

“This is a post-peer-review, pre-copyedit version of an article published in Environmental science and pollution research international. ***The final authenticated version is available online at:***
<http://dx.doi.org/10.1007/s11356-018-2162-4>

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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 4th, 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), Σ 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 (~ 4 ng/L, $n = 2$). The short chain perfluorobutanoate (PFBA) was
14 predominant in lake water (60-80% of the Σ 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 Σ PFAS concentrations were determined for sea
22 water (5 - 6 ng/L), whereas high Σ 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 Σ 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 Σ 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 (PFSA), 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 C₄-C₁₃ PFCAs (i.e., PFBA,
11 PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA) C₄, C₆,
12 C₈ PFSA (i.e., PFBS, PFHxS, PFOS) and 6:2 fluorotelomer sulfonate (6:2 FTSA) (see
13 Table S1 in the Supplementary Material (SM)).

14

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.

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 ($n = 22$).

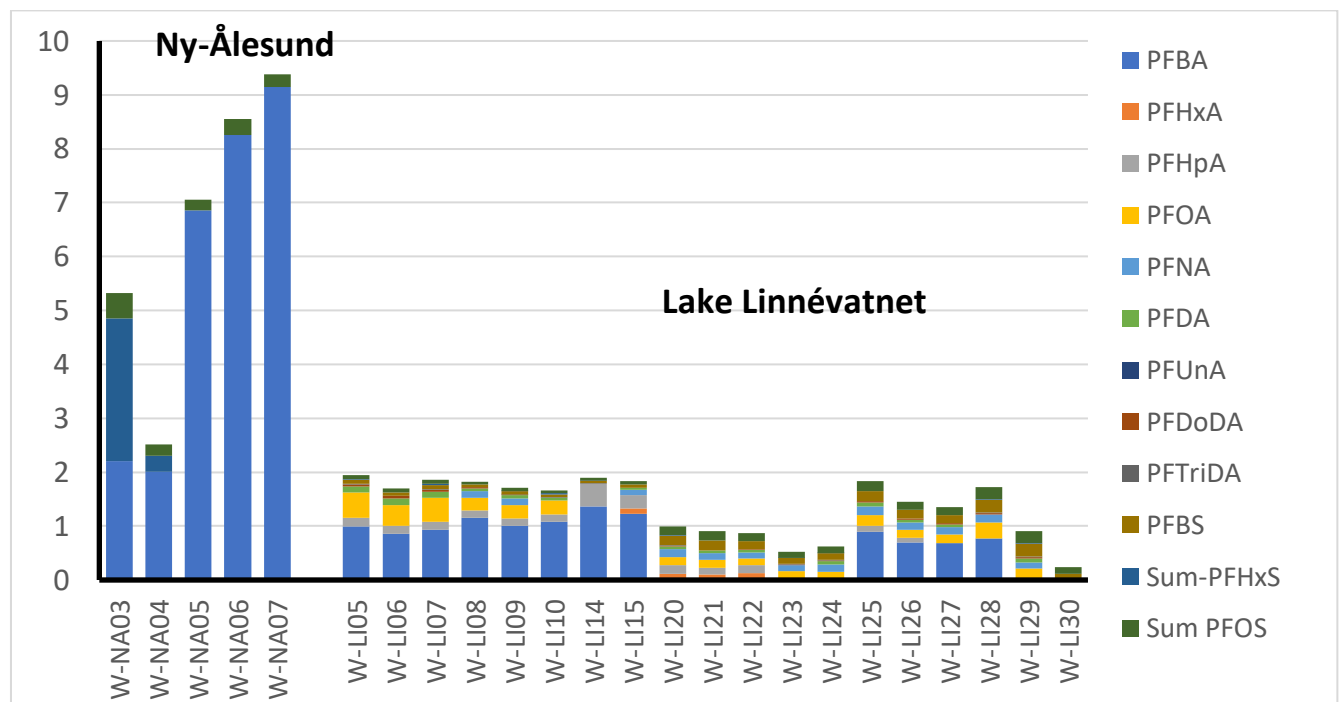
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Table 1: Σ PFASs concentrations in the sample matrices analysed (for details, see Table S7 – S9 in the SM)

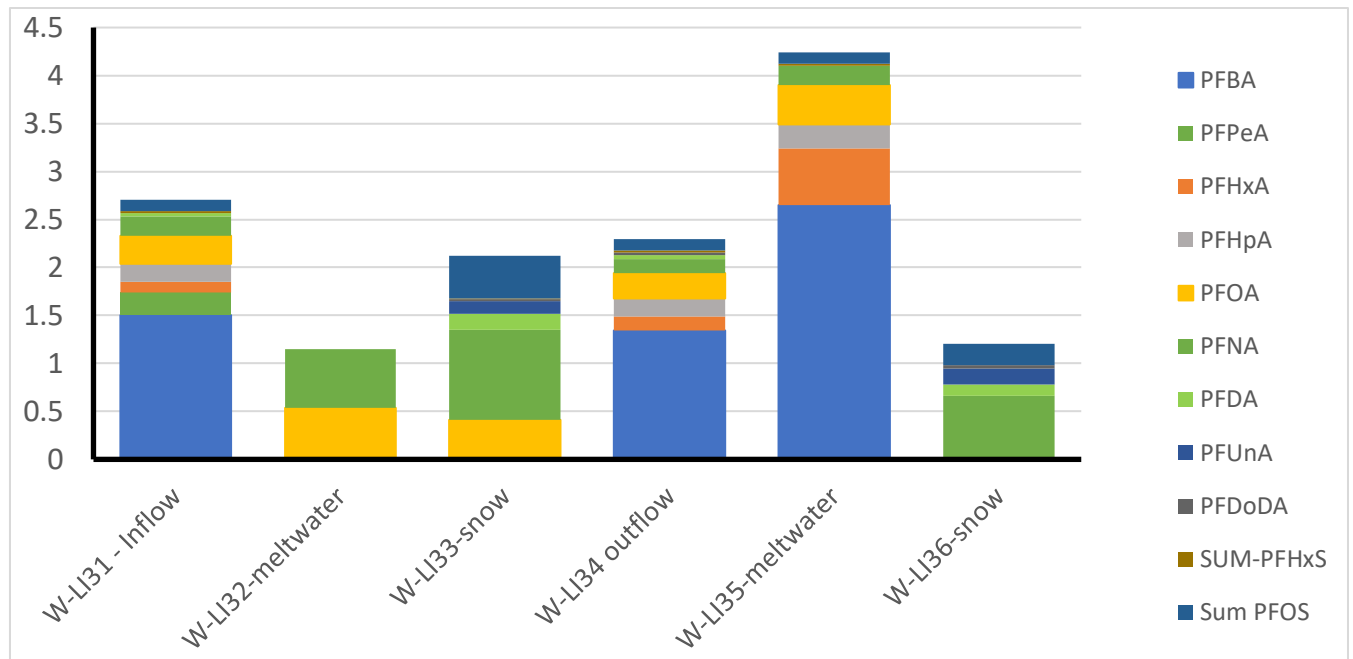
Location	Matrix	No of samples	Range Sum PFAS
Ny-Ålesund	Sea water	6	0.2 – 1.6 ng/L
	Soil	9	<0,03 - 800 ng/g dw
	Freshwater	8	2.7 – 13 ng/L
	Run-off water	2	544 – 1156 ng/L
Longyearbyen	Sea water	2	5 – 6 ng/L
	Run-off water	3	110 – 120 ng/L
Lake Linnévatnet	Lake water	22	<0,03 – 4.1 ng/L
	Meltwater	2	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 (Σ 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 Σ 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 Σ PFASs) with small contribution of PFHxS (5-10% of
20 Σ PFASs) and PFOS (2-5 % of Σ 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).

