Supporting Information

Climatic Influence on Temporal Trends of Polychlorinated Biphenyls and Organochlorine Pesticides in Landlocked Char from Lakes in the Canadian High Arctic

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1. Analytical Methods

1.1. Extraction and isolation steps for Arctic char *(Salvelinus alpinus)* samples: Extraction and isolation steps generally followed US EPA Method 1699. Prior to the extraction, char muscle+skin samples were homogenized using a hand blender. Briefly, up to 15 g of the homogenized sample were spiked with δ-HCH, and PCBs 30 and 204 prior the extraction for GC-ECD analysis. For GC-MS analysis, samples were spiked with ^{13}C- and deuterated OCPs ($^{13}C_6$-Pentachlorobenzene, $^{13}C_6$-HCB, $^{13}C_6$-alpha-HCH, $^{13}C_6$ -gamma-HCH, $^{13}C_{10}$-heptachlor, $^{13}C_{10}$-oxychlordane, $^{13}C_{10}$-trans-nonachlor, $^{13}C_{10}$-dieldrin, $^{13}C_{10}$-endrin, d_4-Endosulfan, d_8-4,4'-DDE, $^{13}C_{12}$-4,4'-DDT, d_6-methoxychlor, $^{13}C_8$-mirex) and $^{13}C_{12}$-labelled PCBs (CB-1, CB-3, CB-4, CB-9, CB-15, CB-19, CB-37, CB-52, CB-54, CB-77, CB-81, CB-104, CB-105,CB-114, CB-118, CB-123, CB-126, CB-155, CB-156, CB-157, CB-167, CB-169, CB-188, CB-189, CB-202, CB-205, CB-206, CB-208, CB-209). Samples were extracted in a Soxhlet during 24h with dichloromethane (DCM). Extracts were reduced with a roto-vap to 2 ml and then subjected to lipid removal by gel permeation chromatography (GPC) using Biobeads SX3. The percent extractable lipid content was determined (±0.1%) gravimetrically by evaporating the first GPC fraction (NLET) or by use of approximately ¼ of the extract (ALS Environmental) prior to GPC. $^{13}C_{12}$-CB-133 was added as a recovery standard for GPC performance. ALS Environmental split the GPC elution into two separate fractions. The OCP fraction was chromatographed on a 2% deactivated silica gel column then reduced to 0.05 mL for analysis using GC high-resolution mass spectrometry (GC-HRMS). The PCB fraction was cleaned up on an acid-silica gel column (45% w/w H_2SO_4 on Silica Gel topped with neutral Silica Gel) then reduced to 0.04 mL for GC-low resolution MS (GC-LRMS) analysis. A laboratory blank consisting of all reagents and a two NIST reference material (fish muscle SRM 1946 or SRM 1947 lake trout) were analyzed with each batch of 9 samples.

PCBs (87 congeners from mono to decachloro PCBs) and OCPs (HCHs, including α-HCH, β-HCH and γ-HCH; DDTs including o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD o,p'-DDT, p,p'-DDT and HCB) (Table S1) were analyzed by gas chromatography coupled with electron capture detector (GC-ECD) for samples collected between 1990 to 2010 and by GC-LRMS in electron-ionization mode all others from 2011. GC-high resolution mass spectrometry (HRMS) was used to analyse final extracts of samples from 2011-2015 for HCB as well as HCHs and DDTs related compounds at ≥8,000 mass resolution. Analyses of toxaphene for all sample years were performed with gas chromatography-low resolution mass spectrometry (GC-LRMS), using negative chemical ionization (NCI) on an Agilent 7890A GC-5975C MS system run in selective ion monitoring (SIM) mode. Toxaphene was quantified using a technical toxaphene standard (Glassmeyer et al, 1999). Individual toxaphene congeners (P26, P50, P62) were determined in a subset of samples from all four lakes (2012-2015) using authentic external standards of each compound obtained from Ehrenstorfer Labs (www.LGCstandards.com). Further details on individual compounds and GC-ECD and GC-MS conditions are provided below. The analysis of the samples was performed every year after sampling, with the available technology at each moment, which is why different techniques were used in this study.

GC-ECD Analysis: GC-ECD analysis was conducted using an Agilent 6890 gas chromatograph with a 63Ni-electron capture detector (ECD) and a 30 m x 0.25 mm (i.d.) DB-5 column (internal film thickness 0.25 mm; J&W Scientific, Folsom, CA, USA) with H2 carrier gas (constant flow rate 0.91 ml min^{-1}). Ultra-pure N2 was used as the makeup gas for the ECD (detector temperature: 325 C). The GC-ECD quantification of OCs in each sample was performed using a 4 point external standard calibration curve. Calibration standards were quantified after every 10 samples. A list of individual PCB and OCP analytes is given in Table S1.

GC-LRMS and GC-HRMS analysis: Final extracts were analyzed by GC-HRMS for 31 OCPs related compounds (Table S1) using GC-HRMS at ≥ 8,000 resolution and for 87 individual + co-eluting PCB congeners GC-LRMS, using isotope dilution. Separation was achieved on a HP-5ms column (30m length x 0.25mm id x 0.25 um film thickness).

Quality assurance: Method detection limits (MDLs) for PCBs and OCPs were calculated for all analytes based on results from 6 laboratory blanks that were analysed in the same laboratory at approximately the same time, where MDL = 3x standard deviation of the blanks. For analytes with non-detectable blank values, the instrument detection limit (IDL), based on a signal to noise of approximately 10:1 was used. Results that were <IDL were replaced with the IDL for statistical calculations.

Analysis of the NIST SRMs 1946 and 1947 showed good agreement with all analytes quantified to within ±25% of certified values of OCPs (17 compounds) and PCBs (29 congeners).

Table S1. List of individual organohalogen analytes along with their MDLs (ng/g wet wt) using either GC with high- or low resolution MS, GC-NCIMS, or GC-ECD

