The fate of poly- and perfluoroalkyl substances in a marine food web influenced by land-based sources in the Norwegian Arctic

SUPPLEMENTARY INFORMATION

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Sample collection data

Table S1. Properties of investigated water samples

Location	Water samples (L)	Field		Laboratory	
		EC (µS/cm)	Temp. (^{0}C)	pH	Temp. (^{0}C)
Landfill	1.5	2550	5.20	7.60	13.20
LY-river	1.5	218	2.40	6.10	11.00
FFTS-pond	1.5	308	3.90	6.50	11.20
FFTS-creek	1.5	237	6.90	6.40	10.40
Ref-creek	1.5	62.4	-	5.80	19.90

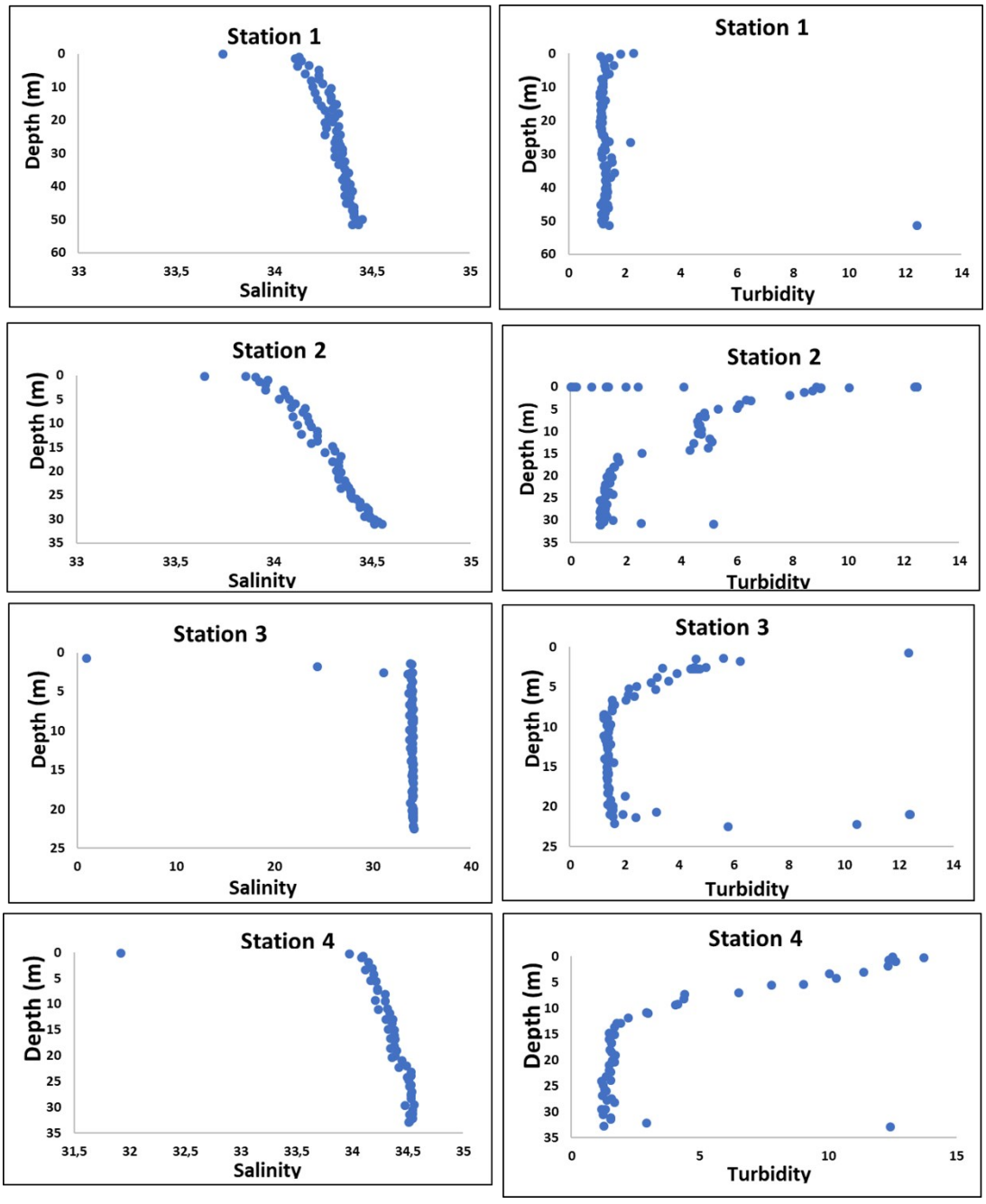


Figure S1. Turbidity and salinity depth profile at stations 1-4 in Adventfjorden.

Table S2. Weight of collected plankton and polychaete samples from different stations

Station	**Plankton (g)**	**Polychaete (g)**
St1	24.88	11.08
St2	25.59	10.84
St3	16.51	12.14
St4	36.01	9.00

Table S3. Biological parameters for collected crab individuals (Hyas araneus) from Adventfjorden/Isfjorden.

Station	Sample Name	Sex	Wdith (cm)	Length (cm)	Weight (g)
St1	Cr-1	M	6.5	8.6	123.89
	Cr-2	M	5.3	6.8	77.45
	Cr-3	M	5.1	7.2	77.45
	Cr-4	M	3.6	5.5	43.84
	Cr-5	M	3.6	5.3	38.09
	Cr-6	F	3.4	4.6	21.71
	Cr-7	M	4.4	6.3	54.88
	Cr-8	M	4.2	5.8	38.78
	Cr-9	M	5.5	7.4	116.01
	Cr-10	M	3.4	4.6	22.29
	Cr-11	M	2.7	4.0	13.27
	Cr-12	M	5.1	7.4	97.95
	Cr-13	M	5.1	7.4	93.72
	Cr-14	M	4.7	6.9	96.38
St2	Cr-1	M	6.1	7.6	108.51
	Cr-2	M	5.4	7.3	88.08
	Cr-3	F	4.4	5.4	32.19
	Cr-4	M	6.2	8.7	108.59
	Cr-5	M	3.3	4.5	19.83
	Cr-6	M	6.7	8.9	184.38
	Cr-7	M	6.3	8.4	118.62
	Cr-8	F	3.2	4.6	20.02
	Cr-9	M	3.5	4.9	25.56
	Cr-10	M	2.9	4.3	17.83
	Cr-11	M	6.1	8.0	157.00
	Cr-12	M	4.3	5.9	53.88
	Cr-13	M	5.7	7.9	107.90
	Cr-14	M	6.0	8.2	155.52
St3	Cr-1	-	-	-	-
St4	CR-1	F	5.6	6.8	59.48
	Cr-2	F	5.0	6.6	64.12
	Cr-3	F	4.6	5.8	46.13
	Cr-4	F	4.8	6.5	69.15
	Cr-5	M	6.4	8.2	150.49
	Cr-6	M	6.5	8.4	126.04
	Cr-7	M	4.9	6.4	76.93
	Cr-8	F	5.0	6.8	73.33
	CR-9	F	5.0	6.9	91.29

Table S4. Biological parameters of collected fish. SC: sculpin (Myoxocephalus scorpius). WF: wolffish (Anarhichas lupus).

Station	Sample Name	Weight (g)	Length (cm)	Liver weight (g)	Muscle sample (g)
St1	SC-1	126.6	21.6	2.97	11.77
	SC-2	61.79	18.1	1.49	6.95
	SC-3	529.88	30.9	32.61	21.1
	SC-4	263.76	25.7	15.07	14.18
	WF-1	856.1	45.8	26.1	30.76
St2	SC-1	279.3	27	11.79	20.58
	SC-2	243.9	25.9	17.18	16.73
	SC-3	358.22	27.5	22.02	17.3
	WF-1	2220	56.4	41.01	50.09
St3	SC-1	151.08	22.8	6.16	21.17
	SC-2	109.8	21	1.57	18.17
	SC-3	127.17	21.1	5.93	6.69
	SC-4	367.97	29.3	14.2	19.86
	SC-5	217.39	22.5	15.45	12.3
	SC-6	78.49	18.2	2.09	6.93
	SC-7	229.4	24.4	10.94	15.12
	SC-8	88.48	16.9	2.88	1213
	SC-9	73.22	18.1	1.34	9.14
	SC-10	69.92	17.4	1.89	5.54
	SC-11	102.77	19.1	7.78	9.37
St4	SC-1	75.35	18.2	2.13	14.81
	SC-2	131.72	21.18	3.32	19.45
	SC-3	62.52	17.8	0.56	8.76
	SC-4	87.93	20.3	1.71	11.46
	SC-5	232.83	26.4	5.39	24.28
	SC-6	89.69	19.4	2.75	11.99
	SC-7	113.88	21.3	3.23	7.19
	SC-8	92.3	19.1	2.66	7.48
	WF-1	780	43.5	13.56	62.93

