS level of 4556.15 ng/g ww, a linPFOS concentration of 2539.57 ng/g ww and a total PFOS concentration of 3233.53 ng/g ww, which is within the range, and exceeding, the levels reported for adverse effects. However, the levels found in this single sample far exceeded the levels found in the remaining samples collected at the same location. Therefore it is likely that this sample is the cause of the high Σ_{22}PFAS levels found at Longyearbyen airport. If the outlier were to be removed from the dataset, the Σ_{22}PFAS at Longyearbyen airport would become 58.12 ng/g ww, well below the toxicity thresholds reported.

Other studies by Molina et al. (2006) and O'Brien et al. (2009) estimated LD_{50} values for PFOS in eggs of domestic white leghorn chicken in range of 4900 ng/g to 93000 ng/g [O'Brien et al., 2009, Molina et al., 2006]. Based on reduced hatchability, Molina et al. estimated a lowest observed adverse effect level (LOAEL) of 100 ng/g for PFOS in eggs. Newsted et al. (2005) estimated a toxicity reference value (TRV) of 1700 ng/mL and a PNEC of 1000 ng/mL for PFOS in eggs (1 mL egg corresponds to approximately 1 g) [Newsted et al., 2005, Braune and Letcher, 2012]. Of the samples collected in the present study, there was one sample with PFOS levels higher than the LOAEL of 100 ng/g ww. The remaining samples were below the LOAEL, but certain samples were close to reaching 100 ng/g ww and indicate that the snow buntings residing close to Longyearbyen airport and Ny-Ålesund might be in danger of experiencing adverse effects caused by PFOS.

Custer et al. (2014) found a negative association between PFOS in eggs and hatching success of tree swallows (*Tachycineta bicolor*) at PFOS concentrations of 150-200 ng/g ww, which is lower than most of the effect levels found in the previous studies mentioned. However, the author mentions a low sample size (n=5-20) and the possibility that tree swallows are unusually sensitive to PFASs compared to other species. Values should thus be considered with caution [Custer et al., 2014]. However, Peden-Adams et al. (2009) examined the effects of PFOS from *in ovo* exposure in white leghorn hatchlings and observed effects at the lowest dose which averaged 154 ng/g. This indicate that effect can occur following *in ovo* exposure to environmentally relevant concentrations of PFOS [Peden-Adams et al., 2009]. When comparing these effect levels to those found in the present study, the average PFOS burden experienced for snow buntings living in proximity to Longyearbyen airport (mean$_{Ar}$ of 256.86 ng/g ww) exceeds the adverse effect levels estimated for these possible sensitive species at $\sim$ 150 ng/g ww. However, the mean$_{Geo}$ PFOS from Longyearbyen airport (28.70 ng/g ww) was below the threshold level. The location with the second highest PFOS level was Ny-Ålesund, with a mean$_{Ar}$ PFOS level of 28.25 ng/g ww, well below the suggested threshold level. The high mean$_{Ar}$ PFOS level at Longyearbyen airport was driven by one sample with extremely high detection of all PFASs analysed, likely causing the high mean$_{Ar}$ PFOS concentration at this location. The outlier was not removed considering it was sampled right next to the old fire fighting station, and likely represent actual values of PFAS burden experienced there. However, if this outlier was removed from the dataset, the mean$_{Ar}$ for PFOS would become 28.39 $\pm$ 21.92 ng/g ww, which is below the reported toxicity thresholds.

Three swallows are, in comparison with snow buntings, also a passerine species. Therefore, it is likely that the effect study performed on tree swallows are more comparable regarding adverse effect levels than the other birds, considering these two species belong to the same order. However, species specific effect studies for the snow bunting should be performed in order to elucidate whether or not the snow bunting is a sensitive species at a similar PFOS level as the tree swallows.

PFAS toxicity appear to be dependent on the specific profile in addition to the total burden experienced. Longer chain compounds with a sulfonate group appear to exhibit stronger biological effects than short chain compounds with a carboxylate group [Liao et al., 2009]. Few studies have focused on toxicity of perfluorooctane sulfonamide (PFOSA), however it appear to differ from PFOS in terms of toxicity. PFOSA has shown stronger neurotoxicity than PFOS, which might be due to an increased hydrophobicity [Slotkin et al., 2008]. PFOSA was not found at high levels in this study. Highest PFOSA levels were found for Ny-Ålesund with a mean$_{Ar}$ of 2.91 ng/g ww.

A study by Galatius et al. (2013) examined the PFAS profiles of three North Sea top predators (white-beaked dolphin (*Lagenorhynchus albirostris*), harbor porpoise (*Phocoena phocoena*), and harbor seal (*Phoca vitulina*)) in the same area and found differences in contaminant profiles, most likely as a result of different metabolic capacities [Galatius et al., 2013]. It has also been suggested that habitat use and local sources of contamination are more important determinants of PFASs in biota than trophic level. This is, however, suggested for freshwater food webs in the Canadian Arctic [Lescord et al., 2015]. This show that type of PFAS, type of organism and evolutionary traits (such as diet (vegetational vs animal-based) and metabolic capacity) need to be accounted for. Additionally, it has been suggested that annual variations in environmental conditions (such as seasonal emaciation) could increase the internal concentration of PFASs, and thus their toxic potential of exposure to PFASs [Aas et al., 2014, Bustnes et al., 2015].

The mean Σ_{22}PFAS concentration at Longyearbyen airport were in the range of 450 ng/g ww, with a mean$_{Ar}$ PFOS concentration of 250 ng/g ww. When comparing these levels to previously performed toxicity studies, the levels are mainly lower than most reported threshold levels. But the levels are also in range with what has been suggested as threshold for sensitive species ($\sim$ 150 ng/g ww PFOS for tree swallows). Hence, it should be further elucidated whether or not the snow bunting is in fact a sensitive species. Additionally, the lowest LOAEL estimated for eggs (100 ng/g ww PFOS), are in range with the levels found in snow bunting eggs from the present study. Based on these toxicity studies, it is assumed that snow buntings residing in close proximity to Longyearbyen airport could be at risk of adverse effects resulting from PFAS exposure.

Another interesting finding is that PFTriA are among the compounds found to dominate the contamination pattern at all locations examined. PFTriA have previously been found to have a potent binding affinity for the thyroid hormone receptor (TH) [Haugerud, 2011, Mortensen, 2015]. Mortensen (2015) also found higher binding affinity for the PFCAs compared to PFSAs to transthyretin (TTR) [Mortensen, 2015]. This competitive binding of PFASs with THs for the transport proteins globulin, albumin and TTR can potentially disrupt circulation of THs and possibly affect the TH homeostasis and TH-dependent functions. This is of concern because THs are important for thermogenesis, reproduction, growth and differentiation [Mortensen, 2015].

A number of toxicity threshold studies have been performed, both in different bird species and their eggs. However, the threshold levels vary greatly, making evaluations difficult as to whether or not the levels found in snow bunting eggs from the present study are high enough to cause adverse effects. Further studies should focus on identifying if the snow bunting is a sensitive species. It is also possible that some of the compounds found to dominate the contamination profile could be potent endocrine disruptors by affecting the thyroid hormone system, which is important for growth and reproduction.

5.4 Levels at the airport - AFFFs as a local source

Previous studies have linked the usage of AFFFs at fire fighting training facilities to the contamination of the environment with PFASs [Moody et al., 2002, Moody et al., 2003, Kärrman et al., 2011, Awad et al., 2011, De Solla et al., 2012]. Longyearbyen airport appear to be a point source to PFASs for the local biota living in immediate vicinity.

The present study show that the levels of PFAS contamination in eggs from snow buntings living in proximity to Longyearbyen airport is significantly more contaminated with PFASs than the remaining locations. Up to 90 x higher Σ_{22}PFAS levels were found for snow buntings residing around Longyearbyen airport compared to Adventdalen, indicating the fire fighting training facilities as a point source of PFAS contamination for the local biota. There is, however, one sample from the airport with extremely high PFAS levels measured. This sample was not removed from the dataset because it was sampled right next to the old fire fighting station at the airport. Hence, it was concluded that the sample did provide levels relevant for the sampling location. The data were tested in order to examine if PFAS levels would still be significantly higher at Longyearbyen airport compared to the other locations with the removal of this data point from the dataset. This was indeed the case (student t.test, p = 0.003).

Perfluorinated surfactants are more thermally stable than their corresponding hydrocarbon analogues. In particular, PFCAs and PFSAs are considered the most thermally stable fluorinated surfactants. In addition to thermal stability, perfluorinated surfactants are stable to acids, bases, oxidants, and reductants. This stability allows fluorinated surfactants to remain intact in environments where hydrocarbon surfactants are degraded [Moody and Field, 2000]. This is consistent with the present study, as the main PFASs found in eggs from snow buntings mainly consist of PFSAs and PFCAs. It was also found that the level of PFAS contamination decreased with increasing distance from Longyearbyen airport, through Longyearbyen and towards Adventdalen. Another interesting finding was that the ratio of PFSA:PFCA also decreased with increasing distance from Longyearbyen airport (3.0) to Longyearbyen (1.3) and dropping to 0.5 for Adventdalen. This indicates that the PFSAs are not easily transported from the local point source, or that the PFSAs are degraded to PFCAs during this short-range transport.

The intermediate levels found in Longyearbyen could be a result of transport of PFASs from Longyearbyen airport as a local source, or it could be a result of skiing activities which can release PFASs that have been used in products such as jackets or waxes applied on skiing equipment used by local residents and tourists [Freberg et al., 2010, Nilsson et al., 2010].

Realistic toxicity evaluations of AFFF mixtures and AFFF wastewater in the environment are difficult because AFFFs are complex mixtures that contain AFFF components and primary pollutants, as well as toxic burn products. Differential degradation during transport of AFFF components will also change the mixture composition and toxicity over distance and time. Additionally, the toxicity of these types of complex mixtures are difficult to assess because of the potential synergistic effects between mixture components, making it difficult to predict *a priori* the toxicity of these mixtures in the environment [Moody and Field, 2000]. The AFFF mixture also have to be considered in relation to the already complex mixture of contaminants that the snow buntings are exposed to [Choy et al., 2010, Kristoffersen, 2012], putting additional strain on the contamination burden snow buntings already experience.

Not only does AFFF compounds present their own environmental and toxiocological concerns, they also could be potential sources of perfluoroalkyl carboxylates through environmental and anthropogenic transformation [Place and Field, 2012]. The uncertainty about the original product formulations and degradation pathways, as well as studies reporting the occurrence and distribution of PFASs in environmental samples from AFFF-impacted sites are still somewhat scarce [Awad et al., 2011, D'Agostino and Mabury, 2013, Filipovic et al., 2015], and a significant data gap exists regarding the magnitude of PFASs associated with other AFFF release sites [Baduel et al., 2015]. However, from the present study it appears that the fraction of PFSAs compared to PFCAs decrease with increasing distance from a potential local point source.

5.5 Levels in Ny-Ålesund

The PFAS levels in Ny-Ålesund were dominated by PFSAs followed by PFCAs and FTSs, respectively. Main PFAS components in biota from Ny-Ålesund were PFOS, PFOSA, PFUnA, PFDoA and brPFOS. Together, these 5 PFAS components constituted 91.5% of the contamination load of the 16 different PFASs detected at this location. PFOS was the main contributor, and made up as much as 74.5% of the total PFAS burden. PFOS was found at the highest proportion at the Ny-Ålesund location. However, it is important to note that even though the proportion of PFOS is higher in Ny-Ålesund compared to Longyearbyen airport, this does not necessarily mean that the overall level of sum total PFOS (Σ_{Total}PFOS; linPFOS + brPFOS) or Σ_{22}PFASs is higher. In fact, it is actually lower in Ny-Ålesund.

When comparing the contaminant ratio between PFSA:PFCA and linPFOS:brPFOS, Ny-Ålesund separates from the other locations with a level of PFSA:PFCA ratio more than twice of what was found at Longyearbyen airport (6.9 at Ny-Ålesund compared to 3.2 at Longyearbyen airport), and a linPFOS:brPFOS ratio almost 5 fold higher than that of Longyearbyen airport (20.5 at Ny-Ålesund and 4.3 at Longyearbyen airport). Regarding the linPFOS:brPFOS ratio in Ny-Ålesund, it can be seen that the ratio is quite similar to the one found in Adventdalen. It appears that Ny-Ålesund, as well as Adventdalen, are exposed to old sources of PFASs or PFASs that have been long range transported to these locations, in contrast to Longyearbyen airport and Longyearbyen, which appear to be locations close to local point sources.

The PFSA:PFCA ratio in Ny-Ålesund were higher than all other locations. The cause of this is difficult to address, however, it could result from Ny-Ålesund being exposed to different PFAS sources compared to the other locations. It appears to be a dissimilarity between Ny-Ålesund and Longyearbyen as settlements regarding the ratio of PFSA:PFCA and linPFOS:brPFOS, possibly indicating different sources of exposure to PFASs between these two locations.

