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2 **Elucidation of contamination sources for poly- and perfluoroalkyl**  
3 **substances (PFASs) on Svalbard (Norwegian Arctic)**  
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24 **Keywords:** Perfluoroalkyl substances, PFASs, Arctic, Svalbard, Environment,  
25 pollution, source elucidation  
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30 *This publication is dedicated to the memory of the eight Russian Arctic environmental*  
31 *scientists and crew members who tragically lost their life in a recent helicopter accident*  
32 *near Barentsburg, Svalbard (November 4<sup>th</sup>, 2017).*

1 **Abstract** A combination of local (i.e. firefighting training facilities) and remote sources  
2 (i.e., long-range transport) are assumed to be responsible for the occurrence of per-  
3 and polyfluoroalkyl substances (PFASs) in Svalbard (Norwegian Arctic). However, no  
4 systematic elucidation of local PFASs sources have been conducted yet. Therefore, a  
5 survey was performed aiming at identifying local PFASs pollution sources on the island  
6 of Spitsbergen (Svalbard, Norway). Soil, fresh water (lake, draining rivers), sea water,  
7 melt-water run-off, surface snow and coastal sediment samples were collected from  
8 Longyearbyen (Norwegian mining town), Ny-Ålesund (research facility) and the Lake  
9 Linnévatnet area (background site) during several campaigns (2014-2016) and  
10 analysed for 14 individual target PFASs. For background site (Linnévatnet area,  
11 sampling during April to June 2015),  $\Sigma$ PFAS levels ranged from 0.4 – 4 ng/L in surface  
12 lake water ( $n = 20$ ). PFAS in melt water from the contributing glaciers showed similar  
13 concentrations ( $\sim 4$  ng/L,  $n = 2$ ). The short chain perfluorobutanoate (PFBA) was  
14 predominant in lake water (60-80% of the  $\Sigma$ PFASs), meltwater (20-30 %) and run-off  
15 water (40 %). Long range transport is assumed to be the major PFAS source. In  
16 Longyearbyen, 5 water samples (i.e. 2 seawater, 3 run-off) were collected near the  
17 local firefighting training site (FFTS) in November 2014 and June 2015, respectively.  
18 The highest PFAS levels were found in FFTS melt water run-off (118 ng/L). PFOS was  
19 the most abundant compound in the FFTS meltwater run-off (53 – 58 % PFASs). At  
20 the research station Ny-Ålesund, sea water ( $n = 6$ ), soil ( $n = 9$ ) and fresh water ( $n =$   
21 10) were collected in June 2016. Low  $\Sigma$ PFAS concentrations were determined for sea  
22 water (5 - 6 ng/L), whereas high  $\Sigma$ PFAS concentrations were found in run-off water  
23 (113 – 119 ng/L) and soil (211 – 800 ng/g dry weight (dw)) collected close to the local  
24 FFTS. In addition, high  $\Sigma$ PFAS levels (127 ng/L) were also found in fresh water from  
25 lake Solvatnet close to former sewage treatment facility. Overall, at both FFTS  
26 affected sites (soil, water), PFOS was the most abundant compound (60 – 69% of  
27  $\Sigma$ PFASs). FFTS and landfill locations were identified as major PFASs sources for  
28 Svalbard settlements.

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30 **Keywords:** Arctic, poly- and perfluoroalkyl substances, PFAS, local sources,  
31 pollutants, environment

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# 1 **Introduction**

2 Per- and polyfluoroalkyl substances (PFASs) were first identified as global  
3 environmental pollutants in a survey more than 15 years ago (Giesy and Kannan 2001;  
4 Kannan et al. 2001). Since then, a vast number of scientific publications have  
5 confirmed that PFASs are environmental pollutants with the potential to adversely  
6 affect human- and the environmental health (Banzhaf et al. 2017; Conder et al. 2008;  
7 Hekster et al. 2003; Jahnke and Berger 2009; Pabel et al. 2017; Valsecchi et al. 2013;  
8 Young and Mabury 2010). In 2009, perfluorooctane sulfonic acid (PFOS) and its  
9 precursors were added to the list of persistent organic pollutants (POPs) of the  
10 Stockholm Convention (under annex B = restricted usage). However, many other  
11 PFASs are still being produced for a variety of applications (Gao et al. 2015; Lam et  
12 al. 2016; Pan et al. 2017). In addition, precursor compounds which can be transformed  
13 to perfluoroalkyl carboxylates (PFCAs) and perfluoroalkyl sulfonates (PFASs), are  
14 continued to be produced and released into the environment (Gao et al. 2015; Kwok  
15 et al. 2015; Mailler et al. 2017; Myers et al. 2012).

