



Environmental distribution of per- and polyfluoroalkyl substances (PFAS) on Svalbard: Local sources and long-range transport to the Arctic

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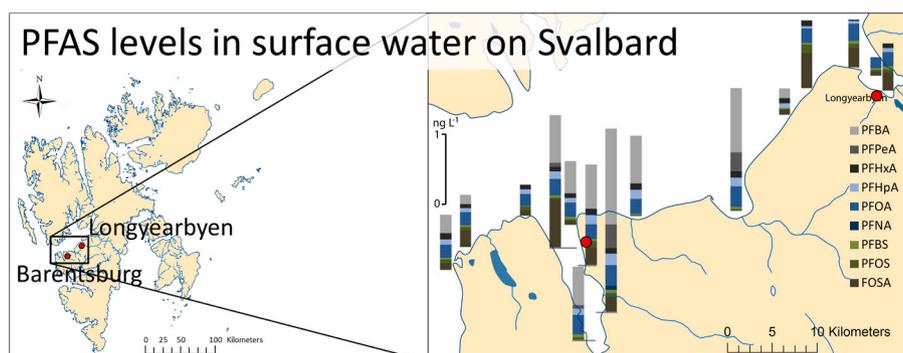
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HIGHLIGHTS

- Distribution of PFASs in water, snow, sediment, and soil in the Arctic.
- Fire training sites, landfills and wastewater were identified as local PFAS sources.
- PFCAs were dominant at sites impacted by LRAT.
- PFASs and 6:2 FTSA were dominant at sites impacted by local sources.
- Melting snow and ice is an important PFAS source in the Arctic.

GRAPHICAL ABSTRACT



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ABSTRACT

The environmental distribution of per- and polyfluoroalkyl substances (PFAS) in water, snow, sediment and soil samples taken along the west coast of Spitsbergen in the Svalbard archipelago, Norwegian Arctic, was determined. The contribution of potential local primary sources (wastewater, firefighting training site at Svalbard airport, landfill) to PFAS concentrations and long-range transport (atmosphere, ocean currents) were then compared, based on measured PFAS levels and composition profiles. In remote coastal and inland areas of Spitsbergen, meltwater had the highest mean Σ PFAS concentration ($6.5 \pm 1.3 \text{ ng L}^{-1}$), followed by surface snow ($2.5 \pm 1.7 \text{ ng L}^{-1}$), freshwater ($2.3 \pm 1.1 \text{ ng L}^{-1}$), seawater ($1.05 \pm 0.64 \text{ ng L}^{-1}$), lake sediments ($0.084 \pm 0.038 \text{ ng g}^{-1}$ dry weight (dw)) and marine sediments ($< \text{method detection limit (MDL)} - 0.46 \text{ ng g}^{-1}$ dw, median 0.015 ng g^{-1} dw). Perfluoroalkyl sulfonates (PFSA) and 6:2 fluorotelomer sulfonate (FTSA) were predominant in water and soil samples influenced by local sources, while perfluoroalkyl carboxylates (PFCA) were predominant in water and sediment from remote coastal and inland areas of Svalbard. The PFAS composition profiles observed in remote areas indicated that atmospheric transport and oxidation of volatile precursors is an important source of PFCA on Svalbard. Shorter-chain PFAS such as perfluorobutanoate (PFBA) were the predominant PFAS in freshwater, reflecting replacement of C_8 -chained PFAS with shorter-chained compounds. The comparatively high PFAS (especially PFBA) concentration in meltwater indicated that melting of snow and ice during the Arctic

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spring is an important diffuse local PFAS source. This source may become even more important with climate warming-induced melting of Arctic glaciers and ice sheets. Further studies of mobilisation and transport of PFAS in the Arctic region are needed to confirm this trend.

1. Introduction

Poly- and perfluoroalkyl substances (PFAS) are ubiquitously distributed in remote environments such as the Arctic (Butt et al., 2010; Kwok et al., 2013; Lescord et al., 2015; Stock et al., 2007; Young et al., 2007; AMAP, 2017). Concerns about their environmental persistency, bioaccumulation potential and possible toxic effects on humans and wildlife have led to national and international regulations restricting production and use of certain PFAS (Buck et al., 2011). For example, perfluorooctane sulfonate (PFOS; $C_8F_{17}SO_3^-$) and perfluorooctanoate (PFOA; $C_7F_{15}COO^-$) and their salts have been added to the persistent organic pollutants (POPs) list of the Stockholm Convention (under Annex B and Annex A, respectively). However, alternative PFAS (e.g. hexafluoropropylene oxide dimer acid (HFPO-DA)) and precursors, which can degrade to persistent perfluoroalkyl sulfonates (PFSA; $C_nF_{2n+1}SO_3^-$) and perfluoroalkyl carboxylates (PFCA; $C_nF_{2n+1}COO^-$) (Ellis et al., 2004; Martin et al., 2006), are still being produced, used and ultimately emitted to the environment (Wang et al., 2014).