Class	Common name	Chemical or other name	Instrumental analysis	MDL	MDL
				GC-MS	GC-ECD
OCP	Hexachloro-benzene		GC-HRMS, GC-ECD	<0.026	<0.002
OCP	α-HCH	α-hexachlorocyclohexane	GC-HRMS, GC-ECD	<0.059	0.015
OCP	β-HCH	β-hexachlorocyclohexane	GC-HRMS, GC-ECD	<0.1	0.013
OCP	γ-HCH	Lindane	GC-HRMS, GC-ECD	<0.075	<0.002
OCP	24'-DDE		GC-HRMS, GC-ECD	<0.01	0.001
OCP	44'-DDE		GC-HRMS, GC-ECD	<0.01	0.021
OCP	24'-DDD		GC-HRMS, GC-ECD	<0.01	0.014
OCP	44'-DDD		GC-HRMS, GC-ECD	<0.01	0.02
OCP	24'-DDT		GC-HRMS, GC-ECD	<0.01	0.02
OCP	44'-DDT		GC-HRMS, GC-ECD	<0.01	0.02
OCP	Toxaphene	Total toxaphene	GC-NCIMS	<1.0	
OCP	P26	B8-1413	GC- NCIMS	<0.01	
OCP	P50	B9-1679	GC- NCIMS	<0.01	
OCP	P62	B9-1025	GC- NCIMS	<0.01	
PCBs	PCB-1	monochlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB-3	monochlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB4/10	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.166
PCBs	PCB7/9	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.158
PCBs	PCB6	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.042
PCBs	PCB8/5	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.078
PCBs	PCB12/13	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	1.11
PCBs	PCB15	dichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.036
PCBs	PCB19	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	<0.002
PCBs	PCB18	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.211
PCBs	PCB17	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB27/24	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.036
PCBs	PCB16/32	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.119
PCBs	PCB26	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.112
PCBs	PCB25	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.019
PCBs	PCB31	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.445
PCBs	PCB28	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.05
PCBs	PCB20/33/21	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.155
PCBs	PCB22	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.097
PCBs	PCB37	trichlorobiphenyl	GC-LRMS, GC-ECD	<0.002	
PCBs	PCB53	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.117
PCBs	PCB45	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.136
PCBs	PCB46	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.058
PCBs	PCB73/52	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.913
PCBs	PCB43/49	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.446
PCBs	PCB48/47/75	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	5.152
PCBs	PCB44	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.554
PCBs	PCB59/42	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.21
PCBs	PCB71/41/68/64	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.417
PCBs	PCB100	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.052

PCBs	PCB63	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.024
PCBs	PCB74/61	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.241
PCBs	PCB70/76	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.663
PCBs	PCB80/66	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.375
PCBs	PCB56/60	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.229
PCBs	PCB81	tetrachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.413
PCBs	PCB95/93	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.641
PCBs	PCB91	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.146
PCBs	PCB92	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.156
PCBs	PCB84/90/101/89	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.26
PCBs	PCB89-101	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.692
PCBs	PCB99	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.319
PCBs	PCB119	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.022
PCBs	PCB83/108	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.089
PCBs	PCB97	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.172
PCBs	PCB86/111/125/117/87/116/115	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB120/85	pentachlorobiphenyl	GC-LRMS, GC-ECD	0.055	0.092
PCBs	PCB110	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.461
PCBs	PCB136	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.114
PCBs	PCB82	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.049
PCBs	PCB107/109	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.059
PCBs	PCB123	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.406
PCBs	PCB118/106	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.298
PCBs	PCB114	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.024
PCBs	PCB105/127	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.141
PCBs	PCB126	pentachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB151	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.173
PCBs	PCB135/144	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.651
PCBs	PCB139/149	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB131/165/142	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.021
PCBs	PCB146	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.056
PCBs	PCB153	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.293
PCBs	PCB132/168	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.135
PCBs	PCB141	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.068
PCBs	PCB137	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	3.971
PCBs	PCB163/164/138	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.283
PCBs	PCB158/160	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.044
PCBs	PCB129	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.018
PCBs	PCB159	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB128	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.028
PCBs	PCB167	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.01
PCBs	PCB156	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.013
PCBs	PCB157	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	<0.001
PCBs	PCB169	hexachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB182/187	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.109
PCBs	PCB183	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.046
PCBs	PCB174/181	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.072
PCBs	PCB177	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.143
PCBs	PCB171	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.042
PCBs	PCB172/192	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.018

PCBs	PCB197	octachloorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.007
PCBs	PCB180	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.087
PCBs	PCB193	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.132
PCBs	PCB191	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	<0.001
PCBs	PCB170/190	heptachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.03
PCBs	PCB-202	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	na
PCBs	PCB199	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.008
PCBs	PCB196/203	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.022
PCBs	PCB195	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.015
PCBs	PCB194	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.005
PCBs	PCB205	octachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	<0.001
PCBs	PCB208	nonachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.002
PCBs	PCB207	nonachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	<0.001
PCBs	PCB206	nonachlorobiphenyl	GC-LRMS, GC-ECD	<0.002	0.013
PCBs	PCB209	decachlorobiphenyl	GC-LRMS, GC-ECD	0.08	0.004

1.2 Extraction and isolation steps for soil samples: Approximately 35 g of wet soil samples were homogeneized and mixed with equal parts of ASE-cleaned Hydromatrix using a mortar and a pestle. Hydromatrix (diatomaceous earth; Dionex) was packed in 100 mL ASE cells and cleaned prior to sample extraction. Hydromatrix cleaning consisted of 2 methods run consecutively on the ASE using acetone:hexane and DCM (Table S2). Homogenized soil samples were packed in stainless steel ASE cells with cellulose filters and spiked with $^{13}C_{12}$-labelled PCBs (CB-28, CB-52, CB-101, CB-153, CB-138, CB-180, CB-209). All samples were then extracted in an accelerated solvent extractor (ASE 300; Dionex) using the same methods previously used to clean the hydromatrix as described in Table S2. All methods were set to reach a maximum extraction temperature and pressure of 100°C and 1500 psi respectively. After extraction, samples were dried on sodium sulphate, evaporated and clean up on 5 g of activate silica (60-200 mesh silica gel). The fraction containing the PCBs and HCB was eluted with 60 ml of hexane, concentrated on a TurboVap to 0.5 ml and reduced to 100µl under N_2. The fraction containing the PCBs and HCB were solvent exchange to isooctane and a mix containing PCB 30 and $^{13}C_{12}$-labelled PCBs (CB-77, CB-81, CB-126, CB-169) were added and used as internal standards.

Table S2. ASE Methods details for cleaning Hydromatrix and Soil Extraction for POPs

	Solvent	Pre-Heat (min)	Heat (min)	Static (min)	Purge (s)	Cycles
Cleaning Method 1	Acetone:hexane (1:1)	1	5	5	60	1
Cleaning Method 2	DCM	1	5	5	60	1
Soil Samples Method 1	Acetone:hexane (1:1)	1	5	5	60	1
Soil Samples Method 2	DCM	1	5	5	60	1

All solvent lots were quality checked prior to use. Sodium sulphate, silica and glass wool were fired at 450 ºC for at least 12 h prior to use. All glassware was thoroughly rinsed with hot tap water, deionized water, and placed in a 6% Extran™ (Merck) bath for at least 8 h as described in Morris et al, 2014. Glassware was then rinsed and soaked in clean hot water for 1 h, rinsed, air dried, acetone rinsed and fired at 150C for 4-8 h.

Soil water content (SWC) (%): was determined using a known amount of wet soil and freeze dried until soil weight becomes constant.

$$\%SWC = \frac{fw - fd}{fw} * 100$$

where *fw* and *fd* are the amount of wet and dry soil respectively. SWC was used to determined the soil dry weight extracted.

