

1 **Text S1. Supplementary methods, hydrocarbon, diatom and inferred primary production**
2 **results and discussion.**

3

4 **Supplementary Methods**

5 **Analyses of water chemistry:**

6 Water samples were collected from the central portion of each lake approximately 1 m
7 below the surface, and were kept cool until transferred to the Taiga Environmental Laboratory
8 (Aboriginal Affairs and Northern Development Canada) in Yellowknife, NT. The Taiga
9 laboratory is an ISO/IEC 17025 accredited laboratory and a member of the Canadian Association
10 for Laboratory Accreditation (CALA). Accreditation results can be viewed by entering member
11 number 2635 on the CALA website. The Taiga Environmental Laboratory also regularly
12 participates in QA/QC activities and exchanges with other accredited laboratories, including the
13 National Laboratory for Environmental Testing (NLET; Environment Canada; Burlington, ON).
14 Chemical determination followed a variety of standard protocols depending on specific analyte.
15 Major ions were measured using ion chromatography following modified method SM4110B.
16 Metals were analyzed by inductively coupled plasma – mass spectrometry following EPA
17 method 200.8. Turbidity was analyzed by nephelometry following modified method SM2130B.
18 True and apparent colour were analyzed via spectrophotometer following method SM2120C.
19 Only chemical variables where the majority of samples occurred above the detection limit were
20 included in further analyses.

21

22 **Sediment collection, radiometric dating and sediment age determination:**

23 Sediment cores from the three study lakes were collected from a raft in the central,
24 deepest portion of the lake using a Glew-type gravity corer (internal diameter: 7.62 cm; [1]) and
25 sectioned at 0.5 cm resolution by vertical extrusion [2]. Sediment core intervals were dated using
26 ^{210}Pb and ^{137}Cs radiometric techniques, via gamma spectroscopy [3, 4]. Selected sediment
27 intervals were lyophilized, homogenized, entombed in epoxy resin and allowed to equilibrate for
28 a minimum of two weeks. Samples were then analyzed for ^{210}Pb , ^{214}Bi and ^{137}Cs activity using
29 an Ortec[®] well-type Ge crystal detector for 80,000 seconds each. Sediment age determination
30 was conducted using the constant rate of supply (CRS) model [5], with the supported ^{210}Pb
31 activity determined via the method established by [6]. The radioisotopic activities, as well as
32 respective CRS-derived sediment ages, are presented in Fig. S1. For each core the relationship
33 between the cumulative dry mass of the sediment interval and the unsupported ^{210}Pb activity (log
34 transformed) was examined and found to be strong ($r > 0.8$).

35

36 **Supplementary PAH methods:**

37 Wet sediments were homogenized and mixed in an approximate 1:1 ratio with Agilent
38 brand Hydromatrix[®]. The samples were spiked with ^{13}C labelled PAHs (Cambridge Isotope
39 Laboratories). PAHs were extracted from the wet sediments using accelerated solvent extraction
40 (ASE, Dionex) at 100°C with hexane:dichloromethane (DCM), followed by 35% acetone:65%
41 hexane. The non-polar extract (PAHs, hexane and DCM) was separated from the polar extract
42 (water and acetone) by combining the entire sample with DCM, 3% NaCl and saturated Na_2SO_4
43 in a series of liquid-liquid extractions. Sulphur compounds and pigments were removed from the
44 final extract on an Agilent 1100 Preparative Liquid Chromatograph using Waters Envirogel
45 Columns and USEPA method 3640A. The samples were evaporated to 1ml and fractioned,

46 according to USEPA method 3630C, on Davisil 635 silica (60-100 mesh, pore size 60). The
47 PAH fraction was analyzed by gas chromatography (Agilent 6890) and mass spectrometry
48 (Agilent 5973). 1µl injections were made in pulsed splitless mode at 280°C on a DB-XLB 30m x
49 0.18µm x 180µm column. An initial oven temperature of 60°C was held for 2 minutes then
50 increased at a rate of 6°C per minute to 300°C and held for 10 minutes. A constant flow rate of
51 39cm per second of helium was used for a total run time of 52 minutes. The mass spectrometer
52 was set to have a transfer line temperature of 280°C, with a source temperature of 230°C and
53 quadrupole temperature of 150°C.