PFAS are widely used in a variety of industrial and consumer products, such as stain- and water-repellent textiles, non-stick products, paints, waxes and aqueous film-forming foams (AFFFs) (Buck et al., 2011). During their production, usage and end-of-life cycle, PFAS can be emitted to the environment (Prevedouros et al., 2006) and subsequently transported to remote areas, such as the Arctic region (Armitage et al., 2006; Stock et al., 2007; Wallington et al., 2006; Zhao et al., 2012). Two major long-range transport pathways (atmospheric, oceanic) to the Arctic have been identified (Armitage et al., 2009; Kwok et al., 2013; Stock et al., 2007; Young et al., 2007). Long-range atmospheric transport (LRAT) occurs as atmospheric transport of volatile precursors, followed by atmospheric degradation to PFCA and PFSA (Stock et al., 2007; Young et al., 2007; Zhao et al., 2012) or atmospheric transport of neutral PFCA in the gas phase and aerosol-mediated transport of particle bound PFAS (Ahrens et al., 2012; McMurdo et al., 2008). As a result, volatile PFCA and PFSA precursors such as fluorotelomer alcohols (FTOHs; $C_nF_{2n+1}CH_2CH_2OH$), perfluorooctane sulfonamides (FOSAs; $C_8F_{17}SO_2N(C_nH_{2n+1})H$) and perfluorooctane sulfonamidoethanols (FOSEs, $C_8F_{17}SO_2N(C_nH_{2n+1})CH_2CH_2OH$) have been detected in the Arctic atmosphere (Ahrens et al., 2011a; Shoeib et al., 2006; Stock et al., 2007) and can undergo atmospheric degradation to persistent PFCA and PFSA (Ellis et al., 2004; Young and Mabury, 2010). Long-range oceanic transport of PFAS has also been identified as a major pathway to the Arctic (Ahrens et al., 2010; Armitage et al., 2009; Prevedouros et al., 2006), but the relative importance of atmospheric and oceanic transport is not yet fully understood (Butt et al., 2010; Wang et al., 2015; Boucher et al., 2019). Possible exchange of PFAS between the sea-surface microlayer and sea-spray aerosols adds to the complexity of PFAS transport, distribution and fate in remote environments (Del Vento et al., 2012; Casas et al., 2020).

In addition to long-range transport, local sources of PFAS, such as discharge from wastewater treatment plants (Becker et al., 2008; Sinclair and Kannan, 2006), firefighting training sites (Ahrens et al., 2015; Kärrman et al., 2011; Mejia-Avendaño et al., 2017) and landfills (Ahrens et al., 2011b; Busch et al., 2010; Lang et al., 2017) are considered important direct sources of PFAS contamination in the Arctic region (Ali et al., 2021; Skaar et al., 2019; Lescord et al., 2015; Stock et al., 2007).

In this study, PFAS were analysed in representative samples (water, snow, sediment, soil) from different environmental compartments collected along the west coast of Spitsbergen in the Svalbard archipelago, Norwegian Arctic. The samples included *i*) suspected local PFAS sources and the surrounding environment, *ii*) coastal waters from the

bay of Longyearbyen (~2400 inhabitants, 2020) past the mining town of Barentsburg (~450 inhabitants, 2020) to a remote western region by Kapp Linné and *iii*) multimedia samples from a remote region around an inland lake, Lake Linnévatnet. The research objectives were *i*) to identify local PFAS sources, *ii*) to evaluate the environmental fate of PFAS emitted from suspected local sources (i.e. wastewater, firefighting training site at the local airport, landfill) and *iii*) to compare the PFAS contribution from local point sources to that from diffuse secondary sources such as long-range transport (atmospheric and oceanic) and inputs from melting of glaciers and ice. Characteristic PFAS composition profiles (fingerprints) were identified and used in this comparison.

2. Materials and method

2.1. Sampling

Sampling was conducted on Nordenskiöld land on the west coast of Spitsbergen, the main island in the Svalbard archipelago, in early summer 2015 (June–July). All samples were collected in polypropylene (PP) bottles as grab samples. Surface seawater samples (denoted W1–W11, W15, W17, W20, W23–W24) and marine sediment samples (S1–S8, S12, S14–S15) were collected along a ~55 km coastal transect westward from Longyearbyen (~2400 inhabitants, 2020) to the southern side of the outlet of Isfjorden (Kapp Linné); including sampling along Grønnfjorden near the Russian mining town Barentsburg (~500 inhabitants in 2015) (Fig. 1). A surface seawater sample was also collected from Kongsfjord, a bay close to Ny-Ålesund airport (W1). In addition, samples of wastewater (W12–13, W18), drainage water from old and current landfills (W19 and W22, respectively) and water from a reference stream (W21) were taken in the Longyeardalen valley and the adjacent Adventdalen valley. In order to evaluate PFAS distribution patterns around the active firefighting training site at Svalbard airport, soil and water samples were collected from representative locations at and near the site (S9–S11, W14) and from farther downhill (S13, W16). A region located near Kapp Linné was chosen to represent background conditions, with sampling performed in Lake Linnévatnet and the surrounding area. The lake receives its water from glacial meltwater and the outlet drains into Isfjorden seawater. There is a small research station in the vicinity (~3 km) and Barentsburg is the closest settlement, ~10 km to the east across a mountain and Grønnfjorden. The samples included surface lake water collected under the ice cover in Lake Linnévatnet (W28–W30, W33), lake bottom sediment (S16–S19) and surface water from the lake inlet (W34) and outlet (W27) streams as well as a nearby reference lake (W25). Meltwater uphill from Lake Linnévatnet (W26, W31–W32) and surface snow (Sn1–Sn3) uphill from the meltwater sampling sites were also sampled (Fig. 1). Some of the sampling sites at Lake Linnévatnet and the surrounding area were similar to those used in a previous study (Skaar et al., 2019). For further details of all samples analysed, see Supplementary Tables S1 and S2.

