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# Perfluoroalkyl substances (PFASs) in white whales (*Delphinapterus leucas*) from Svalbard – A comparison of concentrations in plasma sampled 15 years apart☆

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ABSTRACT

The objective of the present study was to investigate recent concentrations of perfluoroalkyl substances (PFASs) in white whales (*Delphinapterus leucas*) from Svalbard and compare them to concentrations found in white whales sampled from that same area 15 years ago. Plasma collected from live-captured white whales from two time periods (2013–2014, n = 9, and 1996–2001, n = 11) were analysed for 19 different PFASs. The 11 PFASs detected included seven $C_8$–$C_{14}$ perfluoroalkyl carboxylates (PFCAs) and three $C_6$–$C_8$ perfluoroalkyl sulfonates (PFSAs) as well as perfluorooctane sulfonamide (FOSA). Recent plasma concentrations (2013–2014) of the dominant PFAS in white whales, perfluorooctane sulfonate (PFOS; geometric mean = 22.8 ng/mL), was close to an order of magnitude lower than reported in polar bears (*Ursus maritimus*) from Svalbard. PFOS concentrations in white whales were about half the concentrations in harbour (*Phoca vitulina*) and ringed (*Pusa hispida*) seals, similar to hooded seals (*Cystophora cristata*) and higher than in walruses (*Odobenus rosmarus*) from that same area. From 1996 to 2001 to 2013–2014, plasma concentrations of PFOS decreased by 44%, whereas four $C_{9\text{-}12}$ PFCAs and total PFCAs increased by 35–141%. These results follow a similar trend to what has been reported in other studies of Arctic marine mammals from Svalbard. The most dramatic change has been the decline of PFOS concentrations since 2000, corresponding to the production phase-out of PFOS and related compounds in many countries around the year 2000 and a global restriction on these substances in 2009. Still, the continued dominance of PFOS in white whales, and increasing concentration trends for several PFCAs, even though exposure is relatively low, calls for continued monitoring of concentrations of both PFCAs and PFSAs and investigation of biological effects.



## 1. Introduction

Perfluoroalkyl substances (PFASs), in particular perfluoroalkyl sulfonates (PFSAs) and perfluoroalkyl carboxylates (PFCAs), are persistent, long-range transported contaminants found in relatively high concentrations in human and wildlife populations worldwide, including in Arctic marine mammals (Armitage et al., 2009; Butt et al., 2010; Dreyer et al., 2009; Haug et al., 2018; Muir et al., 2019). These chemicals have been produced and used in large volumes since the 1950s and are still widely used in consumer products and for industrial purposes (OECD, 2011; Wang et al., 2017; Xie et al., 2013). PFASs have for example been used in electric and electronic devices, fire-fighting foam, photo imaging, hydraulic fluids, metal plating and textiles (Key et al., 1997; Prevedouros et al., 2006).

The emission history of PFCAs and PFSAs is complicated (Wang et al., 2014a; Wang et al., 2017). In 2001–2002, a major producer (3M) in the USA started production phase-out of perfluorooctane sulfonate (PFOS) and related compounds (3M 2000; Paul et al., 2009). In 2009, PFOS, its salts and perfluorooctane sulfonyl

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fluoride (PFOSF) were listed in Annex B (i.e. restricted, not completely banned) of the Stockholm Convention on Persistent Organic Pollutants (Stockholm Convention, 2009). For PFCAs, there was high production in Western countries and Japan until 2002 (Wang et al., 2014a). After this time, perfluorooctanoate (PFOA) and other PFCAs with ≥8 carbon chain-lengths, as well as perfluorohexane sulfonate (PFHxS), have been gradually phased-out of production in these countries. At the same time, there has been an increase in production and emission of PFCAs and some precursors from China and other emerging economies in Asia (Wang et al., 2014a). PFOA and PFHxS are now being considered for restriction under the Stockholm convention (Stockholm Convention, 2015). PFOA will be banned under chemical regulations in the European Union in 2020 and $C_{9\text{-}14}$ PFCAs and PFHxS are currently under consideration (www.echa.europa.eu).

PFASs have been emitted into the environment mainly from North-America, Europe and Asia (Wang et al., 2017; Wang et al., 2014a). PFASs emitted into aquatic environments can reach the Arctic by slow oceanic transport (Zhao et al., 2012). Formation of sea spray aerosols charged with PFASs can speed up their transport northwards (Johansson et al., 2019). PFASs are also transported to the Arctic via atmospheric transport, with contributions from melting glaciers and snowpacks (Xie et al., 2015; Zhao et al., 2012). Degradation of volatile precursors during atmospheric transport or after deposition may also be a source of both PFSAs and PFCAs in remote areas (D'Eon et al., 2006; Ellis et al., 2004; Martin et al., 2006; Taniyasu et al., 2013). However, the exact transport pathways to the Arctic for various PFASs or their precursors are not yet fully understood, especially regional differences in the relative importance of oceanic versus atmospheric input.

As with other organohalogen contaminants (OHCs; e.g. polychlorinated biphenyls, PCBs, and organochlorine pesticides), several PFSAs and PFCAs have long half-lives and they bioaccumulate in organisms leading to biomagnification in Arctic marine food webs (i.e. with increasing trophic level), resulting in higher concentrations in upper trophic animals, such as marine mammals (Kelly et al., 2009; Martin et al., 2004; Muir et al., 2019; Tomy et al., 2009). Internal biotransformation processes in organisms, including in marine mammals, convert some precursors into more persistent compounds (e.g. perfluorooctane sulfonamide, FOSA, or other precursors to PFOS) although these processes vary among species (Galatius et al., 2013; Letcher et al., 2014).

Contrary to other OHCs that accumulate predominantly in lipid-rich tissues, PFASs are mainly found in protein-rich tissues, such as liver and blood, due to their high affinity for proteins (Ahrens et al., 2009; Bischel et al., 2010; Jones et al., 2003). PFASs are generally found at higher concentrations than lipid-soluble OHCs in plasma of Arctic marine top predators (Grønnestad et al., 2018; Nøst et al., 2012; Routti et al., 2019a). The presence of PFASs in Arctic wildlife is of concern due to their toxicity. Both experimental animal and human epidemiological studies have associated PFAS exposure, in particular PFOS and PFOA, with adverse outcomes such as endocrine disruption (thyroid and sex hormones), disruption of lipid homeostasis, immunotoxicity, liver toxicity, and developmental toxicity (e.g. lowered pre- and postnatal growth, and neurobehavioral effects) (Blum et al., 2015; Lau et al., 2007; Liew et al., 2018; Rappazzo et al., 2017; Sunderland et al., 2019).

