

Supporting Information for:

Concentrations and biomagnification of polyfluorinated compounds through lake food webs in the Canadian high Arctic.

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1. Additional data on lake characteristics and compounds analyzed

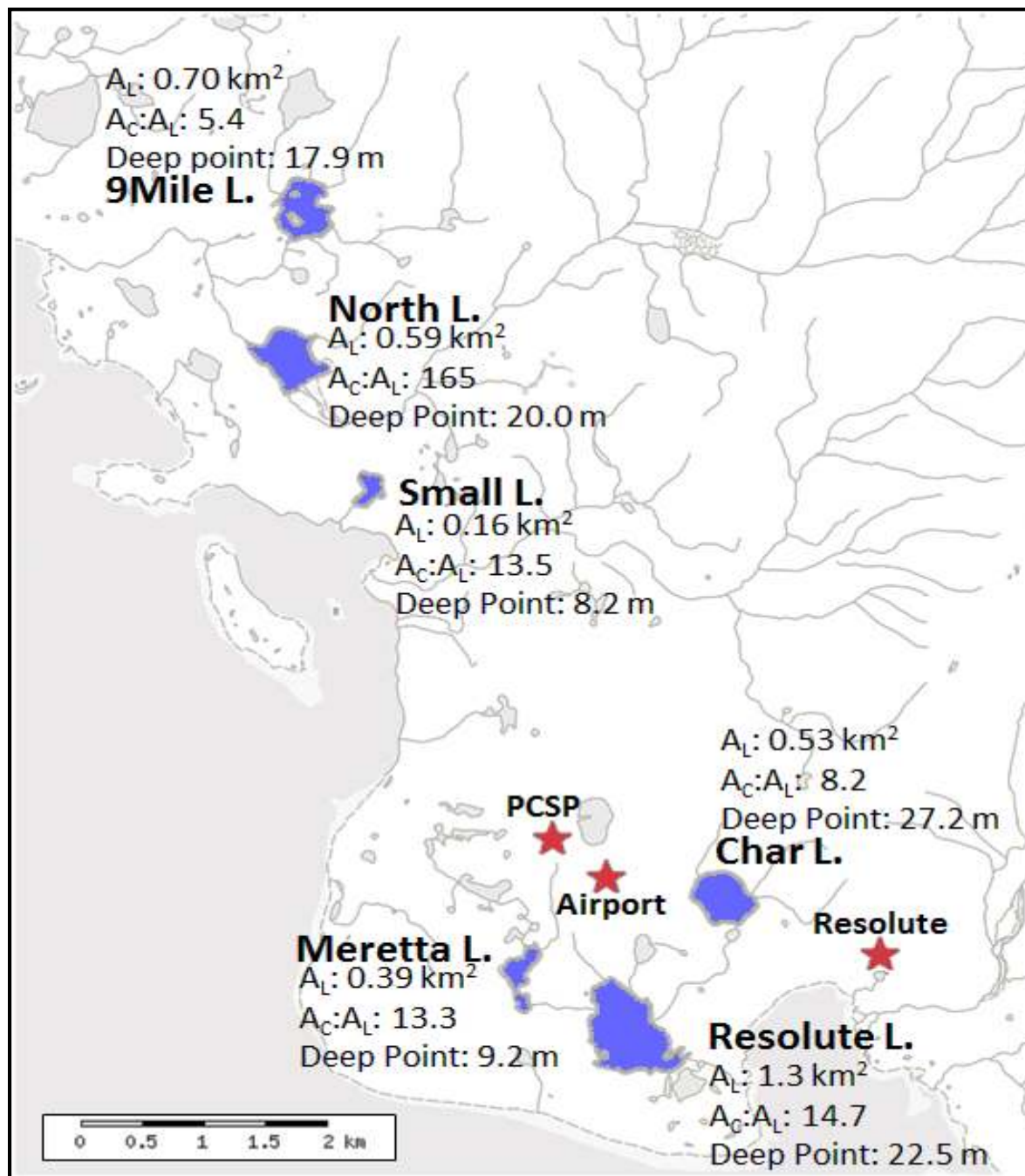


Figure SI-1 – A regional map showing locations of the six sample lakes relative to the airport and town of Resolute Bay, Nunavut. Adapted from Lescord et al. (2014).

Table SI-1 - Mean concentrations and measures of lake characteristics and water chemistry parameters for the six lakes studied on Cornwallis Island, NU. Note: Cond = conductivity; Chl α = Chlorophyll α ; DOC = dissolved organic carbon; POC = particulate organic carbon; AC:AL = Catchment:Lake Area ratio.

Lake	n	Cond. ($\mu\text{S}/\text{cm}$)	pH (pH units)	Chl α ($\mu\text{g}/\text{L}$)	DOC (mg/L)	POC (mg/L)	Depth (m)	Area (km^2)	A _C :A _L (ratio)
Meretta	7	230.4 $\pm$ 34.7	7.90 $\pm$ 0.06	1.29 $\pm$ 1.04	1.74 $\pm$ 0.14	0.24 $\pm$ 0.08	9.2	0.39	13.3
Resolute	7	360.3 $\pm$ 26.2	8.09 $\pm$ 0.05	1.30 $\pm$ 1.14	1.00 $\pm$ 0.06	0.20 $\pm$ 0.03	22.5	1.3	14.7
Char	8	263.6 $\pm$ 21.7	8.10 $\pm$ 0.05	1.28 $\pm$ 1.36	0.53 $\pm$ 0.07	0.14 $\pm$ 0.02	27.2	0.53	8.2
Small	7	277.3 $\pm$ 28.3	8.06 $\pm$ 0.08	1.43 $\pm$ 1.03	1.86 $\pm$ 0.14	0.36 $\pm$ 0.07	8.2	0.16	13.5
North	7	167.3 $\pm$ 53.1	7.91 $\pm$ 0.09	2.23 $\pm$ 1.51	0.83 $\pm$ 0.18	0.21 $\pm$ 0.05	20.0	0.59	165
9-Mile	7	220.3 $\pm$ 90.1	8.02 $\pm$ 0.08	1.93 $\pm$ 1.61	1.72 $\pm$ 1.30	0.22 $\pm$ 0.02	17.9	0.70	5.4

Table SI-2 - Complete list of targeted PFASs, chemical formula and mass spectral precursor-product ion transition (Wellington Laboratories).^{8, 50}