54 The following fragments were monitored for PAHs in Single Ion Monitoring (SIM)
55 mode: m/z 128 for naphthalene, m/z 152 for acenaphthylene, m/z 153 for acenaphthene, m/z 166
56 for fluorine, m/z 178 for phenanthrene and anthracene, m/z 202 for fluoranthene and pyrene, m/z
57 228 for benz(a)anthracene and chrysene, m/z 240 for chrysene, m/z 252 for
58 benzo(b)fluoranthene, benzo(k)fluoranthene and benzo(a)pyrene, m/z 278 for indeno(123-
59 cd)pyrene and deibenz(a,h)anthracene, and m/z 276 for benzo(g,h,i)perylene. Indeno(123-
60 cd)pyrene and dibenz(a,h)anthracene were combined because of poor peak resolution.
61 Quantification was performed using ¹³C labelled PAHs (Cambridge Isotope Laboratories) for all
62 of the compounds listed above.

63 The following fragments were monitored for alkylated PAHs in SIM mode: m/z 142 for 1- and
64 2-methylnaphthalene, m/z 156 for C2-naphthalene, m/z 170 for C3-naphthalene, m/z 184 for C4-
65 naphthalene, m/z 180 for C1-fluorine, m/z 194 for C2-fluorine, m/z 208 for C3-fluorine, m/z 192
66 for C1-phenanthrene and C1-anthracene, m/z 206 for C2-phenanthrene and C2-anthracene, m/z
67 220 for C3-phenanthrene and C3-anthracene, m/a 234 for C4-phenanthrene and C4-anthracene,
68 m/z 216 for C1-fluoranthene and C1-pyrene, m/z 230 for C2-fluoranthene and C2-pyrene, m/z

69 for 244 for C3-fluoroanthene and C3-pyrene, m/z 258 for C4-fluoroanthene and C4-pyrene, m/z
70 242 for C1-benz(a)anthracene and C1-chrysene, m/z 256 for C2-benz(a)anthracene and C2-
71 chrysene, m/z C3-benz(a)anthracene and C3-chrysene, m/z 266 for C1-benzfluoroanthene and C1-
72 benzopyrene, m/z 280 for C2-benzofluoroanthene and C2-benzopyrene. Quantification was
73 performed using ¹³C labelled non-alkylated PAHs (Cambridge Isotope Laboratories) for all of
74 the compounds listed above.

75

76 **Supplementary Results and Discussion**

77 **Present-day lakewater chemical comparisons:**

78 Comparisons of present-day physical and chemical limnological variables reveal major
79 differences between the three *a priori* defined groups (all comparisons made using two-way
80 ANOVA, df=2, 98, followed by Tukey HSD post-hoc test run on normalized data) (Fig. S2).
81 Drilling sump lakes exhibited significantly higher Cl⁻ concentrations than either permafrost thaw
82 or control lakes (F=7.91, p<0.001). Permafrost thaw lakes exhibited significantly increased K⁺
83 (F=23.6, p<0.001), Ca²⁺ (F=50.2, p<0.001), and Na⁺ (F=30.7, p<0.001) concentrations, specific
84 conductivity (F=47.3, p<0.001) and total dissolved solids (F=18.8, p<0.001) than the drilling
85 sump lakes which were all significantly greater than in the control lakes (Fig. S2). The drilling
86 sump lakes were significantly shallower than the permafrost thaw or control lakes (F=19.7,
87 p<0.001). Lakes with permafrost thaw slumps had significantly higher apparent colour (F=37.0,
88 p<0.001), SO₄²⁻ concentration (F=36.0, p<0.001) and lower total suspended solids (F=4.52,
89 p=0.01) than the other two groups. The control lakes had lower pH than either the drilling sump
90 or permafrost thaw lakes (F=22.1, p<0.001). No significant difference was observed in water
91 turbidity between the three groups (F=1.46, p=0.24). Analysis of similarity (ANOSIM) and