As previously mentioned, the report from Granberg et al. identify that there is no overview of chemical use or spills from the airport in Ny-Ålesund, however, PFAS containing AFFFs have likely been used in Ny-Ålesund, and perhaps in conjunction with the mines [Granberg et al., 2016]. The present study indicate that there is PFAS contamination in Ny-Ålesund, but whether or not these levels are a result of PFASs in AFFFs is uncertain. Out of the 5 samples collected in Ny-Ålesund, only 1 sample was collected in immediate vicinity to the airport and 2 samples were collected close to the old mining area, making it difficult to address if the PFAS levels found in these samples can be linked to the possible use of PFAS containing AFFF at the airport in Ny-Ålesund, the mines or other sources. Level of PFAS contamination in Ny-Ålesund were within the range found for Longyearbyen, the largest settlement in Svalbard. Whether or not the levels of PFASs found in Ny-Ålesund originate from local pollution or LRT should be further investigated.

Despite being considered a pristine area with little or no known sources of contamination, what is apparent from the present study is that level of contamination in Ny-Ålesund was within the range found in Longyearbyen. However, there was a PFSA:PFCA ratio and linPFOS:brPFOS ratio that distinguished Ny-Ålesund from the other locations. This was unexpected and warrants further study. It is also recommended to elucidate whether or not the PFAS sources in Ny-Ålesund are LRT or local, and if the magnitude of contamination by PFASs affect the local biota.

6 Concluding remarks

The results from the present study report higher PFAS levels in eggs from snow buntings residing in proximity to Longyearbyen airport compared to those residing at locations further away (Longyearbyen and Adventdalen). This indicate the fire-fighting training facilities at Longyearbyen airport as a local point source of PFASs for the local biota that contribute to the bioaccumulation of PFASs in snow buntings residing here.

Levels of PFASs found in the present study were similar to the results for eggs from snow buntings sampled in Longyearbyen and Ny-Ålesund from a previous study. PFAS levels reported in the present study also appeared to be higher than levels in eggs from glaucous gulls sampled in Ny-Ålesund, peregrine falcons sampled at Greenland and tawny owl eggs sampled in Norway. However, comparisons of snow buntings with these predatory birds, terrestrial or marine, should be performed with caution.

As previously discussed, the literature on toxic effect thresholds for PFASs are contradictive and ranging from 100 ng/g ww to 93.000 ng/g ww. Snow bunting eggs collected at Longyearbyen airport had an mean$_{Ar}$ Σ_{22}PFASs of concentration of 467.03 ng/g ww and mean$_{Geo}$ Σ_{22}PFASs of concentration of 65.16 ng/g ww, indicating that there are individuals at the airport that might experience adverse effects, however this should be further elucidated. The remaining locations examined had mean$_{Ar}$ Σ_{22}PFASs concentrations ranging from 5.02-37.91 ng/g ww, indicating that at these locations the levels of PFASs do not appear to be in range of possible adverse effects.

All locations had the highest contaminant burden resulting from two individual PFASs; PFOS and PFUnA. In general, the contaminant burden was dominated by the PFAS group PFSAs, followed by PFCAs and FTSs, respectively. The isomer profile between linPFOS and brPFOS was dominated by linPFOS for all locations. The observed isomer profile in snow bunting eggs indicate that Longyearbyen airport and Longyearbyen are exposed to local point sources with technical mixtures produced through ECF, while Adventdalen and Ny-Ålesund are exposed to old legacy sources or LRT.

7 Future Perspectives

Preliminary statistics with blm indicated significantly thinner eggshell for individuals with a higher PFAS burden, however glm did not prove any further significance. Eggshell thinning in relation to increasing PFAS burden should be further investigated in order to elucidate whether or not PFASs actually do have an effect on eggshell thickness, as seen for other POPs such as organochlorides [Tucker and Haegele, 1970, Elliott et al., 1988, Holm et al., 2006].

The range of adverse effect thresholds reported in the literature (100-93.000 ng/g ww) make it difficult to evaluate if the snow buntings are in the range of potential adverse effects. Species specific toxicity threshold studies for the snow bunting should therefore be performed. The lowest effect levels reported (100-200 ng/g ww) were reported for eggs and for presumably sensitive passerine species. Considering the snow bunting is a passerine species it should be examined if the snow bunting is a sensitive species or not. More experimental research is also needed to elucidate mechanism(s) of action in addition to more field studies.

Regarding field studies, it would be of interest to perform sampling of blood, and other tissues such as the liver, from adult female birds in order to evaluate if levels found in eggs actually can be used as a non-invasive method of sampling representative for the PFAS levels in the female that laid the egg. Elucidating possible sex differences regarding PFAS levels and accumulation would also be of interest.

Vegetational and insect studies, which examine both level of PFASs at the different locations and the BCFs and BAFs in relation to the the feed, should be performed to elucidate if the PFAS burden can be attributed to either consumption of plant materials, such as seed, or insects, or both of these diets.

Considering snow bunting nests are predated by Arctic foxes, it would be of interest to further examine if the high PFAS levels found for snow bunting eggs at Longyearbyen airport result in high levels of PFASs in Arctic foxes as well, or if their territories are large enough to offset the high levels they can encounter from feeding at eggs close to Longyearbyen airport.

There were unexpectedly high levels of PFASs in Ny-Ålesund, where PFASs contamination was expected, but not known. However, levels were lower than found at Longyearbyen airport and at a similar level of what was found for Longyearbyen (the largest settlement in Svalbard). Future studies focusing on source elucidation in Ny-Ålesund is recommended.

PFASs have proved to cause a wide range of adverse effects in both laboratory test and field studies. Seeing as how PFASs have proved to be long-lasting and stable in the environment and to cause adverse effects, the use and production of PFASs should be completely halted, and alternative compounds utilized. The use of fluorine-free foams have been debated, and alternative foams that do not contain PFASs have been developed. However, the overall performance relative to AFFF is questionable [Baduel et al., 2015].

References

[Aas et al., 2014] Aas, C. B., Fuglei, E., Herzke, D., Yoccoz, N. G., and Routti, H. (2014). Effect of body condition on tissue distribution of perfluoroalkyl substances (PFASs) in Arctic fox (*Vulpes lagopus*). *Environmental science & technology*, 48(19):11654–11661.

[Ahrens et al., 2010] Ahrens, L., Ebinghaus, R., Taniyasu, S., Yeung, L. W., Yamashita, N., and Lam, P. K. (2010). Distribution of polyfluoroalkyl compounds in water, suspended particulate matter and sediment from Tokyo Bay, Japan. *Chemosphere*, 79(3):266–272.

[Ahrens et al., 2011] Ahrens, L., Herzke, D., Huber, S., Bustnes, J. O., Bangjord, G., and Ebinghaus, R. (2011). Temporal trends and pattern of polyfluoroalkyl compounds in tawny owl (*Strix aluco*) eggs from Norway, 1986-2009. *Environmental science & technology*, 45(19):8090–8097.

[AMAP, 2007] AMAP (2007). Final report of phase I of the ACAP project on brominated flame retardants (BFRs) phase I: Inventory of sources and identification of BFR alternatives and management strategies. AMAP Report 2007: 6. Technical report, Arctic Monitoring and Assessment Programme (AMAP).

[Avinor, 2012] Avinor (2012). Miljøprosjektet DP2 miljøtekniske grunnundersøkelser Svalbard Lufthavn.

[Awad et al., 2011] Awad, E., Zhang, X., Bhavsar, S. P., Petro, S., Crozier, P. W., Reiner, E. J., Fletcher, R., Tittlemier, S. A., and Braekevelt, E. (2011). Long-term environmental fate of perfluorinated compounds after accidental release at Toronto airport. *Environmental science & technology*, 45(19):8081–8089.

[Axelson, 2014] Axelson, S. (2014). Perfluoroalkyl substances in Arctic birds.

[Baduel et al., 2015] Baduel, C., Paxman, C. J., and Mueller, J. F. (2015). Perfluoroalkyl substances in a firefighting training ground (FTG), distribution and potential future release. *Journal of Hazardous Materials*, 296:46 – 53.

[Banks et al., 1989] Banks, K. W., Clark, H., Mackay, I. R., Mackay, S. G., and Sellers, R. M. (1989). Biometrics and pre-migratory fattening in the snow bunting (*Plectrophenax nivalis*). *Ringing and Migration*, 10(3):141–158.

[Barrie et al., 1992] Barrie, L., Gregor, D., Hargrave, B., Lake, R., Muir, D., Shearer, R., Tracey, B., and Bidleman, T. (1992). Arctic contaminants: Sources, occurrence and pathways. *Science of The Total Environment*, 122(1–2):1 – 74.

[Benskin et al., 2009] Benskin, J. P., De Silva, A. O., Martin, L. J., Arsenault, G., McCrindle, R., Riddell, N., Mabury, S. A., and Martin, J. W. (2009). Disposition of perfluorinated acid isomers in sprague-dawley rats; Part 1: Single dose. *Environmental toxicology and chemistry*, 28(3):542–554.

[Berthiaume and Wallace, 2002] Berthiaume, J. and Wallace, K. B. (2002). Perfluorooctanoate, perflourooctanesulfonate, and n-ethyl perfluorooctanesulfonamido ethanol; peroxisome proliferation and mitochondrial biogenesis. *Toxicology letters*, 129(1):23–32.

[Blais et al., 2005] Blais, J. M., Kimpe, L. E., McMahon, D., Keatley, B. E., Mallory, M. L., Douglas, M. S., and Smol, J. P. (2005). Arctic seabirds transport marine-derived contaminants. *Science*, 309(5733):445–445.

[Bolduc and Guillemette, 2003] Bolduc, F. and Guillemette, M. (2003). Human disturbance and nesting success of common eiders: Interaction between visitors and gulls. *Biological Conservation*, 110(1):77–83.

[Braune and Letcher, 2012] Braune, B. M. and Letcher, R. J. (2012). Perfluorinated sulfonate and carboxylate compounds in eggs of seabirds breeding in the Canadian Arctic: Temporal trends (1975–2011) and interspecies comparison. *Environmental science & technology*, 47(1):616–624.

[Buck et al., 2011] 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. (2011). Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integrated Environmental Assessment and Management*, 7(4):513–541.

[Burkow and Kallenborn, 2000] Burkow, I. C. and Kallenborn, R. (2000). Sources and transport of persistent pollutants to the Arctic. *Toxicology Letters*, 112–113:87 – 92.

[Bustnes et al., 2015] Bustnes, J. O., Bangjord, G., Ahrens, L., Herzke, D., and Yoccoz, N. G. (2015). Perfluoroalkyl substance concentrations in a terrestrial raptor: Relationships to environmental conditions and individual traits. *Environmental toxicology and chemistry*, 34(1):184–191.

[Butt et al., 2010] Butt, C. M., Berger, U., Bossi, R., and Tomy, G. T. (2010). Levels and trends of poly- and perfluorinated compounds in the Arctic environment. *Science of The Total Environment*, 408(15):2936 – 2965.

[Cancilla et al., 1998] Cancilla, D. A., Martinez, J., and Van Aggelen, G. C. (1998). Detection of aircraft deicing/antiicing fluid additives in a perched water monitoring well at an international airport. *Environmental science & technology*, 32(23):3834–3835.

[Choy et al., 2010] Choy, E. S., Kimpe, L. E., Mallory, M. L., Smol, J. P., and Blais, J. M. (2010). Contamination of an arctic terrestrial food web with marine-derived persistent organic pollutants transported by breeding seabirds. *Environmental Pollution*, 158(11):3431 – 3438.

[Chu and Letcher, 2009] Chu, S. and Letcher, R. J. (2009). Linear and branched perfluorooctane sulfonate isomers in technical product and environmental samples by in-port derivatization-gas chromatography-mass spectrometry. *Analytical Chemistry*, 81(11):4256–4262. PMID: 19402680.

[Custer et al., 2014] Custer, C. M., Custer, T. W., Dummer, P. M., Etterson, M. A., Thogmartin, W. E., Wu, Q., Kannan, K., Trowbridge, A., and McKann, P. C. (2014). Exposure and effects of perfluoroalkyl substances in tree swallows nesting in Minnesota and Wisconsin, USA. *Archives of environmental contamination and toxicology*, 66(1):120–138.

[D'Agostino and Mabury, 2013] D'Agostino, L. A. and Mabury, S. A. (2013). Identification of novel fluorinated surfactants in aqueous film forming foams and commercial surfactant concentrates. *Environmental science & technology*, 48(1):121–129.

[Dauwe et al., 2003] Dauwe, T., Chu, S., Covaci, A., Schepens, P., and Eens, M. (2003). Great tit (*Parus major*) nestlings as biomonitors of organochlorine pollution. *Archives of environmental contamination and toxicology*, 44(1):0089–0096.

[Dauwe et al., 2007] Dauwe, T., Van de Vijver, K., De Coen, W., and Eens, M. (2007). PFOS levels in the blood and liver of a small insectivorous songbird near a fluorochemical plant. *Environment international*, 33(3):357–361.

[De Silva and Mabury, 2004] De Silva, A. O. and Mabury, S. A. (2004). Isolating isomers of perfluorocarboxylates in polar bears (*Ursus maritimus*) from two geographical locations. *Environmental science & technology*, 38(24):6538–6545.

[De Silva et al., 2009a] De Silva, A. O., Mabury, S. A., and Tseng, P. J. (2009a). Toxicokinetics of perfluorocarboxylate isomers in rainbow trout. *Environmental toxicology and chemistry*, 28(2):330–337.