Perfluorocarboxylic acids (PFCAs)				
Chemical name	Abbreviation	Formula	Transitions (m/z)	IS used
Perfluorohexanoic acid	PFHxA (C6)	$\text{CF}_3(\text{CF}_2)_4\text{COO}^-$	313/269*; 313/119	MPFHxA
Perfluoroheptanoic acid	PFHpA (C7)	$\text{CF}_3(\text{CF}_2)_5\text{COO}^-$	363/319*; 363/169	MPFHpA
Perfluorooctanoic acid	PFOA (C8)	$\text{CF}_3(\text{CF}_2)_6\text{COO}^-$	413/369*; 413/169	MPFOA
Perfluorononanoic acid	PFNA (C9)	$\text{CF}_3(\text{CF}_2)_7\text{COO}^-$	463/419*; 463/169	MPFNA
Perfluorodecanoic acid	PFDA (C10)	$\text{CF}_3(\text{CF}_2)_8\text{COO}^-$	513/469*; 513/169	MPFDA
Perfluoroundecanoic acid	PFUnA (C11)	$\text{CF}_3(\text{CF}_2)_9\text{COO}^-$	563/519*; 563/169	MPFUnA
Perfluorododecanoic acid	PFDoA (C12)	$\text{CF}_3(\text{CF}_2)_{10}\text{COO}^-$	613/569*; 613/169	MPFDoA
Perfluorotridecanoic acid	PFTra (C13)	$\text{CF}_3(\text{CF}_2)_{11}\text{COO}^-$	663/619*; 663/169	MPFDoA
Perfluorotetradecanoic acid	PFTA (C14)	$\text{CF}_3(\text{CF}_2)_{12}\text{COO}^-$	713/669*; 713/169	MPFDoA
Perfluoroalkylsulfonates (PFSA)s				
Chemical name	Abbreviation	Formula	Transition (m/z)	IS used
Perfluorobutanesulfonate	PFBS	$\text{CF}_3(\text{CF}_2)_3\text{SO}_3^-$	299/99*; 299/80	MPFHxS
Perfluorohexanesulfonate	PFHxS	$\text{CF}_3(\text{CF}_2)_5\text{SO}_3^-$	399/99*; 399/80	MPFHxS
Perfluoroheptanesulfonate	PFHpS	$\text{CF}_3(\text{CF}_2)_6\text{SO}_3^-$	449/99*; 449/80	MPFHpS
Perfluorooctanesulfonate	PFOS	$\text{CF}_3(\text{CF}_2)_7\text{SO}_3^-$	499/99*; 499/80	MPFOS
Perfluorooctanesulfonamide	FOSA	$\text{CF}_3(\text{CF}_2)_7\text{SO}_2\text{NH}_2^-$	498/78*	MFOSA
Perfluoroethyl-cyclohexanesulfonate	PFECHS	$\text{C}_8\text{F}_{15}\text{SO}_3^-$	461/99*; 461/381	MPFHxS
Perfluorodecanesulfonate	PFDS	$\text{CF}_3(\text{CF}_2)_9\text{SO}_3^-$	599/99*; 599/80	MPFOS
Fluorotelomer Sulfonates (FTSs)				
Chemical name	Abbreviation	Formula	Transition (m/z)	IS used
4:2 Fluorotelomer sulfonate	4:2 FTS	$\text{CF}_3(\text{CF}_2)_3\text{CH}_2\text{CH}_2\text{SO}_3^-$	327/307*; 327/287	MPFHxS
6:2 Fluorotelomer sulfonate	6:2 FTS	$\text{CF}_3(\text{CF}_2)_5\text{CH}_2\text{CH}_2\text{SO}_3^-$	427/407*; 427/387	MPFHxS
8:2 Fluorotelomer sulfonate	8:2 FTS	$\text{CF}_3(\text{CF}_2)_7\text{CH}_2\text{CH}_2\text{SO}_3^-$	527/507*; 527/487	MPFOS

*Transition used for quantification

Table SI-3 - Mean ($\pm$ SD) $\delta^{15}\text{N}$ values of periphyton samples collected from the 6 sample lakes. Mean signatures were subtracted from biotic isotope signatures before comparing between lakes (i.e. ANCOVA analyses).

Lake	n	$\delta^{15}\text{N}$ (‰)
9 Mile	6	2.92 $\pm$ 0.60
Char	6	1.61 $\pm$ 0.57
Meretta	6	2.44 $\pm$ 0.46
North	6	2.54 $\pm$ 0.80
Resolute	6	3.66 $\pm$ 1.05
Small	6	1.04 $\pm$ 0.44

2. Additional information of collection methods

Adult and larval chironomids were collected using aspirators and nets from the ice and shorelines, respectively, between 1 and 5 times per lake in 2010 and 2011. For each sample, chironomids were identified and separated based on trophic ecology; predatory and herbivorous chironomids in the subfamilies Tanypodinae and Orthoclaadiinae or Chironomidae (respectively) were separated for all PFSA and isotope analyses. However, due to low biomass, these data were combined for all statistical analyses and calculation of means. Zooplankton were collected using a 20 μm mesh Wisconsin® net. Adult and juvenile char were collected using gill nets and electrofishers, respectively, at the end of each season. Fish were weighed, measured, and dissected for dorsal muscle.

A total of 3 water samples/lake/depth were collected in 2010 and 2011, logistics permitting. All water samples were collected in 500 mL polypropylene bottles and stored in a cold room ($\sim 4\text{-}5^\circ\text{C}$) until analyzed. Littoral sediment samples ($n = 3\text{-}5/\text{lake}$) were taken with a gravity corer and the top 0.5 cm was collected and frozen. Periphyton samples were collected by hand at three separate sites on each lake for baseline measurement of carbon (C) and nitrogen (N) isotopes.³⁹

3. Additional information of extraction methods

Biotic extraction: Briefly, samples were extracted twice (5.0 mL acetonitrile, 10 to 30 minutes of shaking, and 20 minutes of centrifuging per extraction), dried using an Organomation® Analytical Nitrogen Evaporator (N-EVAP) and later reconstituted in 1.0 mL of methanol (MeOH). Samples were passed through pre-conditioned (5.0 mL MeOH and 2.0 mL glacial acetic acid) carbon SPE columns (Supelclean ENVI-Carb 1 mL 100 mg) and eluted using MeOH. The resulting extracts were dried and reconstituted to 1.0 mL in a 50:50 solution of methanol:water. For compound detection, all biotic samples were run through a Hypersil GOLD® pentafluorophenyl (PFP) column (150 mm X 3 mm, 3 μm particle size) using a solvent gradient similar to Houde et al. (2008) and De Silva et al. (2011). Other samples, run on the Waters™ ultra-performance liquid chromatography (UPLC), were run through a Phenomenex® Gemini NX-C18 column (50 mm X 3 mm X 3 μm) to quantify FTSS. The internal temperatures of both columns were kept consistent at 35.0°C.

Water extractions: Water samples ($n = 30$) were extracted using Oasis® WAX SPE columns as per Taniyasu et al. (2005). Briefly, 500 mL (± 10 mL) samples were warmed to room temperature, spiked with appropriate internal standards, and passed through WAX (weak anion exchange) SPE columns, prepped with 5.0 mL of MeOH basicified with ammonia, 5.0 mL of MeOH, and 5.0 mL of HPLC grade water. PFAS were eluted with 6.0 mL of neutral MeOH for fraction 1 (FOSA only) and 8.0 mL of basic MeOH was used for fraction 2 (all other PFA and PFS compounds). Each fraction was dried and reconstituted in 1.0 mL 50:50 ratio of MeOH:water. 20 μL from each fraction were injected into the Waters™ UPLC, interfaced with the 4000 Q Trap™ LC/MS/MS using a Phenomenex® Gemini NX-C8 column. Sediments were extracted using base digestion and solvent extraction as per Yeung et al (2013)⁵¹; 50 mL of the sediment extract was subjected to further clean up using the WAX method described above.