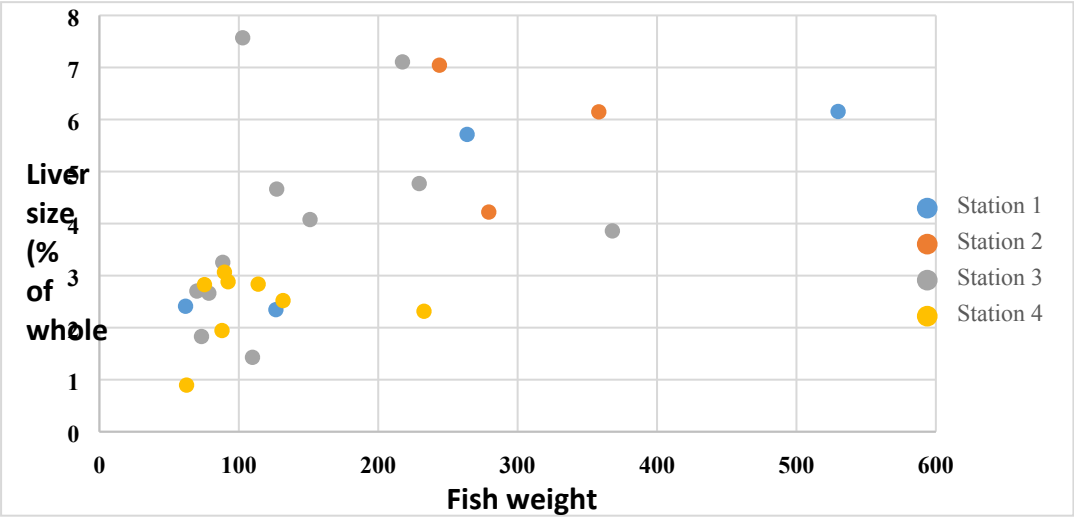


Figure S2. Sculpin (Myoxocephalus scorpius) relative liver size versus individual weight

Sample Code	Sex	Body mass (g)	Liver mass (g)	Gonad mass (g)	Heart mass (g)
G-1	male	1860	38.94	2.2	18.07
G-2	female	1340	30.71	1.25	12.07
G-3	female	1580	36.22	1.83	14.9
G-4	female	1610	37.7	1.81	11.6
G-5	female	1573	41.26	2.11	13.31
G-6	female	1855	39.45	1.79	14.97
G-7	female	1371	37.11	1.27	11.27
G-8	male	2196	40	3.11	15.91
G-9	male	2141	38.87	na	15.57
G-10	male	1947	44.17	2.78	16.84
G-11	female	1355	35.46	1.32	11.25
G-12	male	1943	40.19	5.19	14.38
G-13	female	1434	33.01	0.49	11.8
G-14	female	1363	26.1	1.62	12.61
G-15	male	2026	43.2	3.92	18.12
G-16	male	2210	43.9	3.41	17.12
G-17	female	1700	50.06	4.3	12.99
G-18	female	1640	36.7	1.98	13.91
G-19	male	2011	29.21	2.38	16.3
G-20	female	1407	45.29	0.61	11.92

Table S5. Biological parameters of collected glaucous gulls (Larus hyperboreus).

Detailed description of the chemical analysis

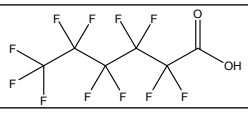
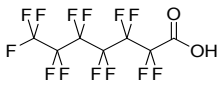
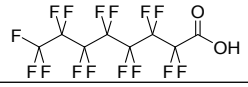
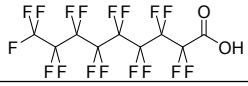
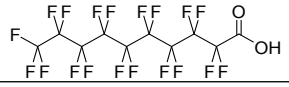
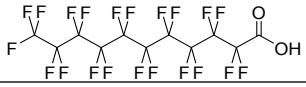
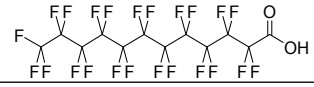
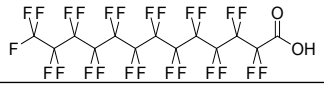
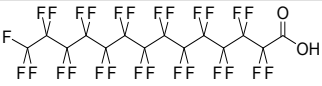
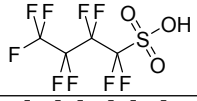
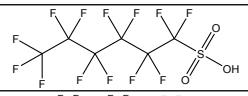
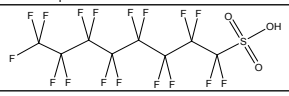
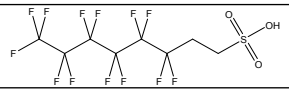
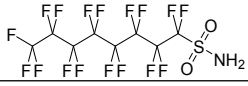
Standards and Chemicals

The following chemicals were applied for sample preparation and clean-up for subsequent LC/MS analysis: Glacial acetic acid (CH_3COOH, ACS reagent, ≥99.7%, Merck KGaA, Darmstadt, Germany), Q-guard 1 (MilliQ water purification, Billerica, MA, USA), Methanol (MeOH, HPLC grade, VWR International AS, Oslo, Norway), ammonium acetate ($NH_4CH_3CO_2$, 98.0%, Merck, Darmstadt, Germany), sodium hydroxide (NaOH, Sigma-Aldrich, ≥97.0%, St. Louis, MO), hydrochloric acid (HCl, 35%, Sigma-Aldrich, St. Louis, MO). All PFAS and their isotopically labeled internal standards (ISTDs) (Table S6) and analytical standards (all >98%) were produced by Wellington Laboratories (Guelph, Canada) and supplied by Greyhound Chromatography and Allied Chemicals (Merseyside, England).

1. Preparation of water samples

Water samples were prepared according to [1, 2] with some modifications. Unfiltered water samples (750 mL each, spiked 50µL of 500 ng mL^{-1} ISTDs mix) were extracted by solid-phase extraction (SPE) using mixed mode reverse phase/weak anion exchange (WAX); Oasis® WAX (500 mg, 6 cc, 60 µm, Waters, Milford MA, USA). The SPE cartridges were placed on a vacuum manifold and were conditioned by 4 mL of 0.1 % ammonia in methanol followed by 4 mL of methanol and 4 mL of Mill-Q water. SPE-cartridges were kept wet using additional 4 mL of Milli-Q water before loading the samples through a reservoir adapter and polypropylene (PP) tubing (o. d. 1/8”). Samples were loaded by the aid of a mild vacuum (water jet) and the flow rate was kept at 1-3 drop per second. The cartridges were cleaned with 4 mL acetate buffer of pH 4 and dried under vacuum for 15 min. Afterwards, the cartridges were eluted using 4 mL of methanol followed by 4 mL of 0.1% NH_3 in methanol. The eluates were collected in 15 mL polypropylene centrifuge tubes (VWR International, Radnor, PA, USA). The resulting eluates were dried at 37 °C under a gentle stream of N_2 (AGA, Oslo, Norway, N_2 5.0 quality) using a Reacti-Therm III evaporating unit (Thermo Fisher Scientific Inc., Rockford, USA). Twenty microliters of recovery standard (500 ng mL^{-1}) were added to each sample followed by addition of 450 µL MeOH. The residues were re-dissolved in 0.5 mL of methanol using vortex mixing (VWR mixer mini vortex 230V EU, VWR International, Radnor, PA, USA), filtered through a 0.2 µm nylon membrane microcentrifuge filter (Spin-X, Costar, Corning Inc., Corning, NY, USA), and transferred into PP vials for HPLC-MS/MS.

Table S6. The 14 poly- and perfluoroalkyl substances (PFAS) quantified in the Longyearbyen study

Analyte Name	Acronym	CAS #	Formula	Structure*	LogP**	LogD (7.4)**	Log K_{OC}
PFCA							
Perfluorohexanoic acid	PFHxA	307-24-4	$F(CF_2)_5COOH$		5.97	0.15	1.31[3]
Perfluoroheptanoic acid	PFHpA	375-85-9	$F(CF_2)_6COOH$		6.86	1.11	1.63[3], 2.1[4]
Perfluorooctanoic acid	PFOA	335-67-1	$F(CF_2)_7COOH$		7.75	1.82	1.89–3.5[5-7 3, 8]
Perfluorononanoic acid	PFNA	375-95-1	$F(CF_2)_8COOH$		8.64	2.84	2.36–4.0[3-5, 7]
Perfluorodecanoic acid	PFDA	335-76-2	$F(CF_2)_9COOH$		9.53	3.62	2.96–4.6[3-5, 7]
Perfluoroundecanoic acid	PFUnDA	2058-94-8	$F(CF_2)_{10}COOH$		10.42	4.23	3.3–5.1[3-5, 7]
Perfluorododecanoic acid	PFDoDA	307-55-1	$F(CF_2)_{11}COOH$		11.31	4.58	5.6 ± 0.2[4]
perfluorotetradecanoic acid	PFTriDA	72629-94-8	$F(CF_2)_{12}COOH$		12.19	4.97	-
Perfluorododecanoate	PFTeDA	376-06-7	$F(CF_2)_{13}COOH$		13.08	5.77	-
PFSA							
Perfluorobutane sulfonic acid	PFBS	375-73-5	$F(CF_2)_4SO_3H$		3.68	-1.56	1.22[8], 1.79[3]
Perfluorohexane sulfonic acid	PFHxS	355-46-4	$F(CF_2)_6SO_3H$		5.25	-0.54	2.05–3.7[3, 4, 7]
Perfluorooctane sulfonic acid	PFOS	1763-23-1	$F(CF_2)_8SO_3H$		7.03	0.66	2.6–3.8[3-8]
FTSA							
6:2 Fluorotelomer sulfonic acid	6:2 FTS	27619-97-2	$F(CF_2)_6(CH_2)_2SO_3H$		3.47	-1.00	-
preFOS							
Perfluorooctane sulfonamide	FOSA	754-91-6	$C_8H_2F_{17}NO_2S$				4.2–4.5[6, 7]

**All structures were prepared with ChemDraw Professional (version 15.0.0.106), PerkinElmer Informatics, Inc. (Boston, Massachusetts, USA) ** Predicted data is calculated with ACD/Labs Percepta Platform − PhysChem Module, Toronto, CA*

2. Preparation of snow sample

A snow sample was collected according to [1]. An aluminum shovel was precleaned, rinsing with MilliQ and then MeOH and transported wrapped in MeOH rinsed aluminum foil. An LDPE container was precleaned by washing with soapy water, rinsing several times with tap water, rinsed 5 times with

MilliQ and then rinsed 3 times with MeOH. Surface snow was then collected with the precleaned shovel and the sampling container sealed. The snow was then melted at 5°C and two 1.5 L duplicates were taken. This was then extracted unfiltered the same as the water samples in Section 1.

