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Perfluorinated Chemicals in Meromictic Lakes on the Northern Coast of Ellesmere Island, High Arctic Canada

JULIE VEILLETTE,[1,2] DEREK C.G. MUIR,[3] DERMOT ANTONIADES,[1] JEFF M. SMALL,[3] CHRISTINE SPENCER,[3] TRACEY N. LOEWEN,[4] JOHN A. BABALUK,[4] JAMES D. REIST[4] and WARWICK F. VINCENT[1]

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APPENDIX 1: SUPPLEMENTARY FIGURE AND TABLES

TABLE S1. Table of definitions of perfluorinated chemicals found in the environment.

Perfluorosulfonates	**PFSAs**
Perfluorobutane sulfonate	PFBS
Perfluorohexane sulfonate	PFHxS
Perfluoroheptane sulfonate	PFHpS
Perfluorooctane sulfonate	PFOS
Perfluorodecane sulfonate	PFDS
Perfluorocarboxylates	**PFCAs**
Perfluoroheptanoic acid	PFHpA
Perfluorooctanoic acid	PFOA
Perfluorononanoic acid	PFNA
Perfluorodecanoic acid	PFDA
Perfluororoundecanoic acid	PFUnA
Perfluorododecanoic acid	PFDoA
Perfluorosulfonamide	
Perfluorooctane sulfonylamide	PFOSA
Unsaturated fluorotelomer acids	
Perfluoroundecanoic acid	6:2 PFUA
Perfluoroundecanoic acid	8:2 PFUA
Perfluoroundecanoic acid	10:2 PFUA
Precursor alcohols	
Perfluorosulfonamido alcohols	PFSOHs
Fluorotelomer alcohols	FTOHs

[1] Département de Biologie & Centre d'études nordiques, Université Laval, Québec City, Québec G1V 0A6, Canada
[2] Corresponding author: julie.veillette.2@ulaval.ca
[3] Environment Canada, 867 Lakeshore Road, Burlington, Ontario L7R 4A6, Canada
[4] Fisheries and Oceans Canada, Central and Arctic Region, 501 University Crescent, Winnipeg, Manitoba R3T 2N6, Canada

TABLE S2. Samples analyzed for the Lake A catchment study of PFCs. Two subsamples of a single sample were taken at every site or depth, except for water at the outflow of Lake A (July 2007) and snow from the shore (August 2008), which are from one sample, and inflow at delta (inflow A; July 2007), which is the average of two samples. The average of the two subsamples from a single sample site is presented in this study. Surface water in the lake (centre and littoral) was sampled at 2 m depth. Inflow, moat, and outflow waters were sampled just below the surface. Snow from the north shore of the lake was sampled less than two days after snowfall.

Component	Date	Number of samples
Water (depth profile)	30 May 2008	1 profile (2, 10, 32 m)
	20 August 2008	1 profile (2, 10, 32 m)
Sediments	31 May 2008	1 core
Water (surface, 0–2 m)	12, 14 July 2007	5 sites
	30 May 2008	2 sites
	20 August 2008	4 sites
Snow	30 May 2008	2 sites (high and low in the catchment)
	19 August 2008	1 site

TABLE S3. Number of samples analyzed for the food web study. Whole individuals of zooplankton and whole body homogenate (*) or dorsal muscle of arctic char were analyzed.

Lake	Component	Date	Number of samples
A	Zooplankton	24 August 2008	1 (3 subsamples of the same tow)
	Arctic char	27–28 July 2002*	27
		16 July 2007	1
		30 May 2008	3
		24 August 2008	1
C2	Arctic char	25 May 2006	14

TABLE S4. Instrument detection limit (IDLs) and method detection limit (MDLs) for sediments, zooplankton and fish, and water and melted snow samples.