4. Quality Assurance and Control Data:

Table SI-4 – Percent recovery (%R) of PFASs from spiked char and invertebrate samples and analysis of standard reference material fish tissue (1947 Lake Superior Fish Tissue, NIST) compared to reference values presented by Reiner et al. (2012) and percent recoveries (%R) were measured.

Sample	PFNA		PFDA		PFUnA		PFTriA		PFOS	
	(ng/g)	%R	(ng/g)	%R	(ng/g)	%R	(ng/g)	%R	(ng/g)	%R
Char	0.19	100	0.18	100	0.28	100	0.21	100	4.49	100
Char	0.22	112.1	0.21	115.7	0.33	116.1	0.29	138.3	4.91	109.4
Char	0.18	91.6	0.17	97.8	0.27	96.1	0.21	99.1	3.83	85.2
Char	0.24	126.2	0.22	125.2	0.39	138.2	0.33	157.7	5.83	129.8
Char	0.23	117.3	0.21	118.8	0.32	114.2	0.27	127.2	5.34	118.9
Char	0.22	113.8	0.21	115.4	0.33	117.1	0.27	129.7	5.45	121.3
Char	0.19	98.7	0.18	99.8	0.28	97	0.22	105.6	4.67	104
Char	0.21	109.7	0.2	113.1	0.32	113.3	0.29	139.5	4.84	107.8
Char	0.17	89.8	0.17	95.4	0.26	93.2	0.22	103.2	3.9	86.7
Char	0.24	123.6	0.22	123	0.39	136.9	0.33	158.9	5.9	131.4
Char	0.22	114.6	0.21	120.1	0.31	110.7	0.27	131.3	5.34	118.9
Char	0.22	111.9	0.2	112.1	0.33	114.9	0.25	118.2	5.49	122.1
Char	0.24	122.4	0.21	119	0.27	94.8	0.22	104.5	4.59	102.1
Char	0.11	55.3	0.13	73.1	0.21	75.1	0.21	101.4	5.28	117.5
Char	0.15	76.4	0.24	133.4	0.21	74.3	0.33	158.3	6.2	138
Invertebrate	0.45	233.7	0.4	225.8	0.47	167.3	0.38	179.7	10.66	237.3
Invertebrate	0.4	205.7	0.43	238.9	0.63	222.1	0.57	270.8	12.68	282.2
Invertebrate	0.44	230	0.47	264.1	0.62	217.4	0.59	281.5	12.11	269.4
Reference value for NIST SRM⁴⁰		0.2		0.26		0.28		0.2		5.9
Mean for SRM in this study (ng/g)		0.26		0.25		0.36		0.31		6.65
Mean %Recovery		129		97.8		126.9		151.8		112.6

Table SI-5 - Native spiked sample recovery (SR) for PFCA compounds.

Sample	n	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnA	PFDoA	PFTriA	PFTA
Juvenile Char	5	80±11	84±12	81±12	89±9	81±11	83±2	84±11	84±14	99±67
Adult Char	8	83±8	83±8	85±10	87±10	81±7	84±9	78±10	76±14	76±14
Inverts	13	85±26	83±28	92±33	91±36	90±33	89±24	88±27	93±23	100±31
Water	7	108±6	105±4	88±9	106±21	112±7	100±9	83±6	87±16	89±17
Sediments	3	98±19	75±13	70±7	72±5	83±10	83±16	81±11	66±11	55±8

Table SI-5 - continued - Native spiked sample recovery (SR) for PFSA compounds.

Sample	n	PFBS	PFHxS	PFHpS	PFOS	PFDS	FOSA	PFECHS
Juvenile Char	5	34±39	88±12	97±8	138±88	78±9	ND	113±28
Adult Char	8	147±20	85±8	85±6	84±8	83±7	ND	87±9
Inverts	13	64±32	68±7	97±32	14±99	108±28	87±36	106±18
Water	7	150±59	118±5	ND	99±7	106±14	ND	198±31
Sediments	3	144±58	77±11	81±13	64±26	67±6	100±26	83±8

Table SI-6 –Blank concentrations and corresponding method detection limits (MDL) for the 9 PFCA compounds analyzed in this study.

Compound	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnA	PFDoA	PFTriA	PFTA
Adult and Juvenile Char									
Conc. (pg/mL)	1.7±2.9	2.6±4.7	3.8±6.0	1.5±2.6	0.2±0.4	1.7±3.6	2.7±4.3	3.7±10.8	0.8±1.6
n	8	10	8	10	10	10	10	10	10
MDL (ng/g)	0.036	0.056	0.072	0.032	0.0040	0.044	0.052	0.13	0.02
Invertebrates									
Conc. (pg/mL)	1.0±2.6	6.2±2.4	13±5.3	2.2±2.4	3.9±3.1	2.5±3.9	3.7±5.0	4.8±6.5	8.0±5.0
n	28	28	28	30	28	27	28	27	28
MDL (ng/g)	0.050	0.043	0.10	0.044	0.056	0.075	0.094	0.12	0.094

Note: MDLs are based on a mean sample weight of 0.25g for fish and 0.16g for invertebrates. Procedural blanks concentrations consisted of extraction using the full suite of reagents and internal standards without any sample. For mean calculations, if no signal was detected, blank concentrations were assumed to be half of the IDL for the corresponding analyte.

Table SI-6 continued – Blank concentrations and corresponding method detection limits (MDL) for the 9 PFCA compounds analyzed in this study.

Compound	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnA	PFDoA	PFTriA	PFTA
Sediments									
Conc. (pg/mL)	29±13	13±5.9	23±3.2	18±4.2	1.1±1.7	16±3.5	14±4.8	14±5.4	29±13
n	7	6	6	5	6	5	5	6	7
MDL (ng/g)	0.08	0.036	0.020	0.026	0.010	0.022	0.028	0.032	0.080
Water									
Conc. (pg/mL)	5.5±8.4	0.44±0.47	2.4±3.5	0.40±0.30	0.10±0.06	0.31±0.31	0.39±0.55	0.40±0.50	0.09±0.06
n	4	4	4	4	4	4	4	4	4
MDL (ng/L)	0.050	0.0028	0.021	0.0018	0.0003	0.0018	0.0033	0.0030	0.0004

Note: MDLs are based on a mean sample weight of 0.50 g for sediments and a volume of 0.5 L.

Table SI-7 – Blank concentrations and corresponding method detection limits (MDL) for the 10 PFSA compounds analyzed in this study.