3. Preparation of biota and sediment samples

A previously published method was adopted with some modification[9]. About 0.5-2.5 g of homogenized biological material was weighed in a 15 ml Falcon tube (VWR International. LLC Radnor. USA). Fifty microliters of ^{13}C-labeled PFAS internal standards mix (Wellington laboratories) mixture solution (500 ng mL^{-1}) was added. Ten milliliters of MeOH were added to each sample and the sample was further homogenized with a sharp spatula. Samples were then sonicated at room temperature (20-22 °C) for ten minutes. Subsequently, the samples were mechanically shaken using Stuart Reciprocating shaker (SSL2, Bibby Scientific Ltd., Staffordshire, UK) for 30 minutes. The shaking was followed by centrifugation 10 min at 3000 rpm. The supernatant was removed with a plastic pipette and transferred to a new plastic tube. This stage was repeated by adding 4 ml of MeOH in the second extraction. The volume of the combined supernatants is reduced to 4 mL (37 °C) under a gentle flow of nitrogen gas (N_2) (AGA, Oslo, Norway, N_2 5.0 quality) using a Reacti-Therm III evaporating unit (Thermo Fisher Scientific Inc., Rockford, USA). Regarding the sediment sample, air-dried 5 g sediment was extracted as biota with an additional step with addition of 2 mL of NaOH solution (200 mM) prior to the extraction and 200 µL HCl solution (2 M) after the extraction according to [10]. About 0.5g of active coal Envi-Carb (ENVI-Carb™ 122, Sigma-Aldrich, Oslo, Norway) was added to each sample extract, the extract was then mixed and centrifuged for 10 min. The supernatant was removed and transferred to a new plastic tube. This step was repeated by adding an aliquot of methanol (1 mL) to the remaining deposits, and the supernatant was added to the corresponding tube. The supernatant was dried at 37 °C under N_2-gas (AGA, Oslo, Norway, N_2 5.0 quality) using a Reacti-Therm III evaporating unit (Thermo Fisher Scientific Inc., Rockford, USA). Twenty microliters of recovery standard (500 ng mL^{-1}) were added to each sample followed by addition of 450 µL MeOH. The samples were then vortexed (VWR mixer mini vortex 230V EU, VWR International, Radnor, PA, USA), and subsequently filtered through a 0.2 µm microcentrifuge filter (Spin-X, Costar, Corning Inc., Corning, NY, USA). The resulting sample was finally transferred to polypropylene vials, and immediately analysed with HPLC–MS -QqQ.

4. HPLC–ESI–QqQ analysis

An Agilent 1200 series HPLC (Agilent Technologies, Waldbronn, Germany) coupled with an Agilent 6460 (Agilent Technologies, Santa Clara, CA, USA) triple quadrupole mass spectrometer (MS-QqQ) was used for quantification of PFASs. A Zorbax Eclipse plus C18 RRHD (2.1 x 100 mm, 1.8 µm) (Agilent, Palo Alto, USA) column was used for separation and quantification of the target compounds at 25 ˚C. The injection volume was 10 µL. The analytical column was protected with a respective Guard Cartridge (4 µm x 3.0 mm ID). Separation was conducted by a binary mobile phase gradient consisting of (A) Aqueous solution of $NH_4CH_3CO_2$ (5 mM) and (B) pure MeOH with a mobile phase flow rate of 0.2 mL/min. The initial mobile phase proportion was 15 % (B) which was held for 5 min. B was then linearly increased to 99 % over 5 min and held for 7 min. B was then linearly changed to 1% until the end of the quantitative analysis (total run = 26 min.). Analytes were ionized in an Agilent Jet Stream electrospray ion source in negative ionization mode. The ion source parameters were as follows; gas temperature was 300 °C and gas flow was 5 l/min, nebulizer pressure was 25 psi, sheath gas temperature was 400 °C, sheath gas flow was 8 l/min.

The produced ions were monitored in negative dynamic multiple reaction monitoring (dMRM). Table S2 contains information on the ion transitions monitored and their individual settings. Agilent MassHunter software (Version B.07.00 /Build 7.0.457.0, 2008) was used for instrument control, method validation and quantification. Combined chromatograms of the MRM transition for the product ions for each analyte are shown in Figure S3.

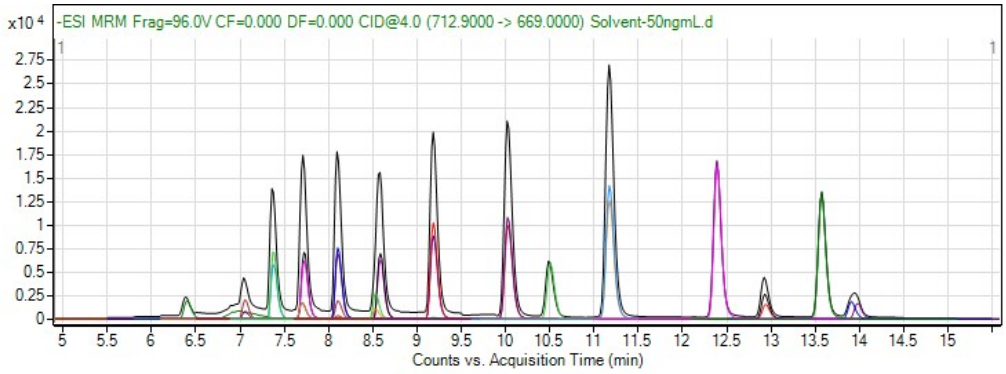


Figure S3. Extracted and Overlaid MRM of all compounds at 50 pg/µL

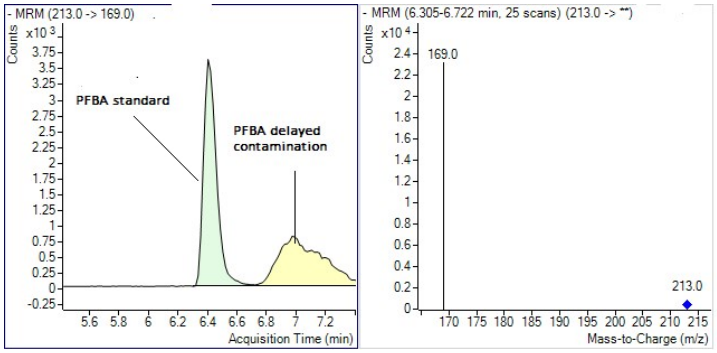


Figure S4. Extracted MRM of PFBA at 50 pg/µL

Table S7. Monitored ion transitions (dMRM) of target PFASs and their individual instrument settings; Retention time (RT) Precursor ion (Prec Ion), product ion (PI), Collision Energy (CE), Fragmentor voltage (FV), Retention time Window (ΔRT), and Polarity. For Acronyms and structure information on the target compounds, see table S6. ISTD used: internal standard used.

Acronym	ISTD	RT (min)	ΔRT	Prec Ion (m/z)	PI 1 (m/z)	PI 2 (m/z)	PI 3 (m/z)	CE1 (V)	CE2 (V)	CE3	Fragmentor (V)
PFBA	$[^{13}C_5]$-PFHxA	6.4	4	213	169			1			61
PFHxA	$[^{13}C_5]$-PFHxA	7.6	4	313	269	119		0	19		66
PFHpA	$[^{13}C_4]$-PFHpA	7.9	4	363	319	168.9		0	10		71
PFOA	$[^{13}C_4]$-PFOA	8.3	4	413	369	169		0	12		76
PFNA	$[^{13}C_5]$-PFNA	8.7	4	463	419	219		4	4		86
PFDA	$[^{13}C_2]$-PFDA	9.2	4	513	469	169		4	4		86
PFUnDA	$[^{13}C_2]$-PFUnDA	9.9	4	563	519	169		4	10		86
PFDoDA	$[^{13}C_2]$-PFDoDA	10.6	4	613	569			4			96
PFBS	$[^{18}O_2]$-PFHxS	7.2	4	299	99	80		25	35		121
PFHxS	$[^{18}O_2]$-PFHxS	7.9	4	398.9	99	80		45	45		151
Br-PFOS	$[^{18}O_2]$-PFHxS	8.3	4	499	99	80	130	61	61	61	200
L-PFOS	$[^{13}C_4]$-PFOS	8.4	4	499	99	80	130	61	61	61	200
6:2 FTS	$[^{18}O_2]$-PFHxS	8.1	4	427	407	81		15	15		145
FOSA	$[^{2}H_3]$-MeFOSA	10.5	4	497.9	78			33			141

Table S8. Monitored ion transitions (dMRM) of internal standards used and their individual instrument settings; Retention time (RT) Precursor ion (Prec Ion), product ion (PI), Collision Energy (CE), Fragmentor voltage (FV), Retention time Window (ΔRT), and Polarity.

Acronym	RT (min)	ΔRT	Prec Ion (m/z)	PI (m/z)	CE (V)	Fragmentor
[$^{13}C_8$]-PFOA	8.34	4	421	376	0	76
[$^{13}C_4$]-PFBA	6.58	4	217	172	1	61
[$^{13}C_5$]-PFHxA	7.66	4	318	273	0	66
[$^{13}C_4$]-PFHpA	7.99	4	367	322	0	66
[$^{13}C_4$]-PFOA	8.35	4	417	372.1	0	76
[$^{13}C_5$]-PFNA	8.79	4	468	423	4	76
[$^{13}C_2$]-PFDA	9.34	4	515	470	4	86
[$^{13}C_2$]-PFUnDA	9.94	4	565	520	4	96
[$^{13}C_2$]-PFDoDA	10.86	4	615	570	4	96
[$^{18}O_2$]-PFHxS	7.97	4	403	84	49	146
[$^{13}C_4$]-PFOS	8.74	4	503	80	61	180
[$^{2}H_3$]-MeFOSA	9.34	4	515	169	25	136

Validation of the Analytical Method

1. Recovery and detection limits

The apparent recoveries for all PFAS and their ISTDs from spiked samples were calculated using the following equation:

$$Apparent\ Recovery\% = 100 * \frac{C\ (sample) - C\ (matrix\ blank)}{C\ (spiked)}$$

Where C (sample) is the calculated concentration in the spiked sample, C (matrix blank) is the calculated background concentration in the matrix used, and C(spiked) is the concentration spiked into the sample.