2.2. Chemicals

A total of 26 target PFAS were investigated, comprising: 13 C_3 – C_{13} , C_{15} , C_{17} PFCA (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDODA, PFTrIDA, PFTeDA, PFHxDA, PFOcDA), four C_4 , C_6 , C_8 , C_{10} PFSA (PFBS, PFHxS, PFOS, PFDS), three FOSAs (FOA, MeFOA, EtFOA), two FOSEs (MeFOSE, EtFOSE), three perfluorooctane sulfonamidoacetic acids (FOSAA, $C_8F_{17}SO_2N(C_nH_{2n+1})CH_2CO_2H$) (FOSAA, MeFOSAA, EtFOSAA) and 6:2 fluorotelomer sulfonate (FTSA, $C_6F_{13}CH_2CH_2SO_3^-$). The target PFAS were selected based on previous

detection of PFAS on Svalbard and availability of analytical standards (Kwok et al., 2013). Sixteen internal standards (IS), namely $^{13}\text{C}_4$ PFBA, $^{13}\text{C}_2$ PFHxA, $^{13}\text{C}_4$ PFOA, $^{13}\text{C}_5$ PFNA, $^{13}\text{C}_2$ PFDA, $^{13}\text{C}_2$ PFUnDA, $^{13}\text{C}_2$ PFDODA, $^{18}\text{O}_2$ PFHxS, $^{13}\text{C}_4$ PFOS, $^{13}\text{C}_8$ FOXA, d_3 -MeFOXA, d_5 -EtFOXA, d_7 -MeFOXA, d_9 -EtFOXA, d_3 -MeFOSAA and d_5 -EtFOSAA, all purchased from Wellington Laboratories Inc., Guelph, Canada, were used for quality assurance. For further details, see [Supplementary Table S3](#).

2.3. Sample preparation and analysis

Before extraction, all samples were spiked with 100 μL of IS mixture ($c = 0.1 \text{ ng } \mu\text{L}^{-1}$). The water samples were then subjected to solid-phase extraction (SPE) using Waters Oasis WAX cartridges (6 cm^3 , 500 mg) as previously described (Yamashita et al., 2004; Ahrens et al., 2009a). The sediment and soil samples were oven-dried (25 $^\circ\text{C}$) and sieved (mesh size: 2 mm), and analysed. Aliquots of 5 g dry weight (dw) were then extracted using methanol and a clean-up was performed using 25 mg ENVI-Carb (Supelco, Bellefonte, PA, USA) as described previously (Powley et al., 2005; Ahrens et al., 2009b) (for details see Supplementary Information).

The instrumental analysis was performed using high-performance liquid chromatography (HPLC, Agilent Technologies 1200 Series, Palo Alto, CA, USA) coupled to tandem mass spectrometry interfaced with an electrospray ionization source in negative-ion mode ((-)ESI-MS/MS, Agilent 6460 Triple Quadrupole System, Palo Alto, CA, USA) as described in Ahrens et al. (2009a).

Aliquots of 10 mL were injected on a Hypersil Gold pre-column (10 \times 2.1 mm, 5 mm particle size, Thermo Scientific, Waltham, MA, USA) coupled with a Betasil C_{18} column (50 \times 2.1 mm, 5 mm particle size, Thermo Scientific, Waltham, MA, USA) using a gradient of 0.350 mL/min Millipore water and methanol (both with 10 mM aqueous

ammonium acetate solution

2.4. Quality assurance

Mean blank concentrations, method detection limits (MDLs), recoveries and standard deviations in percentages for triplicate samples are summarised in [Supplementary Tables S4–S6](#). The instrument performance was monitored with the injection of quality control standards and the correlation coefficient (R^2) of the calibration curve was >0.99 . Blanks for the water phase ($n = 6$) and solid phase ($n = 4$) were prepared as the natural samples but without the sample media. The MDL was calculated as the mean blank concentration plus three times the absolute standard deviation of the blank for water ($n = 6$) and sediment/soil ($n = 4$). If a compound was not detected in the blanks, the MDL was set to the lowest calibration standard level (0.05 ng L^{-1} for water, 0.01 ng g^{-1} dw for sediment). The MDL ranged between 0.0020 and 0.11 ng L^{-1} in water and between 0.0026 and 0.12 ng g^{-1} dw (1.2 ng g^{-1} dw for the outlier 6:2 FTSA) in sediment/soil. For triplicate samples, the mean standard deviation was 15% for water ($n = 4$, [Supplementary Table S4](#)) and 15% for sediment/soil ($n = 3$, [Supplementary Table S5](#)). The recovery is given for individual mass-labelled IS in the respective matrix of the water and solid phase covering almost all detected PFAS. Mean recovery of the quantified PFAS ranged from 59% to 103% for water (except MeFOXA, with 31%) and 42%–101% for sediment/soil ([Supplementary Table S6](#)). Additional details about the quality assurance procedure are provided in Supplementary Information.

2.5. Statistical analysis

The Mann-Whitney U test ($\alpha = 0.05$) was used to compare the mean Σ PFAS concentrations in different environmental matrices, while