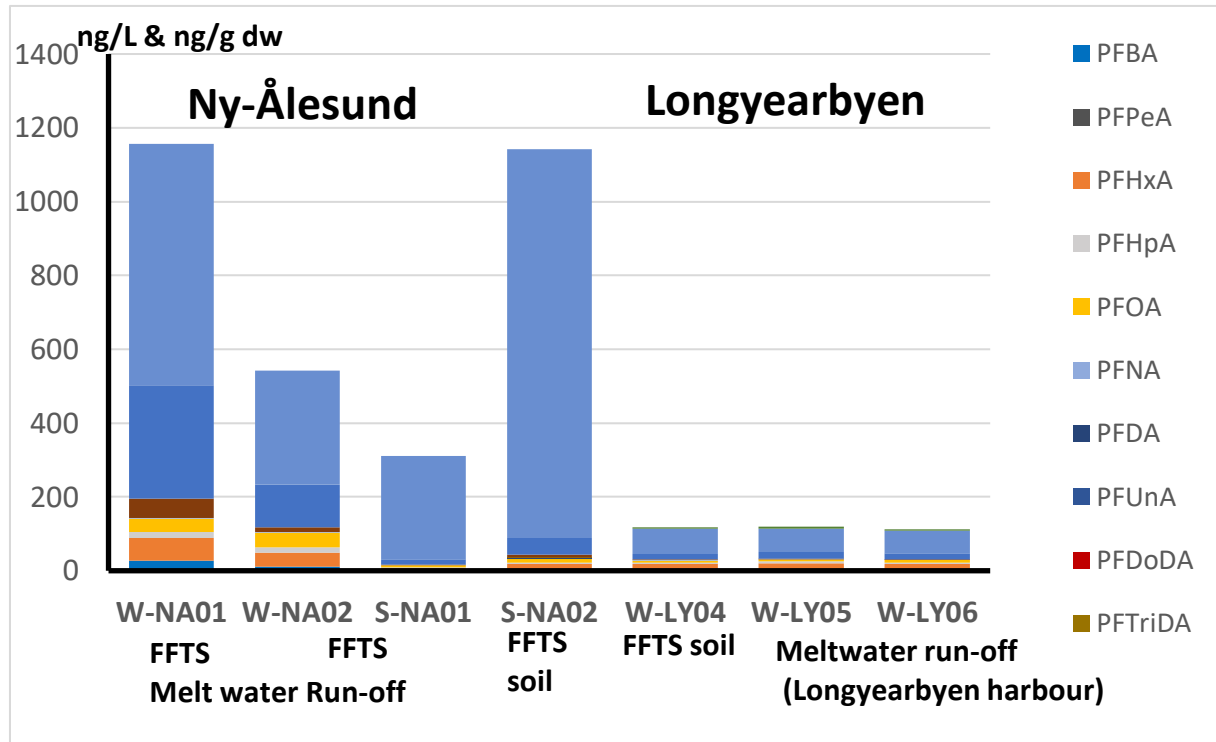
25 In Longyearbyen, both sea water samples ($n = 3$) and run-off water ($n = 3$) were
26 collected draining from the local FFTS (Figure S1 and Table S2). The seawater
27 samples were found in the same concentration range as the Ny-Ålesund freshwater
28 samples (max 6 ng/L). Branched and linear (n-) PFOS 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 Σ PFAS concentrations compared to background levels near the same location (Figure 3 and Tables S8-S13 in the SM).

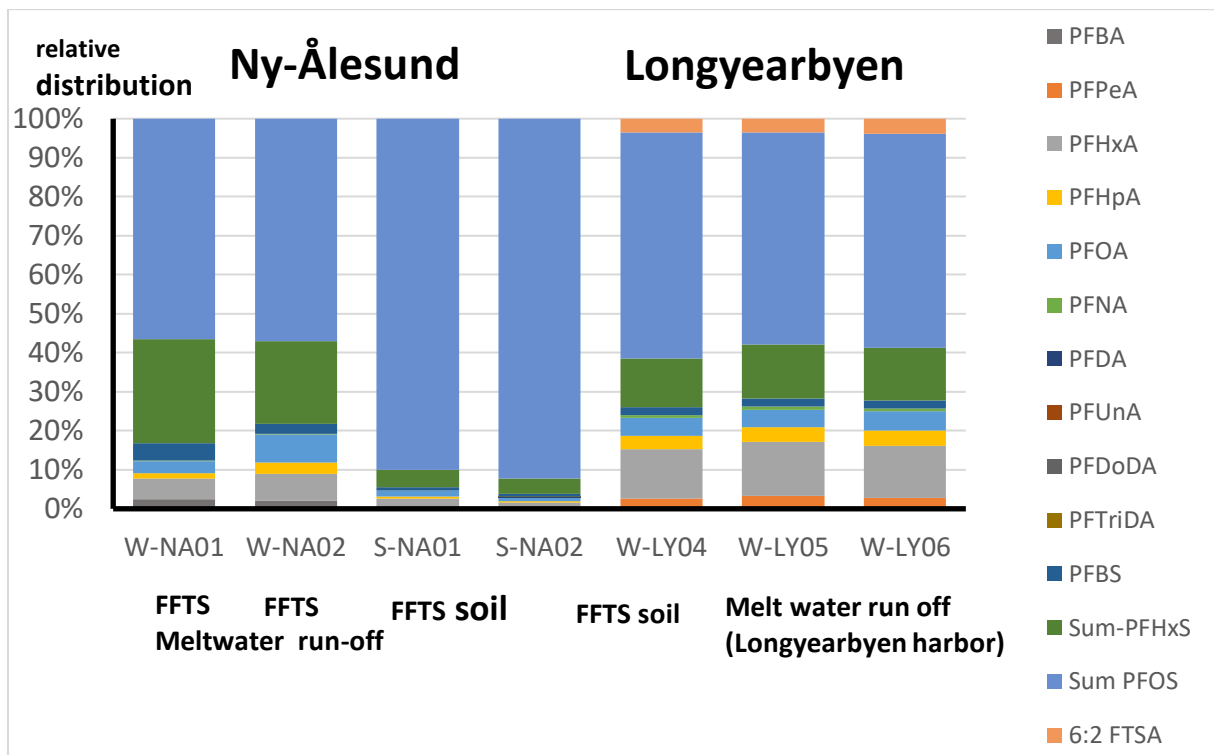


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Figure 3: PFAS concentrations near contaminated locations at Ny-Ålesund and Longyearbyen on Svalbard (for details see tables S8-14).