Compound	PFBS	PFHxS	PFHpS	PFOS	PFDS	FOSA	PFECHS	42 FTS	62 FTS	82 FTS
Adult and Juvenile Char										
Conc. (pg/mL)	2.9±8.2	1.2±3.0	1.6±3.2	34±65	3.1±12	0.5±0.2	0.1±0.1	1.0±1.2	5.7±16	1.1±1.4
n	9	10	10	10	10	9	9	11	12	12
MDL (ng/g)	0.10	0.036	0.036	0.78	0.14	0.004	0.001	0.016	0.19	0.016
Invertebrates										
Conc. (pg/mL)	2.3±4.7	3.7±3.7	1.6±2.4	3.0±6.3	1.1±2.4	0.4±0.3	4.0±5.3	28±21.0	72±71	13±10
n	28	28	28	29	29	29	28	32	32	29
MDL (ng/g)	0.088	0.069	0.04	0.12	0.044	0.0063	0.10	0.39	1.3	0.19
Sediments										
Conc. (pg/mL)	12±5.6	43±35	17±5.0	5.3±5.6	17±13	8.3±3.1	0.0±0.0	10±4.7	0.1±0.1	13±11
n	6	7	6	6	6	6	0	6	6	6
MDL (ng/g)	0.034	0.21	0.030	0.034	0.074	0.018	0	0.028	0	0.066
Water										
Conc. (pg/mL)	0.33±0.32	0.30±0.28	0.14±0.10	1.4±1.9	0.29±0.36	0.65±0.44	18.4±28.1	0.80±0.91	1.7±2.6	0.77±0.83
n	4	4	0	4	4	4	4	4	4	4
MDL (ng/L)	0.0019	0.0017	0.0006	0.012	0.0022	0.0026	0.17	0.0055	0.015	0.005

Note: MDLs are based on a mean sample weight of 0.25, 0.16, 0.50 g, and 0.5 L for fish, invertebrates, sediments, and water, respectively

Table SI-8 – Instrument detection limits (IDL) and related data for 19 PFAS compounds analyzed in this study.

Compound	STD Conc. (ng/mL)	Mean S:N	IDL (ng/mL)	IDL (ng/g)	STD Conc. (ng/mL)	Mean S:N	IDL (ng/mL)	IDL (ng/g)
	Sediments/Water				Biota			
PFHxA	0.018	42.3	0.0024	0.0048	0.012	70.5	0.0008	0.0040
PFHpA	0.018	140.8	0.0007	0.0014	0.012	130.7	0.0003	0.0017
PFOA	0.018	172.8	0.0006	0.0012	0.012	115.8	0.0002	0.0013
PFNA	0.018	277.1	0.0004	0.0007	0.012	207.2	0.0002	0.0011
PFDA	0.018	262.0	0.0004	0.0008	0.012	128.9	0.0003	0.0017
PFUnA	0.018	238.9	0.0004	0.0009	0.012	37.3	0.0003	0.0023
PFDoA	0.018	329.2	0.0003	0.0006	0.012	157.5	0.0002	0.0014
PFTriA	0.018	297.5	0.0003	0.0007	0.012	128.0	0.0009	0.0070
PFTA	0.018	294.6	0.0003	0.0007	0.018	56.1	0.0007	0.0045
PFBS	0.030	28.2	0.0032	0.0064	0.010	68.2	0.0006	0.0037
PFHxS	0.032	60.6	0.0016	0.0032	0.012	83.5	0.0004	0.0024
PFHpS	0.032	146.6	0.0007	0.0013	0.011	63.6	0.0003	0.0014
PFOS	0.032	135.8	0.0007	0.0014	0.011	17.1	0.0010	0.0068
PFDS	0.032	187.0	0.0005	0.0010	0.011	24.3	0.0010	0.0068
FOSA	0.029	31.5	0.0030	0.0060	0.010	97.6	0.0003	0.0018
PFECHS	0.031	485.6	0.0002	0.0004	0.012	17.6	0.0014	0.0102
42 FTS	0.027	83.2	0.0010	0.0020	0.015	78.3	0.0005	0.0041
62 FTS	0.028	350.6	0.0002	0.0005	0.015	203.6	0.0002	0.0015
82 FTS	0.028	122.8	0.0007	0.0014	0.015	62.5	0.0004	0.0030

Table SI-9 - Total PFAS calculations. Sum of Total PFC include all 19 compounds analyzed. Total PFCA concentrations were the sum of PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTriA, and PFTA. Total PFSA concentrations are the sum of PFBS, PFHxS, PFHpS, PFDS, PFECHS, FOSA 4:2 FTS, 6:2 FTS, and 8:2 FTS. PFOS concentrations are presented separately. Note: Σ PFSA calculations in Sediments (seds) did not include PFHpS, as data was unavailable. All measures are in ng/g (wet wt.; or ng/L in water samples) except those asterisked (*pg/g or pg/L).

Sample Type	Lake (n)	Σ PFAS	Σ PFCA	Σ PFSA	PFOS
Seds	Meretta (4)	57±10	5.1±7.0	24±34	28±43
Seds	Resolute (3)	64±6.6	4.4±0.53	11±7.0	49±29
Seds	Char (4)	2.7±0.18	1.9±1.3	0.40±0.46	0.44±0.08
Seds	Small (3)	2.1±1.8	0.86±0.16	0.001±0.002	0.22±0.13
Seds	North (4)	0.19±0.03	0.15±0.21	0.02±0.02	0.02±0.03
Seds	9 Mile (5)	1.6±1.9	0.82±0.42	0.73±1.4	0.001±0.001
Water	Meretta (5)	153±14	71±7.9	41±6.7	41±9.3
Water	Resolute (5)	112±20	60±17.9	26±4.6	26±5.2
Water	Char (5)	1.9±0.48	1.50±0.46	0.39±0.05	0.05±0.01
Water	Small (5)	2.4±0.40	1.91±0.41	0.43±0.02	0.09±0.02
Water	North (5)	1.9±0.81	1.68±0.83	0.16±0.03	0.03±0.00
Water	9 Mile (5)	1.9±0.42	1.66±0.40	0.20±0.05	0.02±0.01

Table SI-10 – Mean ($\pm$ SD) concentrations of individual PFAS compounds in various samples collected for six lakes in the Canadian high Arctic. Note ND = non-detect (< 50% of samples below MDL).