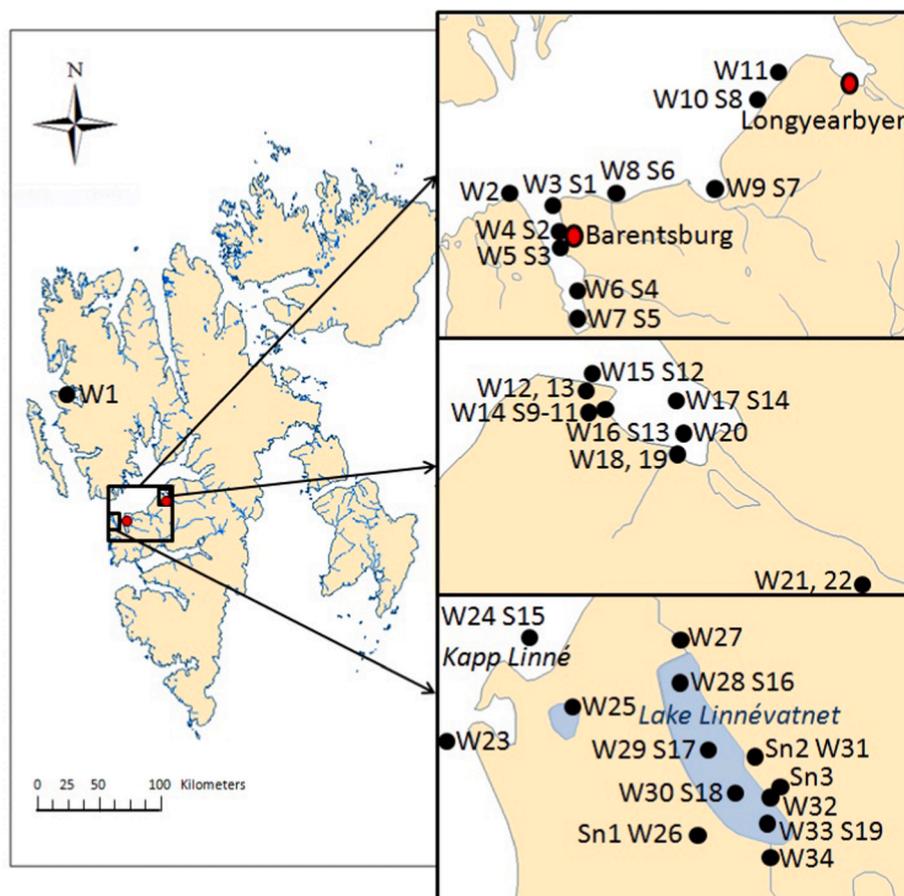


Fig. 1. Sampling locations for water (W), snow (Sn), sediment and soil (S) on Svalbard. Note: sampling location W21 (reference stream) was 700 m uphill from W22.

Spearman's Rho was used to evaluate the correlation (ratio) between PFOA and PFNA. The ration between PFOA and PFNA can be used as an evidence that they originate from the atmospheric degradation of 8:2 FTOH (Young et al., 2007; Kwok et al., 2013). Before performing the statistical tests, all PFAS concentrations that were <MDL were set as half of MDL.

3. Results and discussion

3.1. PFAS distribution in snow, meltwater, stream water, lake water, seawater, lake sediment and marine sediment

Of the 26 target compounds, 13 were detected (>MDL) in surface water (lake water, meltwater, stream water and seawater) and surface snow. These were: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFOS, 6:2 FTSA and FOSA (Supplementary Figs. S1–S3, Table S7). In background and coastal areas, meltwater had the highest mean \sum PFAS concentration (W26, W31–W32; 6.5 ± 1.3 ng L⁻¹), followed by surface snow (Sn1–Sn3; 2.5 ± 1.6 ng L⁻¹), surface lake water including inlet and outlet stream water (W27–30, W33–34; 2.3 ± 1.1 ng L⁻¹) and seawater (1.05 ± 0.64 ng L⁻¹) (Fig. 2). Based on differences in PFAS composition profiles, the seawater samples were divided into two groups (SW1, SW2). The samples in group SW2 (W3–W9) had almost three-fold higher PFAS levels (1.5 ± 0.60 ng L⁻¹) than those in group SW1 (W2, W10–W11, W15, W17, W20, W23–W24) (0.62 ± 0.24 ng L⁻¹) (Fig. 2). This difference in \sum PFAS concentrations in seawater indicates that the sampling sites in group SW2 were affected by the stream outlets (W6–W9) and less dilution in Grønnsfjorden, whereas the sampling locations in group SW1 were only impacted by the open sea, with lower PFAS concentrations.

The \sum PFAS concentrations in meltwater were significantly higher ($p < 0.05$) than those in the stream and lake water recipients (Fig. 2), most likely due to dilution of PFAS in the stream and lake. The \sum PFAS levels in meltwater were also higher (differences not significant, $p = 0.08$) than those in surface snow. Elevated PFAS levels in meltwater from snow, ice and glaciers might be a result of up-concentration caused by repeated evaporation, as suggested in a previous study which found higher PFAS concentrations in glacial water from Longyearbyen glacier than in glacial surface snow in the vicinity of Longyearbyen (Kwok et al., 2013). Samples from the inlet (W34) and outlet (W27) streams of Lake Linnévatnet had twice as high \sum PFAS levels (mean 3.7 ± 0.018 ng L⁻¹) compared to lake water samples (W28–W30, W33; mean 1.6 ± 0.41 ng L⁻¹). This might be due to the influence of meltwater with elevated PFAS levels, since the stream inlet is fed by glacial and snow meltwater and the outlet receives inputs from melting of surrounding snow and ice. Meltwater might thus be an important PFAS source to the nearby aquatic

environment, as observed in previous studies (Filipovic et al., 2015; MacInnis et al., 2019a; Skaar et al., 2019).