[De Silva et al., 2009b] De Silva, A. O., Muir, D. C. G., and Mabury, S. A. (2009b). Distribution of perfluorocarboxylate isomers in select samples from the North American environment. *Environmental Toxicology and Chemistry*, 28(9):1801–1814.

[De Silva et al., 2009c] De Silva, A. O., Riddell, N., Benskin, J. P., Martin, L. J., Arsenault, G., McCrindle, R., Martin, J. W., and Mabury, S. A. (2009c). Disposition of perfluorinated acid isomers in sprague-dawley rats; Part 2: Subchronic dose. *Environmental toxicology and chemistry*, 28(3):555–567.

[De Solla et al., 2012] De Solla, S., De Silva, A., and Letcher, R. (2012). Highly elevated levels of perfluorooctane sulfonate and other perfluorinated acids found in biota and surface water downstream of an international airport, Hamilton, Ontario, Canada. *Environment international*, 39(1):19–26.

[Drent and Daan, 1980] Drent, R. and Daan, S. (1980). The prudent parent: Energetic adjustments in avian breeding 1. *Ardea*, 68:225–252.

[Drouillard and Norstrom, 2001] Drouillard, K. G. and Norstrom, R. J. (2001). Quantifying maternal and dietary sources of 2, 2, 4, 4, 5, 5-hexachlorobiphenyl deposited in eggs of the ring dove (*Streptopelia risoria*). *Environmental Toxicology and Chemistry*, 20(3):561–567.

[Eens et al., 2013] Eens, M., Jaspers, V. L., Van den Steen, E., Bateson, M., Carere, C., Clergeau, P., Costantini, D., Dolenec, Z., Elliott, J. E., Flux, J., et al. (2013). Can starling eggs be useful as a biomonitoring tool to study organohalogenated contaminants on a worldwide scale? *Environment international*, 51:141–149.

[Elliott et al., 1988] Elliott, J., Norstrom, R., and Keith, J. (1988). Organochlorines and eggshell thinning in northern gannets (*Sula bassanus*) from Eastern Canada, 1968–1984. *Environmental Pollution*, 52(2):81–102.

[Erten-Unal et al., 1998] Erten-Unal, M., Paranjape, S., and Schafran, G. C. (1998). Evaluation of the effects of AFFF inputs to the VIP biological nutrient removal process and pass-through toxicity-phase IA. Technical report, DTIC Document.

[Falconer et al., 2008] Falconer, C. M., Mallory, M. L., and Nol, E. (2008). Breeding biology and provisioning of nestling snow buntings in the Canadian High Arctic. *Polar Biology*, 31(4):483–489.

[Fiedler, 2007] Fiedler, H. (2007). National PCDD/PCDF release inventories under the Stockholm convention on persistent organic pollutants. *Chemosphere*, 67(9):S96–S108.

[Filipovic et al., 2015] Filipovic, M., Woldegiorgis, A., Norström, K., Bibi, M., Lindberg, M., and Österås, A.-H. (2015). Historical usage of aqueous film forming foam: A case study of the widespread distribution of perfluoroalkyl acids from a military airport to groundwater, lakes, soils and fish. *Chemosphere*, 129:39–45.

[Fossøy et al., 2014] Fossøy, F., Stokke, B. G., Kåsi, T. K., Dyrset, K., Espmark, Y., Hoset, K. S., Wedege, M. I., and Moksnes, A. (2014). Reproductive success is strongly related to local and regional climate in the Arctic snow bunting (*Plectrophenax nivalis*). *Polar Biology*.

[Freberg et al., 2010] Freberg, B. I., Haug, L. S., Olsen, R., Daae, H. L., Hersson, M., Thomsen, C., Thorud, S., Becher, G., Molander, P., and Ellingsen, D. G. (2010). Occupational exposure to airborne perfluorinated compounds during professional ski waxing. *Environmental science & technology*, 44(19):7723–7728.

[Gabrielsen, 2007] Gabrielsen, G. (2007). Levels and effects of persistent organic pollutants in arctic animals. In Ørbæk, J., Kallenborn, R., Tombre, I., Hegseth, E., Falk-Petersen, S., and Hoel, A., editors, *Arctic Alpine Ecosystems and People in a Changing Environment*, pages 377–412. Springer Berlin Heidelberg.

[Galatius et al., 2013] Galatius, A., Bossi, R., Sonne, C., Rigét, F. F., Kinze, C. C., Lockyer, C., Teilmann, J., and Dietz, R. (2013). PFAS profiles in three North Sea top predators: Metabolic differences among species? *Environmental Science and Pollution Research*, 20(11):8013–8020.

[Gebbink and Letcher, 2012] Gebbink, W. A. and Letcher, R. J. (2012). Comparative tissue and body compartment accumulation and maternal transfer to eggs of perfluoroalkyl sulfonates and carboxylates in Great Lakes herring gulls. *Environmental pollution*, 162:40–47.

[Gewurtz et al., 2013] Gewurtz, S. B., Backus, S. M., Silva, A. O. D., Ahrens, L., Armellin, A., Evans, M., Fraser, S., Gledhill, M., Guerra, P., Harner, T., Helm, P. A., Hung, H., Khera, N., Kim, M. G., King, M., Lee, S. C., Letcher, R. J., Martin, P., Marvin, C., McGoldrick, D. J., Myers, A. L., Pelletier, M., Pomeroy, J., Reiner, E. J., Rondeau, M., Sauve, M.-C., Sekela, M., Shoeib, M., Smith, D. W., Smyth, S. A., Struger, J., Spry, D., Syrgiannis, J., and Waltho, J. (2013). Perfluoroalkyl acids in the Canadian environment: Multi-media assessment of current status and trends. *Environment International*, 59:183 – 200.

[Giesy and Kannan, 2001] Giesy, J. P. and Kannan, K. (2001). Global distribution of perfluorooctane sulfonate in wildlife. *Environmental Science & Technology*, 35(7):1339–1342. PMID: 11348064.

[Glynn et al., 2012] Glynn, A., Berger, U., Bignert, A., Ullah, S., Aune, M., Lignell, S., and Darnerud, P. O. (2012). Perfluorinated alkyl acids in blood serum from primiparous women in Sweden: Serial sampling during pregnancy and nursing, and temporal trends 1996–2010. *Environmental science & technology*, 46(16):9071–9079.

[Granberg et al., 2016] Granberg, M. E., Ask, A., and Gabrielsen, G. (2016). Local contamination on Svalbard – Overview and suggestions for remediation actions. *Short report from the Norwegian Polar Institute, Tromsø, Norway*, (044):1–65.

[Grandjean et al., 2012] Grandjean, P., Andersen, E. W., Budtz-Jørgensen, E., Nielsen, F., Mølbak, K., Weihe, P., and Heilmann, C. (2012). Serum vaccine antibody concentrations in children exposed to perfluorinated compounds. *Jama*, 307(4):391–397.

[Gwiazdowicz et al., 2012] Gwiazdowicz, D., Coulson, S., Grytnes, J.-A., and Pilskog, H. (2012). The bird ectoparasite *Dermanyssus hirundinis* (Acari, Mesostigmata) in the High Arctic; A new parasitic mite to Spitsbergen, Svalbard. *Acta Parasitologica*, 57(4):378–384.

[Han et al., 2003] Han, X., Snow, T. A., Kemper, R. A., and Jepson, G. W. (2003). Binding of perfluorooctanoic acid to rat and human plasma proteins. *Chemical research in toxicology*, 16(6):775–781.

[Hanssen et al., 2013] Hanssen, L., Dudarev, A. A., Huber, S., Øyvind Odland, J., Nieboer, E., and Sandanger, T. M. (2013). 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*, 447:430 – 437.

[Haugerud, 2011] Haugerud, A. J. (2011). Levels and effects of organohalogenated contaminants on thyroid hormone levels in glaucous gulls (*Larus hyperboreus*) from Kongsfjorden, Svalbard. Master's thesis, Institutt for biologi.

[Haukås et al., 2007] Haukås, M., Berger, U., Hop, H., Gulliksen, B., and Gabrielsen, G. W. (2007). Bioaccumulation of per-and polyfluorinated alkyl substances (PFAS) in selected species from the Barents Sea food web. *Environmental Pollution*, 148(1):360–371.

[Helgason et al., 2010] Helgason, L., Sagerup, K., and Gabrielsen, G. (2010). Temporal trends and contaminant profiles of persistent organic pollutants (POPs) in seabird eggs from Northern Norway and Svalbard. *Global contamination trends of persistent organic chemicals. Taylor & Francis*.

[Higgins and Luthy, 2006] Higgins, C. P. and Luthy, R. G. (2006). Sorption of perfluorinated surfactants on sediments. *Environmental Science & Technology*, 40(23):7251–7256.

[Hoff et al., 2005] Hoff, P. T., Van de Vijver, K., Dauwe, T., Covaci, A., Maervoet, J., Eens, M., Blust, R., and De Coen, W. (2005). Evaluation of biochemical effects related to perfluorooctane sulfonic acid exposure in organohalogen-contaminated great tit (*Parus major*) and blue tit (*Parus caeruleus*) nestlings. *Chemosphere*, 61(11):1558–1569.

[Holm et al., 2006] Holm, L., Blomqvist, A., Brandt, I., Brunström, B., Ridderstråle, Y., and Berg, C. (2006). Embryonic exposure to o, p-DDT causes eggshell thinning and altered shell gland carbonic anhydrase expression in the domestic hen. *Environmental toxicology and chemistry*, 25(10):2787–2793.

[Holmström et al., 2010] Holmström, K. E., Johansson, A.-K., Bignert, A., Lindberg, P., and Berger, U. (2010). Temporal trends of perfluorinated surfactants in Swedish peregrine falcon eggs (*Falco peregrinus*), 1974- 2007. *Environmental science and technology*, 44(11):4083–4088.

[Hoset et al., 2004] Hoset, K. S., Espmark, Y., Lier, M., Haugan, T., Ingebrigtsen, M., and Moksnes, A. (2004). Effect of ambient temperature on food provisioning and reproductive success in snow buntings (*Plectrophenax nivalis*) in the High Arctic. *Ardea*, 92(2):239–246.

[Hoset et al., 2009] Hoset, K. S., Espmark, Y., Lier, M., Haugan, T., Moksnes, A., and Wedege, M. (2009). The effects of male mating behaviour and food provisioning on breeding success in snow buntings (*Plectrophenax nivalis*) in the High Arctic. *Polar Biology*, 32(11):1649–1656.

[Houde et al., 2008] Houde, M., Czub, G., Small, J. M., Backus, S., Wang, X., Alaee, M., and Muir, D. C. (2008). Fractionation and bioaccumulation of perfluorooctane sulfonate (PFOS) isomers in a Lake Ontario food web. *Environmental Science & Technology*, 42(24):9397–9403. PMID: 19174922.

[Houde et al., 2006] Houde, M., Martin, J. W., Letcher, R. J., Solomon, K. R., and Muir, D. C. (2006). Biological monitoring of polyfluoroalkyl substances: A review. *Environmental science and technology*, 40(11):3463–3473.

[Hoyt, 1979] Hoyt, D. F. (1979). Practical methods of estimating volume and fresh weight of bird eggs. *The Auk*, pages 73–77.

[Hung et al., 2010] Hung, H., Kallenborn, R., Breivik, K., Su, Y., Brorström-Lundén, E., Olafsdottir, K., Thorlacius, J. M., Leppänen, S., Bossi, R., Skov, H., et al. (2010). Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006. *Science of the Total Environment*, 408(15):2854–2873.

[Jenssen et al., 2007] Jenssen, B. M., Sormo, E. G., Bæk, K., Bytingsvik, J., Gaustad, H., Ruus, A., and Skaare, J. U. (2007). Brominated flame retardants in North-East Atlantic marine ecosystems. *Environmental health perspectives*, 115:35.

[Johansson et al., 2008] Johansson, N., Fredriksson, A., and Eriksson, P. (2008). Neonatal exposure to perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) causes neurobehavioural defects in adult mice. *Neurotoxicology*, 29(1):160–169.

[Jones et al., 2003] Jones, P. D., Hu, W., De Coen, W., Newsted, J. L., and Giesy, J. P. (2003). Binding of perfluorinated fatty acids to serum proteins. *Environmental toxicology and chemistry*, 22(11):2639–2649.

[Kannan, 2011] Kannan, K. (2011). Perfluoroalkyl and polyfluoroalkyl substances: Current and future perspectives. *Environmental chemistry*, 8(4):333–338.

[Kannan et al., 2004] Kannan, K., Corsolini, S., Falandysz, J., Fillmann, G., Kumar, K. S., Loganathan, B. G., Mohd, M. A., Olivero, J., Wouwe, N. V., Yang, J. H., et al. (2004). Perfluorooctanesulfonate and related fluorochemicals in human blood from several countries. *Environmental science & technology*, 38(17):4489–4495.

[Kärrman et al., 2011] Kärrman, A., Elgh-Dalgren, K., Lafossas, C., and Møskeland, T. (2011). Environmental levels and distribution of structural isomers of perfluoroalkyl acids after aqueous fire-fighting foam (AFFF) contamination. *Environmental chemistry*, 8(4):372–380.

[Kärrman et al., 2007] Kärrman, A., Langlois, I., van Bavel, B., Lindström, G., and Oehme, M. (2007). Identification and pattern of perfluorooctane sulfonate (PFOS) isomers in human serum and plasma. *Environment International*, 33(6):782 – 788.

