Levels and trends of poly- and perfluoroalkyl substances in the Arctic environment — An update

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ABSTRACT

Poly- and perfluoroalkyl substances (PFASs) are important environmental contaminants globally and in the early 2000s they were shown to be ubiquitous contaminants in Arctic wildlife. Previous reviews by Butt et al. and Letcher et al. have covered studies on levels and trends of PFASs in the Arctic that were available to 2009. The purpose of this review is to focus on more recent work, generally published between 2009 and 2018, with emphasis on PFASs of emerging concern such as perfluoroalkyl carboxylates (PFCAs) and short-chain perfluoroalkyl sulfonates (PFSAs) and their precursors. Atmospheric measurements over the period 2006—2014 have shown that fluorotelomer alcohols (FTOHs) as well as perfluorobutanoic acid (PFBA) and perfluorooctanoic acid (PFOA) are the most prominent PFASs in the arctic atmosphere, all with increasing concentrations at Alert although PFOA concentrations declined at the Zeppelin Station (Svalbard). Results from ice cores show generally increasing deposition of PFCAs on the Devon Ice cap in the Canadian arctic while declining fluxes were found in a glacier on Svalbard. An extensive dataset exists for long-term trends of long-chain PFCAs that have been reported in Arctic biota with some datasets including archived samples from the 1970s and 1980s. Trends in PFCAs over time vary among the same species across the North American Arctic, East and West Greenland, and Svalbard. Most long term time series show a decline from higher concentrations in the early 2000s. However there have been recent (post 2010) increasing trends of PFCAs in ringed seals in the Canadian Arctic, East Greenland polar bears and in arctic foxes in Svalbard. Annual biological sampling is helping to determine these relatively short term changes. Rising levels of some PFCAs have been explained by continued emissions of long-chain PFCAs and/or their precursors and inflows to the Arctic Ocean, especially from the North Atlantic. While the effectiveness of biological sampling for temporal trends in long-chain PFCAs and PFSAs has been demonstrated, this does not apply to the C_4-C_8—PFCAs, perfluorobutane sulfonamide (FBSA), or perfluorobutane sulfonate (PFBS) which are generally present at low concentrations in biota. In addition to air sampling, sampling abiotic media such as glacial cores, and annual

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sampling of lake waters and seawater would appear to be the best approaches for investigating trends in the less bioaccumulative PFASs.

1. Introduction

Poly- and perfluoroalkyl substances (PFASs) are a large suite of highly fluorinated aliphatic compounds, which differ in their functional groups and the length of the carbon chains [1]. For the perfluoroalkyl substances, all the carbon atoms of the alkyl chain are fully fluorinated as illustrated in Fig. 1, while polyfluoro substances contain non-fluorinated carbons, oxygens and other substituents. The high chemical and thermal stability of the C−F bond and the surfactant-like properties of PFASs are the main characteristics that have made this group of compounds suitable for many different uses in industrial and commercial applications. The exceptional stability of the perfluoroalkyl acids has also led to their ubiquitous presence in the global environment [2,3]. The PFASs that have been detected in environmental media can be further divided into the perfluoro-alkyl sulfonates (PFSA) and perfluorocarboxylates (PFCA) sub-groups, which include some of the most investigated PFASs over the past 18 years, i.e. perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA). Another large subgroup of PFASs is the fluorotelomer alcohols (FTOHs), considered the neutral precursors of the more stable acidic compounds, the PFCAs. PFASs have varying chain lengths. A distinction is often made between long-chain PFSAs (≥ 6 perfluorinated C atoms) and PFCAs (≥ 7 perfluorinated C) and their short-chain analogs [1].

Butt et al. [4] thoroughly reviewed information available for per- and polyfluoroalkyl substances (PFASs) in the Arctic, and Letcher et al. [5] reviewed PFAS exposure in wildlife and fish, both as part of a previous s Arctic Monitoring and Assessment Programme (AMAP) assessment. Therefore the purpose of this review is to focus on more recent work, generally published between 2009 and 2018, with emphasis on PFASs of emerging concern such as PFCAs and short-chain PFSAs and their precursors (Table S1A). This review is an updated and revised version of a chapter in the recent Arctic Monitoring and Assessment Programme (AMAP) report on "Contaminants of Emerging Arctic Concern" [6].

Table 1 summarizes the detection of PFASs in Arctic environmental media. It includes all reports to date, i.e. data summarized in previous reviews [4] as well as more recent published and unpublished results. Each environmental compartment is discussed separately in Section 2 (Environmental concentrations and spatial trends) and Section 3 (Temporal trends).
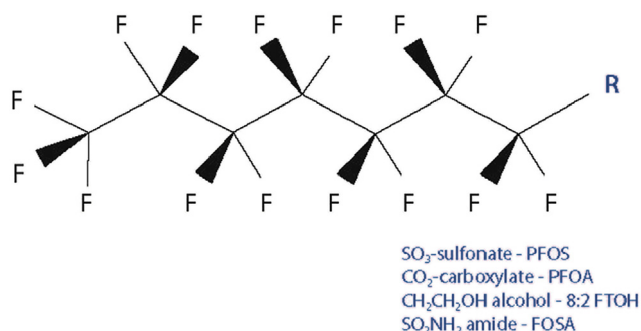
To ensure data comparability, quality assurance (QA) issues, such as field blanks, spike recoveries, and detection limits, were considered when selecting data on spatial and temporal trends of PFASs to include in this review. Recent overviews on temporal trends of persistent organic pollutants (POPs), including PFOS, in arctic biota [7] and of PFASs in arctic air [8] have also included consideration of data quality. A large interlaboratory program operated annually by Canada's Northern Contaminants Program and AMAP and with participation from circumpolar countries, has included analyses of PFASs and evaluation of laboratory performance [9].

1.1. Physical-chemical properties

The environmental fate of PFASs is influenced by their physical-chemical properties, which vary depending on their chain length and functional groups. Wang et al. [10] and Gomis et al. [11] summarized the limited information available on the physical-chemical properties of PFASs, including newer replacement compounds. Selected physical-chemical properties of the PFASs discussed in this section are provided in Table S1B. The PFCAs and PFSAs have very low acid dissociation constants (pKa) and are almost fully ionized under most environmental conditions [12,13]. As anionic substances the PFCAs and PFSAs also bind to proteins and phospholipids (reviewed by Armitage et al. [14]). They are typically found at elevated concentrations in tissues with high protein and phospholipid contents (e.g., liver, kidney, blood) Several PFASs have been shown to accumulate in food chains, including those in remote environments such as the Arctic [4,5].

Perfluorocarbon chain length and functional group are the predominant parameters influencing the partitioning of PFASs in abiotic environments [15]. The organic carbon partition coefficients (log K_{OC}) of PFCAs and PFSAs are relatively low (Table S1B) ranging from ~1.3 to 3.5 compared to neutral halogenated organics. The short-chain PFCAs ($\leq C_7$ chain length) were exclusively found in the dissolved phase, while long-chain PFCAs ($\geq C_8$ chain length), PFSAs, ethylperfluorooctane sulfonamidoacetic acid (EtFOSAA), and PFOSA appeared to bind more strongly to particles [15,16]. The partitioning also varied depending on the conditions (e.g., organic carbon content, pH, metal ions), for example, increasing sorption was found with increasing organic carbon content [15]. As a consequence, the short-chain PFCAs have a higher potential for long-range transport in aquatic environments, while PFSAs, EtFOSAA and perfluorooctane sulfonamide (FOSA) are preferentially distributed in biota or the abiotic environment such as sediments, which could act as a sink for PFASs. The physical-chemical characteristics also influence the type of long-range transport in the aqueous environment (e.g., sea spray, microlayer, surface water, deep ocean water).

1.2. Sources, production, uses, and transport

PFASs have been produced since the 1950s and used in surfactants and polymers [1]. PFASs have been used in polymers developed for stain repellent treatments of textiles and carpets and for use in grease-proof, food-contact paper. As surfactants they have

SO$_3$-sulfonate - PFOS
CO$_2$-carboxylate - PFOA
CH$_2$CH$_2$OH alcohol - 8:2 FTOH
SO$_2$NH$_2$ amide - FOSA

Fig. 1. Molecular structure of perfluorinated alkyl substances.

Table 1
Summary PFASs covered in this review and of Arctic media for which PFAS data have been reported as of 2018. Full nomenclature is provided in Table S1A.

PFAS	Atmosphere		Terrestrial		Freshwater			Marine		
	Air	Snow	Soil	Biota	Water	Sediment	Biota	Water	Sediment	Biota
PFBA	×	×			×		×	×	×	
PFPeA	×	×			×		×	×		
PFHxA	×	×		×	×	×	×	×		×
PFHpA	×	×		×	×	×		×	×	×
PFOA	×	×		×	×	×		×		×
PFNA	×	×		×	×	×	×	×		×
PFDA	×	×		×	×	×		×		×
PFUnDA	×	×		×	×	×	×	×		×
PFDoDA	×	×		×	×	×		×		×
PFTrDA	×			×	×	×				×
PFTeDA	×									×
PFPeDA	×									×
PFHxDA	×									×
6:2-Cl-PFAES		×								×
PFBS	×	×		×	×			×		×
PFHxS	×	×		×	×	×		×		×
PFHpS	×	×		×	×			×		×
PFOS	×	×	×	×	×	×	×	×	×	×
PFDS	×			×				×		×
PFDoS										×
4:2 FTS					×					
6:2 FTS	×				×					×
FBSA										×
FOSA	×	×	×	×	×	×	×	×	×	×
N-EtFOSA	×									
N-EtFOSE	×							×		
N-MeFOSE	×							×		
N-MeFOSEA	×									
PFECHS		×			×			×		×
6:2 FTOH	×									
8:2 FTOH	×									
10:2 FTOH	×									
8:2 FTUCA										×
10:2 FTUCA										×
7:3 FTCA										×
8:2 FTCA										
10:2 FTCA										

been used (in free acid or salt forms) as the processing aids for fluoropolymer manufacture (e.g. Teflon™), in aqueous film-forming foams and in wetting and leveling agents, emulsifiers, foaming agents, and dispersants due to their ability to lower aqueous surface tension [1,17—19]. Worldwide production of perfluorooctane sulfonyl fluoride (POSF), the key building block for PFOS-related compounds, was estimated at 96,000 tonnes (or 122,500 tonnes, when unusable waste production is included) during the period 1972—2002 [18]. After discovering that several PFASs were globally distributed in the abiotic environment, biota, food items and humans, measures have been taken by international authorities to regulate their production and use. PFOS and its salts were included in the Stockholm Convention in 2009 (under Annex B, which allows exemptions for use) owing to their persistence, bioaccumulation and toxicity. As of 2018 PFOA and perfluorohexane sulfonate (PFHxS) are currently under review for listing [20].

PFCAs are degradation products of perfluoro- and polyfluoro-substances including fluorotelomer alcohols, perfluoroalkyl phosphates, phosphonates, acrylates, and other compounds with perfluorinated carbon chains [1]. Some PFCAs, especially PFOA and perfluorononanoate (PFNA), had commercial uses as processing aids for fluoropolymers. Two manufacturing techniques have been used to produce PFOA: (i) electrochemical fluorination (ECF) in which octanoyl fluoride, $C_7H_{15}COF$, undergoes electrolysis in anhydrous hydrogen fluoride, and (ii) telomerization in which pentafluoroethyl iodide is reacted with tetrafluoroethylene to yield a mixture of perfluoroalkyl iodides. The two manufacturing processes for PFOA and other PFCAs are readily distinguishable by the isomeric composition of the final commercial products because ECF yields straight-chain and branched isomers while telomerization retains the geometry of the starting material with the majority being straight-chain isomers with an even number of perfluorocarbons [21]. These differences have been used to identify sources to the Arctic Ocean [22].

Under the United States Environmental Protection Agency (US EPA) PFOA Stewardship Program [23], eight leading fluorochemical manufacturing companies agreed to reduce emissions and product content of PFOA in fluorinated polymers (i.e., containing perfluoroalkyl moieties) by 95% by 2010 and to work toward their elimination by 2015 [24]. Recent trends among the global PFAS producers are to replace the long-chain PFSAs and PFCAs with shorter-chain homologues or other types of fluorinated chemicals [19]. However not all global manufacturers have participated in this voluntary agreement and production of C_8-related perfluorinated chemicals continues in China [25] as well as in Russia and India [26]. Wang et al. [27] concluded that releases from fluoropolymer production contributed most to historical PFCA emissions (e.g., 55—83% in the period 1951—2002). They also noted that since 2002, there has been a geographical shift of industrial sources (particularly fluoropolymer production sites) from North America, Europe and Japan to emerging Asian economies, especially China.

Wang et al. [27] quantified global emissions of C_4—C_{14} - PFCA homologues during the life-cycle of products based on PFOA, PFNA, PFOS and fluorotelomer compounds. Their approach improved

upon previous estimates of emissions of PFOA by assigning an average duration to each stage in the product life-cycle, estimating amounts of PFOA- and PFNA-based derivatives used as fluoropolymer processing aids, and incorporating information on the PFOA reduction process under the US EPA Stewardship Program. Their estimated time trends of PFCAs suggest significant ongoing emissions until at least 2020, depending on lower and higher use and emission scenarios (Fig. 2). These trends can be compared with ongoing monitoring of PFCAs in Arctic wildlife (see Section 3.2).

The long-range transport of PFASs to remote areas has been explained by two processes: (i) transport by ocean currents [28] and (ii) atmospheric oxidative transformations and subsequent wet and dry deposition PFSAs and PFCAs derived from airborne precursors, such as FTOHs, FOSAs, and sulfonamido ethanols (FOSEs) [29,30]. PFSAs and PFCAs are persistent and most often found as anions in aquatic environments due to their relatively low pK_a values [4,31]. The main pathway for PFASs to Northern oceans is transport by ocean currents. However, atmospheric long-range transport, degradation of volatile precursors to PFCAs in the atmosphere (e.g., 8:2 FTOH to PFOA) and secondary sources, such as melting glacier and snow packs also contribute [17,28,31—33]. A recent study suggests that sea spray aerosols could be an important source of PFASs to the atmosphere and, over certain areas where sea spray deposition is important, a significant source to terrestrial environments [34].

1.3. Transformation processes

There are multiple pathways in the environment that can account for the presence of PFCAs and other PFASs in Arctic abiotic and biotic compartments. In addition to ocean transport and atmospheric long-range transport of PFCAs and PFSAs (reviewed by Wang et al. [35]), PFASs can also be produced from perfluoroalkyl-containing precursors that undergo degradation processes such as atmospheric oxidation [36]. FTOHs may also undergo degradation on metal-rich atmospheric particle surfaces [37].

In general, biotransformation pathways are important for explaining the relative proportions of PFCAs and PFSAs in exposed biota [38]. Controlled studies have not been performed on biotransformation of fluorotelomer-related compounds by Arctic biota. Studies with laboratory animals have shown that the 8:2 FTOH, widely used as starting material for fluoropolymers, and present as a volatile residual in the polymer, undergoes metabolism to PFOA and, to a smaller degree, PFNA and shorter chain-length PFCAs [38]. There have been a few studies of other fluorotelomer-

related products including biotransformation of polyfluoroalkyl phosphates (PAPs), 8:2 fluorotelomer acrylate (8:2 FTAC) and fluorotelomer carboxylates (FTCAs, FTUCAs), all of which yielded PFOA and other PFCAs as persistent degradation products. Galatius et al. [39] compared PFCA and short-chain PFSAs in three marine mammal species in the North Sea. They found that harbor porpoises (*Phocoena phocoena*) and harbor seals (*Phoca vitulina*) showed lower concentrations of PFCAs than white-beaked dolphins (*Lagenorhynchus albirostris*), and that the seals had higher proportions of perfluorohexane sulfonate (PFHxS) (C_6). The differences were attributed to more rapid excretion in cetaceans.

Pinnipeds also appear to have a much higher metabolic capacity for transforming PFOSA to PFOS than cetacean species. Letcher et al. [40] reported for polar bears, ringed seal and beluga whale on the (*in vitro*) metabolism of a PFOS precursor, *N*-EtFOSA and FOSA, using enzymatically viable liver microsomes. Greater than 95% of *N*-EtFOSA was depleted by the polar bear microsomes, whereas for seal it was 65% and for beluga there was negligible depletion. The concomitantly formation of FOSA from *N*-EtFOSA was also quantitatively polar bear > ringed seal >>> beluga whale. They suggested that, in part, a source of FOSA in polar bears is the biotransformation of accumulated *N*-EtFOSA in polar bears.

1.4. Modelling studies

Global modelling has been used successfully to assess transport of PFASs to the Arctic and to try to explain empirical observations in air, ocean waters and biota [28,31,32,41]. Processes that transport PFASs to the Arctic include transport of parent compounds in air or water and/or of the precursors that can undergo transformation to the ionic form (PFOS and PFCAs). Butt et al. [4] reviewed the initial modelling work; here more recent (post-2009) literature focuses on PFOA and long-chain PFCAs are included. The initial studies showed that ocean transport of directly emitted PFOA is the main source of this compound to the Arctic Ocean and, with continued transport from lower latitudes, concentrations of PFOA are expected to increase over the next couple of decades. However, with the regulation and phase out of some PFASs, emissions are predicted to decrease and lead over time to reduced levels in the Arctic [27].

Armitage et al. [28,31] used the BETR Global model to show that transport via ocean currents is the major pathway to the Arctic for PFOA and long-chain PFCAs that are directly discharged to aquatic environments of urban and industrial areas at mid-latitudes. They predicted PFOA concentrations in the 30—250 pg/L range for the

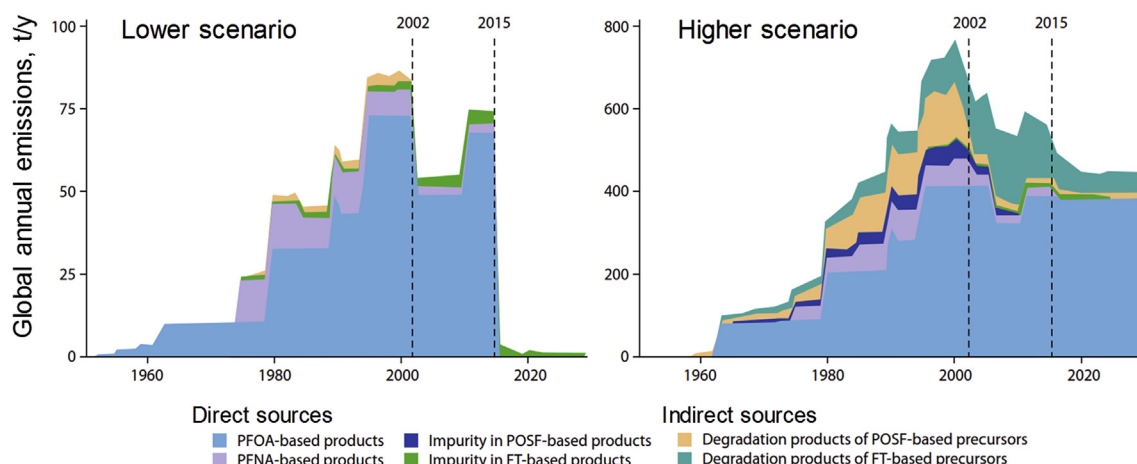**Fig. 2.** Estimated total global annual emissions of C_4—C_{14} PFCAs (1951—2030). Lower and higher production scenarios are shown. Redrawn from Wang et al. [27].
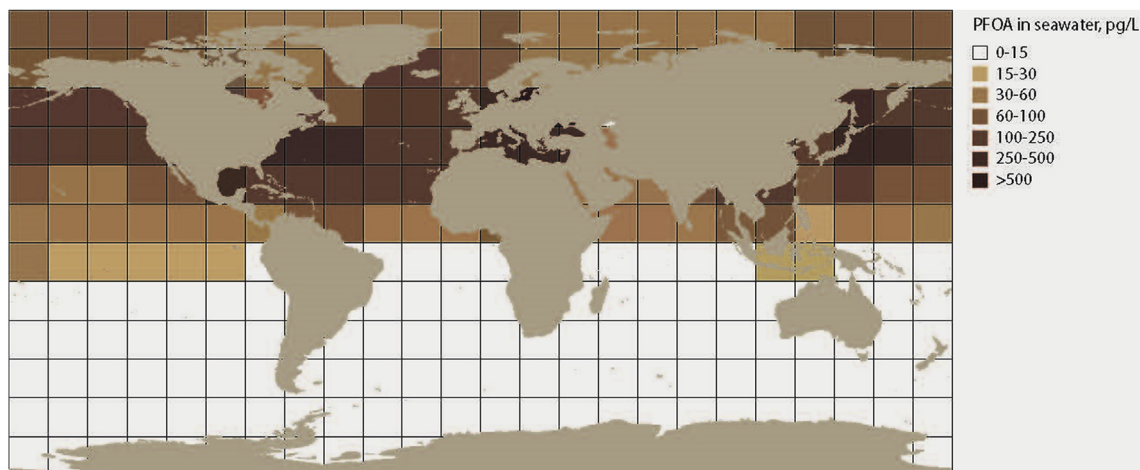
Fig. 3. Modelled concentrations (pg/L) of PFOA in surface ocean waters in 2005. Redrawn from Armitage et al. [31].

Arctic Ocean and adjacent northern seas (Fig. 3) which were within the range observed in seawater monitoring (see Section 2.4).

Stemmler and Lammel [32] used a global multi-compartment model (MPIMCTM) to assess transport of PFOA to the Arctic. This model allowed a comparison of the relative importance of different ocean pathways and showed that the main source of PFOA to the Arctic Ocean was inflow through the Norwegian Sea and that this was strongly influenced by changes in water transport, which thus determined its interannual variability. The atmospheric component of the model suggested that PFOA deposition occurred via episodic transport events (timescale of days) into the Arctic and that wet deposition occurred over land with maxima in winter (Fig. 4).

Yamashita et al. [42] hypothesized that PFASs could be transported globally with the thermohaline circulation system, and that open-ocean water acts as a final sink for PFOS and PFOA. Lohmann et al. [43] showed that a vertical eddy diffusion model could reproduce the observed depth profile of PFOA concentrations at sites in the Pacific and Atlantic Oceans. This process has not been considered in previous modelling of hydrophobic POPs in the ocean, which has assumed they are removed from the water column mainly by settling particles [44].

Zhang et al. [45] modelled the transport of PFOS to the Arctic via the North Atlantic Ocean by developing a 3-D simulation within the MIT general circulation model for the North Atlantic (fully described at http://mitgcm.org). The model assumed only wastewater and riverine inputs were important with North American

coastal releases accounting for 59% of total releases, and the remaining 41% from Europe. Cumulative inputs to the Arctic accounted for 30% of continental discharges. A low release scenario for PFOS matched the observed concentrations of seawater PFOS concentrations in the Labrador Sea observed by Yamashita et al. [42,46]. Atlantic meridional overturning circulation (AMOC), which returns deep water southward, was estimated to reduce northward transport of PFOS by 25%.

Yeung et al. [47] modelled concentrations of PFASs in the Arctic Ocean using a three-compartment geochemical box model adapted from Soerensen et al. [48]. Modelled results suggested atmospheric inputs accounted for 34–59% (~11–19 pg/L) of measured PFOA concentrations in the polar mixed layer (PML) (mean 32 ± 15 pg/L) [47] while for PFOS the contribution of atmospheric deposition to surface water concentrations was minor at <3 pg/L or < 30% of mean observed concentrations. The modelling results from Yeung et al. suggest a greater contribution from atmospheric deposition of PFOA to the Arctic Ocean than predicted by earlier modelling [31,32].

Atmospheric long-range transport and degradation of volatile precursors, such as fluorotelomer alcohols, to PFCAs in the atmosphere are important for Arctic terrestrial environments and could contribute to oceanic inputs via melting glaciers and snow packs (see Section 2.1). Wallington et al. [49] used a three-dimensional global atmospheric chemistry model (IMPACT) to show that 8:2 FTOH, a major precursor of PFOA, would be globally distributed,
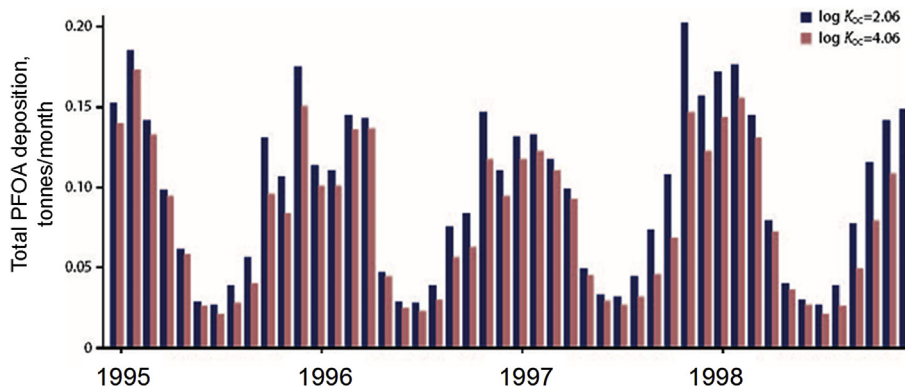

Fig. 4. Four-year time series of monthly sums of total (wet + dry) deposition of PFOA [tonnes/month] to the Arctic predicted by the MPIMCTM model [32]. The blue and red bars represent modelled PFOA deposition assuming a log organic carbon (log Koc) partition coefficient of 2.06 and 4.06, respectively. Redrawn from Stemmler and Lammel [32].

consistent with the measured half-life of approximately 20 days. PFOA was also predicted to be present at higher atmospheric concentrations during the Arctic summer due to degradation of 8:2 FTOH by photo-oxidants, such as OH radical, and very low concentrations during the Arctic winter due to low light intensity and removal in precipitation. Atmospheric oxidative transformations of other airborne precursors, such as short-chain FTOHs and short-chain FOSAs, and FOSEs [29,30], and subsequent wet and dry deposition of PFSAs and PFCAs are also likely to occur although they have not been subject to detailed atmospheric deposition modelling.

2. Environmental concentrations and spatial trends

2.1. Air and precipitation air

To better understand the fate and long-range transport of PFAS to the Arctic, both neutral precursors and ionic PFASs have been measured in the atmosphere at Arctic sites and in the northern Atlantic Ocean. Ionic PFASs have usually been measured in the particulate phase, while the neutral precursors have been measured in both the gas- and particulate phase. Wong et al. [8] reported the first long term temporal trends of neutral and ionic PFASs in air from three arctic stations: Alert (Canada, 2006–2014), Zeppelin (Svalbard, Norway, 2006–2014), and Andøya (Norway, 2010–2014). At Alert, each sample (~2000 m^3) comprised of a glass fiber filter (GFF) and a polyurethane foam (PUF) cartridge while at Zeppelin and Andøya samples (~1200 m^3) were collected only a GFF. Thus only particle-phase PFASs were collected at the Norwegian sites. C$_6$–C$_{11}$–PFCAs and C$_4$–C$_{10}$–PFSAs were determined at all 3 sites while C$_4$, C$_5$, C$_{12-14}$, C$_{16}$, C$_{18}$–PFCAs; 6:2, 8:2, 10:2 fluorotelomer acrylates (FTAC); FOSEs and FOSAs were determined only at Alert; and 1H,1H,2H,2H-perfluorooctanesulfonate (6:2 FTS) and PFOSA were determined only at Zeppelin and Andøya. Among the PFASs that were determined at all sites, the most abundant were C$_6$–C$_{11}$–PFCAs (Fig. 5). These PFCAs were 3 to 30-fold higher at Zeppelin and Andøya compared to Alert. For example, PFOA concentrations in air at Alert (<0.0063–1.3 pg/m^3) were significantly lower than at Zeppelin (<0.12–4.0 pg/m^3) and Andøya (<0.12–5.5 pg/m^3). Wong et al. [8] attributed this to sea spray aerosol due to proximity to the ocean of the Norwegian sites. Alert is located at the northeastern tip of the Ellesmere Island and approximately 4 km from water which is covered by sea ice for most of the year, while the Norwegian sites are closer to open ocean waters. However, for C$_4$–C$_{10}$ PFSAs, the concentrations in air at the 3 sites were similar (Fig. 5; Table S2).

Air concentrations of PFBA in Alert ranged from <0.0063 to 29 pg/m^3) and it was mainly detected in the gas phase. The concentrations were within the same range as the urban sites in Europe (0.93–7.0 pg/m^3) [50–52], which suggests that PFBA may be uniformly distributed in the northern hemisphere. At Alert 6:2, 8:2 and 10:2 FTOHs, sampled with the PUF, had median concentrations of 0.57, 3.7 and 1.3 pg/m^3. These concentrations were much higher than the PFCAs. For example 8:2 FTOH was 25-fold higher than PFOA, the most prominent C$_6$–C$_{11}$-PFCA (Fig. 5). Temporal trends of PFASs reported by Wong et al. [8] are discussed in Section 3.1.