GC/MS-MS analysis for soil samples: PCBs (70 congeners from mono to decachloro PCBs) and HCB were analyzed by gas chromatography (Agilent 7890B) coupled to tandem mass spectrometry (Agilent 7000C) (GC-MS/MS) operating in Multiple Reaction Monitoring (MRM) with 60 m × 0.25 mm i.d. (RESTEK Rxi-5Sil MS) column coated with 5% phenyl-95% methylpolysiloxane (film thickness 0.25 μm). After sample injection (splitless) onto the column (injection port temperature = 250°C), the GC temperature profile was as follows: (1) initial oven temperature was

90°C; held for 2 min (2) ramped at 15°C/min to 190°C, oven held 0.7 min (3) ramped at 3°C/min to 203°C, oven held 5min (4) with the final ramp of 3°C/min to a maximum temperature of 300°C and the oven was held for 5 min. The MS transfer line, source and quadrupoles temperatures were 280 ºC, 300°C, 180°C respectively.

Quality assurance: Laboratory blanks were included in every set of samples extractions (1 blank every 5 samples). Soil blanks consisted in a 100 mL ASE cells + ~30 g ASE-cleaned Hydromatrix. Few PCB congeners were detected in blanks at low concentrations and therefore, samples were blank corrected. Method detection limits were calculated for all analytes based on results from 5 laboratory blanks that were analysed, where MDL = 3x standard deviation of the blanks PCBs. MDL ranged from 0.0001-0.012 ng g dw^{-1}. Recoveries were routinely monitored using $^{13}C_{12}$-labelled PCBs and they ranged from 80 to 110 %.

1.3. Water chemistry analyses: Sample were taken at 0.5-1 m depth by boat generally at mid-lake, or open water site if ice was present, in Resolute, Char and Amituk lakes. At Lake Hazen, a much larger system, samples were generally collected at a position mid-way between John's Island and Hazen camp (approx. 71° 49' N, 71° 19'W). Lescord et al (2015) have described the sample processing. In brief, water samples were stored at 4ºC and filtered through GF/F (47 mm, pore size 1.2 µm for CHLa/POC/PON; 25 mm, pore size 0.45 µm for DOC/DIC) within 24 hrs of collection. Filters were wrapped in aluminum foil and stored at -20C until analysis. Analysis was conducted within 28 days of collection. Chlorophyll a was analysed using NLET method 01-1100 (Environment Canada 2010).

ANNEX II. Fish Biology

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Table S3. Biometric parameters of age, length and weight for Arctic char from each lake.

	Age (yrs)			Length (cm)			Weight (g)		
	N	Mean±SD	Min-Max	N	Mean±SD	Min-Max	N	Mean±SD	Min-Max
Amituk	95	19 ± 4	10 - 31	86	44.1 ± 8.1	29.7 – 68.0	96	688 ± 412	200 - 2391
Char	28	18 ± 8	8 - 33	45	41.3 ± 9.4	20.6 – 62.0	44	758 ± 662	216 - 2537
Hazen	143	20 ± 5	10 - 33	141	45.8 ± 10	29.5 - 81.5	129	982 ± 611	230- 3148
Resolute	161	20 ± 5	11 - 37	165	40.2 ± 4.7	28.6 – 55.0	165	539 ± 211	200 - 1355

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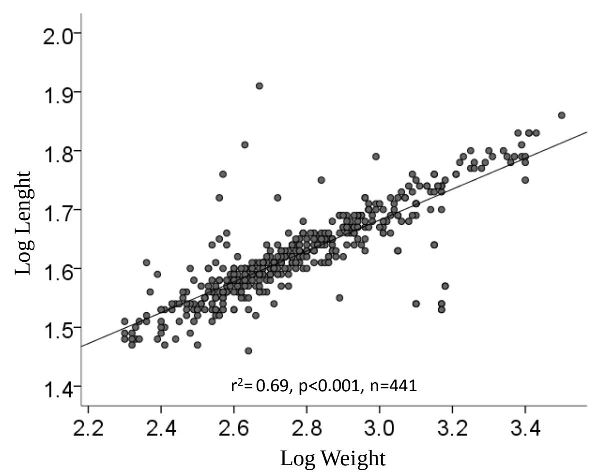

Figure S1. Relationship between Arctic char fork length and weight (in log) within the set of data

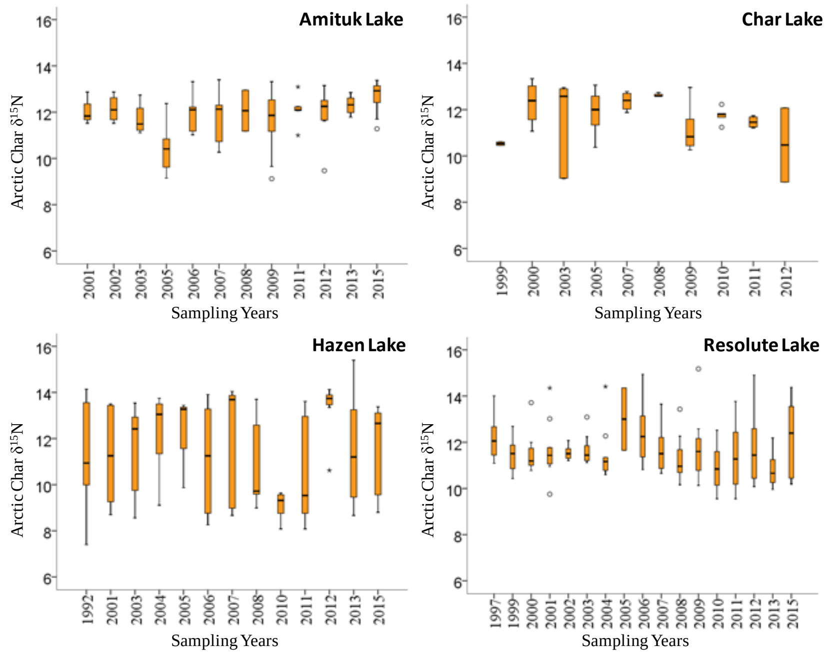

Figure S2. Box-plot comparing the $\delta^{15}N$ isotopic signature for Amituk, Char, Hazen and Resolute lakes through the temporal series of data