[Kemsley, 1996] Kemsley, E. (1996). Discriminant analysis of high-dimensional data: a comparison of principal components analysis and partial least squares data reduction methods. *Chemometrics and intelligent laboratory systems*, 33(1):47–61.

[Key et al., 1997] Key, B. D., Howell, R. D., and Criddle, C. S. (1997). Fluorinated organics in the biosphere. *Environmental Science & Technology*, 31(9):2445–2454.

[Kishi and Arai, 2008] Kishi, T. and Arai, M. (2008). Study on the generation of perfluorooctane sulfonate from the aqueous film-forming foam. *Journal of Hazardous Materials*, 159(1):81 – 86. Papers Presented at the 2006 Annual Symposium of the Mary Kay O'Connor Process Safety Center.

[Kristoffersen, 2012] Kristoffersen, S. (2012). Organohalogenated contaminants in eggs of snow buntings (*Plectrophenax nivalis*) from human settlements in Svalbard.

[Kristoffersen et al., 2012] Kristoffersen, S., Sagerup, K., Jenssen, B. M., Warner, N., Herzke, D., and Gabrielsen, G. W. (2012). Miljøgifter i egg fra snøspurv (*Plectrophenax nivalis*) fra fire bosettinger på Svalbard. *Sluttrapport til Svalbards Miljøvernfond*, pages 1–23.

[Kwadijk et al., 2014] Kwadijk, C. J., Kotterman, M., and Koelmans, A. A. (2014). Partitioning of perfluorooctanesulfonate and perfluorohexanesulfonate in the aquatic environment after an accidental release of aqueous film forming foam at Schiphol Amsterdam airport. *Environmental toxicology and chemistry*, 33(8):1761–1765.

[Langlois et al., 2007] Langlois, I., Berger, U., Zencak, Z., and Oehme, M. (2007). Mass spectral studies of perfluorooctane sulfonate derivatives separated by high-resolution gas chromatography. *Rapid Communications in Mass Spectrometry*, 21(22):3547–3553.

[Lau et al., 2007] Lau, C., Anitole, K., Hodes, C., Lai, D., Pfahles-Hutchens, A., and Seed, J. (2007). Perfluoroalkyl acids: A review of monitoring and toxicological findings. *Toxicological sciences*, 99(2):366–394.

[Lau et al., 2003] Lau, C., Thibodeaux, J. R., Hanson, R. G., Rogers, J. M., Grey, B. E., Stanton, M. E., Butenhoff, J. L., and Stevenson, L. A. (2003). Exposure to perfluorooctane sulfonate during pregnancy in rat and mouse. II: Postnatal evaluation. *Toxicological Sciences*, 74(2):382–392.

[Leat et al., 2013] Leat, E. H. K., Bourgeon, S., Eze, J. I., Muir, D. C., Williamson, M., Bustnes, J. O., Furness, R. W., and Borgå, K. (2013). Perfluoroalkyl substances in eggs and plasma of an avian top predator, great skua (*Stercorarius skua*), in the North Atlantic. *Environmental Toxicology and Chemistry*, 32(3):569–576.

[Lescord et al., 2015] Lescord, G. L., Kidd, K. A., De Silva, A. O., Williamson, M., Spencer, C., Wang, X., and Muir, D. C. (2015). Perfluorinated and polyfluorinated compounds in lake food webs from the Canadian high Arctic. *Environmental science & technology*, 49(5):2694–2702.

[Letcher et al., 2010] Letcher, R. J., Bustnes, J. O., Dietz, R., Jenssen, B. M., Jørgensen, E. H., Sonne, C., Verreault, J., Vijayan, M. M., and Gabrielsen, G. W. (2010). Exposure and effects assessment of persistent organohalogen contaminants in Arctic wildlife and fish. *Science of The Total Environment*, 408(15):2995 – 3043. Levels, trends and effects of legacy and new persistent organic pollutants in the Arctic: An {AMAP} Assessment.

[Liao et al., 2009] Liao, C., Wang, T., Cui, L., Zhou, Q., Duan, S., and Jiang, G. (2009). Changes in synaptic transmission, calcium current, and neurite growth by perfluorinated compounds are dependent on the chain length and functional group. *Environmental science & technology*, 43(6):2099–2104.

[Liu et al., 2010] Liu, X., Liu, W., Jin, Y., Yu, W., Wang, F., and Liu, L. (2010). Effect of gestational and lactational exposure to perfluorooctanesulfonate on calcium-dependent signaling molecules gene expression in rats' hippocampus. *Archives of toxicology*, 84(1):71–79.

[Loveless et al., 2006] 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. (2006). Comparative responses of rats and mice exposed to linear/branched, linear, or branched ammonium perfluorooctanoate (APFO). *Toxicology*, 220(2):203–217.

[Luebker et al., 2002] Luebker, D., Hansen, K., Bass, N., Butenhoff, J., and Seacat, A. (2002). Interactions of fluorochemicals with rat liver fatty acid-binding protein toxicology 176: 175–185. *Find this article online*.

[Luebker et al., 2005] Luebker, D. J., York, R. G., Hansen, K. J., Moore, J. A., and Butenhoff, J. L. (2005). Neonatal mortality from *in utero* exposure to perfluorooctanesulfonate (PFOS) in sprague–dawley rats: dose–response, and biochemical and pharamacokinetic parameters. *Toxicology*, 215(1):149–169.

[Macdonald et al., 2000] Macdonald, R., Barrie, L., Bidleman, T., Diamond, M., Gregor, D., Semkin, R., Strachan, W., Li, Y., Wania, F., Alaee, M., Alexeeva, L., Backus, S., Bailey, R., Bewers, J., Gobeil, C., Halsall, C., Harner, T., Hoff, J., Jantunen, L., Lockhart, W., Mackay, D., Muir, D., Pudykiewicz, J., Reimer, K., Smith, J., Stern, G., Schroeder, W., Wagemann, R., and Yunker, M. (2000). Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. *Science of The Total Environment*, 254(2–3):93 – 234.

[Maher, 1964] Maher, W. J. (1964). Growth rate and development of endothermy in the snow bunting (*Plectrophenax nivalis*) and lapland longspur (*Calcarius lapponicus*) at Barrow, Alaska. *Ecology*, 45(3):520–528.

[Malinverno et al., 2005] Malinverno, G., Colombo, I., and Visca, M. (2005). Toxicological profile of hydrofluoropolyethers. *Regulatory toxicology and pharmacology*, 41(3):228–239.

[Martin et al., 2013] Martin, J. W., Mabury, S. A., Solomon, K. R., and Muir, D. C. (2013). Progress toward understanding the bioaccumulation of perfluorinated alkyl acids. *Environmental toxicology and chemistry*, 32(11):2421–2423.

[Martin et al., 2004] Martin, J. W., Smithwick, M. M., Braune, B. M., Hoekstra, P. F., Muir, D. C. G., and Mabury, S. A. (2004). Identification of long-chain perfluorinated acids in biota from the Canadian Arctic. *Environmental Science & Technology*, 38(2):373–380. PMID: 14750710.

[Melnes, 2014] Melnes, M. (2014). Disruptive effects of organohalogenated contaminants on thyroid hormone levels in glaucous gulls (*Larus hyperboreus*) breeding in Kongsfjorden, Svalbard. Master's thesis, Institutt for biologi.

[Meltofte, 1983] Meltofte, H. (1983). Arrival and pre-nesting period of the snow bunting (*Plectrophenax nivalis*) in East Greenland. *Polar Research*, 1(2):185–198.

[Miljeteig et al., 2012] Miljeteig, C., Gabrielsen, G. W., Strøm, H., Gavrilo, M. V., Lie, E., and Jenssen, B. M. (2012). Eggshell thinning and decreased concentrations of vitamin E are associated with contaminants in eggs of ivory gulls. *Science of the Total Environment*, 431:92–99.

[Miller et al., 1975] Miller, M. L., Clark Jr, L. C., Wesseler, E. P., Stanley, L., Emory, C., and Kaplan, S. (1975). Light microscopic morphometry and fine structure of the liver: A response to perfluorinated liquid emulsions used as artificial blood. *The Alabama journal of medical sciences*, 12(1):84.

[Moe et al., 2012] Moe, M. K., Huber, S., Svenson, J., Hagenaars, A., Pabon, M., Trümper, M., Berger, U., Knapen, D., and Herzke, D. (2012). The structure of the fire fighting foam surfactant Forafac®1157 and its biological and photolytic transformation products. *Chemosphere*, 89(7):869 – 875.

[Molina et al., 2006] Molina, E. D., Balander, R., Fitzgerald, S. D., Giesy, J. P., Kannan, K., Mitchell, R., and Bursian, S. J. (2006). Effects of air cell injection of perfluorooctane sulfonate before incubation on development of the white leghorn chicken (*Gallus domesticus*) embryo. *Environmental Toxicology and Chemistry*, 25(1):227–232.

[Moody and Field, 1999] Moody, C. A. and Field, J. A. (1999). Determination of perfluorocarboxylates in groundwater impacted by fire-fighting activity. *Environmental Science & Technology*, 33(16):2800–2806.

[Moody and Field, 2000] Moody, C. A. and Field, J. A. (2000). Perfluorinated surfactants and the environmental implications of their use in fire-fighting foams. *Environmental Science & Technology*, 34(18):3864–3870.

[Moody et al., 2003] Moody, C. A., Hebert, G. N., Strauss, S. H., and Field, J. A. (2003). Occurrence and persistence of perfluorooctanesulfonate and other perfluorinated surfactants in groundwater at a fire-training area at Wurtsmith Air Force Base, Michigan, USA. *Journal of Environmental Monitoring*, 5(2):341–345.

[Moody et al., 2002] Moody, C. A., Martin, J. W., Kwan, W. C., Muir, D. C., and Mabury, S. A. (2002). Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek. *Environmental science & technology*, 36(4):545–551.

[Mortensen, 2015] Mortensen, Å.-K. (2015). Competitive binding of persistent organic pollutants to the thyroid hormone transport protein transthyretin in glaucous gull (*Larus hyperboreus*). Master's thesis, NTNU.

[Muir and de Wit, 2010] Muir, D. C. and de Wit, C. A. (2010). Trends of legacy and new persistent organic pollutants in the circumpolar Arctic: Overview, conclusions, and recommendations. *Science of The Total Environment*, 408(15):3044 – 3051. Levels, trends and effects of legacy and new persistent organic pollutants in the Arctic: An {AMAP} Assessment.

[Nakayama et al., 2008] Nakayama, K., Iwata, H., Tao, L., Kannan, K., Imoto, M., Kim, E.-Y., Tashiro, K., and Tanabe, S. (2008). Potential effects of perfluorinated compounds in common cormorants from Lake Biwa, Japan: An implication from the hepatic gene expression profiles by microarray. *Environmental Toxicology and Chemistry*, 27(11):2378–2386.

[Newsted et al., 2005] Newsted, J. L., Coady, K., Jones, P. D., and Giesy, J. P. (2005). Avian toxicity reference values for perfluorooctane sulfonate. *Environmental science & technology*, 39(23):9357–9362.

[Newsted et al., 2006] Newsted, J. L., Gallagher, S. P., Giesy, J. P., and Beach, S. A. (2006). Pharmacokinetics and acute lethality of perfluorooctanesulfonate (PFOS) to juvenile mallard and northern bobwhite. *Archives of environmental contamination and toxicology*, 50(3):411–420.

[Nilsson et al., 2010] Nilsson, H., Kärrman, A., Westberg, H., Rotander, A., Van Bavel, B., and Lindström, G. (2010). A time trend study of significantly elevated perfluorocarboxylate levels in humans after using fluorinated ski wax. *Environmental science & technology*, 44(6):2150–2155.

[Nordén et al., 2016] Nordén, M., Berger, U., and Engwall, M. (2016). Developmental toxicity of PFOS and PFOA in great cormorant (*Phalacrocorax carbo sinensis*), herring gull (*Larus argentatus*) and chicken (*Gallus gallus domesticus*). *Environmental Science and Pollution Research*, 23(11):10855–10862.

[O'Brien et al., 2009] O'Brien, J. M., Carew, A. C., Chu, S., Letcher, R. J., and Kennedy, S. W. (2009). Perfluorooctane sulfonate (PFOS) toxicity in domestic chicken (*Gallus gallus domesticus*) embryos in the absence of effects on peroxisome proliferator activated receptor alpha (PPARα)-regulated genes. *Comparative Biochemistry and Physiology Part C: Toxicology & Pharmacology*, 149(4):524–530.

[Oksanen et al., 2017] Oksanen, J., Blanchet, F. G., Friendly, M., Kindt, R., Legendre, P., McGlinn, D., Minchin, P. R., O'Hara, R. B., Simpson, G. L., Solymos, P., Stevens, M. H. H., Szoecs, E., and Wagner, H. (2017). *vegan: Community Ecology Package*. R package version 2.4-2.

[Olsen et al., 2007] Olsen, G. W., Burris, J. M., Ehresman, D. J., Froehlich, J. W., Seacat, A. M., Butenhoff, J. L., and Zobel, L. R. (2007). Half-life of serum elimination of perfluorooctanesulfonate, perfluorohexanesulfonate, and perfluorooctanoate in retired fluorochemical production workers. *Environmental health perspectives*, pages 1298–1305.