Results of long-term air monitoring for PFOS and PFOA in airborne particles have been reported for Svalbard (Zeppelin Station) for the period 2006–2012 [53]. PFOA continues to be the predominant compound among the C$_6$–C$_{11}$–PFCAs in air at Zeppelin (Fig. 6A). PFASs are monitored in air at several locations in Norway, but not many samples have been analyzed and the concentrations of many compounds are below the detection limits. For example, 1H,1H,2H,2H-perfluorooctanesulfonate (6:2 FTS), perfluorobutane sulfonate (PFBS), perfluorodecanoate (PFDA), perfluorodecane sulfonate (PFDS), perfluoroheptanoate (PFHpA), perfluorohexanoate (PFHxA) and PFHxS were not detected in samples from any month at Andøya (northern Norway) in 2014, although they were detectable in 2010–2013. The situation was similar at Zeppelin, although PFDA was found in samples for four months in 2014 (Fig. 6A). PFDA was reported from the southernmost station (Birkenes; southern Norway) as was 6:2 FTS; but they were only found during two months and a single month, respectively [53]. There is no clear south-north trend in Norway for the number of PFASs detected, or for concentrations of PFOA measured (Fig. 6B). All stations show peak concentrations during summer [53].

A recent report on screening for new contaminants at Zeppelin station revealed several poly/perfluoro-compounds perfluoroperhydrophenanthrene (PFPHP), tris(perfluorobutyl)-amine (PFTBA), and 1,2,3,4-tetrachlorohexafluorobutane (TCHFB) in arctic air for the first time. These compounds are very volatile and the authors noted that concentrations reported may therefore be underestimated (due to breakthrough in PUF samplers) although still indicating that these PFASs may undergo long-range transport
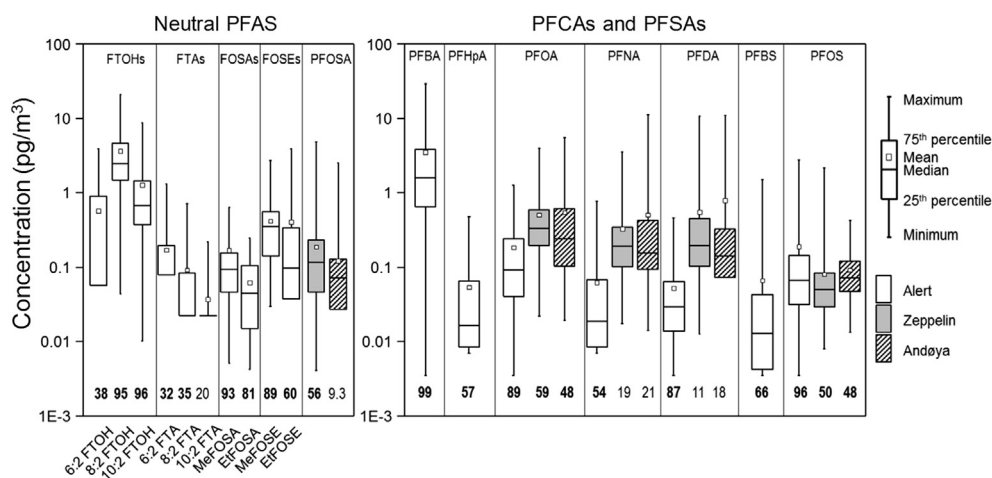

Fig. 5. Box-whisker plots of concentration of PFASs in air at Alert (2006–2014), Zeppelin (2006–2014) and Andøya (2010–2014). Detection frequencies (DF, %) are shown in the bottom of each plot. DF > 30% are shown in bold. Reproduced from Wong et al. [8].
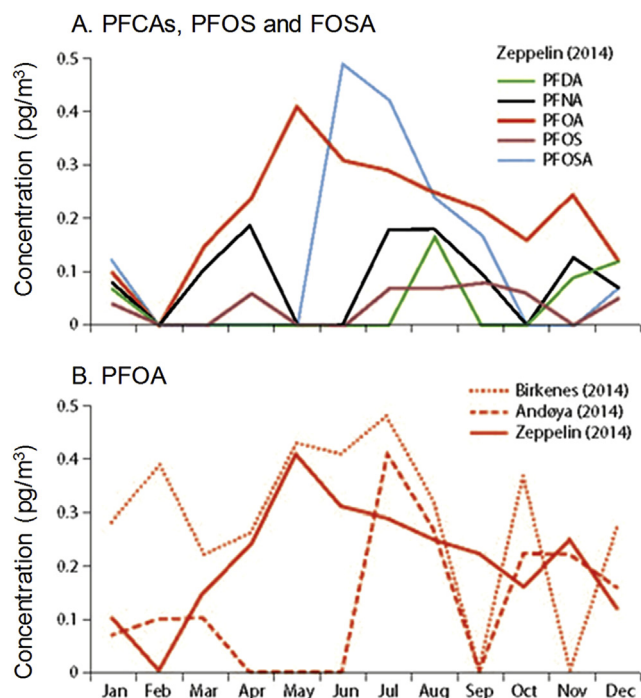
Fig. 6. A) Monthly mean concentrations (pg/m^3) of ionic PFASs and PFOSA in air at Zeppelin, 2014; B) Mean monthly PFOA at Zeppelin, Andøya and Birkenes in 2014 [53].

[54].

Neutral PFAS (FTOH, perfluorosulfonamides and sulfonamido-ethanols) have been measured since 2007 in northern Greenland (Villum Research Station (VRS), Station Nord) and the results covering the period 2007—2015 have been summarized in a recent report [55]. The average yearly sum of the seven measured neutral PFAS (Σ7PFAS) in the atmosphere at VRS ranged from 1.82 to 32.1 pg m-3 (Table 2). The most abundant compound was 8:2 FTOH (44% of Σ7PFAS), followed by 6:2 FTOH (25% of Σ7PFAS) and 10:2 FTOH (14% of Σ7PFAS). The concentrations of FTOH were comparable with those measured at other High Arctic sites (Ny Ålesund (Zeppelin) and Alert).

Atmospheric data for PFASs have also been collected during ship cruises along the northern Atlantic Ocean including Arctic coastal regions. Cai et al. [56] investigated the inter-hemispheric gradient of neutral precursors in atmospheric samples, starting from the Japan Sea and then on a transect towards the Arctic Ocean. Samples north of 66°N were taken in the period July—August 2010. 8:2 FTOH was the compound detected at the highest concentrations (range: 83.4—160 pg/m^3) followed by 10:2 FTOH (range: 6.2—31.2 pg/m^3). Other neutral precursors (FOSA and FOSE) were also detected at concentrations in the pg/m^3 range. Concentrations of fluorotelomer

acrylates (6:2 FTAC, 8:2 FTAC and 10:2 FTAC) were also reported.

Air samples collected on the *Amundsen* icebreaker during annual cruises in the Canadian Arctic since 2007 have been analyzed for PFASs. These data build on earlier studies of PFAS in the Canadian Arctic using oceanographic cruises [57,58]. Fig. 7 shows data for 2010—2011.

Air concentrations of most PFAS compounds measured in the Arctic in 2010 and 2011 were slightly lower or within the same range as those measured in air along a cruise track from coastal Germany to the center of the North Sea in May 2009 by Xie et al. [59].

Xie et al. [60] measured neutral PFASs in air and snow collected from Ny-Ålesund (Svalbard) and determined their air-snow exchange fluxes. Concentrations of the ΣFTOHs in air over a one-year period varied from 5.6 to 34 pg/m^3 with a mean of 14 pg/m^3 (Fig. 8). Among the perfluorosulfonates/amides, MeFBSA, the precursor of PFBS, was the principal component followed by MeFOSA and EtFOSA.

Passive air sampling using polyurethane foam (PUF) or sorbent-impregnated polyurethane foam (SIP) has also been used to measure atmospheric concentrations of neutral PFASs at remote sites. The Global Atmospheric Passive Sampling (GAPS) network currently includes about 60 sites, including five in the Arctic: Alert and Little Fox Lake (Canada); Barrow (Alaska, USA); Ny-Ålesund (Svalbard) and Stórhöfði, (Iceland) [61,62]. A global-scale pilot study in 2009 using SIP disk samplers, demonstrated the feasibility of the method for both neutral and ionizable PFASs [62]. A follow-up study by Rauert et al. [61], which included samples from 2009, 2013 and 2015 for the 5 Arctic sites, found 6:2 FTOH and 8:2 FTOH were the predominant PFASs in air ranging in concentration from 2 to 59 pg/m^3 and 1—11 pg/m^3 (Fig. 9; Table S2). MeFOSE, EtFOSE and EtFOSA were below detection limits at the 5 Arctic sites in 2013 and 2015, while EtFOSA concentrations ranged from 0.083 to 0.17 pg/m^3 with highest concentrations at Barrow and Little Fox Lake. EtFOSA concentrations were in line with levels over the Arctic Ocean in 2010 [56] and at Alert during 2006—2014 [8].

Ionizable PFASs were detected at all 5 locations in 2013 and 2015 with PFBA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUA, PFBS, PFHxS and PFOS, having the highest detection frequencies (44—100%). PFBA was by far the most predominant compound with concentrations ranging from 15 to 2160 pg/m^3 and highest concentrations at Little Fox Lake (Yukon) (Fig. 9; Table S2). Shorter-chain PFSAs, PFBS (0.17—9.8 pg/m^3) and PFHxS (0.063—4.3 pg/m^3) were also detected at all locations and concentrations in 2015 (only sampled from April to June) were 3—4 times higher than observed at Arctic high volume air sampling sites for the same sampling period [8]. Rauert et al. [61] did not comment on the elevated levels of PFBA, however, their results were consistent with observations for high volume sampling [8] and for arctic snow and surface waters (Table S3). PFBA has multiple non-fluorotelomer gas-phase sources, including chlorofluorocarbon replacements HFC-329 ($CF_3(CF_2)_3H$) [63], HFE-7100 ($C_4F_9OCH_3$), and HFE-7200 ($C_4F_9OC_2H_5$) [64,65].

Gawor et al. [66] reported results for neutral PFASs from XAD-type samplers deployed on a global-scale for one-year intervals during the period 2006—2011. These data include the Arctic sites mentioned above and were reported as ng/sampler to allow for comparable spatial and temporal trends to be assessed. Levels of FOSAs and FOSEs tended to decrease globally during the six years of measurements, whereas an initial decline in the concentrations of 6:2, 8:2 and 10:2-FTOHs from 2006 to 2008 did not continue in 2009—2011.

2.1.1. Precipitation/snow

The geographical coverage for PFASs in snow or wet precipitation in the Arctic is very limited, with data available for northern
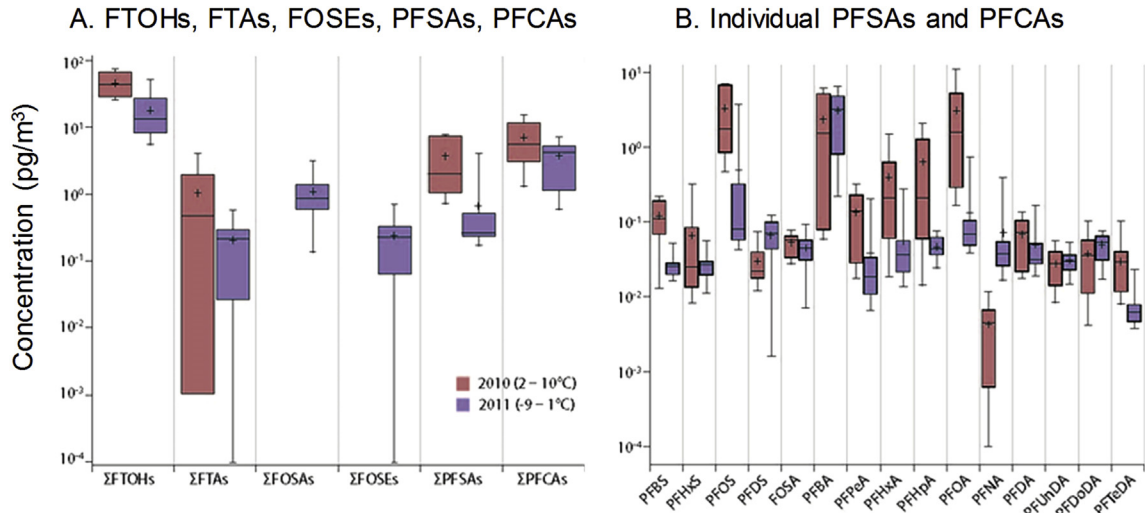
Table 2
Average, median and concentration ranges of neutral PFASs (pg/m^3) in years 2008—2015 (sum of gaseous and particulate phase) determined in northern Greenland (Villum Research Station) [55].

Compound	Average	Median	Range
6:2 FTOH	2.73	1.93	0.23—16.5
8:2 FTOH	4.48	3.37	0.23—22.4
10:2 FTOH	1.32	0.67	0.10—9.68
N-MeFOSA	0.34	0.22	0.10—3.41
N-EtFOSA	0.30	0.22	0.11—1.93
N-MeFOSE	0.48	0.28	0.07—7.46
N-EtFOSE	0.57	0.22	0.06—5.96

A. FTOHs, FTAs, FOSEs, PFSAs, PFCAs

B. Individual PFSAs and PFCAs

Fig. 7. Box and whisker plots of PFAS concentrations for A) PFAS groups, ΣFTOHs, ΣFTAs, ΣFOSAs, ΣFOSEs, ΣPFCAs and ΣPFSAs and B) individual PFSAs and PFCAs in air (sum of particle and gas phases) measured in the Canadian Arctic from the Amundsen icebreaker (2010−11). Boxes correspond to the interquartile range (25th and 75th percentiles), whiskers to the 5th and 95th percentiles. 2010 FOSAs and FOSEs are excluded owing to contamination in 2010 by MeFOSA and MeFOSEs. Results from M. Shoeib, Environment Canada, unpublished data.
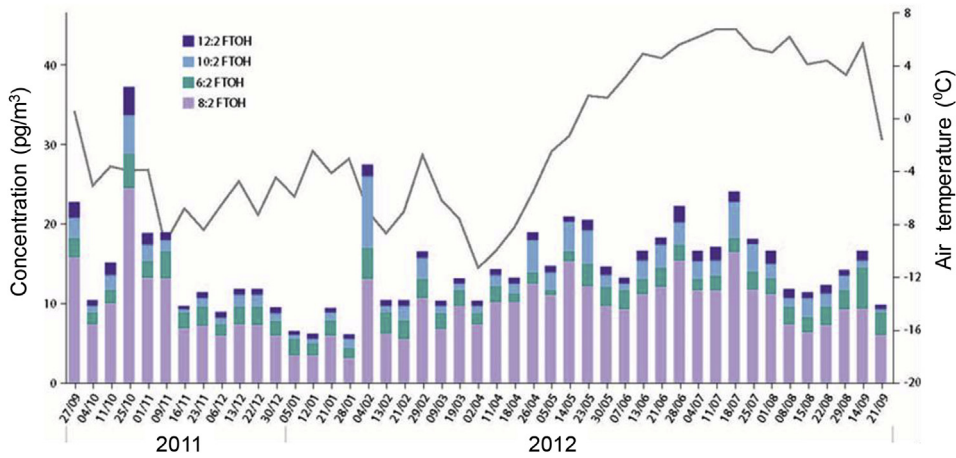
Fig. 8. FTOHs and other neutral PFAS compounds measured in air at Ny-Ålesund (Svalbard) between September 2011 and September 2012 [60]. The grey line and X symbols are average temperatures during the air sampling. Redrawn from Xie et al. [60].
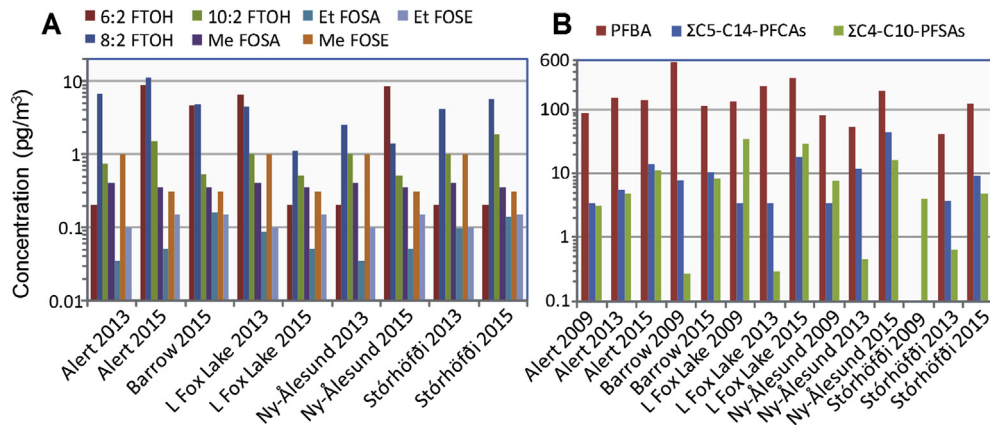

Fig. 9. Concentrations of (A) neutral PFASs and (B) ionizable PFASs in passive air sampler SIP disks in 2013 and 2105 at 5 Arctic locations from Rauert et al. [61]. PFBA is shown separately due to its consistently higher concentrations relative to other longer chain PFCAs.

Sweden, northern Norway, the Canadian Arctic islands, Svalbard and from ice/snow in the Beaufort Sea. Due to their physical-chemical properties, deposited PFSAs and PFCAs accumulate during winter instead of volatilizing back to the atmosphere, and the accumulated PFAS are delivered to the receiving marine systems (from snow on top of sea ice and terrestrial catchment areas) and terrestrial systems (from the snow pack) when the snow/ice melts [67,68]).

Kwok et al. [69] investigated PFAS concentrations in ice cores, surface snow and water samples collected from glaciers and downstream coastal areas of Svalbard. PFBA, PFOA, and PFNA were the predominant compounds found in ice-core samples (Table S3). Kwok et al. [69] found an increase of PFAS concentrations in snow and surface water with increasing proximity to Longyearbyen (Svalbard), suggesting the presence of local sources. PFOA was the main PFAS detected in surface snow, while PFBA and per-fluoropentanoate (PFPeA) were mainly found in surface water samples from glacial meltwater (Table S3).

Deposition of C_6-C_{12} PFCAs and PFOS in snow has been measured at Station Nord (Greenland) since 2008 (R. Bossi, Aarhus University, Roskilde, Denmark, unpubl. data) in samples collected three times per year. PFNA and PFOA were the predominant PFASs detected with average concentrations of 0.45 and 0.53 ng/L, respectively (Table S3).

Macinnis et al. [70] determined concentrations and loads (ng/m^2) of PFASs in integrated snowpack samples from the catchment and ice surface of Lake Hazen by sampling in May/June 2013 and 2014. PFBA was the dominant PFAS, with concentrations averaging 2700 ± 3227 pg/L and.

2656 ± 682 pg/L in light snowpacks during 2013 and 2014, respectively, and 4844 ± 2836 pg/L in dark snowpacks during 2014 (Table S3). PFOS was the dominant PFSA in snow but at concentrations were generally an order of magnitude less than PFCAs (Table S3). Loads of ΣPFASs were lower in snowpacks enriched with light absorbing particles and were attributed to reduced snowpack albedo combined with enhanced post-depositional melting [70].

Veillette et al. [71] reported 6- to 15-fold lower concentrations of PFHpA, PFOA and PFNA in snow from the Lake A catchment (collected in 2008) near the north coast of Ellesmere Island than inland at Lake Hazen [70](Table S3).

In a study of snow collected in 2008 from the Devon Ice Cap (75.2°N, 82.7°W, 2175 m above sea level) in the Canadian Arctic (a follow-up to the original study by Young et al. [72]), Macinnis et al. [73] reported concentrations for PFOA (0.07–0.68 ng/L), 0.030–1.42 ng/L for PFNA, and 0.01–0.23 ng/L for PFDA, which were similar to those at Station Nord. Macinnis et al. [73] found that PFBA was the predominant PFCA, ranging in concentration from 120 to 2000 pg/L, representing from 7 to 70% of ΣC_4-C_{12}-PFCAs). PFBA was not correlated with other PFCAs suggesting a source other than atmospheric oxidation of fluorotelomer compounds. They suggested that the likely source was non-fluorotelomer gases which are known to degrade to PFBA including hydrofluorocarbons (HFC), such as HFC-329 ($CF_3(CF_2)_3H$), and hydrofluoroethers (HFE), such as HFE-7100 ($C_4F_9OCH_3$) and HFE-7200 ($C_4F_9OC_2H_5$) [74]. Concentrations of PFSAs in Devon Ice cap snow were generally much lower than PFCAs, with PFBS predominating (54–226 pg/L). Macinnis et al. [73] also reported perfluoro-4-ethylcyclohexane sulfonate (PFECHS) in Devon Ice cap snow (<0.3–20 pg/L). This was the first report confirming atmospheric deposition of this PFAS in the Arctic. It is reported to be used in aircraft hydraulic fluids [75].

Codling et al. [76] measured PFASs in the snowpack of a boreal forest in northern Sweden (64°N, 19°E). The highest median concentrations ranged from 0.067 to 0.34 ng/L in the order PFBA > PFPeA > PFBS > PFOA. They found evidence of migration to deeper snow layers as melt progressed and a declining proportion

of PFBA, the most water-soluble PFAS. They also noted that PFBS and the longer chain $C_{10}-C_{12}$ PFCAs were retained in the melting snow pack. PFBA and PFPeA concentrations in northern Sweden were similar (within a factor of 2) to those found in snow from the Longyearbyen area while C_7-C_{10} PFCA concentrations were more than 5-fold higher at the Swedish site. PFCA concentrations in snow at Station Nord were similar to those found in northern Sweden [76] and Svalbard (Longyearbyen; [60]) but lower than for snow from the Lake Hazen (Ellesmere Island) catchment. Overall, the levels of C_4-C_{10} PFCAs in snow in the Canadian Arctic and northern Greenland (Lake Hazen, Devon Ice Cap, Station Nord) appear to be higher than in the European Arctic (Svalbard, northern Sweden).

A study of PFAS compounds in snow in northern Norway (Tromsø area) revealed that ΣPFCAs (C_4-C_{12}) was the major group present with concentrations ranging from 0.294 to 5.206 ng/L. PFHpA dominated the samples with a maximum concentration of 3.866 ng/L. Only two of the PFSAs (PFBS and PFOS) were detected, and levels of PFBS varied from below the limit of detection to 0.178 ng/L. Concentrations of particle-bound PFAS were low and often below the detection limit. Particle-associated PFBA showed levels up to 0.383 ng/L, although the detection frequency was highest in samples near the city of Tromsø (Bertrand et al., Lancaster University, Lancaster, UK, unpubl. data). The flux of PFCAs to the snowpack seemed to be related to temperature at the time the snow fell, with higher temperatures linked to larger fluxes. This might be due to greater scavenging of PFCAs from air with wet snow and/or that snowflakes formed a larger surface area during milder conditions, which could facilitate surface-mediated photochemical transformation of PFAS precursors (Bertrand et al., Lancaster University, Lancaster, UK, unpubl. data). Hence, snowfall at mild temperatures can lead to a significant contribution of PFAS to the snowpack, where it accumulates through winter to be released to the surrounding environment at snow melt.

Xie et al. [60] measured neutral PFASs in snow collected from Ny-Ålesund (Svalbard) and determined their air-snow exchange fluxes. In snow, 8:2 FTOH was the predominant species accounting for 45% of ΣFTOHs. Concentrations of ΣFTOHs in snow ranged from 0.218 to 0.507 ng/L (mean: 0.369 ng/L). The composition of PFASs was different in snow compared to air; 10:2 FTOH, MeFOSE and 12:2 FTOH were the three most abundant species in snow. For FTOHs and fluorotelomer acrylates (FTAs), the total air-snow exchange fluxes were positive indicating exchange back into the atmosphere, whereas fluxes for FOSEs were negative [60].

2.2. Terrestrial environment

There are currently fewer data available for the terrestrial environment than for the freshwater and marine environments. Müller et al. [77] detected low levels of PFASs in plants, moss and lichen from locations in the northern Yukon and from western Nunavut near Bathurst Inlet, Canada (Table S4). Overall, average concentrations of ΣPFCAs in lichen in the two regions were similar. However, the PFAS patterns differed widely between vascular plants and lichen (Fig. 10). Highest average ΣPFCA concentrations were found in lichen (*Cladonia mitis/rangiferina* and *Flavocetraria nivalis/cucullata*) (0.17–0.22 ng/g ww) and Arctic willow (*Salix pulchra*) (0.08–0.26 ng/g ww) and were similar in both areas. The PFCA composition differed among vegetation types. PFOA was the predominant PFCA in all plants (grass, sedge, willow and moss, *Rythidium rugosum*), while both lichen species showed a predominance of odd-carbon-chain lengths ($C_8 < C_9$, $C_{10} < C_{11}$, $C_{12} < C_{13}$).

Müller et al. [77] measured PFASs in liver and muscle of caribou (*Rangifer tarandus*) from the Porcupine herd in the northern Yukon and the Bathurst herd from NWT-western Nunavut as part of a terrestrial food web study. Highest PFAS liver concentrations were
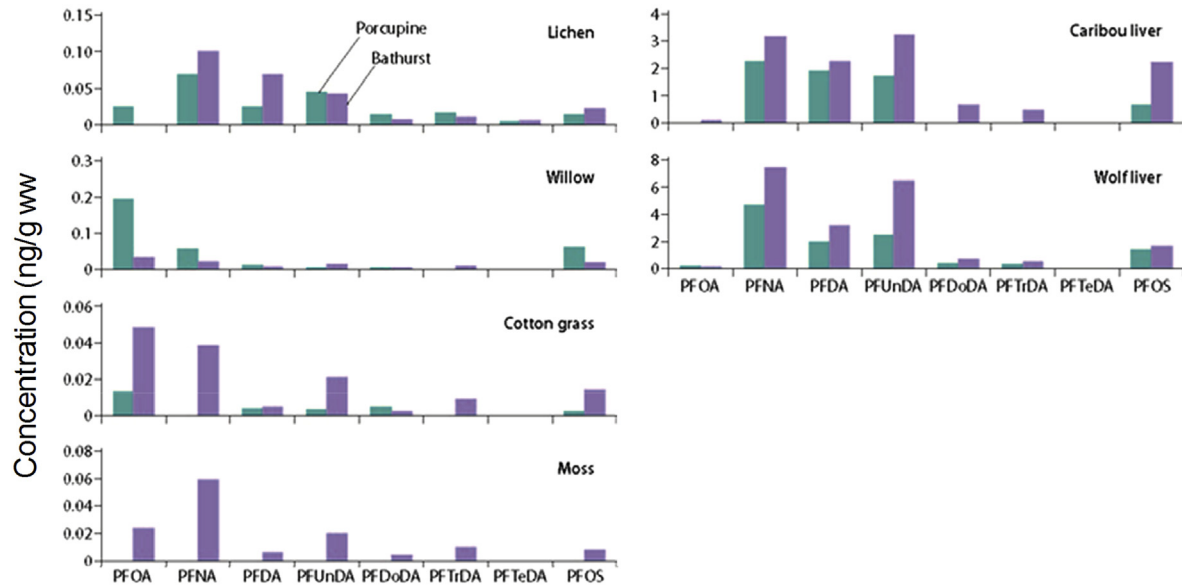
Fig. 10. PFCA homologue composition of vegetation, caribou and wolf in the Porcupine (Yukon) and Bathurst (Nunavut) caribou summer grazing areas [77]. Grass refers to cotton grass. Wolf and caribou homologue distribution shown here are from liver samples.

found for PFNA (2.2 ± 0.2and 3.2 ± 0.4 ng/g ww for the Porcupine and Bathurst herds, respectively) followed by PFDA (1.9 ± 0.1 and 2.2 ± 0.2 ng/g ww, respectively) and PFUnDA (1.7 ± 0.1 and 3.2 ± 0.2 ng/g ww, respectively). In general, ΣPFCAs and PFOS were about 2-fold higher in the Bathurst herd. This geographical difference may be due to the more remote location of the Porcupine herd relative to PFAS sources although concentrations in lichen were similar (Fig. 10). Differences in diet may also play a role.

Müller et al. [77] evaluated the biomagnification of PFAS in vegetation to caribou and wolves (*Canis lupus*). The biomagnification factors were tissue-specific and highest for C_9–C_{11} PFCAs (2.2–2.9) and PFOS (2.3–2.6) although these factors were lower compared to earlier studies of the marine food web [77]. Not surprisingly, wolf liver contained the highest concentrations of ΣPFAS, followed by caribou liver and ΣPFCAs/PFOS ratios were >10 in vegetation and 5–10 in mammals (Fig. 10). Muscle and kidney contained 10–20 times lower concentrations (Table S4).