16 Recently, application of PFAS containing aqueous film forming firefighting foams  
17 (AFFF) at firefighting training sites (FFTSs) have been identified as important source  
18 for PFASs in the environment (Anderson et al. 2016; Banzhaf et al. 2017; Barzen-  
19 Hanson et al. 2017a; Barzen-Hanson et al. 2017b; D'Agostino and Mabury 2014;  
20 Hansen et al. 2016). Especially the required regular training at FFTSs at large  
21 installations (i.e. civil and military air fields, industrial complexes and off shore  
22 installations) was identified as a major PFASs source for the nearby aqueous  
23 environment (Banzhaf et al. 2017; Barzen-Hanson et al. 2017a; Conder et al. 2008).

24 The transport processes, distribution and fate of PFASs has been a major research  
25 focus in Arctic environmental pollutant research (Bossi et al. 2015; Cai et al. 2012;  
26 Kelly et al. 2009; Koponen et al. 2013; Lescord et al. 2015; Smithwick et al. 2006;  
27 Young et al. 2007). Little emphasis, however, was placed upon potential direct  
28 emissions and primary sources for PFASs in the Arctic (Carlsson et al. 2016; Hansen  
29 et al. 2016; Kwok et al. 2013). Nevertheless, we must assume that, similar as already  
30 reported from middle latitude regions (Ahrens et al. 2015; Awad et al. 2011; Dauchy et  
31 al. 2017; Hansen et al. 2016; Hu et al. 2016; Zhang et al. 2016), airport facilities and  
32 active/abandoned industrial facilities may pose an imminent risk for PFAS release into

1 the Arctic aqueous environment with potential for accumulation and uncontrolled  
2 spreading.

3 Therefore, we conducted a first general survey on PFASs in sea water ( $n = 8$ ),  
4 freshwater ( $n = 30$ ) and soil ( $n = 9$ ) at the Arctic island Svalbard around the major  
5 settlements (Longyearbyen and Ny-Ålesund) as well as a background location (Lake  
6 Linnévatnet area) aiming at identifying and characterizing major local and remote  
7 PFASs contamination sources.

## 8 **Material and Methods**

### 9 **Target PFASs**

10 A total of 14 PFASs were investigated in this study including C<sub>4</sub>-C<sub>13</sub> PFCAs (i.e., PFBA,  
11 PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA) C<sub>4</sub>, C<sub>6</sub>,  
12 C<sub>8</sub> PFSA (i.e., PFBS, PFHxS, PFOS) and 6:2 fluorotelomer sulfonate (6:2 FTSA) (see  
13 Table S1 in the Supplementary Material (SM)).

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### 15 **Sampling**

16 In total, 49 aqueous samples (seawater ( $n = 9$ ), fresh water ( $n = 33$ ), snow melt ( $n = 4$ )  
17 and run-off ( $n = 3$ ) were collected in Ny-Ålesund, Longyearbyen and Lake Linnévatnet  
18 area, and 9 soil samples were collected in Ny-Ålesund only. The detailed sample list  
19 can be found in Table S2 in the SM. All sample locations are depicted in figure S1.

20 In Ny Ålesund, the fieldwork was conducted in and around the international research  
21 facilities (coordinates: N78,92 E11,91) in June 2016. The sample characteristics are  
22 listed in Table S2 in the SM and the location is presented in Figure S1 in the SM.  
23 Aqueous grab samples from 16 locations and soil samples from 9 locations were  
24 selected in close collaboration with representatives from the Kings Bay company which  
25 is responsible for the station administration and logistics.

26 Longyearbyen (N,78,24 E15,53) is currently the largest settlement on Svalbard with  
27 around 2000 inhabitants. Water samples (i.e., seawater ( $n = 3$ ) and freshwater ( $n = 3$ )  
28 samples were collected in November 2014 and June 2015, respectively, 600 m  
29 downstream to the local FFTS at Longyear airport (see Table S2 and Figure S1 in the  
30 SM).

31 Lake Linnévatnet (N7803 E13,82) is a fresh water lake on the island of Spitsbergen  
32 (Svalbard Norway). The lake receives its water from melt water of the adjacent glaciers

1 and drains directly in the Isfjorden marine environment. No potential pollution sources  
2 are registered except a small research station (Kapp Linné) in the vicinity (3 km  
3 distance). 26 water samples (i.e. freshwater ( $n = 22$ ): meltwater ( $n = 2$ ), snow ( $n = 2$ )  
4 were collected in the Lake Linnévatnet area in March 2014 and from April to June  
5 2015 (Table S2 and Figure S1 in the SM).

6

### 7 **Sample preparation**

8 All water samples were collected as replicates for separate analysis and quantification.  
9 The soil samples were divided in duplicates prior to sample preparation. 2.5 g of soil  
10 and 1000 – 2300 mL of aqueous samples were collected for further processing (Figure  
11 S2 in the SM). The sample preparation for the water samples (Jahnke et al. 2007;  
12 Möller et al. 2010) and soil samples (Powley et al. 2005) has been performed as  
13 described in previous publications (for details see text in the SM).