The \sum PFAS concentrations in Lake Linnévatnet (mean 1.6 ± 0.41 ng L⁻¹, W28–30, W33) were within the range reported previously for Lake Linnévatnet (<0.03–4.1 ng L⁻¹) by Skaar et al. (2019) and for four remote lakes in the Canadian Arctic (1.9 ± 0.42 to 2.4 ± 0.40 ng L⁻¹) by Lescord et al. (2015) (for details see Supplementary Table S8). Furthermore, the \sum PFAS concentrations in surface snow (mean 2.5 ± 1.6 ng L⁻¹) were in agreement with the level observed in surface snow sampled downstream from Longyearbyen glacier in 2006 (1.5 ng L⁻¹), but were three-fold higher than the levels recorded in glacial snow in that study (0.46 ng L⁻¹) (Kwok et al., 2013). PFAS are affected by LRAT and oxidation of volatile precursors, and also by sea spray, mineral dust and combustion aerosols (MacInnis et al., 2019a; Hartz et al., 2023). Thus, PFAS concentrations in snow may vary considerably, since the deposition of PFAS depends on recent weather events, adding to the integrated pollutant levels in the weathered surface snow. Nevertheless, the observed PFAS levels in snow and meltwater support the assumption that atmospheric deposition sources and pathways are the major source of PFAS in remote Arctic locations.

Three of the 26 target compounds (PFOA, PFNA and PFUnDA) were detected in sediment from Lake Linnévatnet (Supplementary Fig. S4, Table S9). These three compounds were also among the predominant PFAS detected in sediment cores from Canadian Arctic lakes and were showing increasing temporal trends in sediment cores from Lake Hazen in Arctic Canada (MacInnis et al., 2019b; Hartz et al., 2023). Mean \sum PFAS concentrations in Lake Linnévatnet sediment (0.083 ± 0.038 ng g⁻¹ dw) (Fig. 2, Supplementary Table S9) were lower than previously observed in four remote lakes in the Canadian Arctic (range 0.19 ± 0.03 to 2.7 ± 0.18 ng g⁻¹ dw) (Lescord et al., 2015). In marine sediments, only two PFAS were detected (FOSA and PFUnDA) and \sum PFAS levels ranged between < MDL and 0.46 ng g⁻¹ dw (Fig. 2, Supplementary Table S9). Thus, marine sediments at Svalbard showed low contamination with PFAS. Furthermore, FOSA and PFUnDA have a higher particle-water partitioning coefficient (K_d value) than shorter-chain PFCA and PFSA (Ahrens et al., 2015; Higgins and Luthy, 2006; Gellrich et al., 2012), which may explain their presence in marine sediments. The \sum PFAS levels in marine sediments were generally within the range reported for sediment from Adventfjorden bay near Longyearbyen (\sum PFAS 0.16 – 4.6 ng g⁻¹ dw; $n = 7$ detected PFAS) (Ali et al., 2021) and in the Bering Sea and the western Arctic (\sum PFAS 0.06 – 1.7 ng g⁻¹ dw; $n = 14$ detected PFAS) (Lin et al., 2020) (Supplementary Table S8).

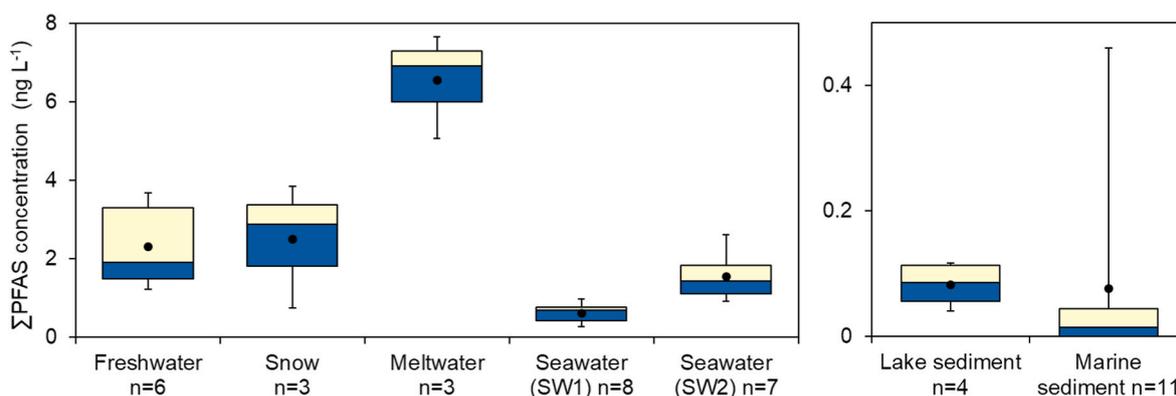


Fig. 2. Distribution of \sum PFAS concentrations in freshwater (lake, inlet and outlet streams), snow, meltwater and seawater (ng L⁻¹), and in lake and marine sediment (ng g⁻¹ dw). Seawater and marine sediment samples were collected along the coast from Longyearbyen to Kapp Linné, including Grønnsfjorden. All other samples were collected from the region representing background levels around Lake Linnévatnet. Box-and-whisker plots show minimum and maximum concentrations, boxes show 25th, median and 75th percentiles, and dots show arithmetic means.

3.2. Impact of local point sources (wastewater, landfill leachate, firefighting training site) and atmospheric deposition on PFAS in the environment

The highest \sum PFAS concentrations were detected in a soil sample (S9) from burn pits at the firefighting training site ($1600 \text{ ng g}^{-1} \text{ dw}$) and in a sample in ponding water on waterlogged soil (W14) downhill of the firefighting training site ($\sim 2750 \text{ ng L}^{-1}$). However, determination of some PFAS in sample W14 was semi-quantitative due to peak interferences (Supplementary Table S7). In a drainage ditch 600 m downhill of the firefighting training site, the \sum PFAS concentration was $3.9 \text{ ng g}^{-1} \text{ dw}$ in streambed sediment (S13) and $120 \pm 0.56 \text{ ng L}^{-1}$ in stream water (W16) (Supplementary Figs. S2 and S5, Tables S7 and S8). This shows that PFAS used at the local firefighting training site were transported off by drainage water and meltwater runoff. Some of these PFAS were retained in streambed sediments, but the fraction detected in stream water will ultimately be discharged into surrounding coastal waters. This is in agreement with previous reports of high levels of PFAS in meltwater runoff from a firefighting training site in Ny-Ålesund and from the same firefighting training site as the present study (Skaar et al., 2019), confirming that meltwater runoff is an important factor for (re) mobilisation of PFAS.