The distribution pattern of PFAS compounds varied with tissue and trophic level. PFOA predominated in plants (willow, grass) while PFNA and PFDA predominated in lichen and moss. PFNA and PFUnDA predominated in caribou and wolf. The odd-carbon-chain PFCAs predominated in caribou, wolf, grasses and moss.

PFAS concentrations in terrestrial animals are summarized in Table S4. PFNA predominated in wolf, moose and caribou liver among terrestrial mammals in northern Canada, and in reindeer (*Rangifer tarandus*) from Svalbard [77]. PFNA was followed by PFOS, PFUnDA, PFDA, perfluorotridecanoate (PFTrDA) and PFHxA as the predominant PFASs, although the pattern varied between these animals.

Concentrations of PFCAs in muscle tissue from Svalbard reindeer were dominated by the long-chained compounds perfluorododecanoate (PFDoDA) and PFTrDA, while PFNA and PFTrDA predominated in liver closely followed by PFOS and PFHxS (Fig. 11). Long-chain PFCAs were also detectable in reindeer fat. Caribou muscle from the central Canadian arctic has high proportions of PFNA, PFDA and PFUnA [77]. No results for caribou fat are available from Canada or Greenland. However, comparing data for liver, the Svalbard reindeer differ in having much higher PFTrDA levels (Fig. 11). PFOS was the PFAS with the highest concentration

(1.42 ng/g ww) in reindeer liver from southern Greenland followed by PFNA (0.84 ng/g ww) and PFUnDA (0.45 ng/g ww) [78]. A long-term study of reindeer and moose (*Alces alces*) during 1987–2006 from Sweden reported PFOSA, PFOS and PFOA above detection limits in reindeer muscle, but not at concentrations that could be quantified [79].

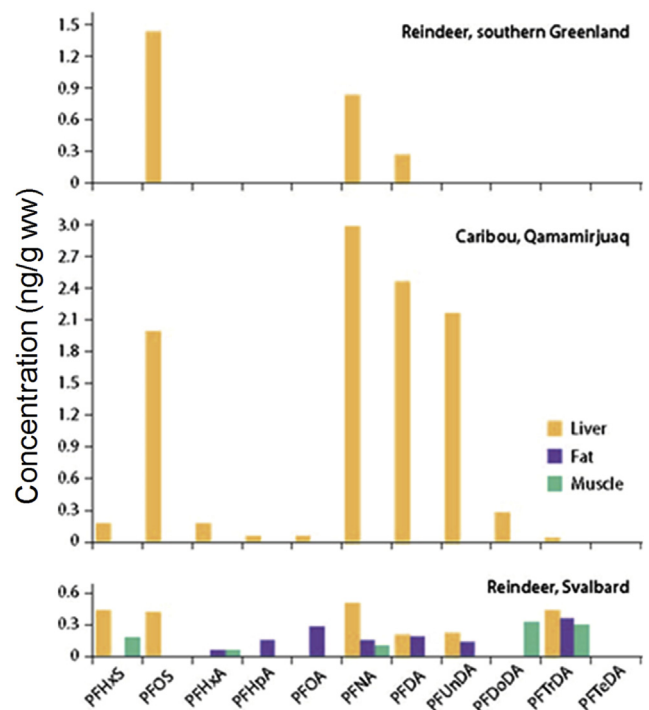Norwegian moose livers showed a slightly different pattern,

Fig. 11. Comparison of PFAS profiles (ng/g ww) in caribou liver from the Qamamirjuaq herd (near Arviat, Canada) and reindeer liver, fat and muscle from Svalbard. Results from southern Greenland from Bossi et al. [78], Qamamirjuaq data from Gamberg and Muir (M. Gamberg and D. Muir, Gamberg Consulting, Whitehorse, Canada 2015, unpubl. data), and Svalbard data from Carlsson et al. [80].

with PFOS as the predominant compound, followed by PFNA, PFUnDA and PFDA. The PFAS pattern varies with species; liver from wood mice (*Apodemus* sp.) and shrews (*Soricidae*) from the same area had the highest concentrations of PFTrDA, followed by perfluorotetradecanoate (PFTeDA) and PFUnDA and PFOS [81].

Liver samples from Arctic fox (*Vulpes lagopus*) from Svalbard had the same PFAS distribution pattern as the Norwegian moose, except that PFTrDA was present at similar levels to PFUnDA [82]. Concentrations of PFAS in these foxes varied with body condition. Lean foxes had higher wet weight concentrations of ΣPFSA and ΣPFCA in their adipose tissue compared to fat foxes. Availability of terrestrial food such as reindeer carcasses contributes to lower concentrations of long-chained PFCAs in arctic foxes compared to when the foxes are feeding on a more marine diet. Routti et al. [83] determined PFASs in livers of arctic foxes from Svalbard collected over the period 1997–2014. PFAS patterns are in agreement with previous studies on arctic foxes showing that PFOS is the dominant PFAS followed by odd chain length PFCAs. Precursors to PFCAs such as 6:2 and 8:2 FTSAs (33% and 13% detection frequency, respectively) were also detected in these foxes [83].

Larter et al. [84] analyzed PFASs in the liver of moose from the Dehcho Region in the southwestern Northwest Territories. PFASs were the major POPs in moose liver ranking ahead of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs): ΣPFCAs > PFOS > ΣPCBs > ΣPBDEs. Concentrations of PFASs in moose liver ranged from 2.2 to 4.3 ng/g ww for ΣPFCAs and the C_9 to C_{11} PFCAs (PFNA–PFUnDA) predominated.

Bossi et al. [78] reported PFAS concentrations in liver samples from terrestrial biota (birds and mammals) from Greenland. Samples from ptarmigan (*Lagopus muta*; western Greenland), reindeer (southwestern Greenland) and muskox (*Ovibos moschatus*; eastern Greenland) were analyzed. PFAS concentrations in ptarmigan were mostly below detection limits but PFNA was detected in all samples analyzed [78]. The mean PFNA concentration was significantly higher in ptarmigan from Nuuk than from Qeqertarsuaq (ANOVA, $p < 0.01$). PFNA, PFDA, PFUnDA and PFDoDA were detected in all samples. Interestingly, ptarmigan had much lower concentrations and different patterns of PFCAs than piscivorus seabirds where long-chain C_9–C_{13} PFCAs predominate in liver. In the three muskox liver samples, PFNA, PFDA and PFUnDA were detected at similar concentrations, ranging from 1.18 to 5.25 ng/g ww. PFDoDA and PFTrDA were found at concentrations between 0.21 and 0.72 ng/g ww, whereas PFTeDA was not detected (Table S4). Although sample size was limited, muskox from East Greenland had much higher concentrations of PFNA, PFDA, PFUnDA, PFDoDA and PFTrDA than reindeer from southwest Greenland.

2.3. Freshwater environment

2.3.1. Lake and river waters

Studies on PFASs in water and sediments from Arctic freshwater environments are very limited. As of 2015, results for water and sediment samples were available for the Canadian Arctic, Faroe Islands, and Norway (Svalbard) and are summarized in Table S3.

Stock et al. [85] determined PFASs in water from Amituk, Char, Merretta and Resolute Lakes on Cornwallis Island in Arctic Canada. Further studies were conducted on PFASs in these and other lakes on Cornwallis Island by Lescord et al. [86]. The analyses included C_4–C_6 PFCAs as well as the first report of a new cyclic perfluorinated alkyl sulfonate (perfluoro-4-ethylcyclohexane sulfonate) (PFECHS) in the Arctic (Table S4). Concentrations of all PFAS congeners in the remote lakes on Cornwallis Island were very similar, with mean concentrations of PFOA of 0.20–0.27 ng/L. PFAS ratios of PFOA:PFNA and PFDA:PFUnDA in Amituk and Char Lakes were generally consistent with ratios observed in Arctic glacial ice

caps by Young et al. [72], and in precipitation from rural and remote sites in North America [87] suggestive of a common atmospheric source.

The study by Lescord et al. [86] compared PFAS concentrations in Resolute and Meretta Lakes, which Stock et al. [85] had shown to be contaminated with high levels of PFOS, with other lakes near Resolute Bay. Other PFASs were identified in Resolute and Meretta Lakes including PFECHS and fluorotelomer sulfonates (FTS) (4:2-, 6:2-, and 8:2 FTS). Stock et al. [85] concluded that the pattern of contamination observed in water samples from Resolute and Meretta Lakes, with extremely high levels of PFHxS and PFOS, in addition to PFHpA and PFOA, relative to background sites (e.g. Char Lake) was consistent with the use of aqueous film-forming foams (AFFFs). PFASs found in AFFFs have been detected following spills at airports [88] and in groundwater at military bases in Canada [89] and the United States [90,91]. The 4:2 and 8:2 FTSs are also be associated with AFFFs [92], while PFECHS is used as an abrasion inhibitor for hydraulic fluids in aircraft and thus also associated with airport emissions [93].

PFBA was the most prominent PFAS in water from Lake Hazen, a large lake on Ellesmere Island, as well as in snow from the lake surface and nearby catchment [70]. The depth profile for PFASs in Lake Hazen (Fig. 12) showed a sharp increase in the near surface waters during the snow melt indicative of inflow of snow melt water having much higher concentrations than deeper waters. Under ice free and completely ice covered (pre-melt) conditions concentrations were similar over the depth profile (Fig. 12).

Lowest PFAS concentrations in Arctic lakes were found in Lake A, on northern Ellesmere Island [71] and in Pingualuk, an isolated crater lake in northern Québec [94] (Table S3). However, concentrations of PFOA, PFNA, PFDA and PFOS in Lake A catchment snow were similar to spring-summer concentrations in accumulated snow from the Devon Ice Cap (8° in latitude south of Lake A)
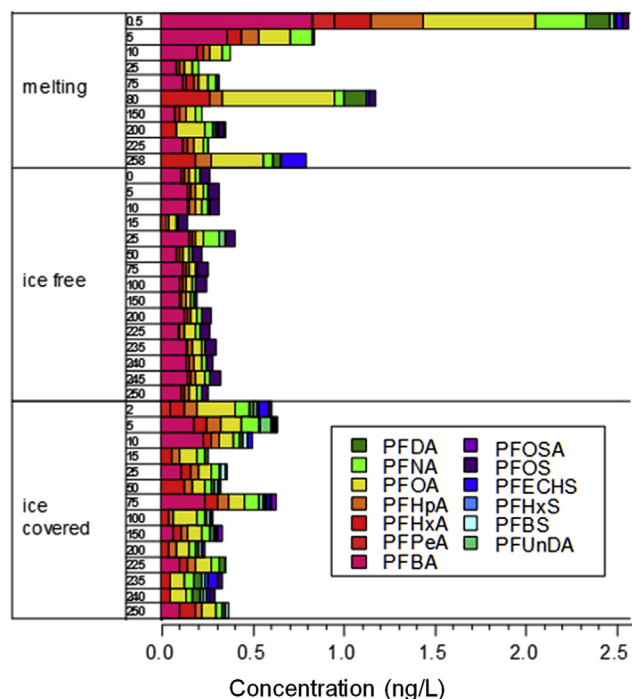

Fig. 12. Depth profiles of PFASs in the water column of Lake Hazen during snowmelt period (late May/early July), ice-free period (late July), and ice-covered period (mid May). Based on samples collected prior to melting in May 2013 (2–250 m), during snowmelt on June 2012 and late May 2014, as well as during ice-free open water period on July 2015 [70].

sampled in spring 2015 [95] and to "light" snow collected at Lake Hazen in 2013-14 [70]. Thus, the loading of PFASs from snow melt is likely to be relatively similar over a large area of the Canadian archipelago. The Lake A, Pingualuk, and Lake Hazen snow sampling was carried out prior to snowmelt freshet which would deliver snow-bound PFAS as illustrated for Lake Hazen in Fig. 12.

Four lakes on the Faroe Islands used for drinking water supply had low C_4–C_{10} PFCA concentrations [96], broadly similar to those in remote lakes on Cornwallis Island (Table S3).

Kwok et al. [69] reported a wide range of PFCAs in river water at Longyearbyen. PFBA and PFPeA were predominant in the surface water samples. Approximately 2–3 times higher average PFAS concentrations were detected in both surface snow and water samples collected from the locations downstream of the glacier near Longyearbyen compared to glacial water. Because the river flows near the town of Longyearbyen there may be an influence of local sources in addition to long-range atmospheric transport. St. Louis and Muir [97] reported PFCAs in water from the Abbe River, which drains a glaciated region near Lake Hazen (Ellesmere Island). PFBA and PFOA were the prominent PFCAs (Table S3).

2.3.2. Lake sediments

Limited data are available for PFASs in Arctic lake sediments and concentrations in lakes unimpacted by local development are very low with ΣPFCAs <1 ng/g dry wt (Table S3). C_6–C_{11}–PFCAs predominated in remote lake sediments in northern Norway and in the Canadian Arctic [81,86,98]. Of the 10 lakes for which data are available, lowest concentrations of ΣC_6-C_{11}-PFCAs were found in surface sediments (0–1 cm) of Lake Hazen in northern Ellesmere Island (0.10 ng/g dw). An exception are Resolute and Meretta Lakes located near the Resolute Bay (NU) airport, where PFOS concentrations averaged 27.8 and 48.5 ng/g, respectively. PFOA and PFNA concentrations in Meretta and Resolute surface sediments were similar (averaging 1.8 and 1.5 ng/g, respectively) but were 6 to 10-fold higher than in remote lakes 5–14 km from the airport [86].

2.3.3. Freshwater fish

A large number of measurements have been made on PFASs in freshwater fish, particularly in the Canadian Arctic and Norway, and recently in Greenland and the Faroe Islands [78,81,86,99]. Detailed results for PFCAs are provided in Table S5. The long-chained (C_9–C_{12}) PFCAs predominate in freshwater fish, however the pattern differs in European Arctic compared with Greenland and Canada. This may be due in part to use of fish liver in Greenland, the Faroes and Svalbard versus fish muscle in Canada (Fig. 13).

Muscle from Arctic char (*Salvelinus alpinus*) collected in 2010 and 2013 from Lake Linnévatnet, a remote lake near Barentsburg and Longyearbyen on Svalbard showed a different pattern of PFASs compared to other locations. PFBA and PFUnDA were the most prominent congeners detected in more than 50% of samples (Fig. 13). PFOA was present in lower concentrations than expected; 0.05 ng/g ww in muscle from Arctic char collected in 2010 and 0.02–0.04 ng/g ww in 2013. However, PFOA was only detected in 20–25% of Arctic char muscle samples. Three years earlier (in 2010), PFUnDA was the most prominent PFAS in char samples from the same lake, together with 6:2 FTS and PFNA. There is insufficient information about local PFAS sources on Svalbard to explain the different PFAS patterns compared to the Norwegian mainland. Longer-chain (C_9–C_{14}) PFCAs predominated in Arctic char from Greenland and the Faroe Islands (Fig. 13).

Concentrations of PFOS, PFDoDA and PFTrDA were similar in char from Lake á Mýranar (Faroe Islands) and the unnamed lake in southern Greenland, whereas concentrations of PFNA, PFDA and PFUnDA were higher in Greenland. In contrast, landlocked char from the Canadian Arctic had overall lower concentrations of all

PFASs than in the Faroe Islands or Greenland and much lower proportions of C_{12}–C_{14} PFCAs.

Low levels of PFASs in lake trout (*Salvelinus namaycush*) have been reported from Great Slave Lake (M. Evans and D. Muir, Environment Canada, Saskatoon, Canada 2015, unpubl. data; Muir et al. [99]), Lake Laberge and Kusawa Lake in Canada (Muir et al. [99]; Stern et al. [100]) and in brown trout (*Salmo trutta*) from the Faroe Islands [78] and Telemark, Norway [81]. PFCAs predominated in all trout samples (Fig. 14), similar to the pattern found in landlocked char. The pattern differs between the European Arctic and Canada, with C_9 to C_{14} PFCAs predominating in Norway, the Faroe Islands and southern Greenland samples (Fig. 14). The C_{13} and C_{14} PFCAs were not detected (<0.01 ng/g ww) in lake trout from northern Canada. The pattern in brown trout from Telemark was: PFTrDA < PFUnDA < PFDoDA (and PFOS). However, none of these PFCAs were above the detection limits in the water or sediment, and PFTrDA was only detected in 30% of brown trout samples [81]. For those samples in which it was detected, the levels of PFTrDA were higher (mean 16.3 ng/g ww) than for other PFCAs, which varied from 5.07 ng/g ww (PFDoDA) to 0.09 ng/g ww (PFOA). Mean ΣPFCA was 32 ng/g ww, which is slightly higher than ΣPFCA reported in liver (mean 5.9–21.0 ng/g ww) from burbot (*Lota lota*) in the Northwest Territories, Fort Good Hope, Canada [99,102].

PFCAs were also prominent components of total PFASs in burbot liver from Fort Good Hope [102,103] and Great Slave Lake (Muir et al. [99]; Evans and Muir, unpubl. data). PFNA was the predominant PFCA (2.0 ng/g in burbot from both locations). However, burbot liver from Great Slave Lake differed from lake trout muscle by having detectable long-chain (C_{13}–C_{16}) PFCAs with mean concentrations of 0.05–0.25 ng/g ww (Table S5).

PFASs were determined in sea-run char muscle from Cambridge Bay, Pond Inlet and Nain in Arctic Canada (Evans and Muir, unpubl. data). PFOS was detectable in all samples along with PFOA, PFNA, PFDA, PFUnDA, PFDoDA, and PFTrDA (Table S6). PFOS and ΣPFCA concentrations were very low, averaging 0.04 ± 0.014 and 0.12 ± 0.03 ng/g ww, respectively, at Cambridge Bay and 0.07 ± 0.06 and 0.12 ± 0.15 ng/g ww, respectively, at Pond Inlet. The range in PFAS concentrations in sea-run char was 2- to 3-fold lower than in landlocked Arctic char in Lake Hazen and Amituk Lake, which are remote lakes (Table S5). This difference may be due to the fact that landlocked char are older and have slower growth rates than sea-run char.

2.4. Marine environment

2.4.1. Seawater

PFASs are globally distributed in the marine environment [46]. The movement of PFASs, from coastal areas influenced by urban emissions, to sub-Arctic and Arctic Ocean waters, was illustrated by Ahrens et al. [104], who found C_6–C_{10} PFCAs averaging about 700 pg/L in coastal seawater of southern Norway and at detection limits (~10 pg/L) in the open Norwegian Sea. Zhang et al. [45] have modelled the inflow of PFOS to the Arctic Ocean and their results likely apply to other ionic PFASs. Overall, ocean currents and related dilution effects have a crucial influence on PFAS distribution in seawater, in which industrial and coastal areas and atmospheric deposition are considered as sources of PFASs, and ocean waters are important as sinks [43] and for transportation of these compounds [104].

Yeung et al. [47] determined C_6–C_{12} PFCAs and C_4–C_{10}–PFSAs, MeFOSAA, EtFOSAA, and FOSA in the Arctic Ocean including deep ocean and shelf waters. PFASs were generally only detectable above 150 m depth in the polar mixed layer (PML). Vertical profiles at 4 locations (Amundsen Basin and Nansen Basin) showed that PFOA and PFOS were the predominant PFASs, averaging 50 and 47 pg/L,
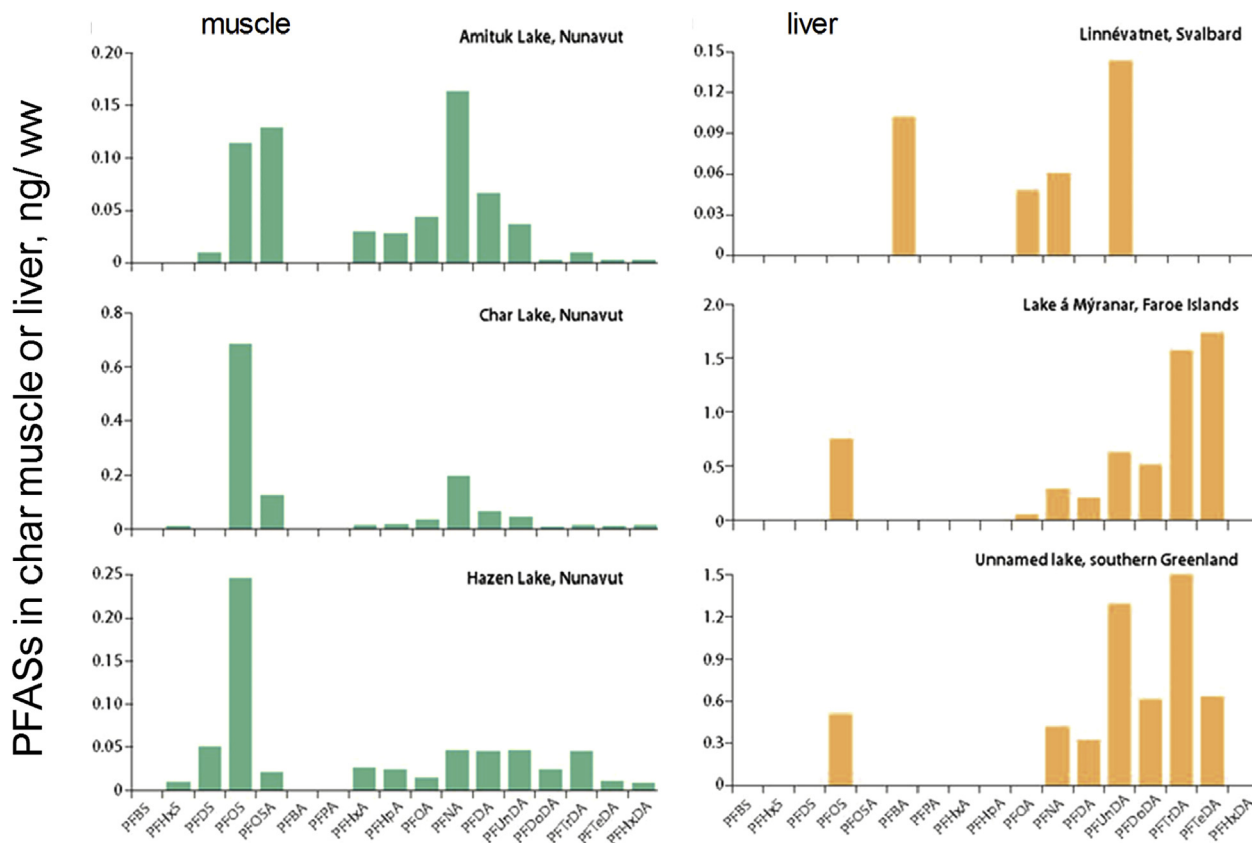
Fig. 13. Comparison of PFAS concentrations and profiles in landlocked Arctic char from the Canadian Arctic, Svalbard, Faroe Islands and southern Greenland. Results from Canada are for muscle while liver was analyzed at the other sites. All data are for the period 2010–2013 (Table S5). Data from Muir et al. Environment Canada, Burlington, Canada, unpublished data 2015, Garsjø [101], and Bossi et al. [78].

respectively. PFBS (40 pg/L), PFNA (39 pg/L), PFHxA (37 pg/L) and PFHpA (35 pg/L) were widely detectable. Concentrations of PFASs in Alaskan continental shelf waters were similar to the ocean with PFOA predominating in almost all samples (detection frequency: 100%; mean 44 ng/L, median 42 ng/L) (Fig. 15). A wider range of PFASs (PFUnDA.

FOSA, and EtFOSAA) were detected in melt pond water and snow samples, collected at stations over the Nansen and Amundsen Basins, compared to seawater from the Arctic shelf and central basin illustrating the importance of atmospheric deposition inputs to surface waters. Other reports for PFCAs and PFBS in the Arctic Ocean and adjacent waters show higher levels than reported by Ahrens and co-workers with the sum of C_6–C_{10} PFCAs (the PFASs that have been determined in all studies) averaging 80–245 pg/L for the 9 studies available as of December 2018 (Table S6). Benskin et al. [22,106] found that PFHxA, PFHpA and PFOA were the major PFASs in seawater in the Canadian Arctic Archipelago and Beaufort/Chukchi Sea waters. However, those studies did not include PFBA and PFPeA. More recent measurements of PFCAs show that PFBA and PFPeA are the major PFAS in seawater at Barrow Strait in the central Canadian Arctic Archipelago [108] and in Chukchi Sea waters [56] with concentrations 2- to 3-fold higher than PFOA. Kwok et al. [69] also found PFBA to be the predominant PFAS in Adventfjorden/Isfjorden, Svalbard. However, PFBA was not reported in two seawater studies that sampled the Greenland Sea and Greenland coastal waters [33,109]. The short-chain sulfonate, PFBS is also widely detected [56,69,104]. In Adventfjorden/Isfjorden, PFPeA was 5-fold higher than PFOA [69], however, this sampling site may have been influenced by the nearby town of Longyearbyen.

Analysis of PFBA in water is challenging due to matrix effects, possibly from early eluting natural organic acids [110].

PFAS concentrations were generally higher in Greenland coastal waters than in open ocean measurements in the northern North Atlantic analyzed in the same studies [33,109]. This may reflect the influence of freshwater inputs to these nearshore waters, as well as to sampling during the period of ice melt. In the case of the southern Beaufort Sea and Canadian Arctic Archipelago, there are major inflows from the Mackenzie, Coppermine and Back Rivers which peak in June and July [22,111,112]. Thus, geographical trends, if any, are obscured by the influence of seasonality. All ship-based sampling has been conducted in open waters or with limited ice cover.

Bertrand et al. [68,113] showed that PFAS concentrations are higher in ice than in the underlying seawater in samples from the Canadian and European Arctic (Fig. 16). In the southeastern Beaufort Sea ΣPFCAs (C_5–C_{12}) were similar in snow and ice but about 4-fold higher than in water. In the Barents Sea near Svalbard, PFOA and other PFCAs were 5- to 10-fold higher in ice than seawater (Fig. 16). Thus, ice is a source of PFASs to surface waters during spring-summer in polar regions while sealing off surface waters from precipitation inputs during winter.

Ongoing measurements of PFOA and other PFASs in the Canadian Arctic Archipelago have shown that concentrations are often elevated in surface seawaters during ice melt each year [108]. This is illustrated in Fig. 17 for depth profiles of PFCAs (sum of C_6–C_{10}) in waters from Barrow Strait near Resolute collected in May–June 2007, 2008 and 2010.

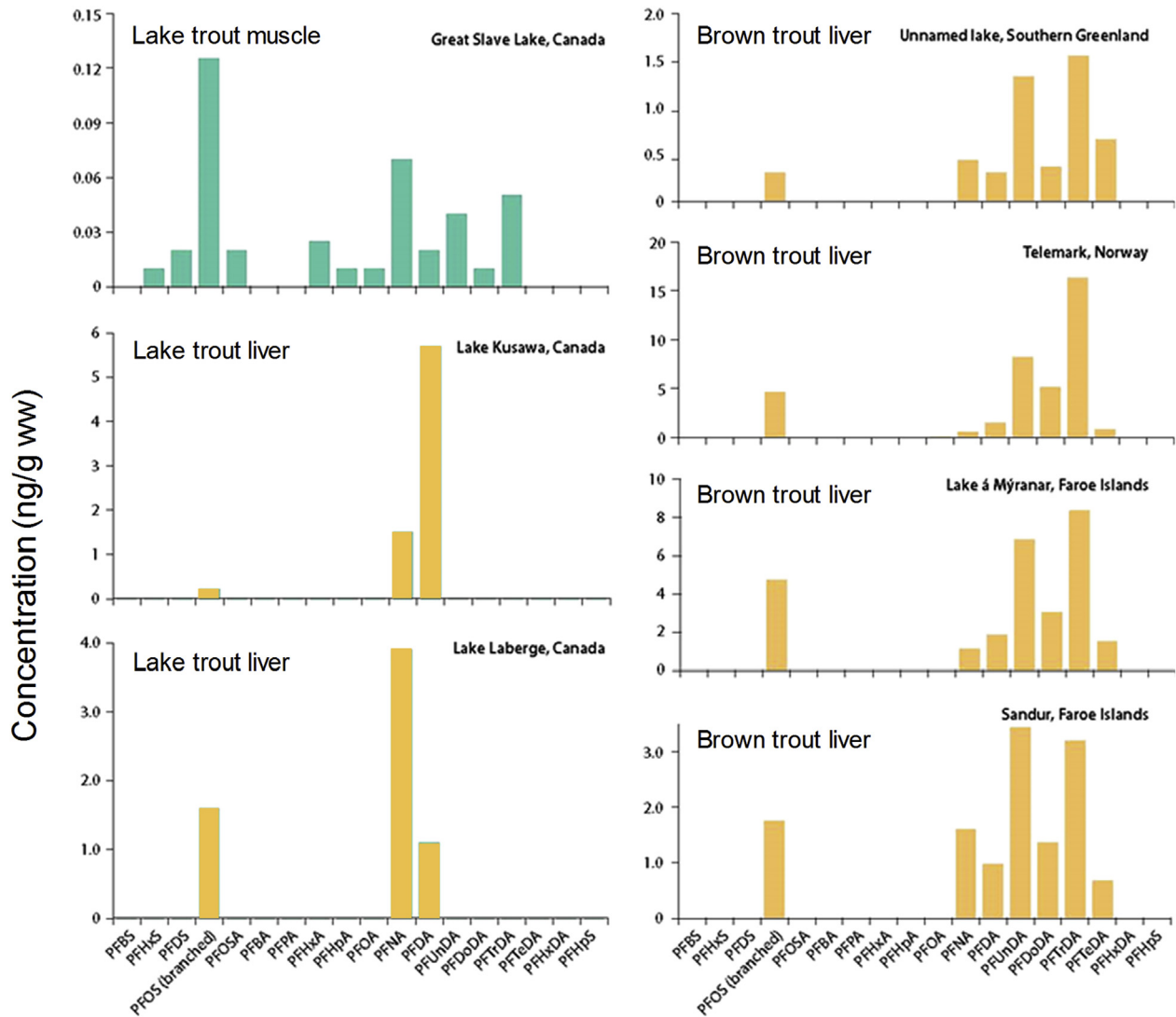Benskin et al. [22] measured PFOA isomers in seawater from the

Fig. 14. Comparison of PFAS concentrations and profiles in brown trout (liver) from northern Norway and the Faroe Islands, as well as lake trout muscle from Great Slave Lake, and lake trout liver from Kusawa Lake, and Lake Laberge in Canada. Results are from Muir et al. [99]), Norwegian Environment Agency [81]); Stern et al. [100], Bossi et al. [78] and M. Evans and D. Muir (Environment Canada, Saskatoon, Canada. unpubl.).