[Parsons et al., 2008] Parsons, J. R., Sáez, M., Dolfing, J., and de Voogt, P. (2008). Biodegradation of perfluorinated compounds. In *Reviews of Environmental Contamination and Toxicology Vol 196*, pages 53–71. Springer.

[Paul et al., 2008] Paul, A. G., Jones, K. C., and Sweetman, A. J. (2008). A first global production, emission, and environmental inventory for perfluorooctane sulfonate. *Environmental Science & Technology*, 43(2):386–392.

[Peden-Adams et al., 2009] Peden-Adams, M. M., Stuckey, J. E., Gaworecki, K. M., Berger-Ritchie, J., Bryant, K., Jodice, P. G., Scott, T. R., Ferrario, J. B., Guan, B., Vigo, C., et al. (2009). Developmental toxicity in white leghorn chickens following *in ovo* exposure to perfluorooctane sulfonate (PFOS). *Reproductive Toxicology*, 27(3):307–318.

[Persson et al., 2013] Persson, S., Rotander, A., Kärrman, A., van Bavel, B., and Magnusson, U. (2013). Perfluoroalkyl acids in subarctic wild male mink (*Neovison vison*) in relation to age, season and geographical area. *Environment international*, 59:425–430.

[Pilskog et al., 2014] Pilskog, H. E., Solhøy, T., Gwiazdowicz, D. J., Grytnes, J.-A., and Coulson, S. J. (2014). Invertebrate communities inhabiting nests of migrating passerine, wild fowl and sea birds breeding in the High Arctic, Svalbard. *Polar Biology*, 37(7):981–998.

[Place and Field, 2012] Place, B. J. and Field, J. A. (2012). Identification of novel fluorochemicals in aqueous film-forming foams used by the US military. *Environmental science & technology*, 46(13):7120–7127.

[Powley et al., 2005] Powley, C. R., George, S. W., Buck, R. C., and Ryan, T. W. (2005). Matrix effect-free analytical methods for determination of perfluorinated carboxylic acids in environmental matrixes. *Analytical Chemistry*, 77(19):6353–6358. PMID: 16194099.

[Powley et al., 2008] Powley, C. R., George, S. W., Russell, M. H., Hoke, R. A., and Buck, R. C. (2008). Polyfluorinated chemicals in a spatially and temporally integrated food web in the Western Arctic. *Chemosphere*, 70(4):664 – 672.

[Pérez et al., 2013] Pérez, F., Nadal, M., Navarro-Ortega, A., Fàbrega, F., Domingo, J. L., Barceló, D., and Farré, M. (2013). Accumulation of perfluoroalkyl substances in human tissues. *Environment International*, 59:354 – 362.

[R Core Team, 2016] R Core Team (2016). *R: A Language and Environment for Statistical Computing*. R Foundation for Statistical Computing, Vienna, Austria.

[Rayne et al., 2008] Rayne, S., Forest, K., and Friesen, K. J. (2008). Congener-specific numbering systems for the environmentally relevant C4 through C8 perfluorinated homologue groups of alkyl sulfonates, carboxylates, telomer alcohols, olefins, and acids, and their derivatives. *Journal of Environmental Science and Health Part A*, 43(12):1391–1401.

[Riddell et al., 2009] Riddell, N., Arsenault, G., Benskin, J. P., Chittim, B., Martin, J. W., McAlees, A., and McCrindle, R. (2009). Branched perfluorooctane sulfonate isomer quantification and characterization in blood serum samples by HPLC/ESI-MS(/MS). *Environmental Science & Technology*, 43(20):7902–7908. PMID: 19921912.

[Romanoff and Romanoff, 1949] Romanoff, A. L. and Romanoff, A. J. (1949). The avian egg.

[Ryzhanovsky, 2015] Ryzhanovsky, V. (2015). Comparative ecology of horned lark (*Eremophila alpestris flava* gm.) and snow bunting (*Plectrophenax nivalis* l.) in subarctic and Arctic zones. *Contemporary Problems of Ecology*, 8(3):309–316.

[Schultz et al., 2004] Schultz, M. M., Barofsky, D. F., and Field, J. A. (2004). Quantitative determination of fluorotelomer sulfonates in groundwater by LC MS/MS. *Environmental science & technology*, 38(6):1828–1835.

[Skøien, 2015] Skøien, E. A. (2015). Causes and consequences of breeding synchrony in the snow bunting (*Plectrophenax nivalis*). Master's thesis, NTNU.

[Slotkin et al., 2008] Slotkin, T. A., MacKillop, E. A., Melnick, R. L., Thayer, K. A., and Seidler, F. J. (2008). Developmental neurotoxicity of perfluorinated chemicals modeled *in vitro*. *Environmental health perspectives*, 116(6):716.

[Smith, 1994] Smith, R. D. (1994). Snow buntings (*Plectrophenax nivalis*): The behavioural ecology and site use of an itinerant flock species in the nonbreeding season. PhD thesis.

[Stahl et al., 2011] Stahl, T., Mattern, D., and Brunn, H. (2011). Toxicology of perfluorinated compounds. *Environmental Sciences Europe*, 23(1):38.

[StockholmConvention, 2002] StockholmConvention (2002). Stockholm convention on persistent organic pollutants (Annex C). *America*, 8.

[Tucker and Haegele, 1970] Tucker, R. K. and Haegele, H. (1970). Eggshell thinning as influenced by method of DDT exposure. *Bulletin of environmental contamination and toxicology*, 5(3):191–194.

[van Paassen et al., 1984] van Paassen, A. G., Veldman, D. H., and Beintema, A. J. (1984). A simple device for determination of incubation stages in eggs. *Wildfowl*, 35(35).

[Verreault et al., 2005] Verreault, J., Houde, M., Gabrielsen, G. W., Berger, U., Haukås, M., Letcher, R. J., and Muir, D. C. (2005). Perfluorinated alkyl substances in plasma, liver, brain, and eggs of glaucous gulls (*Larus hyperboreus*) from the Norwegian Arctic. *Environmental science & technology*, 39(19):7439–7445.

[Vicente et al., 2012] Vicente, J., Bertolero, A., Meyer, J., Viana, P., and Lacorte, S. (2012). Distribution of perfluorinated compounds in yellow-legged gull eggs (*Larus michahellis*) from the Iberian Peninsula. *Science of the total environment*, 416:468–475.

[Vorkamp et al., 2005] Vorkamp, K., Thomsen, M., Falk, K., Leslie, H., Møller, S., and Sørensen, P. B. (2005). Temporal development of brominated flame retardants in peregrine falcon (*Falco peregrinus*) eggs from South Greenland (1986- 2003). *Environmental science & technology*, 39(21):8199–8206.

[Wang et al., 2016] Wang, Y., Vestergren, R., Shi, Y., Cao, D., Xu, L., Cai, Y., Zhao, X., and Wu, F. (2016). Identification, tissue distribution, and bioaccumulation potential of cyclic perfluorinated sulfonic acids isomers in an airport impacted ecosystem. *Environmental Science & Technology*, 50(20):10923–10932. PMID: 27672706.

[Weiss et al., 2012] Weiss, O., Wiesmüller, G. A., Bunte, A., Göen, T., Schmidt, C. K., Wilhelm, M., and Hölzer, J. (2012). Perfluorinated compounds in the vicinity of a fire training area–human biomonitoring among 10 persons drinking water from contaminated private wells in Cologne, Germany. *International journal of hygiene and environmental health*, 215(2):212–215.

[Wickham, 2009] Wickham, H. (2009). *ggplot2: Elegant Graphics for Data Analysis*. Springer-Verlag New York.

[Wickham, 2011] Wickham, H. (2011). The split-apply-combine strategy for data analysis. *Journal of Statistical Software*, 40(1):1–29.

[Xie et al., 2013] Xie, S., Wang, T., Liu, S., Jones, K. C., Sweetman, A. J., and Lu, Y. (2013). Industrial source identification and emission estimation of perfluorooctane sulfonate in China. *Environment international*, 52:1–8.

[Yoo et al., 2009] Yoo, H., Guruge, K. S., Yamanaka, N., Sato, C., Mikami, O., Miyazaki, S., Yamashita, N., and Giesy, J. P. (2009). Depuration kinetics and tissue disposition of PFOA and PFOS in white leghorn chickens (*Gallus gallus*) administered by subcutaneous implantation. *Ecotoxicology and environmental safety*, 72(1):26–36.

[Zhang et al., 2012] Zhang, L., Liu, J., Hu, J., Liu, C., Guo, W., Wang, Q., and Wang, H. (2012). The inventory of sources, environmental releases and risk assessment for perfluorooctane sulfonate in China. *Environmental Pollution*, 165:193 – 198. Chemicals Management and Environmental Assessment of Chemicals in China.

[Zhao et al., 2014] Zhao, L., Zhang, Y., Fang, S., Zhu, L., and Liu, Z. (2014). Comparative sorption and desorption behaviors of {PFHxS} and {PFOS} on sequentially extracted humic substances. *Journal of Environmental Sciences*, 26(12):2517 – 2525.

A Appendices

A.1 Duplicate Samples

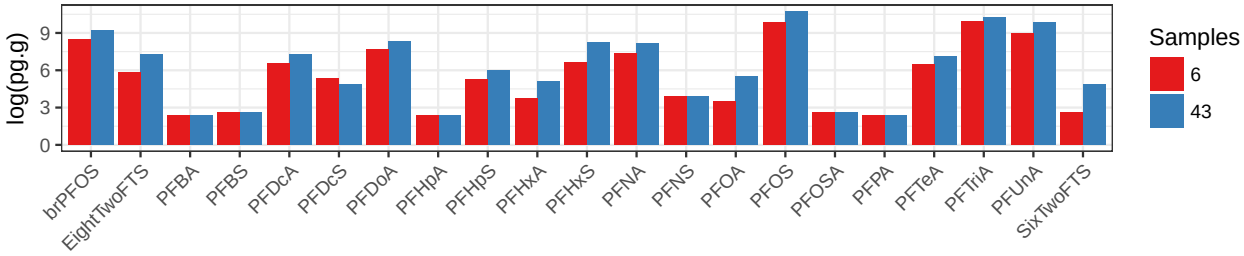


Figure A.1: Summary of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) found in the first duplicate sampled eggs of the same nest from snow buntings (*Plectrophenax nivalis*) in Longyearbyen, Svalbard and Ny-Ålesund, Svalbard in 2016. Concentrations are log-transformed.

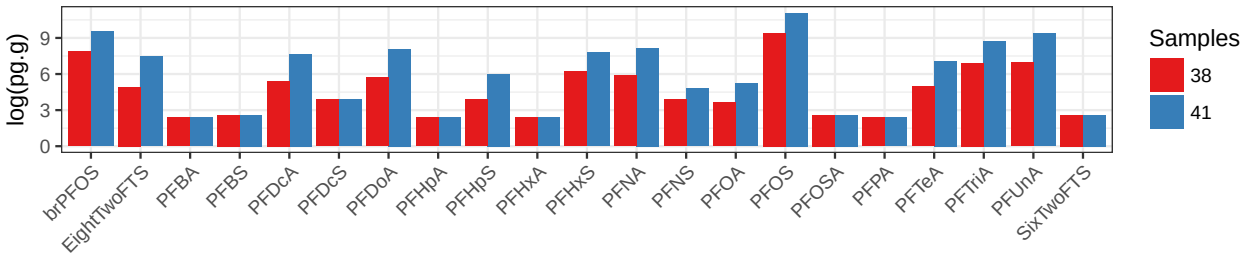
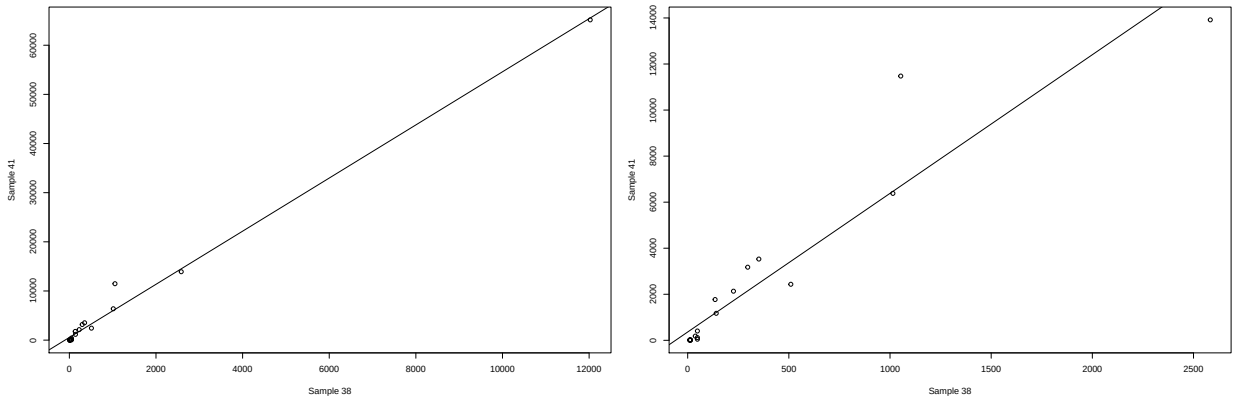
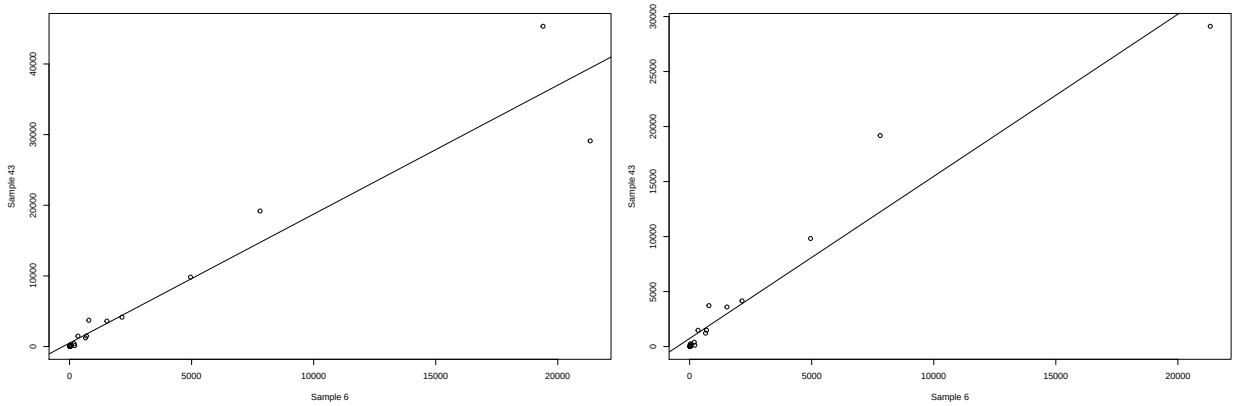