92 similarity percentages (SIMPER) were used in order to assess differences in the *a priori* defined
93 groups, as well as determine which variables contributed to any statistical differences. SIMPER
94 analysis, run on a Euclidean distance matrix, determined that Cl⁻, Si, and Mn concentrations, as
95 well as maximum depth, contributed significantly to the difference between the drilling sump
96 and control lakes as well as the drilling sump and permafrost thaw lakes (Table S2). Ca²⁺, Si, and
97 Mn concentrations, as well as specific conductivity, were found to contribute to the dissimilarity
98 between the permafrost thaw and control lakes, though notably chloride concentration was not
99 observed to be important (Table S2).

100

101 **Polycyclic aromatic hydrocarbons (PAHs):**

102 Sedimentary concentrations of the 12 priority PAHs, including the most common PAHs
103 from petrogenic sources, did not change at the time of, or subsequent to, drilling sump
104 construction in any of the three study lakes (Fig. S3). Included in this summation are the
105 following compounds: phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene,
106 chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene,
107 dibenz[a,h]anthracene, and benzo[g,h,i]perylene. This suggests that, unlike the impact of large-
108 scale extraction operations such as the Alberta oilsands [7, 8], impacts from exploratory drilling
109 activities in this region of the Arctic do not appear related to contamination by hydrocarbons
110 themselves. This is not surprising, given that these exploratory operations extract comparatively
111 few hydrocarbons during the process of test well development, and given that relatively high,
112 stable background PAH concentrations were observed in the sediments of all three lakes
113 throughout the past few hundred years (Fig. S3).

114

115 **Sedimentary diatoms:**

116 Sedimentary diatom assemblages from Lake I20, impacted by a failing drilling sump, do
117 not show any response coincident with, or subsequent to, sump construction (Fig. S4). The
118 modern assemblage is dominated by the planktonic taxon *Cyclostephanos tholiformis*, as well as
119 a variety of small benthic taxa of the group *Fragilaria sensu lato* (primarily *Staurosirella*
120 *pinnata*, *Staurosira venter* and *Pseudostaurosira brevistriata*). Prior to approximately 1950,
121 when a gradual increase in *C. tholiformis* began, the assemblage was dominated by small benthic
122 fragilarioids at greater than 70% relative abundance (Fig. S4). Similarly, in Lake C23, the
123 assemblage is dominated by small benthic fragilarioid taxa throughout the recent past. At ~1910
124 an increase in planktonic taxa, including *Discostella pseudostelligera* (synonymous with
125 *Cyclotella pseudostelligera*), as well as a number of pennate planktonic taxa occurred, coincident
126 with the onset of warming in this region as recorded by several independent proxies [9]. The
127 primary diatom changes in both I20 and control Lake C23 occur earlier than the known
128 construction of drilling sumps in the vicinity of the lakes, and are characterized by increased
129 planktonic diatom abundance, indicative of climate warming [10], similar to the timing and
130 nature observed in many other lakes in this region [9]. The changes in diatom assemblages in I20
131 and C23 occur simultaneously with increases in inferred primary production (Fig. S3),
132 suggesting an influence of warming on overall primary production in these lakes. No major
133 changes were observed in the diatom record from C1A (Fig. S4), which is composed of an
134 assemblage dominated by small *Fragilaria sensu lato* taxa as well as a variety of other periphytic
135 and benthic species. The inferences of primary production in C1A were below detection limits
136 (Fig. S3). In no lake was an increase in saline-tolerant taxa observed, such as those recorded
137 following saline intrusion in the nearby outer Mackenzie Delta [11]. The lack of diatom response

138 to changes in lakewater chemistry as a result of drilling sump failure is not surprising as diatoms
139 were found to respond to changes in aquatic habitat, and not chemistry, following the
140 development of large permafrost thaw slumps; systems which our results show have similar
141 chemical compositions.