Canadian Arctic Archipelago and Beaufort/Chukchi Seas. They found a distinct spatial trend whereby PFOA in seawater originating from the Atlantic was predominantly historic (up to 99% ECF), whereas water in the Archipelago along the ship transect from Barrow Strait to the Bering/Chukchi Seas (Figs. 15 and 18) had a significant telomer PFOA contribution indicating that it was predominantly of contemporary origin (Fig. 18A). The % ECF was also significantly correlated with salinity ($r = 0.58$, $p < 0.01$; Fig. 18B) but not with seawater temperature (Fig. 18C). This suggests that water collected in the Archipelago is more influenced by riverine discharges, which would presumably contain PFOA derived from an atmospheric source (i.e. FTOH). Summer salinity gradients in the Arctic Ocean show a major influence of freshwater inputs in the southern Beaufort Sea and in the southern Archipelago waters [111,112] which supports this hypothesis.

Total extractable organofluorine (EOF) measurements have been made in non-Arctic seawater, which indicates that the PFASs so far identified represent only a small proportion of the EOF (10−40%) [114]. The majority of the EOF has not been identified, which suggests the presence of other PFASs in addition to the known PFASs.

No EOF measurements appear to have been conducted on Arctic seawaters.

2.4.2. Marine invertebrates

Invertebrates, including copepods and ice amphipods, were included in food web studies conducted in the mid-2000s and reviewed by Butt et al. [4]. There are very few recent studies of PFASs in marine invertebrates and plankton. Recent studies of shrimp from northern Norway revealed that they contained higher PFAS levels than found in cod liver or in halibut muscle samples from northern Norway (Fig. 19). The reason for difference is unclear and requires further study.

2.4.3. Marine fish

There are few recent studies on PFASs in Arctic marine fish species. Earlier work on marine fish was based mainly on food web studies and was reviewed by Butt et al. [4]. PFAS concentrations were below detection limits in fish from West Greenland, Iceland and the Faroe Islands [96,115,116]. Studies from the Barents Sea (polar cod, *Boreogadus saida*), Beaufort Sea (polar cod) and the
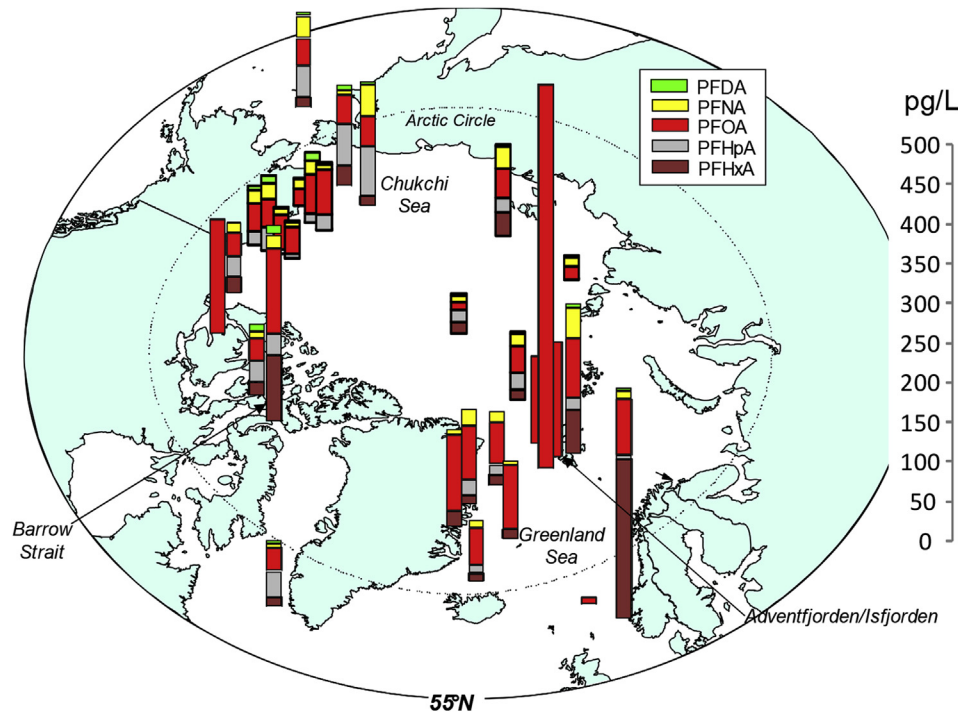
Fig. 15. Reported average concentrations (pg/L) of C_6–C_{10} PFCAs in Arctic seawater based on 9 studies [33,46,47,56,69,81,105–107]. Full details on the data are provided in Table S6. Sampling areas (Barrow Strait, Adventfjorden/Isfjorden) mentioned in the text are identified.
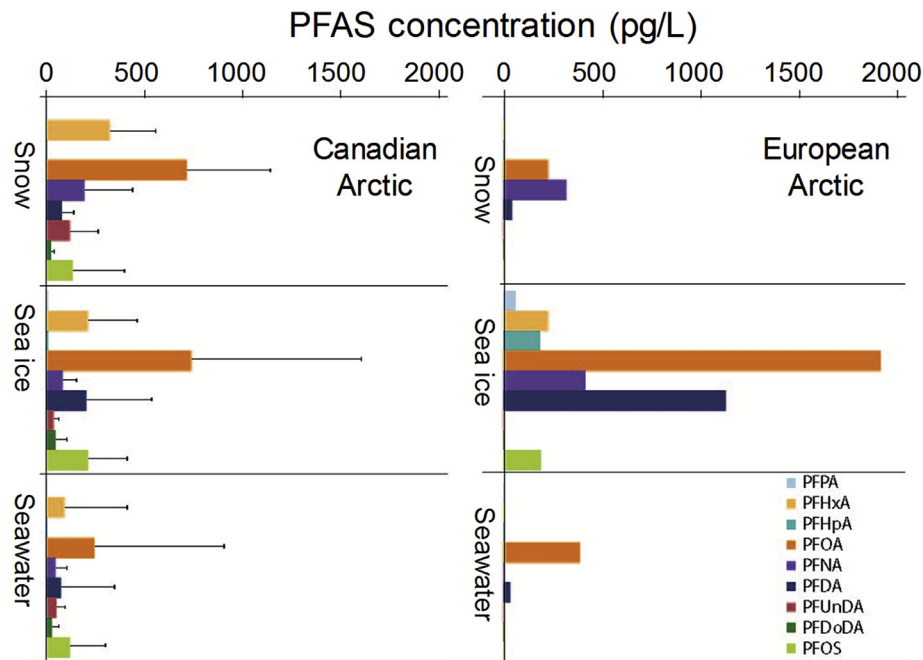

Fig. 16. Enrichment of perfluoroalkyl carboxylates (C_5–C_{12}) (PFPeA = perfluoropentanoate, PFHxA = perfluorohexanoate etc) and perfluorooctane sulfonate (PFOS) in snow/first-year ice of the Canadian Arctic (A) and snow/multi-year ice of the European Arctic (B) relative to underlying seawater. Data from Bertrand et al. [68].

Norwegian west coast (halibut, *Hippoglossus hippoglossus*) have shown low levels of PFASs in fish [115,117,118]. PFOA was not detected in these studies. The levels of PFNA and PFHxA in the Barents Sea samples were 0.19 and 2.06 ng/g ww, respectively [118]).

Braune et al. [119] determined C_6–C_{15}– PFCAs and PFBS, and PFHxS in forage fish from Coats Island (northern Hudson Bay,

Nunavut). PFUnDA and PFTrDA were the most prominent PFCAs, with concentrations (whole fish) of <0.1–0.68 ng/g ww. Arctic cod had the highest concentrations of ΣC_6-C_{15}-PFCAs (1.45, range 1.0–2.1 ng/g ww) followed by sculpins (1.40, range 1.0–1.7 ng/g ww) (Table S7).

Median concentrations of long-chain PFCAs (C_9–C_{13}) in halibut muscle from the Norwegian west coast were 0.07–0.49 ng/g ww,
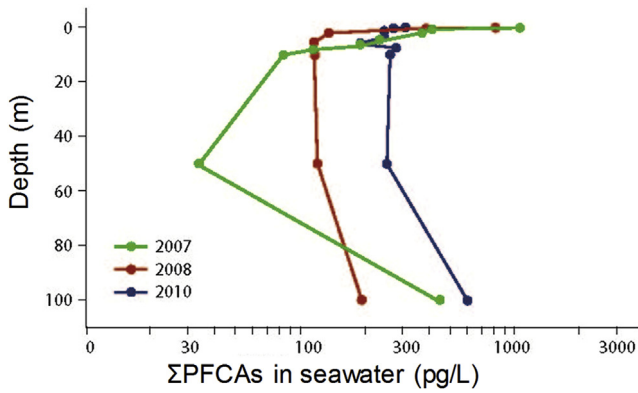
Fig. 17. ΣPFCAs (sum of C_6–C_{10}-perfluorocarboxylates) in seawater from Barrow Strait (Resolute, NU). Sample collection was conducted from the ice during late May to early June each year [108].
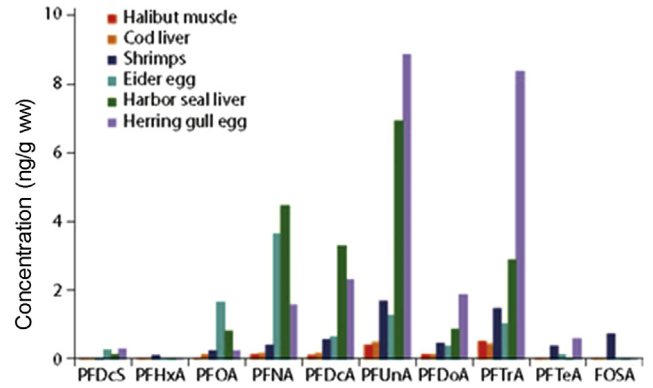

Fig. 19. Concentrations (ng/g ww) of PFAS (not including PFOS) in halibut muscle, shrimp, cod liver, eider and herring gull eggs, and harbor seal liver from northern Norway [81,115].

with PFTrDA and PFUnDA present at the highest levels (Fig. 19). The short-chain PFCAs and PFSAs were below detection limits [115]. Atlantic cod liver from Lofoten (Norway) and Svalbard showed comparable levels of all PFASs analyzed, except for PFOS (0.59 and 0.28 ng/g ww, respectively) and PFTrDA (0.40 and 0.18 ng/g ww, respectively) [81]. These levels were comparable to PFASs in halibut muscle from northern Norway. PFTrDA concentrations in halibut muscle (0.49 ng/g ww) and cod liver from Lofoten were similar, but PFOS was higher in the halibut than the Lofoten cod; 0.95 and 0.59 ng/g ww, respectively [81,115].

2.4.4. Marine mammals

Early studies on spatial and temporal trends of PFASs in Arctic marine mammals were reviewed by Butt et al. [4] and recently temporal trends of PFOS in marine biota have been included in the AMAP temporal trend assessment [120]. This sub-section focuses on more recent reports for PFCAs and on newly detected PFASs. Rigét et al. [121] reported that in ringed seal (*Pusa hispida*) liver

samples collected up until 2010, PFOS was still by far the most predominant compound constituting 92% (West Greenland seals) and 88% (East Greenland seals) of ΣPFASs. However PFCAs constituted an increasing proportion of ΣPFASs in ringed seals from Greenland and in the Canadian Arctic. A similar pattern has been reported for plasma samples from ringed seal from Svalbard; PFOS and PFCAs constituted 60% and 35%, of ΣPFASs, respectively [122]. PFSA and PFCA concentrations in the ringed seals from fjords (sampled from 2002 to 2010) in Svalbard influenced by Atlantic Ocean waters (Kongsfjorden) were approximately 2x higher than those in fjords influenced by Arctic waters ie glacier runoff with limited Atlantic water exchange [122]. This factor was more important than sex, age and body conditions among the seals. This difference may be attributed to the greater influence of glacial meltwater as well as to dietary differences between the ringed seals in the fjords.

Low concentrations of PFASs were found in plasma of walruses (*Odobenus rosmarus rosmarus*) from Svalbard [123] compared to
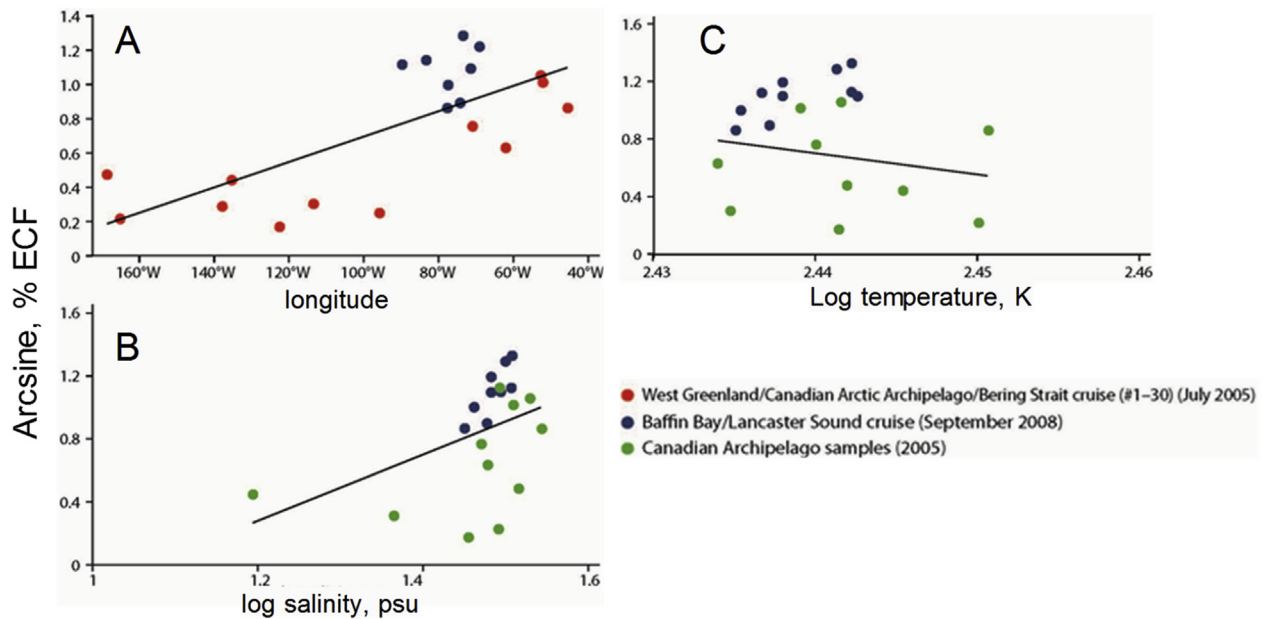

Fig. 18. A. Correlation between % ECF PFOA and longitude. In both graphics, red markers represent the July 2005 West Greenland/Canadian Arctic Archipelago/Bering Strait cruise (#1–30), while blue markers represent the September 2008 Baffin Bay/Lancaster Sound cruise. B. Plot of arcsine (% ECF) versus log (salinity, psu) and, C. Arcsine (% ECF) versus log (water temperature, K) for the same sampling locations. Redrawn from Benskin et al. [22].

harbor seals from the same area, who are pelagic feeders [124]. Walruses feed mainly on mussels, i.e. they are part of a benthic foodweb which may be one of the major reasons for why walruses show much lower concentrations. The difference was a factor of 20 for ΣPFAS between walrus and harbor seal plasma. This is an example of that emerging contaminants may not behave similar to legacy POPs in the environment, which has to be taken into account when fate and environmental impact of CECs are evaluated.

ΣPFCA concentrations in liver from harbor seal (*P. vitulina*) liver from northern Norway were 15 ng/g ww [81]. The longer, odd-chain PFASs predominated (PFUnDA, PFNA and PFTrDA), which is the commonly observed trend among biological samples in all Arctic mammals and is thought to be related to degradation of x:2 FTOHs to both even and odd chain PFCAs via beta- and alpha-oxidation, in the atmosphere as well *in vivo*, with the longer chain length bioaccumulating to a greater extent [38,125].

Data on less reported and new emerging PFASs in ringed seals have been reported by Rotander et al. [126], Muir et al. [127] and Gebbink et al. [128] and are summarized in Fig. 20 and in Table S7. Rotander et al. [126] reported a suite of PFCAs in liver of ringed seals from northwest Greenland and hooded seals (*Cystophora cristata*) from the Greenland Sea (west ice). They found relatively high levels of PFTriDA especially in hooded seals (21 ng/g ww). Gebbink et al. [128] found (C_{14})-PFTeDA and (C_{15})-PFPeCA in ringed seal liver from East Greenland and Muir et al. [129] detected the C_{16}-PFCA in seal liver (Fig. 20).

Gebbink et al. [128] reported the first detection of F53B (also known as 6:2-Cl-PFAES, a chlorinated polyfluorinated ether sulfonic acid) in Arctic biota. This compound has previously been reported in rivers in China (~40 ng/L) and sewage sludge (~2 ng/g) at comparable concentrations to PFOS [130,131]. 6:2-Cl-PFAES was detected in ringed seal liver from East Greenland at 0.045 ± 0.004 ng/g ww (Fig. 20). Muir et al. [132] reported the detection of PFECHS, the cyclic analog of PFOS, in ringed seal liver

from the Canadian Arctic. This perfluoro-compound was previously reported in ice cap snow [73], as well as lake water and landlocked char from Cornwallis Island [86]. In seal liver, PFECHS was present at similar concentrations as PFHxS and PFTeDA. Gebbink et al. [128] analyzed seal liver samples for polyfluoroalkyl phosphate esters (mono- and diPAPs) but did not detect them (<0.5 ng/g wet wt).

Data for new or less studied PFASs in whales from the Norwegian or Canadian Arctic are more limited than for other marine mammals. Butt et al. [4] reviewed results prior to 2010 and additional data for PFASs in beluga (*Delphinapterus leucas*) were reported in the Canadian Arctic POPs assessment [99]. Reiner et al. [133] determined PFASs in liver of beluga from Cook Inlet (southeast Alaska) and the eastern Chukchi Sea. The C_9−C_{12}−PFCAs along with PFOS and PFOSA were major PFASs; PFOA, PFHpA, and PFHxA were detected infrequently (<2% of samples). Total C_9−C_{14}−PFCAs were nearly 3-fold higher in Cook Inlet beluga (70.4 ± 46.1 ng/g wet wt; 2001−2006) than in eastern Chukchi Sea animals (24.0 ± 22.5 ng/g; 1999−2000). Total PFCAs in Cook Inlet beluga were higher than reported for beluga from Hudson Bay [134] or Southern Beaufort Sea [135] (Table S7) mainly due to higher proportions of PFUnDA, PFDoDA, PFTrDA and PFTeDA. Smythe et al. [135] reported concentrations of C_8−C_{12} PFCAs, FOSA and PFOS in 3 Canadian arctic beluga stocks (Southern Beaufort Sea, Cumberland Sound and Southern Hudson Bay). Mean concentrations of ΣC_8-C_{12} PFCAs were similar in all three stocks for samples collected from 2003 to 2013, ranging from 33.3 (range 11.6−163) ng/g ww in Hudson Bay to 35.0 (4.9−132) ng/g ww in Cumberland Sound animals.

Rotander et al. [126] reported concentrations of a suite of PFCAs in liver of whales from the northern North Atlantic, including the Faroe Islands, west Iceland and Greenland, based on samples collected in the mid-2000s as well as older archived samples. PFUnDA was the most prominent PFCA with concentrations ranging from 0.4 ng/g (fin whales, *Balaenoptera physalus*) to 52 ng/g (white-
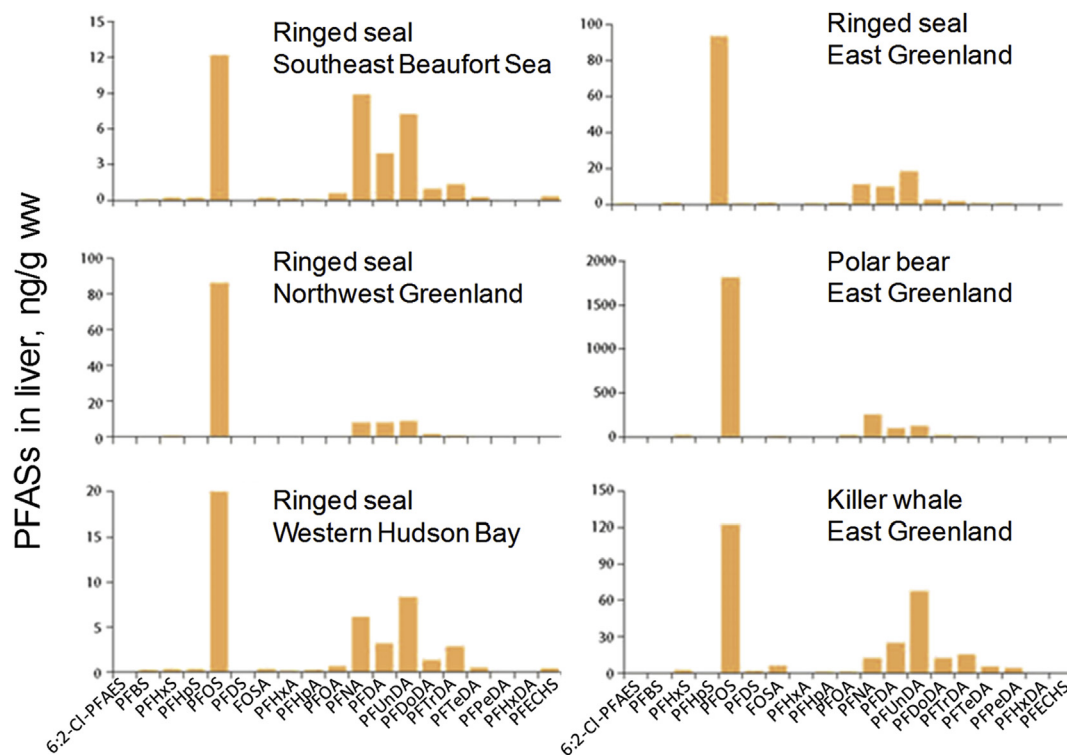

Fig. 20. Concentrations and patterns of a large suite of PFASs in ringed seal liver from northern Canada and east and west Greenland (na = not analyzed). Based on data from Muir et al. [129] and Gebbink et al. [128].

sided dolphin, *Lagenorhynchus acutus*) (Table S7). Gebbink et al. [128] reported a suite of PFASs in liver of killer whales (*Orcinus orca*) collected off East Greenland; the first report for PFASs in this species. Long-chain (C$_9$–C$_{15}$)–PFCAs were prominent contaminants in killer whale liver ranging from 3.4 ng/g (PFPeDA) to 67 ng/g (PFUnDA). Gebbink et al. [128] also detected 6:2-Cl-PFAES in killer whale liver. Levels of PFAES (0.023 ng/g) were about 10-fold lower than in polar bear (*Ursus maritimus*) liver and relatively low compared to PFOA or PFOS.

There is more information available about new or emerging PFASs in polar bears in bears from sub-populations in East Greenland and in southern and western Hudson Bay in Canada compared to the Norwegian Arctic (Svalbard) although a few studies have been published from Svalbard area during the last years. One recent study investigated levels of PFASs in plasma from Svalbard polar bears. PFOS was the predominant PFAS (205 ng/mL plasma), followed by the longer odd-chained PFCAs; PFNA and PFUnDA at 37.6 and 25.5 ng/mL, respectively [81]. The same distribution pattern was observed in a polar bear mother-cub pair study with samples from Svalbard in 1998 and 2008 [136]. Mean levels of PFOS were 432 and 309 ng/mL plasma in the polar bear mothers from 1998 to 2008, respectively, while PFNA was present at 28 and 38 ng/mL plasma in 1998 and 2008 (Table S7). Most of the PFCAs increased over the same period. Maternal transfer is an important exposure pathway for the cubs, although it is less important than for the lipophilic, legacy POPs [136]. Diet is the most important pathway and polar bears with a high marine proportion in the diet have higher PFAS concentrations compared to polar bears with more terrestrial diet [137]. This feature has also been shown in a study on female polar bears offshore versus coastal polar bears on Svalbard. The offshore bears had access to better food quality compared to the coastal bears, but also had around 30–35% higher PFAS concentrations in plasma compared to the coastal bears [138].

Recent studies have focused on environmental/biological factors that impact concentrations of legacy POPs and emerging PFAS in polar bears. One factor with synergistic, negative impact on energy metabolism in combination with e.g. PFAS and POP concentrations is reduced sea ice [139]. Less sea ice may force polar bears towards a more terrestrial diet which decreases their exposure towards PFAS, but, at the same may lead to reduced food quality, which leads to starving polar bears and hence, higher PFAS body burden [137]. In East Greenland polar bears, PFSA and PFCA concentrations were reported to be highest in liver followed by blood > brain > muscle ≈ adipose [140]. Also, liver and blood samples contained proportionally more of the shorter/medium-chain (C$_6$ to C$_{11}$) PFCAs, whereas fat and brain samples were dominated by longer-chain (C$_{13}$ to C$_{15}$) PFCAs. In different brain region samples for the same bears, PFOS and the longer-chain PFCAs (C$_{10}$–C$_{15}$) were found to be significantly and positively correlated with lipid content for all brain regions [141].

C$_9$–C$_{11}$ PFCAs were prominent, with PFNA dominating, in polar bear liver samples collected in 2014 from the southern and western Hudson Bay sub-populations [142]. This is similar to the pattern found in East Greenland bears. In addition to PFOS, PFHxS and several 'Pre-FOS' precursors (precursors to PFOS) were quantifiable, for example, *N*-EtFOSA and FOSA, at low levels [142]. The composition of the PFASs was identical between subpopulations (ΣPFSA = 60–61%, ΣPFCA = 39%, and other FOSAs ∼ 1%), with PFOS and PFNA accounting for 60% and 25% of the ΣPFAS concentrations.

Low ng/g concentrations of C$_4$ perfluorobutane sulfonamide (FBSA) were reported for the first time in polar bear liver [142]. FBSA was detectable at a frequency of 12% in livers of western Hudson Bay bears, but totally non-detectable in liver of southern Hudson Bay bears. This was the first detection of FBSA, a precursor

of PFBS, in an Arctic biota sample. PFBS was also detectable (frequency 53–54%) in the Hudson Bay polar bear liver samples [142]. It has also been shown that ringed seal and polar bear, have been shown to rapidly dealkylate *N*-EtFOSA to FOSA *in vitro* in liver microsomes from these Arctic species [40] and the same pathway probably applies to FBSA. Gebbink et al. [128] reported the first detection of 6:2-Cl-PFAES in polar bear liver. Concentrations (0.27 ng/g) were about 6-fold higher than in seal liver but relatively low compared to PFOA or PFOS.

PFBA was not detected in plasma of Svalbard bears [136] nor in liver of Hudson Bay bears. However, the cyclic analog of PFOS, PFECHS was quantifiable in almost all Hudson Bay bear liver samples (0.09–1.45 ng/g ww) [142]. Detection is consistent with its detection in ringed seals from the same region [132].