### 14 **Analysis and quantification**

15 All collected samples were analysed for 14 PFASs (see Table S1 in the SM) applying  
16 a validated quantification methods for the trace analytical determination of PFASs in  
17 the respective sample types (see text in the SM and Tables S3-S5 in the  
18 supplementary material). The quantitative trace analysis for all samples was done at  
19 the laboratories of the Norwegian University of Life Sciences (NMBU) and is described  
20 in the SM. Details on the quality control including detection limits (LOD), quantification  
21 limits (LOQ) and recoveries can be found in Table S6 and S7, respectively in the SM  
22 and recoveries in Table S7 in the SM.

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## 24 **Results and Discussion**

### 25 **Background levels and diffusive sources**

26 Background seawater, freshwater (lake and marine) as well as soil samples (Ny-  
27 Ålesund) were analysed for PFASs from the three selected locations on Svalbard  
28 (Figures 1 and 2, Tables S8-S13 in the SM). The Ny-Ålesund freshwater samples were  
29 collected as run-off surface water ( $n = 6$ ) or were taken from ponds ( $n = 3$ ) and streams  
30 ( $n = 1$ ) near the settlement, while the Linnévatnet freshwater samples were all surface  
31 lake water ( $n = 22$ ).

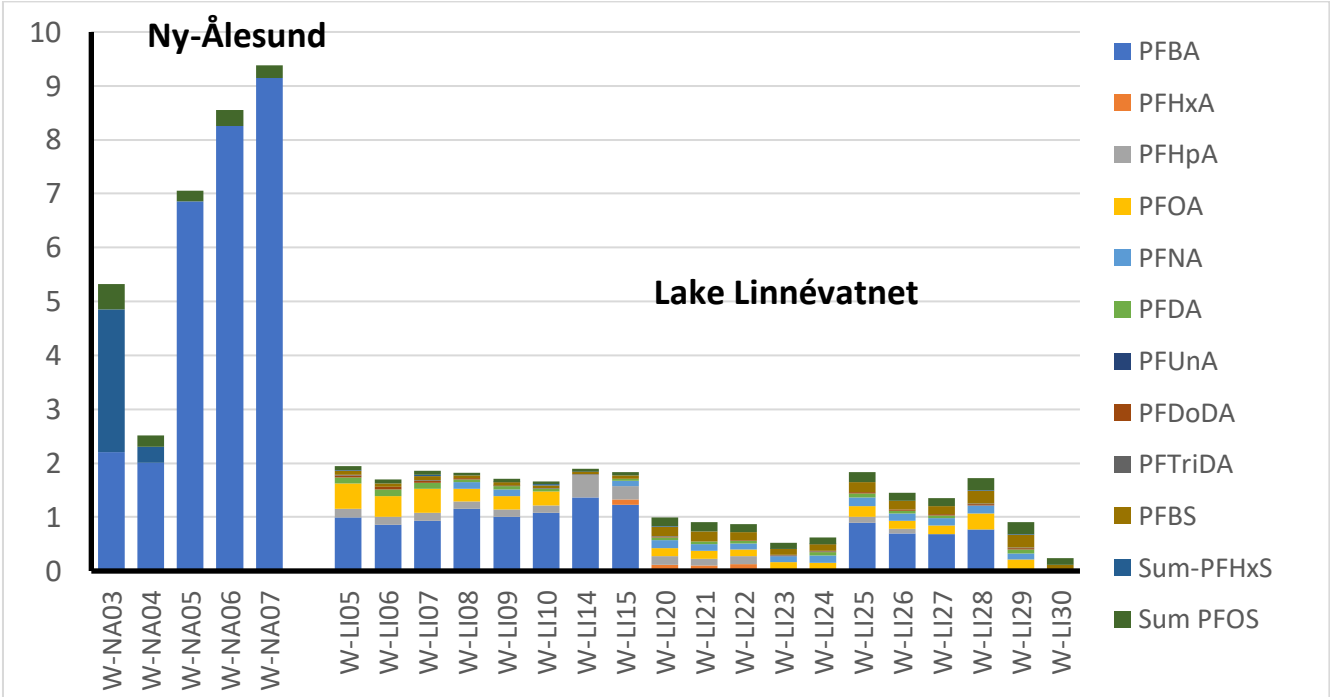
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Table 1:  $\Sigma$ PFASs concentrations in the sample matrices analysed (for details, see Table S7 – S9 in the SM)