The predominant PFAS in soils sampled at and downhill from the firefighting training site (S9–S11, S13) were PFOS (51% of \sum PFAS), 6:2 FTSA (17%) and PFHxS (7.9%). This was similar to the PFAS composition in the ponding water sample (W14) taken downhill of the firefighting training site (6:2 FTSA 53% of \sum PFAS; PFOS 17%; PFHxS 10%) (Fig. 3). This corresponds to previous findings that PFOS and 6:2 FTSA are generally the predominant PFAS at AFFF-affected sites (Ahrens et al., 2015; Kärrman et al., 2011). In this study, the PFAS composition in soil and water changed with increasing distance from the firefighting training site. For example, 6:2 FTSA was the major compound in the burn pits at location S9 (56% of \sum PFAS), but it was not detected in soils a few hundred metres away (S11, S13) (Supplementary Figs. S6–S7), suggesting that 6:2 FTSA was degraded and/or retained in the soil.

PFHxA (18% of \sum PFAS) and PFPeA (7.2%) were amongst the predominant compounds in drainage water downhill of the firefighting training site (W16), while 6:2 FTSA comprised only 4.2% of the total (Fig. 3), indicating that 6:2 FTSA had degraded to PFPeA and PFHxA (Wang et al., 2011). High \sum PFAS concentrations were also observed in untreated landfill leachate (W22; \sum PFAS = 410 ng L^{-1}), in which PFOS (30% of \sum PFAS) and PFOA (22%) were the predominant compounds (Fig. 3). However, a recent study indicates that shorter-chain PFAS are getting more dominant in landfill leachate (Knutsen et al., 2019). In addition, elevated \sum PFAS concentrations were detected in wastewater from the airport workshop (W12; 570 ng L^{-1} , Svalbard airport (W13; 170 ng L^{-1}) and Longyearbyen city (W18; 65 ng L^{-1}) (Supplementary Table S7). The major PFAS in wastewater were PFHxS (74% of \sum PFAS) for the airport workshop (W12), PFOS (57%) and 6:2 FTSA (28%) for Svalbard airport (W13), and PFNA (56%) and PFOA (17%) for Longyearbyen city wastewater (W18) (Fig. 3). In order to decrease the environmental impact of PFAS from AFFF-contaminated sites on Svalbard, use of fluorine-free AFFFs and soil remediation has been suggested (Gobelius et al., 2017; Söregård et al., 2019) as well as implementation of efficient wastewater treatment techniques for removal of PFAS (Arvaniti and Stasinakis, 2015).

In the remote inland region around Lake Linnévatnet, PFCA were the major PFAS in freshwater, including surface lake water and lake inlet and outlet stream water (W27–W30, W33–W34; 93% of \sum PFAS), surface snow (Sn1–Sn3; 80%) and meltwater (W26, W31–W32; 86%); in contrast to the predominance of mainly PFSA and 6:2 FTSA in locally contaminated areas (Fig. 3). This confirms previous claims that LRAT and oxidation of volatile precursors (e.g. FTOHs), which can degrade to PFCA, is an important PFCA source in Arctic regions (Ellis et al., 2004; Stock et al., 2007; Wallington et al., 2006; Young et al., 2007). The mean ratio of PFOA to PFNA in meltwater ($n = 3$), freshwater ($n = 6$) and surface snow ($n = 3$) around Lake Linnévatnet (1.7 ± 0.8) was comparable to that previously observed in atmospherically supplied Canadian Arctic ice cores (1.5 ± 0.8) (Young et al., 2007) and in glacial ice cores on Svalbard (1.9 ± 0.7 and 1.3 ± 0.4) (Kwok et al., 2013).