Liu et al. [143] detected 3 new classes PFSAs in pooled polar bear liver plasma from Western Hudson Bay and the Southern Beaufort Sea populations using a nontarget high resolution mass spectrometry method (Table 3). Class I were cyclic or unsaturated PFSAs indicating a ring or double-bond in the core structures. Class 2 had unique [C$_2$F$_5$O]$^-$ ions and were characterized by the simultaneous detection of PFSA-specific ions (e.g. [SO$_3$]$^-$ and [SO$_3$F]$^-$). Class 3 included unsaturated ether-, cyclic ether-, or carbonyl-PFSAs. Class 4 consisted of x:2 chlorine-substituted perfluoroalkyl ether sulfonates including F–53B previously reported by Gebbink et al. [128] in polar bear liver, the C6 and C7-homologs were also detected.

2.4.5. Seabirds

Butt et al. [4] previously reviewed spatial and temporal trends of PFSAs in seabirds and therefore only reports published after 2009 are considered here. More recent work has involved analyses of seabird liver or eggs from Norway, the Faroe Islands, and Greenland, as well as Nunavut, and is summarized in Table S7. However, comparing locations is difficult because different species have been sampled. The data are summarized here with an emphasis on the PFCA pattern in liver (Fig. 21).

Miljeteig et al. [145] determined PFASs in ivory gull (*Pagophila eburnea*) eggs from four colonies in the Norwegian (Svalbard) and Russian Arctic islands (Franz Josef Land and Severnaya Zemlya). C$_{11}$–C$_{13}$–PFCAs predominated at all locations and the range in concentration was similar among colonies (Table S7). Lucia et al. [146] analyzed PFOA and PFNA in ivory gull eggs from Station Nord (Greenland) and found that concentrations were significantly higher compared results from Miljeteig et al. [145] implying higher exposure further west. However no data for PFASs in ivory gulls are available from the Canadian Arctic or Alaska for comparison.

Nøst et al. [147] determined PFASs in plasma of black-legged kittiwake (*Rissa tridactyla*) and northern fulmar (*Fulmarus glacialis*) chicks from Kongsfjorden (Svalbard). Median concentrations of total (C$_8$–C$_{14}$)–PFCAs were 3-fold higher in fulmar than kittiwakes. Tartu et al. [148] reported on C$_4$–C$_{14}$–PFCAs in plasma of adult chick-rearing kittiwakes from the same population on Svalbard. The pattern of PFCAs was similar to that in livers of other seabirds (Fig. 21) with PFUnDA and PFTrDA predominating.

Braune et al. [149] determined PFASs in liver of adult thick-billed murres (*Uria lomvia*) and northern fulmars from several locations in the eastern Canadian Arctic during 2007–2008. ΣPFCA varied significantly among the five murre colonies with the highest concentrations found at Coats.

Island in Hudson Strait. Patterns varied among colonies and, at some colonies, between sexes, most notably for murres at Akpatok Island. The PFAS patterns and concentrations noted between murres could be at least partially because Pacific waters influence Lancaster Sound while Atlantic waters and freshwater sources influence Hudson Bay. While no results for liver of murres or fulmars are available for the European Arctic there are nevertheless distinct

Table 3
New classes of perfluoroalkyl sulfonates in polar bear plasma detected by high resolution mass spectrometry [143].

Class of PFSA	Molecular formula	Total analytes detected	Confidence Level[a]
[1] Cyclic or unsaturated PFSAs	$C_nF_{2n-1}SO_3$, n = 8–10	8	3
[2] Ether PFSAs	$C_nF_{2n+1}SO_4$, n = 6–9	13	3
[3] Unsaturated ether-, cyclic ether- or carbonyl PFSAs	$C_nF_{2n-1}SO_4$, n = 7–9	11	3
[4] x:2 Chlorinated perfluoroalkyl ether sulfonates	$ClC_nF_{2n}SO_4$, n = 6–8	3	1&2

[a] Confidence level of the proposed structure, assigned based on criteria proposed by Schymanski et al. [144]. Level 1&2 indicates some compounds were confirmed with authentic standards.
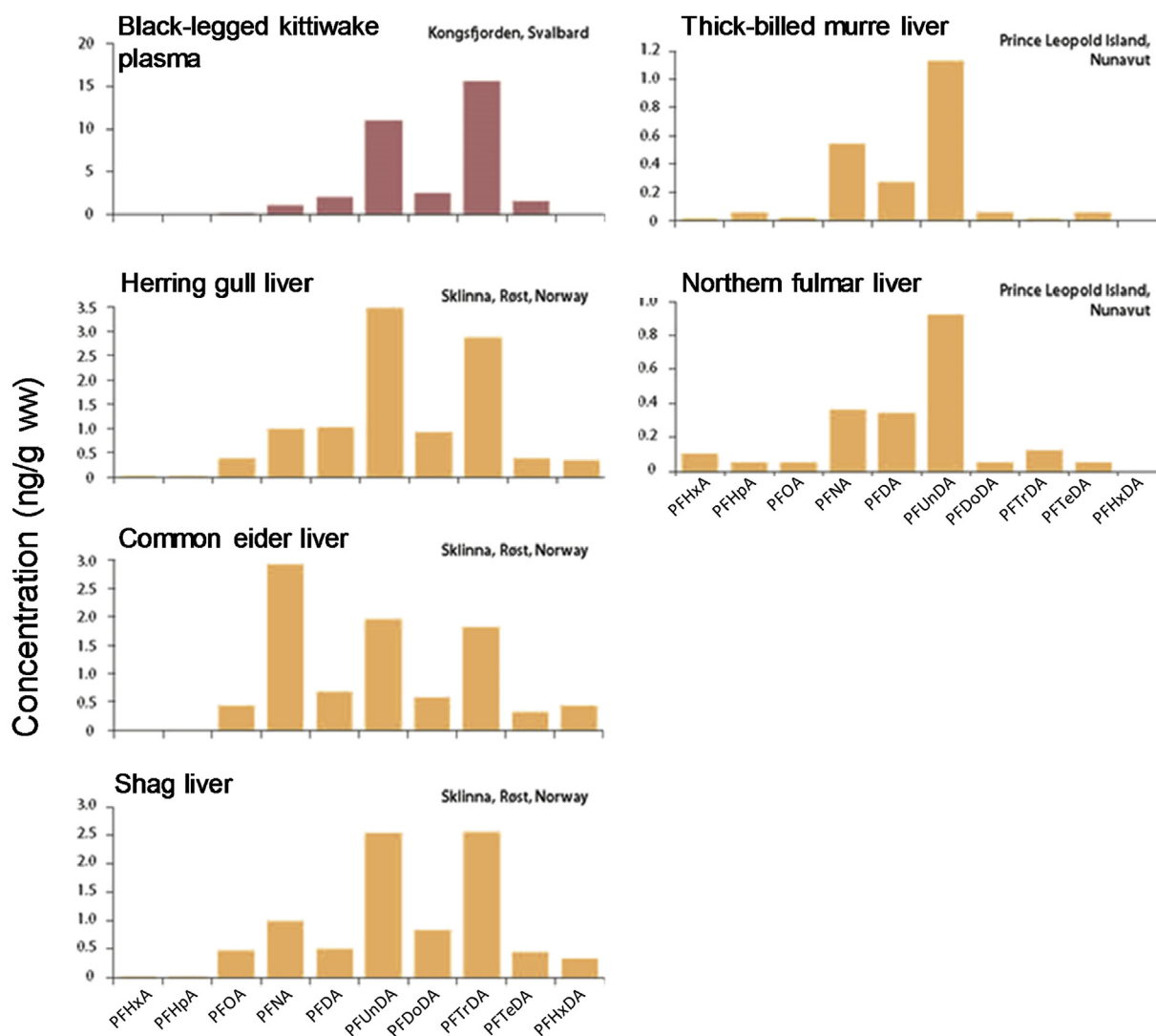
Fig. 21. Concentrations and patterns of C_6–C_{16}–PFCAs in liver of common eider (Sklinna, Røst, Norway), herring gull and shag (Sklinna, Røst, Norway), thick-billed murre and northern fulmar (Prince Leopold Island, Lancaster Sound, Nunavut) and in plasma of black-legged kittiwakes (Kongsfjorden, Svalbard). Results from Norway are from Huber et al. [152] and Tartu et al. [148], and for Nunavut from Braune et al. [149].

differences when comparing the proportions of PFCAs in common eider (*Somateria mollissima*), shag (*Phalacrocorax aristotelis*), kittiwakes, and herring gull (*Larus argentatus*) from Norway. C_{12}–C_{14} PFCAs are more prominent in the Norwegian species (Fig. 21).

Braune and Letcher [150] examined the profile of C_6–C_{15}–PFCAs in eggs of five species of seabird from the Canadian Arctic, collected in 2008. PFUnDA was the predominant congener in glaucous gull (*Larus hyperboreus*), thick-billed murre, and black guillemot (*Cepphus grylle*), while PFTrDA predominated in black-legged kittiwake and northern fulmar (Fig. 22). Although long-chain PFCAs predominated in eggs, the C_9–C_{11} PFCAs predominated in liver of adult

murres and fulmars from Prince Leopold Island (Lancaster Sound) [149]. This may be due to preferential accumulation of long-chain PFCAs in egg yolk as demonstrated in herring gull eggs [151].

Braune and Letcher [150] also detected PFBS in the murre and fulmar eggs. This was the first detection in these species of PFBS, which is a degradation product of perfluorobutane sulfonyl products, which have replaced PFOS-related chemicals. However, it was detected only in samples from 2010 to 2011 and at low levels ranging from means of 0.04 ng/g (murres) to 0.57 ng/g (fulmars). By comparison, mean PFOS concentrations for 2011 ranged from 19.8 ng/g (fulmars) to 23.8 ng/g (murres). Tartu et al. [148] did not
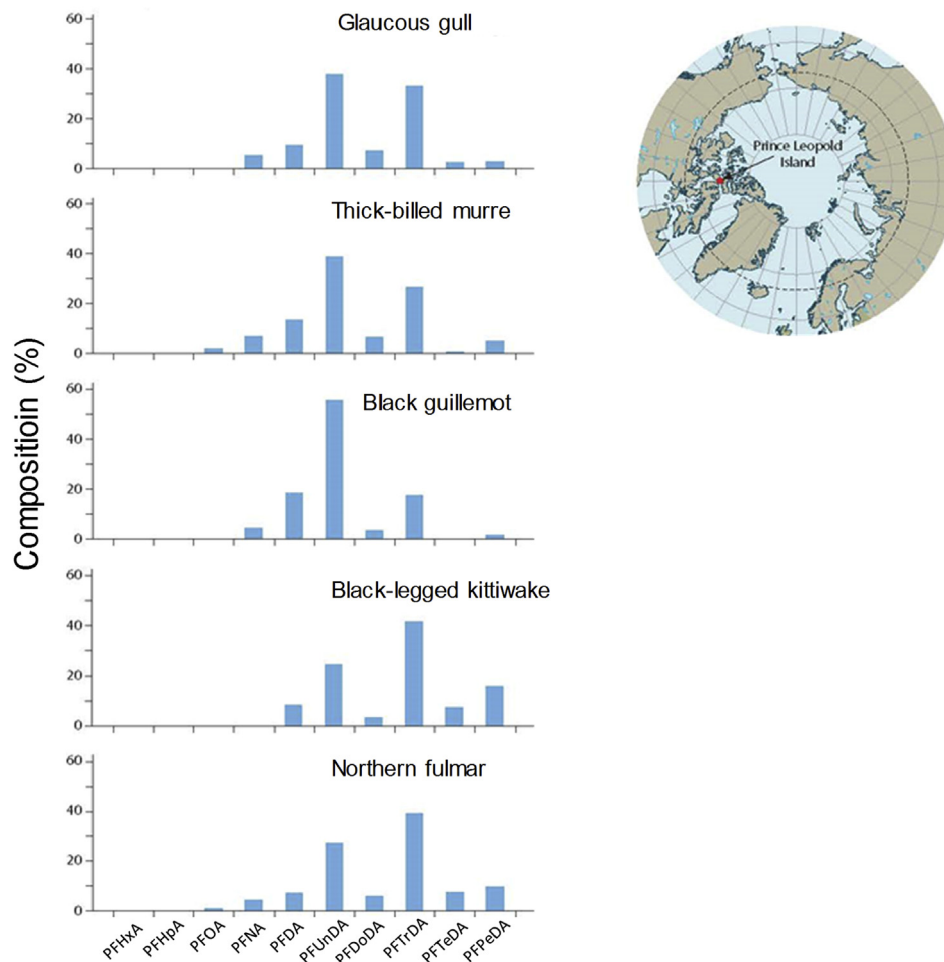
Fig. 22. Mean contributions (%) of $C_6-C_{15}-$PFCAs (PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, and PFPeDA) to ΣPFCA in eggs of black guillemot, thick-billed murre, northern fulmar, glaucous gull, and black-legged kittiwake sampled from Prince Leopold Island, Nunavut, 2008 [150].

detect PFBS or short-chain (C_4-C_6) PFCAs in plasma of chick-feeding black kittiwakes from Svalbard. However, the pattern of PFCAs in the kittiwakes from Svalbard was very similar to that for kittiwake eggs from Prince Leopold Island, Nunavut.

2.5. Temporal trends

Long-term temporal trend data sets are available for $C_8-C_{12}-$PFCAs in selected Arctic freshwater and marine biota and for PFOA and FTOH precursors, as well as for MeFOSE and EtFOSE in atmospheric samples. Butt et al. [4] reviewed temporal trends of PFASs for studies published to 2009. The Canadian Arctic Contaminants Assessment Report on POPs reported temporal trends of PFASs to 2011 in freshwater fish, seabirds and marine mammals [99]. This section includes a review of recently published and unpublished trend data with a focus on results for PFCAs reported since 2009.

2.6. Air measurements

Wong et al. [8] reported the first time temporal trends for PFASs in air from three arctic stations: Alert (Canada, 2006–2014); Zeppelin (Svalbard, Norway, 2006–2014) and Andøya (Norway, 2010–2014). Results for PFOA, PFBA and PFBS are shown in Fig. 23. Increasing trends in air concentrations were observed at Alert with doubling times (t2) of 3.7 years (y) for PFOA, 2.9 y for PFOS, 2.5 y for

PFBA, 5.0 y for 8:2 FTOH and 7.0 y for 10:2 FTOH. In contrast, declining or non-changing trends, were observed for PFOA and PFOS at Zeppelin (PFOA, half-life, t1/2 = 7.2 y; PFOS t1/2 = 67 y), and Andøya (PFOA t1/2 = 1.9 y; PFOS t1/2 = 11 y). FOSEs and FOSAs were near detection limits but visual inspection of the trend plots showed that these chemicals were declining. Although the overall trends (i.e. 2006–2014) of 8:2 and 10:2 FTOH were increasing at Alert, they appeared to be declining from 2012 [8].

Gawor et al. (2014) reported results for neutral PFASs from XAD-type samplers deployed on a global-scale for one-year intervals during the period 2006–2011. Levels of FOSAs and FOSEs tended to decrease globally during the six years of measurements, whereas an initial decline in the concentrations of FTOHs from 2006 to 2008 did not continue in 2009–2011.

2.7. Seawater

Time series measurements are lacking for PFASs in seawater although annual measurements at fixed locations such as Barrow Strait (Nunavut, Canada) have begun [153]. Additional studies are necessary to investigate seasonality and long-term changes.

2.8. Ice cores

Kwok et al. [69] reported historical profiles of PFCAs from two ice cores collected on the glacier near Longyearbyen in 2006.
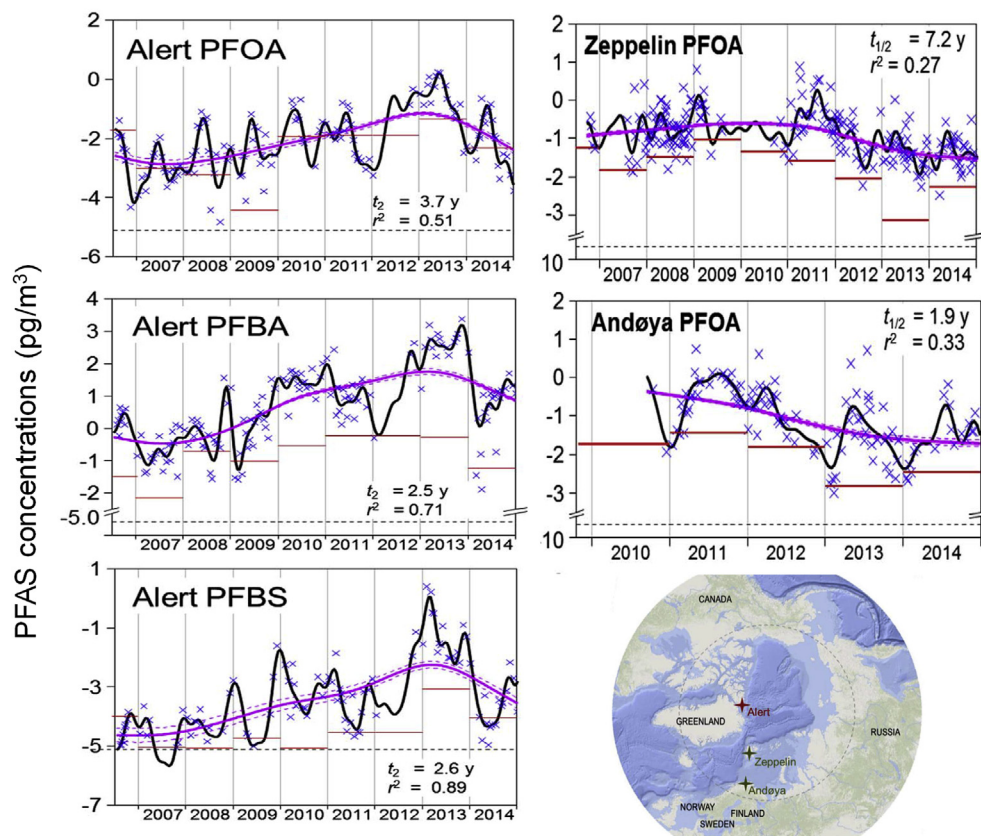
Fig. 23. Trends of major ionic PFASs in air at Alert(filter+PUF), Zeppelin (filter) and Andøya (filter). Doubling times (t2) or half-lives (t$_{1/2}$) are shown in units of years (y). Measured data are shown as blue crosses, the black line is the seasonal cycle, and the pink line is the trend derived from a Digital Filtration model [8]. Dash pink lines indicate the lower and upper 95% confidence limits of the trend. r^2 indicates the correlation coefficient between the trend and measured data. IDL and MDL are shown as dash black and red lines respectively. PFBA and PFBS were not determined at Zeppelin or Andøya.

Consistent patterns were observed in the vertical profiles of PFCAs in both ice cores with PFBA predominating (39%), followed by PFOA (17%) and PFNA (11%). PFCAs were the major PFASs in the ice cores. Fluxes (concentrations in pg/L multiplied by water equivalents in L/m^2 for each year) showed maximum PFCA deposition occurred in the late 1990s (Fig. 24). Young et al. [72] also observed elevated PFOA and PFNA deposition from the late 1990s in a snow pit from the Devon Ice Cap collected in 2006 along with higher deposition in near-surface samples. However, fluxes of PFOA, PFNA, PFDA and PFUnDA found in the Norwegian ice cores [69] were two-to 10-fold lower than in the Canadian Arctic ice cap samples [72]. This result is consistent with oceanographic cruise measurements of FTOHs in Nunavut compared to the Svalbard and Norwegian Sea region [57,154].

MacInnis et al. [73] determined temporal trends of deposition of PFASs using samples collected from a 6 m deep snow pit on the Devon Ice Cap in 2008. As noted in Section 2.1, PFBA was the predominant PFCA, with fluxes ranging from 15 to 244 ng/m^2 ·yr. Maximum PFBA fluxes were observed in horizons dated to 2005 (Fig. 25). PFNA was the next largest PFCA ranging from 7.5 to 220 ng/m^2 ·yr, with greatest fluxes in samples dated to 2006 and 2007. Deposition of FOSA, which is also a possible volatile precursor of PFOS, increased in the 1990s, peaking around the year 2000, followed by a decrease up to 2007. This is in accordance with the voluntary phase-out of the C$_8$-based chemistry in 2001 [155].

Pickard et al. [95,156] analysed historical trends of PFAS deposition using a 15.5-m ice core collected from the summit of Devon Ice Cap in May 2015, representing 37 prior years of deposition. They found that PFOA and PFNA fluxes increased from 1977 up until 1995

(Fig. 26). In the period post-1995 until 2013, fluxes have plateaued, with <25 ng/(m^2 · yr) variance in annual flux. PFOA and PFNA fluxes declined from 2012 to 2015. The decline corresponded to anticipated.

PFCA emission reductions through the US EPA PFOA Stewardship Program [23]. The PFOS was detected at consistent levels below 10 ng/(m^2 · yr), with an anomaly (80 ng/(m^2 · yr)/detected in 2013 (Fig. 26C). FOSA was measured in almost every year from 1977 to 2000 with fluxes increasing until 1995. After 2000, FOSA was only detected in three samples with levels <0.76 ng/(m^2 · yr) (Fig. 26D).FOSA is a known volatile precursor and can degrade to PFOS [30]. The profile for PFOS does not reflect known global production while for FOSA the lack of detection post 2000 agrees relatively well with known phase out of POSF chemistry by the main manufacturer (3 M Company).

2.9. Lake sediment cores

MacInnis et al. [98] studied the temporal trends of PFASs using dated sediment cores from Lake Hazen a large lake (82° N) in northern Ellesmere Island, and Lake B35, a small, remote, lake near Baker Lake NU (64° N). In Lake Hazen, increasing fluxes (concentration (ng/g dw) multiplied by sedimentation rate (g/(m^2 yr^1) were observed for PFOA, PFDA, PFBS and PFOS during the period 1963–2011 (Fig. 27). In Lake B35, increasing fluxes were observed for PFHpA, PFOA, PFNA and PFDA for the period 1952–2009. The depositional record in both lakes was broadly consistent with global use and emissions of PFOS and PFCA-related compounds. However, in Lake Hazen, climate–induced glacier melting within
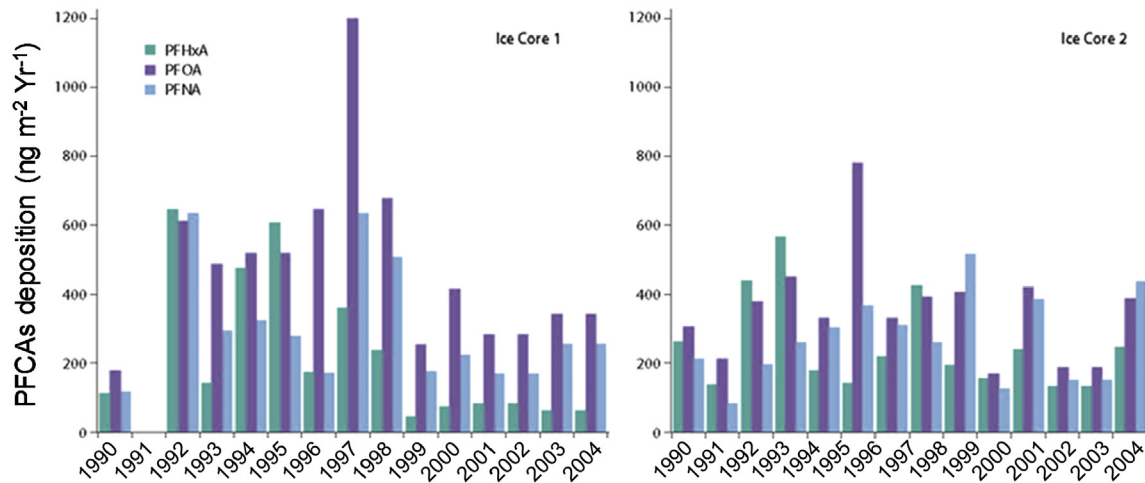
Fig. 24. Estimated annual fluxes of PFCAs (PFHxA, PFOA, and PFNA) in ice cores from Svalbard, Norway [69]. [Note the original graph published by Kwok et al. [69] showed fg/(m^2 · yr) flux units whereas the correct units are shown here.].

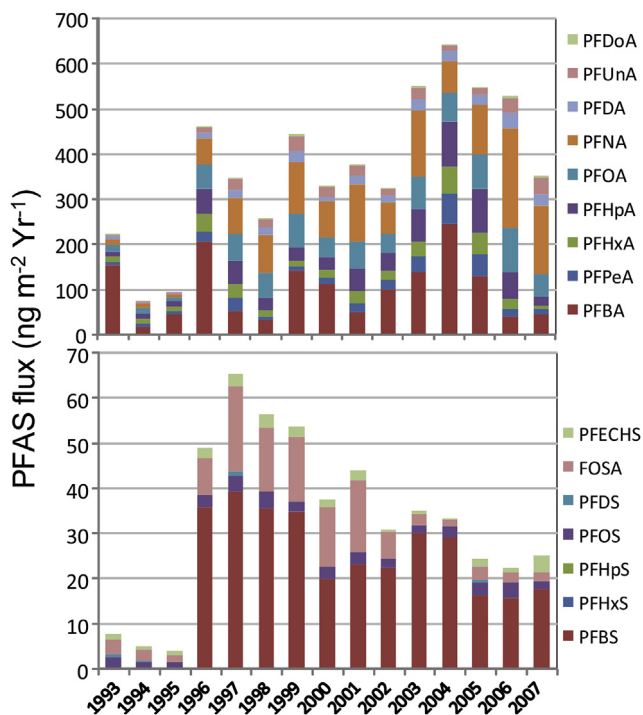**Fig. 25.** Estimated annual fluxes (ng/(m^2 · yr) of A. PFCAs (C$_4$–C$_1$2) and B. PFSAs (C$_4$–C$_{12}$ + PFECHS) in a snow pit from the summit of the Devon Ice cap (Devon Island, Nunavut). Results from Macinnis et al. [73].

the watershed post-2005 gave rise to much higher sedimentation rates [158]. The sharp increase in fluxes of PFASs post-2005 was related to glacier inputs. Fluxes of PFOA, PFBS, and PFOS were correlated with annual glacial meltwater discharge [98].

2.10. Temporal trends in biota

2.10.1. Freshwater fish

The Canadian Arctic Contaminants Assessment Report on POPs reported temporal trends of PFCAs to 2011 in Arctic char, lake trout and burbot [99]. Updated results from that report are provided here. C$_7$–C$_{14}$ PFCAs appear to be declining in landlocked Arctic char

from Lake Hazen, Char Lake and Amituk Lake (Canada), from their peak in the period 2006–2009 (Fig. 28). The mean annual declines between 2009 and 2013 are quite rapid ranging from 8.8% for Amituk Lake to 26% for Lake Hazen using the AMAP PIA program [159].

Declining trends for C$_7$–C$_{14}$ PFCAs were also observed in lake trout from Great Slave Lake (Northwest Territories) over the period 2000–2011 (Muir et al. [99]; M. Evans and D. Muir, Environment Canada, Saskatoon, Canada, unpubl. data) (Fig. 29). These represented declines of about 40% per year from the early 2000s to 2010. Between 2011 and 2013 concentrations levelled off or increased slightly. In lake trout from Lake Laberge and Kusawa Lake (Yukon) total C$_9$–C$_{11}$-PFCA concentrations declined over the period 2007–2012 at very similar rates in both lakes.

Trends for PFCAs in burbot liver were distinctly different from those for lake trout (Fig. 29) possibly reflecting differences in contamination of the benthic versus pelagic food webs. Rising C$_7$–C$_{14}$-PFCA concentrations were observed in the West Basin of Great Slave Lake over the period 2007–2012, while concentrations remained the same in burbot from the East Arm. At Fort Good Hope, C$_9$–C$_{11}$-PFCA concentrations declined in burbot from 2003 but increased between 2010 and 2013.

2.10.2. Seals

Rigét et al. [121] found declining trends in PFOA, PFNA, PFDA and PFUnDA in ringed seals from Qeqertarsuaq (West Greenland) occurred over the period 2008–2010 after a steady increase. The decline was particularly striking for PFOA with a clearing half-life of approximately one year. While PFOA in seal liver also declined (half-life ∼ 2 years) at Ittoqqortoormiit (East Greenland), PFNA, PFDA and PFUnDA continued to increase (Fig. 30).

