

# Supporting Information

## Transport of legacy perfluoroalkyl substances and the replacement compound HFPO-DA through the Atlantic Gateway to the Arctic Ocean – Is the Arctic a sink or a source?

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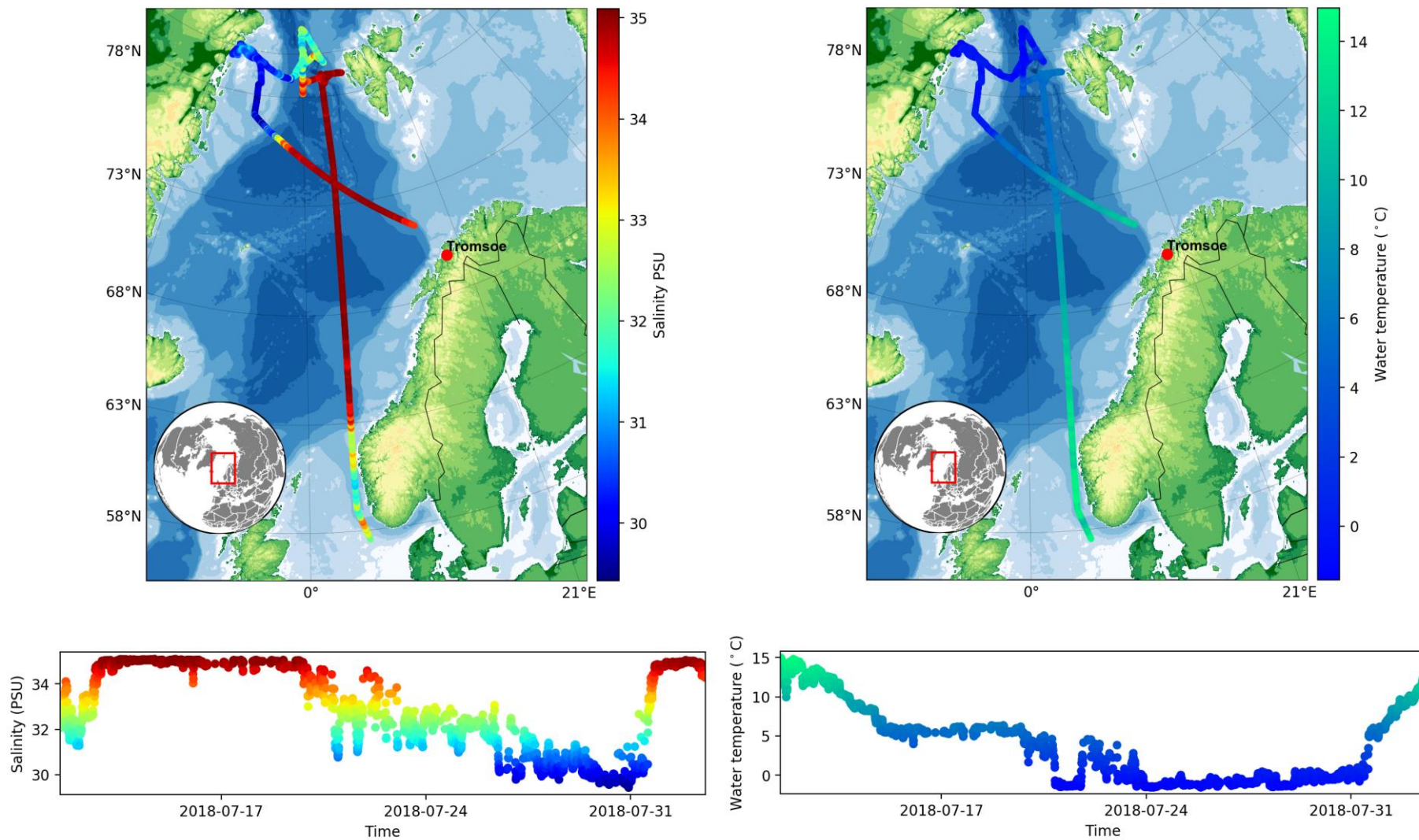
**Table S1: Sampling locations of surface seawater samples analyzed in this study and physicochemical parameters measured in the water phase.**

sample name	latitude °N	longitude °E	date   time of sampling (UTC)	water depth [m]	temperature [°C] <sup>1</sup>	pH <sup>1</sup>	salinity [PSU] <sup>1</sup>
<b>Latitudinal transect</b> from the European continent to the Arctic (57°N to 79°N at ~5°E) distance between N1 and N21: 2,432 km							
N1	57.137	5.409	11.07.2018   09:29	51	na <sup>2</sup>	na <sup>2</sup>	na <sup>2</sup>
N2	57.735	5.023	11.07.2018   12:53	96	14.52	na <sup>2</sup>	32.22
N3	58.539	4.488	11.07.2018   17:17	277	11.14	na <sup>2</sup>	33.73
N4	60.618	4.169	12.07.2018   04:12	300	14.08	na <sup>2</sup>	31.78
N5	62.007	4.203	12.07.2018   11:16	148	13.08	8.19	32.59
N6	63.109	4.256	12.07.2018   16:50	978	13.04	8.28	34.69
N7	64.035	4.305	12.07.2018   21:40	1410	12.93	8.27	34.87
N8	65.301	4.374	13.07.2018   04:18	976	12.20	8.24	35.02
N9	66.139	4.423	13.07.2018   08:32	1219	11.36	8.27	34.86
N10	67.042	4.474	13.07.2018   13:11	1331	10.36	8.22	35.05
N11	68.140	4.540	13.07.2018   18:47	1765	9.91	8.23	34.92
N12	68.937	4.590	13.07.2018   22:45	3159	8.87	8.24	35.02
N13	70.126	4.672	14.07.2018   04:51	3153	8.79	8.24	35.01
N14	71.087	4.739	14.07.2018   09:40	3048	8.26	8.25	35.06
N15	72.240	4.824	14.07.2018   15:31	2450	8.24	8.27	35.05
N16	73.334	4.917	14.07.2018   21:02	2558	7.10	8.26	35.03
N17	74.803	5.025	15.07.2018   04:20	3203	5.45	8.30	35.02
N18	75.755	4.887	15.07.2018   09:10	2657	5.94	8.28	35.04
N19	76.606	4.753	15.07.2018   13:27	2806	5.61	8.34	35.04
N20	77.722	4.568	15.07.2018   19:15	2890	5.36	8.37	35.02
N21	79.021	4.335	16.07.2018   14:36	2476	5.48	8.38	34.86
<b>Longitudinal transect</b> across Fram Strait (9°E to 18°W at ~79°N) distance between F1 and F15: 558 km							
F1	79.000	9.300	19.07.2018   00:10	218.0	6.12	8.37	34.92
F2	79.001	8.901	18.07.2018   23:41	216.7	5.83	8.36	34.93
F3	79.018	8.459	18.07.2018   22:02	476.0	6.28	8.36	34.99
F4	79.002	7.758	18.07.2018   17:20	1194.1	6.31	8.29	35.03
F5 (=N21) <sup>3</sup>	79.021	4.335	16.07.2018   14:36	2476.0	5.48	8.38	34.86
F6	79.259	-1.371	26.07.2018   04:43	2639.4	-0.52	8.24	31.50
F7	78.968	-5.089	27.07.2018   05:36	1216.2	-0.93	8.28	30.00
F8	79.402	-9.004	29.07.2018   13:38	77.8	-0.36	8.17	30.13
F9	79.440	-10.623	28.07.2018   04:20	104.0	0.45	8.09	30.37
F10	79.819	-12.273	27.07.2018   22:13	195.5	-0.36	8.13	30.46
F11	79.676	-13.606	29.07.2018   06:42	140.0	-0.14	8.13	30.77
F12	80.038	-14.643	28.07.2018   01:34	171.1	0.07	8.11	31.42
F13	79.618	-16.591	28.07.2018   19:07	274.0	-1.43	8.14	30.61
F14	80.125	-16.642	28.07.2018   07:18	327.0	-1.41	8.18	30.24
F15	79.619	-17.840	28.07.2018   15:14	439.0	-1.38	8.16	30.00
<b>Latitudinal transect</b> in Fram Strait (78°N to 81°N at ~0°EW) distance between P1 and P5: 284 km							
P1	78.240	0.001	20.07.2018   09:16	3031.3	4.41	8.35	34.44
P2	78.819	0.000	20.07.2018   13:02	2630.1	2.40	8.35	33.25
P3	79.455	0.073	21.07.2018   14:09	2817.3	-1.25	8.30	32.41
P4	80.170	0.125	25.07.2018   06:04	3095.4	-1.26	8.13	32.09
P5	80.792	0.057	24.07.2018   20:30	3168.8	-0.87	8.19	31.74

<sup>1</sup> Data was taken from von Appen and Rohardt.<sup>1</sup>

<sup>2</sup> As systems measuring the physicochemical parameters had to be set up and calibrated on the first day of the cruise, continuous measurements for those only started on the second day.

<sup>3</sup> At sampling location N21/F5, the latitudinal transect from the European continent to the Arctic and the longitudinal transect across Fram Strait crossed. Consequently, this sample was used for evaluation of both sampling transects.



**Figure S1: A) Salinity data and B) water temperature data from Polarstern cruise PS114, measured by sensor TSG1 (SBE21-3189, SBE38-118).** The figure was taken from von Appen and Rohardt.<sup>1</sup> General information on the cruise can be found in the expedition report.<sup>2</sup>

**Table S2: Sampling information for depth profiles analyzed in this study and physicochemical parameters measured in samples from the respective sampling depths.<sup>1</sup>**

event name <sup>2</sup>	sample name	latitude °N	longitude °E	date   time (UTC, profile start)	water depth [m]	sampling depth [m] <sup>2</sup>	salinity [PSU] <sup>2</sup>	temperature [°C] <sup>2</sup>	$\theta$ [°C] <sup>3</sup>	$\sigma_0$ [kg/m <sup>3</sup> ] <sup>4</sup>	water mass <sup>5</sup>
<b>vertical profile 1 (V1)</b>											
PS114_12-4	V1/1	79.012	7.033	2018-07-18   15:35	1277	1239	34.91	-0.80	-0.85	28.08	DW
PS114_12-4	V1/2	79.012	7.033	2018-07-18   15:35	1277	1000	34.91	-0.71	-0.75	28.06	DW
PS114_12-1	V1/3	79.012	7.035	2018-07-18   10:41	1277	500	34.99	2.02	1.99	28.01	AW
PS114_12-1	V1/4	79.012	7.035	2018-07-18   10:41	1277	250	35.05	3.52	3.50	27.94	AW
PS114_12-1	V1/5	79.012	7.035	2018-07-18   10:41	1277	100	35.06	4.37	4.36	27.88	AW
PS114_12-1	V1/6	79.012	7.035	2018-07-18   10:41	1277	75	35.05	4.76	4.76	27.85	AW
PS114_12-1	V1/7	79.012	7.035	2018-07-18   10:41	1277	50	35.04	5.35	5.35	27.74	AW
PS114_12-1	V1/8	79.012	7.035	2018-07-18   10:41	1277	10	34.97	6.02	6.01	27.59	AW
<b>vertical profile 2 (V2)</b>											
PS114_9-4	V2/1	78.607	5.047	2018-07-18   00:37	2344	2286	34.92	-0.74	-0.86	28.07	DW
PS114_9-4	V2/2	78.607	5.047	2018-07-18   00:37	2344	1000	34.91	-0.41	-0.45	28.07	IW
PS114_9-1	V2/3	78.607	5.041	2018-07-17   18:53	2347	500	34.93	0.72	0.70	27.96	AW
PS114_9-1	V2/4	78.607	5.041	2018-07-17   18:53	2347	250	35.01	2.52	2.50	27.88	AW
PS114_9-1	V2/5	78.607	5.041	2018-07-17   18:53	2347	100	35.04	3.45	3.44	27.79	AW
PS114_9-1	V2/6	78.607	5.041	2018-07-17   18:53	2347	75	35.03	3.64	3.64	27.74	AW
PS114_9-1	V2/7	78.607	5.041	2018-07-17   18:53	2347	40	35.01	4.57	4.56	27.67	AW
PS114_9-1	V2/8	78.607	5.041	2018-07-17   18:53	2347	10	34.96	5.46	5.46	27.53	AW
<b>vertical profile 3 (V3)</b>											
PS114_25-2	V3/1	78.830	-0.025	2018-07-20   15:53	2636	2573	34.92	-0.72	-0.86	28.08	DW
PS114_25-2	V3/2	78.830	-0.025	2018-07-20   15:53	2636	1001	34.91	-0.23	-0.27	28.05	IW
PS114_25-2	V3/3	78.830	-0.025	2018-07-20   15:53	2636	501	34.92	1.09	1.06	27.98	AW
PS114_25-2	V3/4	78.830	-0.025	2018-07-20   15:53	2636	250	35.00	2.75	2.73	27.91	AW
PS114_25-2	V3/5	78.830	-0.025	2018-07-20   15:53	2636	100	34.93	2.59	2.58	27.87	AW
PS114_25-2	V3/6	78.830	-0.025	2018-07-20   15:53	2636	75	34.89	2.58	2.57	27.84	AW
PS114_25-2	V3/7	78.830	-0.025	2018-07-20   15:53	2636	50	34.72	1.49	1.48	27.78	AW
PS114_25-2	V3/8	78.830	-0.025	2018-07-20   15:53	2636	10	34.51	4.62	4.62	27.33	AW
<b>vertical profile 4 (V4)</b>											
PS114_43-2	V4/1	78.820	-2.779	2018-07-26   14:32	2595	2600	34.93	-0.73	-0.86	28.09	DW
PS114_43-2	V4/2	78.820	-2.779	2018-07-26   14:32	2595	2000	34.92	-0.62	-0.72	28.08	DW
PS114_43-2	V4/3	78.820	-2.779	2018-07-26   14:32	2595	1200	34.91	-0.26	-0.32	28.05	IW
PS114_43-2	V4/4	78.820	-2.779	2018-07-26   14:32	2595	1000	34.90	-0.10	-0.15	28.03	IW

PS114_43-2	V4/5	78.820	-2.779	2018-07-26	14:32	2595	700	34.88	0.21	0.18	28.00	RAW/AAW
PS114_43-2	V4/6	78.820	-2.779	2018-07-26	14:32	2595	500	34.87	0.57	0.54	27.97	RAW/AAW
PS114_43-4	V4/7	78.818	-2.769	2018-07-26	18:32	2598	250	34.91	1.77	1.75	27.92	RAW/AAW
PS114_43-4	V4/8	78.818	-2.769	2018-07-26	18:32	2598	100	34.50	0.02	0.02	27.70	PW
PS114_43-4	V4/9	78.818	-2.769	2018-07-26	18:32	2598	75	34.23	-1.01	-1.02	27.53	PW
PS114_43-4	V4/10	78.818	-2.769	2018-07-26	18:32	2598	60	34.06	-1.57	-1.57	27.41	PW
PS114_43-4	V4/11	78.818	-2.769	2018-07-26	18:32	2598	40	33.92	-1.40	-1.40	27.30	PW
PS114_43-4	V4/12	78.818	-2.769	2018-07-26	18:32	2598	15	33.06	-1.51	-1.51	26.60	PW
PS114_43-4	V4/13	78.818	-2.769	2018-07-26	18:32	2598	5	30.24	-0.69	-0.69	24.29	PW