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143 **References**

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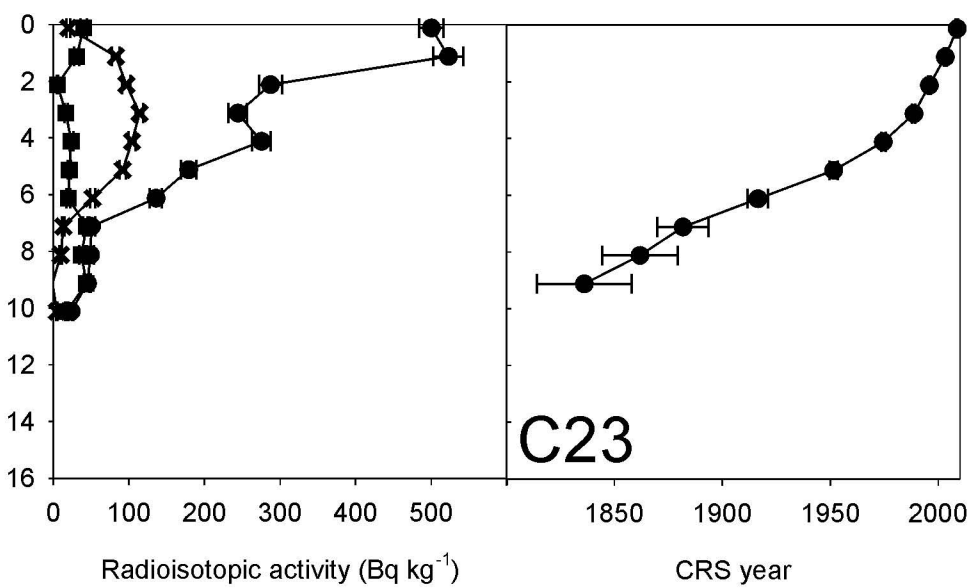
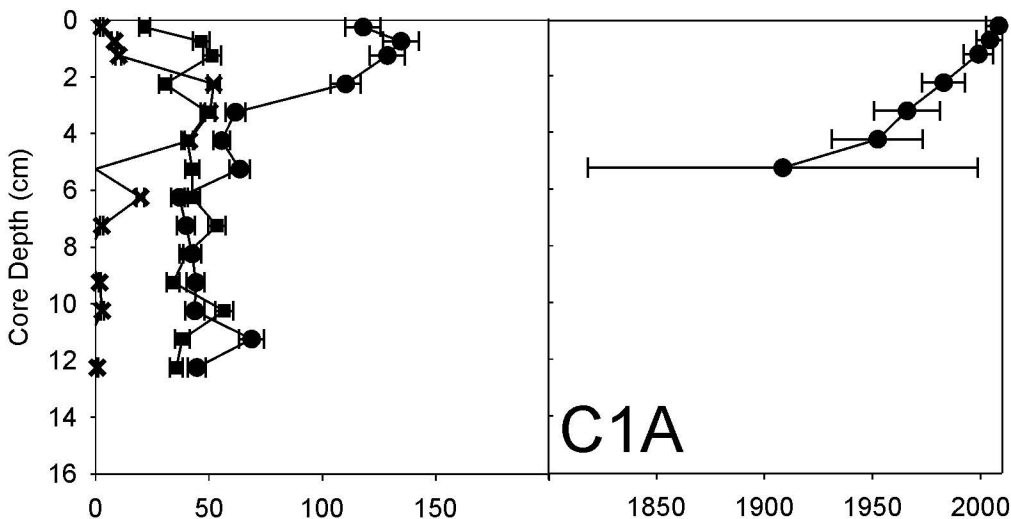
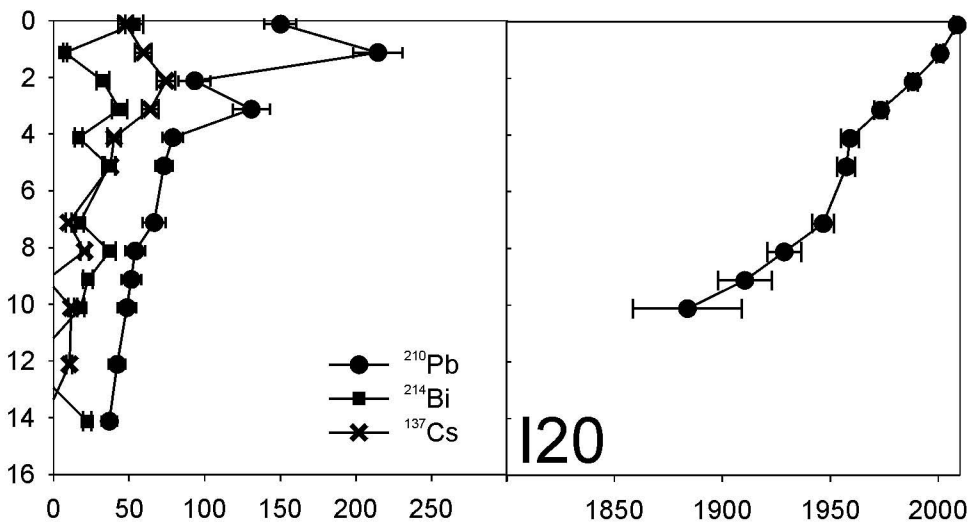