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

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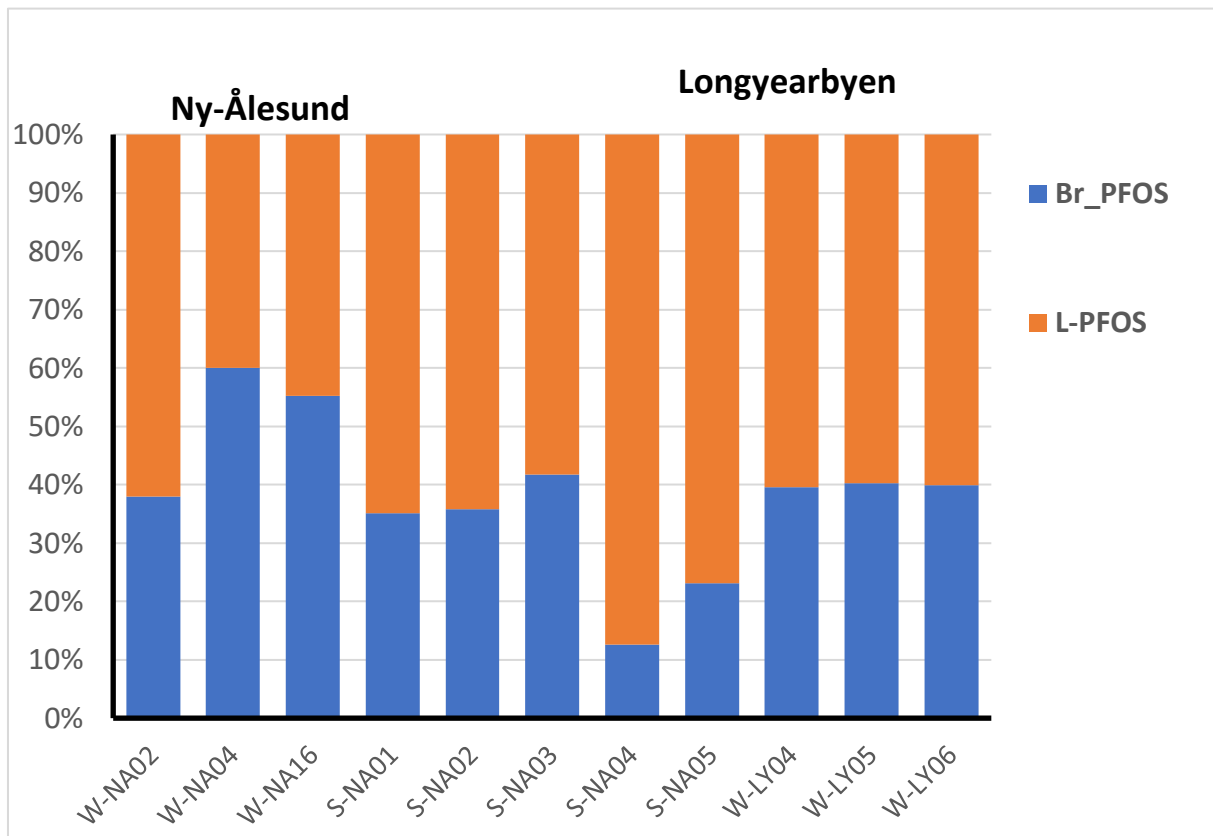
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3 Figure 4: Relative PFAS composition profile in percent of PFASs near contaminated
4 locations at Ny-Ålesund and Longyearbyen on Svalbard (for details see tables S8-14)

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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 Σ PFAS concentration was reduced to ~50% compared to the
17 source levels (Σ 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 Σ 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 Σ 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 Σ 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 Σ 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 Σ PFAS concentrations (7.1
17 ng/g dw and 2.1 ng/g dw, respectively) but still largely dominated by PFOS (> 90 % of
18 Σ 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)
Svalbard, Ny-Ålesund airport	1140	This study

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.

1 Acknowledgement

2 The here performed study was financially supported by the Arctic Monitoring and
3 Assessment Program (AMAP)/ Nordic Council of Ministers (RCN): Project Combined
4 effects of Pollutants and Climate in the Arctic (2014-2016); KingsBay, Ny-Ålesund:
5 Local PFAS contamination in Ny-Ålesund and Research Council of Norway (RCN):
6 BareLab - Integration of the New Lab Facility for Chemical Analyses in Barentsburg in
7 the International Cooperation in the Arctic. The University Centre in Svalbard (UNIS)
8 supported the study with internal funding. We thank the UNIS laboratory engineer
9 Jessica Bosch and PhD scientist Tatiana Drotikova for the laboratory and logistical
10 support during sample preparation at UNIS. The NMBU Master student Stig Magnus
11 Lunde and the SLU Master students Jelena Rakovic and Siri Axelsson supported the
12 field work at the Lake Linnévatnet location. The Lake Linnévatnet and Longyearbyen
13 results were produced as a Master of Science thesis of Jøran Solnes Skaar at the
14 Faculty of Chemistry, Biotechnology and Food Sciences (KBM) of the Norwegian
15 University of Life Sciences (NMBU) in close collaboration with the Faculty of Veterinary
16 Sciences. The authors appreciate the open and trustful collaboration with Kings Bay
17 AS and UNIS logistics during the sampling campaigns. The University Centre in
18 Svalbard allowed access to the Environmental Chemistry laboratory for sample
19 preparation and storage.

20

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21

SUPPLEMENTARY MATERIAL:

**Elucidation of contamination sources for poly- and perfluoroalkyl
substances (PFASs) on Svalbard (Norwegian Arctic)**

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1

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Table S1:

List of target PFASs for quantitative analysis

Analyte	Acronym	CAS#	Formula
PFCAs			
Perfluorobutanoic acid	PFBA	375-22-4	$F(CF_2)_3COOH$
Perfluoropentanoic acid	PFPeA	2706-90-3	$F(CF_2)_4COOH$
Perfluorohexanoic acid	PFHxA	307-24-4	$F(CF_2)_5COOH$
Perfluoroheptanoic acid	PFHpA	375-85-9	$F(CF_2)_6COOH$
Perfluorooctanoic acid	PFOA	335-67-1	$F(CF_2)_7COOH$
Perfluorononanoic acid	PFNA	375-95-1	$F(CF_2)_8COOH$
Perfluorodecanoic acid	PFDA	335-76-2	$F(CF_2)_9COOH$
Perfluoroundecanoic acid	PFUnDA	2058-94-8	$F(CF_2)_{10}COOH$
Perfluorododecanoic acid	PFDoDA	307-55-1	$F(CF_2)_{11}COOH$
Perfluorotridecanoic acid	PFTriDA	72629-94-8	$F(CF_2)_{12}COOH$
PFSAs			
Perfluorobutanoic sulfonic acid	PFBS	375-73-5 or 59933-66-3	$F(CF_2)_4SO_3H$
Perfluorohexanoic sulfonic acid	PFHxS	355-46-4	$F(CF_2)_6SO_3H$
Perfluorooctanoic sulfonic acid	PFOS	1763-23-1	$F(CF_2)_8SO_3H$
FTSAs			
6:2 Fluorotelomer sulfonic acid	6:2 FTSA	27619-97-2	$F(CF_2)_6(CH_2)_2SO_3H$

Table S2: Sample characteristics for the collected environmental samples from A) Ny-Ålesund, B.) Longyearbyen, C.) Lake Linnevatnet