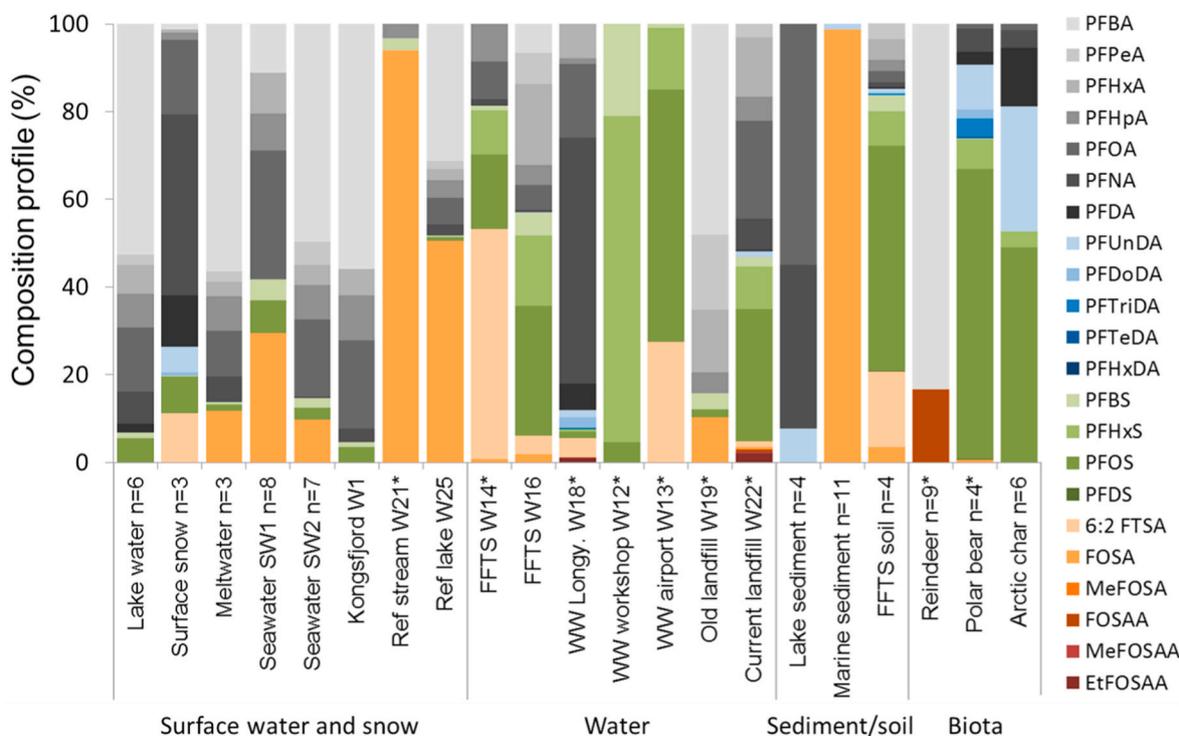


Fig. 3. PFAS composition profiles (%) of individual samples and sample groups. Some PFCA, FOSAs and FOSAAs were not quantified due to matrix interference (samples marked *) (Supplementary Table S9). Freshwater includes Lake Linnévatnet and the inlet and outlet streams. WW = wastewater, FFTS = firefighting training site.

(Supplementary Table S11), indicating atmospheric deposition as a source. Moreover, a positive correlation (Spearman $\rho = 0.62$, $p < 0.05$, $n = 13$) was observed between PFOA and PFNA in freshwater and in surface snow around Lake Linnévatnet, again suggesting that atmospheric deposition is most likely the source of these PFAS.

The PFAS composition in meltwater uphill of Lake Linnévatnet was similar to that in recipient freshwater (lake, inlet and outlet streams), with PFBA (57% and 53% of \sum PFAS, respectively), PFOA (10% and 15%, respectively), PFHpA (7.8% in both) and PFNA (6.0% and 7.3%, respectively) as the major compounds (Fig. 3). The predominance of PFBA in meltwater and freshwater complies with the assumption regarding conversion of C₈-chained PFAS (PFOS and PFOA) to short-chain PFAS (Möller et al., 2010), which also seemed to be widely distributed according to the results. In surface snow, on the other hand, PFNA predominated (41% of \sum PFAS), followed by PFOA (17%), PFDA (12%) and PFUnDA (5.9%) (Fig. 3). This indicates that PFAS retention and mobility in snowpack may be compound-specific (Codling et al., 2014). For example, PFBA predominated in meltwater from snow and ice in the present study (W26 and W31–W32; 57% of \sum PFAS, 3.7 ± 0.92 ng L⁻¹), but was not a major compound in snow (<0.096 ng L⁻¹ in samples Sn1 and Sn3, 0.14 ng L⁻¹ in sample Sn2). This is in agreement with previous findings indicating that PFBA is more mobile than other longer-chained PFAS (Kwok et al., 2013; Skaar et al., 2019). Kwok et al. (2013) found higher PFBA concentrations in meltwater from a glacier (0.29 ± 0.057 ng L⁻¹) than in surface snow and ice cores. They concluded that the PFBA in meltwater originated from continental areas, since the concentrations correlated with levels of non-sea salt sulphate (SO₄²⁻) in glacial ice cores (Kwok et al., 2013).

The differences in \sum PFAS concentrations and composition profiles observed in seawater in the present study (Figs. 2 and 3) were mainly attributable to the absence of PFBA in several seawater samples (W2, W11, W15, W17, W20) (Figs. 1 and 4). The seawater samples with the highest \sum PFAS concentrations had a high proportion of PFBA (on average 49% of \sum PFAS) and were located near the outlet of rivers fed by meltwater from glaciers and snow (W4–W9) (Figs. 1 and 4). Moreover, PFBA was the major compound in meltwater and recipient freshwater (Fig. 3) and \sum PFAS concentrations in meltwater were higher than in stream, lake and seawater (Fig. 2), indicating that meltwater may be an important local diffuse PFAS source. On the other hand, Ali et al. (2021)

reported that PFOS was the dominant PFAS (>80%) in seawater samples from Adventfjord and Isfjord, near the Svalbard Airport, and the PFOS isomer profile indicates that the PFOS originated from local sources. However, it is not fully clear why PFBA was not detected in the seawater samples taken near Longyearbyen (W11, W15, W17, W20) and the adjacent river outlets. The presence and absence of PFBA might be influenced by differences in sampling dates, because the Longyearbyen seawater samples were collected 5–6 days before all other seawater samples (except W9) but one (W9) of the seawater samples with high PFBA levels (Supplementary Table S1). When the sampling was performed, the summer snow melt may have led to elevated inputs of PFBA to coastal waters in the later period. Wide variations in \sum PFAS concentrations in the Arctic Ocean have previously been ascribed to varying inputs from sea ice, with PFBA suggested as a marker for meltwater from sea ice (Cai et al., 2012). Climate change could accelerate PFAS release into seawater, due to melting snowpack and glaciers (MacInnis et al., 2019b). As shown previously, atmospheric deposition is important factor for assessing the Arctic as a sink or source of PFAS (Joerss et al., 2020).