**vertical profile 5 (V5)**

PS114_46-1	V5/1	78.993	-5.436	2018-07-27	07:16	997	959	34.89	-0.02	-0.06	28.02	IW
PS114_46-1	V5/2	78.993	-5.436	2018-07-27	07:16	997	500	34.87	0.54	0.52	27.97	RAW/AAW
PS114_46-1	V5/3	78.993	-5.436	2018-07-27	07:16	997	250	34.84	1.00	0.99	27.92	RAW/AAW
PS114_46-8	V5/4	79.012	-5.285	2018-07-27	15:45	1139	100	34.32	-0.76	-0.76	27.60	PW
PS114_46-8	V5/5	79.012	-5.285	2018-07-27	15:45	1139	75	33.98	-1.33	-1.33	27.34	PW
PS114_46-8	V5/6	79.012	-5.285	2018-07-27	15:45	1139	50	33.61	-1.50	-1.50	27.05	PW
PS114_46-8	V5/7	79.012	-5.285	2018-07-27	15:45	1139	26	33.17	-1.59	-1.59	26.69	PW
PS114_46-8	V5/8	79.012	-5.285	2018-07-27	15:45	1139	5	31.32	-1.01	-1.01	25.17	PW

**vertical profile 6 (V6)**

PS114_49-2	V6/1	79.615	-16.525	2018-07-28	20:34	281	271	34.81	0.96	0.95	27.89	RAW/AAW
PS114_49-2	V6/2	79.615	-16.525	2018-07-28	20:34	281	150	34.42	0.44	0.43	27.61	RAW/AAW
PS114_49-2	V6/3	79.615	-16.525	2018-07-28	20:34	281	100	33.68	-0.89	-0.89	27.08	PW
PS114_49-2	V6/4	79.615	-16.525	2018-07-28	20:34	281	50	31.70	-1.68	-1.68	25.50	PW
PS114_49-2	V6/5	79.615	-16.525	2018-07-28	20:34	281	10	31.25	-1.53	-1.53	25.12	PW

**vertical profile 7 (V7)**

PS114_36-2	V7/1	80.855	-0.140	2018-07-24	12:48	3181	3117	34.93	-0.70	-0.88	28.09	DW
PS114_36-2	V7/2	80.855	-0.140	2018-07-24	12:48	3181	2000	34.92	-0.67	-0.77	28.08	DW
PS114_36-2	V7/3	80.855	-0.140	2018-07-24	12:48	3181	1000	34.90	-0.23	-0.27	28.04	IW
PS114_36-2	V7/4	80.855	-0.140	2018-07-24	12:48	3181	500	34.90	0.99	0.97	27.97	RAW/AAW
PS114_36-2	V7/5	80.855	-0.140	2018-07-24	12:48	3181	250	34.99	2.76	2.75	27.90	RAW/AAW
PS114_36-2	V7/6	80.855	-0.140	2018-07-24	12:48	3181	100	34.73	1.13	1.13	27.82	RAW/AAW
PS114_36-2	V7/7	80.855	-0.140	2018-07-24	12:48	3181	50	34.22	-1.43	-1.43	27.54	PW
PS114_36-2	V7/8	80.855	-0.140	2018-07-24	12:48	3181	10	33.28	-1.51	-1.51	26.77	PW

<sup>1</sup> At the CTD/rosette stations, physicochemical data was recorded during the casts<sup>3</sup> and measured in water samples taken from the Niskin bottles mounted on the CTD rosette.<sup>4</sup>

Data shown here was taken from von Appen et al.<sup>4</sup>

<sup>2</sup> Event names were taken from the expedition report<sup>2</sup> and are structured as follows: cruise\_station\_cast.<sup>3</sup> $\theta$ : potential temperature; <sup>4</sup> $\sigma_0$ : potential density referenced to sea surface

<sup>5</sup> Water masses were classified according to Rudels et al.<sup>5</sup>, based on measured physicochemical parameters (see Table S21 for further explanation).

**Table S3: Overview of analytical standards, CAS numbers, the standard suppliers, purity and concentration/amount.**

acronym <sup>1</sup>	analytical standard	CAS number <sup>2</sup>	supplier, purity and concentration/amount <sup>3</sup>
PFBA	perfluoro- <i>n</i> -butanoic acid	375-22-4 (acid)	
PFPeA	perfluoro- <i>n</i> -pentanoic acid	2706-90-3 (acid)	
PFHxA	perfluoro- <i>n</i> -hexanoic acid	307-24-4 (acid)	
PFHpA	perfluoro- <i>n</i> -heptanoic acid	375-85-9 (acid)	PFC-MXA (mixture)
PFOA	perfluoro- <i>n</i> -octanoic acid	335-67-1 (acid)	Wellington Laboratories, > 98 %
PFNA	perfluoro- <i>n</i> -nonanoic acid	375-95-1 (acid)	
PFDA	perfluoro- <i>n</i> -decanoic acid	335-76-2 (acid)	2.0 µg/mL ± 5 %
PFUnDA	perfluoro- <i>n</i> -undecanoic acid	2058-94-8 (acid)	of the single compounds
PFDoDA	perfluoro- <i>n</i> -dodecanoic acid	307-55-1 (acid)	
PFTTrDA	perfluoro- <i>n</i> -tridecanoic acid	72629-94-8 (acid)	
PFTeDA	perfluoro- <i>n</i> -tetradecanoic acid	376-06-7 (acid)	
PFBS	potassium perfluoro- <i>n</i> -butanesulfonate	29420-49-3 (K <sup>+</sup> salt) 375-73-5 (acid)	
PFHxS	sodium perfluoro- <i>n</i> -hexanesulfonate	82382-12-5 (Na <sup>+</sup> salt) 355-46-4 (acid)	PFS-MXA (mixture)
PFHpS	sodium perfluoro- <i>n</i> -heptanesulfonate	22767-50-6 (Na <sup>+</sup> salt) 375-92-8 (acid)	Wellington Laboratories, > 98 %
PFOS	sodium perfluoro- <i>n</i> -octanesulfonate	4021-47-0 (Na <sup>+</sup> salt) 1763-23-1 (acid)	2.0 µg/mL ± 5 %
PFDS	sodium perfluoro- <i>n</i> -decanesulfonate	2806-15-7 (Na <sup>+</sup> salt) 335-77-3 (acid)	of the single compounds
PFECHS	potassium perfluoro-4-ethylcyclohexane-sulfonate (isomeric mixture <i>cis/trans</i> 1:3)	335-24-0 (K <sup>+</sup> salt) 646-83-3 (acid)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
DONA	sodium 2,2,3-trifluoro-3-[1,1,2,2,3,3-hexafluoro-3-(trifluoromethoxy)propoxy]-propionate	958445-44-8 (Na <sup>+</sup> salt) 919005-14-4 (acid)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
HFPO-DA	2,3,3,3-tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)propanoic acid	13252-13-6 (acid) 62037-80-3 (NH <sub>4</sub> <sup>+</sup> salt, GenX)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
HFPO-TrA	2,3,3,3-tetrafluoro-2-[1,1,2,2,3,3,3-hexafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)propoxy]-propanoic acid	13252-14-7 (acid)	ABCR, 95 %, 5 g
HFPO-TeA	2,3,3,3-tetrafluoro-2-[1,1,2,2,3,3,3-hexafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)propoxy]propanoic acid	65294-16-8 (acid)	Apollo Scientific, 98 %, 5 g
6:2 Cl-PFESA	potassium 2-[(6-chloro-1,1,2,2,3,3,3,4,4,5,5,6,6-dodecafluorohexyl)oxy]-1,1,2,2-tetrafluoroethanesulfonate	73606-19-6 (K <sup>+</sup> salt) 756426-58-1 (acid)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
8:2 Cl-PFESA	potassium 2-[(8-Chloro-1,1,2,2,3,3,3,4,4,5,5,6,-6,7,7,8,8-hexadecafluorooctyl)oxy]-1,1,2,2-tetrafluoroethanesulfonate	83329-89-9 (K <sup>+</sup> salt)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
6:6 PFPiA	sodium bis(perfluorohexyl)phosphinate	70609-44-8 (Na <sup>+</sup> salt) 40143-77-9 (acid)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
6:8 PFPiA	sodium perfluorohexylperfluorooctylphosphinate	610800-34-5 (acid)	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
FOSA	perfluorooctane sulfonamide	754-91-6	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
4:2 FTSA	sodium 4:2 fluorotelomer sulfonate	757124-72-4 (acid)	Wellington Laboratories,



6:2 FTSA	sodium 6:2 fluorotelomer sulfonate	27619-94-9 (Na <sup>+</sup> salt) 27619-97-2 (acid)	> 98 %, (50 ± 2.5) µg/mL Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL Wellington Laboratories,
8:2 FTSA	sodium 8:2 fluorotelomer sulfonate	39108-34-4 (acid)	> 98 %, (50 ± 2.5) µg/mL
<sup>13</sup> C <sub>4</sub> -PFBA	perfluoro- <i>n</i> -[ <sup>13</sup> C <sub>4</sub> ]-butanoic acid	-	
<sup>13</sup> C <sub>2</sub> -PFHxA	perfluoro- <i>n</i> -[1,2- <sup>13</sup> C <sub>2</sub> ]-hexanoic acid	-	
<sup>13</sup> C <sub>4</sub> -PFOA	perfluoro- <i>n</i> -[1,2,3,4- <sup>13</sup> C <sub>4</sub> ]-octanoic acid	-	
<sup>13</sup> C <sub>5</sub> -PFNA	perfluoro- <i>n</i> -[1,2,3,4,5- <sup>13</sup> C <sub>5</sub> ]-nonanoic acid	-	MPFAC-MXA (mixture)
<sup>13</sup> C <sub>2</sub> -PFDA	perfluoro- <i>n</i> -[1,2- <sup>13</sup> C <sub>2</sub> ]-decanoic acid	-	Wellington Laboratories,
<sup>13</sup> C <sub>2</sub> -PFUnDA	perfluoro- <i>n</i> -[1,2- <sup>13</sup> C <sub>2</sub> ]-undecanoic acid	-	> 98 %, 2.0 µg/mL ± 5 % of the single compounds
<sup>13</sup> C <sub>2</sub> -PFDoDA	perfluoro- <i>n</i> -[1,2- <sup>13</sup> C <sub>2</sub> ]-dodecanoic acid	-	
<sup>18</sup> O <sub>2</sub> -PFHxS	sodium perfluorohexane- <i>n</i> -[ <sup>18</sup> O <sub>2</sub> ]-sulfonate	-	
<sup>13</sup> C <sub>4</sub> -PFOS	sodium perfluoro- <i>n</i> -[1,2,3,4- <sup>13</sup> C <sub>4</sub> ]-octanesulfo-nate	-	
<sup>13</sup> C <sub>3</sub> -HFPO-DA	2,3,3,3-tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)-[ <sup>13</sup> C <sub>3</sub> ]-propanoic acid	-	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL
<sup>13</sup> C <sub>8</sub> -PFOA	perfluoro-[ <sup>13</sup> C <sub>8</sub> ]-octanoic acid (injection standard)	-	Wellington Laboratories, > 98 %, (50 ± 2.5) µg/mL

<sup>1</sup> The standards are prepared from salts or the free acid (see column 2), whereas the measured form of the target analyte is the anion. The acronyms are used for all three forms. To name the compounds, the nomenclature proposed by Buck et al. was followed, where applicable.<sup>6</sup> For emerging PFASs, commonly used acronyms were used and the IUPAC names for the respective compounds are given in column 2 of the table.

<sup>2</sup> If standards are provided as dissolved salts, the CAS number of the salt and the free acid counterpart is given (if available).

<sup>3</sup> Given concentrations refer to the salts.

**Table S4: Chemicals used for sample preparation and cleaning.**

chemical	specification by supplier	supplier
methanol	for liquid chromatography, LiChrosolv	Merck, Germany
ultrapure water	supplied by Milli-Q Integral 5 system	Merck, Germany
acetic acid	≥ 99.8% eluent additive, for LC-MS	Honeywell Fluka, USA
ammonium acetate	for mass spectrometry, eluent additive for LC-MS	Merck, Germany
ammonia solution 25 %	Suprapur	Merck, Germany

## Section S1: Sample extraction

Solid phase extraction for 1 L samples was performed as described previously.<sup>7</sup> Weak anion exchange cartridges (Oasis WAX, 6 cc, 500 mg sorbent, 60 µm particle size, Waters, USA) were preconditioned by 6 mL 0.1% ammonium hydroxide in methanol, 6 mL methanol and 6 mL water. An internal standard mix was added to the samples (400 pg of each standard) before they were loaded onto the cartridges. A washing step using an 80:20 (v/v) water/methanol solution followed.<sup>8</sup> To remove matrix components, especially sea salt, and to improve recovery rates for long-chain PFASs, a comparatively large volume of 15 mL of the washing solution was used per sample. After vacuum drying of the cartridges and storage at -20 °C, the target compounds were eluted using 6 mL methanol and 6 mL 0.1% ammonium hydroxide in methanol. Subsequently, the combined fractions were reduced to 150 µL under nitrogen. Before the measurement, 40 µL water (20 % v/v) as well as 400 pg of the injection standard <sup>13</sup>C<sub>8</sub>-PFOA (10 µL of a 40 pg/µL solution) were added to the sample.

**Table S5: LC parameters and MS/MS parameters of the instrumental method for PFAS analysis.**

LC parameters				
instrument	HP 1100 (Agilent Technologies, USA)			
analytical column	Synergi Fusion-RP C <sub>18</sub> , 150 mm x 2 mm, particle size 4 µm, pore size 80 Å (Phenomenex, USA)			
guard column	SecurityGuard cartridge for Fusion-RP LC columns, 4 mm x 2 mm (Phenomenex, USA)			
injection volume	10 µL (needle rinsed twice with methanol before injection)			
column temperature	30 °C			
flow rate	0.2 mL/min			
mobile phases	A: 2 mM ammonium acetate aqueous solution B: 0.05 % acetic acid in methanol			
gradient	Time [min]	A [%]	B [%]	note
	-8	70	30	equilibration
	0	70	30	
	3	30	70	
	29	10	90	
	31	0	100	purging
	45	0	100	
MS/MS parameters				
instrument	API 4000 triple quadrupole mass spectrometer (AB Sciex, USA)			
ion source	Turbo V Ion Source (AB Sciex, USA)			
ionization	electrospray ionization (ESI) in negative mode			
ion spray voltage	-4500 V			
source temperature	400 °C			
gas 1 (nebulizer gas)	N <sub>2</sub> , 4.2 bar			
gas 2 (heater gas)	N <sub>2</sub> , 2.8 bar			
curtain gas	N <sub>2</sub> , 1.0 bar			
collision gas	N <sub>2</sub> , 0.6 bar			
scan type	Scheduled Multiple Reaction Monitoring (MRM)			
	retention time window: 180 s, target scan time: 2 s			