In the Canadian Arctic, declining trends for total (C$_7$–C$_{14}$)–PFCAs were observed in ringed seals from four locations in the period 2005–2010 (Fig. 31). However, more recent results suggest an increase in total PFCAs in Hudson Bay and Lancaster Sound; the declining trend occurred later in the southeastern Beaufort Sea ringed seals. The trend for total PFCAs reflects changes in the major components (PFNA, PFDA, PFUnDA; Fig. 20) which have very similar individual trends [127,132]. Similar differences in trends between ringed seals in the Beaufort Sea and in Hudson Bay and Lancaster Sound have been observed for β-HCH isomers [160]. They are thought to reflect the influence of Pacific Ocean waters and Russian rivers as sources for the Beaufort Sea [161] whereas sources for
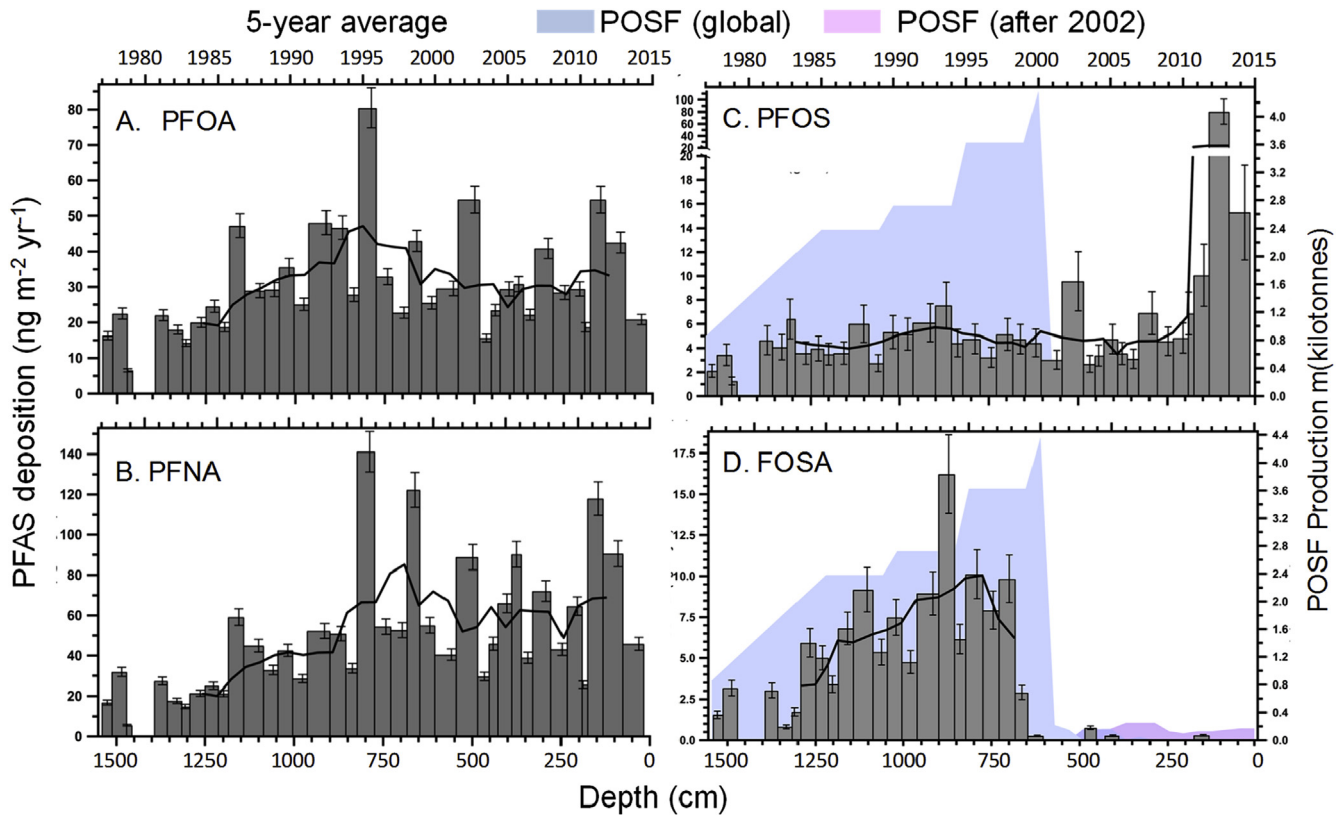
Fig. 26. Estimated annual deposition fluxes (ng/(m² · yr) on the Devon Ice Cap: (A) PFOA, (b) PFNA, (C) PFOS and (D) FOSA, with global POSF production from 1977 to 2003 and POSF production for Western Europe, Japan, United States and China from 2003 to 2015 [27,157]. Solid black lines represent 5-year moving averages. Redrawn from Pickard et al. [95].
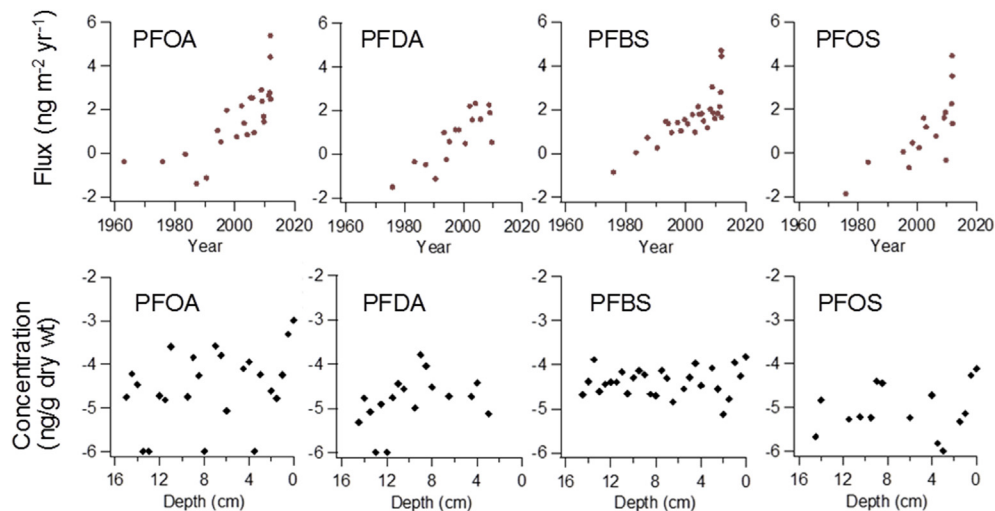

Fig. 27. Deposition fluxes (ng/(m² · yr) and concentrations (ng/g dry wt) of PFOA, PFDA, PFBS and PFOS in a dated sediment core from Lake Hazen (82° N). Redrawn from Macinnis et al. [98].

western Hudson Bay reflect inflowing rivers and atmospheric deposition from North American sources [28].

2.10.3. Whales

Rotander et al. (2012) reported increasing trends for PFNA, PFDA and PFUnDA in pilot whales (*Globicephala melas*) and white-sided dolphins from the Faroe Islands. Samples were from the period 1986–2006 (pilot whales) and 2001–2006 (dolphins). Calculated

on an annual basis the increases were generally over 10% per year, reflecting the sharp increases seen for other Arctic marine species up to the mid-2000s. However only three sampling years were available for pilot whales and two for dolphins.

Reiner et al. [133] examined temporal trends of PFCAs in beluga from Cook Inlet and the Chukchi Sea (northwestern Alaska). Although sample sizes were limited (n = 1 or 2 per year for Cook Inlet 2001–2006) the overall trend appears to show a rapid
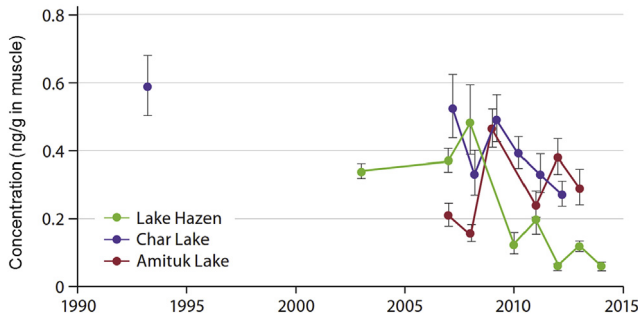
Fig. 28. Trends of C$_7$–C$_{14}$ perfluorocarboxylates (ΣPFCAs) in landlocked char muscle from Lake Hazen, Char Lake and Amituk Lake in the Canadian Arctic. (D. Muir, Environment Canada, Burlington, Canada. Unpublished data).

increase in concentration of total PFCAs over the period 1993–2003 with a decline thereafter (Fig. 32A). However no significant trend was evident for the Chukchi Sea beluga.

Smythe et al. [135] found that ΣC$_8$-C$_{12}$-PFCA concentrations in beluga from the southern Beaufort Sea population (Henderson Island) decreased by 6.0%/year, t$_{1/2}$ = 12 years) between 1984 (157 ± 15 ng/g ww) and 2013 (65 ± 11 ng/g ww) (Fig. 32B). In contrast, ΣC$_8$-C$_{12}$-PFCA concentrations in beluga harvested at Pangnirtung, in the Eastern Canadian arctic, increased by 6.7% annually (t2 = 10 years) from 9.3 ± 2.5 ng/g ww in 1982 to 25 ± 18 ng/g ww in 2010, although they also had an earlier maximum in 2000 (Fig. 26). Beluga from southern Hudson Bay (Sanikiluaq) showed a continuous decline (12%/year) from 2003 to 2014 (Fig. 32B). Total PFCA concentrations in the Beaufort Sea beluga in the 1990s were generally higher than reported for the Chukchi Sea animals.

2.10.4. Arctic foxes

Routti et al. [83] determined temporal trends of PFCAs and PFSAs in arctic foxes from western Spitsbergen in Svalbard. Liver samples from 11 years over the period 1997 to 2014 were available. PFDA, PFUnDA, and PFTrDA increased also significantly in arctic foxes (~4% per year) during 1997–2014, whereas the increasing trend of ~2% per year for PFNA and PFDoDA was not significant (Fig. 33). Concentrations or detection frequency of PFHxA, PFHpA, and PFOA in arctic foxes, did not change over time [83]. ΣPFCAs increased in arctic fox. Correspondingly, PFHpS, PFOS, and PFTrDA concentrations in arctic foxes increased with increasing sea ice cover, whereas the amount of reindeer carcasses were negatively related to concentrations of C9−12 PFCAs.

2.10.5. Polar bears

Hudson Bay polar bears appeared to have relatively constant concentrations of ΣPFCAs for the period 2007–2014 while PFOS appears to have declined at least in southern Hudson Bay (Fig. 34). Even though concentrations were uncorrected for sex, age, and diet, R. Letcher et al. (Environment Canada, Ottawa, Canada, 2015, unpublished data) found that in polar bear livers collected for all years between 2007 and 2014, mean concentrations in the Hudson Bay population were continuously very high at >400 ng/g ww (ΣPFCAs) and >900 ng/g ww (PFOS). This stresses the importance of PFCA and PFOS precursors as sources, which are transported to the Arctic and/or degraded in bears and/or their prey/food web.

Rigét et al. [121] reported that PFOS was still the most predominant PFAS in polar bear liver samples up to 2010, constituting 85% in East Greenland polar bears, and until 2006 had doubling times of about 14 years. PFOS as well as PFHxS and FOSA concentrations showed decreasing trends in recent years as did PFDA and PFUnDA. Rigét et al. [121] updated the temporal trends of PFCAs in
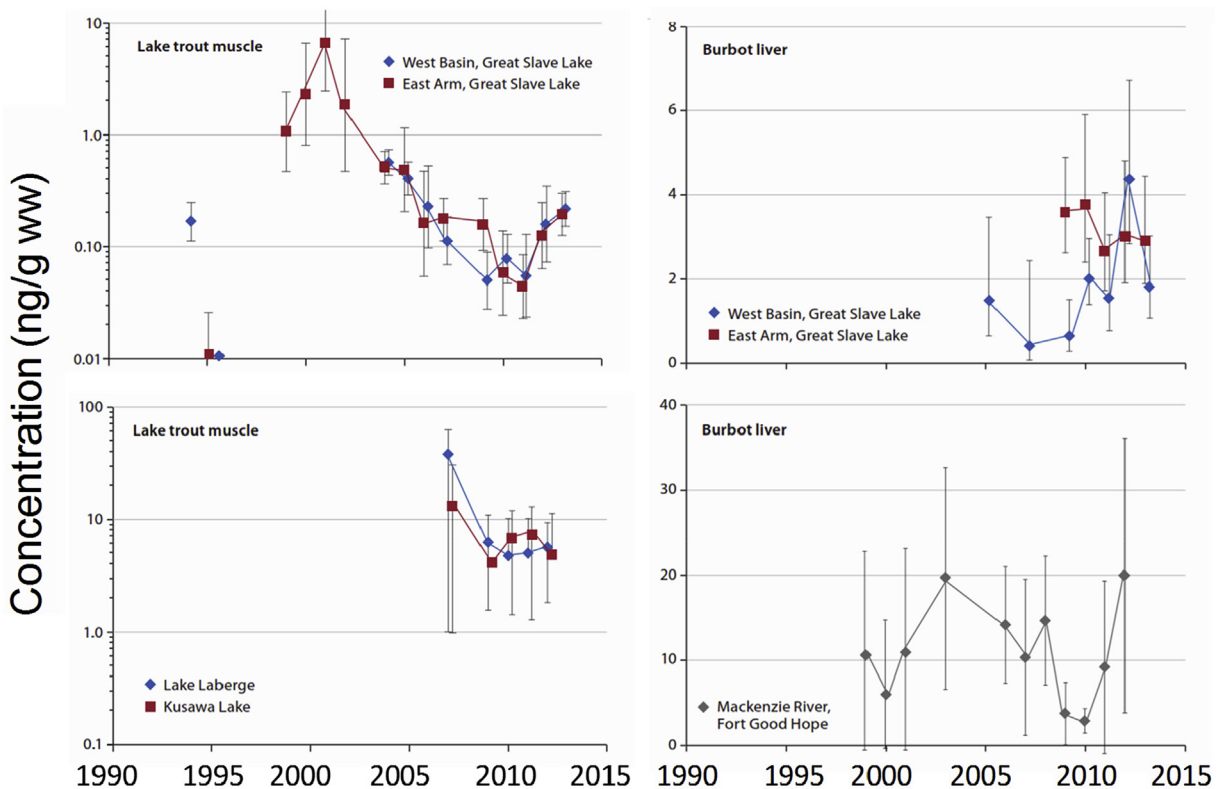

Fig. 29. Trends in C$_7$–C$_{14}$ perfluorocarboxylate concentration in lake trout muscle and burbot liver from Great Slave Lake (GSL) (Muir et al. [99]; Evans and Muir, unpubl. data), lake trout muscle from Lake Laberge and Kusawa Lake (Stern et al. [100]), and burbot liver from Fort Good Hope, Mackenzie River [102].
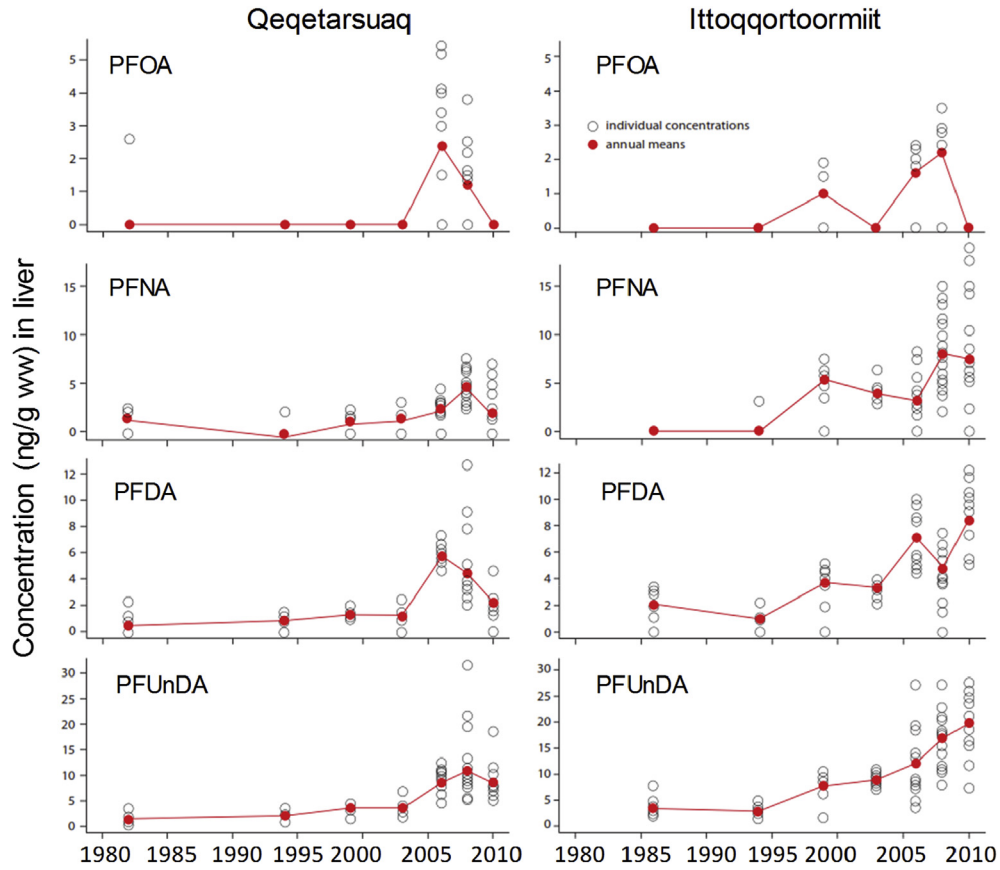
Fig. 30. Temporal trends in PFOA, PFNA, PFDA, PFUnDA (ng/g ww) in liver tissue of ringed seals from Qeqertarsuaq (West Greenland) and Ittoqqortoormiit (East Greenland). Results reproduced from Rigét et al. [121]. Black circles represent individual concentrations and red points represent annual means linked by red lines.
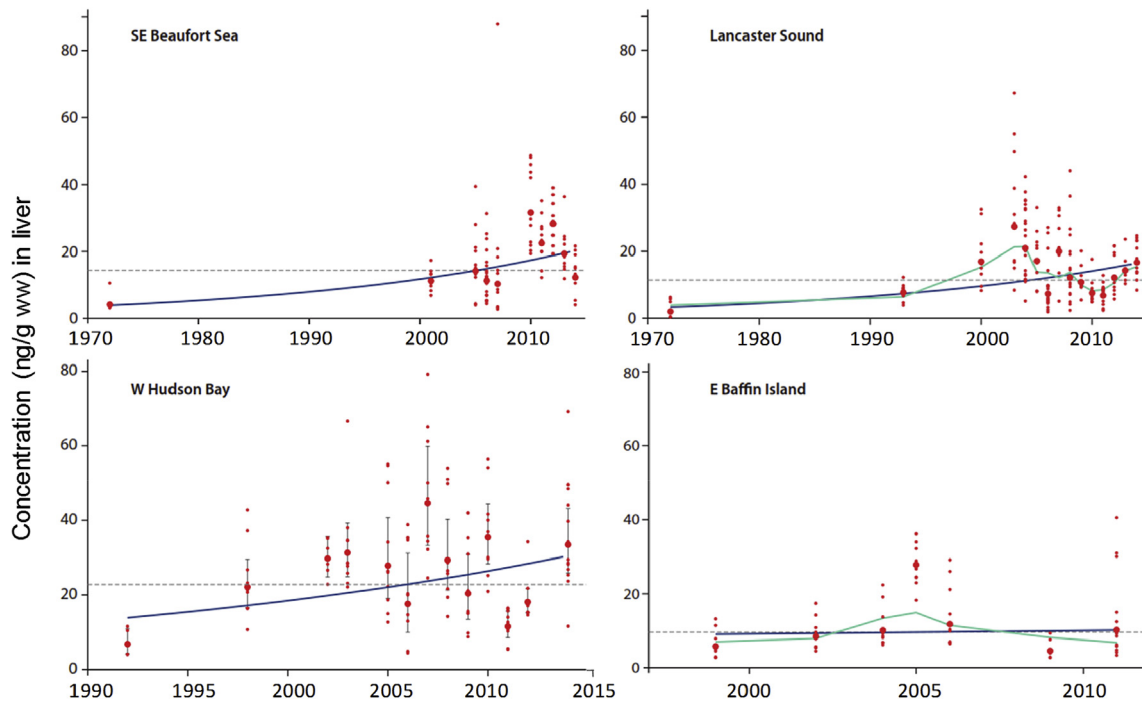

Fig. 31. Temporal trends of total PFCAs (sum C_7–C_{14} PFCAs) in ringed seal liver from four locations (SE Beaufort Sea = Sachs Harbor and Ulukhaktok; Lancaster Sound = Resolute, Grise Fiord and Arctic Bay; W. Hudson Bay = Arviat; E Baffin Is = Pangnirtung, Pond Inlet and Qikiqtarjuaq) in the Canadian Arctic [127,132]. Green lines represent 2-year moving average. Red circles and vertical lines are annual means and standard deviations.
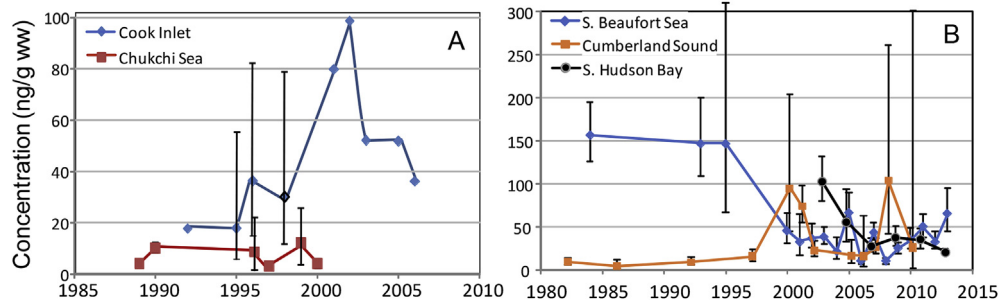
Fig. 32. Temporal trends in ΣPFCAs in (A) beluga liver samples from the Cook Inlet and Chukchi Sea (northwest Alaska) reported by Reiner et al. [133] and (B) beluga liver from the southern Beaufort Sea, Cumberland Sound, and Southern Hudson Bay stocks [135]. Symbols represent geometric mean concentrations. Vertical bars are upper and lower 95% confidence intervals.
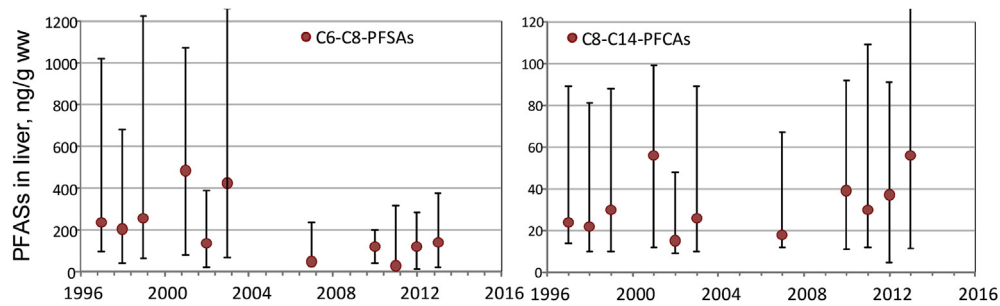

Fig. 33. Temporal trends in median (range) of concentrations (ng/g ww) of ΣC_6-C_8-PFSAs and ΣC_8-C_{14}-PFCAs liver samples from young arctic foxes collected from Svalbard during the period 1997–2013. Data from Routti et al. [83].

East Greenland polar bears that were first reported by Dietz et al. [162]. PFOA, PFNA and PFUnDA all showed declining trends in polar bear liver over the period 2006 to 2011 (Fig. 35) with annual declines ranging from 6% (PFNA) to 15% (PFOA).

Routti et al. [83] reported temporal trends of PFASs in blood plasma of female polar bears from the Barents Sea subpopulation were captured annually in the period 2000–2014 (some animals were recaptured and sampled multiple years). Concentrations of C_6–C_8 PFSAs in the bears peaked in 2001–2003, after which they declined by 9–14% per year (Fig. 36). This decline, also noted for arctic foxes in Svalbard, as well as seals and polar bears in Canada and Greenland, corresponds to the phaseout of POSF-based products and their C_6 and C_7 homologues as discussed in Section 1.2.

Concentrations of C9–13 PFCAs in polar bear plasma, adjusted for biological and environmental variables, increased ~3% per year over period 2000–2014, while the increase in C12–14 PFCA concentrations leveled off as of 2009. The leveling off post-2009 agrees with projected trends in global emissions of PFCA related compounds proposed by Wang et al. [27]. Adjustment of the data for feeding habits based on $\delta^{15}N$ and $\delta^{13}C$ showed only a small influence on trends in concentrations of PFCAs in polar bears and none for PFSAs.

2.10.6. Seabirds

Braune and Letcher [150] and Braune [163] examined temporal trends in PFCAs in thick-billed murre and northern fulmar eggs over the period 1975–2014 (14 sampling years including annual
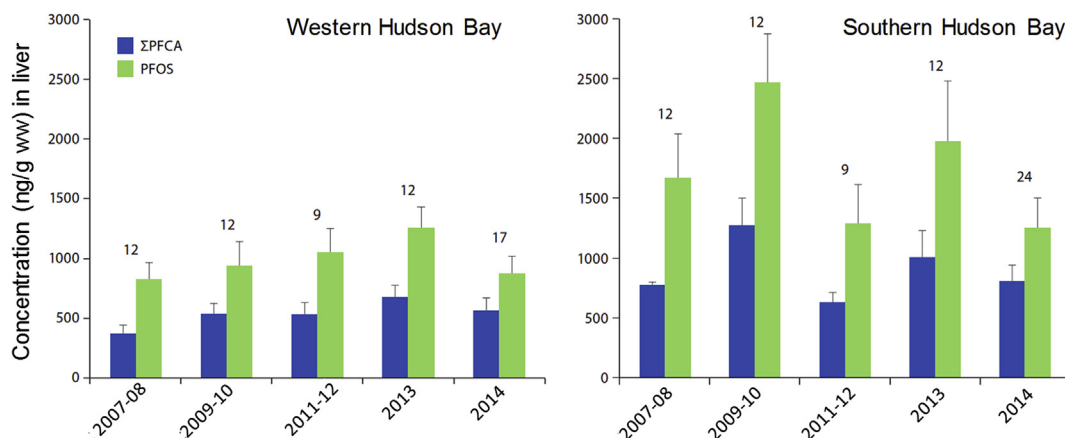

Fig. 34. Temporal trends in geometric mean concentration of ΣPFCA and PFOS in the liver of western Hudson Bay bears (2007–2014) and southern Hudson Bay bears (2007–2014) R. Letcher et al. (Environment Canada, Ottawa, Canada, 2015, unpublished data). Error bars are standard deviations (SDs). Sample numbers are shown above bars. Data not corrected for sex, age or diet.
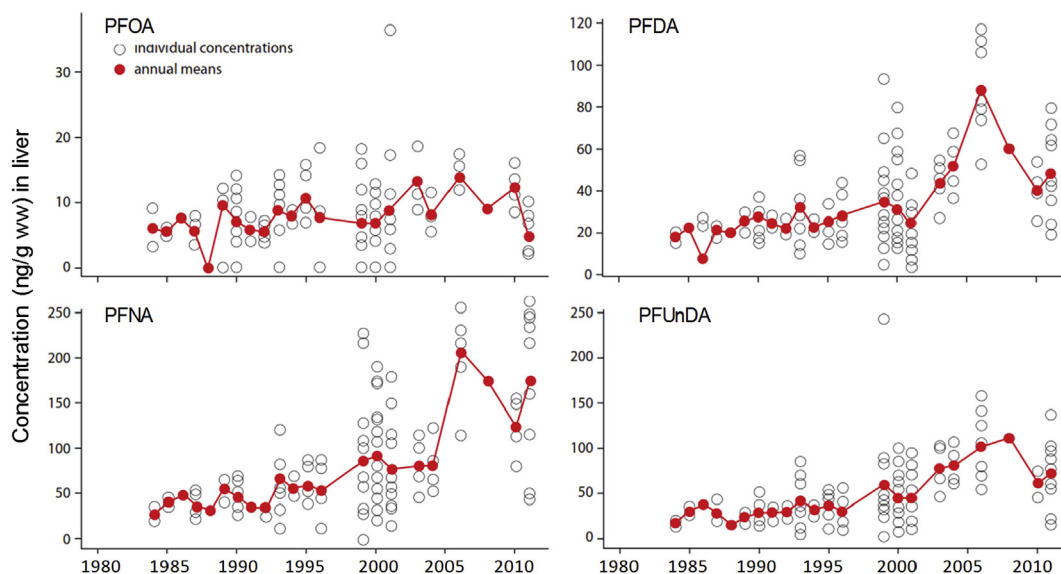
Fig. 35. Temporal trends in PFOA, PFNA, PFDA, and PFUnDA (ng/g ww) in polar bear liver from Ittoqqortoormiit (East Greenland). Black circles represent individual concentrations and red points represent annual means (Rigét et al. [121]).

samples from 2006 to 2014). Increasing trends were observed in fulmars until 2008 and until 2010 in murres. From 2010 to 2014 PFCAs declined rapidly in fulmar eggs (~12%/y) while remaining more or less constant in murres (Fig. 37). The long-chain (C_9–C_{15}) PFCAs contributed to the increasing ΣPFCA concentrations in the murres and fulmars during the period 1993–2014. PFUnDA and PFTrDA were the predominant PFCAs and together constituted over 60% of ΣPFCA in all years, with PFTrDA dominating the fulmar PFCA profile (except in 1975) and PFUnDA generally dominating the murre PFCA profile. PFOA (C_8) was detected (>0.08 ng/g ww) in eggs of both species from 2008 onward and comprised <3% of the PFCA profile in murres and northern fulmars.