A.) Ny-Ålesund

Sample ID (see fig S1 &S2)	Sample type	Sampling date	GPS Coordinates (WGS84)
W-NA01	Freshwater	22/06/2016	N78.92694 E11.91112
W-NA02	Freshwater	22/06/2016	N78.92851 E11.91476
W- NA03	Freshwater	22/06/2016	N78.92258 E11.88621
W- NA04	Freshwater	22/06/2016	N78.92771 E11.84532
W-NA05	Freshwater	22/06/2016	N78.91825 E11.92187
W-NA06	Freshwater	22/06/2016	N78.91680 E11.93807
W-NA07	Freshwater	22/06/2016	N78.91609 E11.92270
W-NA08	Freshwater	22/06/2016	N78.92445 E11.90311
W-NA09	Seawater	23/06/2016	N78.92743 E11.90141
W-NA10	Seawater	23/06/2016	N78.92855 E11.89795
W-NA11	Seawater	23/06/2016	N78.92987 E11.88421
W-NA12	Seawater	23/06/2016	N78.93246 E11.87649
W-NA13	Seawater	23/06/2016	N78.92860 E11.92930
W-NA14	Seawater	23/06/2016	N78.92980 E11.92205
W-NA15	Freshwater	22/06/2016	N78.91738 E11.86061
W-NA16	Freshwater	23/06/2016	N78.92619 E11.94336
S-NA01	Soil	22/06/2016	N78.92877 E11.91242
S-NA02	Soil	22/06/2016	N78.92880 E11.91109
S-NA03	Soil	24/06/2016	N78.92755 E11.88072
S-NA04	Soil	24/06/2016	N78.92827 E11.87646
S-NA05	Soil	22/06/2016	N78.91875 E11.92477
S-NA06	Soil	22/06/2016	N78.91647 E11.94133
S-NA07	Soil	22/06/2016	N78.91597 E11.91987
S-NA08	Soil	22/06/2016	N78.91535 E11.92624
S-NA10	Soil	23/06/2016	N78.92434 E11.94621

B.) Longyearbyen

Sample ID (see fig S1 &S2)	Sample type	Sampling date	GPS Coordinates (WGS84)
W-LY01	Seawater Marina	14/11/2014	N 78.24022 E15.55480
W-LY02	Seawater Marina	14/11/2014	N 78.24022 E15.55480
W- LY03	Seawater Marina	14/11/2014	N 78.24022 E15.55480
W- LY04	Freshwater run-off (FFTS)	05/06/2015	N78.19498 E15.53396
W-LY05	Freshwater run-off (FFTS)	05/06/2015	N78.19498 E15.53396
W-LY06	Freshwater run-off (FFTS)	05/06/2015	N78.19498 E15.53396

C.) Lake Linnevatnet

Sample ID (see fig S1 &S2)	Sample type	Sampling date	GPS Coordinates (WGS84)
W-LI01	Freshwater	22/03/2014	N78.058160 E13.77365
W-LI05	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI06	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI07	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI08	Freshwater	18/04/2015	N78.039256 E13.83773
W-LI09	Freshwater	18/04/2015	N78.039256 E13.83773
W-LI10	Freshwater	18/04/2015	N78.039256 E13.83773
W-LI14	Freshwater	18/04/2015	N78.06440 E13.77834
W-LI15	Freshwater	18/04/2015	N78.06440 E13.77834
W-LI20	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI21	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI22	Freshwater	18/04/2015	N78.033555 E13.86948
W-LI23	Freshwater	18/04/2015	N78.039256 E13.83773
W-LI24	Freshwater	18/04/2015	N78.039256 E13.83773
W-LI25	Freshwater	18/04/2015	N78.05126 E13.79859
W-LI26	Freshwater	15/06/2015	N78.05126 E13.79859
W-LI27	Freshwater	15/06/2015	N78.05126 E13.79859
W-LI28	Freshwater	15/06/2015	N78.06440 E13.77834
W-LI29	Freshwater	15/06/2015	N78.06440 E13.77834
W-LI30	Freshwater	15/06/2015	N78.06440 E13.77834
W-LI31	inflow LV	14/06/2015	N78.03028 E13.86909
W-LI32	meltwater	16/06/2015	N78.035121 E13.79493
W-LI33	snow	15/06/2015	N78.035121 E13.79493
W-LI34	outflow LV	14/06/2015	N78.06504 E13.782119
W-LI35	Meltwater	16/06/2015	N78.04024 E13.87870
W-LI36	snow	15/06/2015	N78.04024 E13.87870

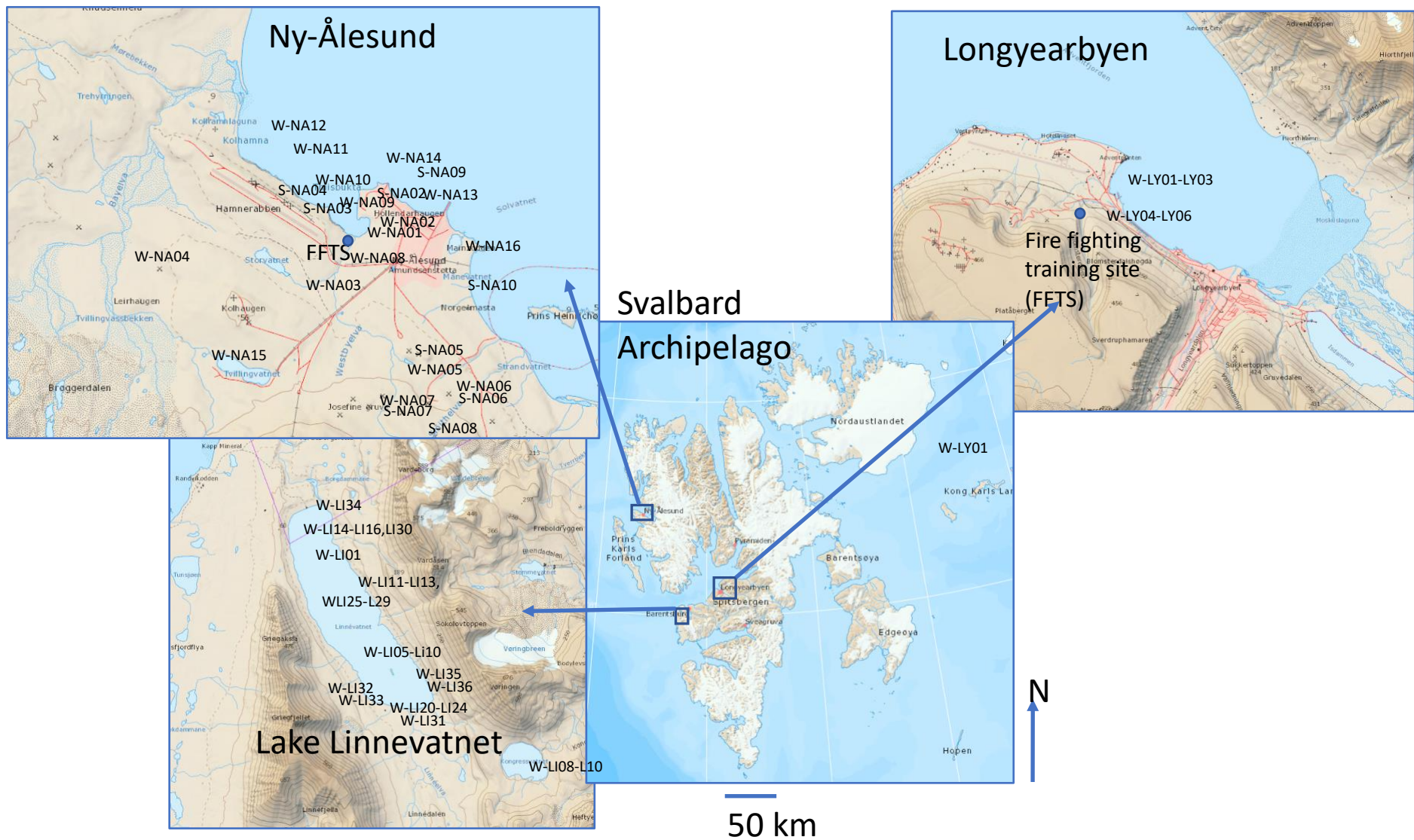


Figure S1:

Figure S1: Sample locations for PFASs screening on Svalbard (Norwegian Arctic). Source, TopoSvalbard, Norwegian Polar Institute (NPI)

Ny-Ålesund



Longyearbyen



Figure S2: Firefighting training sites (FFTS) in Ny-Ålesund and Longyearbyen (source: Google maps).