**Table S6: Retention times, precursor ion, monitored mass transitions, transition-specific parameters and corresponding internal standards used for quantification of the target analytes.**

acronym analyte	tr <sup>1</sup> [min]	molecular formula precursor ion	monitored mass transition s <sup>2</sup> [m/z]	transition-specific parameters [V]				internal standard <sup>7</sup>
				DP <sup>3</sup>	EP <sup>4</sup>	CE <sup>5</sup>	CXP <sup>6</sup>	
PFBA	10.1	[C <sub>4</sub> F <sub>7</sub> O <sub>2</sub> ] <sup>-</sup>	213>169*	-30	-5	-13	-9	<sup>13</sup> C <sub>4</sub> -PFBA
PFPeA	11.9	[C <sub>5</sub> F <sub>9</sub> O <sub>2</sub> ] <sup>-</sup>	263>219*	-26	-4	-12	-13	<sup>13</sup> C <sub>2</sub> -PFHxA
PFHxA	13.4	[C <sub>6</sub> F <sub>11</sub> O <sub>2</sub> ] <sup>-</sup>	313>269*	-28	-4	-13	-16	<sup>13</sup> C <sub>2</sub> -PFHxA
			313>119	-28	-4	-30	-5	
PFHpA	15.1	[C <sub>7</sub> F <sub>13</sub> O <sub>2</sub> ] <sup>-</sup>	363>319	-29	-4	-14	-19	<sup>13</sup> C <sub>4</sub> -PFOA
			363>169*	-29	-4	-25	-8	
PFOA	17.1	[C <sub>8</sub> F <sub>15</sub> O <sub>2</sub> ] <sup>-</sup>	413>369*	-24	-4	-15	-8	<sup>13</sup> C <sub>4</sub> -PFOA
			413>169	-24	-4	-28	-8	
PFNA	19.7	[C <sub>9</sub> F <sub>17</sub> O <sub>2</sub> ] <sup>-</sup>	463>419*	-34	-4	-15	-9	<sup>13</sup> C <sub>5</sub> -PFNA
			463>219	-34	-4	-24	-12	
PFDA	22.6	[C <sub>10</sub> F <sub>19</sub> O <sub>2</sub> ] <sup>-</sup>	513>469*	-35	-6	-15	-11	<sup>13</sup> C <sub>2</sub> -PFDA
			513>219	-35	-6	-29	-12	
PFUnDA	25.6	[C <sub>11</sub> F <sub>21</sub> O <sub>2</sub> ] <sup>-</sup>	563>519*	-35	-5	-17	-13	<sup>13</sup> C <sub>2</sub> -PFUnDA
			563>169	-35	-5	-37	-8	
PFDoDA	28.7	[C <sub>12</sub> F <sub>23</sub> O <sub>2</sub> ] <sup>-</sup>	613>569*	-38	-9	-17	-15	<sup>13</sup> C <sub>2</sub> -PFDoDA
			613>169	-38	-9	-38	-8	
PFTTrDA	31.7	[C <sub>13</sub> F <sub>25</sub> O <sub>2</sub> ] <sup>-</sup>	663>619*	-39	-8	-18	-14	<sup>13</sup> C <sub>2</sub> -PFDoDA
			663>169	-39	-8	-41	-8	
PFTeDA	34.6	[C <sub>14</sub> F <sub>27</sub> O <sub>2</sub> ] <sup>-</sup>	713>669*	-36	-9	-22	-15	<sup>13</sup> C <sub>2</sub> -PFDoDA
			713>169	-36	-9	-40	-8	
PFBS	12.0	[C <sub>4</sub> F <sub>9</sub> O <sub>3</sub> S] <sup>-</sup>	299>99	-66	-12	-42	-16	<sup>18</sup> O <sub>2</sub> -PFHxS
			299>80*	-66	-12	-60	-2	
PFHxS	15.0	[C <sub>6</sub> F <sub>13</sub> O <sub>3</sub> S] <sup>-</sup>	399>99	-70	-14	-50	-15	<sup>18</sup> O <sub>2</sub> -PFHxS
			399>80*	-70	-14	-66	-2	
PFHpS	17.0	[C <sub>7</sub> F <sub>15</sub> O <sub>3</sub> S] <sup>-</sup>	449>99	-80	-12	-61	-16	<sup>13</sup> C <sub>4</sub> -PFOS
			449>80*	-80	-12	-85	-2	
L-PFOS	19.5	[C <sub>8</sub> F <sub>17</sub> O <sub>3</sub> S] <sup>-</sup>	499>99	-73	-12	-74	-17	<sup>13</sup> C <sub>4</sub> -PFOS
			499>80*	-73	-12	-90	-2	
Br-PFOS	18.7/18.4	[C <sub>8</sub> F <sub>17</sub> O <sub>3</sub> S] <sup>-</sup>	499>99	-73	-12	-74	-17	<sup>13</sup> C <sub>4</sub> -PFOS
			499>80*	-73	-12	-90	-2	
PFDS	25.3	[C <sub>10</sub> F <sub>21</sub> O <sub>3</sub> S] <sup>-</sup>	599>99	-80	-14	-62	-3	<sup>13</sup> C <sub>4</sub> -PFOS
			599>80*	-80	-14	-90	-2	
PFECHS	16.7	[C <sub>8</sub> F <sub>15</sub> O <sub>3</sub> S] <sup>-</sup>	461>381*	-60	-10	-40	-20	<sup>18</sup> O <sub>2</sub> -PFHxS
			461>99	-60	-10	-55	-4	
DONA	15.1	[C <sub>7</sub> HF <sub>12</sub> O <sub>4</sub> ] <sup>-</sup>	377>251*	-33	-6	-22	-16	<sup>13</sup> C <sub>4</sub> -PFOA
			377>85	-33	-6	-50	-14	
HFPO-DA	14.0	[C <sub>6</sub> F <sub>11</sub> O <sub>3</sub> ] <sup>-</sup>	329>285	-22	-4	-7	-18	<sup>13</sup> C <sub>3</sub> -HFPO-DA
			329>169*	-22	-4	-18	-8	
HFPO-TrA	20.7	[C <sub>9</sub> F <sub>17</sub> O <sub>4</sub> ] <sup>-</sup>	495>185*	-15	-4	-9	-12	<sup>13</sup> C <sub>2</sub> -PFDA
HFPO-TeA	28.3	[C <sub>12</sub> F <sub>23</sub> O <sub>5</sub> ] <sup>-</sup>	661>351*	-8	-8	-8	-2	<sup>13</sup> C <sub>2</sub> -PFDoDA
			661>185	-30	-4	-45	-14	
6:2 Cl-PFESA	20.9	[C <sub>8</sub> ClF <sub>16</sub> O <sub>4</sub> S] <sup>-</sup>	531>351*	-60	-14	-39	-20	<sup>13</sup> C <sub>4</sub> -PFOS
			531>83	-60	-14	-61	-14	
8:2 Cl-PFESA	26.8	[C <sub>10</sub> ClF <sub>20</sub> O <sub>4</sub> S] <sup>-</sup>	631>451*	-75	-13	-44	-20	<sup>13</sup> C <sub>4</sub> -PFOS
			631>83	-75	-13	-82	-2	
6:6 PFPiA	29.5	[C <sub>12</sub> F <sub>26</sub> O <sub>2</sub> P] <sup>-</sup>	701>401*	-75	-8	-77	-16	-
			701>63	-75	-8	-89	-10	
6:8 PFPiA	34.1	[C <sub>14</sub> F <sub>30</sub> O <sub>2</sub> P] <sup>-</sup>	801>501*	-57	-11	-83	-20	-
			801>401	-57	-11	-82	-18	
L-FOSA	20.9	[C <sub>8</sub> HF <sub>17</sub> NO <sub>2</sub> S] <sup>-</sup>	498>78*	-84	-10	-69	-2	<sup>13</sup> C <sub>8</sub> -FOSA
Br-FOSA	19.5/19.9	[C <sub>8</sub> HF <sub>17</sub> NO <sub>2</sub> S] <sup>-</sup>	498>78*	-84	-10	-69	-2	<sup>13</sup> C <sub>8</sub> -FOSA
4:2 FTSA	13.0	[C <sub>6</sub> H <sub>4</sub> F <sub>9</sub> O <sub>3</sub> S] <sup>-</sup>	327>307	-60	-10	-28	-18	<sup>13</sup> C <sub>2</sub> -6:2 FTSA
			327>81*	-60	-10	-48	-2	
6:2 FTSA	16.7	[C <sub>8</sub> H <sub>4</sub> F <sub>13</sub> O <sub>3</sub> S] <sup>-</sup>	427>407	-70	-13	-34	-20	<sup>13</sup> C <sub>2</sub> -6:2 FTSA
			427>81*	-70	-13	-68	-2	
8:2 FTSA	22.2	[C <sub>10</sub> H <sub>4</sub> F <sub>17</sub> O <sub>3</sub> S] <sup>-</sup>	527>507	-80	-14	-40	-20	<sup>13</sup> C <sub>2</sub> -6:2 FTSA
			527>81*	-80	-14	-76	-2	
<sup>13</sup> C <sub>4</sub> -PFBA	10.1	[ <sup>13</sup> C <sub>4</sub> F <sub>7</sub> O <sub>2</sub> ] <sup>-</sup>	217>172*	-22	-4	-13	-9	
<sup>13</sup> C <sub>2</sub> -PFHxA	13.4	[ <sup>13</sup> C <sub>2</sub> <sup>12</sup> C <sub>4</sub> F <sub>11</sub> O <sub>2</sub> ] <sup>-</sup>	315>270*	-23	-6	-12	-16	
			315>120	-23	-6	-31	-4	

<sup>13</sup> C <sub>4</sub> -PFOA	17.1	[ <sup>13</sup> C <sub>4</sub> <sup>12</sup> C <sub>4</sub> F <sub>15</sub> O <sub>2</sub> ] <sup>-</sup>	417>372*	-32	-4	-13	-8
			417>169	-32	-4	-27	-8
<sup>13</sup> C <sub>8</sub> -PFOA <sup>8</sup>	17.1	[ <sup>13</sup> C <sub>8</sub> F <sub>15</sub> O <sub>2</sub> ] <sup>-</sup>	421>376*	-25	-6	-14	-8
			421>172	-25	-6	-26	-8
<sup>13</sup> C <sub>5</sub> -PFNA	19.7	[ <sup>13</sup> C <sub>5</sub> <sup>12</sup> C <sub>4</sub> F <sub>17</sub> O <sub>2</sub> ] <sup>-</sup>	468>423*	-30	-7	-14	-10
			468>223	-30	-7	-24	-12
<sup>13</sup> C <sub>2</sub> -PFDA	22.6	[ <sup>13</sup> C <sub>2</sub> <sup>12</sup> C <sub>8</sub> F <sub>19</sub> O <sub>2</sub> ] <sup>-</sup>	515>470*	-39	-6	-16	-10
			515>220	-39	-6	-26	-12
<sup>13</sup> C <sub>2</sub> -PFUnDA	25.6	[ <sup>13</sup> C <sub>2</sub> <sup>12</sup> C <sub>9</sub> F <sub>21</sub> O <sub>2</sub> ] <sup>-</sup>	565>520*	-33	-6	-16	-13
			565>169	-33	-6	-34	-8
<sup>13</sup> C <sub>2</sub> -PFDoDA	28.7	[ <sup>13</sup> C <sub>2</sub> <sup>12</sup> C <sub>10</sub> F <sub>23</sub> O <sub>2</sub> ] <sup>-</sup>	615>570*	-38	-9	-17	-15
			615>169	-38	-9	-41	-8
<sup>18</sup> O <sub>2</sub> -PFHxS	15.0	[C <sub>6</sub> F <sub>13</sub> <sup>18</sup> O <sub>2</sub> <sup>16</sup> OS] <sup>-</sup>	403>103	-82	-10	-55	-4
			403>84*	-82	-10	-79	-2
<sup>13</sup> C <sub>4</sub> -PFOS	19.5	[ <sup>13</sup> C <sub>4</sub> <sup>12</sup> C <sub>4</sub> F <sub>17</sub> O <sub>3</sub> S] <sup>-</sup>	503>99	-65	-12	-64	-4
			503>80*	-65	-12	-92	-2
<sup>13</sup> C <sub>3</sub> -HFPO-DA	14.0	[ <sup>13</sup> C <sub>3</sub> <sup>12</sup> C <sub>3</sub> F <sub>11</sub> O <sub>3</sub> ] <sup>-</sup>	332>287	-21	-3	-7	-14
			332>169*	-21	-3	-23	-8

<sup>1</sup> t<sub>R</sub>: retention time

<sup>2</sup> Asterisks mark the product ion which was used as quantifier, whereas the second product ion was used as qualifier. The most intense ion was taken as quantifier except for HFPO-DA and PFHpA. For these compounds, the product ion with the second highest intensity was taken for quantification because of matrix interferences, which affected the first mass transition. For PFBA, PFPeA and FOSA, only one mass transition was monitored because product ion scans showed the generation of only one product ion with a high enough intensity. This is consistent with literature.<sup>8-9</sup>

<sup>3</sup>DP: declustering potential, <sup>4</sup>EP: entrance potential, <sup>5</sup>CE: collision energy, <sup>6</sup>CXP: cell exit potential

<sup>7</sup> The column lists the internal standards used for quantification of the compounds.

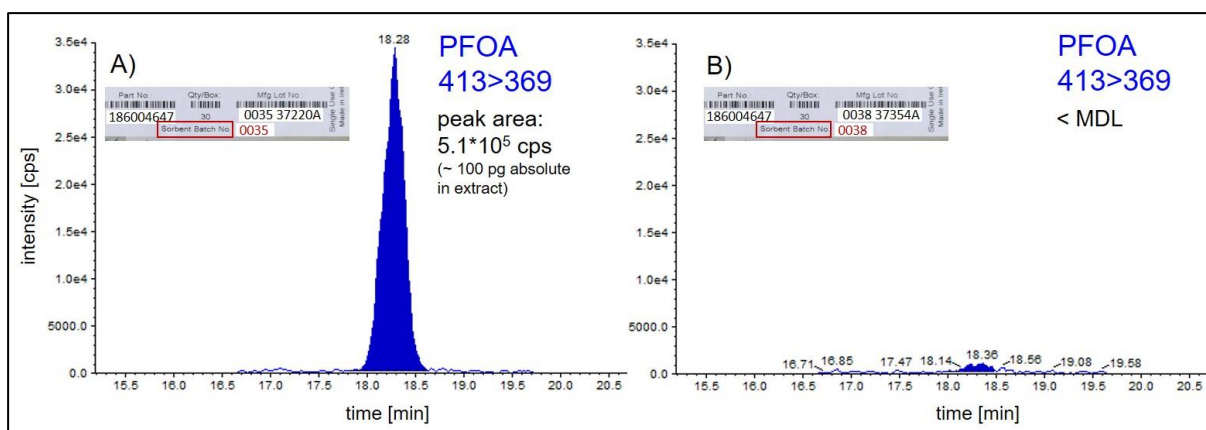
<sup>8</sup> <sup>13</sup>C<sub>8</sub>-PFOA was added as injection standard.

## **Section S2: Quantification of the target analytes**

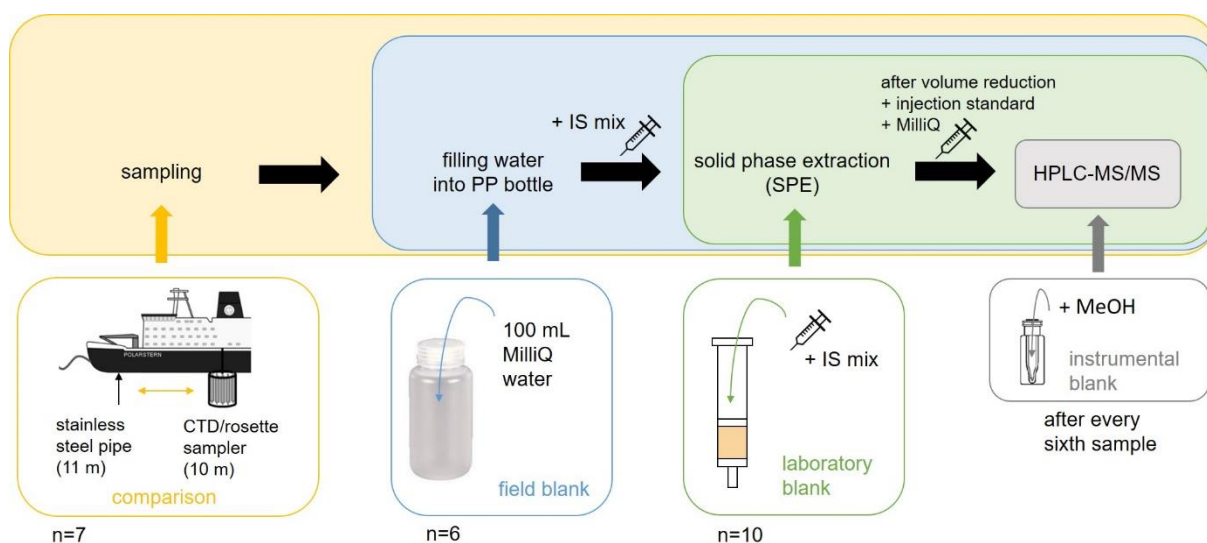
The internal standard method was used for quantification, based on a 11-point calibration curve ranging from 0 to 10 pg/ $\mu$ L. Standards were prepared in 80:20 (v/v) methanol/water. A 1/x weighting factor was applied to improve accuracy for the low concentration region of the curve. All reported results were calculated on anion basis, i.e. corrected for the molecular weight of the salts, which were used to prepare the standard solutions.

