SUPPORTING INFORMATION

High Methylmercury in Arctic and Subarctic Ponds is Related to Nutrient Levels in the Warming Eastern Canadian Arctic

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Supporting Information consists of 15 pages including detailed materials and methods, interlaboratory calibrations for aqueous THg and MeHg concentrations, 4 tables and 3 figures.

SI: MATERIALS AND METHODS

Study Sites

Bylot Island is a polar oasis located above the Arctic Circle, where conditions are cold and dry, with long-term averages of -34 °C in February and 6 °C in July, and average yearly precipitation of 191 mm.[1] Kuujjuarapik-Whapmagoostui is located in the Subarctic region of Quebec on the eastern shore of Hudson Bay[2], with long-term average temperatures of -23 °C in February and 11 °C in July, and with average yearly precipitation of 649 mm.[1]

Bylot thaw ponds are part of a dynamic network of syngenetic ice-wedge polygons that developed in the outwash plain of glacier C-79 after 6000 BP, modified by the accumulation of wind-blown and organic sediments that began after 3670 ± 110 BP and now reaching 2-3 m deep.[3] Bylot Island thaw ponds can be classified into two types: 1) low-center polygonal ponds created by the rise of peat polygon ridges generating cuvettes that fill with water and offer a stable environment for the establishment of thick microbial mats; and 2) trough ponds that form over melted ice wedges between the polygon mounds, an elongated aquatic system featuring peat erosion and higher turbidity therefore classified as thermokarstic.[4] Both types of ponds examined were no more than a few meters in diameter and generally less than 1.5 m in depth. At this site, the active soil layer is approximately 40 to 60 cm deep.[3] A few lakes were also sampled at this site, two kettle lakes formed on glacier ice melt depressions of about 11 m deep (named BYL36 and BYL37), a thermokarst lake 4.5 m deep (BYL66; Fig 2b), and some nearby oligotrophic lakes on rocky substrate (BYL39, BYL40, BYL49, BYL50; not shown). These seven larger aquatic systems on Bylot were categorized as “lakes” for this study, given their much larger surface area and depth than the polygonal and trough ponds.

The Subarctic sample sites included thermokarst, taiga/rock basin ponds sampled near the village of Kuujjuarapik-Whapmagoostui. At the Kwakwatanikapistikw lithalsa peatland (or "KWK") and Sasapimakwananisikw palsa bog ("SAS") (local Cree names of nearby rivers), thermokarst thaw ponds develop in depressions left after the ice has melted below mineral (KWK) or besides organic (SAS) permafrost mounds.[5,6] The ponds are surrounded by dense vegetation, mainly shrubs, small trees and mosses. They are 10 to 30 m in diameter and have a maximum depth of 3.5 m.[4,5] The taiga/rock basin ponds are formed on granite or carbonate-derived bedrock respectively and are 10-20 m in diameter with a maximum depth of 1 m.

Physico-chemical Sampling

DOC concentrations were measured using a Shimadzu TOC-5000A carbon analyzer (2006-2011) or an IO Aurora 1030 carbon analyzer (2012-2013), calibrated with potassium biphthalate. To measure the optical properties of DOM, water samples were filtered through pre-rinsed cellulose acetate filters (0.2 µm pore size; Advantec Micro Filtration Systems), and scanned from 250 to 800 nm on a spectrophotometer (Varian Cary 300). The absorption coefficient at 320 nm (a_{320}) was used as a proxy to quantify the water colour.

Total nitrogen was measured by flow injection analysis (Lachat Instruments) on unfiltered samples fixed by H_2SO_4 (0.15% final concentration) and digested with potassium persulfate. Using the same water samples as above for TN, total phosphorus (TP) was measured by spectrophotometry[7] (2006-2011) or by flow injection analysis (Astoria) (2012-2013).

To measure Chla concentration, water samples were passed through glass fiber filters (0.7 µm nominal pore size; Advantec MFS) and kept frozen at -80 °C until the extraction of pigments in 95% boiling ethanol.[8] The fluorescence was measured with a spectrofluorometer (Varian Cary

Eclipse) before and after acidification (to correct for interference by pheophytin) and the Chla concentration calculated as in Jeffrey and Welschmeyer (1997).

Water samples for anion and cation analysis were filtered through pre-rinsed cellulose acetate filters or pre-combusted glass-fibre filters (0.7 um pore size, Whatman GF-F), and they were preserved with HNO_3 (0.15% final concentration). Anions were quantified using ion chromatography (either Dionex Corp. or Waters, IC-Pak A and C columns). Cations were analyzed with inductively coupled plasma optical emission spectrometry (ICP-OES, Varian VISTA AX CCD) in 2006-2011, or with an atomic absorption spectrophotometer (AAS, Agilent) in 2012-2013. Water samples for major metals were filtered and preserved with HNO_3 (2%) before analysis by inductively coupled plasma mass spectrometry (ICP-MS, Perkin-Elmer NexION 300x).

Dissolved CO_2 and CH_4 were determined following Hesslein et al. (1990)[10] using the equilibration of 2 liters of water into 20 mL of ambient air for 3 min. The headspace was sampled in duplicated vials (red stopper Vacutainer®) previously flushed with helium and vacuumed. Gas samples were taken within 5 minutes after collecting the water and were kept at 4 °C until analysis by gas chromatography (Varian 3800 with a COMBI PAL head space injection system and a CP-Poraplot Q 25 m × 0.53 mm column and flame ionization detector).