Time-trend analyses of PFAS concentrations in Arctic biota are important in order to assess the environmental impact of regulatory-based changes in production rates of PFASs and their precursors. Also, ongoing climate change in the Arctic could potentially alter transport pathways and predator-prey interactions, and thus contribute to time-related changes in PFAS exposure in wildlife species in the Arctic (McKinney et al., 2015; Muir et al., 2019; Routti et al., 2017; Smythe et al., 2018). Although many studies have investigated temporal trends of concentrations of these chemicals in Arctic biota, including white whales (*Delphinapterus leucas*) and other marine mammals, these studies have mainly been performed in Greenland and in the North-American Arctic (AMAP. AMAP Assessment, 2015; Butt et al., 2010; Muir et al., 2019). Additionally, few studies have taken place in the past decade (AMAP. AMAP Assessment, 2015; Muir et al., 2019). Recent studies on polar bears (*Ursus maritimus*), Arctic foxes (*Vulpes lagopus*) and ringed seals (*Pusa hispida*) from Svalbard show a decline in PFOS concentrations since about 2000, whereas PFCA trends show more variability related to chain length, as well as species or area investigated (Routti et al., 2017; Routti et al., 2016).

The white whale is an odontocete whale with a circumpolar Arctic distribution (O'Corry-Crowe et al., 2009). In the Svalbard Archipelago, it is the most commonly observed cetacean and satellite telemetry shows that they reside year-round in this area (Lydersen et al., 2001; Vacquié-Garcia et al., 2018). White whales feed high in the marine food chain (mainly fish), have a long lifespan, and seem to have little capacity to metabolize and excrete contaminants, which makes them susceptible to accumulation of OHCs (AMAP, 1998; Letcher et al., 2014; McKinney et al., 2004; Wolkers et al., 2004). Thus, it is an important species for monitoring long-range transported OHCs that are biomagnified in Arctic marine food webs. Previous studies report relatively high concentrations of chlorinated and brominated OHCs in white whales from Svalbard compared to other marine mammals, such as polar bears and seals, in this area (Andersen et al., 2006; Andersen et al., 2001; Villanger et al., 2011; Wolkers et al., 2006; Wolkers et al., 2004). While studies of white whale populations in Canada and Alaska have shown the presence of PFASs in liver and blood at relatively high concentrations (Kelly et al., 2009; Muir et al., 2013; Reiner et al., 2011; Smythe et al., 2018; Tomy et al., 2009; Tomy et al., 2004), no studies have yet reported concentrations of PFASs in white whales from Svalbard.

The objective of the present study was to investigate recent concentrations of PFASs in white whales from Svalbard (2013–2014) and compare them to concentrations found in white whales sampled 15 years ago (1996–2001).

## 2. Materials and methods

### 2.1. Data collection

Plasma was collected from live-captured white whales from two time periods in Svalbard, Norway (Fig. 1): Recent: August 2013 and 2014 (n = 9; 4 adult females and 5 adult males); Past: July to October, 1996–2001 (n = 11; 4 adult males, 1 adult female, 4 subadult males, and 2 subadult females). Capture and sampling procedures are described in detail elsewhere (Villanger et al., 2011). Briefly, the whales were restrained and handled for up to 30 min, while being measured, instrumented and sampled for biological studies, prior to release. All handling was done under permits from the Norwegian Animal Research Authority and the Governor of Svalbard. Standard length and girth (girth behind the flippers) were measured. Age group (adult or subadult) and sex were determined by examination of genitalia, body size and skin colour (Brodie, 1971). Sex was confirmed by DNA analyses at Bioforsk Svanhovd (Svanvik, Norway). About 100 mL of blood was sampled from the caudal vein of each whale using heparinized blood collection tubes and spun (4000 *g* for 10 min) to collect plasma. Plasma samples were stored at −20 °C until analysis.

### 2.2. Chemical analysis of perfluoroalkyl substances (PFASs)

Nineteen PFASs were analysed in white whale plasma using high

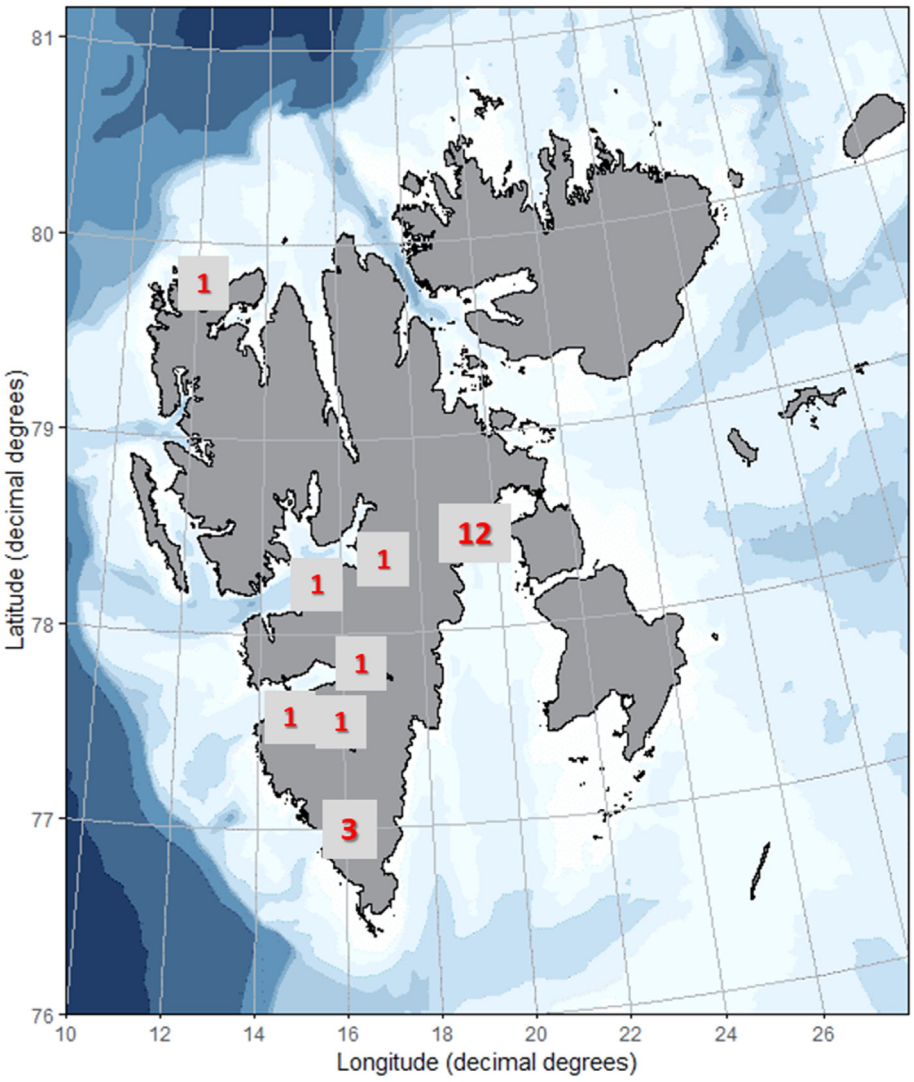


**Fig. 1.** Sampling locations of white whales in the coastal waters of Svalbard, Norway. Number of animals sampled in each location is given.