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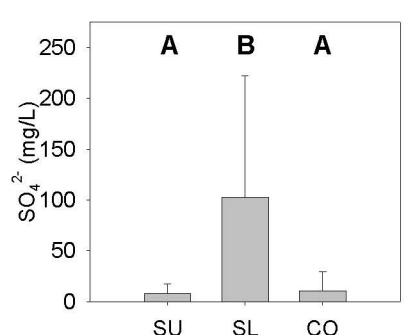
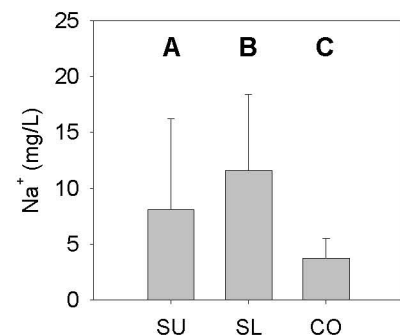
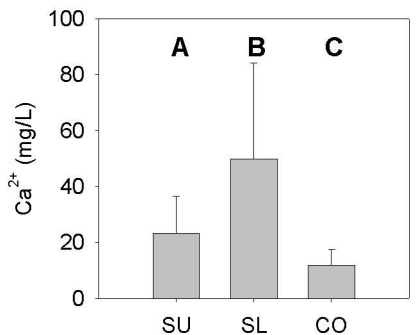
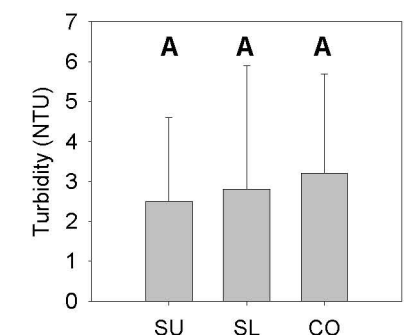
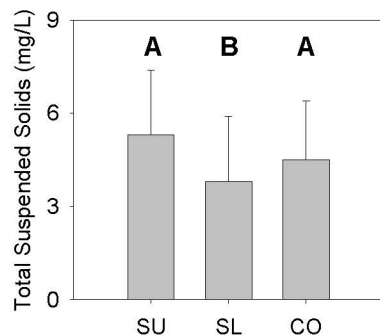