Sample preparation and analysis of aqueous samples

Samples of water and melted snow were extracted by solid-phase extraction (SPE) using mixed mode reverse phase/weak anion exchange (WAX) resin. The SPE cartridges, Waters Oasis® WAX (500 mg, 6 cc, 60 µm, Waters, Milford MA, USA), were placed on the vacuum Manifold after conditioning (according to manufacturers recommendation). An additional 4 mL of methanol was added and retained in the SPE-cartridge to prevent from drying out during preparation for application of the sample, and a reservoir adapter was placed on top of the cartridge. As far as possible, sample triplicates and field blanks from each location were extracted at the same time. The sample bottle was placed on top of a lab jack elevated above the SPE-assembly, the bottle opening was covered by aluminum foil and the sample bottle was connected to the SPE cartridge through a fitting length of polypropylene tubing (o. d. 1/8"). The loading of the sample was started with vacuum pump at light vacuum (ca.400 mbar). Loading speed was maximum 5 mL/min (ca. 2 drops/second). The internal standard mixture (50 µl of 200 ng/µl Mix of [13C4]-PFBA, [13C5]-PFHxA,[13C4]-PFHpA,[13C4]-PFOA,[13C5]-PFNA,[13C2]-PFDA,[13C2]-PFUnDA,[13C2]-PFDODA,[18O2]-PFHxS,[13C4]-PFOS) was added before pumping. Typical loading time for a 2 L sample was approx. 10 – 24 hours.

The SPE cartridges were cleaned/conditioned with 4 mL acetate buffer to remove salts and other interferences and improve adsorption of target analytes to the sorbent (Taniyasu, et al. 2005, Van Leeuwen, et al. 2009), the eluate was discarded. Afterwards, the cartridges were centrifuged at 1500 rpm for 2 minutes to remove residual solvent.

The cartridges were placed in two 15 mL polypropylene tubes per cartridge and eluted in two different fractions. Fraction 1, containing neutral PFASs, by using 4 mL of methanol and fraction 2, containing ionic PFASs, by 4 mL of 0.1% NH₃ in methanol. The polypropylene tubes were stored at 4 °C before transportation and further treatment and analysis.

Soil and sediment samples

After sampling all samples were carefully dried in an oven for several days at 30 C. The water loss was monitored and the dry weight for PFAS concentration calculation was determined. 2,5 g aliquots were weighed into 50 ml tubes. 1 ml 200mM NaOH was added and the samples allowed to rest for 30 min. 10 ml MeOH was added, along

with internal standard mixture (20 µl of 0,5ng/µl mix of [13C4]-PFBA, [13C5]-PFHxA,[13C4]-PFHpA,[13C4]-PFOA,[13C5]-PFNA,[13C2]-PFDA,[13C2]-PFUnDA,[13C2]-PFDoDA,[18O2]-PFHxS,[13C4]-PFOS) and shaken for 30 min. 100 µL of 2M Hydrochloric acid (HCl) was added and the extract was shaken shortly and centrifuged for 20 min at 3000 rpm. The supernatant was transferred to a 15 mL PP vial and the previous extraction procedure was repeated with 3 ml MeOH. The resulting supernatants were unified and the volume was reduced to 5mL on a Tubovap evaporator (Biotage, Stockholm, Sweden). The extract was further cleaned by adding 0.25 g EnviCarb (Supelco, Sigma Aldrich, Darmstadt Germany). The mixture was centrifuged again (3000 rpm) and the supernatant was transferred to a 15 mL pp vial. 2 ml. methanol was added and the volume was reduced to 0.5 mL on a Turbovap evaporator.

The final extract was transferred to an analytical vial for HPLC-ESI-QqQ analysis and recovery standards ¹³C₈-PFOA was added (50µl of 0,2 ng/µl).

HPLC separation

Chromatographic separation was performed on a Zorbax Eclipse Plus C-18 column (Agilent, 3,5 µm, 2,1 x 150 mm) and a supelguard Discovery C-18 guard column (Supelco, 20 mm x 2,1 mm). As mobile phase 10% methanol in water [A] and methanol [B] were used, both contained 2mM ammonium acetate as ionisation agent. Two different chromatographic separation methods were used for PFCAs and for PFSAAs. For PFCAs, the gradient started with 85 % [B] held for 5 minutes, then increased linearly over 5 minutes to 99 % [B], this was held constant for 7 minutes then changed linearly over 1 minute to 1 % [B] until end of analysis at 26 minutes.

For PFSAAs, FASAAs and FASEs the gradient started with 85 % [B] held for 5 minutes, then increased linearly over 5 minutes to 99 % [B], this was held constant for 7 minutes then changed linearly over 1 minute to 10 % [B] and held for 7 minutes before increased linearly over 2 minutes to 85 % [B].

Quantitative analysis for soil and aqueous samples

Detection and characterisation were done on two triple quadrupole mass spectrometers in multiple reaction monitoring (MRM) mode. For the analysis of the Lake Linnévatnet and the Longyearbyen samples, an Agilent 1200 HPLC system coupled to an Agilent 6460 series triple quadrupole MS/MS system was used (Agilent,

Santa Clara, USA). The Agilent QqQ mass spectrometer was operated with the Agilent jet stream electrospray ionization (AJS-ESI) source. Ion source parameters, MS/MS parameters and MRM transitions are given in tables S3-S4. For the Ny-Ålesund samples, an Agilent 1200 HPLC system coupled an API 3000 triple quadrupole Mass spectrometer (Sciex, Stockholm, Sweden) with instrumental parameters as described in tables S5-S6 was used.

Instrumental parameters

Table S3. MRM transitions and MS/MS parameters. Agilent 6460, instrument operated in ESI⁻

Acronym	ISTD used	Precursor ion (m/z)	Product ion 1 (Quantifier) (m/z)	Product ion 2 (Qualifier) (m/z)	Qualifier relative abundance (%)	CE (V)	Fragmentor (V)
PFBA	[¹³ C ₄]-PFBA	213	169			1	61
PFPeA	[¹³ C ₅]-PFHxA	263	219			1	61
PFHxA	[¹³ C ₅]-PFHxA	313	269	119	4.6	0 (12)	66
PFHpA	[¹³ C ₄]-PFHpA	363	319	169	13.2	0 (8)	71
PFOA	[¹³ C ₄]-PFOA	413	369	169	30.1	0 (12)	76
PFNA	[¹³ C ₅]-PFNA	463	419	219	9.9	4 (8)	86
PFDA	[¹³ C ₂]-PFDA	513	469	219	12.9	4 (12)	86
PFUnDA	[¹³ C ₂]-PFUnDA	563	519			4	86
PFDODA	[¹³ C ₂]-PFDODA	613	569			4	96
PFTriDA	[¹³ C ₂]-PFDODA	663	619			4	106
PFBS	[¹⁸ O ₂]-PFHxS	299	99	80	39.9	25 (33)	121
PFHxS	[¹⁸ O ₂]-PFHxS	399	99	80	52.1	45	151
Br-PFOS	[¹³ C ₄]-PFOS	499	99	80	16.7	61	166
L-PFOS	[¹³ C ₄]-PFOS	499	99	80	46.2	61	166
6:2 FTSA	[¹⁸ O ₂]-PFHxS	427	407	81	14.3	15	145