Four and six replicates of each sample type (water, sediment, fish muscle, fish liver, plankton, worms, crab, and gull liver) were prepared and spiked with a mixture of all target PFASs and their ISTDs, while matrix blanks were spiked with only ISTDs. Six seawater samples were spiked at final concentration of 25 ng/L and four seawater samples were spiked at final concentration of 3 ng/L. For sediment and biota, 4 samples were spiked at 1 ng/g and six samples were spiked at 25 ng/g. All spiked samples with their blanks were then extracted and treated as real sample. The obtained recoveries are presented in Table S11. In general, most PFAS showed satisfying recoveries (42 – 120%) in the initial method validation as shown in Table S11. For N-MeFOSE, N-MeFOSA, N-EtFOSE, and N-EtFOSA, however, unacceptable recovery rates from some matrices were achieved, and were consequently excluded from further quantitative analysis from these matrices (red highlighted in Table S11). Sample specific recovery rates for all internal standards (ISTD) from different samples were calculated applying known concentrations of [$^{13}C_8$]-PFOA as a recovery standard (Table S12). Instrumental limit of detection (LOD), instrumental lower limit of quantification (LOQ) , and method detection limit (MDL) in abiotic and biotic samples, values are summarised in Table S9.

2. Matrix effect

Matrix effects (ME) caused by some co-eluting components from the residual matrix, are common in HPLC-ESI-QqQ methods [11]. ME influences the ionization efficiency of the target analytes. Typically, this results in suppression or enhancement of target compound signal [12]. In general, MEs are not reproducible among various sample sets or even replicates of the same set of samples and, therefore, compromise the quantitative analysis if not appropriately assessed [13], [14]. Therefore, here we evaluated MEs as we have investigated complex environmental samples. ME was estimated applying the following equation, where Sm and Ss represent the slopes of the matrix matched and solvent matched calibration curves respectively.

$$ME\% = \left[\left(\frac{Sm}{Ss}\right) - 1\right] \times 100$$

The results are summarized n Table S10. ME% values outside the range −20% to +20% indicate significant effects. Therefore, in sediment and biota samples, all PFASs experienced significant effect. We found that the use of ISTDs was not compensated for these effects, therefore, calibration standards including both native and ISTDs were prepared in similar matrix extracts using real samples collected from the reference in order to confirm the linear range of the method for reliable quantification.

3. Blank samples

Milli-Q water and clean sodium sulfate were used in the field during abiotic and biotic sample collection in order to monitor for possible contamination during sample collection, transport, and storage and analyzed as field blanks. Certain PFAS; PFBA, PFOA, and 6:2 FTS were detected in some of these blank samples at average concentration of 0.12, 0.010, and 0.034 ng/mL, respectively.

Table S9. Instrument limit of detection (ILOD), instrument limit of quantification (ILOQ), and method detection limit (MDL). For abbreviations and structure information on the target compounds see table S6

Compound	ILOD (ng/mL)	ILOQ (ng/mL)	MDL-Water (ng/L)	MDL-Fish liver (ng/g)	MDL-Fish Muscle (ng/g)	MDL-Sediment (ng/g)	MDL-Crab (ng/g)	MDL-Plankton (ng/g)	MDL-Worms (ng/g)	MDL-Gull liver (ng/g)
PFBA	0.007	0.022	0.150	0.094	0.02	0.320	0.80	0.600	5.3	0.08
PFHxA	0.012	0.040	0.270	0.060	0.030	0.003	0.003	0.003	0.040	0.045
PFHpA	0.015	0.051	0.340	0.015	0.020	0.002	0.003	0.003	0.011	0.0033
PFOA	0.004	0.015	0.180	0.020	0.010	0.045	0.012	0.003	0.017	0.010
PFNA	0.010	0.033	0.070	0.040	0.050	0.008	0.010	0.003	0.015	0.013
PFDA	0.004	0.014	0.080	0.030	0.090	0.040	0.003	0.002	0.060	0.040
PFUnDA	0.010	0.030	0.220	0.030	0.020	0.020	0.020	0.003	0.060	0.010
PFDoDA	0.050	0.160	0.110	0.070	0.050	0.030	0.060	0.002	0.018	2.24
PFTrDA	0.040	0.150	0.100	0.030	0.010	0.030	0.040	0.005	0.015	0.17
PFTeDA	0.040	0.140	0.090	0.090	0.070	0.030	0.040	0.009	0.014	0.80
PFBS	0.050	0.160	0.110	0.016	0.050	0.018	0.020	0.005	0.016	0.27
PFHxS	0.021	0.070	0.790	0.016	0.010	0.011	0.101	0.090	0.010	0.20
PFOS	0.060	0.190	0.240	0.050	0.040	0.014	0.063	0.030	0.010	0.30
6:2 FTS	0.040	0.010	1.08	0.015	0.060	0.030	0.050	0.410	0.05	0.38
FOSA	0.010	0.030	0.070	0.003	0.010	0.010	0.150	4.55	0.04	0.06
N-MeFOSE	0.070	0.220	0.150	0.079	0.010	2.00	0.070	1.30	0.22	0.51
N-MeFOSA	0.010	0.040	0.030	0.047	0.010	0.08	0.170	6.25	0.04	0.51
N-EtFOSE	0.010	0.040	0.030	0.015	0.014	0.030	1.11	1.54	0.04	2.50
N-EtFOSA	0.040	0.130	0.090	0.016	0.016	0.010	0.270	8.82	0.13	0.17

Table S10. Matrix effect (ME %); negative values indicate ion suppression and positive values indicate signal enhancement. For abbreviations and structure information on the target compounds

Compound	Water	Sediment	Crab	Plankton	Gull liver	Fish Liver	Fish Muscle
PFBA	-95.2	-80.0	-98.6	-89.9	-96.6	-99.0	-79.1
PFHxA	-86.2	-74.5	-96.5	-63.1	-89.1	-99.4	-76.9
PFHpA	-70.1	-68.1	-96.6	-50.5	-84.3	-98.6	-77.2
PFOA	-56.4	-66.6	-92.7	-51.5	-92.3	-98	-77.2
PFNA	-40.0	-64.9	-90.4	-34.5	-76.0	-96.9	-49.9
PFDA	-21.2	-68.3	-86.3	-38.8	-79.3	-97.4	-48.7
PFUnDA	1.80	-67.1	-80.3	-40.9	-73.0	-95.3	-41.8
PFDoDA	-12.1	-64.4	-71.3	-65.4	-73.0	-91.8	-29.3
PFTrDA	-16.1	-65.9	-77.0	-39.0	-89.4	-96.1	-40.8
PFTeDA	-16.1	-68.5	-97.4	-67.4	-94.0	-95.3	-40.2
PFBS	-61.5	-77.3	-93.5	-69.6	-89.1	-98.0	-67.2
PFHxS	-7.90	-70.0	-92.0	-55.3	-84.8	-97.9	-62.9
PFOS	-23.8	-70.8	-87.8	-55.4	-79.2	-97.8	-51.3
6:2 FTS	126.5	-57.7	-45.8	126.1	-21.1	-83.8	166.8
FOSA	-28.6	-68.9	-81.8	-67.8	-88.6	-99.5	-53.2
N-MeFOSE	-27.9	-69.2	-95.6	-96.0	-96.7	-99.8	-71.1
N-MeFOSA	-28.2	-70.1	-93.6	-88.6	-96.9	-98.3	-67.5
N-EtFOSE	-27.6	-78.5	-99.4	-96.3	-99.2	-99.9	-94.3
N-EtFOSA	-24.2	-74.8	-97.1	-92.5	-98.2	-98.7	-88.3

see table S6

Table S11. Recovery rates percent (mean± relative standard deviation) for all target compounds determined by repeated spiking of different type of samples. For abbreviations and structure information on the target compounds, see table S6 (colored cells indicate compounds with low recoveries, that were not quantified). Accepted recovery % :40-125%. Recoveries of 6:2 FTS were determined using matrix matched calibration curves.