performance liquid chromatography/tandem mass spectrometry (HPLC-MS/MS) at the Department of Environmental Exposure and Epidemiology, Norwegian Institute of Public Health, in Oslo, Norway: perfluorobutanoate (PFBA); perfluoropentanoate (PFPeA); perfluorohexanoate (PFHxA); perfluoroheptanoate (PFHpA); perfluorooctanoate (PFOA); perfluorononanoate (PFNA); perfluorodecanoate (PFDA); perfluoroundecanoate (PFUnDA); perfluorododecanoate (PFDoDA); perfluorotridecanoate (PFTrDA); perfluorotetradecanoat (PFTeDA); perfluorobutane sulfonate (PFBS); perfluorohexane sulfonate (PFHxS); perfluoroheptane sulfonate (PFHpS); perfluorooctane sulfonate (PFOS); perfluorodecane sulfonate (PFDS); perfluorooctane sulfonamide (FOSA); N-methyl-perfluorooctane sulfonamide (MeFOSA), and N-ethyl-perfluorooctane sulfonamide (EtFOSA) (Table S1). For quantification the following $^{13}C$ and deuterium-labeled compounds were used as internal standards: MPFBA, MPFHxA, MPFOA, MPFNA, MPFDA, MPFDoDA, MPFHxS, MPFOS, $D_3$-N-MeFOSA and $D_3$-N-EtFOSA. All PFASs as well as their internal standards were purchased from Wellington Laboratories (Guelph, Ontario, Canada). Methanol, acetonitrile and water (J.T. Baker LC-MS grade) were obtained from Avantor Performance Materials (Gliwice, Poland). 2-propanol (HPLC grade) was purchased from Alfa Aesar (Karlsruhe, Germany). Ammonium hydroxide (puriss PA grade) was from Fluka (Steinheim, Germany). Formic acid (GPR Rectapur grade) was obtained from VWR BDH Prolabo (Fontenay-sous-Bois, France) and acetic acid (PA grade) was purchased from Merck (Darmstadt, Germany). Argon of purity 99.9999% was obtained from AGA (Oslo, Norway) and nitrogen gas was supplied by a Maxigas Nitrogen generator from Dominick Hunter Ltd. (Gateshead, UK). All glassware was washed in 2.5% Extran MA 01 from Merck (Darmstadt, Germany), rinsed with distilled water and subsequently heated at 450 °C for 4 h (volumetric equipment was not heated).

### 2.3. Extraction procedure and analyses using HPLC-MS/MS

The procedures, as described in detail elsewhere (Haug et al., 2009), were followed with some minor modifications. Briefly, internal standards were added to 150 μL plasma and then protein precipitation was performed by the addition of methanol. After centrifugation, the supernatant was transferred to a glass vial and 0.1M formic acid was added. Calibration solutions were prepared in the same way, where calf (*Bos taurus*) plasma was used as a matrix with analytes, internal standards, methanol and 0.1 M formic acid added. The sample extract was then injected onto the on-line

column switching HPLC-MS/MS system. The column switching LC system consisted of a HTC PAL autosampler (CTC analysis AG, Zwingen, Switzerland) with one six port and one ten port switching valve attached (CTC), and two Surveyor LC pumps (Thermo Finnigan, San Jose, CA, USA). To eliminate possible contamination of PFASs from Teflon parts in the LC pumps, a Hypercarb guard column (10 mm × 4 mm x 5 μm particles) from Thermo Scientific (San Jose, CA, USA) was installed between each of the LC pumps and the switching valves. A Betasil C8 column (10 mm × 4 mm x 5 μm particles) from Thermo Scientific was used as on-line SPE column and a Betasil C8 (50 mm × 2.1 mm x 3 μm particles) from Thermo Scientific as analytical column. The mass spectrometric detector was a TSQ Quantum (Thermo Finnigan) triple quadrupole MS.

The samples, controls and calibration solutions were analysed and quantification of the white whale samples were done within the linear range of the ten-point calibration curve. PFAS concentrations are given as ng/mL plasma. The limit of detection (LOD) and limit of quantification (LOQ) of the method have previously been estimated based on the signal/noise ratio of three and ten, respectively, from four calibration solutions run in triplicate (Haug et al., 2009; Table S1). LOQ was 0.05 ng/mL except for PFBA (LOQ = 0.1 ng/mL), and for PFTeDA and PFDS (LOQs = 0.20 ng/mL) (Haug et al., 2009; Table S1).

### 2.4. Quality control and assurance

This analytical method has been thoroughly validated and shown to be a sensitive and reliable method that requires only small amounts of plasma or serum (Haug et al., 2009). The method has been used for determination of PFASs in large-scale studies on human serum/plasma samples (Halldorsson et al., 2012; Haug et al., 2018; Starling et al., 2014; Whitworth et al., 2012), and the method has also been used for determination of PFASs in serum from mice (*Mus musculus*) (Ngo et al., 2014). The laboratory used in this study has standard procedures for quality assurance (blanks, controls, quantification within linear range of the calibration curves), ensuring that the precision, linearity, and sensitivity of the analyses are within the original parameters described in the method validation (Haug et al., 2009). Three internal quality control samples were analysed in the same run as the white whale plasma samples, and the results were in accordance with the laboratory's previous measurements of this control sample. The laboratory has also participated in an inter-laboratory comparison on PFASs around the time when the white whale samples were analysed, with satisfactory results (z-score < 2) (AMAP, 2015).

### 2.5. Data analyses

Statistical analyses were conducted using SPSS (Version 23, standard version, SPSS Inc., Chicago, IL, USA). Compounds below LOD for >25% of the whale samples were noted as not detected and excluded from further statistical analysis (PFBA, PFPeA, PFHxA, PFHpA, PFBS, PFDS, MeFOSA, and EtFOSA; Table S1). Missing values (i.e. < LOD) were assigned a random number between LOD and zero for PFOA (n = 5 of 20) and PFTeDA (n = 4 of 20). Group sums of PFAS concentrations were calculated; sum (Σ) of all 11 detected PFASs ($\Sigma_{11}$PFAS: PFOA, PFNA, PFDA, PFUnDA, PFDoDa, PFTrDa, PFTeDa, PFHxS, PFHpS, PFOS and FOSA), sum of seven carboxylates ($\Sigma_7$PFCA: PFOA, PFNA, PFDA, PFUnDA, PFDoDa, PFTrDa, and PFTeDa) and sum of three sulfonates ($\Sigma_3$PFSAs: PFHxS, PFHpS, and PFOS).