3.3. Conclusions and future implication

In this study, PFAS were ubiquitously detected in environmental samples taken along the coast of western Spitsbergen. Comparisons of findings for different locations revealed that PFAS concentrations and composition profiles were mainly influenced by a combination of long range atmospheric transport and local sources. PFCA predominated in remote abiotic environments (e.g. snow, surface water), while PFSA and 6:2 FTSA generally were the most dominant compounds present in locally contaminated media, which suggests that LRAT and oxidation of volatile precursors is a major transport route for PFCA to the Arctic region. In freshwater PFBA was the predominant compound, indicating replacement of C₈-based PFAS (i.e. PFOS and PFOA) with short-chain PFAS, as widely reported on global scale. Landfill leachate, AFFF-contaminated water and wastewater were identified as important local point sources, with \sum PFAS concentrations (mean ~ 680 ng L⁻¹) that were approximately 200-fold higher than background levels (mean ~ 3.5 ng L⁻¹) in water and snow around Lake Linnévatnet. In order to reduce the PFAS loads from wastewater, it is important to reduce the

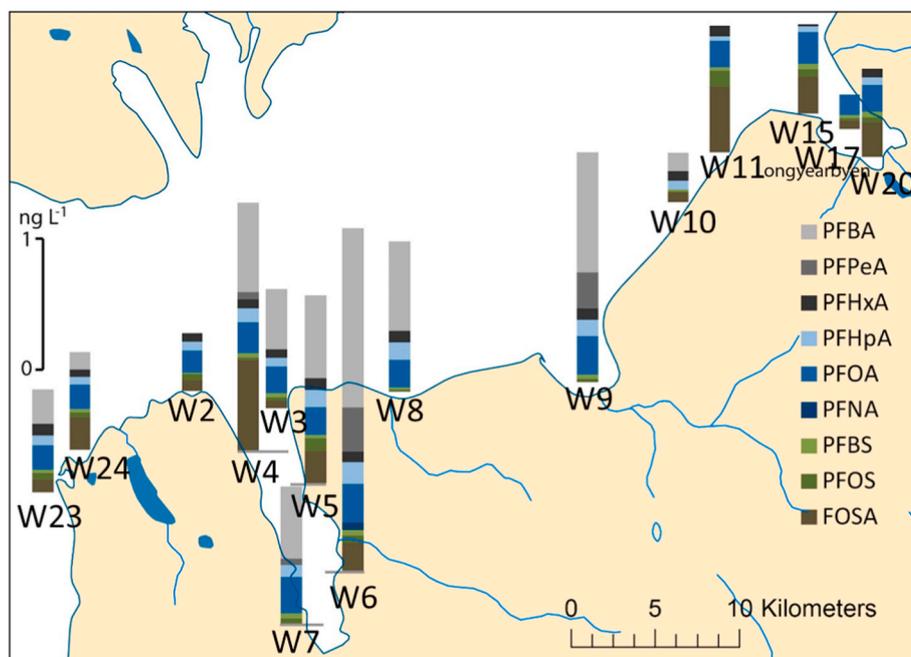


Fig. 4. PFAS concentrations and composition profiles in seawater samples taken along the coast of Spitsbergen from Longyearbyen to Kapp Linné.

usage or even ban the use of PFAS and efficient wastewater treatment techniques need to be implemented for historical and current usage. In soils around the firefighting training site at Svalbard airport (S9–S10), the Σ PFAS concentrations were approximately 25-fold to 20,000-fold higher than in remote inland lake sediments (0.083 ng g^{-1}). Further, the Σ PFAS concentrations in drainage water 600 m downhill from the firefighting training site were approximately 20-fold higher than in meltwater from the remote inland area ($6.5 \pm 1.3 \text{ ng L}^{-1}$) and the proportion of PFSA was higher (51% of Σ PFAS, compared with 2%). This shows that PFAS are transported from the AFFF-contaminated site by water draining through the landscape, and ultimately reach the marine environment. In order to decrease the environmental impact of PFAS from AFFF-contaminated sites on Svalbard, fluorine-free AFFFs and soil remediation should be implemented. The local contamination with PFAS indicates a potential risk to wildlife, particularly top predators in the region (e.g. polar bears), and to humans due to contamination of drinking water and local food (e.g. fish, reindeer). Previous studies have shown the detection of PFAS in fish (Ali et al., 2021), caribou and reindeer (Roos et al., 2022) which are a risk for human consumption. Furthermore, based on the European Food Safety Authority (EFSA) recommendation a new tolerable weekly intake (TWI) of 4.4 ng/kg body weight per week for the sum of PFOS, PFOA, PFNA and PFHxS has been established for the consumption of food and drinking water (EU, 2022). This study confirmed that meltwater from snow and ice, which contains accumulated atmospheric deposition of PFAS, is an important diffuse PFAS source in the Arctic region and likely caused the observed differences in seawater PFAS levels. Global warming could accelerate such inputs of PFAS to the aquatic environment, due to melting of glaciers and ice sheets in the Arctic.

Author statement

Lutz Ahrens: Funding acquisition, Conceptualization, Investigation, Writing – review & editing and Project administration. Jelena Rakovic: Investigation, Writing – review & editing, Data curation and Methodology. Siri Axelson: Investigation, Writing – review & editing and Methodology. Roland Kallenborn: Funding acquisition, Conceptualization, and Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2023.140463>.

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