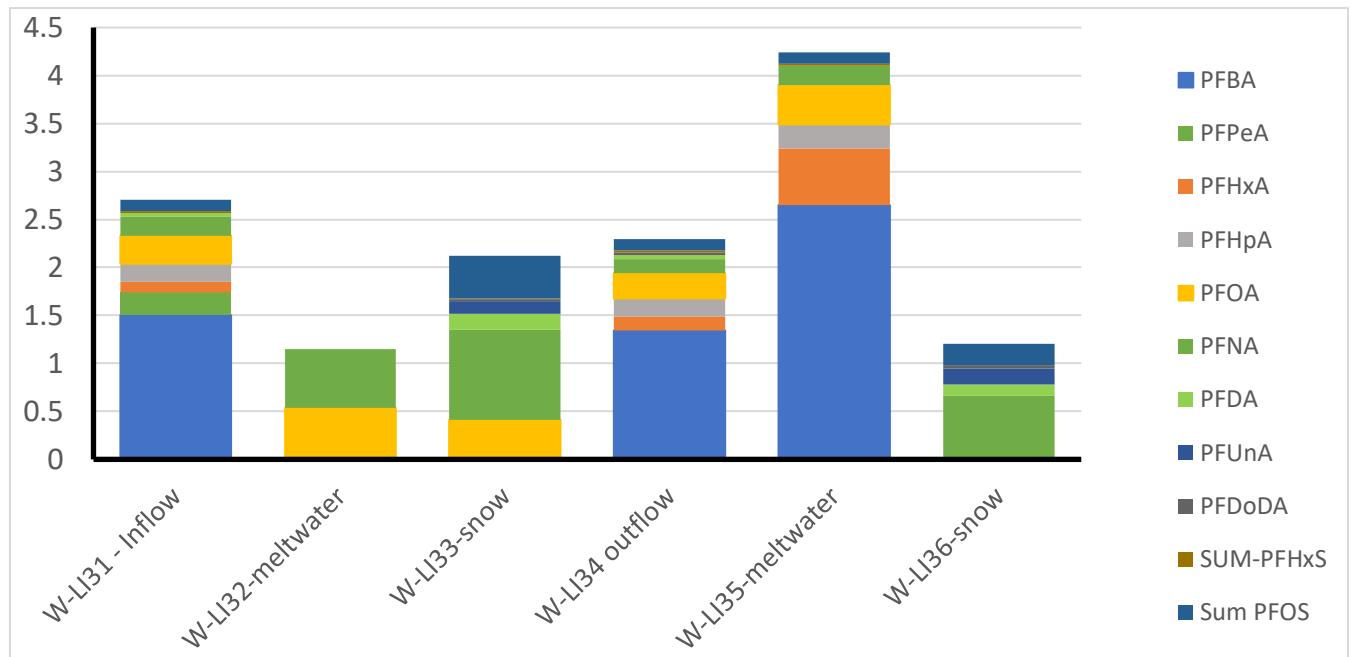
Location	Matrix	No of samples	Range Sum PFAS
Ny-Ålesund	Sea water	6	0.2 – 1.6 ng/L
	Soil	9	<0,03 - 800 ng/g dw
	Freshwater	8	2.7 – 13 ng/L
	Run-off water	2	544 – 1156 ng/L
Longyearbyen	Sea water	2	5 – 6 ng/L
	Run-off water	3	110 – 120 ng/L
Lake Linnévatnet	Lake water	22	<0,03 – 4.1 ng/L
	Meltwater	2	1.1 - 4.2 ng/L
	Surface snow	2	1 – 2 ng/L

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Figure 1: Background PFASs levels [ng/L] in freshwater water from Ny-Ålesund and lake Linnévatnet. Sum PFOS: Sum branched and linear isomers; Sum PFHxS: Sum branched and linear isomers (for details see tables S8-14)



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3 Figure 2: PFASs levels [ng/L] in fresh water at the in- and outflow of Lake Linnévatnet  
4 as well as from meltwater and snow from the lake Linnévatnet area (data see Table  
5 S9, for locations, see figure S1 in the SM).

6  
7 For Lake Linnévatnet, mainly combined diffusive secondary sources contribute to the  
8 PFASs levels. However, short range atmospheric transport influences cannot entirely  
9 be excluded since Barentsburg, the Russian mining town (78°03'60"N 14°12'60"), is  
10 located about 12 km East of the lake. A considerable variability of the PFAS patterns  
11 is identified in the Lake Linnévatnet surface water samples ( $\Sigma$ PFASs = 0.1 - 4 ng/L),  
12 which seem largely associated with the variable levels of PFBA (<0,03 - 1.1 ng/L)  
13 (Figure 1). The concentrations are greatly dependent on the storage capability of the  
14 respective matrix and the contributing sources. The meltwater runoff from nearby  
15 glaciers and surface snow were probably important sources for the water inflow of the  
16 lake W8-LI31) was dominated by PFBA (Table S8 in the SM), however, both melt water  
17 and surface snow samples collected from the western hills near the lake were rather  
18 dominated by PFOS and PFOA (W-LI32-33). The stages of metamorphosis and  
19 melting of snow were probably affecting the PFAS composition profiles in the snow  
20 and eluting meltwater, where short-chain PFASs elute early during melting leaving  
21 long-chain PFASs in an aged snowpack (Codling et al. 2014; Plassmann et al. 2011),  
22 this coincided with non-detectable PFBA observed in these samples of aged snow. In  
23 general, PFAS levels were low in lake Linnévatnet surface water samples mainly