Compound	Seawater		Fish muscle		Fish liver		Sediment		Crab		Plankton		Worm	Gull	
	3 ng/L	50 ng/L	1 ng/g	25 ng/g	1 ng/g	25 ng/g	1 ng/g	25 ng/g	1 ng/g	25 ng/g	1 ng/g	25 ng/g	25 ng/g	1 ng/g	25 ng/g
	(n=4)	(n=6)	(n=4)	(n=6)	(n=4)	(n=6)	(n=4)	(n=6)	(n=4)	(n=6)	(n=4)	(n=6)	(n=3)	(n=4)	(n=6)
PFBA	54±42	95±14	80±15	107.2±6.7	76±7	98±11	-	68±11	82±17	57±31	138±26	106±25	111±13	113.8±3.5	110.6±1.5
PFHxA	104±5	116±1	81.9±9.9	111.2±5.9	81.9±9.9	111.2±5.9	119.0±8.3	113.5±2.6	124.9±7.6	121.3±9.1	110.3±1.0	113.6±4.3	108.1±3.3	113.8±3.5	110.6±1.5
PFHpA	108±6	116±1	92.4±12.9	95.1±9.2	92.4±12.9	95.1±9.2	110.6±9.6	109.2±3.4	101.4±	108.5±5.9	101.5±6.6	101.2±4.0	97.0±3.9	104.0±3.7	100.4±4.9
PFOA	111±3	118±2	106.8±5.1	113.0±5.4	106.8±5.1	113.0±5.4	96.4±10.7	122.3±5.8	98.8±5.9	123.5±5.7	115.4±4.2	116.1±2.1	114.0±2.9	119.8±2.5	114.2±5.3
PFNA	109±2	117±1	114.3±9.4	107.3±2.8	114.3±9.4	107.3±2.8	89.4±12.8	114.8±4.2	98.9±10.9	125.6±11.3	107.0±5.7	104.8±1.6	106.4±3.2	111.3±2.5	116±4
PFDA	109±2	116±1	114.8±8.1	106.5±3.8	114.8±8.1	106.5±3.8	117.7±4.6	112.6±2.4	116.4±5.4	113.8±10.3	103.1±1.5	109.4±3.9	106.3±3.1	110.5±3.1	108±4
PFUnDA	106±2	119±2	126.0±5.3	115.3±1.6	126.0±5.3	115.3±1.6	126.5±3.9	121.0±2.4	131.7±0.5	122.4±6.0	115.2±0.40	120±4	118.6±2.5	129.3±3.3	118±4
PFDoDA	101±2	130±1	118.3±6.8	102.2±16.4	118.3±6.8	102.2±16.4	118.4±4.0	116.0±3.1	120.3±5.1	111.3±12.7	108.2±1.2	113.3±4.3	110.32.0	114.7±3.2	128.9±2.7
PFTrDA	72±4	117±4	114.7±6.0	100.3±6.7	114.7±6.0	100.3±6.7	128.8±6.5	117.9±12.7	69.0±12.2	94.3±21.7	93.7±3.7	96.8±4.1	102.1±4.2	52.0±18.5	107±4
PFTeDA	50±4	75±4	86.1±5.7	80.2±6.5	86.1±5.7	80.2±6.5	192.2±8.6	109.8±27.6	84.1±4.0	82.3±8.9	40.2±63.	110±10	89.3±15.0	47.3±37	95±16
PFBS	115±6	117±1	59.9±6.9	51.0±14.4	59.9±6.9	51.0±14.4	72.6±5.3	89.0±5.2	121.1±42.4	116.9±9.2	76.8±6.3	75.7±6.1	124.2±16.9	68.4±5.1	75.1±1.8
PFHxS	111±2	120.0±0.8	98.8±9.8	94.8±5.1	98.8±9.8	94.8±5.1	85.5±7.4	112.8±3.1	114.1±9.4	125.5±5.9	102.8±3.2	101.7±1.7	99.4±2.2	102.±2.3	73.4±2.8
L-PFOS	112±3	118±2	120.8±9.7	104.0±1.6	120.8±9.7	104.0±1.6	92.0±10.9	125.4±2.6	113.5±9.6	125.2±5.8	119.7±8.0	113.5±2.1	110.5±4.8	-	121±16
6:2 FTS	94±6	119±3	50±34	88.7±7.1	107±2	108±11	107±10	103±16	88±11	89±7.0	125±10	127±2.0	124±2.0	109±9.0	109±6.0
FOSA	53±51	92±7	51.3±12.7	52.4±4.9	51.3±12.7	52.4±4.9	104.7±25.1	79±26	115.8±11.9	111.5±22.4	52±14	57±6	79.7±9.9	62.7±3.7	72.8±5.0
N-MeFOSE	7±55	6±32	43±45	43±33	34±29	93±29	84±18	67±28	37±7	32±21	33±12	23±46	42±22	84±37	92±14
N-MeFOSA	-	0.3±17	63±25	18±61	64±12	68±10	12±26	13±34	38±6	28±32	26±14	13±29	53±20	84±13	110±17
N-EtFOSE	11±52	9±21	50±13	103±67	105±20	88±13	108±14	44±63	25±4	8±96	7±194	2±27	4±6	95±12	74±11
N-EtFOSA	0.6±83	0.7±11	8±68	8±41	72±13	71±14	44±6	13±36	36±2	11±29	25±67	5±7	13±14	55±32	76±15

Table S12. Recovery rates percent (mean± relative standard deviation) for all internal standards determined by repeated spiking of different type of samples. For abbreviations and structure information on the target compounds, see table S6.

Compound	Seawater (n=8)	Fish muscle (n=8)	Fish liver (n=8)	Glaucous gull (n=8)	Worms (n=3)	Plankton (n=8)	Sediment (n=5)	Crab avg (n=8)
[$^{13}C_4$]-PFBA	11.6±20.6	48.1±23.7	9.6±131.8	18.7±44.6	2.6±173.2	9.0±31.1	20.5±62.6	4.3±97.0
[$^{13}C_5$]-PFHxA	13.0±15.5	48.7±19.6	14.7±13.7	43.7±13.3	9.8±1.8	24.2±9.3	33.8±12.5	17.4±14.1
[$^{13}C_4$]-PFHpA	78.3±2.2	87.4±14.5	78.9±13.4	96.2±15.5	46.6±24.7	51.7±8.9	62.6±24.0	62.2±13.1
[$^{13}C_4$]-PFOA	42.6±9.3	49.6±8.9	28.1±13.1	31.7±12.1	30.5±27.7	35.7±5.9	34.3±39.6	51.2±6.8
[$^{13}C_5$]-PFNA	61.6±5.0	91.6±13.3	55.8±17.8	86.9±14.3	69.1±11.8	52.3±7.9	35.0±36.5	0.0±68.3
[$^{13}C_2$]-PFDA	65.1±4.9	101.1±9.3	60.1±10.6	82.1±14.0	71.4±9.2	45.7±10.2	36.3±26.8	76.0±12.9
[$^{13}C_2$]-PFUnDA	79.0±11.0	112.2±14.0	85.6±20.8	99.8±14.5	71.9±14.0	43.4±9.7	38.5±19.8	119.4±8.9
[$^{13}C_2$]-PFDoDA	65.9±11.5	126.1±8.1	103.8±20.2	76.9±15.2	80.5±10.5	43.4±10.8	40.4±13.3	125.6±9.3
[$^{18}O_2$]-PFHxS	39.3±10.7	59.0±16.0	38.3±15.6	72.6±12.7	22.5±15.1	42.4±5.7	35.0±47.2	24.3±14.8
[$^{13}C_4$]-PFOS	104.3±11.7	115.6±14.8	93.8±17.1	118.8±19.8	101.7±18.2	74.7±15.3	75.5±26.0	109.0±8.4
[$^{2}H_3$]-MeFOSA	82.2±8.6	119.5±13.4	37.6±96.9	83.5±15.5	70.7±4.3	42.0±11.6	36.3±23.3	59.5±76.7

Sample analysis results

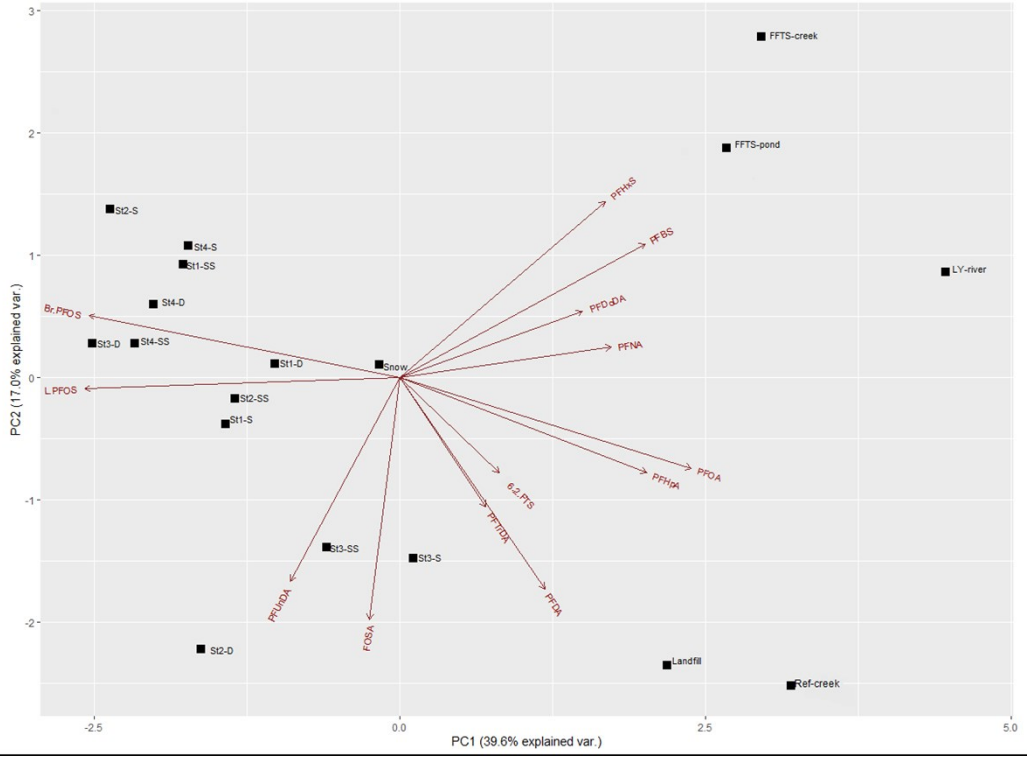


Figure S5. PCA biplots of PCA for PFAS profiles of the various samples (i.e. % of $\sum_{14}$PFAS) with PC1 and 2 explaining more than 56% of the variation.