Both the linear isomer of PFOS (L-PFOS) and the branched isomers (Br-PFOS) were detected in the samples. As their peaks were baseline-separated, L-PFOS and Br-PFOS were quantified individually against the linear calibration standard (mass transition  $m/z$  499 $\rightarrow$ 80).

For most of the PFCAs and PFSAAs, isotopically analogues were available, whereas only  $^{13}\text{C}_3$ -HFPO-DA could be obtained to quantify emerging substances from other groups of PFASs. To assign substances without isotopically labelled analogues to structurally similar internal standards, the results from matrix spike recovery tests were used. The internal standards which were used for quantification of the compounds are given in Table S6.



**Figure S2: LC-MS/MS chromatograms of PFOA showing differences in contamination between cartridges of the same manufacturer and part number, packed with different WAX resin sorbent batches, 0035 (A) and 0038 (B).** Cartridges were preconditioned with 6 mL 0.1 % NH<sub>4</sub>OH in methanol and 6 mL methanol. Then, 6 mL methanol and 6 mL 0.1 % NH<sub>4</sub>OH in methanol were added as for the elution of the target compounds and the eluates were collected. They were reduced to 200  $\mu$ L and analyzed by LC-MS/MS. The peak area of PFOA in A) was comparable to a 0.5  $\mu$ g/ $\mu$ L calibration standard, which contains 100  $\mu$ g PFOA absolute. The different WAX resin sorbent batches were tested before the cruise and only cartridges packed with sorbent lots for which tests showed no contamination with PFOA were used for extraction.



**Figure S3: Preparation and analysis of blank samples.** Methanol was injected as instrumental blank after every sixth sample in a measurement sequence. On board, one laboratory blank was prepared with every SPE batch by adding the IS mix to a preconditioned cartridge, leaving the cartridge on the vacuum manifold during the extraction and treating it as if it was a sample from the washing step on (n=10). In addition, six field blanks were prepared on different sampling days by filling a sampling bottle with 100 mL of pretested MilliQ water and analyzing it like a field sample. When surface water samples were taken with the CTD/rosette sampler (10 m depth), a sample was taken with the stainless steel pipe (11 m depth) at the same time to compare the two sampling techniques (n=7).

**Table S7: Mean concentrations of PFASs in procedural blanks ± standard deviation (SD), method detection limits (MDLs) and method quantification limits (MQLs).**

analyte	laboratory blanks (n=10)	field blanks (n=6)	MDL <sup>2</sup>	MQL <sup>2</sup>
	[pg/L]		[pg/L]	
PFPeA	0.46±0.53	9.7±4.4	23	53
PFHxA	5.2±2.1	5.6±2.1	12	27
PFHpA	4.8±1.8	7.9±1.3	12	21
PFOA	16±4	11±2	16	29
PFNA	nd <sup>1</sup>	nd	6.3	21
PFDA	nd	nd	6.4	21
PFUnDA	nd	nd	3.7	12
PFDoDA	nd	nd	3.3	11
PFTTrDA	nd	nd	4.6	15
PFTeDA	nd	nd	8.8	29
PFBS	nd	nd	13	45
PFHxS	nd	nd	18	61
PFHpS	nd	nd	27	90
L-PFOS	11±3	11±3	21	44
Br-PFOS	4.2±2.1	5.2±2.2	12	28
PFDS	nd	nd	32	93
PFECHS	nd	nd	43	140
HFPO-DA	nd	nd	6.0	20
HFPO-TrA	nd	nd	44	150
HFPO-TeA	nd	nd	75	250
DONA	nd	nd	7.5	25
6:2 Cl-PFESA	nd	nd	6.9	23
8:2 Cl-PFESA	nd	nd	6.8	23
6:6 PFPiA	nd	nd	4.9	16
6:8 PFPiA	nd	nd	8.7	29
L-FOSA	nd	nd	8.0	27
Br-FOSA	nd	nd	11	33
4:2 FTSA	nd	nd	30	100
6:2 FTSA	20±7	25±12	61	150
8:2 FTSA	nd	nd	36	120
PFBA <sup>3</sup>	390±580	330±510	-	-

<sup>1</sup> nd = not detected

<sup>2</sup> For compounds present in blanks, MDLs and MQLs were calculated as the average blank value plus 3 or 10 times the standard deviation, respectively (blue). For PFASs other than these, the MDLs and MQLs were derived from a signal-to-noise ratio of 3 or 10, respectively, observed in low level samples (green) or spiked matrix samples (yellow).

<sup>3</sup> In blank samples processed on board, comparatively high concentrations of PFBA were found, strongly varying between the different SPE batches (red). Consequently, no results are reported for PFBA.

### **Section S3: Data analysis**

Calculation of arithmetic means and further statistical analysis was performed only for PFASs with a detection frequency >50%. Results <MDL were considered as zero and the calculated values between MDL and MQL were used unaltered for calculations. For PFAS mass transport estimates through Fram Strait, different substitution techniques of values <MDL (0,  $\sqrt{2}/2$ \*MDL and MDL) were compared (see Table S22).

OriginPro 2018 (version 9.5) was used for statistical analysis, setting the significance level at  $\alpha=0.05$ . Normality was tested by the Kolmogorov-Smirnov test, before further statistical tests were applied. To investigate the relationship between individual PFASs and between PFASs and physicochemical parameters, Pearson correlation analysis was conducted, if data was normally distributed. Otherwise, Spearman's correlation analysis was applied. To test for significant differences in PFAS concentrations and patterns between different sampling areas, two-sample t-tests were conducted. If data sets did not show homogeneity of variance according to Leuvene's test, the Welch correction was applied.



**Table S8: Coefficients of variation [%] for quantified PFASs, calculated from triplicate analysis of surface water samples taken by seawater intake system (n=2) and samples taken by CTD/rosette sampler and seawater intake system at the same time (n=7).**

compound	coefficient of variation [%]										
	triplicates seawater intake system			comparison between sampling techniques							
	N5	N22	mean	CTD_049	CTD_009	CTD_025	CTD_036	CTD_012	CTD_043	CTD_046	mean
<b>PFHxA</b>	6	6	6	6	22	3	4	12	1	10	8
<b>PFHpA</b>	2	9	5	4	3	19	14	3	11	3	8
<b>PFOA</b>	5	2	5	11	16	13	13	-	-	-	13
<b>PFNA</b>	5	7	6	8	15	9	1	24	3	4	9
<b>PFDA</b>	-	-	-	-	-	-	-	-	-	18	18
<b>PFUnDA</b>	-	1	1	-	19	-	-	-	21	3	14
<b>PFBS</b>	11	12	11	-	-	-	3	-	-	-	3
<b>PFHxS</b>	5	-	5	-	-	-	-	-	-	-	-
<b>L-PFOS</b>	18	16	17	-	20	-	-	-	-	-	20
<b>Br-PFOS</b>	3	11	7	-	-	-	3	-	-	-	3
<b>HFPO-DA</b>	7	-	7	1	-	-	-	1	-	-	1

**Table S9: Matrix spike recoveries<sup>1</sup> for seawater.**

matrix spiking level	relative recovery (mean ± SD)
	[%], n=3 seawater (1 L) 400 pg/L
PFBA	111 ± 5
PFPeA <sup>2</sup>	94 ± 2
PFHxA	108 ± 6
PFHpA <sup>2</sup>	89 ± 3
PFOA	105 ± 0
PFNA	95 ± 2
PFDA	100 ± 0
PFUnDA	95 ± 2
PFDoDA	104 ± 2
PFTTrDA <sup>2</sup>	64 ± 3
PFTeDA <sup>2</sup>	63 ± 5
PFBS	96 ± 3
PFHxS	101 ± 3
PFHpS <sup>2</sup>	109 ± 12
PFOS	97 ± 2
PFDS <sup>2</sup>	78 ± 3
PFECHS <sup>2</sup>	100 ± 1
HFPO-DA	94 ± 2
HFPO-TrA <sup>2</sup>	88 ± 4
HFPO-TeA <sup>2</sup>	73 ± 2
DONA <sup>2</sup>	88 ± 2
6:2 Cl-PFESA <sup>2</sup>	95 ± 3
8:2 Cl-PFESA <sup>2</sup>	101 ± 2
6:6 PFPiA <sup>3</sup>	69 ± 2
6:8 PFPiA <sup>3</sup>	39 ± 2
FOSA	101 ± 4
4:2 FTSA	93 ± 13
6:2 FTSA <sup>2</sup>	98 ± 4
8:2 FTSA	96 ± 11

<sup>1</sup> Matrix spike recoveries [%] were calculated using the following equation:

$$\text{matrix spike recovery} = \frac{m_{\text{spiked sample}} - m_{\text{sample}}}{m_{\text{spike}}} \cdot 100 \%$$

where  $m_{\text{spiked sample}}$  is the amount of the analyte in the spiked sample [pg],  $m_{\text{sample}}$  [pg] is the amount of analyte in the unspiked sample and  $m_{\text{spike}}$  [pg] is the amount of the analyte spiked into the sample. Quantification of the compounds was based on the peak area ratio of the analyte and the assigned internal standard.

<sup>2</sup> Because for these compounds no isotopically labelled analogues were available as internal standards, they were assigned to structurally similar internal standards (Table S6). Differences in the properties of the target analyte and the assigned internal standard can explain lower relative recoveries.

<sup>3</sup> For PFPiAs, absolute values are given as there was no structurally similar internal standard to which they could be assigned.

**Table S10: Percent absolute recoveries of internal standards<sup>1</sup> in analyzed seawater samples (n=109).**

internal standard	mean absolute recovery ± SD
	[%]
<sup>13</sup> C <sub>4</sub> -PFBA	24 ± 12
<sup>13</sup> C <sub>2</sub> -PFHxA	46 ± 7
<sup>13</sup> C <sub>4</sub> -PFOA	51 ± 9
<sup>13</sup> C <sub>5</sub> -PFNA	48 ± 9
<sup>13</sup> C <sub>2</sub> -PFDA	49 ± 10
<sup>13</sup> C <sub>2</sub> -PFUnDA	53 ± 11
<sup>13</sup> C <sub>2</sub> -PFDoDA	50 ± 12
<sup>18</sup> O <sub>2</sub> -PFHxS	51 ± 8
<sup>13</sup> C <sub>4</sub> -PFOS	47 ± 9
<sup>13</sup> C <sub>3</sub> -HFPO-DA	43 ± 6

<sup>1</sup> Percent absolute recoveries of the internal standards were calculated using the following equations:

$$RF = \frac{\text{area (InjS cal)} \cdot c \text{ (IS cal)}}{\text{area (IS cal)} \cdot c \text{ (InjS cal)}}$$

RF response factor  
 area (InjS cal) peak area of the injection standard 13C8-PFOA in the sample [cps]  
 area (IS cal) peak area of the internal standard in the calibration standard [cps]  
 c (IS cal) concentration of the internal standard in the calibration standard [cps]  
 c (InjS cal) concentration of the injection standard 13C8-PFOA in the calibration standard [cps]

$$\text{absolute IS recovery rate [\%]} = RF \text{ (mean)} \cdot \frac{\text{area (IS sample)}}{\text{area (InjS sample)}} \cdot \frac{c \text{ (InjS sample)}}{c \text{ (IS sample)}} \cdot 100 \%$$

RF (mean) mean response factor of all calibration standards  
 area (IS sample) peak area of the internal standard in the sample [cps]  
 area (InjS sample) peak area of the injection standard 13C8-PFOA in the sample [cps]  
 c (InjS sample) concentration of the injection standard 13C8-PFOA in the sample [cps]  
 c (IS sample) concentration of the internal standard in the sample [cps]

**Table S11: Verification of the method's trueness by analysis of the reference material IRMM-428.** The reference material was produced by the European Commission, Joint Research Center (JRC), Institute for Reference Materials and Measurements (IRMM). Tap drinking water from the Netherlands, spiked with a mixture of PFASs, was characterised by intercomparison among 12 laboratories and 6 certified values and 1 indicative value were assigned. The values given in the certification report were compared to the results derived from analysis of the reference material using the routine method in our laboratory.

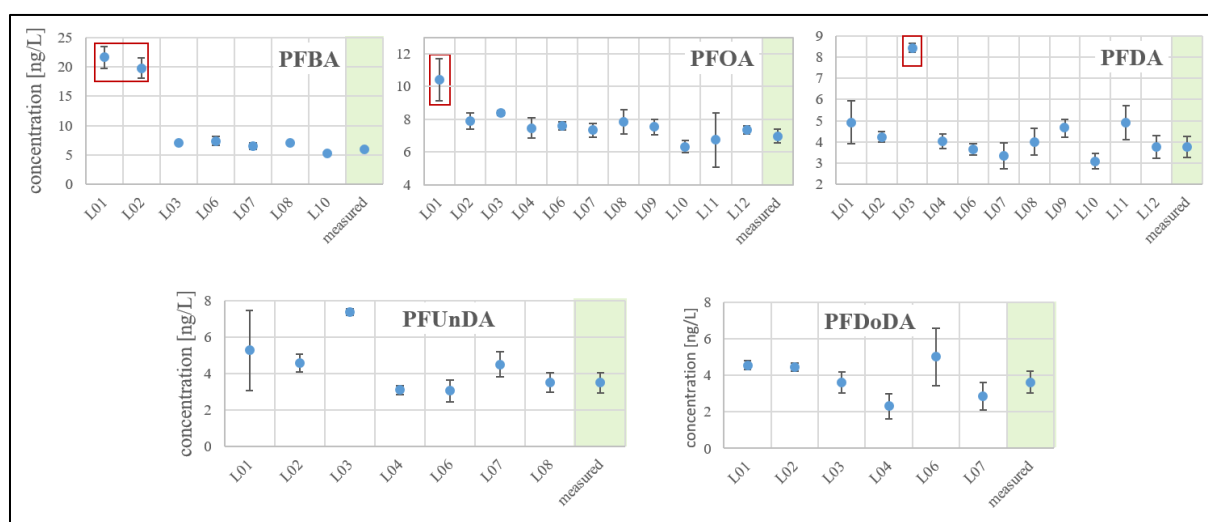
	certified value $\pm U_{Ref}^1$ [ng/L]	measured value $\pm SD^2$ [ng/L]	recovery [%]	z-score <sup>3</sup>
PFPeA	4.0 $\pm$ 1.0	3.7 $\pm$ 0.1	93	-0.57
PFHxA	7.4 $\pm$ 1.0	7.5 $\pm$ 0.2	101	0.15
PFHpA	3.7 $\pm$ 0.7	3.1 $\pm$ 0.1	84	-1.6
PFNA	3.9 $\pm$ 1.4*	3.4 $\pm$ 0.4	87	-0.68
PFBS	5.5 $\pm$ 1.4	6.3 $\pm$ 0.1	115	1.1
PFHxS	3.6 $\pm$ 1.0	3.6 $\pm$ 0.1	101	0.050
L-PFOS	9.6 $\pm$ 1.7	8.8 $\pm$ 1.1	92	-0.71