1 influenced by a variety of diffusive secondary sources. (in contrast to the Ny-Ålesund  
2 back ground fresh water, figure 1). This feature is supported by distinct pattern  
3 differences. All samples with low PFBA were collected at the southern end of the Lake  
4 (figure 1: W-LI20 – 24 & 30). Samples with elevated PFBA (W-Li05-10) stem from  
5 southern locations away from shore (see figure S1). Samples W-Li25-29, indicating  
6 influence of mixed sources, are from the central part of the lake and sample W-Li 14-  
7 16 represent the northern end of the Lake close to the outlet (figure S1). Thus the local  
8 hydrology and surface currents within the lake in combination with the resulting source  
9 strength of different diffusive sources may influence the PFASs patterns.

10 In Ny-Ålesund, seawater samples (W-NA9-14) showed only low levels of PFBA and  
11 PFOS (Table S12 in the SM). Most of the Ny-Ålesund freshwater samples were  
12 collected from run-off meltwater, small ponds and creeks near the settlement and  
13 research installations (Figure S1). The  $\Sigma$ PFASs concentrations in freshwater at the  
14 Ny-Ålesund background sites were found to be, in average, three-times as high (3.4 .  
15 - 9.6 ng/L) compared to Lake Linnévatnet. This can be explained by the fact that the  
16 Ny-Ålesund samples are influenced by a combination of only few both obviously strong  
17 diffusive (mainly melt water run-off) and primary PFAS sources from the Ny-Ålesund  
18 installations and surroundings. Ny-Ålesund freshwater samples were strongly  
19 dominated by PFBA (>90% of  $\Sigma$ PFASs) with small contribution of PFHxS (5-10% of  
20  $\Sigma$ PFASs) and PFOS (2-5 % of  $\Sigma$ PFASs) (Figure 1). This indicates one strong source.  
21 In addition, 6 background soil samples were collected on representative locations at  
22 Ny-Ålesund (Table S13 and Figure S1). In these samples, only minor PFOS levels  
23 were found in three out of eight samples (maximum of 7 ng/g dw for S-NA03; 200 m  
24 North of the local FFTS).

25 In Longyearbyen, both sea water samples ( $n = 3$ ) and run-off water ( $n = 3$ ) were  
26 collected draining from the local FFTS (Figure S1 and Table S2). The seawater  
27 samples were found in the same concentration range as the Ny-Ålesund freshwater  
28 samples (max 6 ng/L). Branched and linear (n-) PFOS were determined and an  
29 approximately 50:50 ratio was found for seawater samples from Longyearbyen and  
30 Ny-Ålesund indicating a significant contribution of diffusive sources to the overall  
31 PFASs contamination (Benskin et al. 2010a; Benskin et al. 2010b).