Sample Type	Lake (n)	PFHxA	PFOA	PFDA	PFHxS	FOSA	4:2 FTS	8:2 FTS
Seds	Meretta (4)	0.23 $\pm$ 0.21	1.8 $\pm$ 2.4	0.039 $\pm$ 0.035	0.67 $\pm$ 0.17	0.13 $\pm$ 0.20	0.20 $\pm$ 0.31	6.3 $\pm$ 5.7
	Resolute (3)	0.065 $\pm$ 0.057	1.8 $\pm$ 0.3	0.023 $\pm$ 0.0001	0.45 $\pm$ 0.043	0.002 $\pm$ 0.002	0.001 $\pm$ 0.014	0.002 $\pm$ 0.029
	Char (4)	0.67 $\pm$ 1.15	0.74 $\pm$ 0.55	0.14 $\pm$ 0.16	0.95 $\pm$ 1.3	0.01 $\pm$ 0.01	0.12 $\pm$ 0.24	13 $\pm$ 20
	Small (3)	ND	0.37 $\pm$ 0.05	0.018 $\pm$ 0.050	0.001 $\pm$ 0.0001	0.02 $\pm$ 0.03	0.001 $\pm$ 0.025	0.001 $\pm$ 0.0001
	North (4)	ND	0.04 $\pm$ 0.03	ND	0.001 $\pm$ 0.0001	0.01 $\pm$ 0.01	0.001 $\pm$ 0.0001	0.001 $\pm$ 0.0001
	9 Mile (5)	ND	0.33 $\pm$ 0.21	0.017 $\pm$ 0.011	0.62 $\pm$ 1.38	ND	0.002 $\pm$ 0.002	0.012 $\pm$ 0.012
Water	Meretta (5)	30 $\pm$ 4.7	17 $\pm$ 1.2	0.13 $\pm$ 0.05	30 $\pm$ 3.5	ND	0.18 $\pm$ 0.12	0.20 $\pm$ 0.26
	Resolute (5)	22 $\pm$ 3.0	9.4 $\pm$ 1.97	0.11 $\pm$ 0.06	19.7 $\pm$ 3.87	ND	ND	0.01 $\pm$ 0.01
	Char (5)	0.43 $\pm$ 0.15	0.62 $\pm$ 0.27	0.04 $\pm$ 0.04	0.12 $\pm$ 0.01	ND	ND	ND
	Small (5)	0.60 $\pm$ 0.12	0.60 $\pm$ 0.22	0.06 $\pm$ 0.03	0.11 $\pm$ 0.02	ND	ND	ND
	North (5)	0.43 $\pm$ 0.33	0.66 $\pm$ 0.41	0.08 $\pm$ 0.04	0.01 $\pm$ <0.001	ND	ND	ND
	9 Mile (5)	0.43 $\pm$ 0.09	0.69 $\pm$ 0.31	0.08 $\pm$ 0.03	0.02 $\pm$ <0.001	ND	ND	ND
Benthic	Meretta (8)	0.38 $\pm$ 0.42	ND	1.7 $\pm$ 1.4	ND	ND	ND	ND
Invertebrates	Resolute (10)	0.34 $\pm$ 0.50	ND	1.7 $\pm$ 2.0	ND	0.75 $\pm$ 0.70	ND	11 $\pm$ 7.9
	Char (6)	0.083 $\pm$ 0.10	ND	0.20 $\pm$ 0.30	ND	ND	ND	0.74 $\pm$ 1.1
	Small (4)	0.36 $\pm$ 0.61	ND	2.0 $\pm$ 1.5	ND	0.01 $\pm$ 0.01	ND	0.39 $\pm$ 0.19
	North (10)	0.18 $\pm$ 0.30	ND	0.4 $\pm$ 0.4	ND	0.42 $\pm$ 1.13	ND	ND
	9-Mile (6)	0.19 $\pm$ 0.22	ND	1.1 $\pm$ 2.5	ND	ND	ND	0.22 $\pm$ 0.19
Pelagic	Meretta (13)	0.13 $\pm$ 0.10	ND	0.13 $\pm$ 0.10	ND	ND	ND	0.54 $\pm$ 0.54
Invertebrates	Resolute (9)	ND	ND	0.13 $\pm$ 0.10	ND	0.01 $\pm$ 0.01	ND	ND
	Char (6)	ND	ND	ND	ND	0.00 $\pm$ 0.00	ND	0.018 $\pm$ 0.027
	Small (7)	0.14 $\pm$ 0.35	ND	ND	ND	0.01 $\pm$ 0.01	ND	ND
	North (5)	0.017 $\pm$ 0.015	ND	ND	ND	0.02 $\pm$ 0.01	ND	ND
	9-Mile (7)	ND	ND	ND	ND	0.01 $\pm$ 0.01	ND	ND

Table SI-10 continued - Mean ($\pm$ SD) concentrations of individual PFAS compounds in various samples collected for six lakes in the Canadian high Arctic. Note ND = non-detect (< 50% of samples below MDL). NA = no data available.

Sample Type	Lake (n)	PFHxA	PFOA	PFDA	PFHxS	FOSA	4:2 FTS	8:2 FTS
Juvenile char (whole body)	Meretta (5)	ND	1.31 $\pm$ 0.65	0.81 $\pm$ 0.72	ND	2.24 $\pm$ 0.28	2.5 $\pm$ 3.7	7.4 $\pm$ 3.4
	Resolute (9)	ND	50.64 $\pm$ 151.49	0.29 $\pm$ 0.64	0.18 $\pm$ 0.06	0.17 $\pm$ 0.18	0.17 $\pm$ 0.20	1.4 $\pm$ 1.5
	Char (5)	ND	ND	0.36 $\pm$ 0.29	ND	ND	0.26 $\pm$ 0.38	ND
	Small (7)	ND	ND	0.11 $\pm$ 0.057	ND	ND	0.18 $\pm$ 0.049	0.05 $\pm$ 0.06
	North (8)	ND	ND	0.08 $\pm$ 0.040	ND	ND	0.55 $\pm$ 0.57	ND
	9-Mile (14)	ND	0.30 $\pm$ 0.49	0.21 $\pm$ 0.27	ND	ND	0.37 $\pm$ 0.21	ND
Juvenile char (muscle)	Meretta (5)	0.001 $\pm$ <0.001	ND	0.84 $\pm$ 0.95	2.0 $\pm$ 1.0	0.55 $\pm$ 0.48	ND	1.4 $\pm$ 0.9
	Resolute (4)	0.001 $\pm$ 0.001	ND	0.09 $\pm$ 0.09	ND	0.16 $\pm$ 0.15	ND	0.11 $\pm$ 0.031
	Char (9)	0.001 $\pm$ <0.001	ND	0.10 $\pm$ 0.07	ND	0.04 $\pm$ 0.04	0.29 $\pm$ 0.25	0.06 $\pm$ 0.05
	Small (9)	0.003 $\pm$ 0.007	ND	ND	ND	ND	0.07 $\pm$ 0.038	ND
	North (5)	0.001 $\pm$ 0.001	ND	0.07 $\pm$ 0.038	ND	0.04 $\pm$ 0.03	3.6 $\pm$ 7.2	3.6 $\pm$ 7.2
	9-Mile (0)	ND	NA	ND	ND	NA	ND	ND
Adult char (muscle)	Meretta (21)	ND	0.10 $\pm$ 0.13	0.080 $\pm$ 0.12	0.28 $\pm$ 0.10	0.93 $\pm$ 0.31	0.0055 $\pm$ 0.0094	0.15 $\pm$ 0.17
	Resolute (18)	0.036 $\pm$ 0.037	0.35 $\pm$ 0.10	0.14 $\pm$ 0.13	1.2 $\pm$ 1.5	117.1 $\pm$ 63.7	ND	0.11 $\pm$ 0.07
	Char (13)	0.058 $\pm$ 0.067	ND	0.22 $\pm$ 0.14	ND	0.27 $\pm$ 0.17	ND	ND
	Small (20)	ND	ND	0.032 $\pm$ 0.015	ND	0.01 $\pm$ 0.01	ND	ND
	North (25)	ND	ND	0.048 $\pm$ 0.022	ND	0.05 $\pm$ 0.04	ND	ND
	9-Mile (23)	ND	ND	0.023 $\pm$ 0.012	ND	0.01 $\pm$ 0.02	ND	ND

Table SI-11 – P-values resulting from Kruskal-Wallis H tests, showing the overall differences in total PFAS concentrations detected between lakes within each biotic group. *significant difference based on $\alpha = 0.05$.