Aqueous THg and MeHg Concentrations

Table A: Results for the CALA Laboratory Intercalibration for THg in 2014.

Matrix	Analyte	Units	Assigned Value	Assigned Uncertainty	Reported Value	S -Value	Z-Score	PT-Score	Summary
Water	Mercury	µg L^{-1}	0.67	0.01	0.63	0.11	-0.364		
Water	Mercury	µg L^{-1}	1.75	0.02	1.62	0.20	-0.650		
Water	Mercury	µg L^{-1}	2.47	0.03	2.33	0.27	-0.519		
Water	Mercury	µg L^{-1}	4.47	0.05	4.24	0.45	-0.511	92	Acceptable

Table B: Results for the Northern Contaminants Interlaboratory Quality Assurance Program (NCP III QA/QC Program 2014-2015)

Matrix	Analyte	Units	Reported Value	Design	Median	SD	n	%RECUP
Water	THg	ng L^{-1}	5053	5000	5200	342	15	101%
Water	MeHg	ng L^{-1}	935	1000	960	26.2	4	93.5%

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TABLE S1. Mercury concentrations and physicochemical characteristics, including temperature (Temp), conductivity (Cond), dissolved oxygen (DO), dissolved organic carbon (DOC), chlorophyll *a (Chla)*, total mercury (THg) and methylmercury (MeHg), of a) trough ponds, b) polygonal ponds and c) lakes on Bylot Island.

Site	**Year Sampled**	**Temp (°C)**	**pH**	**Cond (µS cm^{-1})**	**DO (mg L^{-1})**	**a_{320} (m^{-1})**	**DOC (mg L^{-1})**	**CO_2 (µM)**	**CH_4 (µM)**	**Ca (mg L^{-1})**	**Mg (mg L^{-1})**	**K (mg L^{-1})**
a) Trough Ponds (n = 18)												
BYL24	2009 - 2010	14.52	7.17	43	11.97	32.4	8.67	51.4	2.09	4.82	2.87	0.65
BYL27	2008 - 2010	10.50	6.90	67	10.46	32.4	10.10	87.5	3.40	4.64	3.59	1.52
BYL33	2009	13.31	7.55	100	10.05	18.4	7.70	46.1	4.63	10.15	8.56	1.49
BYL38	2008 - 2010	9.17	6.57	51	8.07	54.3	10.53	123.5	3.70	3.37	2.52	1.12
BYL47	2008 - 2010	12.06	6.84	95	10.17	37.9	12.45	114.1	3.94	6.37	4.73	1.68
BYL48	2009 - 2010	15.32	6.59	79	10.87	32.5	11.53	94.8	3.83	7.13	4.76	1.24
BYL59	2009 - 2010	8.72	6.78	62	9.14	47.4	12.40	186.0	5.17	3.66	2.93	1.36
BYL63	2009 - 2010	12.99	6.44	139	3.81	269.4	33.00	450.7	4.83	17.21	10.16	2.20
BYL64	2009 - 2010	13.68	7.18	61	10.80	44.7	15.95	43.0	5.57	4.89	3.59	1.23
BYL67	2009 - 2010	13.16	6.85	56	11.61	50.0	12.63	73.0	5.70	5.06	3.43	1.10
BYL68	2009	8.03	5.93	62	5.55	25.7	11.30	-	-	4.19	3.27	1.16
BYL69	2009	11.38	6.13	233	4.50	19.1	9.40	609.1	19.90	18.63	10.59	2.23
BYL70	2009	13.55	6.87	448	9.81	41.6	23.20	178.4	9.94	20.37	17.87	2.01