Numerical values of PFAS concentrations are presented as geometric mean in tables and text, unless otherwise mentioned. Shapiro-Wilks tests ($p < 0.05$) and evaluation of skewness values, and histograms showed that standard length and most PFAS variables were approximately normally distributed. PFOS, FOSA, PFOA and $\Sigma_3$PFSAs were natural log (ln)-transformed to approximate normal distribution. Pearson correlation was used to investigate correlation patterns among individual PFASs ($p < 0.05$).

Due to small samples sizes and few individuals within each sex and age group in the two sampling periods (Table 1), it was not statistically feasible to test for differences in PFAS concentrations by age or sex of the animals. Hence, all whales were pooled into their respective groups by years of sampling (1996–2001 or 2013–2014) to investigate temporal trends in PFAS concentrations over this 15-year time period. Linear models were used to investigate differences in PFAS concentrations in time clusters of white whales, using sampling period (1996–2001 and 2013–2014) as a categorical variable. The estimate with 95% confidence interval (CI) was used to calculate the relative change (%) with corresponding CIs between the two sampling periods. For ln-transformed PFASs variables (PFOA, PFOS, FOSA, and $\Sigma_3$PFSAs), relative change and 95% CI were calculated using the formula: $100*(e^{estimate} - 1)$. For the remaining PFAS variables, relative change with 95% CI were estimated using the following equation: estimate/intercept*100. The estimates (and the calculated relative change) are statistically significantly different from zero at the 5% confidence level if the corresponding 95% CIs do not cross zero.

## 3. Results

### 3.1. PFAS concentrations and patterns

Among the 11 detected PFASs, PFOS had the highest concentrations in white whale plasma from both time periods (Tables 1 and S2). When investigating the contribution of individual compounds to $\Sigma_{11}$PFAS in whales sampled in 1996–2001 and 2013–2014, PFOS was the predominant compound (72 and 56% of total PFASs, respectively), followed by PFUnDA (9 and 17%), FOSA (6 and 7%), PFNA (1 and 4%), and PFHxS (1 and 4%) while the contribution of PFDA, PFHpS, PFOA and PFTeDA were even more minor (Fig. 2). Four short-chained ($C_{4-7}$) PFCAs (PFBA, PFPeA, PFHxA, PFHpA) were not detected (LODs = 0.003–0.009 ng/mL; Table S1) in white whale plasma, while seven ($C_{8-14}$) PFCAs were present in all or most of the white whales examined (Tables 1 and S1). Out of five analysed PFSAs, PFBS ($C_4$) and PFDS ($C_{10}$) were not detected in any samples (LODs = 0.009 and 0.05 ng/mL, respectively), while $C_{6-8}$ PFSAs were present in all whales (Tables 1 and S1). FOSA was detected in all samples, while MeFOSA and EtFOSA (LODs = 0.009 ng/mL) were not detected in any of the sampled white whales (Tables 1 and S1).

The correlation among individual PFASs per sampling period showed that PFDA, PFUnDA and PFDoDA were strongly, positively correlated in both sampling periods ($r > 0.94$; Table S3), and these compounds were also positively correlated with PFOS ($r > 0.78$; Table S3). FOSA showed a moderate correlation with PFOS in both time periods, as well as with PFDA, PFUnDA and PFDoDA in the 2013–2014 group but not in the 1996–2001 group of white whales (Table S3).

### 3.2. Temporal trends

There were significant changes in plasma concentrations for all individual and summed PFASs between the two sampling periods, except for PFHxS and PFOA (Table 1). Plasma concentrations of PFOS and FOSA decreased by 44% and 27%, respectively, and PFTrDA and PFTeDA decreased by 30% and 46%, respectively, whereas $C_{9-12}$ PFCAs (PFNA, PFDA, PFUnDA, PFDoDA) and $\Sigma_7$PFCAs as well as the FOSA:PFOS ratio increased by 29–141% from 1996 to 2001 to 2013–2014 (Table 1). Confidence intervals for relative change in concentrations of PFTrDA (95% CI -59, −8.2), PFTeDA (95% CI

**Table 1**
Biological information and concentrations (ng/mL) of individual PFASs and group sums (∑) in plasma of white whales from Svalbard, Norway, sampled in two time periods (1996–2001 and 2013–2014). PFAS concentrations are given as geometric mean with 95% confidence intervals (CIs). Percent change with 95% CIs of PFAS concentrations between the two time periods derived from linear models are presented.

| | Past (1996–2001) | Recent (2013–2014) | | |
|---|---|---|---|---|
| n (female: male) | 11 (3:8) | 9 (4:5) | | |
| n (adult: subadult) | 5:6 | 9:0 | | |
| Length (cm) | 373 ± 56 | 412 ± 30 | | |
| **Concentration (ng/mL)** | **Geometric mean (95% CI)** | | **%change**[a] | **(95% CI)** |
| Perfluoroalkyl carboxylates | | | | |
| PFOA | 0.02 (0.01, 0.05) | 0.01 (0.00, 0.02) | −59 | (-86, 29) |
| PFNA | 0.80 (0.64,0.97) | 1.95 (1.68, 2.37) | 141* | (138, 143) |
| PFDA | 1.25 (0.98, 1.54) | 2.40 (2.05, 2.85) | 85* | (64, 98) |
| PFUnDA | 4.86 (3.90, 5.90) | 7.09 (6.07, 8.25) | 42* | (17, 58) |
| PFDoDA | 0.74 (0.62, 0.87) | 1.02 (0.89, 1.18) | 35* | (14, 51) |
| PFTrDA | 1.45 (1.18, 1.74) | 1.03 (0.86, 1.21) | −30* | (-59, −8.2) |
| PFTeDA | 0.09 (0.06, 0.14) | 0.05 (0.03, 0.08) | −46* | (-110, −7.9) |
| $\sum_7$PFCAs | 9.34 (7.69, 11.1) | 13.7 (12.1, 15.8) | 43* | (22, 58) |
| **Perfluoroalkyl sulfonates** | | | | |
| PFHxS | 1.33 (0.96, 1.64) | 1.29 (1.04, 1.55) | −3.1 | (-42, 23) |
| PFHpS | 0.91 (0.73, 1.13) | 0.49 (0.37, 0.59) | −48* | (-82, −22) |
| PFOS | 40.7 (32.0, 50.6) | 22.8 (19.8, 26.5) | −44* | (-58, −26) |
| **$\sum_3$PFSAs** | 42.9 (34.1, 54.0) | 24.6 (21.6, 29.0) | −43* | (-57, −24) |
| Others | | | | |
| FOSA | 3.59 (2.91, 4.49) | 2.61 (2.20, 3.14) | −27* | (-45, −3.7) |
| FOSA:PFOS ratio | 0.10 (0.07, 0.11) | 0.11 (0.09, 0.14) | 29* | (2.53, 47.23) |