<sup>1</sup> expanded uncertainty reference sample (confidence level 95 %, k=2)

<sup>2</sup> standard deviation of the measurement series in our laboratory (n=3)

<sup>3</sup> The z-score was calculated using the following equation:

$$z\text{-score} = \frac{(C_{\text{measured}} - C_{\text{certified}})}{\sqrt{\left(\frac{U_{\text{ref}}}{2}\right)^2 + \left(\frac{SD}{\sqrt{3}}\right)^2}}$$



**Figure S4: Comparison of own measured values (marked in green) with laboratory intercomparison results (L01–L12) provided in the certification report for the compounds to which no certified value was assigned. The JRC did not assign values to substances for which the interlaboratory comparison results disagreed significantly without a technical reason (PFBA, PFOA, PFDA; outliers marked in red) and to substances which were not included in homogeneity and stability studies (PFUnDA and PFDoDA). The number of interlaboratory comparison results is different from compound to compound because the number of analytes differed between laboratories. Further information on the reference material can be found in the certification report.<sup>10</sup>**

#### **Section S4: Water transport data used for calculation of PFAS mass transport estimates through Fram Strait**

Reported estimates of water transport budgets in and out of Fram Strait vary significantly due to complex recirculation patterns and strong mesoscale activity that complicate data interpretation.<sup>11-14</sup> Here, we focused on the boundary currents defined in accordance with Stöven et al.<sup>15</sup>

- Atlantic Water (AW) advected in the West Spitsbergen Current (WSC): longitude 5-9°E and depth  $\leq 840$  m
- Polar Water (PW) flowing southward in the EGC, defined as mean temperature  $\leq 1^\circ\text{C}$  and depth  $\leq 400$  m
- Recirculating and Arctic Atlantic Water (RAW/AAW), transported through Fram Strait both due to recirculation of Atlantic Water and the long loop of Atlantic Water through the Arctic Ocean: longitude 7°W to 1°E, depth  $\leq 840$  m, not defined as Polar Water

Stöven et al. assumed the exchange flow across Fram Strait below 840 m to be 0 Sv as there are no connections between the Nordic Seas and the Arctic Ocean below the sill depth of the Greenland-Scotland-Ridge (840 m) other than Fram Strait.<sup>15</sup> No vertical displacements of isopycnals in these two basins are observed indicating a non-zero net transport through Fram Strait below 840 m.<sup>16</sup>

Water mass transport estimates with the AW, PW and RAW/AAW were based on the MIT general circulation model (MITgcm) ECCO v4. The MITgcm has a global resolution of  $1^\circ \times 1^\circ$ , with a finer resolution in the Arctic and near the equator and 50 vertical layers spanning 10 m intervals near the surface and 500 m at the bottom of the ocean. Ocean state estimates are derived from the Estimating the Circulation & Climate of the Ocean (ECCO v4) climatology.<sup>17</sup> The water transport masses reported here are averages over the years 2011-2015. While transport velocities change strongly between seasons and on shorter time scales, there is no strong interannual trend in Fram Strait.<sup>12</sup> Therefore, modeled average volume transport over the period 2011-2015 is assumed representative of the volume transport through Fram Strait in 2018, when the samples for PFAS analysis were taken.

Modeled net transport through Fram Strait with the boundary currents is southward. It is in the same range as estimated from an array of 17 moorings at 78°50'N between 2002 and 2010<sup>15</sup> (see table below).

<b>water mass</b>	<b>volume (Sv)</b>	
	<b>MITgcm</b> (2011-2015)	<b>observations<sup>15</sup></b> (mooring array, 2002-2010)
AW	3.7	4.4( $\pm$ 3.2)
PW	-0.6	-1.4( $\pm$ 0.8)
RAW/AAW	-3.8	-3.5( $\pm$ 1.9)
$\Sigma$	-0.7	-0.5

**Table S12: Detection frequencies [%], concentration ranges [pg/L], mean concentrations [pg/L] and median values [pg/L] of detected PFASs in surface water samples of the entire study area and along a latitudinal transect from the European continent to the Arctic.** Values in brackets are between MDL and MQL. Of the physicochemical parameters, temperature [°C], salinity [PSU] and water depth [m] are given for the different sampling areas.

PFASs <sup>1</sup>	entire study area (n=40)				latitudinal transect from the European continent to the Arctic Ocean (57°N to 79°N at ~5°E)											
					North Sea/continental shelf <i>Norwegian Coastal Current (NCC)</i> (n=5)				Norwegian Sea/continental slope <i>Norwegian Atlantic Current (NwAC)</i> (n=6)				Greenland and Norwegian Sea/deep waters (n=10)			
	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]
HFPO-DA	90	<MDL-120	30	21	100	52-120	92	94	100	(19)-33	25	25	80	<MDL-26	(16)	(17)
PFHxA	100	(17)-110	56	53	100	74-110	96	100	100	33-70	51	51	100	(17)-66	35	30
PFHpA	100	(20)-100	40	32	100	67-100	86	88	100	25-44	33	30	100	(20)-30	23	22
PFOA	100	38-170	66	56	100	120-170	140	140	100	41-76	59	59	100	38-56	47	45
PFNA	100	22-55	34	33	100	45-55	49	49	100	35-44	38	38	100	22-45	31	31
PFDA	68	<MDL-(20)	(8.1)	(8.2)	100	13-(20)	(16)	(16)	100	(11)-(19)	(14)	(14)	100	(7.0)-(16)	(11)	(11)
PFUnDA	83	<MDL-17	(7.5)	(6.8)	100	(9.3)-(13)	(11)	(11)	100	(10)-17	13	13	100	(6.4)-16	(10)	(9.3)
PFDoDA	42	<MDL-(10)	-	-	80	<MDL-6.5	(4.1)	(5.0)	67	<MDL-(10)	(4.9)	(5.8)	60	MDL-(10)	(3.4)	(4.0)
PFBS	100	(15)-93	(39)	(38)	100	57-93	75	69	67	(19)-(39)	(31)	(32)	100	(15)-(28)	(23)	(25)
PFHxS	39	<MDL-54	-	-	100	(38)-(54)	(49)	(52)	100	(19)-(24)	(21)	(22)	0	<MDL	-	-
L-PFOS	<sup>2</sup>	<MDL-110	(42)	(32)	100	60-110	91	100	100	(35)-83	56	58	80	<MDL-(32)	(23)	(27)
Br-PFOS	<sup>2</sup>	<MDL-68	(24)	(19)	100	46-68	57	58	100	(19)-35	(28)	(27)	80	<MDL-(19)	(13)	(15)
ΣPFASs	-	140-850	340	280	-	680-850	770	780	-	270-430	370	390	-	140-290	230	240
physicoch. par.		range	mean	median		range	mean	median		range	mean	median		range	mean	median
temperature [°C]	-	-1.43-14.52	5.48	5.72	-	11.14-14.52	13.21	13.58	-	9.91-13.14	11.63	11.78	-	5.36-8.87	6.91	6.52
salinity [psu]	-	30.00-35.06	33.35	34.78	-	31.78-33.73	32.58	32.41	-	34.69-35.05	34.90	34.90	-	34.86-35.06	35.02	35.03
water depth [m]	-	51-3200	1630	1770	-	51-300	170	150	-	976-1765	1280	1280	-	2450-3200	2840	2850

<sup>1</sup> Due to high blanks, no values can be reported for PFBA. In addition, PFPeA could not be evaluated because of matrix interferences.

<sup>2</sup> PFOS could not be evaluated in 10 of the 40 samples because of matrix interferences. The given values refer to only the samples in which no interferences occurred.

**Table S13: Detection frequencies [%], concentration ranges [pg/L], mean concentrations [pg/L] and median values [pg/L] of detected PFASs in surface water samples along a longitudinal and a latitudinal sampling transect in Fram Strait.** Values in brackets are between MDL and MQL. Of the physicochemical parameters, temperature [°C], salinity [PSU] and water depth [m] are given for the different sampling areas.

PFASs <sup>2</sup>	longitudinal transect across Fram Strait (9°E to 18°W at ~ 79°N)								latitudinal transect along the prime meridian (78°N to 81°N at ~ 0°EW) (n=5)			
	east of 0°EW, ice-free (n=4+1 <sup>1</sup> )				west of 0°EW, ice-covered (n=10)				DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]
	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]
HFPO-DA	100	(14)-45	29	29	80	<MDL	(14)	(12)	100	(15)-70	33	28
PFHxA	100	39-51	44	43	100	52-78	64	63	100	39-75	58	57
PFHpA	100	21-28	24	24	100	36-58	46	46	100	26-54	39	41
PFOA	100	38-48	43	42	100	49-66	57	57	100	61-95	73	71
PFNA	100	22-29	25	24	100	31-38	33	32	100	29-36	32	31
PFDA	60	<MDL-(8.2)	<MDL	(7.0)	0	<MDL	-	-	80	<MDL-8.1	<MDL	(6.9)
PFUnDA	100	(4.9)-(7.1)	(6.1)	(6.4)	30	<MDL-(5.7)	-	-	100	5.3-7.4	(6.0)	(5.8)
PFDoDA	40	<MDL-(3.6)	-	-	0	<MDL	-	-	20	<MDL-5.0	-	-
PFBS	100	(21)-(29)	(24)	(23)	100	(38)-56	(43)	(40)	100	23-64	53	59
PFHxS	0	<MDL	-	-	20	<MDL-(24)	-	-	60	<MDL-27	<MDL	(23)
L-PFOS	<sup>3a</sup>	(24)-(33)	(29)	(29)	<sup>3b</sup>	<MDL-(37)	(22)	(28)	<sup>3c</sup>	25-35	(31)	34
Br-PFOS	<sup>3a</sup>	(12)-(19)	(16)	(17)	<sup>3b</sup>	<MDL-(13)	<MDL	(13)	<sup>3c</sup>	15-21	(18)	(17)
ΣPFASs (DF>50%) <sup>4</sup>	-	170-210	190	190	-	230-290	260	260	-	190-310	260	280
physicoch. par.		range	mean	median		range	mean	median		range	mean	median
temperature [°C]	-	5.48-6.31	6.00	6.12	-	-1.43-0.45	-0.6	-0.44	-	-1.26-4.41	0.69	-0.87
salinity [PSU]	-	34.86-35.03	34.95	34.93	-	30.00-31.50	30.55	30.42	-	31.74-34.44	32.79	32.41
water depth [m]	-	220-2500	920	480	-	78-2600	558	235	-	2600-3200	2900	3000

<sup>1</sup> At sampling location N21/F5, the latitudinal transect from the European continent to the Arctic and the longitudinal transect across Fram Strait crossed. Consequently, this sample was used for evaluation of both sampling transects.

<sup>2</sup> Due to high blanks, no values can be reported for PFBA. In addition, PFPeA could not be evaluated because of matrix interferences.

<sup>3a-c</sup> PFOS could not be evaluated in (a) two, (b) seven and (c) two of the samples because of matrix interferences. The given values refer to only the samples in which no interferences occurred.

<sup>4</sup> ΣPFASs only includes the compounds with a detection frequency >50% in samples of the longitudinal sampling transect across Fram Strait (C6-C9 PFCAs, PFBS and HFPO-DA).



**Table S14: Concentrations [pg/L] of detected PFASs in surface seawater samples taken along a latitudinal transect from the European continent to the Arctic (57°N to 79°N at ~5°E).**  
 Values in brackets are between MDL and MQL.

station ID	HFPO-DA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	L-PFOS	Br-PFOS	ΣPFASs
N1	86	74	84	150	48	(16)	(12)	(6.0)	69	(52)	66	46	710
N2	94	110	88	140	45	(13)	(9.3)	<MDL	93	(50)	110	68	820
N3	52	110	67	120	49	(18)	13	(6.5)	57	(38)	100	53	680
N4	100	100	100	170	55	(20)	(9.6)	(5.0)	69	(54)	110	60	850
N5 <sup>1</sup>	120±8	93±5	92±2	140±10	49±2	(14±3)	(11±1)	<MDL	88±9	(53±3)	60±11	58±2	780±30
N6	33	70	44	76	39	(13)	(11)	<MDL	(39)	(23)	58	(27)	430
N7	23	53	28	50	37	(11)	14	(6.2)	(38)	(22)	83	35	400
N8	21	40	29	57	37	(13)	15	(7.5)	(38)	(22)	57	(27)	360
N9	26	58	32	30	35	(14)	(11)	(5.4)	(26)	(24)	60	(27)	380
N10	(19)	33	25	41	39	(15)	(10)	<MDL	(19)	(18)	(35)	(19)	270
N11	29	49	39	74	44	(19)	17	(10)	(25)	(19)	(40)	30	390
N12	26	31	22	47	33	(14)	(12)	(5.5)	(15)	<MDL	(32)	(15)	250
N13	21	29	30	55	45	(16)	16	(10)	(25)	<MDL	(28)	(15)	290
N14	(15)	44	26	53	36	(14)	(10)	(6.2)	(27)	<MDL	(29)	(19)	280
N15	(14)	(26)	24	42	31	(12)	(10)	(4.4)	(25)	<MDL	(27)	(15)	230
N16	<MDL	29	21	39	27	(10)	(6.5)	<MDL	(24)	<MDL	<MDL	<MDL	160
N17	<MDL	(17)	(20)	42	23	(8.8)	(8.2)	<MDL	(21)	<MDL	<MDL	<MDL	140
N18	(15)	(25)	(20)	43	26	(9.2)	(8.5)	<MDL	(28)	<MDL	(29)	(18)	220
N19	24	33	(20)	53	31	(12)	(11)	(4.4)	(25)	<MDL	(27)	(18)	260
N20	(20)	66	23	56	35	(9.7)	(6.8)	<MDL	(21)	<MDL	(30)	(13)	280
N21	21	51	24	37	22	(7.0)	(6.4)	(3.6)	(22)	<MDL	(24)	(19)	240

<sup>1</sup> Mean±SD is given for this sample, as a triplicate of the sample was analyzed.