BYL71	2009	14.20	7.47	100	10.97	20.6	10.70	47.4	3.06	8.11	5.15	1.63
BYL72	2009	14.45	7.57	141	12.25	21.5	11.40	24.3	4.74	12.48	7.41	2.77
BYL74	2009 - 2010	11.43	6.72	85	8.22	76.3	17.35	204.8	13.49	10.07	5.90	1.56
BYL75	2009	15.64	6.73	125	10.04	38.1	11.50	148.0	8.66	15.64	7.21	1.69
BYL76	2009	18.16	7.00	116	9.21	24.9	8.90	141.8	10.52	13.66	6.52	1.81
	Mean	12.79	6.85	115	9.30	49.30	13.26	154.4	6.66	9.47	6.17	1.58
	Median	13.24	6.85	90	10.05	35.19	11.45	114.1	4.83	7.62	4.95	1.54
	Std dev	2.60	0.44	95	2.45	56.89	6.12	153.9	4.58	5.58	3.83	0.50
	Min	8.03	5.93	43	3.81	18.43	7.70	24.3	2.09	3.37	2.52	0.65
	Max	18.16	7.57	448	12.25	269.45	33.00	609.1	19.90	20.37	17.87	2.77
b) Polygonal Ponds (n = 9)												
BYL01	2008 - 2010	13.32	8.44	68	12.06	10.2	7.20	11.1	0.72	4.39	3.24	2.00
BYL22	2008 - 2010	12.14	7.85	51	11.17	16.0	6.55	30.6	1.59	2.41	1.51	2.07
BYL31	2009	14.32	8.21	119	9.98	19.8	8.60	9.1	2.63	13.30	9.71	1.78
BYL44	2008 - 2010	11.89	8.12	56	10.70	11.0	8.38	10.1	1.31	5.42	3.62	1.74
BYL51	2008 - 2010	12.07	8.68	86	12.09	14.2	9.40	10.2	2.98	6.05	3.95	2.37
BYL61	2009	14.82	8.48	70	10.65	8.2	-	-	-	-	-	-
BYL65	2009	14.32	7.93	85	11.46	15.4	10.10	17.3	0.68	8.39	5.11	2.38
BYL73	2009	15.77	6.52	119	6.75	77.9	15.20	280.1	5.05	15.86	6.67	1.21
BYL78	2010	-	-	-	-	-	-	-	-	-	-	-
	Mean	13.58	8.03	82	10.61	21.6	9.35	52.7	2.14	7.98	4.83	1.94
	Median	13.82	8.17	78	10.93	14.8	8.60	11.1	1.59	6.05	3.95	2.00
	Std dev	1.45	0.67	26	1.72	23.1	2.85	100.6	1.56	4.91	2.68	0.41
	Min	11.89	6.52	51	6.75	8.2	6.55	9.1	0.68	2.41	1.51	1.21
	Max	15.77	8.68	119	12.09	77.9	15.20	280.1	5.05	15.86	9.71	2.38
c) Lakes (n = 7)												
BYL36	2009	16.12	7.58	90	10.88	6.29	4.1	18.22	0.91	4.54	2.80	1.30
BYL37	2010	-	-	-	-	-	5.3	-	-	-	-	-
BYL39	2008	6.35	6.58	9	11.93	3.94	1.3	-	-	0.80	0.29	-
BYL40	2008	6.08	6.73	15	12.85	3.56	1.5	-	-	1.29	0.49	-
BYL49	2008	8.89	6.95	12	12.02	3.40	1.5	-	-	5.79	4.23	-
BYL50	2008	13.34	6.74	13	11.19	1.34	1.0	-	-	1.21	0.46	-
BYL66	2009	-	7.1	-	10.43	-	5.3	-	-	-	-	-
	Mean	10.16	6.95	28	11.55	3.71	2.86	-	-	2.73	1.65	-
	Median	8.89	6.85	13	11.56	3.56	1.50	-	-	1.29	0.49	-
	Std dev	4.43	0.36	35	0.88	1.77	1.96	-	-	2.28	1.78	-
	Min	6.08	6.58	9	10.43	1.34	1.00	-	-	0.80	0.29	-
	Max	16.12	7.58	90	12.85	6.29	5.30	-	-	5.79	4.23	-