Concentrations of PFASs above LOD in >80% of the individuals were used, and missing values (i.e. below LOD) were replaced by a random value between zero and LOD for PFOA (n = 5) and PFTeDA (n = 4).
*Significant change (i.e. 95% CIs do not cross zero).
[a] Estimated % change of PFAS concentrations across the two time periods are based on linear models with time (year of sampling) categorized into time period (1996–2001 and 2013–2014) as a dependent group variable and PFAS concentrations as continuous, independent (response) variables. PFOS, FOSA and PFOA and $\Sigma_3$PFSAs were natural log (ln)-transformed before analyses.

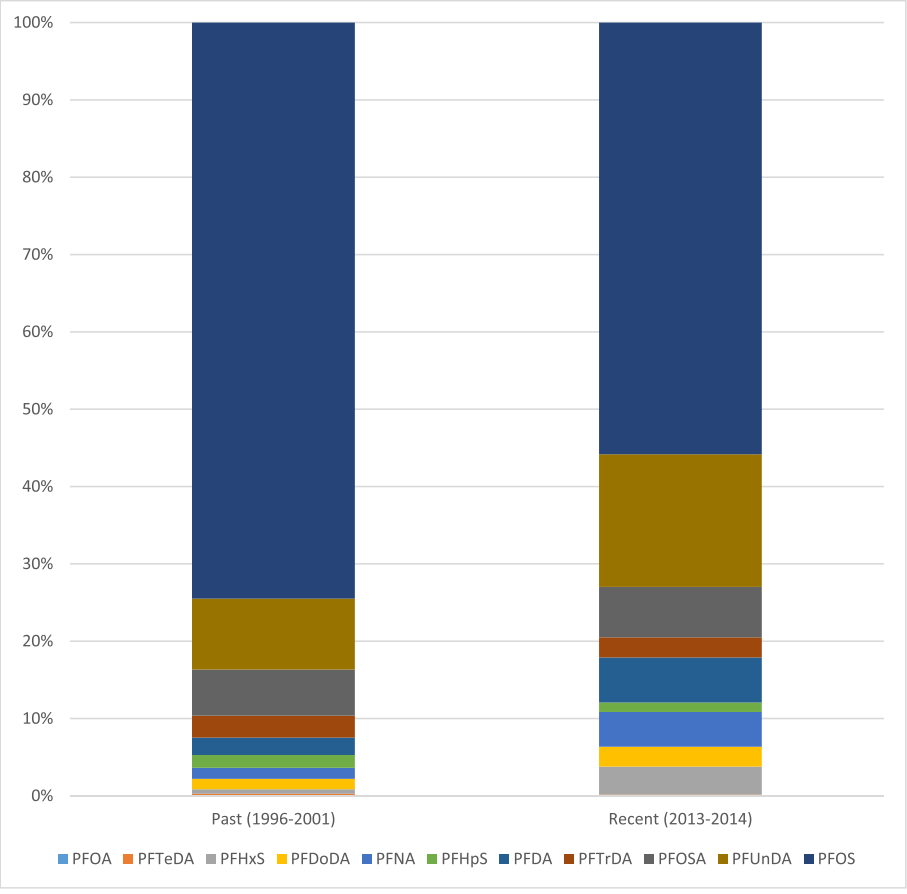


**Fig. 2.** Percent contribution of individual PFASs to the total PFAS load ($\Sigma_{11}$PFAS) in white whales from Svalbard, Norway, sampled in 1996–2001 (n = 11) and 2013–2014 (n = 9).

-110, −7.9), FOSA (95% CI -45, −3.7) and FOSA:PFOS ratio (95% CI 2.53, 47.23) were closer to zero compared to the other compounds (Table 1).

## 4. Discussion

### 4.1. PFAS concentrations and patterns

Similar to other studies of wildlife and humans (Haug et al., 2018; Houde et al., 2011; Muir et al., 2019; Nøst et al., 2012; Sunderland et al., 2019), PFOS was the dominant PFAS in plasma of white whales from Svalbard sampled in 1996–2001 and 2013–2014. PFOS concentrations in the recent time period 2013–2014 (=22.8 ng/mL ~ 22.8 ng/g) were close to an order of magnitude lower than what is reported in plasma of polar bears from Svalbard (Bytingsvik et al., 2012; Tartu et al., 2017a), and about half the concentrations reported for plasma in ringed seals and harbour seals (*Phoca vitulina*) from this area (Routti et al., 2016; Routti et al., 2014), but in the same range as hooded seal (*Cystophora cristata*) mothers and pups from the West-Ice, East of Greenland (Grønnestad et al., 2017), and about ten times higher than in plasma from adult male walruses (*Odobenus rosmarus*) from Svalbard sampled in 2013–2014 (Scotter et al., 2019) (Table 2).

PFOA, which is usually the second most dominant PFAS in human blood (Haug et al., 2018; Sunderland et al., 2019), was just barely above detection limit (LOD = 0.006 ng/mL) in white whales from both time periods (Table 1; Table S1). This is in accordance with the low exposure and/or bioaccumulation of this compound reported in Arctic marine food webs (Kelly et al., 2009; Martin et al., 2004; Routti et al., 2017; Tomy et al., 2009). Short-chain PFASs were not detected in plasma of white whales from Svalbard. This is in agreement with generally low concentrations of these compounds reported in Arctic mammals and birds (Routti et al., 2017; Scotter et al., 2019; Tartu et al., 2014), which may be related to their low bioaccumulation potency (Brendel et al., 2018).

