SUPPLEMENTARY MATERIAL:

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

Jøran Solnes Skaar[1], Jan Ludvig Lyche[2], Erik Magnus Ræder[2], Lutz Ahrens[3], **Roland Kallenborn**[*4,5]

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1. Norwegian Institute for Air Research (NILU), 2027 Kjeller, Norway
2. Faculty of Veterinary Medicine, Norwegian University of Life Sciences (NMBU), 0033 Oslo, Norway
3. Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences (SLU), Box 7050, 750 07 Uppsala, Sweden
4. Faculty of Chemistry, Biotechnology and Food Sciences (KBM), Norwegian University of Life Sciences (NMBU), 1432 Ås, Norway
5. Department of Arctic Technology (AT), University Centre in Svalbard (UNIS), 9171 Longyearbyen, Svalbard, Norway

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

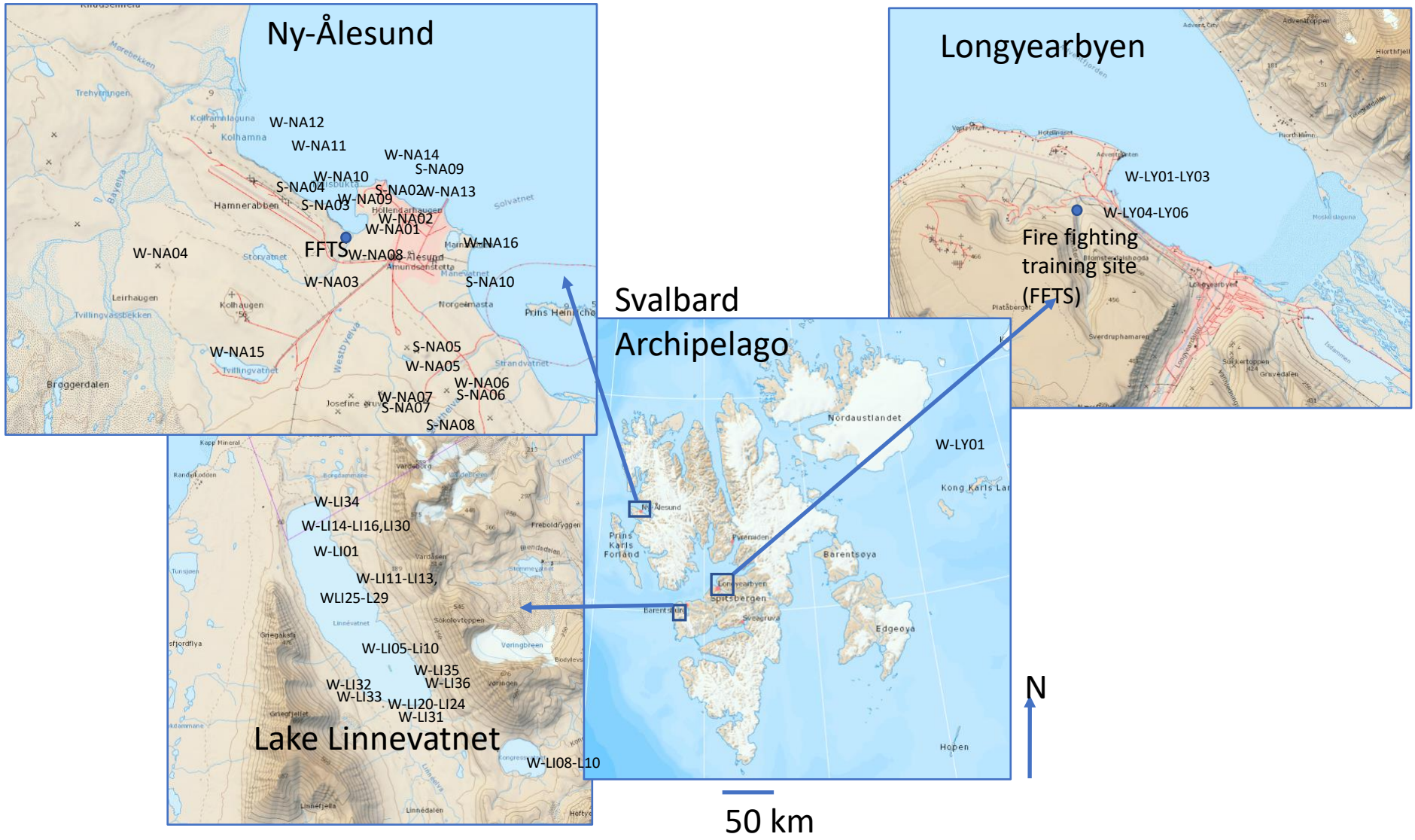

Figure S1:

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% NH_3 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 $^{13}C_8$-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 PFSAs.