TABLE S1 *continued*

Site	**Na ($mg L^{-1}$)**	**Cl ($mg L^{-1}$)**	**SO_4 ($mg L^{-1}$)**	**Fe ($\mu g L^{-1}$)**	**Mn ($\mu g L^{-1}$)**	**TP ($\mu g L^{-1}$)**	**TN ($\mu g L^{-1}$)**	**Chl*a* ($\mu g L^{-1}$)**	**THg ($ng L^{-1}$)**	**MeHg ($ng L^{-1}$)**	**%MeHg (MeHg/THg)**
a) Trough Ponds (n = 18)											
BYL24	2.99	2.75	1.08	101.25	2.99	31.8	387.3	1.46	3.34	0.81	24.3
BYL27	4.46	7.36	1.53	90.50	2.58	26.3	641.2	1.17	2.96	0.75	25.3
BYL33	1.99	2.35	0.33	48.23	2.05	14.5	286.8	0.40	1.45	0.31	21.4
BYL38	3.79	5.92	2.48	385.03	6.55	63.0	586.8	0.99	3.59	1.23	34.3
BYL47	5.19	8.44	2.25	97.79	9.47	45.3	775.5	1.38	3.46	0.97	28.0
BYL48	5.28	6.03	2.14	164.59	5.43	24.3	571.6	1.99	2.36	0.41	17.4
BYL59	4.30	5.93	3.52	109.38	11.36	50.8	811.4	26.60	3.52	0.74	21.0
BYL63	10.49	12.60	0.09	1637.50	556.14	359.8	4366.3	1.89	21.82	10.58	48.5
BYL64	5.17	7.20	0.07	154.00	12.00	31.9	1174.9	1.23	2.41	0.65	27.0
BYL67	3.84	4.79	1.16	220.29	5.34	49.4	776.0	3.71	4.27	1.39	32.6
BYL68	4.57	6.68	3.06	53.03	6.95	-	-	-	2.35	0.61	26.0
BYL69	18.84	28.60	0.73	27.80	140.95	-	-	0.80	1.30	0.51	39.2
BYL70	60.15	56.43	2.35	46.89	20.52	-	-	2.50	2.16	0.34	15.7
BYL71	7.17	9.76	2.77	11.25	3.68	-	-	1.30	1.65	0.39	23.6
BYL72	9.43	13.64	3.59	40.65	4.25	-	-	3.20	1.53	0.50	32.7
BYL74	2.28	1.50	3.13	270.38	20.31	-	645.0	1.29	6.28	2.16	34.4
BYL75	3.74	1.64	2.35	94.88	16.67	-	-	0.70	1.87	0.61	32.6
BYL76	4.06	2.04	1.01	12.13	10.39	-	-	0.90	1.26	0.14	11.1
Mean	8.76	10.20	1.87	198.09	46.54	69.72	1002.1	3.03	3.75	1.28	27.5
Median	4.52	6.36	2.20	96.34	8.21	38.64	645.0	1.30	2.39	0.63	26.5
Std dev	13.42	13.16	1.15	372.18	131.05	102.95	1139.8	6.14	4.68	2.37	9.0
Min	1.99	1.50	0.07	11.25	2.05	14.55	286.8	0.40	1.26	0.14	11.1
Max	60.15	56.43	3.59	1637.50	556.14	359.75	4366.3	26.60	21.82	10.58	48.5
b) Polygonal Ponds (n = 9)											
BYL01	4.12	5.13	1.47	29.94	0.71	17.1	412.0	1.32	1.55	0.29	18.7
BYL22	2.15	5.01	2.19	55.75	1.13	46.8	364.7	1.66	2.93	0.27	9.2
BYL31	2.86	2.08	1.06	43.64	4.62	12.9	334.3	0.60	1.24	0.21	16.9
BYL44	3.69	4.69	1.32	32.51	1.07	15.3	572.3	2.66	1.31	0.23	17.6
BYL51	5.10	7.02	1.47	28.58	0.58	15.6	450.9	1.46	1.66	0.20	12.0
BYL61	-	-	-	-	-	-	-	0.40	1.63	0.08	4.9
BYL65	3.49	4.65	6.08	48.79	1.56	21.4	415.8	1.00	2.33	0.34	14.6
BYL73	3.47	3.21	0.40	352.36	13.94	-	-	0.30	1.34	0.23	17.2
BYL78	-	-	-	-	-	-	-	-	2.30	0.16	7.0
Mean	3.55	4.54	2.00	84.51	3.37	21.5	425.0	1.18	1.81	0.22	13.1
Median	3.49	4.69	1.47	43.64	1.13	16.3	413.9	1.16	1.63	0.23	14.6
Std dev	0.93	1.56	1.88	118.55	4.86	12.72	83.05	0.78	0.58	0.08	5.1
Min	2.15	2.08	0.40	28.58	0.58	12.9	334.3	0.30	1.24	0.08	4.9
Max	5.10	7.02	6.08	352.36	13.94	46.8	572.3	2.66	2.93	0.34	18.7
c) Lakes (n = 7)											
BYL36	6.82	13.29	2.47	6.06	1.30	8.5	194.5	1.7	1.01	0.04	4.0
BYL37	-	-	-	-	-	-	-	-	1.49	0.06	4.0
BYL39	-	0.31	0.75	9.61	-	3.14	105.9	1.67	1.07	0.03	2.8
BYL40	-	0.42	1.43	7.78	-	4.73	104.5	0.91	0.86	0.05	5.8
BYL49	-	0.56	0.66	881.57	-	3.82	137.0	1.19	0.99	0.00	0.0
BYL50	-	0.69	0.81	7.85	-	3.67	94.8	0.62	0.68	0.03	4.4
BYL66	-	8.33	2.26	-	-	-	-	-	1.55	0.02	1.3
Mean	-	3.93	1.40	182.57	-	4.77	127.3	1.22	1.09	0.03	3.2
Median	-	0.63	1.12	7.85	-	3.82	105.9	1.19	1.01	0.03	4.0
Std dev	-	5.55	0.80	390.75	-	2.16	40.8	0.47	0.32	0.02	2.0
Min	-	0.31	0.66	6.06	-	3.14	94.8	0.62	0.68	0.00	0.0
Max	-	13.29	2.47	881.57	-	8.50	194.5	1.70	1.55	0.06	5.8

TABLE S2. Mercury concentrations and physicochemical characteristics (see abbreviations in Table S1) of a) the surface and b) bottom waters of thermokarst ponds and c) in tundra and rock ponds near Kuujjuarapik-Whapmagoostui.