	Sediments (pg g^{-1} dry weight)		Zooplankton and fish (pg g^{-1} wet weight)		Water and snow (pg L^{-1})	
	IDL	MDL	IDL	MDL	IDL	MDL
PFBS	11.0	11.0	11.0	11.0	0.9	0.8
PFHxS	2.8	2.8	2.8	2.8	0.2	0.7
PFHpS	6.0	6.0	6.0	6.0	0.5	0.3
PFOS	6.0	4.3	6.0	6.0	0.5	14.5
PFDS	3.0	3.0	3.0	3.0	0.2	1.0
PFHpA (C7)	2.5	2.5	2.5	2.5	0.2	16.3
PFOA (C8)	1.3	3.4	1.3	1.3	0.1	15.3
PFNA (C9)	1.2	1.2	1.2	1.2	0.1	9.0
PFDA (C10)	1.4	1.4	1.4	1.4	0.1	6.6
PFUnA (C11)	1.0	1.0	1.0	1.0	0.1	3.4
PFDoA (C12)	1.2	1.2	1.2	1.2	0.1	3.6
PFOSA	1.1	1.1	1.1	1.1	0.1	1.0
6:2 PFUA	4.2	4.2	4.2	4.2	0.3	4.4
8:2 PFUA	1.0	1.0	1.0	1.0	0.1	0.1
10:2 PFUA	1.0	1.0	1.0	1.0	0.1	0.1

TABLE S5. Mean method or laboratory blank values for sediments (n = 3), zooplankton and fish (n = 6), and water and melted snow (n = 5). Standard deviations (SD) are indicated between parentheses. Non-detected (nd) means that no analyte was observed above instrument detection limits (IDLs) inferred from extrapolating the calibration curve to a signal-to-noise ratio of three. Sediment method blank consisted of sediments deeper than 5 cm, zooplankton and fish laboratory blanks consisted of methanol, and water method blanks consisted of the solvents and the cartridge.

	Sediments (pg g^{-1} dry weight)	Zooplankton and fish (pg g^{-1} wet weight)	Water and melted snow (pg L^{-1})
PFBS	nd	nd	2.1 (0.3)
PFHxS	nd	nd	0.7 (0.2)
PFHpS	nd	nd	1.0 (0.1)
PFOS	6 (1)	nd	7.5 (4.8)
PFDS	nd	nd	0.8 (0.3)
PFHpA (C7)	nd	nd	4.0 (5.4)
PFOA (C8)	46 (3)	nd	8.3 (5.1)
PFNA (C9)	nd	nd	2.4 (3.0)
PFDA (C10)	nd	nd	1.6 (2.2)
PFUnA (C11)	nd	nd	0.8 (1.1)
PFDoA (C12)	nd	nd	0.9 (1.2)
PFOSA	nd	nd	2.2 (0.3)
6:2 PFUA	nd	nd	2.2 (1.5)
8:2 PFUA	nd	nd	nd
10:2 PFUA	nd	nd	nd

TABLE S6. Mean extraction recoveries (%) of ^{13}C-mass labeled standards added to samples of sediments (n = 9), zooplankton (n = 3), fish (n = 56), water (n = 29) and melted snow (n = 5), with standard deviations (SD) in parentheses. Mean recoveries of PFNA for water and melted snow were calculated with the instrument performance internal standard that was added just before LC/MS/MS analysis. nd: non-detected.

	Sediments	Zooplankton	Fish	Water	Melted snow
PFHxS				180 (23)	168 (18)
PFOS	50 (16)	74 (24)	84 (29)	123 (19)	119 (12)
PFOA (C8)	67 (20)	122 (13)	122 (35)	102 (16)	119 (15)
PFNA (C9)	53 (17)	77 (9)	92 (29)	54 (9)	68 (7)
PFDA (C10)	52 (16)	73 (21)	71 (25)	88 (13)	45 (32)
PFUnA (C11)	52 (16)	73 (21)	71 (25)	72 (19)	28 (22)
PFDoA (C12)	43 (20)	27 (25)	48 (26)	55 (13)	25 (13)
6:2 PFUA	nd	nd	nd	56 (22)	103 (19)
8:2 PFUA	nd	nd	nd	63 (31)	54 (9)
10:2 PFUA	nd	nd	nd	43 (29)	12 (7)

TABLE S7. Mean and range of PFC concentrations (pg L^{-1}) in surface water samples and snow of the lake catchment for all compounds with concentrations above the method detection limits. Lake: sampling sites at the centre of the lake and in the littoral zone (30 m from the delta); delta: sampling sites at the moat around the delta and at the inflow of the delta (inflow A); inflow B: the inflow from Lake B; outflow: the outflow of Lake A to the Arctic Ocean; snow sampled in May 2008 was collected from a higher and lower location in the catchment; snow sampled in August 2008 was collected from the north shore and was sampled less than two days after snowfall.