**Table S15: Concentrations [pg/L] of detected PFASs in surface seawater samples taken along a longitudinal transect across Fram Strait (9°E to 18°W at ~79°N).** Values in brackets are between MDL and MQL.

station ID	HFPO-DA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	L-PFOS	Br-PFOS	ΣPFASs(DF>50%) <sup>2</sup>
F1	45	41	21	45	24	(7.1)	(6.7)	(3.6)	(29)	<MDL	(31)	(18)	205.8
F2	(14)	43	24	41	24	<MDL	(5.3)	<MDL	(25)	<MDL	(27)	(12)	171.0
F3	35	43	22	41	28	(8.2)	(7.1)	<MDL	(21)	<MDL	(33)	(16)	190.4
F4	29	39	28	48	29	<MDL	(5.0)	<MDL	(23)	<MDL	na <sup>1</sup>	na	196.2
F5(=N21)	21	51	24	37	22	(7.0)	(6.4)	(3.6)	(22)	<MDL	(24)	(19)	177.5
F6	<MDL	71	36	63	31	<MDL	<MDL	<MDL	56	<MDL	na	na	258.4
F7	(12)	67	37	48	32	<MDL	<MDL	<MDL	(38)	<MDL	na	na	234.5
F8	(11)	63	45	50	34	<MDL	<MDL	<MDL	(39)	(19)	na	na	241.6
F9	(11)	55	49	58	32	<MDL	<MDL	<MDL	(39)	<MDL	na	na	243.5
F10	<MDL	58	44	56	32	<MDL	<MDL	<MDL	(39)	<MDL	na	na	228.0
F11	38	62	47	54	33	<MDL	(5.4)	<MDL	(44)	<MDL	na	na	278.5
F12	(13)	78	46	58	35	<MDL	<MDL	<MDL	(40)	<MDL	na	na	269.3
F13	(13)	64	49	57	38	<MDL	(5.7)	<MDL	52	(24)	<MDL	<MDL	272.0
F14	(16)	52	54	65	32	<MDL	(4.8)	<MDL	(38)	<MDL	(37)	(13)	257.5
F15	24	69	58	64	32	<MDL	<MDL	<MDL	45	<MDL	(28)	(13)	291.5

<sup>1</sup>na: not analyzable (PFOS could not be evaluated in these samples because of matrix interferences)

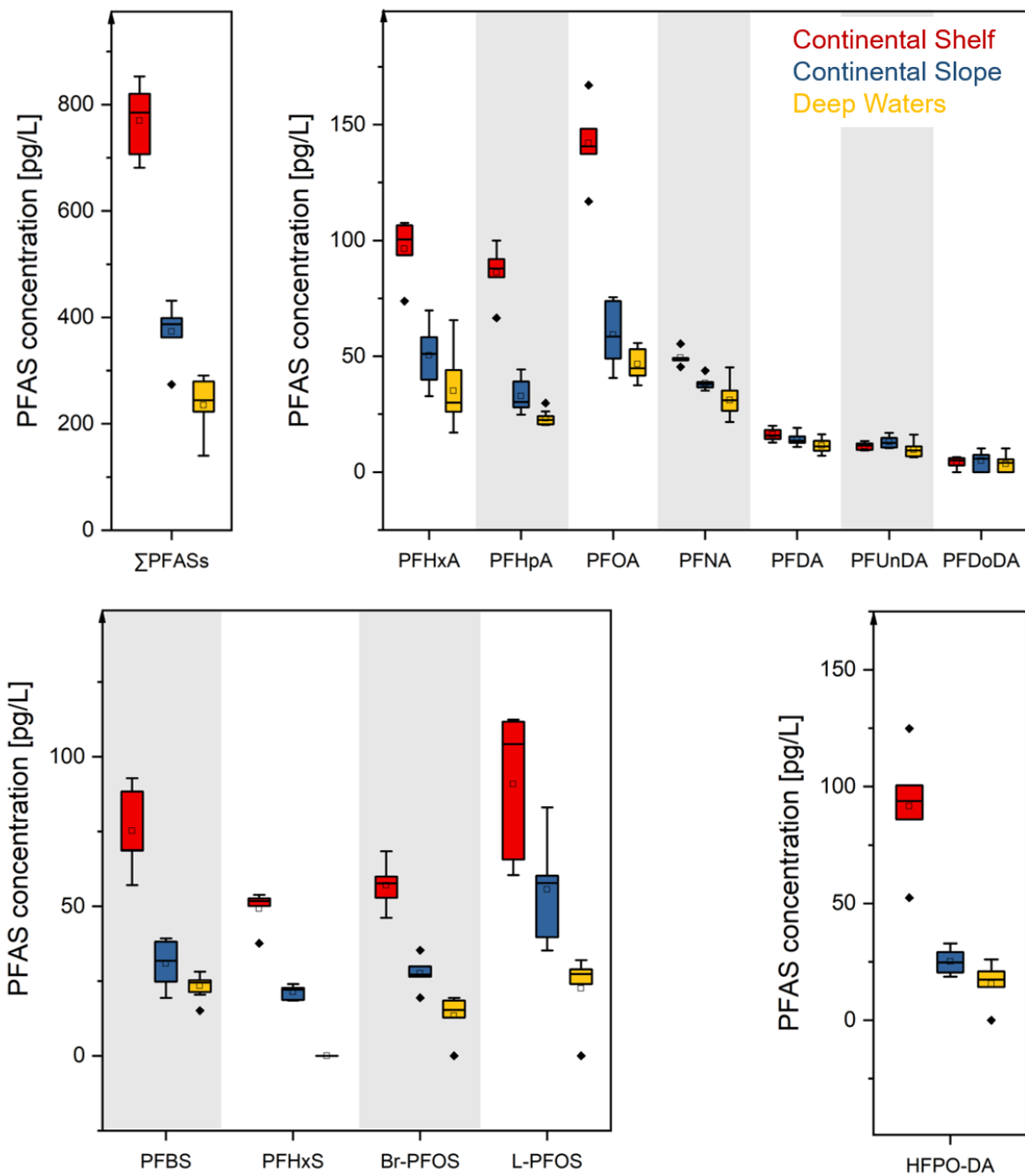
<sup>2</sup>ΣPFASs only includes the compounds with a detection frequency >50% in samples of the longitudinal sampling transect across Fram Strait (C6-C9 PFCAs, PFBS and HFPO-DA).

**Table S16: Concentrations [pg/L] of detected PFASs in surface seawater samples taken along a latitudinal transect along the prime meridian (78 °N to 81 °N at ~ 0°EW).** Values in brackets are between MDL and MQL.

station ID	HFPO-DA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	L-PFOS	Br-PFOS	ΣPFASs(DF>50%) <sup>2</sup>
P1	(15)	39	26	61	29	<MDL	(5.3)	<MDL	(23)	<MDL	(25)	(15)	193.3
P2	28	50	26	64	31	(8.1)	(7.4)	(5.0)	58	<MDL	(34)	(17)	228.9
P3	22	68	41	71	30	(6.9)	(5.3)	<MDL	64	(27)	na <sup>1</sup>	na	296.0
P4	31	75	54	91	33	(7.5)	(6.1)	<MDL	59	(25)	na	na	314.5
P5	70	57	48	77	36	(6.5)	(5.8)	<MDL	60	(23)	(35)	(21)	277.4

<sup>1</sup>na: not analyzable (PFOS could not be evaluated in these samples because of matrix interferences)

<sup>2</sup>ΣPFASs only includes the compounds with a detection frequency >50% in samples of the longitudinal sampling transect across Fram Strait (C6-C9 PFCAs, PFBS and HFPO-DA).

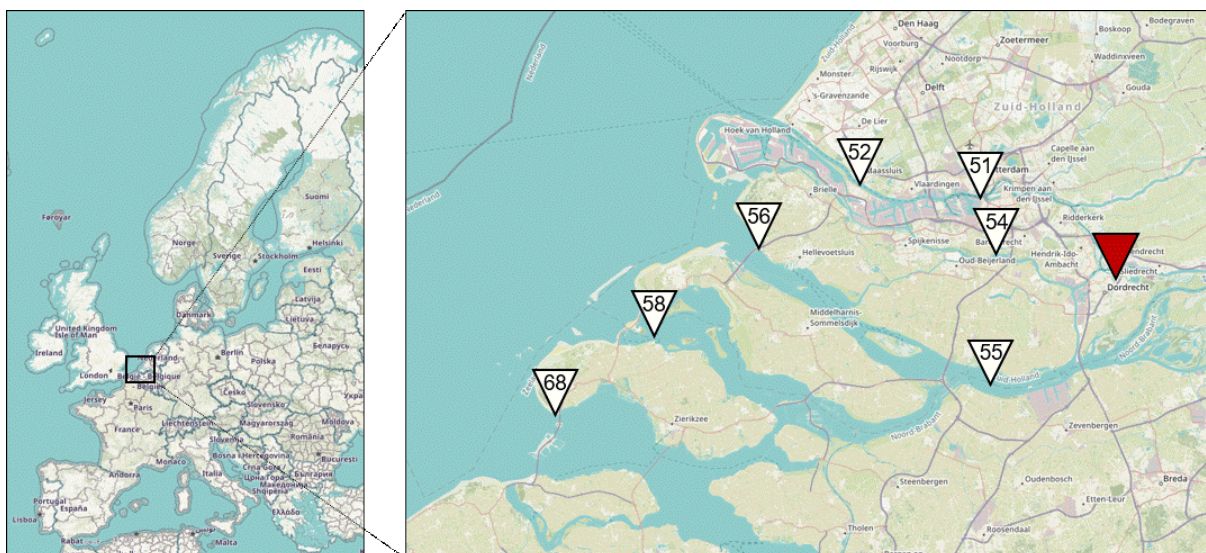


**Figure S5: Boxplots showing PFAS concentrations in surface water samples from the North Sea continental shelf (samples N1-N5, red), the continental slope (N6 to N11, blue) and deep water regions of the Norwegian Sea and Greenland Sea (N12 to N21, yellow). The box represents the 25% to 75% quartile, the median is plotted by the band inside the box and the mean by the blank square. The ends of the whiskers display the lowest and the highest concentration still within 1.5 IQR of the lower and higher quartile. Outliers are plotted with a black diamond.**

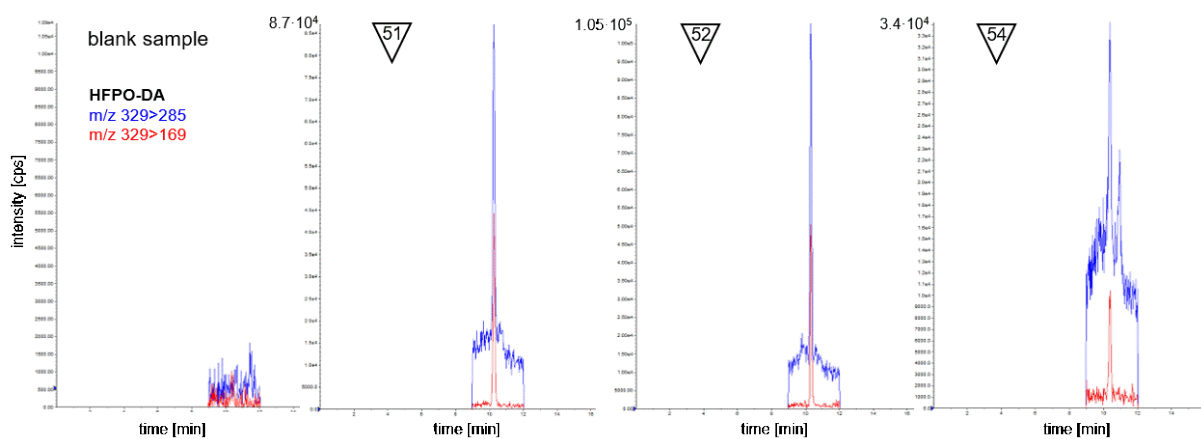
### **Section S5: Potential sources of HFPO-DA to European coastal waters**

A fluoropolymer manufacturing plant in the Rhine-Meuse delta is assumed to be a major source of HFPO-DA in European coastal areas.<sup>18-20</sup> The manufacturer started to replace PFOA by HFPO-DA in the United States in 2005. No information is available on when HFPO-DA production and use started at the plant in the Rhine-Meuse delta, but PFOA emissions decreased strongly from 2002 on and ceased in 2012.<sup>21</sup> Reanalysis of water sample extracts from the Rhine-Meuse delta showed that the compound had been present in 2008 already (Figures S6 and S7). This indicates that at least since then, there have been HFPO-DA emissions into the European environment.

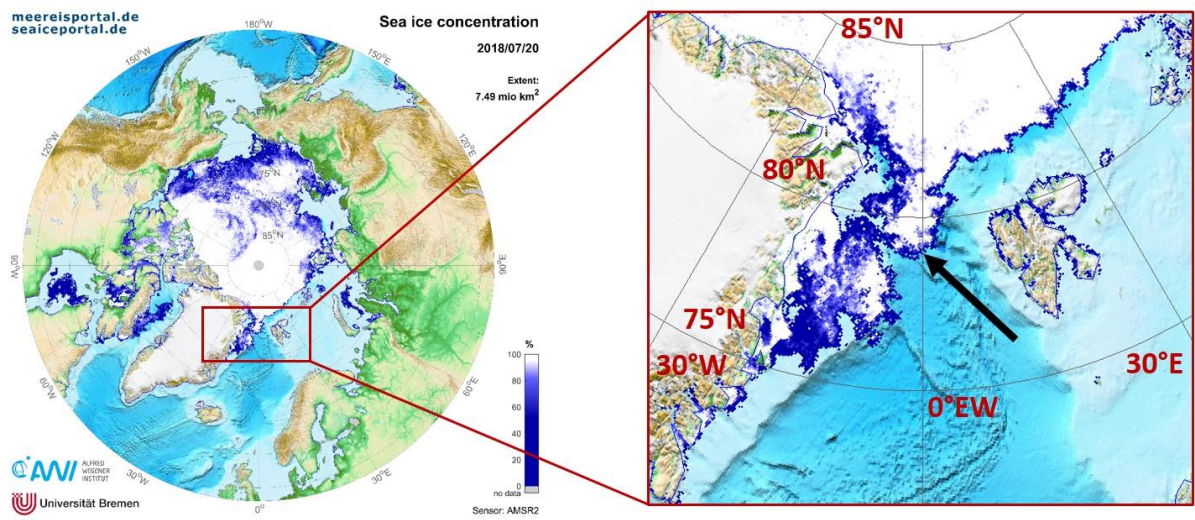
The emission permit of the fluoropolymer manufacturer in the Netherlands of HFPO-DA to surface water was 6400 kg/year from 2012 to 2017, before being lowered to 2035 kg/year in 2017 and to 148 kg/year in 2019.<sup>21</sup> HFPO-DA concentrations up to 812 ng/L were reported for river water samples taken downstream of the chemical park in 2016.<sup>20</sup> It can be hypothesized that HFPO-DA is transported from the Rhine-Meuse delta to North Sea coastal waters, from where radionuclide tracer studies suggest transit times of 2-3 years to Fram Strait.<sup>22</sup>



**Figure S6: Sampling locations of reanalyzed samples, taken in the Rhine-Meuse delta in 2008 by Möller et al.<sup>23</sup>** The red triangle shows the fluoropolymer manufacturing plant, which is assumed to be a major source of HFPO-DA in European coastal areas. The replacement compound HFPO-DA has not yet been under discussion in 2008 and was not included in the analytical method. Results for legacy PFASs can be found in reference <sup>23</sup>. The basic map was taken from Openstreetmap (CC-BY-SA 2.0).



**Figure S7: LC-MS/MS chromatograms of the replacement compound HFPO-DA in sample extracts from 2008, reanalyzed using the current instrumental method.** The extracts had been stored in brown-glass vials at  $-20\text{ }^{\circ}\text{C}$  for ten years. Before instrumental analysis, methanol was topped on to a volume of approximately  $200\text{ }\mu\text{L}$ . The vials were put into an ultrasonic bath for 30 min and vortexed for 5 min. Chromatograms are shown for the blank sample and the samples in which HFPO-DA was detected. In samples 55, 56, 58 and 68, HFPO-DA was not detected.



**Figure S8: Sea ice concentration in the Arctic during the cruise.**<sup>24</sup> The black arrow marks the location at which the longitudinal sampling transect in Fram Strait (~79°N) cuts across the ice edge near the prime meridian (0°EW).