Site	Year Sampled	Temp (°C)	pH	Cond (μS cm^{-1})	DO (mg L^{-1})	A_{320} (m^{-1})	DOC (mg L^{-1})	CO_2 (μM)	CH_4 (μM)	Ca (mg L^{-1})	Mg (mg L^{-1})	K (mg L^{-1})
a) Thaw Ponds: Surface Waters (n = 12)												
Kwk1	2006, 2009	19.09	6.60	46	9.59	42.2	7.85	52.8	0.38	1.12	1.09	0.88
Kwk2	2006, 2009	16.07	6.88	38	8.56	26.3	5.73	73.5	0.44	0.88	0.95	0.59
Kwk6	2006, 2009	17.65	6.59	61	9.81	12.9	3.96	33.9	0.35	1.67	1.77	1.16
Kwk11	2006, 2009	17.80	6.10	27	9.40	53.7	11.01	141.6	1.411	1.41	0.96	0.56
Kwk12	2006, 2009	18.57	6.80	36	9.08	26.5	6.43	51.0	0.24	0.63	0.81	0.43
Kwk16	2006, 2009	24.25	7.24	53	9.25	38.9	7.86	50.3	0.38	1.24	1.41	0.94
Kwk19	2013	16.57	6.46	37	6.52	-	9.81	-	-	1.75	2.09	1.96
Kwk20	2006, 2009	17.98	6.82	-	9.17	42.2	7.41	71.5	0.28	0.88	1.04	0.87
Kwk23	2009	19.14	7.14	48	9.21	32.1	6.64	47.3	0.35	1.06	1.23	0.94
Kwk38	2009	19.72	7.25	204	9.43	25.1	8.53	72.5	0.92	5.50	7.40	2.17
SAS1G	2013	14.76	5.91	84	2.49	-	28.06	-	-	8.70	3.34	1.01
SAS2A	2013	14.35	5.82	49	5.78	-	16.58	-	-	2.56	1.32	0.13
	Mean	17.99	6.63	61	8.19	33.3	9.99	66.0	0.53	2.29	1.95	0.97
	Median	17.89	6.70	48	9.19	32.1	7.85	52.9	0.38	1.33	1.28	0.91
	Std dev	2.60	0.49	47	2.19	12.2	6.52	31.4	0.39	2.40	1.85	0.59
	Min	14.35	5.82	27	2.49	12.9	3.96	33.9	0.24	0.63	0.81	0.13
	Max	24.25	7.25	204	9.81	53.7	28.06	141.6	1.41	8.70	7.40	2.17
b) Thaw Ponds: Bottom Waters (n = 9)												
Kwk1	2009	7.75	6.11	155	0.23	44.2	8.30	405.0	93.8	2.78	1.65	0.99
Kwk2	2009	8.28	6.24	200	0.28	47.9	6.12	-	-	2.49	1.80	0.96
Kwk6	2009	9.06	6.30	265	0.14	19.7	4.17	421.8	145.2	-	-	-
Kwk11	2009	-	-	-	9.31	53.9	11.93	131.3	1.24	0.67	0.90	0.44
Kwk12	2009	7.35	6.08	247	0.19	63.1	7.45	761.2	259.0	3.37	1.93	1.17
Kwk16	2009	4.88	5.94	163	0.19	44.8	7.21	476.9	114.9	1.81	1.54	0.93
Kwk20	2009	5.25	6.14	145	0.15	106.9	9.27	815.5	311.9	3.62	2.08	1.30
Kwk23	2009	13.13	6.87	209	0.18	87.6	7.47	570.1	131.6	2.56	1.79	1.18
Kwk38	2009	7.75	6.11	155	3.69	24.0	6.28	106.9	0.479	5.54	7.45	2.14
	Mean	7.93	6.22	192	1.60	54.7	7.58	461.1	132.3	2.85	2.39	1.14
	Median	7.75	6.13	182	0.19	47.9	7.45	449.4	123.3	2.67	1.79	1.08
	Std dev	2.54	0.28	46	3.12	28.1	2.18	258.2	110.1	1.42	2.07	0.48
	Min	4.88	5.94	145	0.14	19.7	4.17	106.9	0.479	0.67	0.90	0.44
	Max	13.13	6.87	265	9.31	106.9	11.93	815.5	311.9	5.54	7.45	2.14
c) Taïga and Rock Ponds (n = 12)												
R104	2012 - 2013	14.07	6.10	26	8.34	-	11.73	-	-	1.06	0.81	0.57
R202	2012 - 2013	13.09	7.39	189	11.13	-	13.98	-	-	8.22	5.40	1.09
R206	2012 - 2013	12.18	7.63	280	11.46	-	18.27	-	-	12.53	8.25	1.66
R301	2012 - 2013	12.55	7.24	131	9.34	-	14.49	-	-	7.75	3.31	1.40
T102	2012 - 2013	16.30	7.24	55	11.47	-	13.58	-	-	6.99	2.17	0.29
T104	2012 - 2013	14.64	7.15	61	10.55	-	11.69	-	-	8.59	1.56	0.70
T107	2012 - 2013	13.32	6.99	39	11.10	-	15.42	-	-	3.29	1.35	0.41
T207	2012 - 2013	12.83	6.80	61	10.91	-	6.78	-	-	7.04	1.28	0.71
R217	2013	12.84	7.78	514	12.73	-	10.11	-	-	2.90	5.29	3.32
Km2.5	2013	13.71	5.69	26	9.36	-	14.07	-	-	4.55	1.33	0.50
Km4	2013	16.01	7.04	50	10.32	-	9.49	-	-	0.93	0.62	0.35
WP2	2013	11.43	6.60	42	9.03	-	14.88	-	-	3.28	1.19	0.29
	Mean	13.58	6.97	123	10.48	-	12.87	-	-	5.59	2.71	0.94
	Median	13.20	7.09	58	10.73	-	13.78	-	-	5.77	1.45	0.63
	Std dev	1.47	0.60	145	1.25	-	3.08	-	-	3.49	2.39	0.87
	Min	11.43	5.69	26	8.34	-	6.78	-	-	0.93	0.62	0.29
	Max	16.30	7.78	514	12.73	-	18.27	-	-	12.53	8.25	3.32