	12 and 14 July 2007					30 May 2008				19–20 August 2008						
	Lake	Delta				Lake		Snow		Lake				Snow	Surface water	
	Littoral	Moat	Inflow A	Inflow B	Outflow	Centre	Littoral	High	Low	Centre	Littoral	Inflow B	Outflow	Shore	Mean (SD)	Range
PFBS	15	14	18	21	20	11	11	3	4	14	11	24	19	3	16(4)	11–24
PFHxS	4	3	6	9	7	8	6	3	2	10	6	24	13	0.7	9(6)	3–24
PFHpS	0.6	< 0.5	0.7	0.5	0.9	1	1	1	1	1	0.8	3	2	< 0.5	1(1)	0.4–3
PFOS	23	21	13	16	13	19	14	70	14	27	32	71	27	35	25(16)	13–71
PFHpA	69	60	59	93	88	90	100	213	244	98	134	190	165	37	104(42)	59–190
PFOA	131	137	85	145	142	125	126	199	239	113	175	245	216	27	149(46)	85–245
PFNA	91	108	57	89	90	99	109	195	273	143	162	163	192	32	118(41)	57–192
PFDA	23	27	3	8	8	18	12	29	41	15	20	18	18	0.3	15(7)	3–27
PFUnA	5	9	1	3	2	11	8	19	30	10	11	16	14	0.2	8(5)	1–16
ΣPFCs	362	379	243	385	371	382	387	732	848	431	552	754	666	134	446	243–754

TABLE S8. PFC concentrations (pg L^{-1}) at 2, 10, and 32 m in Lake A for all compounds with concentrations above the method detection limits.

	30 May 2008			20 August 2008		
	2 m	10 m	32 m	2 m	10 m	32 m
PFBS	11	11	0.8	14	17	< 0.9
PFHxS	8	7	1	10	11	1.3
PFHpS	1	1	0.9	1.4	2	0.8
PFOS	19	30	16	27	39	4
PFHpA	90	92	7	98	105	< 0.2
PFOA	125	113	25	113	120	9
PFNA	99	97	26	143	125	6
PFDA	18	14	11	15	26	4
PFUnA	11	11	5	10	11	1.4
ΣPFCs	382	376	93	431	456	27

TABLE S9. Results of the statistical analyses that tested for differences in the horizontal and vertical distribution of PFCs. a) Two-way ANOVA testing for differences between lake sites (centre and littoral) at the different sampling times. b) Two-way ANOVA testing for differences between inflows, outflow, lake sites and snow at the three sampling times. c) Two-way ANOVA testing for differences between sites and analytes in July 2007, May 2008, and August 2008. d) Tukey test to find which analytes were different between the lake sites and the snow in May 2008. e) Tukey test to find which sites were different in August 2008. f) Two-way ANOVA testing for differences between depths at the two sampling times. g) Tukey test to find which depths were different for the analytes that were significantly different between depths. Asterisk (*) indicates significance at α = 0.05.

a)	F_{site}	p_{site}	F_{time}	p_{time}
Total PFCs	1.086	0.487	2.822	0.388
PFBS	1.594	0.426	2.815	0.388
PFOS	< 0.001	0.984	2.985	0.379
PFHpA	3.072	0.330	5.793	0.282
PFOA	1.053	0.492	0.312	0.782
PFNA	13.690	0.168	116.311	0.065
PFDA	< 0.001	0.990	0.597	0.675

b)	F_{site}	p_{site}	F_{time}	p_{time}
Total PFCs	2.940	0.264	6.007	0.143
PFBS	4.764	0.178	4.444	0.184
PFOS	0.837	0.585	1.476	0.404
PFHpA	7.242	0.124	9.659	0.094
PFOA	1.205	0.483	2.136	0.319
PFNA	9.066	0.101	15.600	0.060
PFDA	1.758	0.383	0.584	0.631