**Table S17: Pearson correlation coefficients r between physicochemical parameters and PFASs as well as among individual PFASs in surface water samples taken along a longitudinal transect across Fram Strait (9°E to 18°W at ~79°N). Statistically significant values are marked with \* (p<0.05). Correlation analysis was conducted only for compounds with a detection frequency >50% (C6-C9 PFCAs, PFBS and HFPO-DA).**

	temperature	pH	salinity	PFHxA	PFHpA	PFOA	PFNA	PFBS	HFPO-DA	ΣPFASs(DF>50%)	PFHpA/PFNA
<b>temperature</b>	1										
<b>pH</b>	0.82*	1									
<b>salinity</b>	0.98*	0.85*	1								
<b>PFHxA</b>	-0.82*	-0.66*	-0.76*	1							
<b>PFHpA</b>	-0.90*	-0.91*	-0.91*	0.67*	1						
<b>PFOA</b>	-0.82*	-0.79*	-0.78*	0.62*	0.86*	1					
<b>PFNA</b>	-0.83*	-0.85*	-0.82*	0.70*	0.79*	0.71*	1				
<b>PFBS</b>	-0.87*	-0.70*	-0.80*	0.81*	0.73*	0.82*	0.75*	1			
<b>HFPO-DA</b>	0.56	0.42	0.52*	-0.52*	-0.38	-0.40	-0.39	-0.49	1		
<b>ΣPFASs(DF&gt;50%)</b>	-0.88*	-0.84*	-0.85*	0.80*	0.89*	0.86*	0.83*	0.87*	-0.21	1	
<b>PFHpA/PFNA</b>	-0.79*	-0.83*	-0.80*	0.54*	0.94*	0.78*	0.52*	0.58*	-0.33	0.79*	1

**Table S18: Detection frequencies [%], concentration ranges [pg/L], mean concentrations [pg/L] and median values [pg/L] of detected PFASs in all samples and in different water masses (Atlantic Water (AW), Polar Surface Water (PSW)) of the seven depth profiles taken in Fram Strait. Values in brackets are between MDL and MQL.**

PFASs <sup>1</sup>	all samples (n=58)				Atlantic Water (AW) (n=18)				Polar Surface Water (PSW) (n=16)			
	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]
<b>HFPO-DA</b>	21	<MDL-(17)	-	-	39	<MDL-(14)	-	-	19	<MDL-(12)	-	-
<b>PFHxA</b>	91	<MDL-74	37	39	100	(25)-54	41	40	100	41-74	54	53
<b>PFHpA</b>	81	<MDL-56	24	24	100	(14)-32	25	26	100	26-56	42	44
<b>PFOA</b>	<sup>2a</sup>	<MDL-120	49	50	<sup>2b</sup>	44-72	55	52	<sup>2c</sup>	48-120	76	60
<b>PFNA</b>	91	<MDL-51	25	26	100	(19)-39	31	29	100	(21)-51	31	28
<b>PFDA</b>	24	<MDL-(21)	-	-	56	<MDL-(16)	(7.8)	13	25	<MDL-(21)	-	-
<b>PFUnDA</b>	66	<MDL-15	(5.6)	(6.6)	78	<MDL-15	(7.2)	(8.0)	69	<MDL-14	(6.0)	(6.7)
<b>PFBS</b>	57	<MDL-50	16	17	61	<MDL-(34)	(13)	(16)	100	(21)-50	(36)	(37)
<b>PFHxS</b>	34	<MDL-(29)	-	-	17	<MDL-(28)	-	-	75	<MDL-(29)	(17)	(22)
<b>L-PFOS</b>	36	<MDL-44	-	-	83	<MDL-44	(22)	(23)	19	<MDL-(33)	-	-
<b>Br-PFOS</b>	43	<MDL-33	-	-	83	<MDL-27	(13)	(15)	38	<MDL-33	-	-
<b>ΣPFASs(DF&gt;50%)<sup>3</sup></b>	-	<MDL-230	110	110	-	64-140	120	120	-	120-230	170	170

<sup>1</sup> Due to high blanks, no values can be reported for PFBA. In addition, PFPeA could not be evaluated because of matrix interferences.

<sup>2</sup> PFOA could not be evaluated in a) 29, b) 6, c) 11 of the samples because of matrix interferences. The given values refer to only the samples in which no interferences occurred.

<sup>3</sup> ΣPFASs only includes the compounds with a detection frequency >50% in all depth profiles samples (C6, C7, C9, C11 PFCAs and PFBS).



**Table S19: Detection frequencies [%], concentration ranges [pg/L], mean concentrations [pg/L] and median values [pg/L] of detected PFASs in different water masses (Recirculating Atlantic Water/Arctic Atlantic Water (RAW/AAW), Intermediate Waters (IW) and Deep Waters (DW)) of the seven depth profiles taken in Fram Strait. Values in brackets are between MDL and MQL.**

PFASs <sup>1</sup>	Recirculating Atlantic Water/Arctic Atlantic Water (RAW/AAW) (n=10)				Intermediate Waters (IW) (n=6)				Deep Waters (DW) (n=8)			
	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]	DF [%]	range [pg/L]	mean [pg/L]	median [pg/L]
<b>HFPO-DA</b>	20	<MDL-(17)	-	-	0	<MDL	-	-	0	<MDL	-	-
<b>PFHxA</b>	100	(22)-66	34	30	100	(14)-43	28	28	38	<MDL-(20)	-	-
<b>PFHpA</b>	90	<MDL-46	(21)	(20)	50	<MDL-19	8.5	8.0	13	<MDL-(20)	-	-
<b>PFOA</b>	<sup>2a</sup>	36-94	54	48	<sup>2b</sup>	34-50	40	37	<sup>2c</sup>	<MDL	-	-
<b>PFNA</b>	100	(14)-34	22	(20)	100	(16)-26	(21)	20	38	<MDL-(19)	-	-
<b>PFDA</b>	0	<MDL	-	-	0	<MDL	-	-	0	<MDL	-	-
<b>PFUnDA</b>	70	<MDL-(10)	(5.1)	(6.1)	67	<MDL-13	(6.1)	(7.1)	25	<MDL-(7.3)	-	-
<b>PFBS</b>	30	<MDL-(33)	-	-	50	<MDL-(25)	(10)	(7.0)	0	<MDL	-	-
<b>PFHxS</b>	40	<MDL-(23)	-	-	17	<MDL-(23)	-	-	0	<MDL	-	-
<b>L-PFOS</b>	10	<MDL-(33)	-	-	33	<MDL-(36)	-	-	0	<MDL	-	-
<b>Br-PFOS</b>	20	<MDL-31	-	-	33	<MDL-(17)	-	-	0	<MDL	-	-
<b>ΣPFASs(DF&gt;50%)</b>	-	60-180	90	76	-	40-120	73	73	-	<MDL-40	16	13

<sup>1</sup> Due to high blanks, no values can be reported for PFBA. In addition, PFPeA could not be evaluated because of matrix interferences.

<sup>2</sup> PFOA could not be evaluated in a) 4, b) 3, c) 4 of the samples because of matrix interferences. The given values refer to only the samples in which no interferences occurred.

<sup>3</sup> ΣPFASs only includes the compounds with a detection frequency >50% in all depth profiles samples (C6, C7, C9, C11 PFCAs and PFBS).

**Table S20: Concentrations [pg/L] of detected PFASs in depth profiles.** Values in brackets are between MDL and MQL.

sample name	sampling depth	water mass	HFPO-DA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFBS	PFHxS	L-PFOS	Br-PFOS	ΣPFASs(DF>50%) <sup>2</sup>
V1/1	1239	DW	<MDL	<MDL	<MDL	na <sup>1</sup>	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	0.0
V1/2	1000	DW	<MDL	(20)	(20)	na	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	40.0
V1/3	500	AW	<MDL	(25)	(21)	na	(19)	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	64.7
V1/4	250	AW	<MDL	33	24	na	24	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	81.7
V1/5	100	AW	<MDL	28	26	na	23	<MDL	<MDL	(34)	<MDL	<MDL	<MDL	110.5
V1/6	75	AW	<MDL	41	24	na	34	(14)	(8.1)	<MDL	<MDL	(26)	(17)	106.8
V1/7	50	AW	<MDL	36	29	na	28	(13)	(6.4)	<MDL	<MDL	(23)	(16)	99.2
V1/8	10	AW	<MDL	32	26	na	29	(15)	(8.1)	(20)	<MDL	(21)	(13)	114.7
V2/1	2286	DW	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	(7.1)	<MDL	<MDL	<MDL	<MDL	7.1
V2/2	1000	IW	<MDL	30	<MDL	37	26	<MDL	13	(21)	<MDL	<MDL	<MDL	89.4
V2/3	500	AW	<MDL	35	(14)	44	35	<MDL	(9.2)	(18)	<MDL	(28)	(16)	110.6
V2/4	250	AW	<MDL	53	21	52	34	(13)	(8.0)	(16)	<MDL	(24)	(13)	132.7
V2/5	100	AW	<MDL	43	31	50	39	(16)	14	(15)	<MDL	(21)	(16)	141.9
V2/6	75	AW	<MDL	50	32	67	31	(14)	(11)	<MDL	<MDL	(22)	(13)	124.3
V2/7	40	AW	(11)	39	24	55	34	(14)	(10)	<MDL	<MDL	(27)	(13)	107.4
V2/8	10	AW	(9.9)	54	28	60	38	(15)	(15)	<MDL	<MDL	(44)	(27)	135.6
V3/1	2573	DW	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	0.0
V3/2	1001	IW	<MDL	37	(15)	34	(18)	<MDL	(9.8)	<MDL	<MDL	(36)	(17)	80.2
V3/3	501	AW	<MDL	32	(19)	48	28	<MDL	(10)	(19)	<MDL	(21)	(13)	107.6
V3/4	250	AW	(14)	48	29	52	38	(14)	(12)	(15)	<MDL	(21)	(15)	142.5
V3/5	100	AW	(14)	45	27	48	29	<MDL	<MDL	(21)	(21)	(23)	(16)	122.8
V3/6	75	AW	(13)	47	32	72	29	<MDL	(6.6)	(21)	(20)	(22)	(18)	135.2
V3/7	50	AW	(11)	53	23	62	27	<MDL	(6.6)	(28)	(28)	(23)	(16)	138.9
V3/8	10	AW	(8.0)	39	26	46	29	(13)	(5.0)	(30)	<MDL	(40)	(20)	129.6
V4/1	2600	DW	<MDL	<MDL	<MDL	na	(19)	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	19.2
V4/2	2000	DW	<MDL	(14)	<MDL	na	(12)	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	26.4
V4/3	1200	IW	<MDL	(14)	<MDL	na	(18)	<MDL	<MDL	(14)	<MDL	<MDL	<MDL	46.2
V4/4	1000	IW	<MDL	(26)	(19)	na	21	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	65.8
V4/5	700	RAW/AAW	<MDL	(22)	(15)	na	(20)	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	56.7
V4/6	500	RAW/AAW	<MDL	(23)	(18)	na	25	<MDL	(10)	<MDL	<MDL	<MDL	<MDL	75.5
V4/7	250	RAW/AAW	<MDL	28	(19)	na	22	<MDL	(7.8)	<MDL	<MDL	<MDL	<MDL	76.6
V4/8	100	PW	<MDL	41	26	na	25	<MDL	(7.2)	(23)	(22)	<MDL	<MDL	122.8
V4/9	75	PW	<MDL	48	28	na	27	<MDL	(6.3)	(21)	(24)	(22)	(17)	130.3
V4/10	60	PW	<MDL	51	41	na	26	<MDL	(5.6)	(38)	(25)	<MDL	<MDL	161.8
V4/11	40	PW	<MDL	56	40	na	28	<MDL	(9.0)	(32)	(23)	<MDL	<MDL	165.6
V4/12	15	PW	<MDL	59	45	na	36	<MDL	(6.1)	(24)	(23)	<MDL	<MDL	169.5
V4/13	5	PW	<MDL	42	56	na	44	(14)	13	(31)	<MDL	<MDL	<MDL	186.2
V5/1	959	IW	<MDL	(16)	<MDL	na	(16)	<MDL	(4.9)	<MDL	<MDL	<MDL	<MDL	37.7
V5/2	500	RAW/AAW	<MDL	(25)	(16)	na	(18)	<MDL	(5.7)	<MDL	<MDL	<MDL	<MDL	65.2

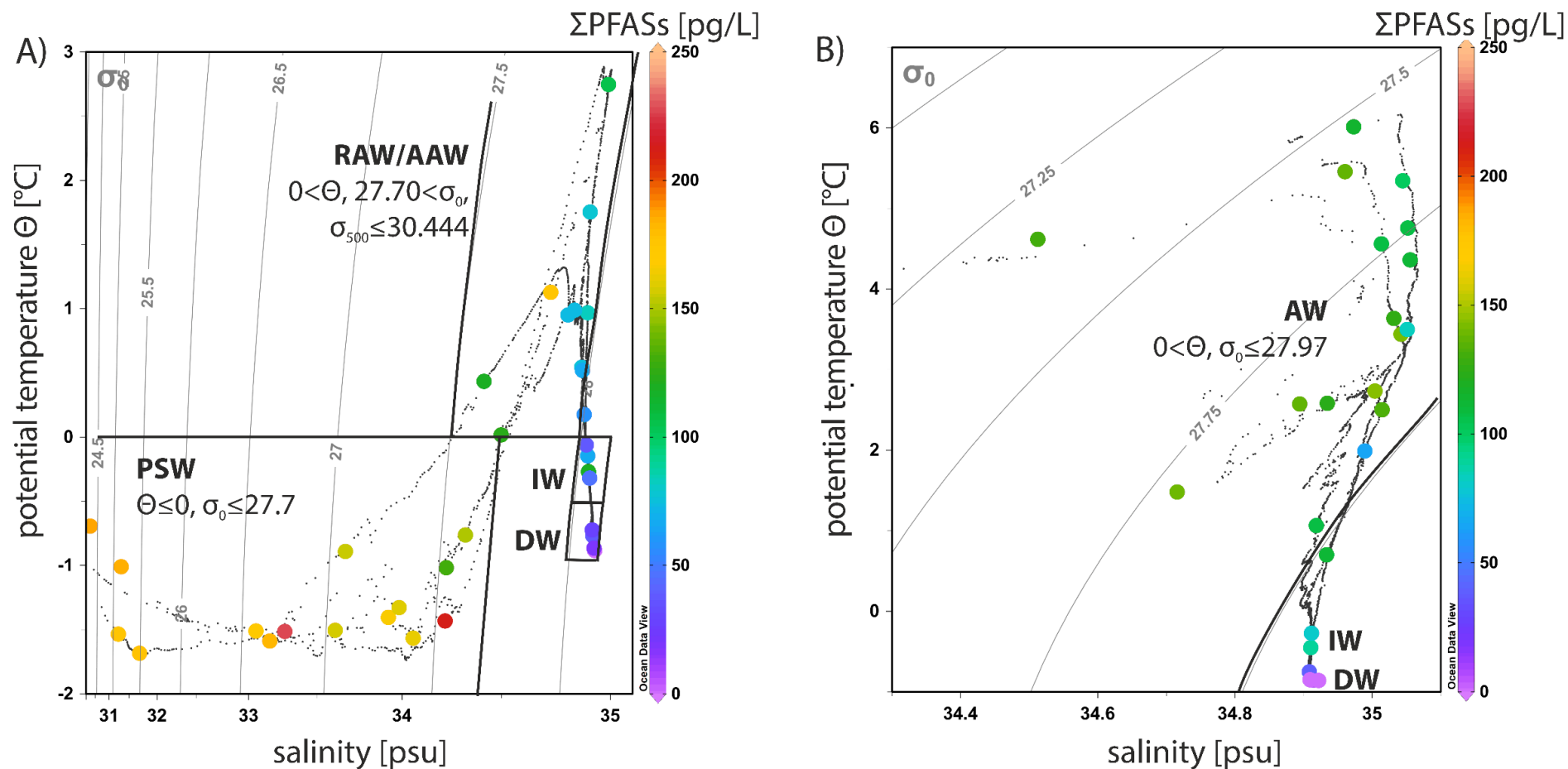
V5/3	250	RAW/AAW	<MDL	(24)	22	na	(20)	<MDL	(7.5)	<MDL	(19)	<MDL	<MDL	73.8
V5/4	100	PW	<MDL	48	31	na	26	<MDL	(8.6)	(36)	(19)	<MDL	<MDL	148.8
V5/5	75	PW	<MDL	52	41	na	(21)	<MDL	<MDL	47	(18)	<MDL	<MDL	160.0
V5/6	50	PW	<MDL	46	50	na	24	<MDL	<MDL	(38)	(25)	<MDL	<MDL	157.5
V5/7	26	PW	<MDL	54	51	na	34	<MDL	(7.7)	(35)	(22)	<MDL	<MDL	182.1
V5/8	5	PW	<MDL	44	46	na	51	(21)	(14)	(28)	(25)	<MDL	<MDL	183.8
V6/1	271	RAW/AAW	<MDL	36	<MDL	36	(14)	<MDL	<MDL	(23)	<MDL	<MDL	<MDL	72.8
V6/2	150	RAW/AAW	(8.4)	44	23	39	(19)	<MDL	<MDL	(33)	<MDL	<MDL	(20)	117.6
V6/3	100	PW	(12)	57	32	48	21	<MDL	<MDL	(44)	<MDL	<MDL	(17)	153.8
V6/4	50	PW	(9.2)	61	47	60	27	<MDL	<MDL	(42)	<MDL	<MDL	(17)	177.7
V6/5	10	PW	(9.8)	56	47	51	28	<MDL	<MDL	(43)	<MDL	<MDL	(12)	174.0
V7/1	3117	DW	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	0.0
V7/2	2000	DW	<MDL	(15)	<MDL	<MDL	(14)	<MDL	(7.3)	<MDL	<MDL	<MDL	<MDL	36.4
V7/3	1000	IW	<MDL	43	(17)	50	24	<MDL	(9.2)	(25)	(23)	(22)	(13)	118.7
V7/4	500	RAW/AAW	<MDL	33	22	48	(20)	<MDL	(6.5)	<MDL	(20)	<MDL	<MDL	81.6
V7/5	250	RAW/AAW	(17)	36	32	52	31	<MDL	(5.6)	<MDL	(20)	<MDL	<MDL	105.0
V7/6	100	RAW/AAW	<MDL	66	46	94	34	<MDL	(7.7)	(21)	(23)	(33)	31	175.0
V7/7	50	PW	<MDL	74	43	120	39	(12)	(9.4)	45	(29)	(30)	(26)	210.9
V7/8	10	PW	<MDL	73	55	110	41	(11)	(8.2)	50	(19)	(33)	33	226.1