White whales in Svalbard had several orders of magnitude higher FOSA concentrations compared to other marine mammal species from Svalbard (Bytingsvik et al., 2012; Routti et al., 2017; Routti et al. 2016; Routti et al. 2014; Scotter et al., 2019) (Table 2). A similar pattern of elevated FOSA in white whales, compared to other non-cetacean marine mammals, has been reported in studies from the Canadian Arctic and Greenland (Gebbink et al., 2016; Kelly et al., 2009; Martin et al., 2004; Tomy et al., 2009). Most studies of white whales report that FOSA is the predominant PFAS in liver, exceeding PFOS concentrations (Kelly et al., 2009; Reiner et al., 2011; Smythe et al., 2018; Tomy et al., 2004; Tomy et al., 2009) (Table S4). Even though FOSA has been shown to biomagnify in Arctic marine food webs (Kelly et al., 2009), it is metabolized to PFOS in most upper trophic mammals. However, cetaceans, including white whales, appear to lack the ability for biotransformation of FOSA into PFOS (Galatius et al., 2013; Gebbink et al., 2016). Thus, white whales and other cetaceans are exposed to PFOS mainly through direct bioaccumulation from prey species (mainly fish), whereas in marine carnivore species (pinnipeds, polar bear) PFOS also originate from internal biotransformation of FOSA and its precursors. Whether FOSA exposure in white whales originates from bioaccumulation or biotransformation is unclear. In vitro depletion of EtFOSA and formation of FOSA was negligible in white whale liver microsomes (Letcher et al., 2014), which suggests dietary origin of FOSA in white whales. FOSA precursors, EtFOSA and MeFOSA, were, however, not detected in plasma of white whales from Svalbard.

Even though white whales from Svalbard sampled in 2013–2014 had lower plasma concentrations of PFSAs compared to polar bears from the same time period (Table 2), this difference was less pronounced for PFCAs. This was especially true for the $C_{9\text{-}12}$ PFCAs, where plasma concentrations in white whales were 20–50% of those found in polar bears (Table 2). Compared to seal species from Svalbard, the white whales showed similar or occasionally somewhat higher concentrations of PFCAs (Table 2). Species-specific dietary preferences (and thus trophic positions) and biotransformation ability, as well as variations in feeding habitats of marine mammals in the Svalbard Archipelago and adjacent waters probably explain differences in PFAS concentrations in white

**Table 2**
Comparison of PFAS concentrations in white whale plasma (ng/mL) from the current study with previously reported PFAS concentrations in plasma (ng/g) of marine mammal species from Svalbard.

| Species (n) | Year | Statistic | PFSAs | | | | FOSA | PFCAs | | | | | | | $\sum_7$PFCAs | $\sum_3$PFSAs |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | PFBS | PFHxS | PFHpS | PFOS | | PFOA | PFNA | PFDA | PFUnDA | PFDoDA | PFTrDA | PFTeDA | | |
| **White whale** adult males + females (n = 9)[a] | 2013-2014 | ME | <0.009 | 1,43 | 0,49 | 22,17 | 2,59 | 0,015 | 1,79 | 2,32 | 6,81 | 1,02 | 1,03 | 0,05 | 13,55 | 39,59 |
| **Polar bear** adult females (n = 112)[b] | 2012–2013 | | | | | | | | | | | | | | | |
| -spring | | ME | n.a. | 27,6 | n.a. | 197 | n.a. | 4,2 | 27,2 | 8,5 | 18,4 | 2,5 | 5,8 | n.a. | 66,6 | 244,4 |
| -autumn | | ME | n.a. | 31,3 | n.a. | 244 | n.a. | 4,4 | 35,3 | 10,8 | 23,6 | 3 | 7 | n.a. | 84,1 | 274,9 |
| **Harbour seal**[c] | 2009–2010 | | | | | | | | | | | | | | | |
| -male (n = 4) | | ME | <0,07 | 1,8 | n.a. | 43 | <0,05 | 0,8 | 4,3 | 3,9 | 10 | 1,3 | 2,9 | 0,24 | 24 | 45 |
| -female (n = 4) | | ME | <0,07 | 1,4 | n.a. | 35 | <0,05 | 0,58 | 4,0 | 3,1 | 8,3 | 1,1 | 2,2 | 0,14 | 20 | 37 |
| -juvenile (n = 4) | | ME | <0,07 | 2,3 | n.a. | 38 | <0,05 | 0,59 | 4,5 | 3,7 | 9,4 | 1,1 | 2,2 | 0,12 | 22 | 41 |
| **Ringed seal** (n = 11 females)[d] | 2010 | AM | <0,07 | 1,29 | n.a. | 48 | <0,0003 | 0,22 | 8,0 | 5,2 | 9,2 | 1,16 | 2,24 | <0,07 | 25,8 | 49 |
| **Hooded seal** [e] | 2008 | ME | n.a. | 0,696 | n.a. | 12,2 | n.a. | 0,278 | 1,99 | 2,30 | 8,51 | 1,41 | 4,72 | n.a. | 19,21 | 12,9 |
| -mothers (n = 15) | | | | | | | | | | | | | | | | |
| -pups (n = 15) | 2008 | ME | n.a. | 2,60 | n.a. | 28,3 | n.a. | 0,336 | 1,33 | 1,46 | 11,1 | 3,37 | 12,6 | n.a. | 30,20 | 30,9 |
| **Walrus** adult males (n = 39)[f] | 2014–2015 | ME | <0,05 | 1.35 | <0,10 | 2,22 | <0,15 | 0,288 | 1,35 | 0,227 | 0,269 | <0,10 | <0,10 | <0,10 | 7,534 | 3,57 |

ME = median. AM = arithmetic mean. n.a. = not analysed.
[a] Current study.
[b] Tartu et al. (2017a).
[c] Routti et al. (2014).
[d] Routti et al. (2016).
[e] Grønnestad et al. (2017).
[f] Scotter et al. (2019).

whales compared to other marine mammal species in this area. Still, the concentrations and patterns of PFASs in white whales from Svalbard were consistent with biomagnification of (in particular) PFOS and $C_8-_{14}$PFCAs in Arctic marine food webs (Kelly et al., 2009).

Studies of other white whale populations in Canada and Alaska have mainly measured PFAS concentrations in liver samples (Kelly et al., 2009; Muir et al., 2013; Reiner et al., 2011; Smythe et al., 2018; Tomy et al., 2004; Tomy et al., 2009) (Table S4), which are not directly comparable to the PFAS results from plasma of white whales in the present study. However, Kelly et al. (2009) also measured PFASs in whole blood from white whales captured in 1999–2003 in the Eastern Hudson Bay (Canada). By using conversion factors described in Jin et al. (2016) with a mean hematocrit of 0.53 for white whales (Norman et al., 2012), plasma-equivalent concentrations of the reported whole blood concentrations in Kelly et al. (2009) were calculated in order to compare with the results herein (Table S4). ∑PFCAs concentrations in white whales from Svalbard sampled in 1996–2001 were generally similar to the plasma equivalent ∑PFCA concentrations in white whales from Hudson Bay sampled in 1999–2003 (Table S4). However, PFOS concentrations in white whales from Svalbard were about two times higher while FOSA was about 40% of the plasma equivalent concentrations in white whales from Hudson Bay (Kelly et al., 2009). Analyses of PFASs in blood samples from different white whale populations would enable a more comprehensive investigation of spatial trends across the species' distribution in the Arctic.