Figure A.2: Summary of the perfluoroalkyl and polyfluoroalkyl substances (PFASs) found in the second duplicate sampled eggs of the same nest from snow buntings (*Plectrophenax nivalis*) in Longyearbyen, Svalbard and Ny-Ålesund, Svalbard in 2016. Concentrations are log-transformed.



(a) Correlation with the whole dataset **(b)** Correlation with one outlier removed

Figure A.3: Correlation regression of the two duplicate samples 6 and 43 , sampled from the airport in Longyearbyen at Svalbard in 2016: **(a)** Whole dataset included in analysis (p-value $<$ 2.2e-16, corr = 0.9956143), **(b)** One outlier removed from the analysis (p-value = 4.89e-10, corr = 0.9430135)



(a) Correlation with whole dataset **(b)** Correlation with one outlier removed

Figure A.4: Correlation regression of the two duplicate samples 38 and 41, sampled from the airport in Longyearbyen at Svalbard in 2016: **(a)** Whole dataset included in analysis (p-value = 8.264e-12, corr = 0.9585103), **(b)** One outlier removed from the analysis (p-value = 4.063e-12, corr = 0.9668573)

A.2 Rawdata

Table A.1: Rawdata containing the concentration of each per- and polyfluorinated alkylated compound (ng/g w.w.) analysed for in eggs of snow buntings (*Plectrophenax nivalis*) collected in Longyearbyen and Ny-Ålesund (NÅ, n = 5), 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). The PFASs analysed for were divided into perfluororalkyl carboxylic acids (PFCAs), perfluoro sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Concentration below the detection limit (LOD) is reported as LOD/2.

Sample	Location	PFSA									PFCA										FTS	
		PFOSA	PFBS	PFPS	PFHxS	PFHpS	brPFOS	linPFOS	PFNS	PFDcS	PFBA	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDcA	PFUnA	PFDoA	PFTriA	PFTeA	6:2 FTS	8:2 FTS
05	AIR	0.01	0.01	0.29	0.08	3.16	12.13	0.05	0.05	0.01	0.01	0.04	0.01	0.06	1.50	1.17	4.18	1.64	4.75	0.84	0.01	3.83	33.85
06	AIR	0.01	0.01	2.26	0.29	7.39	32.37	0.05	0.17	0.01	0.01	0.10	0.01	0.14	2.56	1.09	13.49	3.15	25.22	0.94	0.07	0.91	90.29
13	AIR	2.46	3.67	126.38	21.00	693.96	2539.57	29.89	45.39	3.49	0.29	0.76	0.50	12.62	113.11	46.48	461.90	84.84	343.43	13.74	0.01	12.65	4556.15
14	AIR	0.01	0.01	0.21	0.05	2.06	8.51	0.05	0.05	0.01	0.01	0.06	0.01	0.13	0.61	0.47	1.97	0.01	1.39	0.17	0.01	4.26	20.08
21	AIR	0.01	0.01	1.52	0.05	14.28	51.77	0.05	0.05	0.01	0.01	0.01	0.01	0.16	5.61	1.63	8.83	0.62	3.60	0.37	0.01	14.27	102.90
23	AIR	0.01	0.01	0.32	0.05	2.48	12.67	0.05	0.05	0.01	0.01	0.01	0.01	0.01	2.11	0.35	2.70	0.53	2.32	0.21	0.01	1.72	25.66
24	AIR	0.01	0.01	0.01	0.05	0.74	3.41	0.05	0.05	0.01	0.01	0.01	0.01	0.01	0.66	0.59	2.77	0.49	2.68	0.47	0.01	0.70	12.78
33	AIR	0.01	0.12	5.84	0.46	14.00	59.92	0.05	0.05	0.01	0.01	0.01	0.01	0.05	3.71	2.39	13.30	3.05	12.09	1.11	0.01	3.15	119.37
34	AIR	0.01	0.01	0.08	0.05	0.83	4.09	0.05	0.05	0.01	0.01	0.01	0.01	0.08	0.73	0.52	2.08	0.53	1.97	0.27	0.01	0.39	12.39
36	AIR	0.01	0.01	0.69	0.21	14.79	60.44	0.10	0.05	0.01	0.01	0.02	0.01	0.22	2.49	1.60	5.70	1.49	3.79	0.69	0.01	6.21	98.58
38	AIR	0.01	0.01	1.47	0.23	8.25	38.59	0.08	0.05	0.01	0.01	0.01	0.01	0.11	1.94	1.18	6.26	1.74	3.70	0.66	0.01	0.95	65.31
02	LYB	0.01	0.01	0.38	0.05	4.89	15.76	0.05	0.05	0.56	0.01	0.21	0.13	1.68	3.89	4.07	7.96	3.59	3.77	1.51	0.01	7.15	55.81
19	LYB	0.01	0.01	0.17	0.05	0.05	4.31	0.05	0.05	0.01	0.01	0.01	0.01	0.10	0.44	0.44	1.68	0.01	1.40	0.01	0.01	0.05	9.06
20	LYB	0.01	0.01	1.65	0.05	6.47	21.59	0.05	0.05	0.01	0.01	0.01	0.01	0.32	0.92	1.10	2.38	1.14	1.06	0.20	0.01	1.73	39.03
26	LYB	0.01	0.01	0.01	0.05	0.75	2.36	0.05	0.05	0.01	0.01	0.01	0.01	0.03	0.47	0.70	2.14	0.52	1.38	0.49	0.01	0.05	9.46
01	ADV	0.01	0.01	0.01	0.07	0.05	0.76	3.31	0.05	0.05	0.01	0.01	0.06	0.04	0.69	0.41	1.41	0.69	1.25	0.28	0.01	0.18	9.36
04	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.52	0.05	0.05	0.01	0.01	0.05	0.03	0.17	0.25	0.87	0.49	0.98	0.15	0.01	0.05	3.91
08	ADV	0.45	0.01	0.01	0.01	0.05	0.05	0.92	0.05	0.05	0.01	0.01	0.04	0.01	0.36	0.56	1.54	1.06	1.62	0.50	0.01	0.46	7.80
09	ADV	0.01	0.01	0.01	0.12	0.05	0.51	2.38	0.05	0.05	0.01	0.01	0.08	0.04	0.60	0.84	2.27	0.58	1.62	0.21	0.01	0.05	9.51
10	ADV	0.01	0.01	0.01	0.01	0.05	0.05	1.57	0.05	0.05	0.01	0.01	0.03	0.03	0.19	0.27	0.99	0.27	0.86	0.18	0.01	0.05	4.73
11	ADV	0.01	0.01	0.01	0.01	0.05	0.05	1.67	0.05	0.05	0.01	0.01	0.08	0.03	0.24	0.14	0.01	0.01	0.01	10.32	0.01	0.05	12.86
15	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.05	0.05	0.01	0.01	0.07	0.01	0.33	0.22	0.44	0.01	0.66	0.20	0.01	0.05	2.32
16	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.85	0.05	0.05	0.01	0.01	0.01	0.01	0.24	0.32	0.92	0.46	1.07	0.20	0.01	0.05	4.44
17	ADV	0.01	0.01	0.01	0.01	0.05	0.05	1.52	0.05	0.05	0.01	0.01	0.10	0.06	0.47	0.25	0.70	0.01	0.01	0.01	0.01	0.05	3.47
18	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.89	0.05	0.05	0.01	0.01	0.01	0.01	0.31	0.47	1.61	0.88	1.86	0.27	0.01	0.05	6.66
22	ADV	0.01	0.01	0.01	0.01	0.05	0.05	1.61	0.05	0.05	0.01	0.01	0.01	0.01	0.62	0.06	0.01	0.01	0.01	0.01	0.01	0.05	2.71
25	ADV	0.01	0.01	0.01	0.01	0.05	0.63	2.47	0.05	0.05	0.01	0.01	0.01	0.01	0.50	0.90	2.02	0.01	3.09	0.43	0.01	0.05	10.38
27	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.05	0.05	0.01	0.01	0.01	0.01	0.68	0.47	1.25	0.01	0.01	0.01	0.01	0.05	2.85
29	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.32	0.05	0.05	0.01	0.01	0.01	0.06	0.14	0.20	0.67	0.01	0.21	0.05	0.01	0.05	2.00
31	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.73	0.05	0.05	0.01	0.01	0.01	0.01	0.33	0.43	1.84	0.48	0.96	0.23	0.01	0.05	5.35
32	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.71	0.05	0.05	0.01	0.01	0.01	0.02	0.47	0.28	0.94	0.01	0.73	0.01	0.01	0.05	3.53
35	ADV	0.01	0.01	0.01	0.31	0.05	0.05	2.09	0.05	0.05	0.01	0.01	0.01	0.16	0.80	0.83	1.90	0.56	0.13	0.01	0.01	0.56	7.64
37	ADV	0.01	0.01	0.01	0.01	0.05	0.15	0.61	0.05	0.05	0.01	0.01	0.01	0.01	0.19	0.23	0.52	0.36	0.36	0.07	0.01	0.05	2.79
39	ADV	0.01	0.01	0.01	0.01	0.05	0.05	0.29	0.05	0.05	0.01	0.01	0.01	0.07	0.34	0.09	0.68	0.01	0.01	0.01	0.01	0.05	1.87
40	ADV	0.01	0.01	0.01	0.06	0.05	0.05	1.38	0.05	0.05	0.01	0.01	0.01	0.04	0.42	0.38	0.01	0.01	0.01	0.01	0.01	0.05	2.67
42	ADV	0.31	0.01	0.01	0.01	0.05	0.05	0.05	0.05	0.05	0.01	0.01	0.01	0.01	0.19	0.01	0.01	0.01	0.01	0.01	0.01	0.05	0.97
44	ADV	0.01	0.01	0.01	0.17	0.05	0.05	1.06	0.05	0.05	0.01	0.01	0.01	0.01	0.43	0.50	1.12	0.76	0.26	0.01	0.01	0.05	4.68
45	ADV	0.67	0.01	0.01	0.04	0.05	0.05	1.35	0.05	0.05	0.01	0.01	0.01	0.04	0.19	0.12	0.24	0.01	0.01	0.01	0.01	0.05	3.02
03	NÅ	0.01	0.01	0.72	0.11	4.31	45.39	0.10	0.69	0.01	0.01	0.06	0.01	0.17	0.89	0.91	2.04	0.71	1.52	0.27	0.01	1.12	59.09
07	NÅ	0.01	0.01	0.01	0.05	0.05	1.76	0.05	0.05	0.01	0.01	0.03	0.01	0.05	0.91	0.81	0.89	3.60	0.58	0.01	0.01	0.05	8.99
12	NÅ	14.49	0.01	0.01	0.05	0.05	1.34	0.05	0.05	0.01	0.01	0.11	0.01	0.01	0.53	0.91	1.94	0.01	0.73	1.32	0.01	0.05	21.72
28	NÅ	0.01	0.01	0.01	0.05	0.05	88.21	0.05	0.05	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.05	88.65
30	NÅ	0.01	0.01	0.01	0.05	0.64	4.59	0.05	0.05	0.01	0.01	0.01	0.01	0.04	0.42	0.66	2.04	1.12	1.03	0.26	0.01	0.05	11.10

A.3 LOD and LOQ values

Table A.2: Limit of detection (LOD) and limit of quantification (LOQ) for the 3 different groups of per- and polyfluorinated alykl substances (PFASs) analysed for in eggs of snow buntings (*Plectrophenax nivalis*). The three groups are perfluorinated carboxylic acids (PFCAs), perfluorinated sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). LOD calculated as 3x blank or 3x signal/noise and LOQ as 3x LOD. Both LOD and LOQ are given in pg/g wet weight (ww).