Acronym	Precursor ion (m/z)	Product ion 1 (Quantifier) (m/z)	Product ion 2 (Qualifier) (m/z)	CE (V)	Fragmentor (V)
Recovery standard					
[¹³ C ₈]-PFOA	421	376		0	76
Internal standards					
[¹³ C ₄]-PFBA	217	172		1	61
[¹³ C ₅]-PFHxA	10.42	318	273	0	66
[¹³ C ₄]-PFHpA	367	322		0	66
[¹³ C ₄]-PFOA	417	372		0	76
[¹³ C ₅]-PFNA	468	423		4	76
[¹³ C ₂]-PFDA	515	470		4	86
[¹³ C ₂]-PFUnDA	565	520		4	96
[¹³ C ₂]-PFDoDA	615	570		4	96
[¹⁸ O ₂]-PFHxS	403	84		49	146
[¹³ C ₄]-PFOS	503	80		61	180

a. PFCA instrument method., PFSA/FASA/FASE instrument method, Fragmentor voltages in parenthesis represent qualifier transition, if different from quantifier.

Table S4. Ion source parameters Agilent 6460.

	PFCAs	PFSA, FASAs and FASEs
Gas Flow [L/min]	5	9
Gas temp [°C]	300	350
Nebulizer [psi]	25	30
Sheath Gas Flow [mL/min]	8	8
Sheath Gas Heater [°C]	400	400
Capillary [V]	+5000 / -2500	+5000 / -4000
Charging [V]	+2000 / -500	+2000 / 0

Table S5. MRM transitions and MS/MS parameters. API 3000, instrument operated in ESI-

Acronym	ISTD used	Precursor ion (m/z)	Product ion 1 (Quantifier) (m/z)	Product ion 2 (Qualifier) (m/z)	CE (V)
PFBA	[¹³ C ₄]-PFOA	213	169	119	-13
PFHxA	[¹³ C ₄]-PFOA	313	269	169	-12
PFHpA	[¹³ C ₄]-PFOA	363	319	169	-14
PFOA	[¹³ C ₄]-PFOA	413	369	219	-15
PFNA	[¹³ C ₅]-PFNA	463	419		-15
PFDA	[¹³ C ₂]-PFDA	513	469		-15
PFUnDA	[¹³ C ₂]-PFUnDA	563	519		-15
PFDoDA	[¹³ C ₂]-PFDoDA	613	569		-16
PFTriDA	¹³ C ₂ -PFDoDA	663	619	99	-17
PFBS	[¹⁸ O ₂]-PFHxS	299	80	99	-50
PFHxS	[¹⁸ O ₂]-PFHxS	399	80	99	-57
Br-PFOS	[¹³ C ₄]-PFOS	499	80	99	-72
L-PFOS	[¹³ C ₄]-PFOS	499	80		-72
Recovery standard					
	[¹³ C ₆]-PFOA	421	376		
Internal standards					
	[¹³ C ₄]-PFOA	417	372		-15
	[¹³ C ₅]-PFNA	468	423		-15
	[¹³ C ₂]-PFDA	515	470		-15
	[¹³ C ₂]-PFUnDA	565	520		-15
	[¹³ C ₂]-PFDoDA	615	570		-15
	[¹⁸ O ₂]-PFHxS	403	103		-57
	[¹³ C ₄]-PFOS	503	99		-74

Table S6. Ion source parameters API 3000.

	PFCAs	PFSAs, FASAs and FASEs
Nebulizer	8	7
Curtain gas	8	8
Collision gas	9	10
Ion spray	-1500	-3000
Temperature	500	450

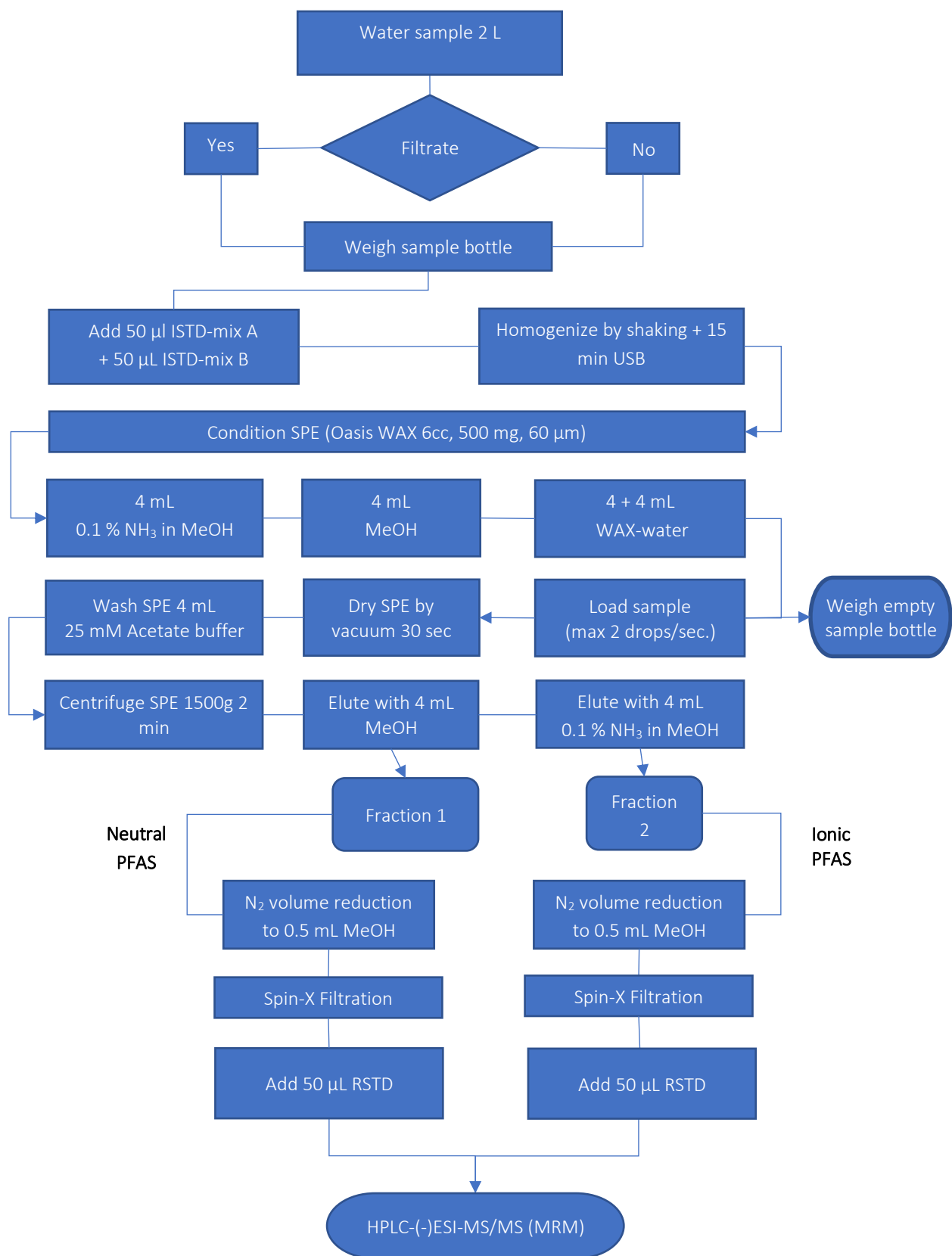


Figure S2: Sample preparation scheme for the quantitative analysis of PFASs in aqueous samples (fresh water and Sea water).