### 4.2. Temporal trends

#### 4.2.1. PFSA and FOSA concentrations

Plasma concentrations of PFOS in white whales from Svalbard decreased by 44% (95% CI -58, −26) between the two time periods, 1996–2001 and 2013–2014. This coincides with a decrease in plasma PFOS concentrations in polar bears and Arctic foxes from Svalbard sampled between 1997 and 2014 (Routti et al., 2017). For both polar bears and Arctic foxes, PFOS concentrations peaked around 2003 and then declined 9–14% annually until levelling off around 2009–2010 (Routti et al., 2017). PFOS concentrations in ringed seals from Svalbard also decreased by 11–12% per year from 2004 to 2010 (Routti et al., 2016). PFHxS showed a similar decreasing trend in polar bears, ringed seals and Arctic foxes (Routti et al., 2017; Routti et al., 2016), contrary to the white whales where there was no time trend.

It appears that white whales follow the expected temporal trend observed in human and many wildlife studies, including in marine mammals from Svalbard and other Arctic areas, with a decrease in PFOS since the start of 2000 (Nøst et al., 2014; Rigét et al., 2013; Routti et al., 2017; Routti et al., 2016). This decline can be explained by the phase-out of PFOS and related precursors by the major producer in 2000–2002 (3M 2000), which has led to a lowered atmospheric delivery to the Arctic from the US, Japan and Western Europe of PFOS and other PFSAs and their volatile precursors (Wang et al., 2017). However, PFOS concentrations in polar bears and Arctic foxes levelled off in 2009–2010, and the authors suggested continued delivery of PFSAs to the European Arctic by slower ocean currents as a possible explanation (Routti et al., 2017). Also, studies on pilot whales (*Globicephala melas*) from the North-Atlantic reported increasing PFOS concentrations during the period of 1986–2013, perhaps due to slow removal of PFOS from ocean environments (Dassuncao et al., 2017).

Since white whales do not metabolize FOSA into PFOS, the estimated 27% decrease in FOSA concentrations in white whales from Svalbard over the 15 year period may indicate a lowered atmospheric delivery of FOSA or its volatile precursors to this area of the Arctic. Similarly, the detection rate of FOSA in polar bear and Arctic fox samples decreased during the period of 1997–2014 (Routti et al., 2017). A 34% decrease in muscle concentrations of FOSA was reported in pilot whales from the North-Atlantic from ~2000 to 2013, and this was accompanied by a 79% decline in FOSA:PFOS ratio (Dassuncao et al., 2017). In Alaska, a decline in the FOSA:PFOS ratio was reported in Chukchi Sea white whales between 1989 and 2006, while no trend was observed in white whales from Cook Inlet in this time period (Reiner et al., 2011). In contrast, the FOSA:PFOS ratio increased by an estimated 29% in white whales from Svalbard between 1996-2001 and 2013–2014. This potential change in ratio probably reflects the large decline in PFOS (or other non-FOSA precursors) over this time period. The estimated changes showed, nonetheless, a high uncertainty for FOSA (95% CI -45,-3.7) and FOSA:PFOS ratio (95% CI 2.53, 47.23), and should thus be interpreted with caution.

When comparing temporal PFSA trends in the present study to trends in Canadian white whale populations, the results from Svalbard are consistent with white whales from Western Hudson Bay; PFOS and FOSA concentrations have declined from 2003 to 2013 (Smythe et al., 2018). FOSA concentrations also decreased in white whales from Eastern Beaufort Sea and Cumberland Sound during 1984–2013 and 1982–2010, respectively, whereas the concentrations peaked in Western Hudson Bay in 2005 and decreased thereafter (Smythe et al., 2018). In contrast to the present study, PFOS concentrations increased in white whales from Cumberland Sound from 1982 to 2010 (Smythe et al., 2018). Also, PFOS concentrations appeared to be stable in Eastern Beaufort Sea white whales from 1984 to 2010, and then increased, followed by a decrease (Smythe et al., 2018).

#### 4.2.2. PFCA concentrations

In white whales from Svalbard, $\Sigma_7$PFCAs increased by 43% (95% CI 22, 58) from 1996 to 2001 to 2013–2014. This was due to an increase in four $C_{9-12}$ PFCAs across the sampling periods; PFNA by 141% (95% CI 138, 143), PFDA by 85% (95% CI 64, 98), PFUnDA by 42% (95% CI 17, 58) and PFDoDA by 35% (95% CI 14, 51) (Table 1). $C_{9-12}$ PFCAs also increased in polar bears and Arctic foxes from Svalbard from 2000 to 1997, respectively, until 2014 (Routti et al., 2017). Ringed seals from Svalbard showed an increasing trend for all PFCAs ($C_{9-13}$) from 1990 until 2010 (Routti et al., 2016). A similar increasing PFCA trend was reported for North Atlantic pilot whales from 1986 to 2003 (Rotander et al., 1984) and from 1994 to 2013 (Dassuncao et al., 2017), as well as North-Atlantic white-sided dolphins (*Lagenorhuchus acutus*) from 2001 to 2006 (Rotander er al. 2012). PFOA concentrations in polar bears and Arctic foxes from Svalbard have been stable (Routti et al., 2017), which is similar to the findings in the present study. PFTrDA ($C_{13}$) concentrations in polar bears, Arctic foxes and ringed seals from Svalbard as well as North-Atlantic pilot whales have been reported to increase over time (Dassuncao et al., 2017; Routti et al., 2017; Routti et al., 2016), while the white whales in the present study showed a 30% and 46% decrease in PFTrDA ($C_{13}$) and PFTeDA ($C_{14}$) concentrations, respectively, across the sampling periods, although the estimated changes were uncertain due to wide confidence intervals ((95% CI -59, −8.2) and (95% CI -110, −7.9), respectively).

Compared with other white whale populations in the Canadian Arctic, the results in the present study were similar to Cumberland Sound (1982–2010) and Alaskan (1984–2010) white whales, which have displayed an increase in $\Sigma C_{8-12}$ PFCAs (sum of PFOA, PFNA, PFDA, PFUnDA, and PFDoDA) (Reiner et al., 2011; Smythe et al., 2018). Contrary to the findings herein, decreasing trends of $\Sigma C_{8-12}$ PFCAs in a comparable time period have been reported for Eastern Beaufort Sea and Western Hudson Bay populations (Smythe et al., 2018).

Variations in temporal trends of PFCAs across Arctic white whale populations, may be explained by regional differences in exposure sources and transport mechanisms to the different Arctic regions, which is probably linked to the complex emission history of PFCAs. Since 2002, direct emissions of PFCAs from Europe, the US and Japan have declined (Wang et al., 2014a). Nonetheless, PFCAs are still present in the marine environment and transported towards the Arctic through slow oceanic transport. PFCA emissions have increased rapidly from China and other developing countries in Asia since 2003 (Wang et al., 2014a). Additionally, fluorotelomer-based precursors increasingly contribute to global PFCA emissions (Wang et al., 2014a) and there are probably several unquantified sources that contribute to global PFCA emissions (Wang et al., 2014b).