3. Conclusions and recommendations

New data are available for a wider range of perfluoro- and polyfluorinated substances in the Arctic since the previous detailed review [4]. New PFASs detected include PFECHS (a cyclic analog of PFOS), FBSA (a precursor of PFBS), a series polyfluorinated ether sulfonates including the chlorinated PFOS related compound, 6:2-Cl-PFAES, and cyclic or unsaturated PFSAs [86,128,143]. Concentrations of these new substances are not elevated relative to PFOS or most PFCAs, however, particularly in the case of 6:2-Cl-PFAES and FBSA, they are replacement compounds for PFOS-related uses, and deserve additional scrutiny.

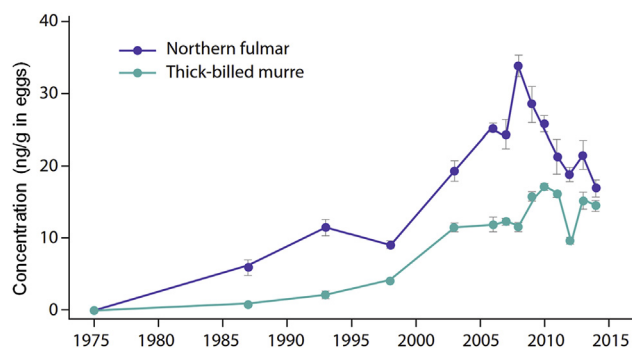While spatial and temporal coverage of PFASs in wildlife, as well

Fig. 37. Mean (±SE) annual concentrations (ng/g ww) of total perfluorinated carboxylates (sum of C_6 to C_{14}-PFCA) in eggs of northern fulmars and thick-billed murres from Prince Leopold Island, 1975–2014 [163].

as in seawater, is quite extensive across a wide portion of the Arctic from the Bering Sea, eastward to the Barents Sea, there is limited or no information for the Russian Arctic. This is a concern especially given the potential local uses and emissions of PFASs that might be used at airports, harbours, oil development sites, and military bases, in a region that has much larger urban and industrial centers compared with the North American Arctic and Greenland [164].

More studies on the partitioning behavior of PFASs are needed, such as between solid/water and air/water phases [104] to get a
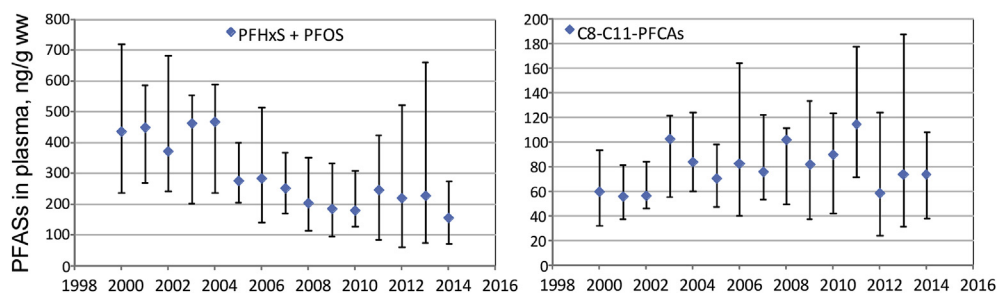

Fig. 36. Trends in median (range) concentrations (ng/g ww) of PFHxS+PFOSs and ΣC_8-C_{11}-PFCAs in plasma from polar bears at Svalbard (2000–2014). Data from Routti et al. [83].

better understanding of the relationship between sources, seawater concentrations and the mechanisms of long-range global transport. Also more follow-up of the results suggesting that sea spray aerosols could be an important source of PFASs to coastal environments is needed [165].

Although C_{13}–C_{15}–PFCAs had been reported previously in Arctic marine biota, particularly in seabirds [166] additional measurements have shown that these compounds are present in most top predators. Information on the tissue distribution of these long-chain PFCAs, which are more hydrophobic than PFOS or more commonly detected C_8–C_{12} PFCAs, is limited. For example, results from analysis of reindeer showed that C_{13}–C_{15} PFCAs were detectable in fatty tissue. Similarly, seabird eggs have higher proportions of C_{13}–C_{15} PFCAs than seabird liver [150]. Accumulation of long-chain PFCAs in egg yolk as demonstrated in herring gull eggs [151] may be the reason. Additional studies of fatty tissues for other mammals are needed. The relative proportions or pattern of the PFCA congeners also differs between the European Arctic, Greenland and Canada. For example, C_{13}- and C_{14}–PFCAs were not detected in lake trout from northern Canada but readily detected in fish in the European Arctic, the Faroe Islands and Greenland. However, additional studies are needed because most analyses of fish from the European Arctic have used liver, while muscle has generally been used in Canada (except for burbot). The differences may thus also reflect tissue distribution.

There are an increasing number of measurements of PFBA in freshwater, seawater, snow and air samples which indicate that it is the most prominent PFCA in these abiotic media. For example, PFBA predominated in all air samples collected by the GAPS program using sorbent-impregnated polyurethane foam or polyurethane foam disks. PFBA has multiple non-fluorotelomer gas-phase sources, including chlorofluorocarbon replacements HFC-329 ($CF_3(CF_2)_3H$), HFE-7100162 ($C_4F_9OCH_3$), and HFE-7200 ($C_4F_9OC_2H_5$), and this is likely to explain its predominance. PFBA has also been reported in fish (e.g. in landlocked char from a lake in Svalbard) although it was not detected in liver of polar bears from Hudson Bay. However, some authors have noted that further confirmation of PFBA and PFPeA is needed because of the availability of only a single quantifier transition in the analysis by low-resolution MS/MS. Although it is expected to be eliminated quickly due to its high water solubility, the predominance of PFBA in abiotic samples suggests that it could be present, particularly in fish and invertebrates that are continuously exposed via gill respiration. Thus further measurements of PFBA in biota are needed.

Recent studies have shown that there can be substantial EOF in aquatic biota that is not explained by measured PFASs [167]. However as of late 2018 no EOF measurements appear to have been conducted on Arctic abiotic or biota samples. This is an important knowledge gap which could be addressed using samples from environmental specimen banks.

An extensive dataset exists for long-term trends of long-chain PFCAs in Arctic biota with some datasets including archived samples from the 1970s and 1980s. Trends in PFCAs over time vary across the North American Arctic and between East and West Greenland. Rising levels of some PFCAs have been explained by continued emissions of long-chain PFCAs and/or their precursors [27]. Continuing production of precursors of the long-chain PFCAs in Asia (including India and Russia) while reductions of these precursors occur in Europe and North America may explain some of this pattern.

Another temporal pattern that has emerged since 2010 is increasing PFCAs in biota following previous declines from higher concentrations in the early 2000s. This is most evident in ringed seals in the Canadian Arctic, East Greenland polar bears, and in arctic foxes in Svalbard. Annual biological sampling is helping to define this trend. The trend has not appeared in the data for atmospheric sampling of volatile precursors (FTOHs) or of PFOA. Nevertheless, the year to year variation is best addressed by continued annual sampling in order to maintain statistical power. Rigét et al. [7] have pointed out that only 13% of the datasets for persistent organochlorine compounds had sufficient sampling years to detect a 5% annual change with 80% power with $\alpha < 0.05$. Currently only a limited number of datasets for PFCAs (PFNA, PFDA, PFUnDA) would be in or near this category for arctic biota.

While the effectiveness of biological sampling for temporal trends in long-chain PFCAs has been demonstrated, this does not apply to the C_4–C_8–PFCAs or PFBS, which are generally present at low concentrations in biota. In addition to air sampling, sampling abiotic media such as glacial cores, and annual sampling of lake waters and seawater would appear to be the best approaches for investigating trends in the less bioaccumulative PFASs.

Personal communications and unpublished data

Acknowledgements

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.emcon.2019.06.002.

References

[1] R.C. Buck, J. Franklin, U. Berger, J.M. Conder, I.T. Cousins, P.D. Voogt, A.A. Jensen, K. Kannan, S.A. Mabury, S.P.J. van Leeuwen, Perfluoroalkyl and polyfluoroalkyl substances in the environment: terminology, classification, and origins, Integr. Environ. Assess. Manag. 7 (2011) 513–541.

[2] N. Yamashita, L.W.Y. Yeung, S. Taniyasu, K.Y. Kwok, G. Petrick, T. Gamo, K.S. Guruge, P.K.S. Lam, B.G. Loganathan, Global distribution of PFOS and related chemicals, in: B.G. Loganathan, P.K.S. Lam (Eds.), Global Contamination Trends of Persistent Organic Chemicals, Taylor & Francis Group, 2012, pp. 593–628.

[3] M. Houde, A. De Silva, R.J. Letcher, D.C.G. Muir, Monitoring of perfluorinated compounds in aquatic biota: an updated review, Environ. Sci. Technol. 45 (19) (2011) 7962–7973.

[4] C.M. Butt, U. Berger, R. Bossi, G.T. Tomy, Levels and trends of poly- and perfluorinated compounds in the arctic environment, Sci. Total Environ. 408 (2010) 2936–2965.

[5] R.J. Letcher, J.O. Bustnes, R. Dietz, B.M. Jenssen, E.H. Jørgensen, C. Sonne, J. Verreault, M.M. Vijayan, G.W. Gabrielsen, Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish, Sci. Total Environ. 408 (2010) 2995–3043.

[6] AMAP, AMAP Assessment, Chemicals of Emerging Arctic Concern, in: Arctic Monitoring and Assessment Programme (AMAP), 2016. Oslo, Norway.

xvi+353, 2017.

[7] F. Rigét, A. Bignert, B. Braune, M. Dam, R. Dietz, M. Evans, N. Green, H. Gunnlaugsdóttir, K.S. Hoydal, J. Kucklick, R. Letcher, D. Muir, S. Schuur, C. Sonne, G. Stern, G. Tomy, K. Vorkamp, S. Wilson, Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota, Sci. Total Environ. 649 (2019) 99—110.

[8] F. Wong, M. Shoeib, A. Katsoyiannis, S. Eckhardt, A. Stohl, P. Bohlin-Nizzetto, H. Li, P. Fellin, Y. Su, H. Hung, Assessing temporal trends and source regions of per- and polyfluoroalkyl substances (PFASs) in air under the Arctic Monitoring and Assessment Programme (AMAP), Atmos. Environ. 172 (2018) 65—73.

[9] A. Myers, E. Reiner, Northern contaminants program and arctic monitoring and assessment Programme interlaboratory study (NCP/AMAP — phase 11), in: Ontario Ministry of Environment & Climate Change, 2017, p. 74. Toronto ON + Appendix).

[10] Z. Wang, M. MacLeod, I.T. Cousins, M. Scheringer, K. Hungerbühler, Using COSMOtherm to predict physicochemical properties of poly- and per-fluorinated alkyl substances (PFASs), Environ. Chem. 8 (2011) 389—398.

[11] M.I. Gomis, Z. Wang, M. Scheringer, I.T. Cousins, A modeling assessment of the physicochemical properties and environmental fate of emerging and novel per- and polyfluoroalkyl substances, Sci. Total Environ. 505 (2015) 981—991.

[12] J. Cheng, E. Psillakis, M.R. Hoffmann, A.J. Colussi, Acid dissociation versus molecular association of perfluoroalkyl oxoacids: environmental implications, J. Phys. Chem. A 113 (2009) 8152—8156.

[13] L. Vierke, U. Berger, I.T. Cousins, Estimation of the acid dissociation constant of perfluoroalkyl carboxylic acids through an experimental investigation of their water-to-air transport, Environ. Sci. Technol. 47 (2013) 11032—11039.

[14] J.M. Armitage, J.A. Arnot, F. Wania, D. Mackay, Development and evaluation of a mechanistic bioconcentration model for ionogenic organic chemicals in fish, Environ. Toxicol. Chem. 32 (2013) 115—128.

[15] L. Ahrens, W. Gerwinski, N. Theobald, R. Ebinghaus, Sources of poly-fluoroalkyl compounds in the North Sea, Baltic Sea and Norwegian Sea: evidence from their spatial distribution in surface water, Mar. Pollut. Bull. 60 (2010) 255—260.

[16] C.P. Higgins, R.G. Luthy, Sorption of perfluorinated surfactants on sediments, Environ. Sci. Technol. 40 (2006) 7251—7256.

[17] K. Prevedouros, I.T. Cousins, R.C. Buck, S.H. Korzeniowski, Sources, fate and transport of perfluorocarboxylates, Environ. Sci. Technol. 40 (2006) 32—44.

[18] A.G. Paul, K.C. Jones, A.J. Sweetman, A first global production, emission, and environmental inventory for perfluorooctane sulfonate, Environ. Sci. Technol. 43 (2009) 386—392.

[19] Z. Wang, I.T. Cousins, M. Scheringer, R.C. Buck, K. Hungerbühler, Global emission inventories for C4-C14 perfluoroalkyl carboxylic acid (PFCA) ho-mologues from 1951 to 2030, part II: the remaining pieces of the puzzle, Environ. Int. 69 (2014) 166—176.

[20] Stockholm Convention, Chemicals proposed for listing, persistent organic pollutants review committee, in: Stockholm Convention on Persistent Organic Pollutants, United Nations Environment Programme, Geneva, CH, 2018. http://www.pops.int/TheConvention/ThePOPs/in.

[21] J.P. Benskin, A.O. De Silva, J.W. Martin, Isomer Profiling of Perfluorinated Substances as a Tool for Source Tracking, in: P. De Voogt (Ed.), Reviews of Environmental Contamination and Toxicology, Vol. 208, 2010, pp. 111—160.

[22] J.P. Benskin, L. Ahrens, D.C.G. Muir, B.F. Scott, C. Spencer, B. Rosenberg, G. Tomy, H. Kylin, R. Lohmann, J.W. Martin, Manufacturing origin of per-fluorooctanoate (PFOA) in Atlantic and Canadian Arctic seawater, Environ. Sci. Technol. 46 (2012) 677—685.

[23] US EPA, PFOA Stewardship Program Docket ID Number EPA-HQ-OPPT-2006-0621, US Environmental Protection Agency, Washington, DC, 2006.

[24] US EPA, EPA's Actions to Restrict PFOA and Similar Chemicals Yield Signifi-cant Human Health and Environmental Benefits, 2015. http://yosemite.epa.gov/opa/admpress.nsf/bd4379a92ceceeac8525735900400c27/d604d2dbf5f9c55c85257dce0065dfcf!OpenDocument. Accessed June 2015.

[25] W. Jiang, Y. Zhang, L. Yang, X. Chu, L. Zhu, Perfluoroalkyl acids (PFAAs) with isomer analysis in the commercial PFOS and PFOA products in China, Che-mosphere 127 (2015) 180—187.

[26] SAICM/ICCM4, Going global towards sound management of per-and poly-fluorinated chemistries, in: International Conference on Chemicals Man-agement (ICCM4), The Strategic Approach to International Chemicals Management, UNEP, Geneva, Switzerland, 2015.

[27] Z. Wang, I.T. Cousins, M. Scheringer, R.C. Buck, K. Hungerbühler, Global emission inventories for C4-C14 perfluoroalkyl carboxylic acid (PFCA) ho-mologues from 1951 to 2030, Part I: production and emissions from quan-tifiable sources, Environ. Int. 70 (2014) 62—75.

[28] J.M. Armitage, U. Schenker, M. Scheringer, J.W. Martin, M. MacLeod, I.T. Cousins, Modeling the global fate and transport of perfluorooctane sul-fonate (PFOS) and precursor compounds in relation to temporal trends in wildlife exposure, Environ. Sci. Technol. 43 (2009) 9274—9280.

[29] D.A. Ellis, J.W. Martin, A.O. De Silva, S.A. Mabury, M.D. Hurley, M.P.S. Andersen, T.J. Wallington, Degradation of fluorotelomer alcohols: a likely atmospheric source of perfluorinated carboxylic acids, Environ. Sci. Technol. 38 (2004) 3316—3321.

[30] J.C. D'Eon, M.D. Hurley, T.J. Wallington, S.A. Mabury, Atmospheric chemistry of N-methyl perfluorobutane sulfonamidoethanol, C4F9SO2N(CH3) CH2CH2OH: kinetics and mechanism of reaction with OH, Environ. Sci.

Technol. 40 (2006) 1862—1868.

[31] J. Armitage, M. MacLeod, I.T. Cousins, Modeling the global fate and transport of perfluorooctanoic acid (PFOA) and perfluorooctanoate (PFO) emitted from direct sources usinga multispecies mass balance model, Environ. Sci. Tech-nol. 43 (2009) 1134—1140.

[32] I. Stemmler, G. Lammel, Pathways of PFOA to the Arctic: variabilities and contributions of oceanic currents and atmospheric transport and chemistry sources, Atmos. Chem. Phys. 10 (2010) 9965—9980.

[33] Z. Zhao, Z. Xie, A. Möller, R. Sturm, J. Tang, G. Zhang, R. Ebinghaus, Distri-bution and long-range transport of polyfluoroalkyl substances in the Arctic, Atlantic Ocean and Antarctic coast, Environ. Pollut. 170 (2012) 71—77.

[34] J.H. Johansson, M.E. Salter, J.C. Acosta Navarro, C. Leck, E.D. Nilsson, I.T. Cousins, Global transport of perfluoroalkyl acids via sea spray aerosol, Environ. Sci. Processes Impacts 21 (2019) 635—649.

[35] Z. Wang, I.T. Cousins, M. Scheringer, K. Hungerbühler, Fluorinated alterna-tives to long-chain perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkane sulfonic acids (PFSAs) and their potential precursors, Environ. Int. 60 (2013) 242—248.

[36] C.J. Young, S.A. Mabury, Atmospheric perfluorinated acid precursors: chemistry, occurrence, and impacts, Rev. Environ. Contam. Toxicol. 208 (2010), 1-109.

[37] S.A. Styler, A.L. Myers, D.J. Donaldson, Heterogeneous photooxidation of fluorotelomer alcohols: a new source of aerosol-phase perfluorinated car-boxylic acids, Environ. Sci. Technol. 47 (2013) 6358—6367.

[38] C.M. Butt, D.C.G. Muir, S.A. Mabury, Biotransformation pathways of fluorotelomer-based polyfluoroalkyl substances: a review, Environ. Toxicol. Chem. 33 (2014) 243—267.

[39] A. Galatius, R. Bossi, C. Sonne, F.F. Rigét, C.C. Kinze, C. Lockyer, J. Teilmann, R. Dietz, PFAS profiles in three North Sea top predators: metabolic differ-ences among species? Environ. Sci. Pollut. Control Ser. 20 (2013) 8013—8020.

[40] R.J. Letcher, S. Chu, M.A. McKinney, G.T. Tomy, C. Sonne, R. Dietz, Compar-ative hepatic in vitro depletion and metabolite formation of major per-fluorooctane sulfonate precursors in arctic polar bear, beluga whale, and ringed seal, Chemosphere 112 (2014) 225—231.

[41] F. Wania, A global mass balance analysis of the source of perfluorocarboxylic acids in the Arctic Ocean, Environ. Sci. Technol. 41 (2007) 4529—4535.

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

[43] R. Lohmann, E. Jurado, H.A. Dijkstra, J. Dachs, Vertical eddy diffusion as a key mechanism for removing perfluorooctanoic acid (PFOA) from the global surface oceans, Environ. Pollut. 179 (2013) 88—94.

[44] R. Lohmann, E. Jurado, M.E.Q. Pilson, J. Dachs, Oceanic deep water formation as a sink of persistent organic pollutants, Geophys. Res. Lett. 33 (2006).

[45] X. Zhang, Y. Zhang, C. Dassuncao, R. Lohmann, E.M. Sunderland, North Atlantic Deep Water formation inhibits high Arctic contamination by con-tinental perfluorooctane sulfonate discharges, Glob. Biogeochem. Cycles 31 (2017) 1332—1343.

[46] N. Yamashita, K. Kannan, S. Taniyasu, Y. Horii, G. Petrick, T. Gamo, A global survey of perfluorinated acids in oceans, Mar. Pollut. Bull. 51 (2005) 658—668.

[47] L.W.Y. Yeung, C. Dassuncao, S. Mabury, E.M. Sunderland, X. Zhang, R. Lohmann, Vertical profiles, sources, and transport of PFASs in the Arctic Ocean, Environ. Sci. Technol. 51 (2017) 6735—6744.

[48] A.L. Soerensen, D.J. Jacob, A.T. Schartup, J.A. Fisher, I. Lehnherr, V.L. St Louis, L.-E. Heimbürger, J.E. Sonke, D.P. Krabbenhoft, E.M. Sunderland, A mass budget for mercury and methylmercury in the Arctic Ocean, Glob. Bio-geochem. Cycles 30 (2016) 560—575.

[49] T.J. Wallington, M.D. Hurley, J. Xia, D.J. Wuebbles, S. Sillman, A. Ito, J.E. Penner, D.A. Ellis, J. Martin, S.A. Mabury, O.J. Nielsen, M.P. Sulbaek Andersen, formation of C7F15COOH (PFOA) and other perfluorocarboxylic acids during the atmospheric oxidation of 8:2 fluorotelomer alcohol, Envi-ron. Sci. Technol. 40 (2006) 924—930.

[50] A. Dreyer, R. Ebinghaus, Polyfluorinated compounds in ambient air from ship- and land-based measurements in northern Germany, Atmos. Environ. 43 (2009) 1527—1535.

[51] C.E. Müller, A.C. Gerecke, C. Bogdal, Z. Wang, M. Scheringer, K. Hungerbühler, Atmospheric fate of poly- and perfluorinated alkyl substances (PFASs): I. Day-night patterns of air concentrations in summer in Zurich, Switzerland, Environ. Pollut. 169 (2012) 196—203.

[52] L. Ahrens, T. Harner, M. Shoeib, M. Koblizkova, E.J. Reiner, Characterization of two passive air samplers for per- and polyfluoroalkyl substances, Environ. Sci. Technol. 47 (2013) 14024—14033.

[53] NILU, Monitoring of environmental contaminants in air and precipitation, annual report 2014. M-368, in: D. Herzke (Ed.), NILU, Norwegian Institute for Air Research, Kjeller Norway, 2015, p. 90.

[54] M. Schlabach, B. van Bavel, J.A. Baz Lomba, A. Borgen, G.W. Gabrielsen, A. Götsch, A.-K. Halse, L. Hanssen, I. Sunde Krogseth, V. Nikiforov, T. Nygård, P. Bohlin Nizzetto, M. Reid, P. Rostkowski, S. Samanipour, Screening Pro-gramme, AMAP Assessment Compounds. M-1080, Norwegian Institute for Air Research, Kjeller, No. 2018, 2018, p. 93.

[55] H. Skov, A. Massling, I.E. Nielsen, I. Nordstrøm, R. Bossi, K. Vorkamp, J. Christensen, M.M. Larsen, K.M. Hansen, J. Liisberg, M.B. Poulsen, AMAP CORE - atmospheric part. Results from Villum Research Station. Technical report from DCE - Danish centre for environment and energy No. 101, in:

Aarhus University, DCE - Danish Centre for Environment and Energy, 2017, p. 77.

[56] M. Cai, Z. Zhao, Z. Yin, L. Ahrens, P. Huang, M. Cai, H. Yang, J. He, R. Sturm, R. Ebinghaus, Z. Xie, Occurrence of perfluoroalkyl compounds in surface waters from the North Pacific to the Arctic Ocean, Environ. Sci. Technol. 46 (2012) 661–668.

[57] M. Shoeib, T. Harner, P. Vlahos, Perfluorinated chemicals in the arctic atmosphere, Environ. Sci. Technol. 40 (2006) 7577–7583.

[58] L. Ahrens, M. Shoeib, S. Del Vento, G. Codling, C. Halsall, Polyfluoroalkyl compounds in the Canadian Arctic atmosphere, Environ. Chem. 8 (2011) 399–406.

[59] Z. Xie, Z. Zhao, A. Möller, H. Wolschke, L. Ahrens, R. Sturm, R. Ebinghaus, Neutral poly- and perfluoroalkyl substances in air and seawater of the North Sea, Environ. Sci. Pollut. Res. 20 (2013) 7988–8000.

[60] Z. Xie, Z. Wang, W. Mi, A. Möller, H. Wolschke, R. Ebinghaus, Neutral poly-/perfluoroalkyl substances in air and snow from the arctic, Sci. Rep. 5 (2015).

[61] C. Rauert, M. Shoieb, J.K. Schuster, A. Eng, T. Harner, Atmospheric concentrations and trends of poly- and perfluoroalkyl substances (PFAS) and volatile methyl siloxanes (VMS) over 7 years of sampling in the Global Atmospheric Passive Sampling (GAPS) network, Environ. Pollut. 238 (2018) 94–102.

[62] S. Genualdi, S.C. Lee, M. Shoeib, A. Gawor, L. Ahrens, T. Harner, Global pilot study of legacy and emerging persistent organic pollutants using sorbent-impregnated polyurethane foam disk passive air samplers, Environ. Sci. Technol. 44 (2010) 5534–5539.

[63] C.J. Young, M.D. Hurley, T.J. Wallington, S.A. Mabury, Atmospheric chemistry of CF_3CF_2H and $CF_3CF_2CF_2H$: kinetics and products of gas-phase reactions with Cl atoms and OH radicals, infrared spectra, and formation of perfluorocarboxylic acids, Chem. Phys. Lett. 473 (2009) 251–256.

[64] T.J. Wallington, W.F. Schneider, J. Sehested, M. Bilde, J. Platz, O.J. Nielsen, L.K. Christensen, M.J. Molina, L.T. Molina, P.W. Wooldridge, Atmospheric chemistry of HFE-7100 (C4F9OCH3): reaction with OH radicals, UV spectra and kinetic data for C4F9OCH2· and C4F9OCH2O2· radicals, and the atmospheric fate of C4F9OCH2O· radicals, J. Phys. Chem. A 101 (1997) 8264–8274.

[65] L. Chen, T. Uchimaru, S. Kutsuna, K. Tokuhashi, A. Sekiya, Kinetics and mechanism of gas-phase reactions of n-C4F9OCH3, i-C4F9OCH3, n-C4F9OC(O)H, and i-C4F9OC(O)H with OH radicals in an environmental reaction chamber at 253–328 K, Chem. Phys. Lett. 514 (2011) 207–213.

[66] A. Gawor, C. Shunthirasingham, S.J. Hayward, Y.D. Lei, T. Gouin, B.T. Mmereki, W. Masamba, C. Ruepert, L.E. Castillo, M. Shoeib, S.C. Lee, T. Harner, F. Wania, Neutral polyfluoroalkyl substances in the global Atmosphere, Environ. Sci.: Processes Impacts 16 (2014) 404–413.

[67] AMAP, ArcRisk (arctic health risks: impacts on health in the arctic and Europe owing to climate-induced changes in contaminant cycling) results overview, in: Arctic Monitoring and Assessment Program Oslo, 2014, p. 62.

[68] O.R. Bertrand, C.J. Halsall, D. Herzke, S. Huber, T. Nordstad, S. del Vento, E.S. Heimstad, Short-term fluctuations in the levels of poly- and perfluoroalkyl Substances (PFASs) in the seasonal Snowpack, in: Arctic Frontiers, Tromsoe, Norway, 2014.

[69] K.Y. Kwok, E. Yamazaki, N. Yamashita, S. Taniyasu, M.B. Murphy, Y. Horii, G. Petrick, R. Kallerborn, K. Kannan, K. Murano, P.K.S. Lam, Transport of Perfluoroalkyl substances (PFAS) from an arctic glacier to downstream locations: implications for sources, Sci. Total Environ. 447 (2013) 46–55.

[70] J.J. MacInnis, I. Lehnherr, D.C.G. Muir, R. Quinlan, A.O. De Silva, Fate and transport of perfluoroalkyl substances from snowpacks into a lake in the High Arctic of Canada, Sci. Total Environ. 666 (2019) 414–422.

[71] J. Veillette, D.C.G. Muir, D. Antoniades, J.M. Small, C. Spencer, T.N. Loewen, J.A. Babaluk, J.D. Reist, W.F. Vincent, Perfluorinated chemicals in meromictic lakes on the northern coast of Ellesmere Island, High Arctic Canada, Arctic 65 (2012) 245–256.