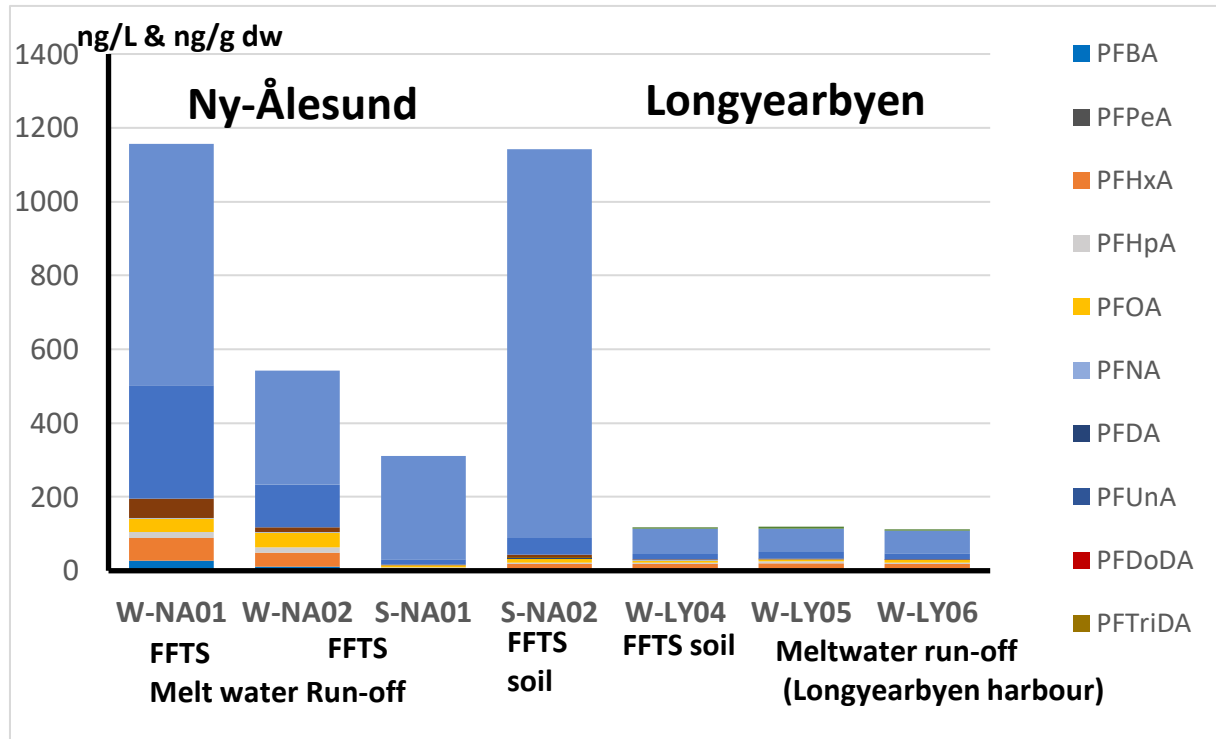
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1 Identification and characterisation of primary sources

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High PFAS concentrations were confirmed for distinct local contamination sources in both Ny-Ålesund and Longyearbyen with up to 100-fold higher  $\Sigma$ PFAS concentrations compared to background levels near the same location (Figure 3 and Tables S8-S13 in the SM).

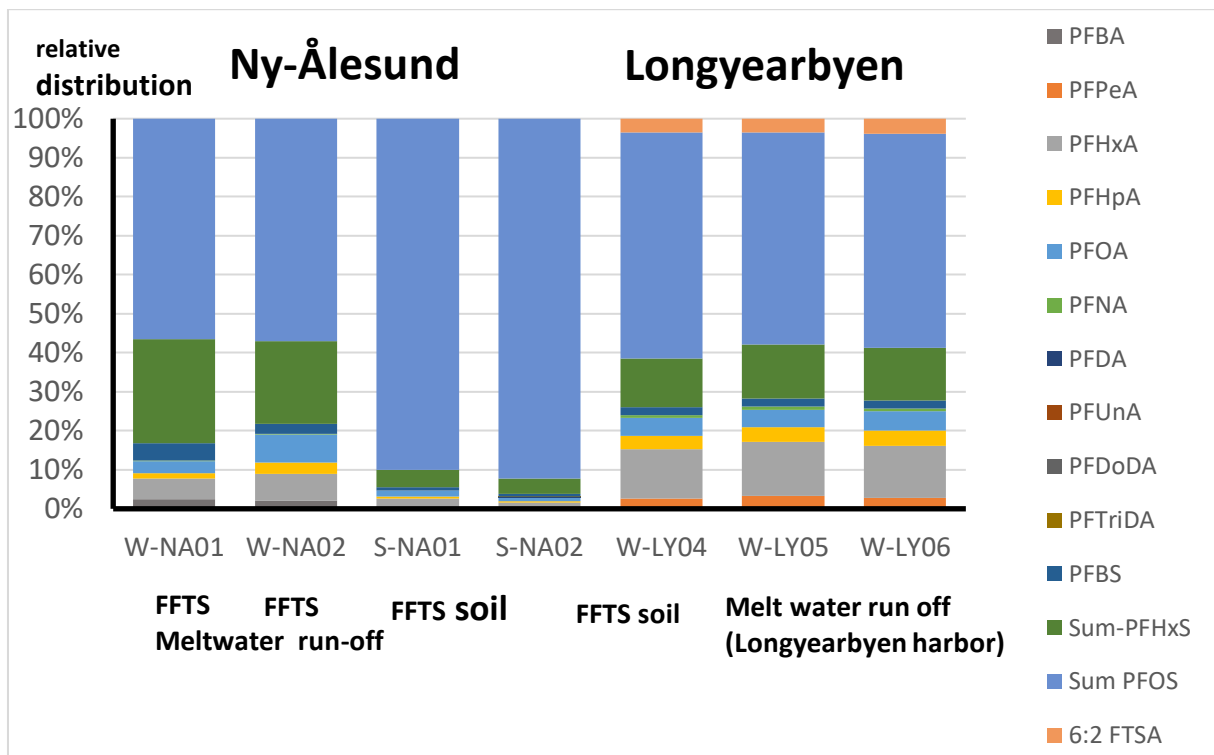


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Figure 3: PFAS concentrations near contaminated locations at Ny-Ålesund and Longyearbyen on Svalbard (for details see tables S8-14).