TABLE S2 *Continued*

Site	Na (mg L^{-1})	Cl (mg L^{-1})	SO$_4$ (mg L^{-1})	Fe (µg L^{-1})	Mn (µg L^{-1})	TP (µg L^{-1})	TN (µg L^{-1})	Chl*a* (µg L^{-1})	THg (ng L^{-1})	MeHg (ng L^{-1})	%MeHg (MeHg/THg)
a) Thaw Ponds: Surface Waters (n = 12)											
Kwk1	3.13	3.89	0.54	477.32	7.55	43.1	251	3.91	3.05	0.09	3.0
Kwk2	2.76	3.80	0.24	293.69	5.90	35.6	289	4.30	1.80	0.07	4.1
Kwk6	2.86	4.23	0.47	136.13	11.00	34.9	228	8.20	0.76	0.02	2.7
Kwk11	3.57	3.82	0.10	271.61	4.16	43.2	597	8.72	3.36	0.12	3.4
Kwk12	2.74	3.72	0.05	234.93	4.67	24.3	312	2.76	1.53	0.14	8.9
Kwk16	2.94	3.57	0.41	462.17	8.01	74.7	234	8.56	1.74	0.09	5.1
Kwk19	4.79	3.54	0.39	930.11	7.79	96.3	673	10.40	2.89	0.12	4.2
Kwk20	3.15	-	-	45.884	1.146	80.0	263	9.65	2.07	0.19	9.2
Kwk23	3.08	4.17	0.06	303.48	11.12	70.2	228	12.71	1.94	0.18	9.3
Kwk38	7.34	4.03	12.52	500.68	8.70	69.9	431	3.67	1.57	0.28	17.7
SAS1G	6.14	5.38	9.09	2462.28	32.40	237.3	2899	4.67	4.35	3.56	81.9
SAS2A	6.85	5.26	0.10	492.22	5.06	15.3	599	1.97	2.79	0.08	2.9
Mean	4.11	4.13	2.18	550.87	8.96	68.73	584	7.01	2.32	0.41	12.7
Median	3.14	3.89	0.39	382.83	7.67	56.56	301	6.43	2.01	0.12	4.7
Std dev	1.72	0.63	4.34	643.05	7.92	58.56	747	4.06	0.99	0.99	22.2
Min	2.74	3.54	0.05	45.88	1.15	15.30	228	1.97	0.76	0.02	2.7
Max	7.34	5.38	12.52	2462.28	32.40	237.31	2899	14.30	4.35	3.56	81.9
b) Thaw Ponds: Bottom Waters (n = 9)											
Kwk1	3.07	4.89	0.62	83.98	11.82	175.9	267	66.4	7.03	0.55	7.8
Kwk2	3.29	5.18	0.25	250.75	30.44	341.5	496	180.2	8.56	3.07	35.9
Kwk6	-	-	0.24	105.70	na	197.7	389	87.2	2.02	0.52	25.7
Kwk11	3.07	3.68	0.08	31.61	0.61	48.1	409	14.6	2.04	0.13	6.4
Kwk12	3.03	5.17	0.25	286.48	22.61	207.1	448	158.9	3.02	2.36	78.1
Kwk16	2.63	3.37	0.30	131.75	11.27	167.1	399	22.4	4.44	1.55	34.9
Kwk20	3.99	6.60	0.28	512.29	17.37	377.0	289	203.4	5.62	2.13	37.9
Kwk23	3.13	4.55	0.17	410.61	15.07	431.8	267	37.1	4.53	3.01	66.4
Kwk38	7.34	4.03	12.47	40.37	1.26	75.4	351	7.4	1.38	0.35	25.4
Mean	3.69	4.68	1.63	205.95	13.81	224.6	368	86.4	4.29	1.52	35.4
Median	3.10	4.72	0.25	131.75	13.44	197.7	389	66.4	4.44	1.55	34.9
Std dev	1.52	1.03	4.07	170.47	10.07	132.3	81	75.9	2.45	1.17	24.0
Min	2.63	3.37	0.08	31.61	0.61	48.1	267	7.4	1.38	0.13	6.4
Max	7.34	6.60	12.47	512.29	30.44	431.8	496	203.4	8.56	3.07	78.1
c) Taïga and Rock Ponds (n = 12)											
R104	3.97	4.66	0.77	491.27	7.80	43.5	736	4.76	10.62	1.24	11.7
R202	16.69	34.14	12.87	278.78	9.33	15.1	707	2.05	4.59	0.59	12.9
R206	23.73	34.07	4.05	371.34	22.88	12.2	705	0.96	5.14	0.41	8.0
R301	10.96	15.64	1.96	313.66	4.91	19.7	628	1.53	7.80	0.57	7.4
T102	3.79	4.41	1.48	67.43	1.51	11.8	610	1.16	7.22	0.20	2.7
T104	3.87	5.50	2.58	78.20	2.17	7.8	603	1.57	4.71	0.25	5.2
T107	4.18	12.00	2.57	103.42	3.41	24.8	741	2.99	7.04	0.28	3.9
T207	3.78	5.03	1.84	53.64	1.53	5.2	208	0.98	3.47	0.11	3.2
R217	52.01	56.31	8.82	519.68	8.97	65.3	804	1.74	9.07	0.80	8.8
Km2.5	4.28	4.78	1.67	164.91	5.51	12.4	466	1.33	11.16	0.14	1.3
Km4	3.55	5.13	0.74	281.19	47.82	5.3	258	0.46	4.80	0.30	6.2
WP2	4.27	5.42	1.49	156.91	2.43	12.6	394	0.62	7.22	0.21	2.9
Mean	11.26	15.59	3.40	240.03	9.86	19.6	572	1.68	6.90	0.42	6.2
Median	4.23	5.46	1.90	221.84	5.21	12.5	619	1.43	7.13	0.29	5.7
Std dev	14.37	16.91	3.69	162.12	13.33	17.8	197	1.18	2.47	0.33	3.7
Min	3.55	4.41	0.74	53.64	1.51	5.2	208	0.46	3.47	0.11	1.3
Max	52.01	56.31	12.87	519.68	47.82	65.3	804	4.76	11.16	1.24	12.9

TABLE S3. GPS Coordinates (dd°mm'ss") from trough ponds, polygonal ponds and lakes on Bylot Island (Nunavut), and thaw ponds and tundra/rock ponds near Kuujjuarapik-Whapmagoostui (Nunavik, QC).