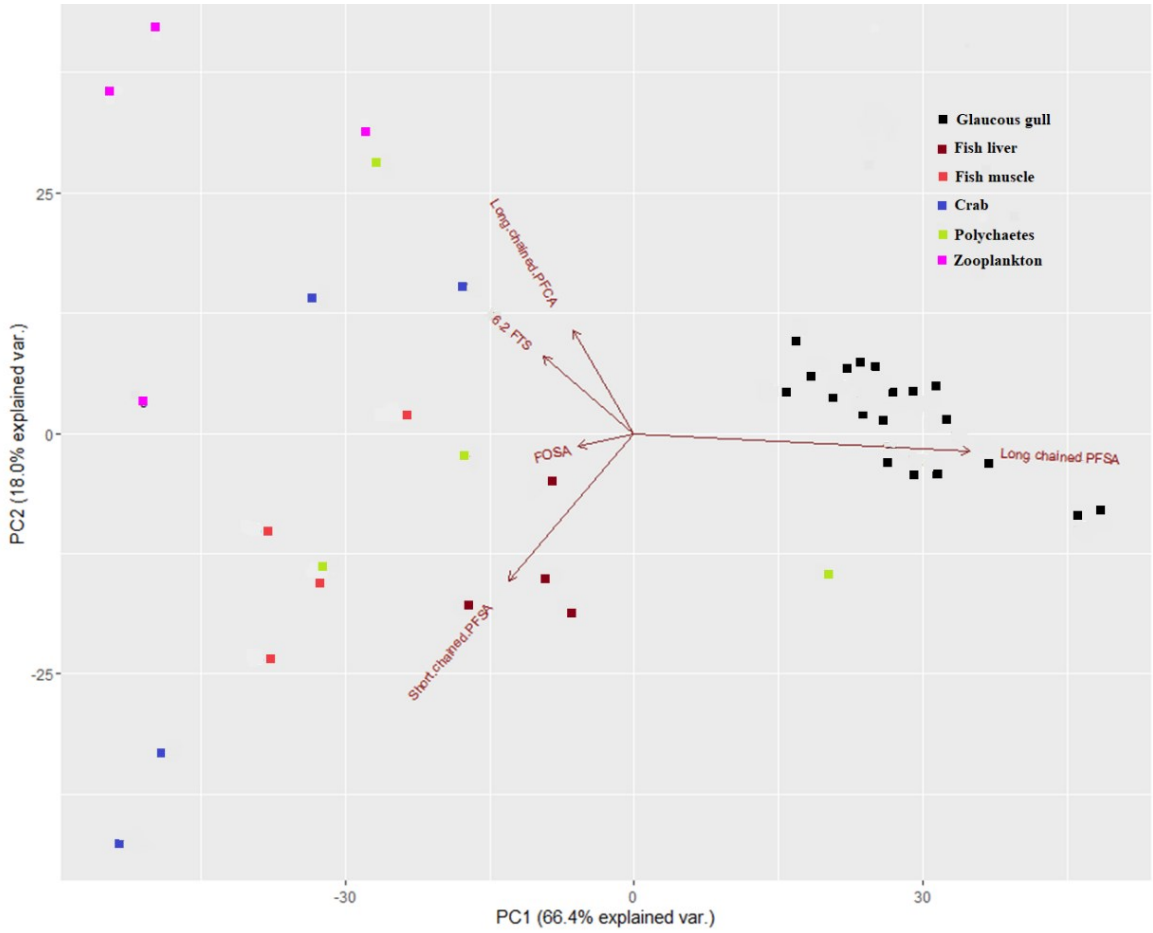


Figure S6. PCA biplots of PCA for PFAS profiles of the biota samples collected from the Adventfjord and the Isfjord, near the Svalbard Airport. (i.e. % of ∑14PFAS) with PC1 and 2 explaining more than 84% of the variation.s (Levels <LOQ were treated as zero in this figure). Short-chained PFCA: PFHxA and PFHpA; long-chained PFCA: C8-C14; Short-chained PFSA: PFBS and PFHxS; Long-chained PFSA: PFOS.

Sample analysis results

Table S13. PFAS concentrations in seawater and fresh water (ng L^{-1}± standard error of the mean) collected in the vicinity of Longyearbyen (Svalbard).

Sample	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFHxS	∑PFOS	6:2 FTS	FOSA	$\sum_{14}$PFAS
St1-surface(n=2)	nd	nd	0.19±0.03	(0.035)	nd	(0.11)	nd	nd	nd	nd	(0.38)	4.37±1.31	(0.54)	(0.03)	5.12±0.97
St1-subsurface (n=2)	nd	nd	0.14±0.09	(0.035)	nd	(0.11)	nd	nd	nd	(0.055)	(0.38)	6.41±2.09	(0.54)	nd	7.71±1.87
St1-deep (n=2)	(0.13)	(0.17)	0.19±0.14	(0.035)	nd	(0.11)	nd	nd	nd	(0.055)	0.61±0.23	5.16±3.92	(0.54)	(0.03)	6.98±2.87
St2-surface(n=2)	(0.13)	(0.17)	0.17±0.01	(0.035)	nd	(0.11)	nd	nd	nd	nd	(0.38)	9.16±3.23	nd	nd	10.06±1.17
St2-subsurface (n=2)	nd	(0.17)	0.28±0.04	(0.035)	(0.04)	(0.11)	nd	nd	nd	nd	0.61±0.24	6.51±1.65	nd	(0.03)	7.59±0.45
St2-deep (n=2)	(0.13)	(0.17)	0.17±0.01	(0.035)	(0.04)	0.35±0.24	nd	nd	nd	(0.055)	(0.38)	4.92± 0.52	nd	(0.03)	6.20±2.6
St3-surface (n=4)	0.32±0.19	(0.17)	0.80±0.54	(0.035)	(0.04)	(0.11)	nd	nd	nd	(0.055)	(0.38)	4.02±1.68	(0.54)	(0.03)	4.45±0.59
St3-subsurface(n=4)	(0.13)	(0.17)	0.22±0.02	(0.035)	(0.04)	(0.11)	nd	nd	nd	nd	(0.38)	4.37±3.51	(0.54)	(0.03)	6.11±0.63
St3-deep (n=2)	(0.13)	(0.17)	0.21±0.02	(0.035)	nd	0.25±0.09	nd	nd	nd	nd	(0.38)	8.61±1.45	nd	(0.03)	9.75±1.87
St4-surface (n=2)	nd	(0.17)	0.12±0.09	(0.035)	nd	(0.11)	nd	nd	nd	(0.055)	(0.38)	4.41±2.10	nd	nd	5.27±1.77
St4-subsurface (n=2)	(0.13)	nd	0.16±0.01	(0.035)	nd	(0.11)	nd	nd	nd	(0.055)	(0.38)	5.13±1.77	nd	(0.03)	5.97±1.19
St4-deep (n=2)	nd	nd	0.13±0.11	(0.035)	(0.04)	(0.11)	nd	nd	nd	(0.055)	(0.38)	7.90±4.02	nd	nd	8.64±2.94
Ref-creek (n=2)	0.38±0.01	0.93±0.03	1.00±0.01	0.83±0.11	(0.04)	(0.11)	nd	nd	nd	(0.055)	0.38±0.06	1.58±0.20	<LOQ	(0.03)	5.86±0.1
LY (n=2)	0.67±0.17	0.73±0.01	0.73±0.10	(0.035)	(0.04)	(0.11)	(0.55)	nd	nd	0.32±0.01	0.69±0.32	2.48±0.80	<LOQ	nd	7.054±2
Snow (n=2)	0.290±0.003	<LOQ	0.360±0.007	0.220±0.001	0.087±0.001	(0.11)	(0.55)	nd	nd	0.260±0.007	1.950±0.023	14.52±0.033	<LOQ	0.140±0.001	18.70±0.04
Landfill (n=2)	67.3±2.2	30.0±0.84	141.0±3.0	41.3±1.6	3.24±0.11	12.4±0.56	0.18±0.001	(0.05)	nd	5.92±0.19	44.9±0.81	289.3±29.15	<LOQ	3.28±0.07	643.6±84
FFTS-pond (n=2)	15.9±0.40	4.97±0.08	44.4±0.27	111.0±0.37	1.21±0.05	1.07±0.03	(0.55)	nd	nd	3.18±0.01	63.91±0.44	115.8±1.31	4.47±1.02	0.16±0.002	365.4±8.00
FFTS-creek (n=2)	10.4±0.43	3.63±0.13	6.73±0.04	0.78±0.02	(0.04)	(0.11)	(0.55)	nd	nd	1.90±0.02	13.19±0.74	19.09±0.87	1.46±0.08	(0.03)	57.4±4.0

Table S14. PFAS concentrations in sediment samples (μg kg^{-1}d.w. ± standard error of the mean) collected in the marine environment and a leachate channel of a landfill in the vicinity of Longyearbyen (Svalbard)