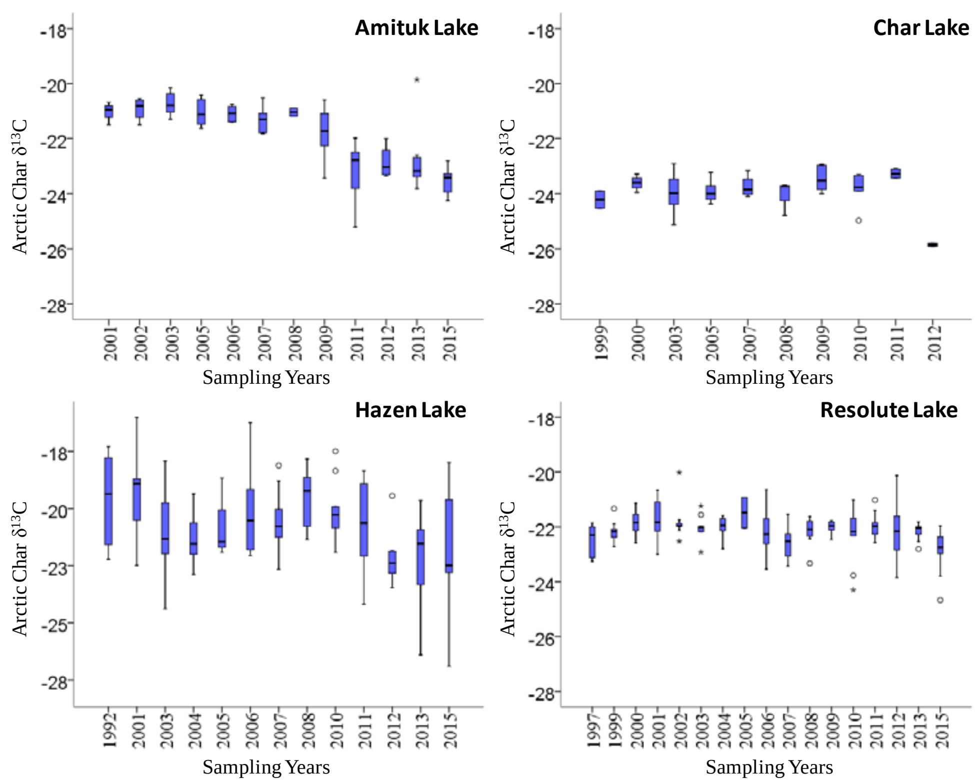

Figure S3. Box-plot comparing the $\delta^{13}C$ isotopic signature for Amituk, Char, Hazen and Resolute lakes through the temporal series of data

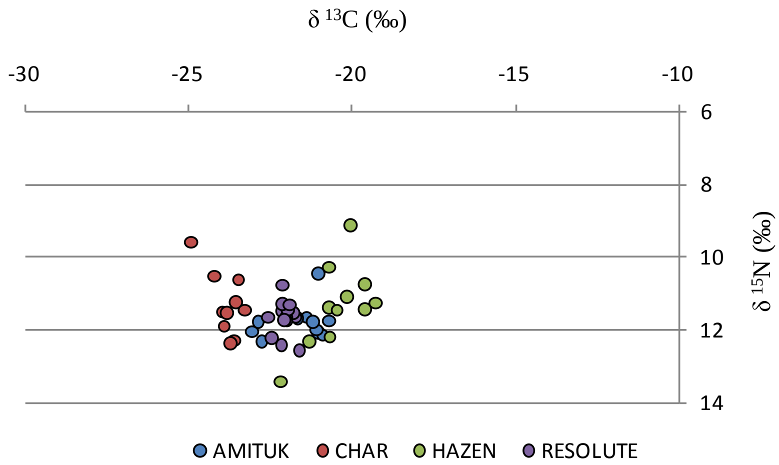

Figure S4. Geomeans $\delta^{13}C$ vs $\delta^{15}N$ isotopic signatures for Amituk, Char, Hazen and Resolute lakes

ANNEX III. Occurrence of legacy POPs in Arctic char

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Table S4. Geometric mean concentration (ng/g wet weight) of ΣPCBs, OCPs (ΣDDTs, ΣHCHs and HCB) and Toxaphene in Resolute, Hazen, Char and Amituk lakes along the temporal series

***na: not available data**

		ΣPCBs	ΣDDTs	ΣHCHs	HCB	Toxaphene
1989	**Resolute**	n.a	n.a	n.a	n.a	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	125.44	61.12	4.24	4.37	151.52
1990	**Resolute**	n.a	n.a	n.a	n.a	n.a
	Hazen	36.97	11.61	2.05	2.46	124.75
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	n.a	n.a	n.a	n.a	n.a
1992	**Resolute**	n.a	n.a	n.a	n.a	n.a
	Hazen	18.93	5.10	1.08	1.24	39.04
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	63.40	28.97	1.53	3.23	171.53

1993	**Resolute**	n.a	n.a	n.a	n.a	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	116.03	51.75	1.18	1.69	22.99
	Amituk	n.a	n.a	n.a	n.a	n.a
1997	**Resolute**	99.08	5.51	0.52	1.23	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	n.a	n.a	n.a	n.a	n.a
1999	**Resolute**	84.15	6.08	0.61	1.29	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	53.14	27.11	2.75	0.86	4.23
	Amituk	n.a	n.a	n.a	n.a	n.a
2000	**Resolute**	119.98	7.77	0.66	1.83	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	117.67	31.57	0.27	0.97	n.a
	Amituk	n.a	n.a	n.a	n.a	n.a
2001	**Resolute**	109.32	8.64	0.84	2.41	n.a
	Hazen	12.12	2.80	0.70	0.95	20.78
	Char	127.07	18.64	1.06	2.47	17.29
	Amituk	37.91	15.01	0.83	1.34	24.74
2002	**Resolute**	74.95	3.67	0.41	1.07	n.a
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	48.26	19.43	0.35	2.44	104.29
2003	**Resolute**	88.34	2.52	0.38	1.39	1.96
	Hazen	15.96	3.09	0.60	1.02	26.21
	Char	106.98	20.33	0.50	1.61	14.11
	Amituk	31.79	1.06	0.31	1.13	73.74
2004	**Resolute**	84.36	1.99	0.38	1.40	1.60
	Hazen	12.80	1.44	0.65	0.86	25.16
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	n.a	n.a	n.a	n.a	n.a
2005	**Resolute**	96.93	4.57	0.15	1.34	1.70
	Hazen	22.75	2.80	1.08	1.67	16.95
	Char	40.16	13.19	0.10	0.59	4.17
	Amituk	11.67	5.36	0.13	0.53	18.99
2006	**Resolute**	87.62	4.03	0.14	0.86	2.17
	Hazen	6.47	0.76	0.34	0.70	13.45
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	37.18	11.47	0.12	1.09	46.67
2007	**Resolute**	83.78	4.81	0.22	1.51	6.82
	Hazen	11.93	1.39	0.49	0.68	18.17
	Char	76.13	19.87	0.17	1.52	19.25
	Amituk	29.62	8.46	0.12	1.29	71.67
2008	**Resolute**	77.75	4.62	0.32	1.53	5.46
	Hazen	6.57	0.61	0.22	0.48	10.50
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	22.07	7.11	0.20	1.05	16.94
2009	**Resolute**	32.48	4.66	0.27	1.50	3.44
	Hazen	n.a	n.a	n.a	n.a	n.a
	Char	18.04	9.60	0.11	0.69	4.91