#### 4.2.3. Limitations and confounding factors

Age, sex and season may confound investigations of time trends when comparing OHC concentrations, including PFASs, of different animals. In the present study, the whales from the two time periods were sampled at approximately the same time of the year. However, small sample sizes and varying number of whales belonging to each age and sex group across the two sampling-periods, limited investigation of the relationships between PFASs and age or sex. Also, age information (i.e. by counting growth layer groups in dentine) was not available, only an approximation based on body length. However, many marine mammal studies report lack of relationships between age or sex with PFAS concentrations; e.g. in ringed seals (Butt et al., 2008; Routti et al., 2016), harbour seals (Routti et al., 2014), polar bears and Arctic foxes (Routti et al., 2017), pilot whales (Dassuncao et al., 2017), and white whales from the Canadian Arctic (Kelly et al., 2009; Smythe et al., 2018; Tomy et al., 2009). Nevertheless, in a study of Alaskan white whales ($n = 68$), Reiner et al. (2011) found that males generally had higher concentrations of most PFASs compared to females. Apart from significant but weak, negative associations between body length (as an indicator of age) and PFNA and PFOS, there were no relationships with other PFASs (Reiner et al., 2011). Although not adjusting for sex or age may present a limitation in the time trend analyses herein, the major trends were probably not profoundly biased as the majority of marine mammal studies to date do not show large impacts of age and sex on PFAS concentrations (Butt et al., 2008; Dassuncao et al., 2017; Kelly et al., 2009; Routti et al., 2016; Routti et al., 2014; Smythe et al., 2018; Tomy et al., 2009).

Another limitation of the present study is that the time trend analysis was based on a few samples from two time periods 1.5 decades apart, and did not capture yearly variations in PFAS concentrations between 2002 and 2012. Also, no repeated measurements of the same individuals were obtained. In addition to time-related changes in production of specific PFASs and their emission sources, climate-change related alterations in oceanographic and atmospheric systems, increased temperature and thus precipitation and ice/snow-smelting, and altered predator-prey interaction can modify transport pathways to Arctic regions and exposure to PFASs and their precursors in Arctic wildlife (McKinney et al., 2015; McKinney et al., 2013; Routti et al., 2017; Smythe et al., 2018). A satellite-telemetry study of white whales from Svalbard reports a habitat change compared to 20 years ago. The whales now spend more time foraging out in the fjords and less time near glacier-fronts, suggesting a change in diet from only Arctic fish species towards a diet that includes more Atlantic fishes, which have become more common in the fjords of Svalbard with increased influxes of Atlantic Water (Fossheim et al., 2015; Spielhagen et al., 2011; Vacquié-Garcia et al., 2018). Thus, a shift in prey species in the two time periods of the sampled white whales herein may also have contributed to altered PFAS concentrations and patterns from 1996 to 2001 to 2013–2014. Despite the limitations, the consistency with other time trend studies of marine mammals in the European Arctic suggests that the current study depicts the main PFAS trends over the study period.

### 4.3. Toxicological implications

The possible toxicological impacts of PFASs on white whale health are presently unknown. Even with the large decrease in PFOS in white whales from Svalbard found in the present study (from 40.7 to 22.8 ng/mL), it is still by far the most dominant PFAS in white whale plasma. Even if the PFCA concentrations are much lower, the increase in the most persistent ($C_{9\text{-}12}$) PFCAs by 35–141% (e.g. PFUnDA increased from 4.86 to 7.09 ng/mL) in white whales from Svalbard over the last 1.5 decades also causes concern for possible adverse effects. Long-term chronic toxicological effects of PFCAs (except PFOA) are much less studied than PFOS and little is known about how these compounds impact vulnerable subpopulations such as developing offspring (Mariussen, 2012; Saikat et al., 2013; Sunderland et al., 2019).

Generally, there is a lack of studies investigating the health impact of any PFAS in Arctic marine mammals. The few existing studies have focused on polar bear subpopulations in Svalbard and East-Greenland (recently reviewed by Routti et al., 2019a); studies report possible endocrine disruption (thyroid and steroid hormones) (Bourgeon et al., 2017; Pedersen et al., 2016), neuroendocrine modulations in the brain (Pedersen et al., 2015; Pedersen et al., 2016), and alterations in lipid and energy metabolism (Routti et al., 2019b; Tartu et al., 2017b), as well as potentially carcinogenic, reproductive and immune system effects (only PFOS; risk assessment based) (Dietz et al., 2015; Sonne, 2010). Although concentrations in the present white whales are considerably lower than in polar bears for most PFASs (except FOSA), a study of hooded seals with similar plasma PFAS concentration as in the present white whales, indicated disruption of thyroid function in the seals (Grønnestad et al., 2018). PFCAs were associated with lowered concentrations of free triiodothyronine (FT3) in hooded seal pups and mothers (Grønnestad et al., 2018). Additionally, PFCAs and PFSAs in hooded seal pups were important predictors for the ratio of free thyroxine (T4) to FT3, also when compared to other potential thyroid disrupting contaminants such as PCBs and brominated OHCs (Grønnestad et al., 2018). Adult male walruses generally show lower PFAS concentrations than white whales (Scotter et al., 2019). However, ΣPFASs was associated with blood transcript levels of genes related to immune response, suggesting that PFASs may negatively impact immune function in walruses (Routti et al., 2019c). Altogether, this may imply that there is potential for similar adverse health effects in white whales. Having previously shown that the thyroid hormone system in white whales might be a sensitive target to OHCs (Villanger et al., 2011), it is important to continuously monitor both legacy and emerging PFASs in white whales from Svalbard, and to investigate possible disruption of thyroid hormone and immune systems, and other health-related outcomes.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## CRediT authorship contribution statement

**Gro D. Villanger:** Conceptualization, Visualization, Investigation, Formal analysis, Writing - original draft, Writing - review &

editing. **Kit M. Kovacs:** Investigation, Writing - original draft, Writing - review & editing. **Christian Lydersen:** Project administration, Funding acquisition, Resources, Writing - original draft. **Line S. Haug:** Resources, Methodology, Writing - original draft, Writing - review & editing. **Azemira Sabaredzovic:** Resources, Methodology, Writing - original draft, Writing - review & editing. **Bjørn M. Jenssen:** Resources, Writing - original draft. **Heli Routti:** Funding acquisition, Investigation, Data curation, Writing - original draft, Writing - review & editing.

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## Appendix A. Supplementary data

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