15 FFTS were identified as the most important local PFAS source near the local airport facilities of Ny-Ålesund and Longyearbyen. The highest concentrations were determined in meltwater run-off water samples taken near the FFTS of the Longyearbyen airport. All three Longyearbyen melt water run-off samples (Figure 3) showed an uniform PFAS profile at  $\Sigma$ PFAS concentrations ranging from 113 – 119 ng/L. PFOS was the predominant compound with 55–58% of the  $\Sigma$ PFASs. Furthermore, the melt-water run-off samples, affected by the Longyearbyen FFTS, contained 6:2 FTSA (2 % of the  $\Sigma$ PFASs). This indicates the use of post-PFOS products applied in modern AFFF as fire prevention tool at the Longyearbyen airport.

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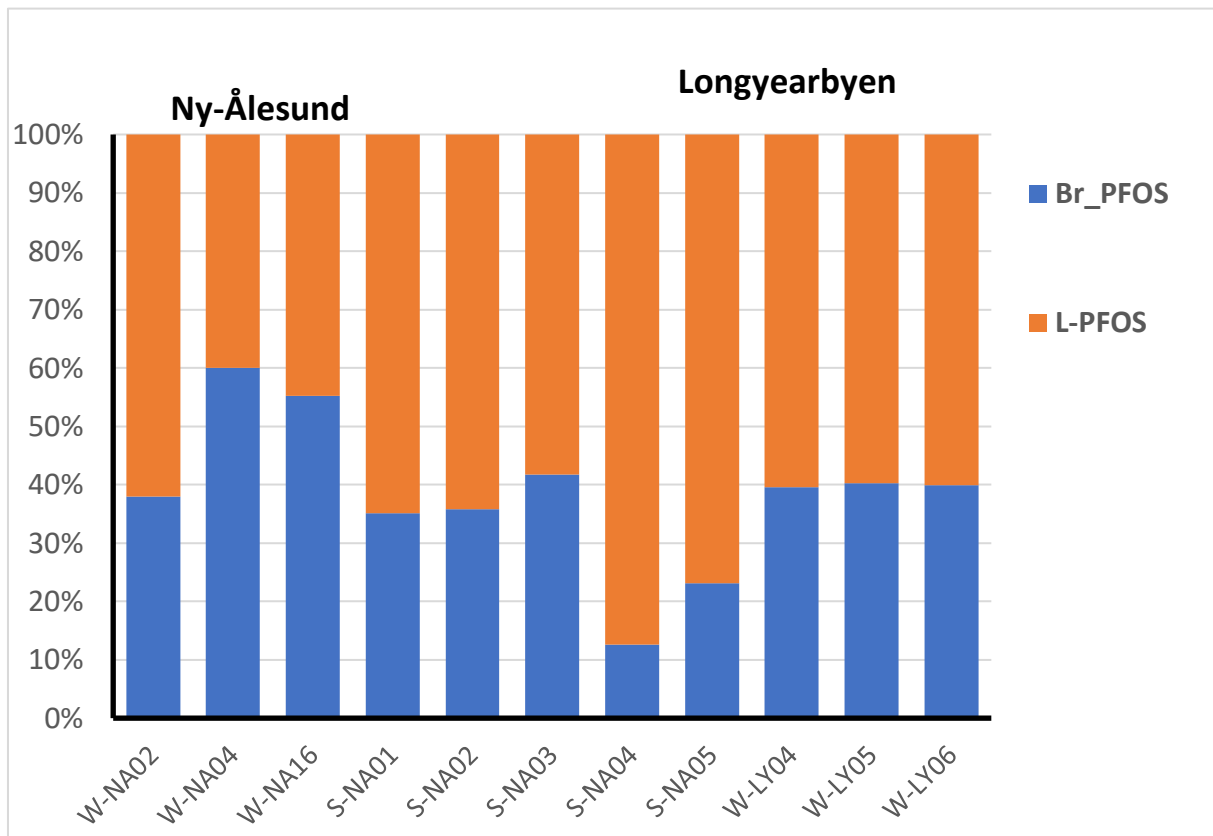
3 Figure 4: Relative PFAS composition profile in percent of PFASs near contaminated  
4 locations at Ny-Ålesund and Longyearbyen on Svalbard (for details see tables S8-14)