	Amituk	20.31	9.48	0.21	1.53	62.14
2010	**Resolute**	78.25	3.92	0.26	1.76	8.61
	Hazen	10.68	0.82	0.23	0.90	33.80
	Char	63.01	8.49	0.13	1.08	12.00
	Amituk	n.a	n.a	n.a	n.a	n.a
2011	**Resolute**	70.77	4.35	0.31	1.84	11.13
	Hazen	4.78	0.54	0.22	0.56	14.79
	Char	32.59	10.29	0.14	0.74	7.45
	Amituk	18.95	6.83	0.10	1.03	39.49
2012	**Resolute**	40.62	3.84	0.39	1.69	9.81
	Hazen	10.81	2.59	0.29	0.88	51.93
	Char	36.50	4.80	0.16	0.88	10.44
	Amituk	12.51	4.01	0.13	0.75	22.84
2013	**Resolute**	42.50	2.72	0.13	1.34	15.39
	Hazen	6.53	0.86	0.22	1.21	23.14
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	15.22	5.40	0.08	1.46	56.33
2015	**Resolute**	91.95	4.38	0.12	2.26	15.34
	Hazen	7.13	0.96	0.14	1.17	13.01
	Char	n.a	n.a	n.a	n.a	n.a
	Amituk	15.71	5.96	0.03	1.21	13.01

Table S5. Geometric mean concentration (ng/g wet weight) of Toxaphene congeners (P26, P50 and P62) in Resolute, Hazen, Char and Amituk lakes in a subset of years from 2005 to 2015

		P26	P50	P62
2005	**Resolute**	n.a	n.a	n.a
	Hazen	4.93	3.90	3.09
	Char	n.a	n.a	n.a
	Amituk	n.a	n.a	n.a
2012	**Resolute**	0.45	0.40	0.03
	Hazen	7.86	9.33	2.02
	Char	0.66	0.70	0.04
	Amituk	1.01	1.77	0.95
2013	**Resolute**	0.40	0.20	n.a
	Hazen	3.08	3.41	0.95
	Char	n.a	n.a	n.a
	Amituk	4.56	7.16	3.37
2015	**Resolute**	0.32	0.20	0.14
	Hazen	1.27	1.32	0.02
	Char	n.a	n.a	n.a
	Amituk	1.46	2.46	0.03

Table S6. Concentrations of ΣPCBs, ΣDDTs, ΣHCHs, HCB and Toxaphene in landlocked Arctic char from this study and comparison with literature available. Concentrations are reported in ng/g wet wt.

***only p,p'-DDE**

Lakes or sampling area	Location	Sample	Sampling period	ΣPCBs	ΣDDTs	ΣHCHs	HCB	Toxaphene	Ref
Resolute Lake	Cornwallis Is. Arctic	Arctic char (*Salvelinus alpinus*)	1997-2015	33-120 (Σ87PCBs)	2-8.7	0.12-0.84	0.86-2.41	1.60-15.39	**This study**
Char Lake	Cornwallis Is. Arctic	Arctic char (*Salvelinus alpinus*)	1993-2012	18-127 (Σ87PCBs)	4.8-52	0.10-2.75	0.59-2.47	4.17-23	**This study**
Amituk Lake	Cornwallis Is. Arctic	Arctic char (*Salvelinus alpinus*)	2001-2015	12-125 (Σ87PCBs)	1.06-61.12	0.03-4.24	0.53-4.37	13.01-171.53	**This study**
Lake Hazen	Ellesmere Is. Arctic	Arctic char (*Salvelinus alpinus*)	1990-2015	4.78-37 (Σ87PCBs)	0.54-11.61	0.14-2.05	0.48-2.46	10.50-125	**This study**
Unnamed lake	Greenland	Arctic char (*Salvelinus alpinus*)	1994-2008	1.53-16.32 (Σ10PCBs)	0.74-7.51	0.05-0.248	0.245-0.725		**Riget et al 2010**
Lake Laberge	Yukon Territory	Lake trout (Salvelinus namaycush)	1993-2010	12-328 (Σ104PCBs)	37.5-391.5	0.10-6.5			**Ryan et al 2013**
Lake near Isortoq	Southwest Greenland	Arctic char (*Salvelinus alpinus*)	1986-2012					0.61 (Σ6Toxaphene)	**Vorkamp et al, 2015**
Lake Ellasjoen	Bear Is, Norway, Arctic	Arctic char (*Salvelinus alpinus*)		694±1009 (Σ7PCBs)	58*				**Evenset et al 2004**
Lake Oyangen	Bear Is, Norway, Arctic	Arctic char (*Salvelinus alpinus*)		49(Σ7PCBs)	3.4*				**Evenset et al 2004**
Lake in Avanersuaq north-west	Greenland	Arctic char (*Salvelinus alpinus*)	1994-1995	8.2±4.7 (Σ11PCBs)	2.9±1.6	0.52±0.28	1.3±0.95	18±14 (Σ4Toxaphene)	**Cleemann et al 2000**
Lake in Nuuk mid-west	Greenland	Arctic char (*Salvelinus alpinus*)	1994-1995	8.9±4.8 (Σ11PCBs)	2.5±1.7	0.66±0.43	0.72±0.59	11±9.5 (Σ4Toxaphene)	**Cleemann et al 2000**
Lake in Oaqortoq south	Greenland	Arctic char (*Salvelinus	1994-1995	16±16 (Σ11PCBs)	7.5±6.7	0.36±0.17	0.72±1.2	11±6.3 (Σ4Toxaphe	**Cleemann et al 2000**

		alpinus)						ne)	
Lake in Tassilaq mid-east	Greenland	Arctic char (*Salvelinus alpinus*)	1994-1995	36±44 (Σ11PCBs)	12±8.8	0.67±0.58	1.8±1.7	49±49 (Σ4Toxaphene)	**Cleemann et al 2000**
Lake Blasjön	Northern Sweden	Arctic char (*Salvelinus alpinus*)_dwards form	1990	13.6 (Σ41PCBs)	3.8*				**Hammar et al, 1993**
Lake Blasjön	Northern Sweden	Arctic char (*Salvelinus alpinus*)_normal form	1990	3.98 (Σ41PCBs)	1.18*				**Hammar et al, 1994**
Peter Lake	Northwest Territories	Arctic char (*Salvelinus alpinus*)	1992-1995	9.88±3.19 (Σ102PCBs)	4.62±1.71	1.68±0.80			**Kidd et al,1998**
Peter Lake	Northwest Territories	Lake trout (Salvelinus namaycush)	1992-1995	81.6±176 (Σ102PCBs)	57.9±148	2.16±4.30			**Kidd et al,1998**
Great Slave Lake (West basin)	Northwest Territories	Lake trout (Salvelinus namaycush)	2011	8.2 ± 2.0 (Σ87PCBs)	1.2 ± 0.49	0.17 ± 0.05	2.0 ± 0.44	7.2± 2.4	**Muir et al. 2013**
Great Slave Lake (East arm)	Northwest Territories	Lake trout (Salvelinus namaycush)	2011	26 ± 11 (Σ87PCBs)	2.8 ± 1.3	0.24 ± 0.14	2.5±0.90	11 ± 3.6	**Muir et al. 2013**
Lac Ste Therese	Northwest Territories	Lake trout (Salvelinus namaycush)	2002	45 ± 13 (Σ87PCBs)	14 ± 4.2	0.41 ± 0.74	1.6 ± 1.2	-	**Muir et al. 2013**
Great Bear Lake	Northwest Territories	Lake trout (Salvelinus namaycush)	2007	65 ± 33 (Σ87PCBs)	16 ± 10	0.49 ± 0.36	2.1 ± 1.2	134 ± 118	**Muir et al. 2013**
Makkovik	Labrador	Arctic char (*Salvelinus alpinus*) sea run		10.7	1.7	1.9			**Muir et al, 2000**
Nain	Labrador	Arctic char (*Salvelinus alpinus*) sea run		30.9	2.5	3.1			**Muir et al, 2000**
Kangirsuk	Nunavik (Quebec)	Arctic char (*Salvelinus alpinus*) sea run		21.4	1.3	1.2			**Muir et al, 2000**
Quaqtaq	Nunavik (Quebec)	Arctic char (*Salvelinus alpinus*) sea run		18.2	1.4	1.2			**Muir et al, 2000**