Bylot	**Site**	**Latitude (°N)**	**Longitude (°W)**	**Kuujjuarapik-Whapmagoostui**	**Site**	**Latitude (°N)**	**Longitude (°W)**
Trough Pond	BYL24	73°09'25	79°58'42	**Thaw Pond**	Kwk1	55°19'51	77°30'09
	BYL27	73°09'20	79°59'14		Kwk2	55°19'50	77°30'10
	BYL33	73°15'11	80°04'12		Kwk6	55°19'56	77°30'06
	BYL38	73°09'19	79°59'04		Kwk11	55°19'49	77°30'13
	BYL47	73°09'37	79°58'50		Kwk12	55°19'48	77°30'14
	BYL48	73°09'23	79°59'13		Kwk16	55°19'47	77°30'11
	BYL59	73°09'23	79°59'02		Kwk19	55°19'47	77°30'16
	BYL63	72°52'55	79°52'58		Kwk20	55°19'56	77°30'08
	BYL64	72°52'55	79°53'01		Kwk23	55°19'57	77°30'06
	BYL67	73°09'15	79°59'14		Kwk38	55°19'58	77°29'58
	BYL68	na	na		SAS1G	55°13'09	77°42'29
	BYL69	73°08'39	80°04'05		SAS2A	55°13'35	77°41'49
	BYL70	73°09'59	80°05'57	**Taiga/ Rock Ponds**	R104	55°17'48	77°45'18
	BYL71	73°08'56	80°02'49		R202	55°18'41	77°44'41
	BYL72	73°09'25	80°00'51		R206	55°18'39	77°44'46
	BYL74	73°10'46	79°52'15		R301	55°18'44	77°44'26
	BYL75	73°11'08	79°50'46		T102	55°16'35	77°44'08
	BYL76	73°11'02	79°48'29		T104	55°16'39	77°49'00
Polygonal Pond	BYL01	73°09'28	79°58'50		T107	55°16'39	77°44'08
	BYL22	73°09'28	79°58'44		T207	55°16'54	77°43'44
	BYL31	73°15'11	80°04'17		R217	55°18'37	77°44'49
	BYL44	73°09'28	79°58'46		Km2.5	55°18'37	77°44'49
	BYL51	73°09'26	79°59'11		Km4-1	55°19'27	77°42'55
	BYL61	73°15'11	80°04'17		WP2	55°16'58	77°44'07
	BYL65	73°09'40	79°58'04				
	BYL73	na	na				
	BYL78	73°06'28	80°07'53				
Lake (Bylot)	BYL36	73°09'34	79°58'13				
	BYL37	73°09'19	79°58'29				
	BYL39	73°02'48	79°25'34				
	BYL40	73°02'40	79°25'45				
	BYL49	73°00'47	79°28'51				
	BYL50	73°00'48	79°27'14				
	BYL66	73°09'14	79°59'34				

Table S4. Simple linear regression models for the most highly correlated environmental variables for surface waters of all data (n=58), separately for each region (Bylot: n = 34 or Kuujjuarapik: n = 24) and on a subset of data from the bottom waters of stratified ponds (n = 9).

Sample Site	n	Modeled Variable	Independent Variables	*p*-value	Adjusted R^2
All Sites: Surface Waters	58	THg	a320	< 0.01	0.57
			DOC	< 0.01	0.45
			TN	0.01	0.33
			Fe	0.02	0.26
		MeHg	TN	< 0.01	0.61
			DOC	< 0.01	0.57
			a320	< 0.01	0.39
			CH_4	< 0.01	0.37
Bylot Island Surface Waters	34	MeHg	Fe	< 0.01	0.65
			TP	< 0.01	0.84
			Mn	< 0.01	0.37
			Hg(II)	< 0.01	0.53
Kuujuarapik Surface Waters	24	MeHg	DOC	< 0.01	0.45
			TN	< 0.01	0.54
			Hg(II)	>0.01	ns
Kuujuarapik Bottom Waters	9	MeHg	Mn	0.01	0.84
			TP	< 0.05	0.79
			Fe	< 0.01	0.85
			CO_2	< 0.05	0.71
			CH_4	< 0.05	0.56
			DO	0.02	0.49

FIGURE S1. Map of the study area, showing Kuujjuarapik-Whapmagoostui (Quebec, Canada) and Bylot Island (Nunavut, Canada).