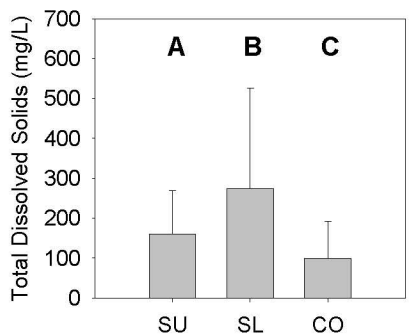
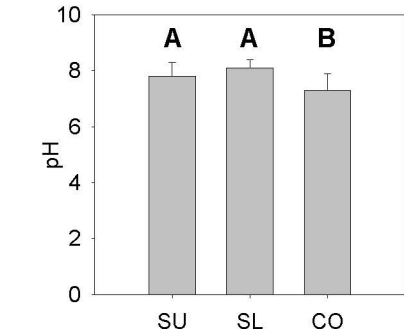
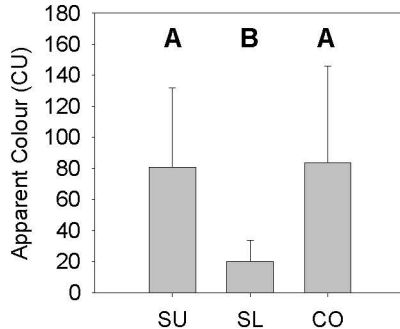
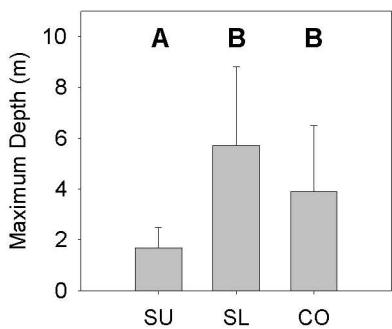
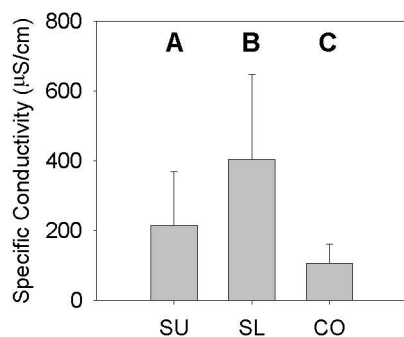
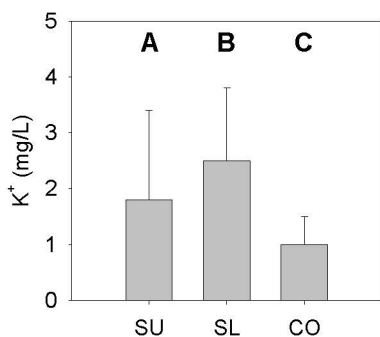