[72] C.J. Young, V.I. Furdui, J. Franklin, R.M. Koerner, D.C.G. Muir, S.A. Mabury, Perfluorinated acids in arctic snow: new evidence for atmospheric formation, Environ. Sci. Technol. 41 (2007) 3455–3461.

[73] J.J. MacInnis, K. French, D.C.G. Muir, C. Spencer, A. Criscitiello, A.O. De Silva, C.J. Young, Emerging investigator series: a 14-year depositional ice record of perfluoroalkyl substances in the High Arctic, Environ. Sci.: Processes Impacts 19 (2017) 22–30.

[74] D.O. Carpenter, S. Reimann, J.B. Burkholder, C. Clerbaux, B.D. Hall, R. Hossaini, J.C. Laube, S.A. Yvon-Lewis, Update on ozone-depleting substances (ODSs) and other gases of interest to the montreal protocol, in: Scientific Assessment of Ozone Depletion: 2014, Global Ozone Research and Monitoring Project - Report No. 55, World Meteorological Organization, Geneva, Switzerland, 2014, p. 416.

[75] S. Posner, S. Roos, P. Brunn Poulsen, H.Ó. Jörundsdottir, H. Gunnlaugsdottir, X. Trier, A. Jensen, A. Katsogiannis, A. Athanasios, D. Herzke, E. Bonefeld-Jörgensen, C. Jönsson, C. Pedersen, G. Alsing, M. Ghisari, S. Jensen, Per and polyfluorinated substances in the Nordic Countries: use, occurrence and toxicology, Nordic Council of Ministers. TeamNord 542 (2013), https://doi.org/10.6027/TN2013-542, 2013.

[76] G. Codling, C. Halsall, L. Ahrens, S. Del Vento, K. Wiberg, M. Bergknut, H. Laudon, R. Ebinghaus, The fate of per- and polyfluoroalkyl substances within a melting snowpack of a boreal forest, Environ. Pollut. 191 (2014) 190–198.

[77] C.E. Müller, A.O. De Silva, J. Small, M. Williamson, X. Wang, A. Morris, S. Katz,

M. Gamberg, D.C.G. Muir, Biomagnification of perfluorinated compounds in a remote terrestrial food chain: lichen-caribou-wolf, Environ. Sci. Technol. 45 (2011) 8665–8673.

[78] R. Bossi, M. Dam, F.F. Rigét, Perfluorinated alkyl substances (PFAS) in terrestrial environments in Greenland and Faroe Islands, Chemosphere 129 (2015) 164–169.

[79] S. Danielsson, T. Odsjö, A. Bignert, M. Remberger, Organic contaminants in moose (*Alces alces*) and reindeer (*Rangifer tarandus*) in Sweden from the past twenty years, in: Department of Contaminant Research, Swedish Museum of Natural History, 2008, p. 43. Stockholm, Sweden.

[80] P. Carlsson, R. Kallenborn, D. Herzke, PFCs in Svalbard reindeer. A combination of educational outreach and environmental research, in: International Polar Year Conference, 2012. Montreal, Canada.

[81] Norwegian Environment Agency, Perfluorinated alkylated substances, brominated flame retardants and chlorinated paraffins in the Norwegian Environment - screening 2013, in: D. Herzke NILU (Ed.), M-40, Norwegian Environment Agency, 2013, p. 110.

[82] C.B. Aas, E. Fuglei, D. Herzke, N.G. Yoccoz, H. Routti, Effect of body condition on tissue distribution of perfluoroalkyl substances (PFASs) in Arctic fox (Vulpes lagopus), Environ. Sci. Technol. 48 (2014) 11654–11661.

[83] H. Routti, J. Aars, E. Fuglei, L. Hanssen, K. Lone, A. Polder, Å.O. Pedersen, S. Tartu, J.M. Welker, N.G. Yoccoz, Emission changes dwarf the influence of feeding habits on temporal trends of per- and polyfluoroalkyl substances in two arctic top predators, Environ. Sci. Technol. 51 (2017) 11996–12006.

[84] N.C. Larter, D.C.G. Muir, X. Wang, D. Allaire, A. Kelly, K. Cox, Persistent organic pollutants in the livers of moose harvested in the Southern Northwest Territories, Canada, Alces 53 (2017) 65–83.

[85] N.L. Stock, V.I. Furdui, D.C.G. Muir, S.A. Mabury, Perfluoroalkyl contaminants in the Canadian arctic: evidence of atmospheric transport and local contamination, Environ. Sci. Technol. 41 (2007) 3529–3536.

[86] G.L. Lescord, K.A. Kidd, A. De Silva, C. Spencer, M. Williamson, X. Wang, D.C.G. Muir, Perfluorinated and polyfluorinated compounds in lake food webs in the Canadian high arctic, Environ. Sci. Technol. 49 (2015) 2694–2702.

[87] B.F. Scott, C. Spencer, S.A. Mabury, D.C.G. Muir, Poly and perfluorinated carboxylates in North American precipitation, Environ. Sci. Technol. 40 (2006) 7167–7174.

[88] C.A. Moody, J.W. Martin, W.C. Kwan, D.C. Muir, S.A. Mabury, Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek, Environ. Sci. Technol. 36 (2002) 545–551.

[89] B. Scott, C. Spencer, D. Muir, Analysis of aqueous film forming foam (AFFF) for the department of national defense (DND). WSTD contribution No. 07-066, in: Water Science & Technology Directorate, Environment Canada, Burlington, 2007, p. 16.

[90] C.A. Moody, J.A. Field, Perfluorinated surfactants and the environmental implications of their use in fire-fighting foams, Environ. Sci. Technol. 34 (2000) 3864–3870.

[91] M.M. Schultz, D.F. Barofsky, J.A. Field, Quantitative determination of fluorotelomer sulfonates in groundwater by LC MS/MS, Environ. Sci. Technol. 38 (2004) 1828–1835.

[92] W.J. Backe, T.C. Day, J.A. Field, Zwitterionic, cationic, and anionic fluorinated chemicals in aqueous film forming foam formulations and groundwater from U.S. military bases by nonaqueous large-volume injection HPLC-MS/MS, Environ. Sci. Technol. 47 (2013) 5226–5234.

[93] A.O. De Silva, C. Spencer, B.F. Scott, S. Backus, D.C.G. Muir, Detection of a cyclic perfluorinated acid, perfluoroethylcyclohexane sulfonate, in the Great Lakes of North America, Environ. Sci. Technol. 45 (2011) 8060–8066.

[94] N. Gantner, J. Veillette, W.K. Michaud, R. Bajno, D.C.G. Muir, W.F. Vincent, M. Power, B. Dixon, J.D. Reist, S. Hausmann, R. Pienitz, Physical and biological factors affecting mercury and perfluorinated contaminants in Arctic char (Salvelinus alpinus) of Pingualuk Crater Lake (Nunavik, Canada), Arctic 65 (2012) 195–206.

[95] H.M. Pickard, A.S. Criscitiello, C. Spencer, M.J. Sharp, D.C.G. Muir, A.O. De Silva, C.J. Young, Continuous non-marine inputs of per- and polyfluoroalkyl substances to the High Arctic: a multi-decadal temporal record, Atmos. Chem. Phys. 18 (2018) 5045–5058.

[96] U. Eriksson, A. Kärrman, A. Rotander, B. Mikkelsen, M. Dam, Perfluoroalkyl substances (PFASs) in food and water from Faroe Islands, Environ. Sci. Pollut. Control Ser. 20 (2013) 7940–7948.

[97] V.L. St Louis, D.C.G. Muir, Quantifying contaminant loadings, water quality and climate change impacts in the World's largest lake north of 74° latitude (Lake Hazen, Quttinirpaaq National Park, northern Ellesmere island, Nunavut), in: Synopsis of Research Conducted under the 2013-2014 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2014, pp. 351–359.

[98] J.J. MacInnis, I. Lehnherr, D.C.G. Muir, R. Quinlan, A.O. De Silva, Characterization of perfluoroalkyl substances in sediment cores from high and low arctic lakes in Canada, Sci. Total Environ. 666 (2019) 414–422.

[99] D.C.G. Muir, P. Kurt-Karakus, J. Stow, J. Blais, B. Braune, E. Choy, M. Evans, B. Kelly, N. Larter, R. Letcher, M. McKinney, A. Morris, G. Stern, G. Tomy, Occurrence and trends in the biological environment. Chapter 4., in: D.C.G. Muir, P. Kurt-Karakus, J.E. Stow (Eds.), Persistent Organic Pollutants in Canada's North, Aboriginal Affairs and Northern Development Canada, 2013, pp. 273–422. Ottawa ON.

[100] G.A. Stern, P. Roach, B. van Dijken, A. Burt, Trace metals and organohalogen contaminants in fish from selected Yukon lakes: a temporal and spatial study, in: Synopsis of Research Conducted under the 2013-2014 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2014, pp. 281–289.

[101] M. Garsjø, Perfluorinated alkylated substances (PFAS) in arctic char (Salvelinus alpinus): a case study from Svalbard. MSc thesis, in: Department of Chemistry, Biotechnology and Food Science, Norwegian University of Life Sciences, University Centre in Svalbard, 2014.

[102] G.A. Stern, J. Carrie, A. Burt, Temporal trend studies of trace metals and halogenated organic contaminants (HOCs), including new and emerging persistent compounds, in Mackenzie River burbot, Fort Good Hope, NWT, in: Synopsis of Research Conducted under the 2013-2014 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, 2014, pp. 271–280. Ottawa, ON.

[103] G.A. Stern, J. Carrie, S. Friesen, J. DeLaronde, C. Fuchs, G. Boila, Temporal trend studies of trace metals and halogenated organic contaminants (HOCs), including new and emerging persistent compounds, in Mackenzie River burbot, Fort Good Hope, NWT, in: Synopsis of Research Conducted under the 2011-2012 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, 2012, pp. 224–235. Ottawa, ON.

[104] L. Ahrens, Z. Xie, R. Ebinghaus, Distribution of perfluoroalkyl compounds in seawater from northern Europe, Atlantic Ocean, and southern ocean, Chemosphere 78 (2010) 1011–1016.

[105] C. Caliebe, W. Gerwinski, N. Theobald, H. Hühnerfuss, Occurrence of perfluorinated organic acids in the water of the north Sea and arctic North Atlantic, in: Fluoros, 2005. Toronto, Canada.

[106] J.P. Benskin, D.C.G. Muir, B.F. Scott, C. Spencer, A.O. De Silva, H. Kylin, J.W. Martin, A. Morris, R. Lohmann, G. Tomy, B. Rosenberg, S. Taniyasu, N. Yamashita, Perfluorinated compounds in the arctic and atlantic oceans, Environ. Sci. Technol. 46 (2012) 5815–5823.

[107] B. Rosenberg, J. DeLaronde, A. MacHutchon, G. Stern, C. Spencer, B. Scott, E. Lopez, D. Muir, G. Tomy, Spatial and vertical distribution of perfluorinated compounds in Canadian arctic and sub-arctic Ocean Water, Organohalogen Compd. 70 (2008) 386–390.

[108] D.C.G. Muir, A. Fisk, I. Lehnherr, R. Lohmann, P. Amarualik, Community based seawater monitoring for organic contaminants and mercury in the Canadian Arctic, in: Synopsis of Research Conducted under the 2014-2015, Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa., Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2015, pp. 289–296.

[109] J. Busch, L. Ahrens, Z. Xie, R. Sturm, R. Ebinghaus, Polyfluoroalkyl compounds in the east Greenland Arctic Ocean, J. Environ. Monit. 12 (2010) 1242–1246.

[110] S.P.J. Van Leeuwen, C.P. Swart, I. Van der Veen, J. De Boer, Significant improvement in the analysis of perfluorinated compounds in water and fish: results from an interlaboratory method evaluation study, J. Chromatogr., A 1216 (2009) 401–409.

[111] J.L. Murray, L. Hacquebord, D.J. Gregor, H. Loeng, Physical/geographical characteristics of the arctic. Chapter 2., in: J.L. Murray (Ed.), AMAP Assessment Report. Arctic Pollution Issues, Arctic Monitoring and Assessment Programme (AMAP), 1998, pp. 9–24. Oslo, Norway.

[112] J.W. McClelland, R.M. Holmes, K.H. Dunton, R.W. Macdonald, The Arctic Ocean estuary, Estuar. Coasts 35 (2012) 353–368.

[113] O. Bertrand, S. Del Vento, C.J. Halsall, G. Codling, L. Ahrens, D. Herzke, S. Huber, P. Carlsson, R. Kallenborn, K.C. Jones, R. Ebinghaus, Enrichment of perfluoroalkyl substances in arctic sea-ice. Poster presentation (MO190), in: SETAC Europe 23rd Annual Meeting, May 2013., 2013. Glasgow, UK.

[114] Y. Miyake, N. Yamashita, P. Rostkowski, M.K. So, S. Taniyasu, P.K.S. Lam, K. Kannan, Determination of trace levels of total fluorine in water using combustion ion chromatography for fluorine: a mass balance approach to determine individual perfluorinated chemicals in water, J. Chromatogr. A 1143 (2007) 98–104.

[115] P. Carlsson, J.D. Crosse, C. Halsall, A. Evenset, E.S. Heimstad, M. Harju, Perfluoroalkylated substances (PFASs) and legacy persistent organic pollutants (POPs) in halibut and shrimp from coastal areas in the far north of Norway: small survey of important dietary foodstuffs for coastal communities, Mar. Pollut. Bull. 105 (2016) 81–87.

[116] H.Ó. Jörundsdóttir, V. Baldursdóttir, N. Desnica, P. Ragnarsdóttir, H. Gunnlaugsdóttir, Undesirable substances in seafood products - results from the Icelandic marine monitoring activities in the year 2011, in: Icelandic Ministry of Fisheries and Agriculture, 2012.

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

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

[119] B.M. Braune, A.J. Gaston, K.H. Elliott, J.F. Provencher, K.J. Woo, M. Chambellant, S.H. Ferguson, R.J. Letcher, Organohalogen contaminants and total mercury in forage fish preyed upon by thick-billed murres in northern Hudson Bay, Mar. Pollut. Bull. 78 (2014) 258–266.

[120] AMAP, Trends in Stockholm convention persistent organic pollutants (POPs) in arctic air, human media and biota. AMAP technical report No. 7, in: Arctic Monitoring and Assessment Program, 2014, p. 62. Oslo, Norway.

[121] F. Rigét, R. Bossi, C. Sonne, K. Vorkamp, R. Dietz, Trends of perfluorochemicals in Greenland ringed seals and polar bears: indications of shifts to decreasing trends, Chemosphere 93 (2013) 1607–1614.

[122] H. Routti, G.W. Gabrielsen, D. Herzke, K.M. Kovacs, C. Lydersen, Spatial and temporal trends in perfluoroalkyl substances (PFASs) in ringed seals (Pusa hispida) from Svalbard, Environ. Pollut. 214 (2016) 230–238.

[123] S.E. Scotter, M. Tryland, I.H. Nymo, L. Hanssen, M. Harju, C. Lydersen, K.M. Kovacs, J. Klein, A.T. Fisk, H. Routti, Contaminants in Atlantic Walruses in Svalbard Part 1: Relationships between Exposure, Diet and Pathogen Prevalence, Environ. Pollut. 244 (2019) 9–18.

[124] H. Routti, C. Lydersen, L. Hanssen, K.M. Kovacs, Contaminant levels in the world's northernmost harbor seals (Phoca vitulina), Mar. Pollut. Bull. 87 (2014) 140–146.

[125] M. Houde, J.W. Martin, R.J. Letcher, K.R. Solomon, D.C.G. Muir, Biological monitoring of polyfluoroalkyl substances: a review, Environ. Sci. Technol. 40 (2006) 3463–3473.

[126] A. Rotander, A. Kärrman, B.van Bavel, A. Polder, F. Rigét, G.A. Auðunsson, G. Víkingsson, G.W. Gabrielsen, D. Bloch, M. Dam, Increasing levels of long-chain perfluorocarboxylic acids (PFCAs) in Arctic and North Atlantic marine mammals, Chemosphere 86 (2012) 278–285, 1984–2009.

[127] D.C.G. Muir, X. Wang, E. Sverko, E. Baressi, M. Williamson, Temporal trends of persistent organic pollutants and metals in ringed seals from the Canadian Arctic, in: Synopsis of Research Conducted under the 2013-2014 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2014, pp. 187–194.

[128] W.A. Gebbink, R. Bossi, F.F. Rigét, A. Rosing-Asvid, C. Sonne, R. Dietz, Observation of emerging per- and polyfluoroalkyl substances (PFASs) in Greenland marine mammals, Chemosphere 144 (2016) 2384–2391.

[129] D.C.G. Muir, X. Wang, M. Houde, Temporal trends of persistent organic pollutants and metals in ringed seals from the Canadian Arctic, in: Synopsis of Research Conducted under the 2014-2015, Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2015, pp. 189–198.

[130] S. Wang, J. Huang, Y. Yang, Y. Hui, Y. Ge, T. Larssen, G. Yu, S. Deng, B. Wang, C. Harman, First report of a Chinese PFOS alternative overlooked for 30 years: its toxicity, persistence, and presence in the environment, Environ. Sci. Technol. 47 (2013) 10163–10170.

[131] T. Ruan, Y. Lin, T. Wang, R. Liu, G.B. Jiang, Identification of novel polyfluorinated ether sulfonates as PFOS alternatives in municipal sewage sludge in China, Environ. Sci. Technol. 49 (2015) 6519–6527.

[132] D.C.G. Muir, M. Houde, T.-L.L. Colson, X. Wang, M. Williamson, C. Butt, S.F. Ferguson, R.F. Addison, Trends of Perfluoro- and Polyfluoroalkyl Substances (PFAS) in Ringed Seals (Phoca Hispida) from the Canadian Arctic, 2019 (1972-2016), Unpublished data, Environment & Climate Change Canada, Burlington ON.

[133] J.L. Reiner, S.G. O'Connell, A.J. Moors, J.R. Kucklick, P.R. Becker, J.M. Keller, Spatial and temporal trends of perfluorinated compounds in beluga whales (Delphinapterus leucas) from Alaska, Environ. Sci. Technol. 45 (2011) 8129–8136.

[134] B.C. Kelly, M.G. Ikonomou, J.D. Blair, B. Surridge, D. Hoover, R. Grace, F.A.P.C. Gobas, Perfluoroalkyl contaminants in an arctic marine food web: trophic magnification and wildlife exposure, Environ. Sci. Technol. 43 (2009) 4037–4043.

[135] T.A. Smythe, L.L. Loseto, A. Bignert, B. Rosenberg, W. Budakowski, T. Halldorson, K. Pleskach, G.T. Tomy, Temporal trends of brominated and fluorinated contaminants in Canadian Arctic beluga (Delphinapterus leucas), Arct. Sci. 4 (2018) 388–404.

[136] J. Bytingsvik, S.P.J. van Leeuwen, T. Hamers, K. Swart, J. Aars, E. Lie, E.M.E. Nilsen, T. Wiig, A.E. Derocher, B.M. Jenssen, Perfluoroalkyl substances in polar bear mother-cub pairs: a comparative study based on plasma levels from 1998 and 2008, Environ. Int. 49 (2012) 92–99.

[137] S. Tartu, S. Bourgeon, J. Aars, M. Andersen, K. Lone, B.M. Jenssen, A. Polder, G.W. Thiemann, V. Torget, J.M. Welker, H. Routti, Diet and metabolic state are the main factors determining concentrations of perfluoroalkyl substances in female polar bears from Svalbard, Environ. Pollut. 229 (2017) 146–158.

[138] S. Tartu, J. Aars, M. Andersen, A. Polder, S. Bourgeon, B. Merkel, A.D. Lowther, J. Bytingsvik, J.M. Welker, A.E. Derocher, B.M. Jenssen, H. Routti, Choose your poison - space-use strategy influences pollutant exposure in Barents Sea polar bears, Environ. Sci. Technol. 52 (2018) 3211–3221.

[139] S. Tartu, R. Lille-Langøy, T.R. Størseth, S. Bourgeon, A. Brunsvik, J. Aars, A. Goksøyr, B.M. Jenssen, A. Polder, G.W. Thiemann, V. Torget, H. Routti, Multiple-stressor effects in an apex predator: combined influence of pollutants and sea ice decline on lipid metabolism in polar bears, Sci. Rep. 7 (2017).

[140] A.K. Greaves, R.J. Letcher, C. Sonne, R. Dietz, E.W. Born, Tissue-specific concentrations and patterns of perfluoroalkyl carboxylates and sulfonates in east Greenland polar bears, Environ. Sci. Technol. 46 (2012) 11575–11583.

[141] A.K. Greaves, R.J. Letcher, C. Sonne, R. Dietz, Brain region distribution and patterns of bioaccumulative perfluoroalkyl carboxylates and sulfonates in east Greenland polar bears (Ursus maritimus), Environ. Toxicol. Chem. 32 (2013) 713–722.

[142] R.J. Letcher, A.D. Morris, M. Dyck, E. Sverko, E.J. Reiner, D.A.D. Blair, S.G. Chu, L. Shen, Legacy and new halogenated persistent organic pollutants in polar bears from a contamination hotspot in the Arctic, Hudson Bay Canada, Sci. Total Environ. 610–611 (2018) 121–136.

[143] Y. Liu, E.S. Richardson, A.E. Derocher, N.J. Lunn, H.-J. Lehmler, X. Li, Y. Zhang, J.Y. Cui, L. Cheng, J.W. Martin, Hundreds of unrecognized halogenated contaminants discovered in polar bear serum, Angew. Chem. Int. Ed. 57 (2018) 16401—16406.

[144] E.L. Schymanski, H.P. Singer, J. Slobodnik, I.M. Ipolyi, P. Oswald, M. Krauss, T. Schulze, P. Haglund, T. Letzel, S. Grosse, N.S. Thomaidis, A. Bletsou, C. Zwiener, M. Ibáñez, T. Portolés, R. De Boer, M.J. Reid, M. Onghena, U. Kunkel, W. Schulz, A. Guillon, N. Noyon, G. Leroy, P. Bados, S. Bogialli, D. Stipaničev, P. Rostkowski, J. Hollender, Non-target screening with high-resolution mass spectrometry: critical review using a collaborative trial on water analysis, Anal. Bioanal. Chem. 407 (2015) 6237—6255.

[145] C. Miljeteig, H. Strøm, M.V. Gavrilo, A. Volkov, B.M. Jenssen, G.W. Gabrielsen, High levels of contaminants in ivory gull pagophila eburnea eggs from the Russian and Norwegian arctic, Environ. Sci. Technol. 43 (2009) 5521—5528.

[146] M. Lucia, N. Verboven, H. Strøm, C. Miljeteig, M.V. Gavrilo, B.M. Braune, D. Boertmann, G.W. Gabrielsen, Circumpolar contamination in eggs of the high-arctic ivory gull Pagophila eburnea, Environ. Toxicol. Chem. 34 (2015) 1552—1561.

[147] T.H. Nøst, L.B. Helgason, M. Harju, E.S. Heimstad, G.W. Gabrielsen, B.M. Jenssen, Halogenated organic contaminants and their correlations with circulating thyroid hormones in developing Arctic seabirds, Sci. Total Environ. 414 (2012) 248—256.

[148] S. Tartu, G.W. Gabrielsen, P. Blévin, H. Ellis, J.O. Bustnes, D. Herzke, O. Chastel, Endocrine and fitness correlates of long-chain perfluorinated carboxylates exposure in arctic breeding black-legged kittiwakes, Environ. Sci. Technol. 48 (2014) 13504—13510.

[149] B.M. Braune, A.J. Gaston, R.J. Letcher, H. Grant Gilchrist, M.L. Mallory, J.F. Provencher, A geographical comparison of chlorinated, brominated and fluorinated compounds in seabirds breeding in the eastern Canadian Arctic, Environ. Res. 134 (2014) 46—56.

[150] B.M. Braune, R.J. Letcher, Perfluorinated sulfonate and carboxylate compounds in eggs of seabirds breeding in the Canadian Arctic: temporal trends (1975-2011) and interspecies comparison, Environ. Sci. Technol. 47 (2013) 616—624.

[151] W.A. Gebbink, R.J. Letcher, Comparative tissue and body compartment accumulation and maternal transfer to eggs of perfluoroalkylsulfonates and carboxylates in Great Lakes herring gulls, Environ. Pollut. 162 (2012) 40—47.

[152] S. Huber, N.A. Warner, T. Nygård, M. Remberger, M. Harju, H.T. Uggerud, L. Kaj, L. Hanssen, A broad cocktail of environmental pollutants found in eggs of three seabird species from remote colonies in Norway, Environ. Toxicol. Chem. 34 (2015) 1296—1308.

[153] D.C.G. Muir, A.O. de Silva, J. Kirk, A. Fisk, I. Lehnherr, R. Lohmann, P. Amarualik, Community based seawater monitoring for organic contaminants and mercury in the Canadian Arctic, in: Synopsis of Research Conducted under the 2014-2015, Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2015, pp. 289—296.

[154] H. Hung, R. Kallenborn, K. Breivik, Y. Su, E. Brorström-Lundén, K. Olafsdottir, J.M. Thorlacius, S. Leppänen, R. Bossi, H. Skov, S. Manø, G.W. Patton, G. Stern, E. Sverko, P. Fellin, Atmospheric monitoring of organic pollutants in the arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993-2006, Sci. Total Environ. 408 (2010) 2854—2873.

[155] 3M Company, Phase-out plan for POSF-based products, in: Specialty Materials Markets Group, U.S. EPA Docket OPPT-2002-0043, St.Paul, MN, 2000, 3M.

[156] H.M. Pickard, A.S. Criscitiello, C. Spencer, M.J. Sharp, D.C.G. Muir, A.O. De Silva, C.J. Young, Long-term Record of Persistent Short-Chain Fluorinated Acids in the Arctic, 2019 submitted for publication.

[157] Z. Wang, J.M. Boucher, M. Scheringer, I.T. Cousins, K. Hungerbühler, Toward a comprehensive global emission inventory of C4—C10 perfluoroalkanesulfonic acids (PFSAs) and related precursors: focus on the life cycle of C8-based products and ongoing industrial transition, Environ. Sci. Technol. 51 (2017) 4482—4493.

[158] I. Lehnherr, V.L. St Louis, M. Sharp, A.S. Gardner, J.P. Smol, S.L. Schiff, D.C.G. Muir, C.A. Mortimer, N. Michelutti, C. Tarnocai, K.A. St Pierre, C.A. Emmerton, J.A. Wiklund, G. Köck, S.F. Lamoureux, C.H. Talbot, The world's largest High Arctic lake responds rapidly to climate warming, Nat. Commun. 9 (2018).

[159] A. Bignert, PIA statistical application developed for use by the Arctic Monitoring and Assessment Programme, Arctic Monitoring and Assessment Programme, Oslo, No, 2007, p. 13, available from: www.amap.no.

[160] R.F. Addison, D.C. Muir, M.G. Ikonomou, L. Harwood, T.G. Smith, J. Alikamik, Temporal trends in "legacy" organochlorine contaminants in blubber of ringed seals (Phoca hispida) from Ulukhaktok, NT, Canada between 1972 and 2010, Sci. Total Environ. 466—467 (2014) 564—576.

[161] Y.F. Li, R.W. Macdonald, Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review, Sci. Total Environ. 342 (2005) 87—106.

[162] R. Dietz, R. Bossi, F.F. Rigét, C. Sonne, E.W. Born, Increasing perfluoroalkyl contaminants in East Greenland polar bears (Ursus maritimus): a new toxic threat to the arctic bears, Environ. Sci. Technol. 42 (2008) 2701—2707.

[163] B. Braune, Temporal trends of contaminants in arctic seabird eggs, in: Synopsis of Research Conducted under the 2013-2014 Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa, ON, 2015, pp. 229—238.

[164] TemaNord, Arctic human development report. Regional processes and global linkages, in: J.N. Larsen, G. Fondahl (Eds.), TemaNord 2014:567, Nordic Council of Ministers, Copenhagen, DK, 2014, p. 507.

[165] J.H. Johansson, Y. Shi, M. Salter, I.T. Cousins, Spatial variation in the atmospheric deposition of perfluoroalkyl acids: source elucidation through analysis of isomer patterns, Environ. Sci.: Processes Impacts 20 (2018) 997—1006.

[166] C.M. Butt, S.A. Mabury, D.C.G. Muir, B.M. Braune, Prevalence of long-chained perfluorinated carboxylates in seabirds from the Canadian arctic between 1975 and 2004, Environ. Sci. Technol. 41 (2007) 3521—3528.

[167] H. Feng, Y. Ruan, K. Zhang, P.K.S. Lam, Current analytical methodologies and gaps for per- and polyfluoroalkyl substances determination in the marine environment, Trac. Trends Anal. Chem. (2019), https://doi.org/10.1016/j.trac.2018.12.026 in press.