<sup>1</sup>na: not analyzable (PFOA could not be evaluated these samples because of matrix interferences)

<sup>2</sup>ΣPFASs only includes the compounds with a detection frequency >50% in all depth profiles samples (C6, C7, C9, C11 PFCA and PFBS).

**Table S21: Water mass definitions used for classification of water samples taken for this study, adapted from Rudels et al.<sup>5</sup>** Boundaries of potential temperature  $\Theta$  [ $^{\circ}\text{C}$ ] and potential density  $\sigma$  [ $\text{kg}/\text{m}^3$ ] are given.  $\sigma_0$  and  $\sigma_{500}$  is potential density at reference pressure 0 m (sea surface) and 500 m. Deviating from the definition by Rudels et al.<sup>5</sup>, water with a potential density  $\sigma$  lower than  $27.70 \text{ kg}/\text{m}^3$  is included in the definition of Atlantic Water to also classify surface water in the West Spitsbergen Current as Atlantic Water. Moreover, our water mass definitions do not differentiate between Recirculating Atlantic Water and Arctic Atlantic Water and between different classes of Intermediate Water and Deep Water.

<b>Water mass (acronym)</b>	<b>Definition</b>
Atlantic Water (AW)	$0 < \Theta, \sigma_0 \leq 27.97$
Polar Surface Water (PSW)	$\Theta \leq 0, \sigma_0 \leq 27.70$
Recirculating and Arctic Atlantic Water (RAW/AAW)	$0 < \Theta, 27.70 < \sigma_0, \sigma_{500} \leq 30.444$
Intermediate Water (IW)	$\Theta \leq 0, 27.97 < \sigma_0, \sigma_{500} \leq 30.444$
Deep Water (DW)	$\Theta \leq 0, 30.444 \leq \sigma_{500}$



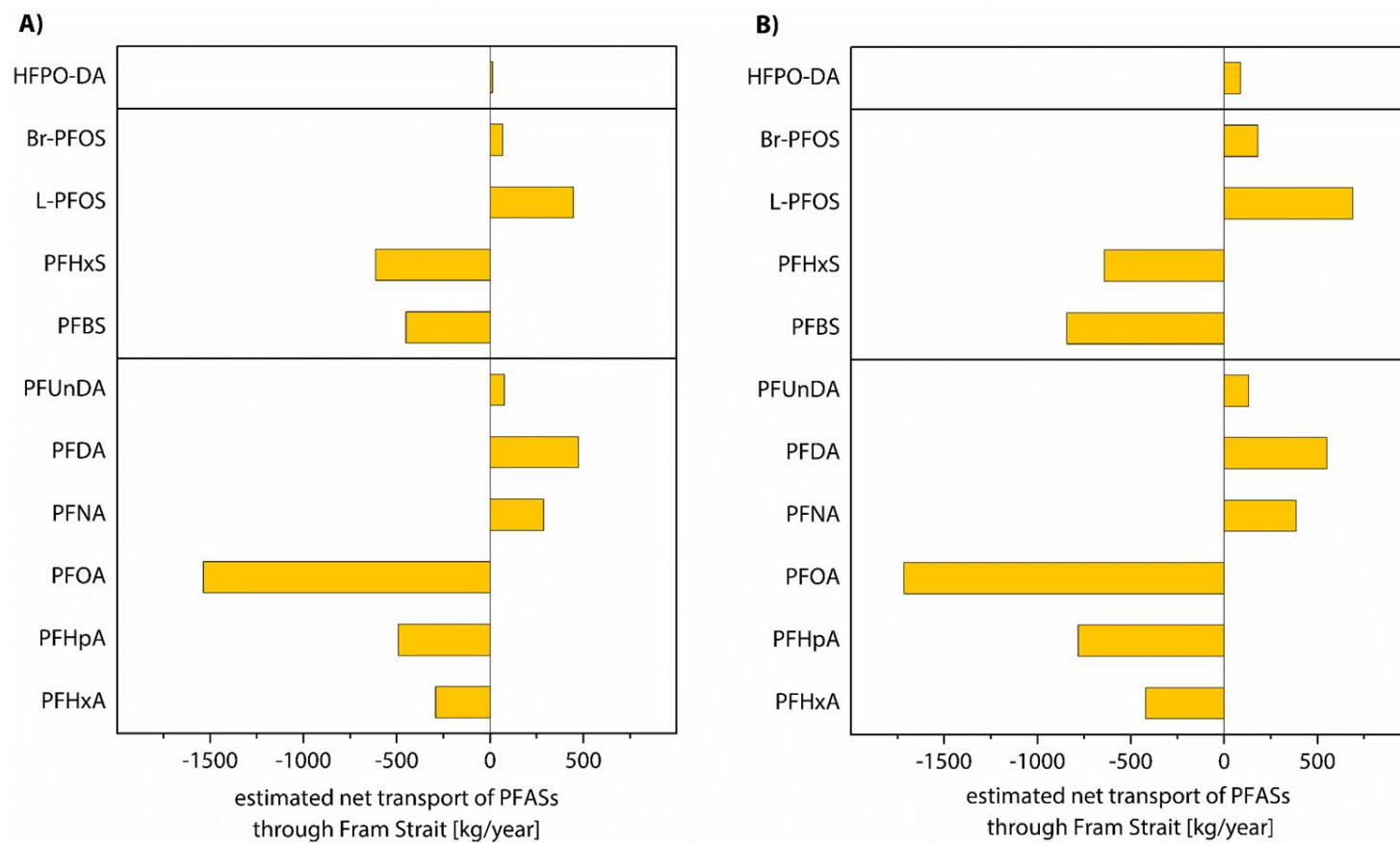
**Figure S9:** Potential temperature  $\theta$  [°C] versus salinity [psu] plots for A) CTD/rosette casts taken west of the surface front between inflowing Atlantic Water (AW) and outflowing Polar Surface Water (PSW) (~79°N) and B) CTD/rosette casts taken east of it. Note that in order to show the vast range of salinity values, the x-scale in A) is non-linear and in B) it is a zoom on the Atlantic water masses that is linear. Temperature and salinity data was taken from references <sup>4</sup> and <sup>3</sup>. Samples analyzed for PFASs are color-coded depending on the sum of PFASs with a detection frequency >50% (C<sub>6</sub>, C<sub>7</sub>, C<sub>9</sub>, C<sub>11</sub> PFCAs and PFBS). Thin gray contour lines show potential density isopycnals referenced to 0 dbar ( $\sigma_0$  [kg/m<sup>3</sup>]). Solid black lines highlight water mass boundaries. Water mass definitions for AW, PSW, Recirculating/Arctic Atlantic Water (RAW/AAW), Intermediate Water (IW;  $\theta \leq 0, 27.97 < \sigma_0, \sigma_{500} \leq 30.444$ ) and Deep Water (DW;  $\theta \leq 0, 30.444 \leq \sigma_{500}$ ) were adapted from reference <sup>5</sup> (Table S21). Data were plotted using Ocean Data View.<sup>25</sup>

**Table S22: PFAS mass transport estimates through Fram Strait via the boundary currents, based on water transport data derived from MITgcm model (section S4).** All values <MDL were replaced by a) zero (blue), b)  $\sqrt{2}$  times the MDL (yellow) c) the MDL (green) to calculate the mean value for each water mass. To calculate the PFAS transport, the mean PFAS concentration of AW, PW and RAW/AAW was multiplied by the transported water volume of the particular water mass, derived from the MITgcm. Positive values describe northward fluxes into the Arctic Ocean, whereas negative values describe southward fluxes to the Nordic Seas. Italicized values are derived from datasets with detection frequencies <50%. If detection frequency of a compound was 100 % in a specific water mass, only one mass transport estimate is given.

Transport through Fram Strait (mean±SD) [kg/y]												
water mass	Substitution of values <MDL	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFBS	PFHxS	L-PFOS	Br-PFOS	HFPO-DA
AW	0					920±852	850±553	1546±1393	446±1041	2536±1383	1587±827	523±629
	$\sqrt{2}$ *MDL	4819±1058	2984±560	6446±1026	3595±656	1159±580	919±446	1980±912	1719±475	2833±886	1753±520	829±450
	MDL					1258±468	947±407	2160±746	2247±267	2956±769	1822±431	956±353
PW	0					<i>-69±130</i>	<i>-116±92</i>		<i>-330±204</i>	<i>-102±224</i>	<i>-148±215</i>	<i>-37±80</i>
	$\sqrt{2}$ *MDL	-1043±190	-819±176	-1471±624	-603±166	<i>-136±93</i>	<i>-132±72</i>	-697±170	<i>-393±100</i>	<i>-340±1120</i>	<i>-251±143</i>	<i>-104±47</i>
	MDL					<i>-163±78</i>	<i>-138±64</i>		<i>-419±65</i>	<i>-438±67</i>	<i>-293±117</i>	<i>-131±34</i>
RAW/AAW	0		<i>-2557±1439</i>			<i>0±0</i>	<i>-616±453</i>	<i>-932±1545</i>	<i>-998±1295</i>	<i>-403±1275</i>	<i>-615±1329</i>	<i>-302±677</i>
	$\sqrt{2}$ *MDL	-4069±1618	<i>-2657±1265</i>	-6512±2843	-2705±741	<i>-551±0</i>	<i>-712±313</i>	<i>-1733±1017</i>	<i>-1938±497</i>	<i>-2047±698</i>	<i>-1435±913</i>	<i>-713±479</i>
	MDL		<i>-2699±1211</i>			<i>-779±0</i>	<i>-751±260</i>	<i>-2065±807</i>	<i>-2328±196</i>	<i>-2728±458</i>	<i>-1774±746</i>	<i>-883±403</i>
Net	0		<i>-392</i>			<i>851</i>	<i>118</i>	<i>-83</i>	<i>-882</i>	<i>2030</i>	<i>824</i>	<i>184</i>
	$\sqrt{2}$ *MDL	-292	<i>-492</i>	-1537	287	<i>472</i>	<i>75</i>	<i>-450</i>	<i>-612</i>	<i>445</i>	<i>68</i>	<i>12</i>
	MDL		<i>-534</i>			<i>315</i>	<i>58</i>	<i>-602</i>	<i>-500</i>	<i>-211</i>	<i>-245</i>	<i>-59</i>

**Table S23: PFAS mass transport estimates through Fram Strait via the boundary currents, based on water transport data derived from observations (mooring array)<sup>15</sup> (section S4).** Values were calculated as explained for Table S22.

Transport through Fram Strait (mean±SD) [kg/y]												
water mass	Substitution of values <MDL	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFBS	PFHxS	L-PFOS	Br-PFOS	HFPO-DA
AW	0					1083±1003	1000±651	1820±1640	525±1226	2985±1628	1868±973	616±814
	$\sqrt{2}$ *MDL	5674±1246	3512±660	7588±1207	4233±772	1364±683	1082±525	2331±1074	2024±559	3335±1043	2064±612	976±529
	MDL					1481±551	1115±479	2543±879	2645±314	3479±906	2145±507	1126±416
PW	0					<i>-159±296</i>	<i>-264±210</i>		<i>-754±466</i>	<i>-233±510</i>	<i>-338±490</i>	<i>-84±182</i>
	$\sqrt{2}$ *MDL	-2380±434	-1868±402	-3357±1425	-1377±379	<i>-310±212</i>	<i>-300±164</i>	-1592±389	<i>-897±228</i>	<i>-775±251</i>	<i>-572±326</i>	<i>-237±107</i>
	MDL					<i>-372±179</i>	<i>-315±146</i>		<i>-956±149</i>	<i>-1000±153</i>	<i>-669±268</i>	<i>-300±77</i>
RAW/AAW	0		<i>-2334±1314</i>			<i>0±0</i>	<i>-563±413</i>	<i>-851±1411</i>	<i>-911±1183</i>	<i>-368±1164</i>	<i>-561±1214</i>	<i>-276±618</i>
	$\sqrt{2}$ *MDL	-3715±1477	<i>-2426±1155</i>	-5945±2596	-2470±677	<i>-503±0</i>	<i>-650±286</i>	<i>-1582±928</i>	<i>-1770±454</i>	<i>-1869±637</i>	<i>-1310±833</i>	<i>-651±438</i>
	MDL		<i>-2464±1105</i>			<i>-712±0</i>	<i>-686±237</i>	<i>-1885±737</i>	<i>-2125±179</i>	<i>-2491±418</i>	<i>-1620±681</i>	<i>-807±368</i>
Net	0		<i>-691</i>			<i>924</i>	<i>174</i>	<i>-622</i>	<i>-1140</i>	<i>2384</i>	<i>969</i>	<i>256</i>
	$\sqrt{2}$ *MDL	-421	<i>-782</i>	-1715	386	<i>551</i>	<i>131</i>	<i>-843</i>	<i>-643</i>	<i>690</i>	<i>182</i>	<i>88</i>
	MDL		<i>-820</i>			<i>397</i>	<i>114</i>	<i>-934</i>	<i>-437</i>	<i>-12</i>	<i>-143</i>	<i>19</i>



**Figure S10:** Comparison of estimated net transport of PFASs through Fram Strait via the boundary columns, calculated by combining measured PFAS concentrations in this study with average water volume transport derived from A) the MITgcm ECCO v4 and B) observational data (mooring array) (section S4).

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