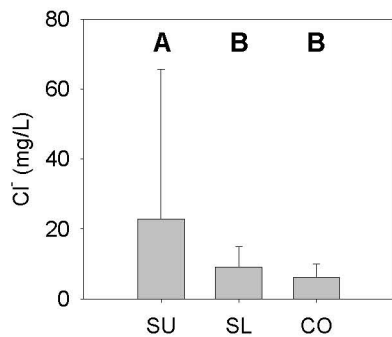
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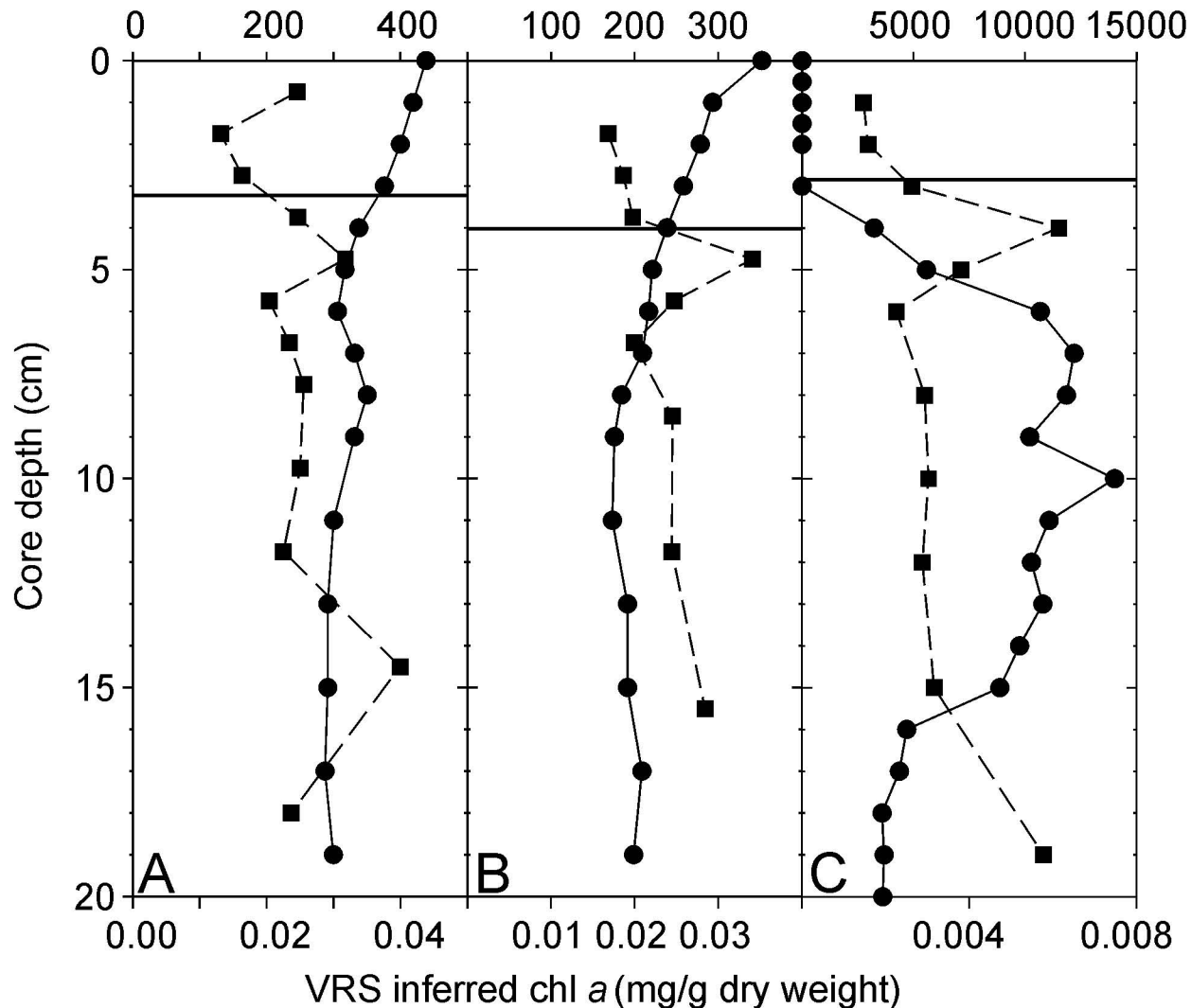
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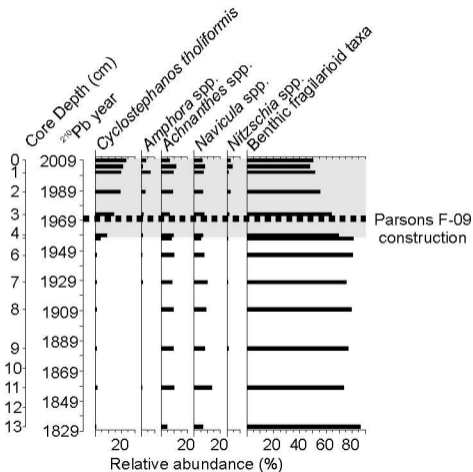