Measure	Char (Adult)	Char (J-M)	Char (J-WB)	Benthic	Pelagic
ΣPFCA	<0.0001*	0.053	0.006*	0.007*	0.005*
ΣPFSA	<0.0001*	0.125	0.005*	<0.001*	0.106
PFOS	<0.0001*	<0.0001*	<0.0001*	0.036*	<0.0001*

Table SI-12 - Post-hoc comparisons (Mann-Whitney U tests) of total PFCA, PFSA, and PFOS concentrations in invertebrates samples from six high arctic lakes. *significant difference based on $\alpha = 0.05$.

Lake Comparison	Benthic (n = 5-14)			Pelagic (n = 6-13)		
	ΣPFCA	ΣPFSA	PFOS	ΣPFCA	ΣPFSA	PFOS
9-Mile - Char	0.485	0.818	0.937	0.295	0.366	0.234
9-Mile - Meretta	0.272	0.012*	0.002*	0.275	0.067	<0.0001*
9-Mile - North	0.713	0.875	0.792	0.999	0.268	0.202
9-Mile - Resolute	0.147	0.011*	0.002*	0.299	0.091	0.001*
9-Mile - Small	0.610	0.352	0.914	0.620	0.383	0.620
Char - Meretta	0.113	0.008*	0.002*	0.007*	0.072	<0.0001*
Char - North	0.635	0.999	0.368	0.329	0.792	0.030*
Char - Resolute	0.022*	0.011*	0.003*	0.012*	0.113	<0.001*
Char - Small	0.257	0.352	0.914	0.731	0.945	0.445
Meretta - North	0.079	0.003*	<0.0001*	0.014*	0.075	<0.0001*
Meretta - Resolute	0.968	0.243	0.968	0.999	0.393	0.999
Meretta - Small	0.414	0.106	0.003*	0.001*	0.183	<0.0001*
North - Resolute	0.019*	0.011*	<0.0001*	0.012*	0.083	0.004*
North - Small	0.188	0.733	0.539	0.755	0.755	0.030*
Resolute - Small	0.945	0.240	0.002*	0.023*	0.174	<0.0001*

*Table SI-13 - Post-hoc comparisons (Mann-Whitney U tests) of total PFCA, PFSA, and PFOS concentrations in adult and juvenile char samples from six high arctic lakes. Note WB = whole body. *significant difference based on $\alpha = 0.05$.*

Lake Comparison	JC-WB (n = 5-14)			Adult Char (n = 13-25)		
	Σ PFCA _s	Σ PFSA _s	PFOS	Σ PFCA _s	Σ PFSA _s	PFOS
9-Mile - Char	0.257	0.559	0.003*	<0.0001*	<0.0001*	<0.0001*
9-Mile - Meretta	0.006*	0.003*	0.003*	<0.0001*	<0.0001*	<0.0001*
9-Mile - North	0.004*	0.868	0.165	0.812	0.044*	0.926
9-Mile - Resolute	0.124	0.007*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
9-Mile - Small	0.094	0.079	0.001*	0.098	0.592	<0.0001*
Char - Meretta	0.071	0.036	0.036*	0.148	<0.0001*	<0.0001*
Char - North	0.045*	0.354	0.002*	<0.0001*	<0.0001*	<0.0001*
Char - Resolute	0.190	0.999	0.001*	0.352	<0.0001*	<0.0001*
Char - Small	0.073	0.432	0.876	<0.0001*	<0.0001*	<0.0001*
Meretta - North	0.012*	0.012*	0.012*	<0.0001*	<0.0001*	<0.0001*
Meretta - Resolute	0.999	0.064	0.145	0.119	0.024*	<0.0001*
Meretta - Small	0.017*	0.017*	0.017*	<0.0001*	<0.0001*	<0.0001*
North - Resolute	0.888	0.036*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
North - Small	0.232	0.463	<0.0001*	0.226	0.005*	<0.0001*
Resolute - Small	0.408	0.008*	<0.0001*	<0.0001*	<0.0001*	<0.0001*

Table SI-14 - Linear Regression results of relationships between sum of PFAS classes and $\delta^{15}\text{N}$ values across lakes. All data were log transformed.

Lake	Measure	ΣPFAS^*	ΣPFCA^*	ΣPFSA^*	PFOS*
Meretta	p	<0.001	<0.001	<0.001	<0.001
	r^2	0.625	0.484	0.662	0.684
	m	-0.19	-0.19	-0.19	-0.16
Resolute	p	0.092	0.005	0.555	0.164
	r^2	0.077	0.195	<0.001	0.053
	m	-0.08	-0.17	-0.03	-0.07
Char	p	0.125	0.796	0.043	0.425
	r^2	0.074	0.002	0.125	0.021
	m	-0.05	-0.01	-0.07	0.06
Small	p	<0.001	<0.001	0.001	0.055
	r^2	0.482	0.501	0.252	0.097
	m	-0.31	-0.36	-0.33	-0.36
North	p	0.002	0.002	0.069	0.108
	r^2	0.209	0.189	0.075	0.059
	m	-0.11	-0.10	-0.08	-0.15
9-Mile	p	<0.001	<0.001	<0.001	0.021
	r^2	0.414	0.359	0.385	0.129
	m	-0.23	-0.22	-0.30	-0.37

ANCOVA interaction terms (Lake $\delta^{15}\text{N}$) significant, indicating a difference between the six slopes.