PFAS	Station 1		Station 2		Station 3		Station 4		Landfill	
	n	Mean±SE	n	Mean±SE	n	Mean±SE	n	Mean±SE	n	Mean±SE
PFHxA	3/3	0.0158±0.0010	2/3	0.0098±0.0051	0/3	nd	1/3	0.0029±0.0029	3/3	0.339±0.010
PFHpA	2/3	0.0098±0.0049	0/3	nd	2/3	0.0133±0.006	1/3	0.0058±0.0057	2/3	0.169±0.009
PFOA	3/3	0.0383±0.0023	3/3	0.0285±0.0004	3/3	0.036±0.0015	3/3	0.0274	3/3	1.203±0.028
PFNA	3/3	0.0678±0.0048	3/3	0.0417±0.0027	3/3	0.0340±0.0023	1/3	0.0087±0.0087	3/3	1.80±0.024
PFDA	3/3	0.0204±0.004	3/3	0.0176±0.0005	3/3	0.0237±0.003	2/3	0.0092±0.0045	3/3	0.860±0.017
PFUnDA	3/3	0.0445±0.0013	3/3	0.0354±0.0032	3/3	0.0292±0.0023	3/3	0.0250±0.0037	3/3	25.5±0.486
PFDoDA	3/3	0.0148±0.0005	3/3	0.0177±0.0011	3/3	0.0296±0.0036	3/3	0.0136±0.0010	3/3	0.693±0.016
PFTrDA	3/3	0.0208±0.0007	3/3	0.0226±0.0026	3/3	0.0337±07.004	3/3	0.0162±0.0021	3/3	4.22±0.111
PFTeDA	3/3	0.0145±0.0004	3/3	0.0178±0.0018	3/3	0.0356±0.0056	3/3	0.0129±0.0004	3/3	0.045±0.023
PFBS	3/3	0.0198±0.0046	2/3	0.0124±0.0075	0/3	nd	1/3	0.0069±0.0068	3/3	0.081±0.012
PFHxS	3/3	0.0398±0.0012	3/3	0.0446±0.0041	3/3	0.0245±0.0026	3/3	0.0229±0.0038	3/3	0.461±0.010
Br-PFOS	3/3	0.0441±0.0001	3/3	0.0341±0.0064	0/3	nd	0/3	nd	3/3	2.59±0.230
L-PFOS	3/3	0.2510±0.0323	3/3	0.1780±0.0007	3/3	0.0491±0.0033	1/3	0.0119±0.0119	3/3	42.8±1.77
6:2 FTSA	3/3	3.99±3.89	0/3	nd	1/3	0.208±0.360	0/3	nd	0/3	nd
FOSA	3/3	0.0128±0.0001	3/3	0.0129±0.0002	3/3	0.0135±0.0001	0/3	nd	3/3	0.782±0.010
∑14PFAS		4.61±3.92		0.474±0.013		0.530±0.181		0.1634±0.0279		81.6±2.13

Table S15. Sediment/water partition coefficients (log K_d in $dm^3\ kg^{-1}$). derived from sediment and water samples taken at the same station. assuming local equilibrium.

PFAS	Landfill sediment (n=3)	Marine sediment (n=12)
PFHxA	0.68±0.03	-
PFHpA	0.85±0.03	-
PFOA	0.94±0.001	-
PFNA	1.6±0.05	-
PFDA	2.4±0.74	-
PFUnDA	3.3±1.4	2.4±1.7
PFBS	0.52±0.32	-
PFHxS	1.0±0.01	-
PFOS	2.0±0.60	1.4±1.2
FOSA	2.7±1.5	-

Table S16. PFAS levels in zooplankton samples from the stations St1-4 in Adventfjorden (µg kg^{-1} ww± standard error of the mean).

PFAS	Station 1		Station 2		Station 3		Station 4	
	n	**Mean±SE**	**n**	**Mean±SE**	**n**	**Mean±SE**	**n**	**Mean±SE**
PFHxA	0/3	nd	0/3	nd	0/3	nd	0/3	nd
PFHpA	0/3	nd	0/3	nd	0/3	nd	0/3	nd
PFOA	3/3	0.0292±0.0039	3/3	0.0270±0.0016	3/3	0.0274±0.0027	3/3	0.0238±0.0050
PFNA	1/3	0.0067±0.0066	1/3	0.0071±0.0071	2/3	0.0155±0.0077	0/3	nd
PFDA	2/3	0.0133±0.0066	3/3	0.0219±0.007	3/3	0.0201±0.0014	3/3	0.0198±0.0001
PFUnDA	3/3	0.0450±0.0045	3/3	0.0367±0.0016	3/3	0.0375±0.0024	3/3	0.0421±0.0008
PFDoDA	3/3	0.0204±0.0023	3/3	0.0181±0.005	3/3	0.0161±0.0001	3/3	0.0175±0.00001
PFTrDA	3/3	0.0274±0.0033	3/3	0.0226±0.0017	3/3	0.0202±0.0013	3/3	0.0269±0.0018
PFTeDA	0/3	nd	0/3	nd	0/3	nd	0/3	nd
PFBS	0/3	nd	1/3	0.0078±0.0078	2/3	0.7356±0.7088	1/3	0.0093±0.0093
PFHxS	0/3	nd	0/3	nd	2/3	0.0131±0.0131	0/3	nd
Br-PFOS	0/3	nd	0/3	nd	2/3	0.0093±0.0083	1/3	0.0156±0.0156
L-PFOS	0/3	nd	1/3	0.0046±0.0046	1/3	0.0146±0.0145	1/3	0.2270±0.02270
6:2 FTS	1/3	0.1895±0.328	3/3	1.87±0.832	3/3	0.6009±0.3392	2/3	0.1667±+.0833
FOSA	2/3	0.0107±0.0054	3/3	0.0146±0.0005	3/3	0.0150±0.0007	0/3	nd
$\sum_{14}$PFAS	-	0.3421±0.1886	-	2.03±0.82	-	1.525±1.397	-	0.5486±0.3044

Table S17. PFAS levels in polychaete samples from marine sediments stations St1-4 in Adventfjorden (µg kg^{-1} ww± standard error of the mean).

PFAS	Station 1		Station 2		Station 2		Station 2	
	n	Mean±SE	n	Mean±SE	n	Mean±SE	n	Mean±SE
PFHxA	0/3	nd	0/3	nd	0/3	nd	0/3	nd
PFHpA	0/3	nd	0/3	nd	1/3	0.1176±0.1176	0/3	nd
PFOA	0/3	nd	0/3	0.0665±0.0283	2/3	0.1602±0.0870	3/3	0.1430±0.0218
PFNA	2/3	0.0677±0.0340	2/3	0.0432±0.0176	2/3	0.0738±0.0368	1/3	0.0307±0.0307
PFDA	3/3	0.0401±0.0029	3/3	0.020±0.0079	2/3	0.0396±0.0200	3/3	0.0457±0.0086
PFUnDA	3/3	0.0745±0.005	3/3	0.0429±0.0178	3/3	0.0903±0.0136	3/3	0.0698±0.0096
PFDoDA	2/3	0.0195±0.0098	2/3	0.0231±0.0100	3/3	0.0420±0.0063	3/3	0.040±0.004
PFTrDA	3/3	0.1750±0.0430	3/3	0.1230±0.0520	3/3	0.1190±0.0118	3/3	0.150±0.026
PFTeDA	3/3	0.0319±0.0052	3/3	0.0144±0.0053	3/3	0.0507±0.0134	3/3	0.049±0.009
PFBS	3/3	0.1543±0.0323	0/3	nd	1/3	0.0621±0.0621	1/3	0.075±0.075
PFHxS	3/3	0.917±0.1247	3/3	0.2056±0.0295	3/3	1.01±0.253	0/3	nd
Br-PFOS	1/3	0.2323±0.2323	1/3	0.0134±0.0133	0/3	nd	1/3	0.0534±0.0534
L-PFOS	3/3	4.54±1.04	2/3	0.2630±0.1315	1/3	2.08±2.08	1/3	0.414±0.414
6:2 FTS	3/3	0.5971±0.2592	0/3	0.0675±0.0675	3/3	0.581±0.378	3/3	nd
FOSA	3/3	0.1922±0.0636	3/3	0.0259±0.0259	3/3	0.1110±0.0128	3/3	0.047
$\sum_{14}$PFAS	-	7.043	-	0.909±0.1394	-	4.54±2.55	-	1.12±0.053

Table S18. Concentrations of PFAS (µg kg^{-1} ww) in crab samples collected from four stations in the vicinity of Longyearbyen (Svalbard).

PFAS	Station 1 (n=5)			Station2 (n=5)			Station 3 (n=1)	Station 4 (n=7)		
	n	Mean±SEM	Median	n	Mean±SEM	Median	St3_Crab	n	Mean±SEM	Median
PFHxA	0/5	nd	nd	0/5	nd	nd	nd	0/7	nd	nd
PFHpA	0/5	nd	nd	0/5	nd	nd	nd	0/7	nd	nd
PFOA	5/5	0.018	0.018	5/5	0.026±0.007	0.018	0.018	7/7	0.040±0.009	0.037
PFNA	3/5	0.033±0.013	0.049	3/5	0.027±0.012	0.023	nd	4/7	0.043±0.019	0.039
PFDA	5/5	0.055±0.007	0.051	5/5	0.030±0.006	0.030	0.033	7/7	0.043±0.007	0.049
PFUnDA	5/5	0.218±0.052	0.204	5/5	0.120±0.026	0.108	0.116	7/7	0.133±0.025	0.146
PFTrDA	5/5	0.255±0.195	0.180	5/5	0.144±0.024	0.133	0.121	7/7	0.122±0.023	0.132
PFTeDA	5/5	0.062±0.022	0.051	5/5	0.035±0.002	0.035	0.030	7/7	0.041±0.006	0.044
PFBS	1/5	0.309±0.691	nd	1/5	4.50±1.87	6.16	1.92	1/7	nd	nd
PFHxS	0/5	nd	nd	0/5	0.144±0.144	nd	nd	0/7	nd	nd
Br-PFOS	2/5	0.033±0.029	nd	2/5	nd	nd	0.034	3/7	0.013±0.008	nd
L-PFOS	4/5	0.276±0.115	0.215	4/5	0.191±0.012	0.195	0.158	7/7	0.312±0.063	0.350
6:2 FTS	2/5	0.66±0.43	nd	2/5	(0.08)	nd	(0.08)	3/7	(0.08)	nd
FOSA	5/5	0.248±0.044	0.210	5/5	0.269±0.022	0.280	0.314	7/7	0.256±0.049	0.235
$\sum_{14}$PFAS	-	1.56±0.491	1.054	-	5.629±1.95	6.943	3.02	-	1.02±0.152	0.956

Table S19. Concentrations of PFAS (µg kg^{-1}; w/w) in muscle of fish collected in the marine in the marine environment in the vicinity of Longyearbyen (Svalbard)