Table S7. Concentration (ng/g dry weight) of PCBs and HCB in soils from catchment areas of Resolute, Amituk and North lakes (Cornwallis Island). <LOD: below detection limit

	Resolute Lake	Amituk Lake	North Lake
PCB1	0.002	<LOD	<LOD
PCB3	0.002	<LOD	<LOD
PCB8	0.003	0.001	0.001
PCB10+4	0.001	<LOD	<LOD
PCB11	0.002	0.001	0.001
PCB15	0.001	0.001	0.001
PCB18	0.006	0.001	0.001
PCB19	0.001	<LOD	<LOD
PCB22	0.003	0.001	0.001
PCB28	0.019	0.005	0.005
PCB33	0.005	0.001	0.001
PCB37	0.005	0.001	0.001
PCB41	0.034	0.004	0.005
PCB49	0.023	0.002	0.004
PCB52	0.096	0.009	0.012
PCB54	<LOD	<LOD	<LOD
PCB60	<LOD	<LOD	<LOD
PCB66	0.019	0.002	0.003
PCB70	0.067	0.006	0.005
PCB74	0.014	0.003	0.002
PCB77	0.018	0.000	0.001
PCB81	<LOD	<LOD	<LOD
PCB87	0.104	0.001	0.003
PCB95	0.121	0.005	0.006
PCB99	0.065	0.001	0.002
PCB101	0.174	0.005	0.006
PCB104	<LOD	<LOD	<LOD
PCB105	0.084	0.000	0.002
PCB110	0.170	0.003	0.005
PCB114	<LOD	<LOD	<LOD
PCB118	0.200	0.002	0.005
PCB119	0.002	<LOD	<LOD
PCB123	<LOD	<LOD	<LOD
PCB126	0.005	<LOD	<LOD
PCB128	0.048	<LOD	<LOD
PCB129	0.013	0.000	0.000
PCB137	0.012	<LOD	<LOD
PCB138	0.209	0.001	0.005
PCB141	0.043	0.000	0.001
PCB149	0.157	0.001	0.005
PCB151	0.051	0.001	0.002
PCB153+168	0.215	0.001	0.005
PCB155	<LOD	<LOD	<LOD
PCB156	0.023	<LOD	<LOD
PCB157	0.005	<LOD	<LOD

PCB158	0.022	<LOD	<LOD
PCB167	0.013	<LOD	<LOD
PCB169	0.000	<LOD	<LOD
PCB170	0.055	<LOD	0.001
PCB171	0.015	<LOD	<LOD
PCB177	0.036	<LOD	<LOD
PCB178	0.021	<LOD	<LOD
PCB180+193	0.165	<LOD	<LOD
PCB183	0.052	<LOD	<LOD
PCB187	0.140	<LOD	<LOD
PCB188	0.001	<LOD	<LOD
PCB189	0.002	<LOD	<LOD
PCB191	0.002	<LOD	<LOD
PCB194	0.047	<LOD	<LOD
PCB199	0.089	<LOD	<LOD
PCB201	0.013	<LOD	<LOD
PCB202	0.020	<LOD	<LOD
PCB203	0.083	<LOD	<LOD
PCB205	0.002	<LOD	<LOD
PCB206	0.024	<LOD	<LOD
PCB208	0.008	<LOD	<LOD
PCB209	0.001	<LOD	<LOD
HCB	0.134	0.013	0.029
Σ1Cl	0.004	<LOD	<LOD
Σ2Cl	0.006	0.002	0.002
Σ3Cl	0.039	0.009	0.010
Σ4Cl	0.272	0.027	0.030
Σ 5Cl	0.926	0.018	0.029
Σ6Cl	0.811	0.005	0.016
Σ7Cl	0.488	<LOD	0.001
Σ8Cl	0.254	<LOD	<LOD
Σ9Cl	0.032	<LOD	<LOD
Σ10Cl	0.001	<LOD	<LOD
Σ_{70}PCBs	**2.834**	**0.061**	**0.089**

Table S8. Correlation coefficients across all char samples and years (n=474) for Σ_{87}PCBs, OCPs (ΣDDTs, ΣHCHs, HCB) and Toxaphene

	ΣPCBs	ΣDDT	ΣHCH	HCB	Toxaphene
Σ_{87}PCBs	1.00				
ΣDDT	0.53**	1.00			
ΣHCH	0.09**	0.10**	1.00		
HCB	0.29**	0.28**	0.33**	1.00	
Toxaphene	p>0.05	0.13*	0.19*	0.21*	1.00

**p<0.001, the data were log-transformed before analysis.

*p<0.05, the data were log-transformed before analysis

Table S9. Influence of Biological Parameters in ΣPCBs, ΣDDTs, ΣHCHs, HCB and Toxaphene Concentrations (in Log ng/g lipid wt) in landlocked char from Resolute Lake over 1997-2015. Table shows regression coefficient (r^2) only if r^2 is >0.1. ** p-value<0.001

	δ^{13}C	**δ^{15}N**	**Log Length**	**Log Weight**	**Log Age**	**Log CF**	**Year**
Log Σ_{87}PCBs		0.18** n=162					
Log ΣDDTs		0.15** n=161					
Log ΣHCHs							0.48** n=164
Log Toxaphene			0.22 ** n=97	0.19** n=97			0.45** n=97
Log HCB							

Table S10. Influence of Biological Parameters in ΣPCBs, ΣDDTs, ΣHCHs, HCB and Toxaphene Concentrations (in Log ng/g lipid wt) in landlocked char from Lake Hazen over 1990-2015. Table shows regression coefficient (r^2) only if r^2 is >0.1. ** p-value<0.001