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6 In all samples affected by PFAS-containing AFFF at FFTSs, except for sites W-NA04  
7 and W-NA16, the linear (n-) PFOS was dominating the PFOS distribution pattern with  
8 60-80 % (Figure 5). The samples W-NA04 and W-NA16 were collected close to the  
9 FFTS at a parking area and from the drainage of a pond (Solvatnet) near the former  
10 waste water sewage station in Ny-Ålesund. It is possible that these two sites were not  
11 directly influenced by the FFTS (Figure S1) and thus aged PFOS (and PFASs) was  
12 dominated by branched PFOS (Benskin et al. 2010a; Benskin et al. 2010b). PFHxS is  
13 the second most abundant PFAS (307 ng/L) with 26% of the total PFAS concentration  
14 (Figure 3). Combined, sum PFOS and PFHxS account for 82% of the total PFAS  
15 burden in the Ny-Ålesund FFTS run-off water in this study. In ~100 m distance from  
16 the FTSS (W-NA02) the  $\Sigma$ PFAS concentration was reduced to ~50% compared to the  
17 source levels ( $\Sigma$ PFAS = 540 ng/L). The rapid decrease may be due to leaching or  
18 sorption to particles or plants during the transport process at the Ny-Ålesund locations.

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3 Figure 5: Branched and linear (n-) PFOS composition profile (relative distribution) in  
4 contaminated water samples from Ny-Ålesund and Longyearbyen.

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6 For surface soil at Ny-Ålesund, a maximum  $\Sigma$ PFAS concentration of 1140 ng/g dw was  
7 found (S-NA02). In this sample, PFOS is the predominant PFAS similar as for the run-  
8 off water sample from the same location with 92% of the  $\Sigma$ PFAS burden (1054 ng/g  
9 dw). Thus, a total of 94% of the here measured PFAS levels in FFTS surface soil is  
10 explained by PFOS and PFHxS only. The  $\Sigma$ PFAS concentration in the soil sample  
11 collected at ~200 m distance from the Ny-Ålesund FFTS (S-NA01) was three times  
12 lower compared to the soil sample near the FFTS (312 ng/g dw). The PFASs patterns  
13 in this soil sample (S-NA01) was also strongly dominated by PFOS (90 % of  $\Sigma$ PFASs;  
14 281 ng/g dw) due to the high sorption potential of PFOS (Ahrens et al., 2010). However,  
15 the soil samples collected uphill across a creek near the FFTS (S-NA03 and S-NA-04)  
16 (~500 m west of the local FFTS) had considerably lower  $\Sigma$ PFAS concentrations (7.1  
17 ng/g dw and 2.1 ng/g dw, respectively) but still largely dominated by PFOS (> 90 % of  
18  $\Sigma$ PFASs) confirming the relatively homogeneous PFAS distribution for all FFTS  
19 affected samples (run-off water and soil).

1 A comparison with other related studies on PFASs in soil collected at FFTS from  
2 Europe and the USA confirmed, that the here detected concentrations are in the same  
3 order of magnitude, but slightly lower as found in FFTS facilities at large airports (Table  
4 2) (Hale et al. 2017).

5

6 **Table 2:** PFOS levels in contaminated soil collected near airport FFTSs

Location	Levels [ng/g dw]	References
Norway: Oslo airport, Gardemoen	2600	(Hale et al. 2017)
USA. Ellsworth Air Force base	2400	(Houtz et al. 2013)
<b>Svalbard, Ny-Ålesund airport</b>	<b>1140</b>	<b>This study</b>

## 7 **Conclusions and recommendations**

8 In general, PFAS levels in freshwater collected from background sites near Lake  
9 Linnévatnet, Longyearbyen and Ny-Ålesund were found to be in low to ultra-low  
10 concentrations (sub nanograms per litre range). Seawater samples collected along the  
11 harbour area were also low contaminated with PFASs, confirming that potential local  
12 PFAS sources do not yet contribute significantly to the local marine and terrestrial  
13 pollution.

14 However, considerable local contamination was identified in drainage water from the  
15 Longyearbyen FFTS and in soil and freshwater samples collected close to the FFTS  
16 in Ny-Ålesund with concentrations >1000 ng/g dw in soil and >1000 ng/L in water  
17 (Figure 3, table S10 in the SM). We, thus, recommend performing an in-depth scientific  
18 source elucidation and risk assessments followed by appropriate remediation and  
19 mitigation measures in close collaboration with the regulatory authorities and the  
20 involved scientific community in Ny-Ålesund and Longyearbyen. Such a risk  
21 assessment is needed for reduce potential exposure risk as well as to avoid potential  
22 PFAS contamination of ongoing sampling and long-term monitoring activities (i.e.,  
23 Zeppelin mountain monitoring programs). Additionally, potential source like the  
24 facilities in Barentsburg and Svea should be investigated in detail for proper risk  
25 evaluation for exposure of the human population and the environment.

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20

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