PFAS*	Station 1			Station 2			Station 3			Station 4		
	n	Mean±SEM	median	n	Mean±SEM	median	n	Mean±SEM	median	n	Mean±SEM	median
PFOA	5/5	0.021±0.006	0.016	3/3	0.037±0.005	0.034	11/11	0.027±0.003	0.027	8/8	0.021±0.002	0.021
PFNA	5/5	0.046±0.008	0.047	3/3	0.055±0.002	0.055	11/11	0.049±0.005	0.047	8/8	0.037±0.006	0.040
PFDA	5/5	0.028±0.003	0.030	3/3	0.028±0.001	0.028	11/11	0.028±0.002	0.029	8/8	0.021±0.002	0.020
PFUnDA	5/5	0.085±0.025	0.087	3/3	0.056±0.005	0.054	11/11	0.062±0.007	0.066	8/8	0.043±0.008	0.033
PFDoDA	5/5	0.025±0.004	0.024	3/3	0.020±0.001	0.020	11/11	0.021±0.001	0.020	8/8	0.016±0.001	0.016
PFTrDA	5/5	0.063±0.021	0.048	3/3	0.033±0.003	0.035	11/11	0.040±0.003	0.042	8/8	0.029±0.003	0.029
PFTeDA	5/5	0.020±0.002	0.018	3/3	0.015±0.0001	0.016	11/11	0.019±0.001	0.018	8/8	0.015±0.001	0.016
PFHxS	5/5	0.48±0.17	0.360	3/3	0.010±0.006	0.099	11/11	0.215±0.033	0.204	7/8	0.0192±0.068	0.147
Br-PFOS	5/5	0.018±0.005	0.015	3/3	0.013±0.005	0.010	10/11	0.017±0.002	0.018	8/8	0.017±0.001	0.018
L-PFOS	5/5	0.149±0.044	0.123	3/3	0.140±0.027	0.160	8/11	0.071±0.018	0.066	8/8	0.087±0.017	0.075
FOSA	5/5	0.212±0.124	0.121	3/3	0.140±0.034	0.112	8/11	0.148±0.063	0.096	8/8	0.045±0.003	0.044
$\sum_{14}$PFAS	-	1.14±0.19	1.244	-	0.640±0.068	0.586	-	0.696±0.068	0.686	-	0.524±0.09	0.475

* PFHxA, PFHpA, 6:2 FTS, PFBS were not detected in any muscle sample.

Table S20. Concentrations of PFAS (µg kg^{-1}; w/w) in liver of fish collected in the marine in the marine environment in the vicinity of Longyearbyen (Svalbard)

PFAS*	Station 1			Station 2			Station 3			Station 4		
	n	Mean±SEM	median	n	Mean±SEM	median	n	Mean±SEM	median	n	Mean±SEM	median
PFHxA	0/5	nd	nd	0/7	nd	0	3/12	0.134±0.090	0.000	0/4	nd	nd
PFHpA	0/5	nd	nd	0/7	nd	0	0/12	nd	0.000	0/4	nd	nd
PFOA	2/5	0.247±0.216	0.000	3/7	0.06±0.030	0	4/12	0.025±0.013	0.000	3/4	0.136±0.053	0.153
PFNA	5/5	0.385±0.142	0.341	7/7	0.138±0.027	0.129667274	12/12	0.241±0.035	0.212	3/4	0.410±0.143	0.486
PFDA	4/5	0.223±0.129	0.128	6/7	0.049±0.010	0.05016601	12/12	0.116±0.014	0.126	3/4	0.174±0.085	0.177
PFUnDA	5/5	0.661±0.246	0.564	7/7	0.164±0.019	0.17319068	12/12	0.357±0.059	0.329	4/4	0.538±0.136	0.583
PFDoDA	5/5	0.179±0.072	0.161	7/7	0.040±0.006	0.037419633	12/12	0.069±0.008	0.068	4/4	0.150±0.032	0.157
PFTrDA	5/5	0.554±0.161	0.557	7/7	0.140±0.028	0.140520246	12/12	0.237±0.053	0.171	4/4	0.495±0.140	0.519
PFTeDA	5/5	0.118±0.062	0.058	6/7	0.022±0.006	0.022609377	11/12	0.0167±0.004	0.041	4/4	0.145±0.058	0.122
PFBS	0/5	nd	0.000	2/7	0.027±0.020	0	0/12	nd	0.000	1/4	0.066±0.065	0.000
PFHxS	4/5	3.02±1.85	1.298	6/7	0.672±0.250	0.367317029	10/12	1.693±0.397	1.587	2/4	0.445±0.280	0.310
Br-PFOS	4/5	0.127±0.054	0.122	6/7	0.103±0.021	0.115676717	12/12	0.154±0.028	0.112	2/4	0.157±0.092	0.137
L-PFOS	4/5	2.812±1.12	2.166	7/7	0.993±0.310	0.574313066	12/12	2.105±0.661	1.416	3/4	2.046±0.792	2.359
6:2 FTS	0/5	nd	0.000	0/7	nd	0	0/12	nd	0.000	nd	nd	0.000
FOSA	4/5	0.234±0.10	0.151	7/7	0.216±0.0739	0.138292607	12/12	0.255±0.089	0.145	4/4	0.447±0.213	0.298
$\sum_{14}$PFAS	-	8.56±3.94	4.318	-	2.624±0.327	2.29086767	-	5.467±0.845	5.300	-	5.2±1.5	6.460

Table S21. Bioaccumulation factors (BAF. L kg^{-1}) for PFAS on Svalbard

PFAS	Log $BAF_{fish\ muscle}$±SE	Log $BAF_{fish\ liver}$±SE
PFOA	2.09±0.103	2.87±0.210
PFNA	2.96±0.141	3.71±0.249
PFDA	3.19±0.161	3.78±0.357
PFUnDA	2.61±0.110	3.41±0.221
PFHxS	2.72±0.140	3.44±0.196
L-PFOS	1.52±0.148	2.61±0.143

Table S22. Concentrations of PFAS µg kg^{-1} (wet weight) detected in glaucous gull collected in Svalbard (n denotes number of samples detected of total number sampled).

PFAS	n	Mean±SEM	Range	Median
PFHxA	nd	nd	nd	nd
PFHpA	2/20	0.006±0.005	nd-0.093	nd
PFOA	13/20	0.101±0.025	nd-0.326	0.093
PFNA	20/20	3.16±0.375	0.796-6.55	3.038
PFDA	20/20	1.95±0.190	0.743-3.61	2.060
PFUnDA	20/20	4.38±0.556	1.40-11.9	4.484
PFDoDA	20/20	0.710±0.078	0.320-1.78	0.685
PFTrDA	20/20	1.55±0.351	0.477-7.94	1.165
PFTeDA	20/20	0.182±0.015	0.009-0.306	0.189
PFBS	nd	nd	nd	nd
PFHxS	19/20	1.75±0.720	nd-15.22	1.054
Br-PFOS	20/20	3.62±2.50	0.156-51.1	0.878
L-PFOS	20/20	51.4±18.0	12.4-382	28.986
6:2 FTS	19/20	nd	nd	nd
FOSA	nd	0.035±0.006	nd-0.122	0.032
$\sum_{14}$PFAS	-	68.9±22.3	16.98-479	44.728

Table S23 Relative distribution (mean±SEM) of the sum [∑] branched PFOS isomer (Br-PFOS) versus linear PFOS (L-PFOS) in abiotic and biotic samples in the Longyearbyen

Sample	Br-PFOS % of sum ΣPFOS	L-PFOS % of ΣPFOS	SEM
Snow	23.4	76.6	0.71
Ref-creek	30.0	70.0	0.41
FFTS-creek	31.2	68.8	0.78
FFTS-pond	39.8	60.2	0.78
Landfil leachate	27.1	72.9	0.91
LY-river	26.3	73.7	0.91
St-1-Seawater	27.9	72.1	0.82
St-2-Seawater	26.3	73.7	1
St-3-Seawater	27.0	73.0	0.65
St-4-Seawater	29.6	70.4	0.47
Marine sediment	20.5	79.5	0.7
Lansdfill sediment	6.7	93.3	0.1
Zooplankton	21.9	78.0	2.2
Polychaetes	15.4	84.6	2.3
Crab	8.7	91.3	3.2
Fish muscle	13.5	86.5	1.3
Fish liver	18.7	81.3	2.34
Glaucous gull	12.8	87.1	3.3

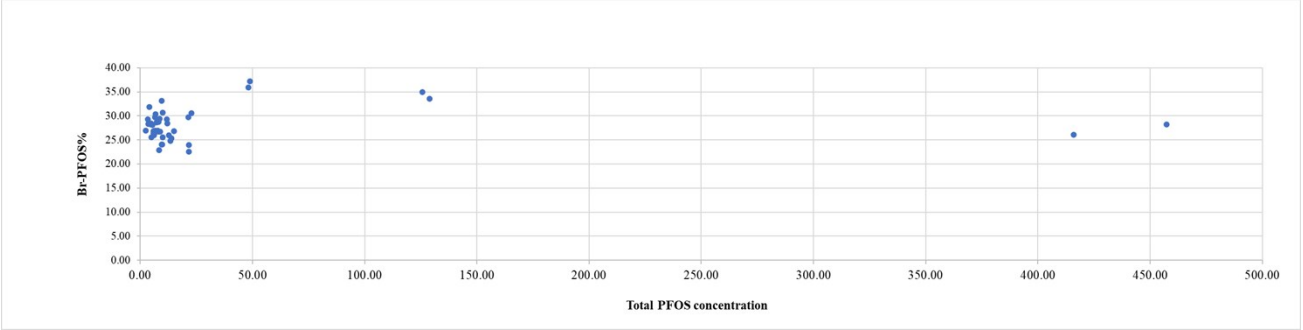


Figure S7. Sum [∑] branched PFOS isomer percentages (Br-PFOS%) versus PFOS total concentration

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