For the samples quantified with the Agilent instrumentation, the MassHunter software (MassHunter Workstation Software: Quantitative analysis for QQQ version B.07.00 / Build 7.0.457.0 Agilent Technologies, Santa Clara, CA, USA) was used. For the samples quantified with ABSiex instrumentation, the Analyst software (Analyst, TF 1.7.1, Sciex, Redwood City CA USA) was applied. All samples were quantified within 3 weeks after sampling in order to avoid prolonged storage time.

Quality control

All solvent and consumables used were of ultra-pure quality purchased by Sigma Aldrich and Merck, Darmstadt, Germany. An integrated quality control program was applied for the here performed survey. All glassware was cleaned in an industrial dishwasher machine the program included rinsing with MilliQ water. After the dishwasher, a manual cleaning followed by rinsing with acetone followed by methanol. Finally, all glass equipment was heated in a muffle furnace at 450 °C for 6 hours. Plastic tubes and other inflammable equipment was cleaned thoroughly with methanol before usage. Pre-cleaned equipment was packed in aluminium foil prior to usage. Fume hoods and other working surfaces were cleaned and rinsed with methanol and the working surfaces were covered with pre-cleaned aluminium foil prior to sample handling. Furthermore, all direct contact with fluoro-polymers, e.g. PTFE, was avoided. Only MilliQ-water, purified through Oasis WAX SPE-cartridges (WAX-water) was used for reagents and blanks.

Instrumental analysis

All fluorinated seals and tubing in the instrument were replaced with non-fluorinated alternatives. A scavenger cartridge between pump and injector was installed to remove contaminants from the degasser, connecting tubes and mobile phase. For every tenth injection of solvent sample or blank, and instrument blank consisting of pure methanol was analysed.

Blanks, detection- and quantification limits

Table S6. Detection and quantification limits for selected PFASs

Acronym	IDL [ng]	LOD (5 g soil) [ng/g dw]	LOQ (5 g soil) [ng/g dw]	LOD (2 L sample) [ng/L]	LOQ (2 L sample) [ng/L]
PFBS	0.003	0.001	0.002	0.003	0.025
PFHxS	0.003	0.001	0.002	0.005	0.006
Br-PFOS	NA	0.001	0.002	0.012	0.019
L-PFOS	0.003	0.001	0.002	0.015	0.020
6:2 FTSA	0.015	0.001	0.002	0.008	0.14
PFBA	0.084	0.02	0.04	0.50	0.68
PFPeA	0.012	0.005	0.01	0.024	0.10
PFHxA	0.025	0.05	0.08	0.028	0.090
PFHpA	0.032	0.06	0.1	0.033	0.089
PFOA	0.059	0.01	0.05	0.031	0.14
PFNA	0.026	0.005	0.01	0.021	0.085
PFDA	0.017	0.003	0.08	0.008	0.036
PFUnDA	NA	0.001	0.002	0.009	0.12
PFDoDA	NA	0.001	0.002	0.005	0.016

- Determined by average field blank + 3*SD.
 - LOQ>MDL, LOQ was determined by average field blank + 10*SD.
 - No blank contamination, LOQ set to S/N x 10 in real sample.
- NA = no standards were available for the calculation.

Field blanks were prepared by filling 250 mL of WAX-water to pre-cleaned 1 L polyethylene- or 2 L polypropylene bottles. The caps of the field blanks were left open for the whole duration of sampling at each site (5 to 10 minutes). The field blanks were transported, stored, extracted and analysed the same way as regular samples. A laboratory/method blank was prepared by adding 250 mL of pre-cleaned water to three 250 mL polyethylene bottles. They were further extracted and analysed according to the here applied method.

For the Lake Linnévatnet and Longyearbyen location a total of 19 field blank samples (9 water and 10 soil blanks) and for the Ny-Ålesund samples 4 blank samples (2 soil, 2 xwater) were analysed. None showed significantly elevated PFAS levels. In addition, for instrumental blank, 10 µL of methanol was injected for every 10 samples or matrix blanks injected. Instrument detection limits (IDL), determined as S/N x 3 in the standard analysis were determined for the three lowest calibrations standards. The method detection limit (LOD) was determined as S/N x 3 for field blank samples. The Limit of quantification (LOQ) was calculated as average LOD plus three times the standard

deviation. For all samples, no blank correction for LOD, LOQ of level determination was performed in the here conducted survey.

Recovery rates.

In addition to individual sample recovery rates, dedicated recovery experiments (matrix addition and solvent mixtures) were performed as integrated part of the method validation (table S7).

Table S7: Recovery rates. The prefix “M” refers to internal ¹³C labelled standards applied for volume correction and quantification.

Acronym	Spiked blank (n = 3)		Spiked sample matrix (n = 4)		Samples (n = 30)		Field blanks (n = 23)		Lab. Blanks (n = 6)	
	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)
PFBA	120	26	92	5.1						
PFPeA	90	15	87	6.0						
PFHxA	71	8.9	83	3.6						
PFHpA	96	12	105	3.9						
PFOA	85	6.0	92	1.9						
PFNA	99	7.4	106	4.9						
PFDA	100	20	95	4.9						
PFBS	78	11	76	1.7						
PFHxS	77	9.9	92	1.2						
PFOS	84	8.0	94	2.4						
6:2 FTSA	85	12	87	1.6						
[¹³ C ₄]-PFBA	100	20	89	4.4	81	19	90	6.3	87	6
[¹³ C ₅]-PFHxA	71	8.7	78	7.1	66	21	57	16	63	17
[¹³ C ₄]-PFHpA	94	9.0	97	7.9	84	24	78	20	82	24
[¹³ C ₄]-PFOA	87	4.9	93	2.0	88	13	86	5.3	87	5.4
[¹³ C ₅]-PFNA	98	5.1	103	4.9	94	15	85	7.3	99	5.5
[¹³ C ₂]-PFDA	99	16	93	6.6	88	16	83	4.2	88	5.9
[¹³ C ₂]-PFUnDA	107	18	93	10	84	20	79	1.4	87	7.8
[¹³ C ₂]-PFDoDA	84	12	66	14	62	18	54	11	67	13
[¹⁸ O ₂]-PFHxS	79	7.5	91	2.8	78	13	80	4.3	86	4.3
[¹³ C ₄]-PFOS	86	8.1	94	3.9	82	14	72	5.6	82	3.8

Table S8: Background PFASs levels [ng/L] in Lake Linnévatnet surface freshwater. Only levels above LOQ are listed.

Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers

Matrix	Name	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFDoDA	SUM-PFHxS	Sum PFOS	SUM PFASs
Freshwater	W-LI01	1.09	<0.01	<0.02	0.26	1.78	<0.03	0.61	0.16	0.023	0.19	4.11
Freshwater	W-LI05	0.99	<0.01	<0.02	0.17	0.47	<0.03	0.11	0.033	0.01	0.079	1.86
Freshwater	W-LI06	0.86	<0.01	<0.02	0.14	0.39	<0.03	0.12	0.047	<0.005	0.068	1.63
Freshwater	W-LI07	0.93	<0.01	<0.02	0.15	0.45	<0.03	0.11	0.029	0.015	0.082	1.77
Freshwater	W-LI08	1.15	<0.01	<0.02	0.14	0.24	0.12	0.05	0.019	<0.005	0.053	1.77
Freshwater	W-LI09	1.01	<0.01	<0.02	0.13	0.25	0.12	0.061	0.015	0.007	0.055	1.65
Freshwater	W-LI10	1.08	<0.01	<0.02	0.14	0.26	<0.03	0.043	<0.02	0.014	0.056	1.59
Freshwater	W-LI14	1.37	<0.01	<0.02	0.42	<0.06	<0.03	<0.02	<0.02	0.007	0.044	1.84
Freshwater	W-LI15	1.23	<0.01	0.1	0.24	<0.06	0.1	0.041	<0.02	<0.005	0.059	1.77
Freshwater	W-LI20	<0.08	<0.01	0.12	0.15	0.16	0.14	0.052	0.017	0.01	0.17	0.82
Freshwater	W-LI21	<0.08	<0.01	0.1	0.13	0.14	0.13	0.048	<0.02	<LOD	0.18	0.73
Freshwater	W-LI22	<0.08	<0.01	0.13	0.14	0.13	0.11	0.049	0.015	<0.005	0.15	0.72
Freshwater	W-LI23	<0.08	<0.01	<0.02	<0.03	0.16	0.11	<0.02	0.021	0.007	0.11	0.41
Freshwater	W-LI24	<0.08	<0.01	<0.02	<0.03	0.15	0.14	0.061	0.014	0.008	0.12	0.49
Freshwater	W-LI25	0.89	<0.01	<0.02	0.11	0.2	0.16	0.076	0.024	<0.005	0.19	1.65
Freshwater	W-LI26	0.7	<0.01	<0.02	0.087	0.14	0.14	0.054	0.016	<0.005	0.16	1.30
Freshwater	W-LI27	0.68	<0.01	<0.02	<0.03	0.16	0.14	0.048	0.021	<0.005	0.15	1.20
Freshwater	W-LI28	0.77	<0.01	<0.02	<0.03	0.3	0.14	<0.02	0.026	0.016	0.23	1.48
Freshwater	W-LI29	<0.08	<0.01	<0.02	<0.03	0.21	0.11	0.083	0.025	0.011	0.23	0.67
Freshwater	W-LI30	<0.08	<0.01	<0.02	<0.03	<0.06	<0.03	<0.02	<0.02	<0.005	0.12	0.12

Table S9: Diffusive PFASs sources, inflow and outflow into Lake Linnévatnet, meltwater and snow [ng/L]. Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers

Matrix	Name	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnD A	PFDoD A	PFBS	Sum- PFHxS	Sum PFOS	SUM PFAS
Seawater	W-NA09	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND
Seawater	W-NA10	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	0.10
Seawater	W-NA11	0.61	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	0.61
Seawater	W-NA12	1.51	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.09	1.60
Seawater	W-NA13	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND
Seawater	W-NA14	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND

Table S10: PFASs levels [ng/L] in Longyearbyen water. Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers.

Matrix	Name	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFBS	Sum-PFHxS	Br-PFOS	L-PFOS	Sum PFOS	6:2 FTSA	Sum PFAS
Sea water	W-LY02	<0.08	1.29	2.66	0.57	0.31	0.03	0.035	0.21	0.088	0.1	0.19	0.02	5.265
	W-LY03	<0.08	1.55	3.02	0.4	0.31	<0.08	0.14	0.43	0.2	0.24	0.44	<0.08	6.29
Run-off water	W-LY04	<0.08	3.16	14.8	4.07	5.53	0.85	2.4	14.8	27	41.2	68.3	4.25	118.16
	W-LY05	<0.08	3.86	16.5	4.48	5.35	0.86	2.41	16.5	26	38.6	64.5	4.17	118.63
	W-LY06	<0.08	3.07	15.2	4.32	5.62	0.87	2.33	15.2	24.7	37.2	61.9	4.35	112.86

Table S11: PFASs levels [water: ng/L & soil ng/g dw] in contaminated Ny-Ålesund and Longyearbyen samples (soil and water: >100 ng/L or ng/g dw). Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers.

Matrix	Name	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnD A	PFDoD A	PFBS	Sum-PFHxS	Sum PFOS	6:2 FTSA	Sum PFAS
Freshwater	W-NA01	28.08	<0.02	61.47	15.43	35.79	1.81	<0.008	<0.009	<0.002	52.55	307.51	653.58	<0.002	1156
	W-NA02	11.33	<0.02	37.48	15.23	39.28	1.24	<0.08	<0.009	<0.002	13.91	114.63	310.01	<0.002	544
	W-NA-16	<0.03	<0.02	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	2.57	30.36	76.59	<0.002	137
Soil	S-NA01	1.14	<0.01	6.86	1.96	4.68	<0.005	<0.05	<0.001	<0.001	2.64	13.82	280,46	<0.001	312
	S-NA02	1.40	<0.01	16.81	4.02	9.92	0.73	0.86	1.18	0.48	7.13	45.02	1054,53	<0.001	1142
Waste water effluent	W-LY04	<0.03	3.16	5,1	0,68	11	36	3,9	11	1,5	<0,02	0,35	1	2,9	65
	W-LY05	<0.03	3.86	16.50	4.48	5.35	0.86	<0.008	<0.009	<0.002	2.41	16.50	64.50	4.17	119
FFTS run-off	W-LY06	<0.03	3.07	15.20	4.32	5.62	0.87	<0.008	<0.009	<0002	2.33	15.20	61.90	4.35	110

Table S12: PFASs in seawater from Ny-Ålesund [ng/L]. Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers.^a

Matrix	Name	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnD A	PFDoD A	PFBS	Sum- PFHxS	Sum PFOS	SUM PFAS
Seawater	W-NA09	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND
Seawater	W-NA10	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	0.10
Seawater	W-NA11	0.61	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	0.61
Seawater	W-NA12	1.51	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.09	1.60
Seawater	W-NA13	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND
Seawater	W-NA14	<0.5	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	<0.02	ND

Table S13: PFASs in background soil from Ny-Ålesund [ng/g/ dw]. Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers.^a

Matrix	Name	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnD A	PFDoD A	PFBS	Sum- PFHxS	Sum PFOS	SUM PFAS
Soil	S-NA03	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	7.06	7.06
Soil	S-NA04	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	2.31	2.31
Soil	S-NA05	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	0.91	0.91
Soil	S-NA06	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	<0.002	ND
Soil	S-NA07	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	<0.002	ND
Soil	S-NA08	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	<0.002	ND
Soil	S-NA09	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	<0.002	ND
Soil	S-NA10	<0.04	<0.05	<0.06	<0.01	<0.005	<0.003	<0.001	<0.001	<0.001	<0.001	<0.002	ND

Table S14: PFASs in background fresh water from Ny-Ålesund [ng/L]. Only levels above LOQ are listed. Sum PFHxS: Sum of branched and linear isomers, Sum PFOS: Sum of Branched and linear isomers.^a

Matrix	Name	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	Sum-PFHxS	Sum PFOS	SUM PFAS
Fresh water	W-NA03	2.2	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	2.65	0.47	8.4
Fresh water	W-NA04	2.0	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	0.3	0.2	3.4
Fresh water	W-NA05	6.9	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.2	7.3
Fresh water	W-NA06	8.25	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.3	8.9
Fresh water	W-NA07	9.1	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.23	9.6
Fresh water	W-NA08	6.9	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	<0.005	0.21	7.1
Fresh water	W-NA15	5.1	<0.03	<0.03	<0.03	<0.02	<0.008	<0.009	<0.005	<0.003	0.15	0.78	6.9

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