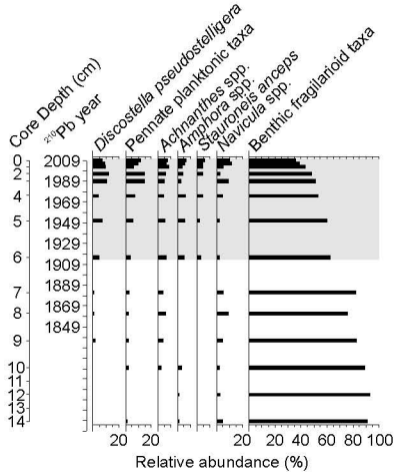
Σ 12 priority PAH concentration (ng PAH/g OC)



A Sump-impacted Lake I20



B Control Lake C23



C Control Lake C1A

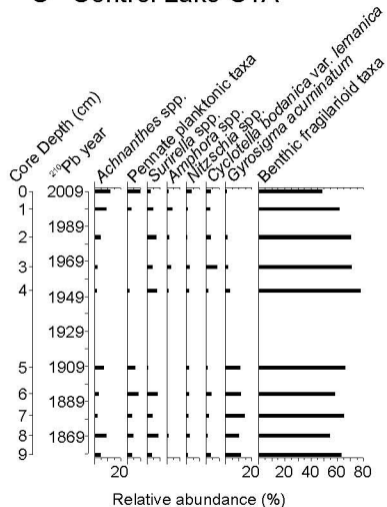


Table S1: Summary statistics of water chemistry data. Sites were sampled in the summers of 2005 and 2007, with samples taken at approximately one metre water depth.

	Sur Area* (ha)	Catch Area* (ha)	Z _{max} * (m)	Alk* (mg/l)	Col* (CU)	Cond* (µS/cm)	pH	TDS* (mg/l)	Turb* (NTU)	Ca (mg/l)	Mg (mg/l)	Na (mg/l)	K (mg/l)	Cl (mg/l)	SO ₄ (mg/l)	Fe (µg/l)	Mn (µg/l)
Drilling-sump lakes (n = 20)																	
Mean	15.8	83.5	1.7	58	81	214	7.8	160	2.5	23.2	10	8.1	1.8	22.8	8	477	46
Median	4.8	27.3	1.5	49	68	144	7.8	132	1.8	18.9	7	4.9	1.3	8.0	6	394	31
STD	22.4	162.9	0.8	35	51	155	0.5	110	2.1	13.2	6	8.1	1.6	42.9	9	397	50
Max	85.7	749.4	3.4	145	243	738	9.0	554	9.6	54.8	27	37.3	6.7	198.0	40	1570	214
Min	0.5	15.5	1.0	12	19	58	6.9	61	0.5	6.9	33	1.8	0.5	1.3	1	50	9
Thaw slump-affected lakes (n = 34)																	
Mean	13.1	50.6	5.7	90	20	404	8.1	275	2.8	49.9	17	11.6	2.5	9.1	102	186	36
Median	6.2	26.0	5.0	86	16	354	8.1	192	1.5	42.3	13	10.9	2.1	6.7	52	131	21
STD	22.6	62.6	3.1	41	14	243	0.3	251	3.1	34.2	12	6.8	1.3	5.9	120	190	47
Max	116.5	254.4	14.2	174	50	1000	8.4	1330	15.8	146.0	53	30.8	6.1	20.6	406	855	253
Min	1.2	4.7	1.7	20	5	84	7.2	52	0.5	6.9	3	2.3	0.4	1.9	2	50	4
Control lakes (n = 47)																	
Mean	5.0	64.8	3.9	29	84	107	7.3	99	3.2	11.8	4	3.7	1.0	6.1	10	895	42
Median	2.9	23.0	3.3	26	74	102	7.5	84	2.5	11.3	4	3.3	1.0	5.9	4	505	25
STD	6.3	151.9	2.8	18	62	54	0.6	93	2.5	5.8	2	1.8	0.5	3.9	19.	1058	71
Max	32.5	749.4	11.2	94	266	287	8.2	694	12.6	27.9	9	7.7	2.5	13.6	118	5960	482
Min	0.5	3.9	1.0	4	5	29	4.3	40	0.3	4.2	1	0.8	0.2	0.9	1	0	2

Table S2: Results of similarity percentages (SIMPER) analysis.

Variable	Dissimilarity Contribution (%)	Cumulative Contribution (%)
Drilling sump and Control		
Chloride	18.54	18.54
Silica	15.60	34.13
Maximum depth	11.14	45.28
Manganese	10.76	56.04
Sodium	8.56	64.59
Drilling sump and Permafrost thaw		
Silica	18.11	18.11
Chloride	13.25	31.36
Manganese	12.43	43.79
Maximum depth	12.25	56.04
Calcium	8.99	65.03
Permafrost thaw and Control		
Calcium	15.42	15.42
Silica	14.31	29.73
Specific conductivity	13.92	43.65
Sodium	11.36	55.01
Manganese	10.02	65.03