Group	Individual PFAS	Abbreviation	LOD (pg/g ww)	LOQ (pg/g ww)
PFCA	Perfluorobutaneoate	PFBA	22	66
	Perfluoropentaneoate	PFPA	22	66
	Perfluorohexanoate	PFHxS	22	66
	Perfluoroheptanoate	PFHpA	22	66
	Perfluorooctaneoate	PFOA	22	66
	Perfluorononanoate	PFNA	22	66
	Perfluorodecanoate	PFDcA	22	66
	Perfluoroundercanoate	PFUnA	27	81
	Perfluorododecanoate	PFDoA	27	81
	Perfluorotridecanoic acid	PFTriA	27	81
	Perfluorotetradecanoate	PFTeA	27	81
PFSA	Perfluorobutane sulfonate	PFBS	27	81
	Perfluoropentanesulfonic acid	PFPS	27	81
	Perfluorohexane sulfonate	PFHxS	27	81
	Perfluoroheptanesulphonic acid	PFHpS	96	288
	Perfluorooctanesulfonamide	PFOSA	27	81
	Perfluorooctane sulfonate	linPFOS	96	288
	Perfluorooctane sulfonate	brPFOS	96	288
	Perfluorononanesulphonic acid	PFNS	96	288
	Perfluorodecane sulfonic acid	PFDcS	96	288
FTS	6:2 Fluorotelomer sulfonic acid	6:2 FTS	27	81
	8:2 Fluorotelomer sulfonic acid	8:2 FTS	96	288

A.4 Histograms

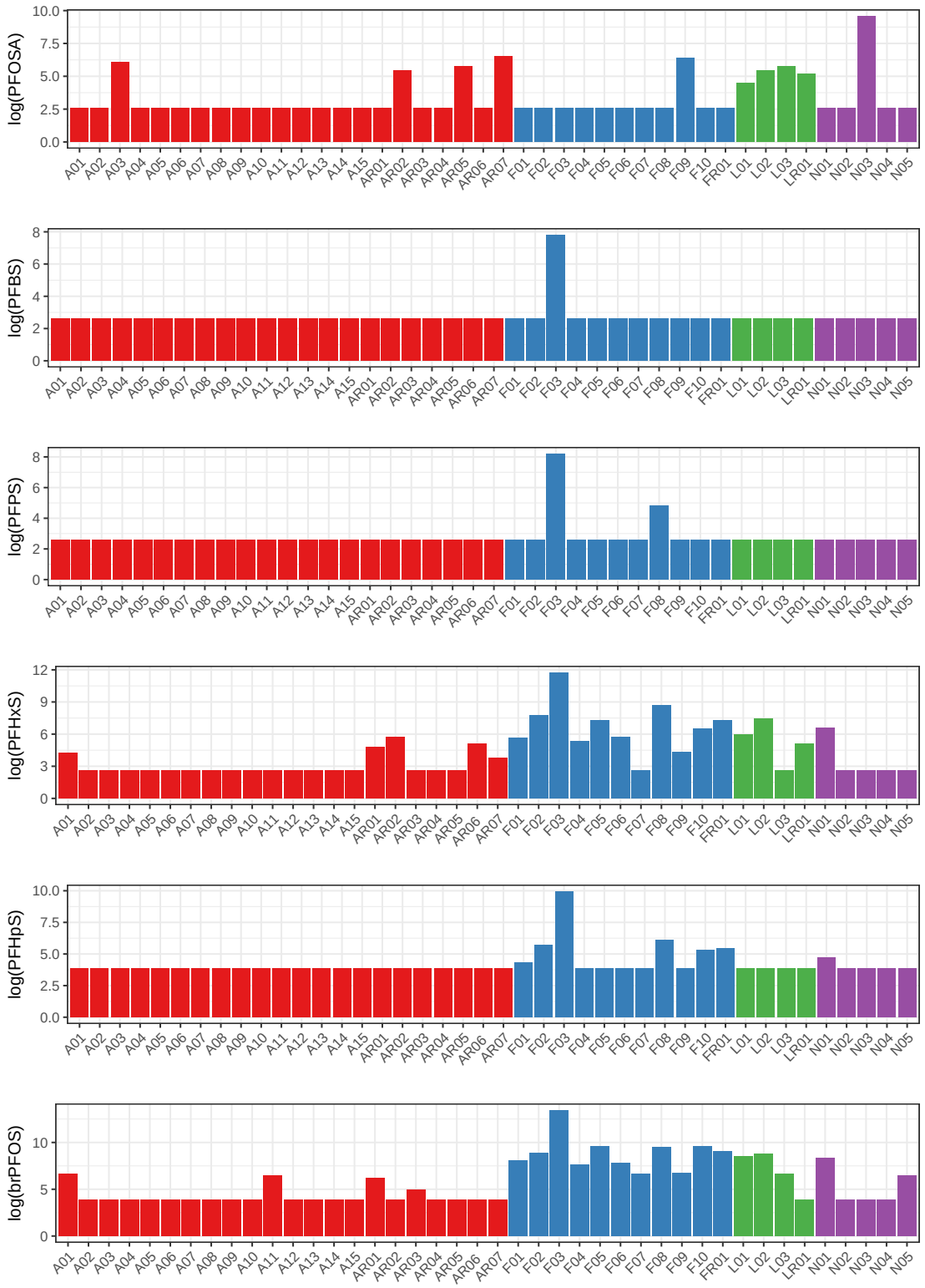


Figure A.5: Histogram showing the concentrations (logtransformed, ng/g ww) of the individual per- and polyfluorinated alkyl substances (PFASs) (PFASs; compound 1-6 out of the 22) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen Airport (F (blue); n=11), Longyearbyen (L (green); n=4) and Adventdalen (A (red); n=23) (Longyearbyen) and in Ny-Ålesund (N (purple); n=5), at Svalbard in 2016. Values are log-transformed and non-detects included with LOD/2 (limit of detection).

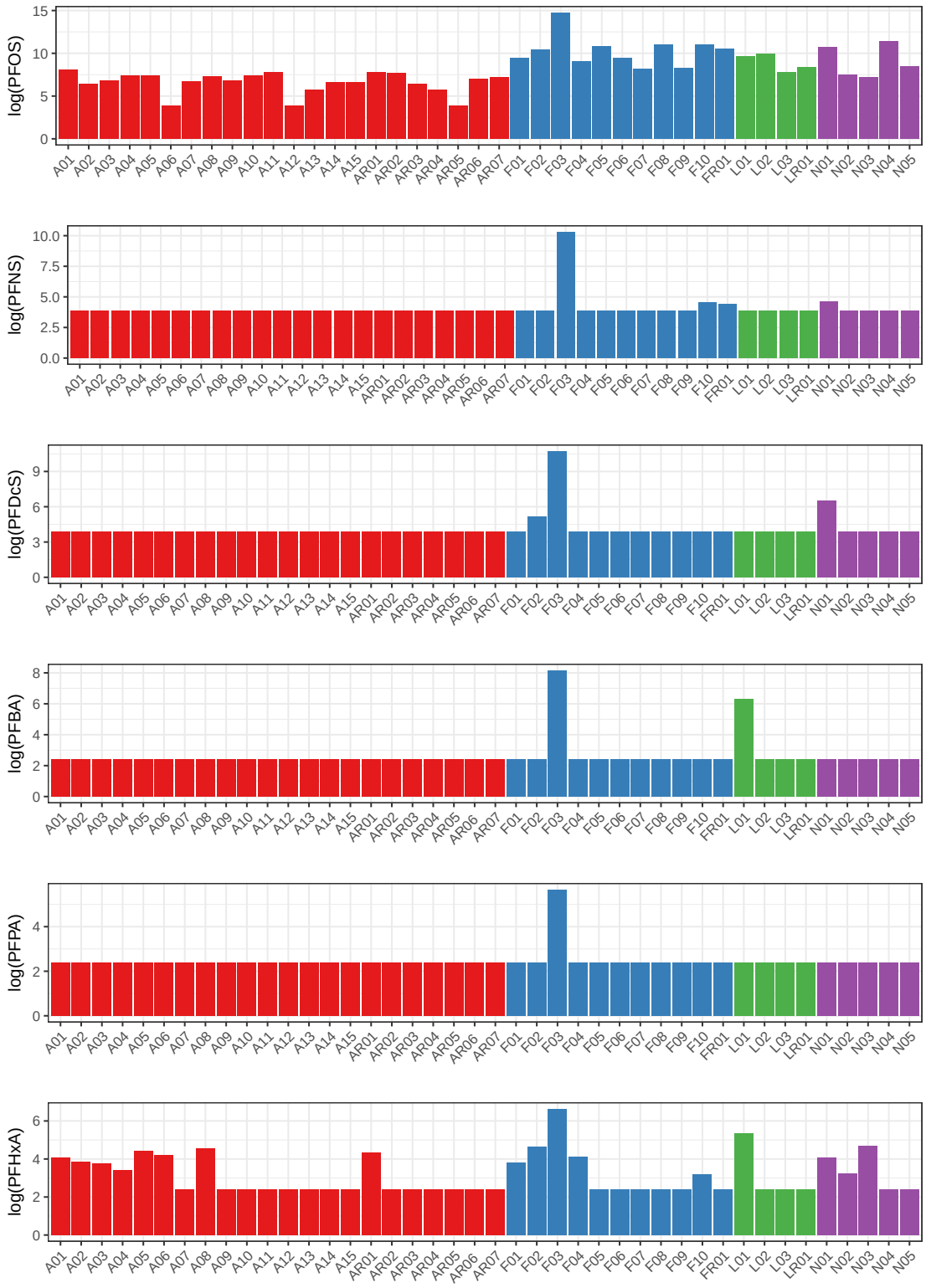


Figure A.6: Histogram showing the concentrations (logtransformed, ng/g ww) of the individual per- and polyfluorinated alkyl substances (PFASs) (PFASs; compound 7-12 out of the 22) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen Airport (F (blue); n=11), Longyearbyen (L (green); n=4) and Adventdalen (A (red); n=23) (Longyearbyen) and in Ny-Ålesund (N (purple); n=5), at Svalbard in 2016. Values are log-transformed and non-detects included with LOD/2 (limit of detection).

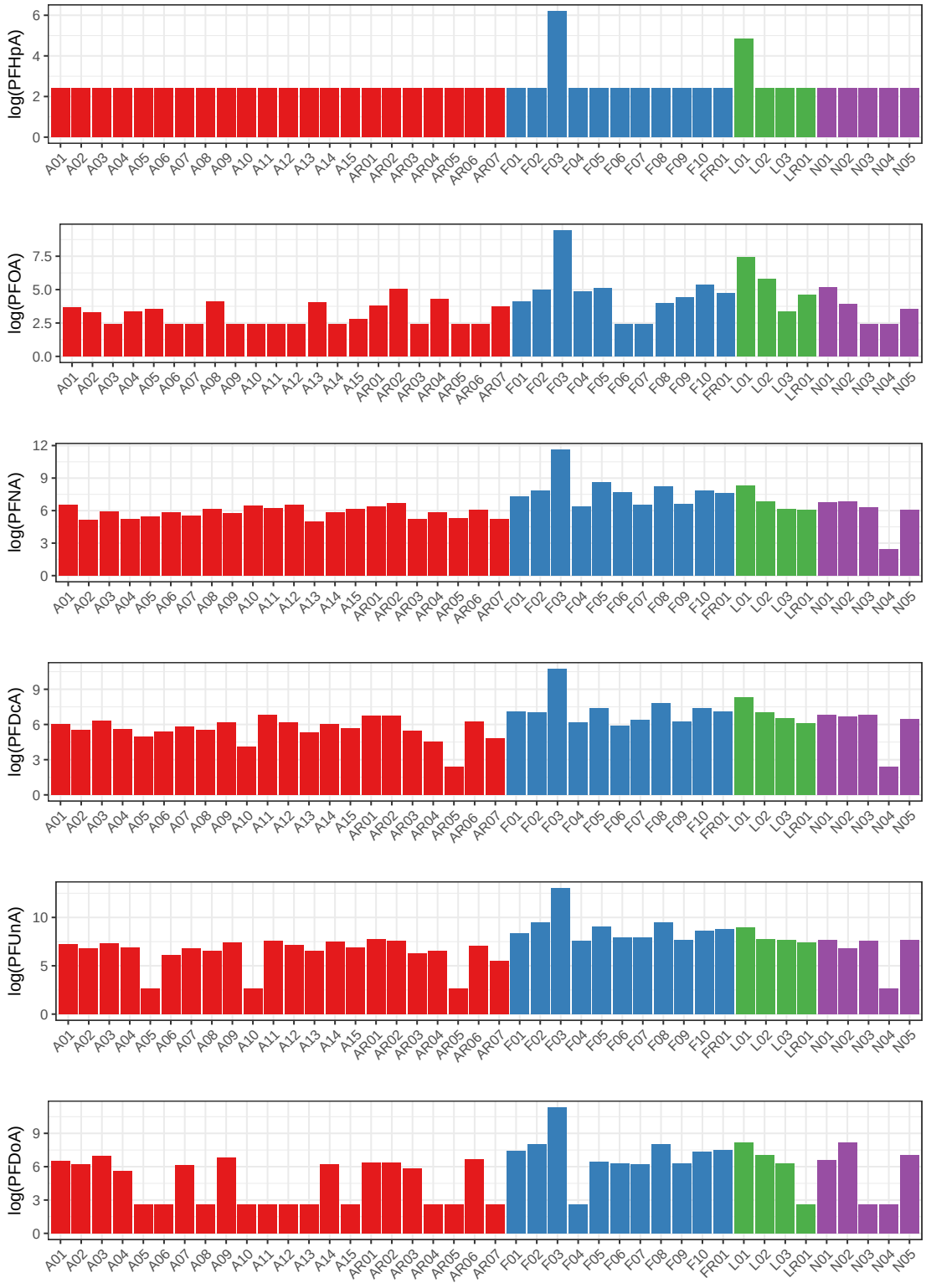


Figure A.7: Histogram showing the concentrations (logtransformed, ng/g ww) of the individual per- and polyfluorinated alkyl substances (PFASs) (PFASs; compound 13-18 out of the 22) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen Airport (F (blue); n=11), Longyearbyen (L (green); n=4) and Adventdalen (A (red); n=23) (Longyearbyen) and in Ny-Ålesund (N (purple); n=5), at Svalbard in 2016. Values are log-transformed and non-detects included with LOD/2 (limit of detection).