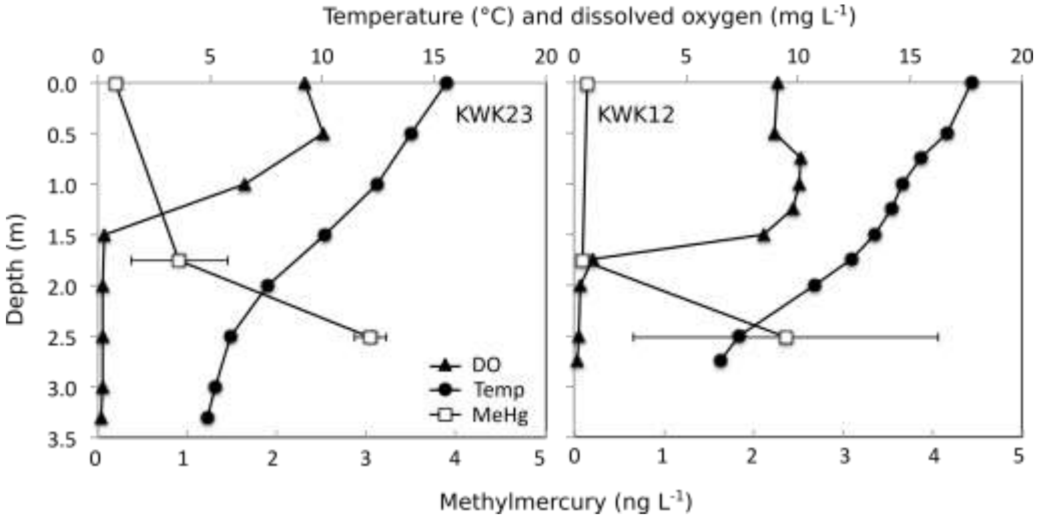

FIGURE S2. Vertical profiles of dissolved oxygen concentrations (DO, mg L^{-1}), temperature (Temp, °C) and MeHg concentrations (ng L^{-1}) for two stratified thermokarst ponds sampled near Kuujjuarapik-Whapmagoostui in August 2009.

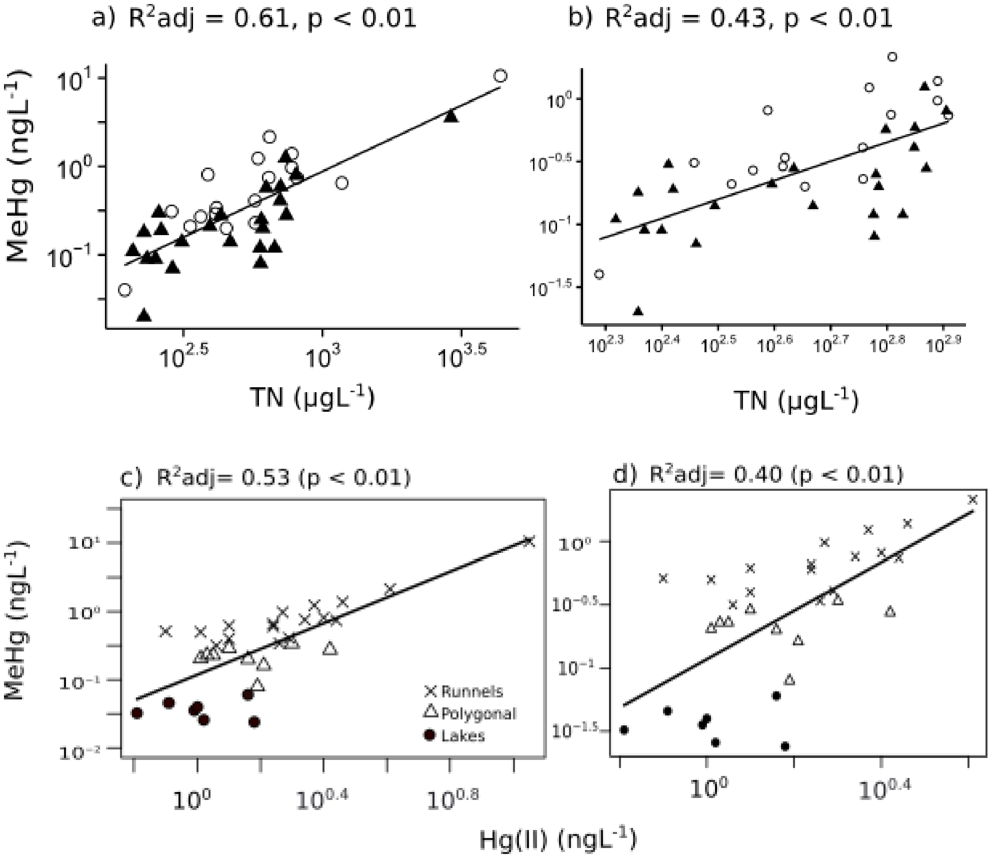

FIGURE S3. Correlations between MeHg concentrations (ngL^{-1}) showing significant positive correlations for a) all sample sites for TN (μgL^{-1}), n = 58 and b) a subset of sample sites for TN (μgL^{-1}) without 3 outliers, n = 55. Correlations between MeHg concentrations (ngL^{-1}) showing significant positive correlations for c) all Bylot sample sites for Hg(II (ngL^{-1}), n = 34 and d) all sites on Bylot Island without the outlier, n = 33. All axes are shown on logarithmic scales and regressions were performed on log-transformed data.