	$\delta^{13}C$	$\delta^{15}N$	Log Length	Log Weight	Log Age	Log CF	Year
Log Σ_{87}PCBs		0.13** n=131	0.13** n=141				0.26** n=146
Log ΣDDTs		0.20** n=131					0.40** n=147
Log ΣHCHs							0.52** n=147
Log Toxaphene							0.11** n=139
Log HCB							0.12** n=147

Table S11. Influence of Biological Parameters in ΣPCBs, ΣDDTs, ΣHCHs, HCB, and Toxaphene Concentrations (in Log ng/g lipid wt) in landlocked char from Amituk Lake over 1989-2015. Table shows regression coefficient (r^2) only if r^2 is >0.1 * p-value<0.05, ** p-value<0.001

	$\delta^{13}C$	$\delta^{15}N$	Log Length	Log Weight	Log Age	Log CF	Year
Log Σ_{87}PCBs	0.10* n=84				0.10* n=95		0.39** n=107
Log ΣDDTs							0.37** n=107
Log ΣHCHs	0.39** n=84						0.84** n=107
Log Toxaphene	0.14* n=82						0.41** n=104
Log HCB							0.37** n=107

Table S12. Influence of Biological Parameters in ΣPCBs, ΣDDTs, ΣHCHs, HCB and Toxaphene Concentrations (in Log ng/g lipid wt) in landlocked char from Char Lake over 1993-2012. Table shows regression coefficient (r^2) only if r^2 is >0.1. * p-value<0.05, ** p-value<0.001

	$\delta^{13}C$	$\delta^{15}N$	Log Length	Log Weight	Log Age	Log CF	Year
Log Σ_{87}PCBs		0.13* n=42		0.11* n=44		0.17* n=44	0.31** n=55
Log ΣDDTs		0.12* n=42			0.31* n=28	0.14* n=44	0.55** n=55
Log ΣHCHs					0.15* n=28		0.74** n=55
Log Toxaphene		0.11* n=38				0.17* n=40	0.38** n=51
Log HCB		0.14* n=42		0.11* n=44		0.23* n=44	0.34** n=55

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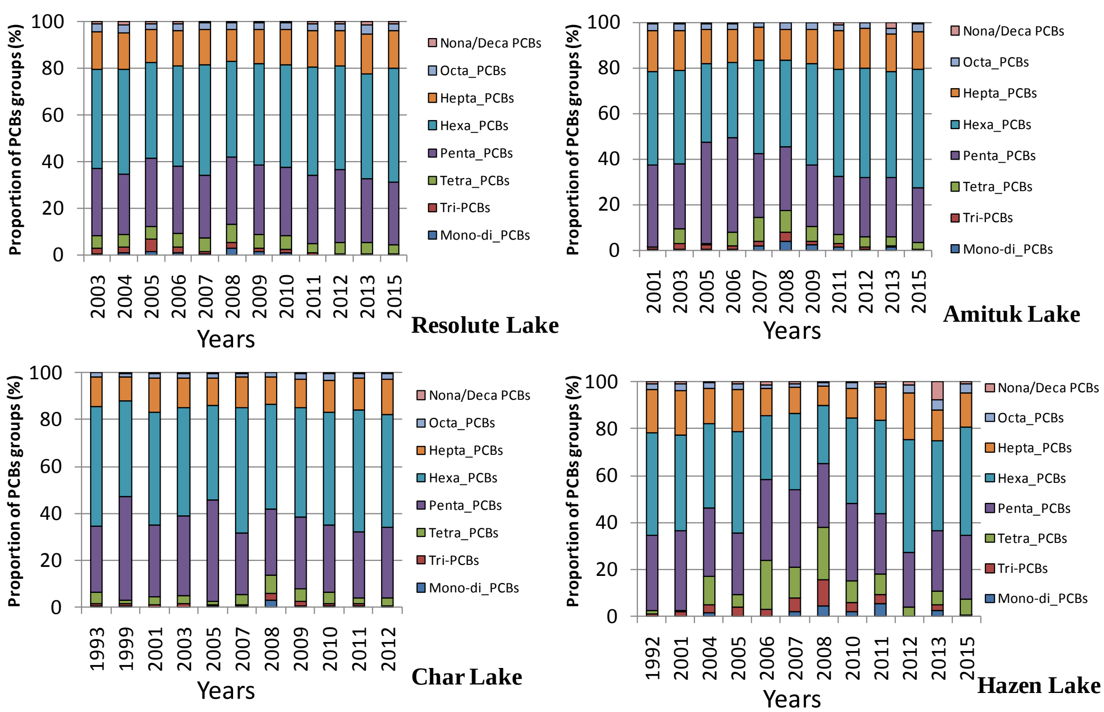

Figure S5. Long term trends in proportion of PCBs congeners groups in landlocked Arctic char from Resolute Lake, Amituk Lake, Char Lake and Lake Hazen.

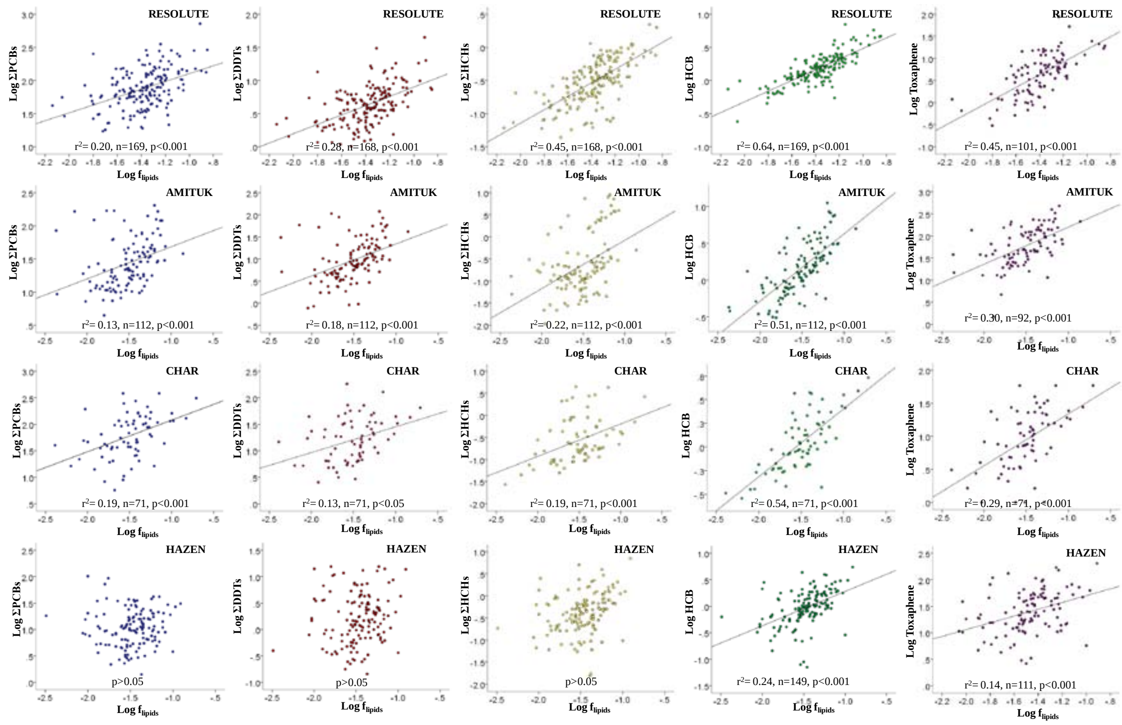

Figure S6. Influence of lipid content on Log ΣPCBs, OC pesticides (ΣDDTs, ΣHCHs and HCB) and Toxaphene (in ng/g wet weight), in Resolute, Amituk, Char and Hazen lakes