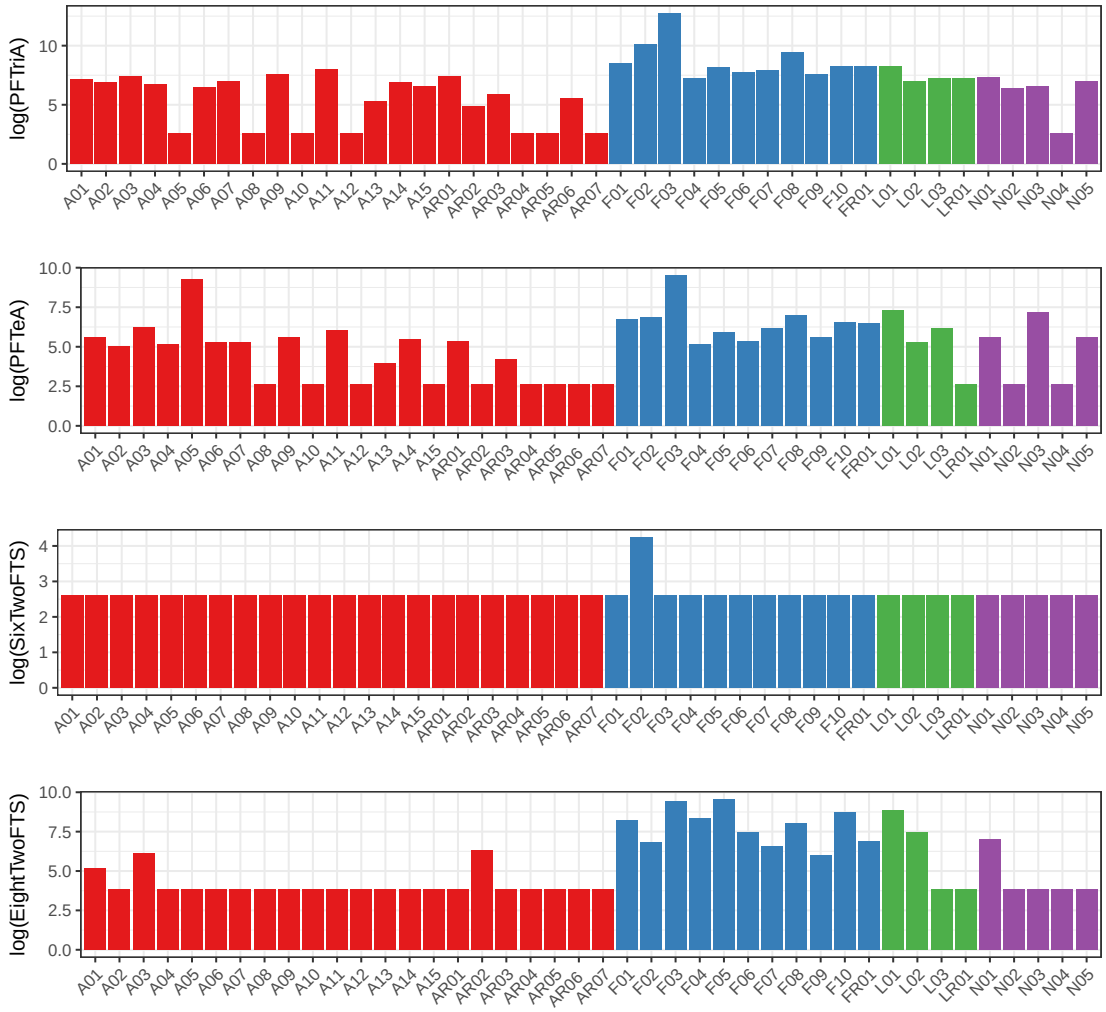


Figure A.8: Histogram showing the concentrations (logtransformed, ng/g ww) of the individual per- and polyfluorinated alkyl substances (PFASs) (PFASs; compound 19-22 out of the 22) analysed for in eggs from snow buntings (*Plectrophenax nivalis*). Samples were collected in Longyearbyen Airport (F (blue); n=11), Longyearbyen (L (green); n=4) and Adventdalen (A (red); n=23) (Longyearbyen) and in Ny-Ålesund (N (purple); n=5), at Svalbard in 2016. Values are log-transformed and non-detects included with LOD/2 (limit of detection).

A.5 Biological Variables

Table A.3: Individual biometric measurements of egg height (mm), width (mm) and volume (ml) as well as eggshell thickness (mm), when the first egg was laid, clutch size, number of hatchlings, fledglings observed for each nest along with if the egg was non-viable or not in eggs of snow buntings (Plectrophenax nivalis). Samples were collected in Longyearbyen and Ny-Ålesund (N; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (F; n=11), Longyearbyen (L; n=4) and Adventdalen (A; n=23). Samples with the additional letter R is indicative of a non-viable egg sampled.

Sample	Height (mm)	Width (mm)	Volume (ml)	Eggshell Thickness	First egg laid	Clutch size	Hatchlings	Fledglings
F01	23.11	15.90	2.98	0.110	June 9	5	4	0
F02	21.12	16.28	2.86	0.114	June 15	6	1	0
F03	23.73	16.18	3.17	0.115		5	4	0
F04	22.94	15.93	2.97	0.110	June 10	6	4	0
F05	23.25	16.59	3.26	0.115	May 28	6	4	0
F06	22.90	15.57	2.83	0.114	May 27	7	6	0
F07	22.18	16.19	2.97	0.115	May 28	6	0	0
F08	22.91	17.01	3.38	0.115	June 4	5	5	0
F09	21.29	16.61	3.00	0.115	June 7	6	0	0
F10	22.96	16.99	3.38	0.115	May 27	6		0
FR01	22.42	16.26	3.02	0.115		7	4	
L01	22.03	16.41	3.03	0.110		6		0
L02	21.51	15.55	2.65	0.110				
L03	22.90	16.23	3.08	0.115	June 3	6		0
LR01	20.84	16.73	2.97	0.115		1		
A01	20.89	16.35	2.85	0.112	June 8	6	5	4
A02	20.84	17.02	3.08	0.115	June 3	4		
A03	23.71	15.48	2.90	0.112	May 31	6	5	4
A04	21.62	16.84	3.13	0.115	June 6	5		
A05	23.42	15.51	2.87	0.110		5		
A06	22.83	16.71	3.25	0.115	May 31	5	4	3
A07	22.14	17.19	3.34	0.110	July 6	5		
⋮								⋮

A08	22.37	16.57	3.13	0.115	June 6	6	4	4
A09	22.45	15.78	2.85	0.115	June 3	6	5	4
A10	24.11	16.11	3.19	0.115	May 30	5	4	3
A11	23.02	16.23	3.09	0.114	June 3	6	5	5
A12	21.90	16.38	3.00	0.115	May 29	6	5	4
A13	20.43	15.91	2.64	0.115	June 4	4	3	3
A14	22.49	16.68	3.19	0.115	June 3	6	5	5
A15	21.99	17.18	3.31	0.115	June 4	4	3	3
AR01	20.48	16.27	2.76	0.110	May 28	4		
AR02	23.22	16.61	3.27	0.115	May 27	5	4	3
AR03				0.115	June 1			
AR04	22.09	17.08	3.29	0.114	May 23	6	5	1
AR05	23.16	16.62	3.26	0.115	May 24	7	6	3
AR06	23.84	16.29	3.23	0.115				
AR07	20.95	15.63	2.61		May 22	7	6	4
N01	22.66	16.70	3.22	0.110	June 9	5		0
N02	22.18	15.63	2.76	0.110	June 12	4		0
N03	20.90	16.73	2.98	0.115	June 11	6		
N04	22.44	15.80	2.86	0.114				0
N05	22.37	15.95	2.90	0.115	June 13	3		0

A.6 PFAS Detection

Table A.4: Table showing the detection percentage (%) of each per- and polyfluorinated alyklated substance (PFASs) analysed for in eggs from snow buntings (*Plectrophenax nivalis*) at each location, in addtition to the overall detection for all the samples. Boldface for samples with detection percentage $\geq 80\%$. Samples were collected in Longyearbyen and Ny-Ålesund (NÅ; n=5) (Svalbard) in 2016. Longyearbyen consist of three sub-locations; Longyearbyen airport (AIR; n=11), Longyearbyen (LYB; n=4) and Adventdalen (ADV; n=23). Overall there were collected a total of 43 samples.

Group	PFAS	Compound detection (%)				
		AIR	LYB	ADV	NÅ	Overall
PFCA	PFBA	8	25	0	0	5
	PFPA	8	0	0	0	2
	PFHxA	46	25	35	60	40
	PFHpA	8	25	0	0	5
	PFOA	**85**	**100**	52	60	65
	PFNA	**100**	**100**	**100**	**80**	**98**
	PFDcA	**100**	**100**	**96**	**80**	**95**
	PFUnA	**100**	**100**	**87**	**80**	**88**
	PFDoA	**92**	75	48	60	63
	PFTriA	**100**	**100**	65	**80**	**81**
	PFTeA	**100**	75	57	60	70
PFSA	PFOSA	8	**100**	17	20	23
	PFBS	8	0	0	0	2
	PFPS	15	0	0	0	5
	PFHxS	**92**	75	26	20	47
	PFHpS	54	0	0	20	16
	brPFOS	**100**	75	17	40	49
	PFOS	**100**	**100**	**87**	**100**	**93**
	PFNS	23	0	0	20	7
	PFDcS	23	0	0	20	7
FTS	6:2 FTS	8	0	0	0	2
	8:2 FTS	**100**	50	13	20	40

A.7 PFAS Concentrations

Table A.5: Summary of the perfluoroalkyl and polyfluoroalkyl substance (PFASs) concentrations (ng/g w.w.) found in eggs sampled from snow buntings (*Plectrophenax nivalis*) in Longyearbyen, Svalbard and Ny-Ålesund, Svalbard in 2016. A total of 43 samples were collected. The PFASs analysed for were divided into perfluororalkyl carboxylic acids (PFCAs), perfluoro sulfonic acids (PFSAs) and fluorotelomer sulfonates (FTSs). Data is summarized as mean and standard deviation (SD), median and range, in addition to portraying number of samples (n) where the detection of the compounds was above limit of detection (LOD).

Group	PFAS	Mean ± SD	Median	Range	n > LOD
PFCA	PFBA	2.02 ± 1.47	2.02	0.56-3.49	2
	PFPA	0.29 ± 0	0.29	0.29	1
	PFHxA	0.11 ± 0.17	0.06	0.02-0.76	17
	PFHpA	0.31 ± 0.19	0.31	0.13-0.50	2
	PFOA	0.59 ± 2.34	0.06	0.02-12.62	28
	PFNA	3.63 ± 17.14	0.51	0.14-113.11	42
	PFDcA	1.84 ± 7.10	0.51	0.06-46.48	41
	PFUnA	14.90 ± 73.55	1.92	0.24-461.90	38
	PFDoA	4.27 ± 15.83	0.71	0.27-84.84	27
	PFTriA	12.71 ± 57.74	1.40	0.13-343.43	34
	PFTeA	1.22 ± 2.94	0.28	0.05-13.74	30
PFSA	PFBS	2.46 ± 0	2.46	2.46	1
	PFPS	1.90 ± 1.77	1.90	0.12-3.67	2
	PFHxS	7.51 ± 28.05	0.32	0.06-126.38	19
	PFHpS	3.20 ± 7.27	0.23	0.08-21.01	7
	PFOSA	1.76 ± 4.25	0.32	0.09-14.49	10
	linPFOS	75.88 ± 395.05	2.37	0.29-2539.57	40
	brPFOS	39.05 ± 150.32	2.82	0.15-693.96	20
	PFNS	10.02 ± 14.05	0.10	0.08-29.89	3
	PFDcS	15.42 ± 21.20	0.69	0.17-45.39	3
FTS	6:2FTS	0.13 ± 0	0.13	0.13	1
	8:2 FTS	3.27 ± 4.02	1.72	0.14-14.27	17