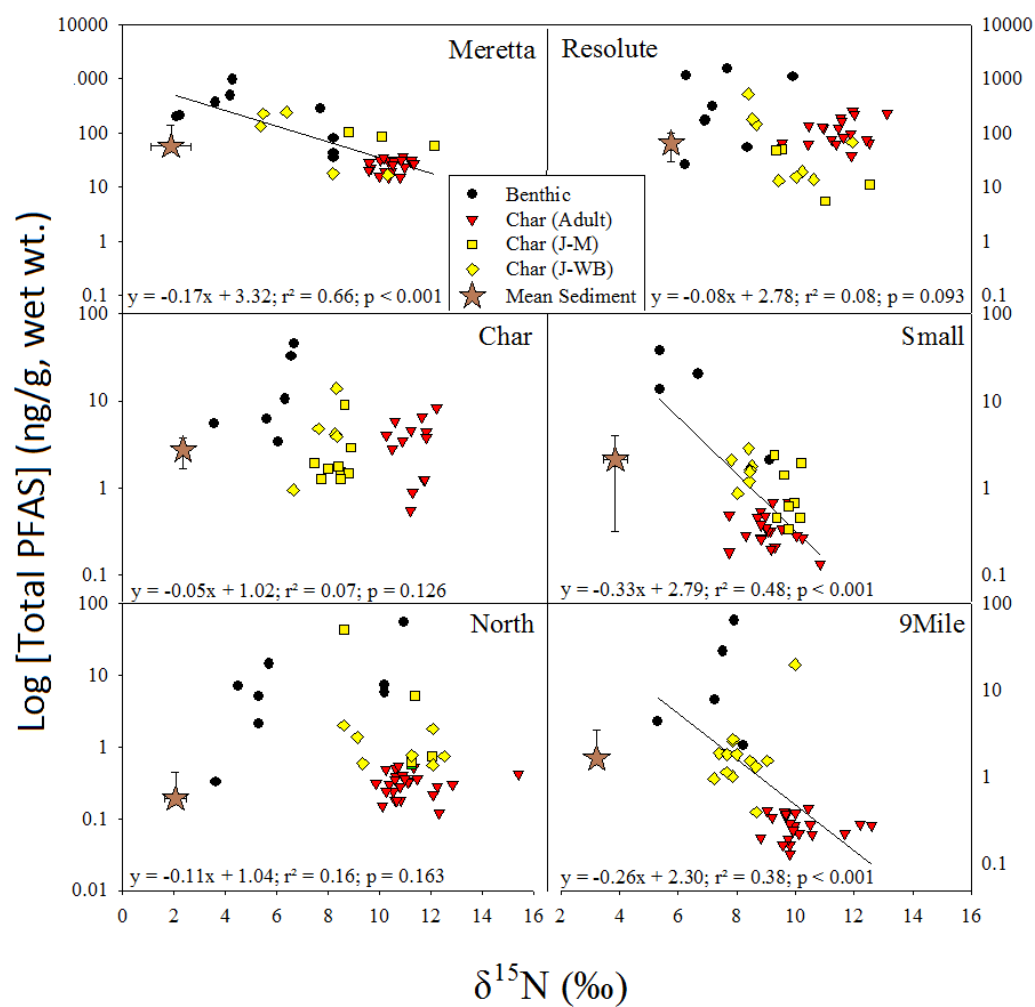


Figure SI-2 - Linear regressions of Total PFAS concentrations and $\delta^{15}\text{N}$ values in adult char, juvenile char (muscle samples [JC-M] and whole body homogenates [JC-WB]), and benthic invertebrates. Mean [PFAS] (ng/g dry wt.) and $\delta^{15}\text{N}$ values of sediment samples are also presented. All data are log-transformed.

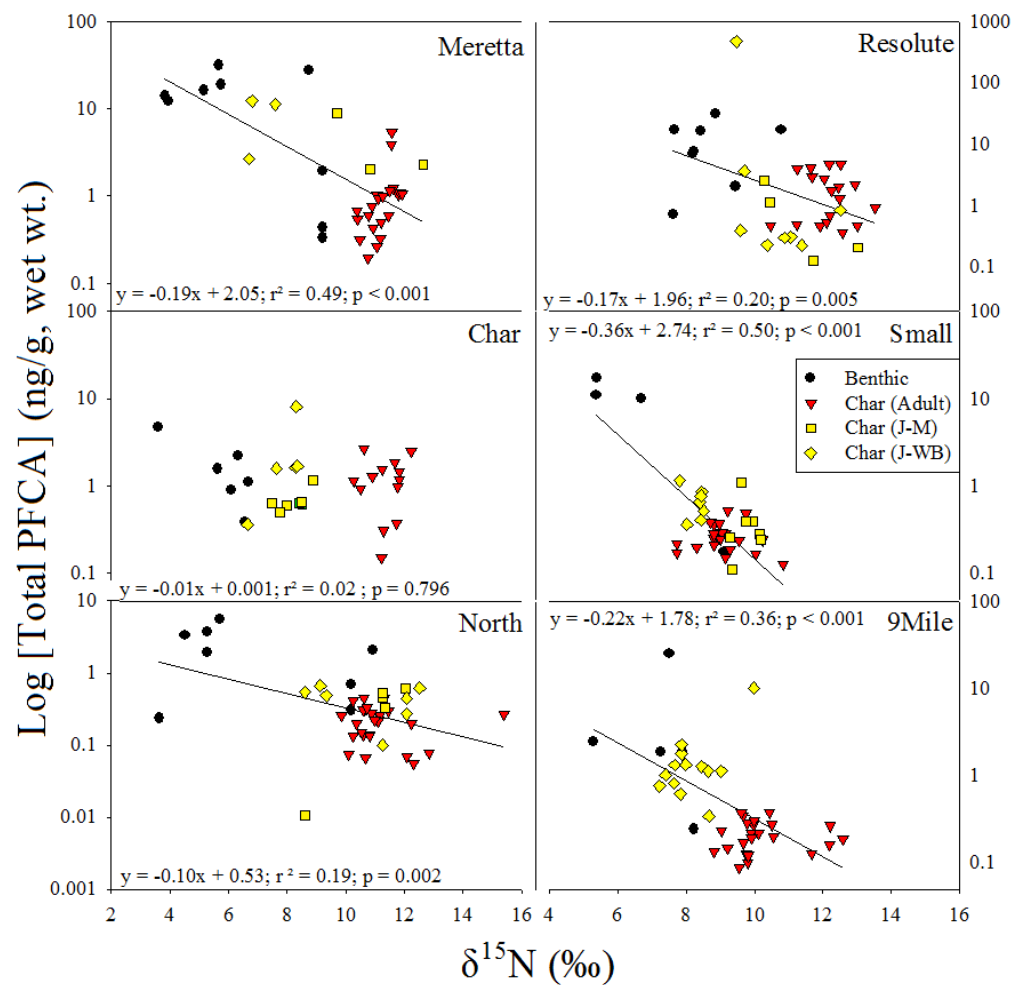


Figure SI-3- Linear regressions of Total PFCA concentrations and $\delta^{15}\text{N}$ values in adult char, juvenile char (muscle samples [JC-M] and whole body homogenates [JC-WB]), and benthic invertebrates. All data are log-transformed.