c)	F_{site}	p_{site}	$F_{analyte}$	$p_{analyte}$
July 2007	1.027	0.376	74.832	< 0.001*
May 2008	101.943	< 0.001*	200.274	< 0.001*
August 2008	3.986	0.018*	70.427	< 0.001*

d)	q	p
Total PFCs	22.576	< 0.001*
PFBS	0.221	0.878
PFHxS	0.368	0.797
PFOS	1.422	0.327
PFHpA	7.349	< 0.001*
PFOA	5.137	0.002*
PFNA	7.180	< 0.001*
PFDA	1.101	0.445
PFUnA	0.818	0.570
PFDoA	0.160	0.911

e)	q	p
Inflow B vs. lake centre	4.655	0.014*
Inflow B vs. lake littoral	2.695	0.250
Inflow B vs. outflow	1.275	0.804
Outflow vs. lake centre	3.380	0.103
Outflow vs. lake littoral	1.420	0.748
Lake centre vs. lake littoral	1.960	0.519

f)	F_{depth}	p_{depth}	F_{time}	p_{time}
Total PFCs	27.485	0.035*	0.151	0.735
PFBS	5.166	0.162	0.177	0.715
PFOS	3.668	0.214	< 0.001	0.984
PFHpA	273.791	0.004*	3.280	0.212
PFOA	57.650	0.017*	1.195	0.388
PFNA	11.802	0.078	0.789	0.468
PFDA	4.220	0.192	0.984	0.426

g)	q_2 vs. 32 m	p_2 vs. 32 m	q_2 vs. 10 m	p_2 vs. 10 m	q_{10} vs. 32 m	p_{10} vs. 32 m
Total PFCs	8.970	0.044*	0.217	0.987	9.187	0.042*
PFHpA	27.955	0.005*	1.361	0.663	29.316	0.004*
PFOA	13.334	0.020*	0.374	0.963	12.960	0.021*

TABLE S10. Linear regression parameters for relationships between total PFCs, fork length, and age.

Regression	Lake A	Lake C2	Both lakes
PFCs vs age:			
n	31	14	45
r^2	0.024	0.118	0.026
p	0.402	0.229	0.288
PFCs vs fork length:			
n	32	14	46
r^2	0.011	0.003	0.024
p	0.575	0.848	0.307

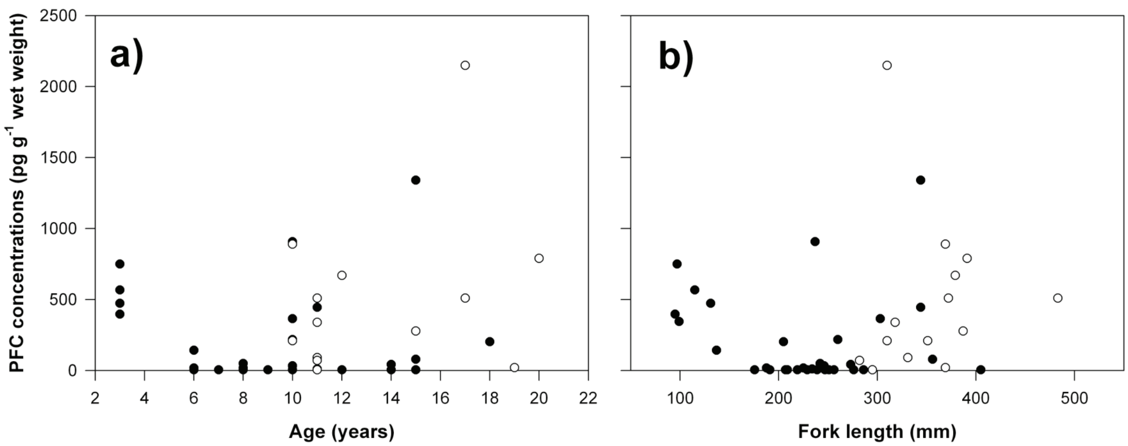

FIG. S1. a) Total PFC concentrations as a function of fish age and b) fork length for the arctic char populations of Lake A (solid circles) and Lake C2 (open circles).