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 PFSAs, FASAs 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	PFSAs, 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					
$[^{13}C_8]$-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

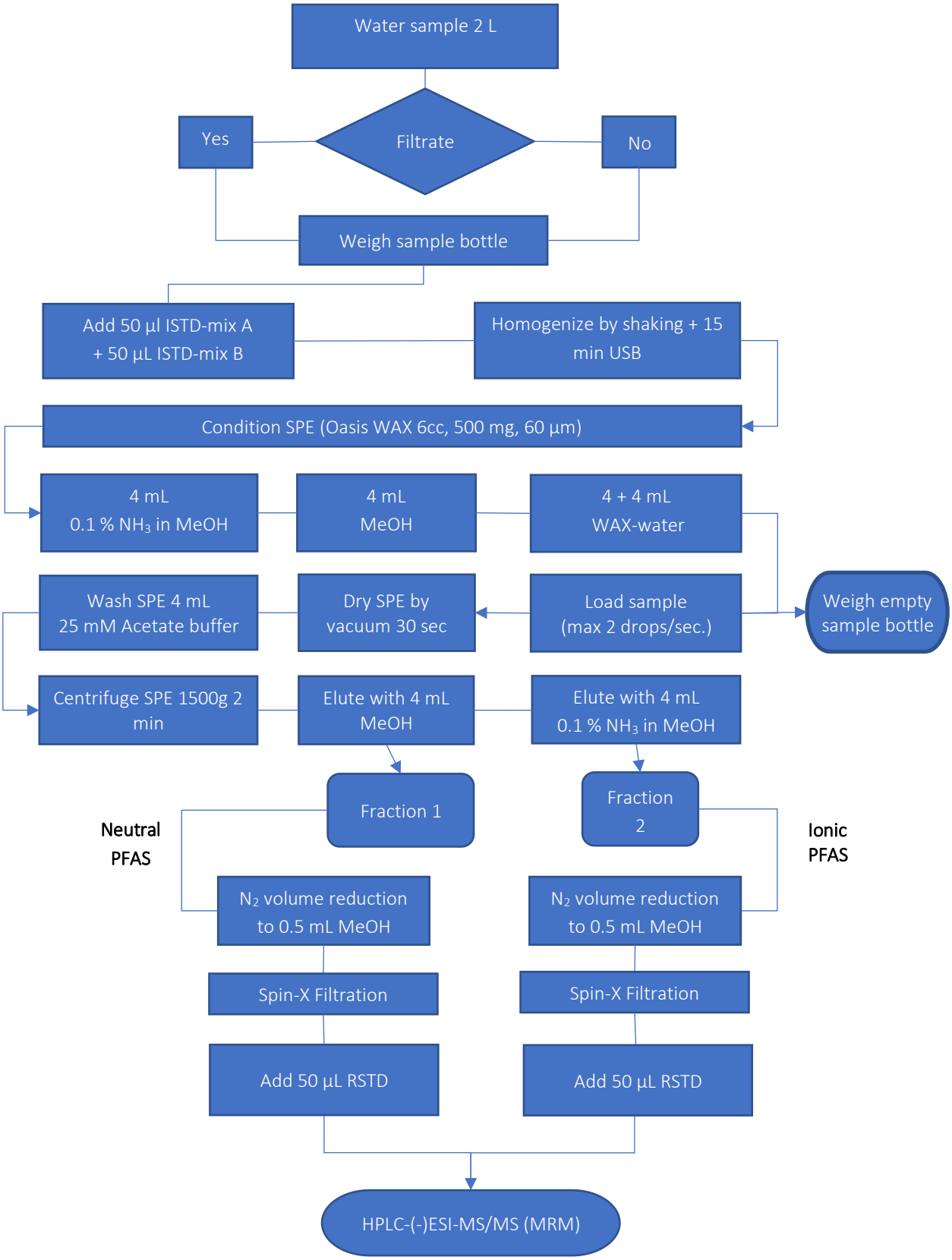

Figure S2: Sample preparation scheme for the quantitative analysis of PFASs in aqueous samples (fresh water and Sea water).

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 and industrial dishwasher machine the program included rinsing with MilliQ water. After the dishwasher, a manually cleaning followed by rinsed 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 precleaned 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

a. Determined by average field blank + 3*SD.
b. LOQ>MDL, LOQ was determined by average field blank + 10*SD.
c. 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 ^{13}C 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	PFUnDA	PFDoDA	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	PFUnDA	PFDoDA	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	PFUnDA	PFDoDA	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	PFUnDA	PFDoDA	PFBS	Sum-PFHxS	Sum PFOS	SUM PFAS
Soil	S-NA03	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	7.06	**7.06**
Soil	S-NA04	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	2.31	**2.31**
Soil	S-NA05	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	0.91	**0.91**
Soil	S-NA06	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	<0.002	**ND**
Soil	S-NA07	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	<0.002	**ND**
Soil	S-NA08	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	<0.002	**ND**
Soil	S-NA09	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<0.001	<0.001	<0.001	<0.002	**ND**
Soil	S-NA10	<0.04	<0.05	<0.06	<0.01	<0005	<0003	<0001	<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	02	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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