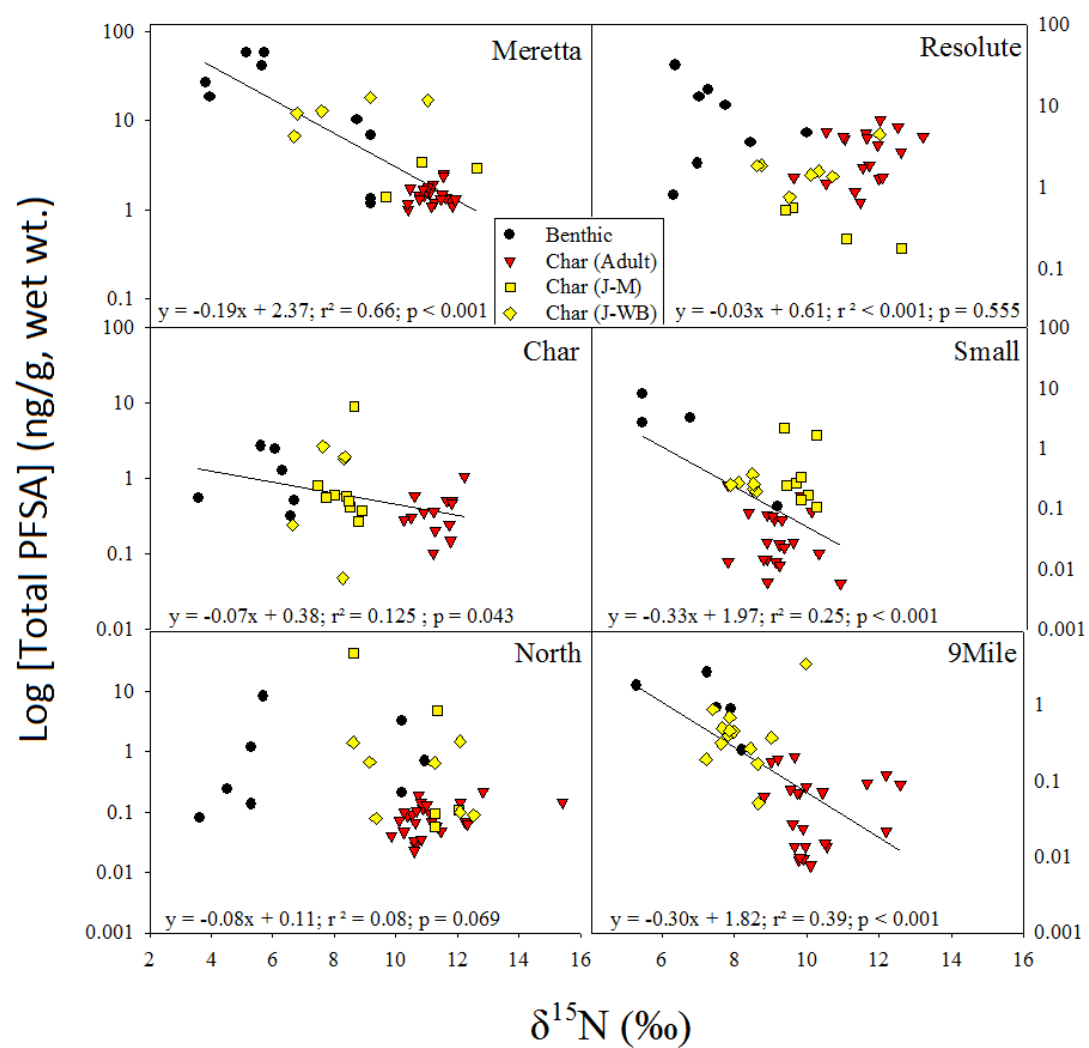


Figure SI-4 - Linear regressions of Total PFSA concentrations and $\delta^{15}\text{N}$ values in adult char, juvenile char (muscle samples [JC-M] and whole body homogenates [JC-WB]), and benthic invertebrates. All data are log-transformed.

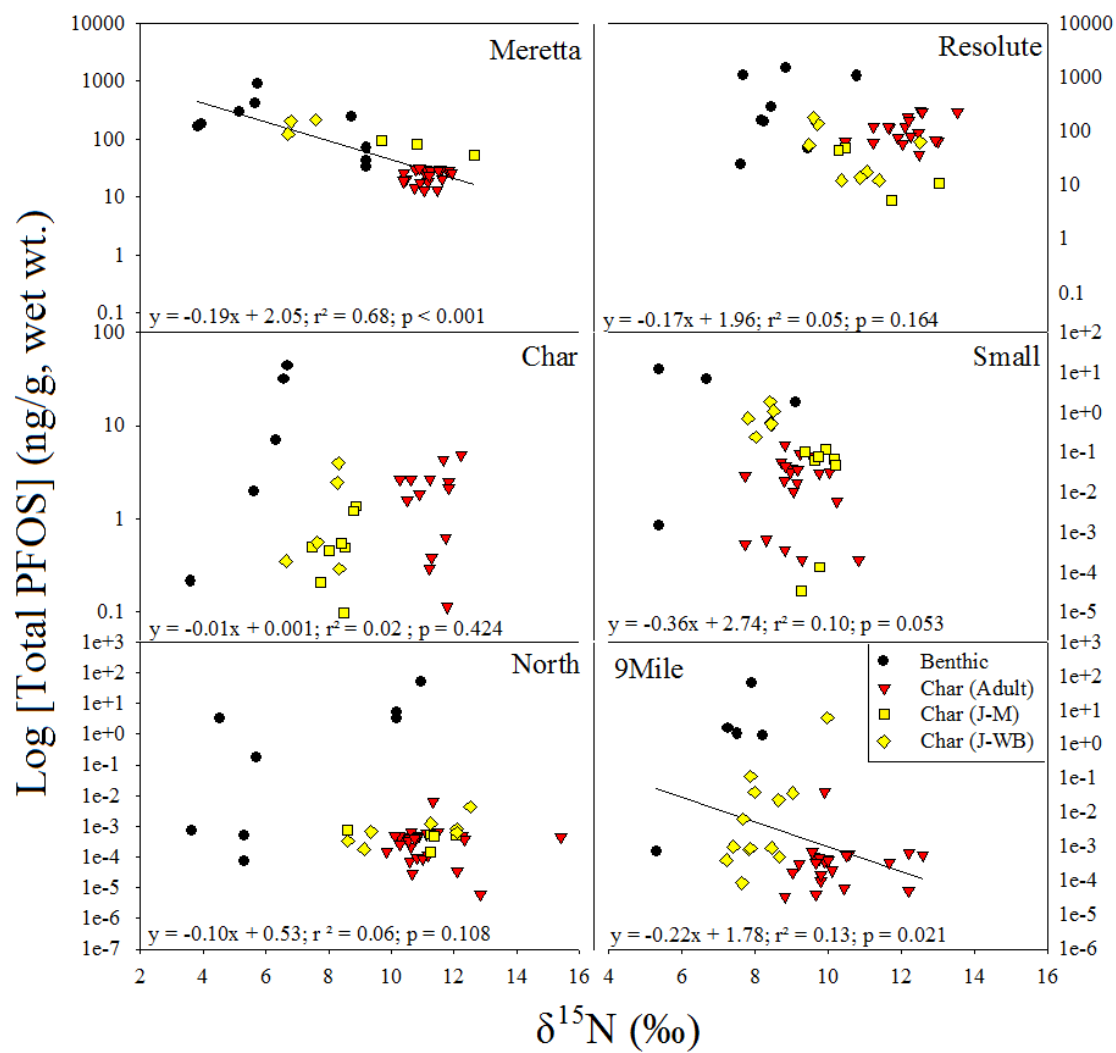


Figure SI-5 - Linear regressions of Total PFOS concentrations and $\delta^{15}\text{N}$ values in adult char, juvenile char (muscle samples [JC-M] and whole body homogenates [JC-WB]), and benthic invertebrates. All data are log-transformed.