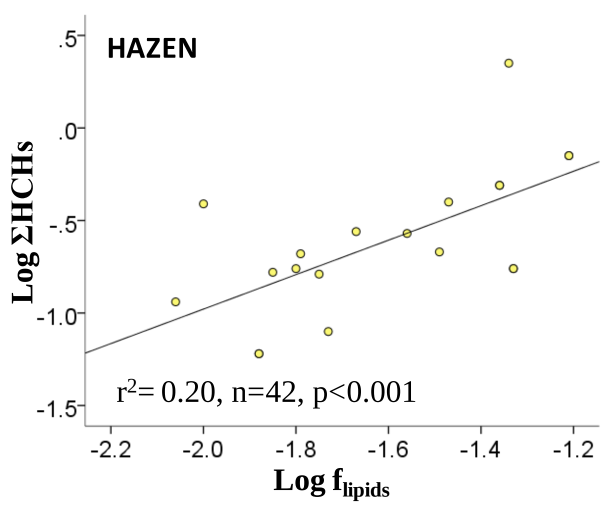

Figure S7. Influence of lipid content on Log ΣHCHs in Hazen Lake for insectivorous Arctic char ($\delta^{15}N$ <12‰)

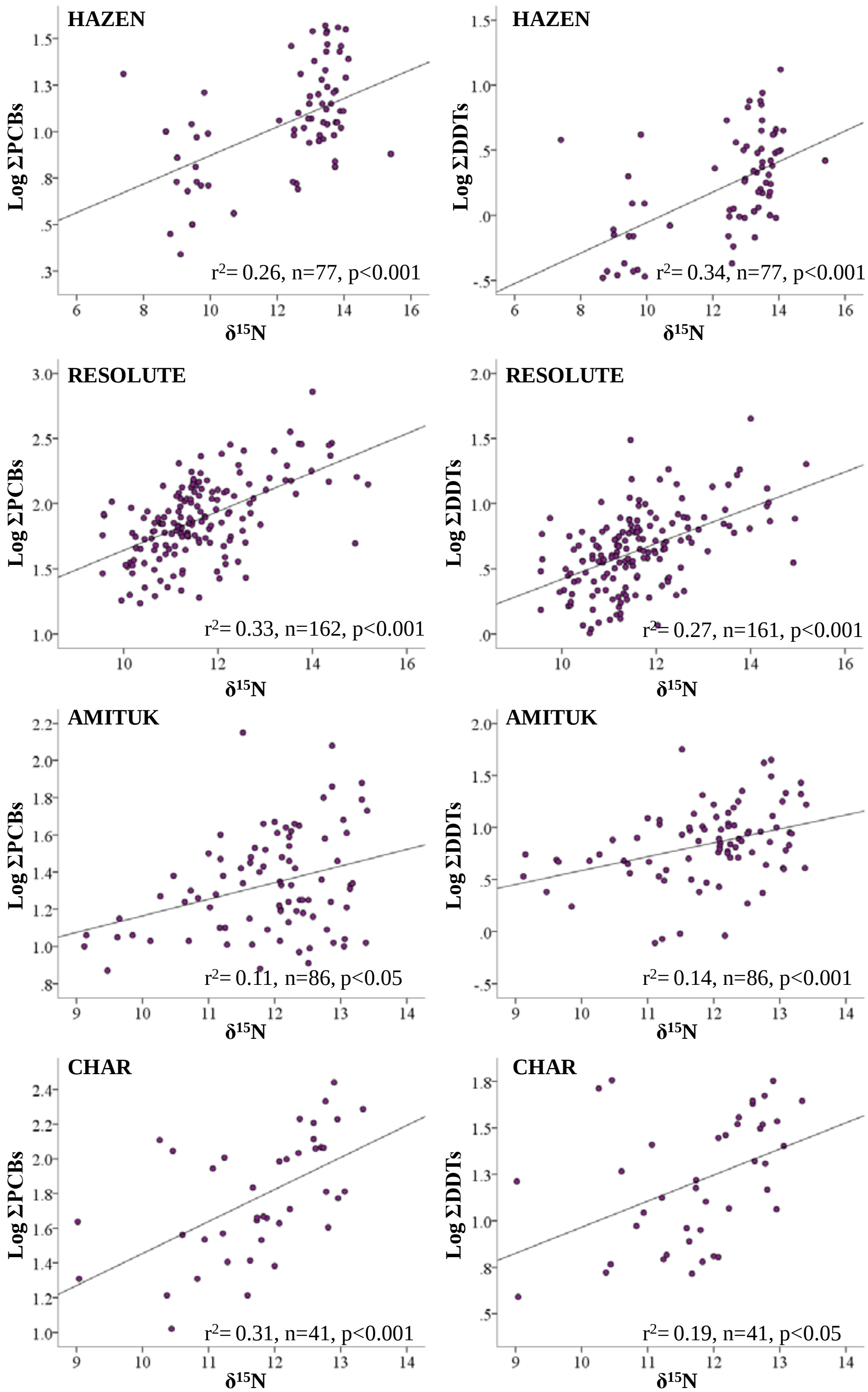

Figure S8. Concentration of PCBs and DDTs vs $\delta^{15}N$ for Lake Hazen, Resolute, Amituk and Char

Annex IV. Trends of legacy POPs in Arctic char

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Table S13. Percent annual decline (negative) and increase (positive) in selected POPs in Arctic Char from from Amituk, Char, Hazen and Resolute Lakes using the PIA program (Bignert, A. (2007). PIA was run using lipid weight concentrations for each sample.

[1]* Indicated statistically significant trend (p<0.05)

[2] Results for toxaphene have 2 or 3 fewer years than other analytes due to use of retrospective analyses for samples from the 1990s and early 2000s

[3] Results for toxaphene in Resolute Lake are from 2003

[4] Sampling of Char Lake was discontinued after 2012.

	Time period	Sampling Years	Toxaphene	ΣPCBs	PCB153	ΣDDTs	α-HCH	β-HCH	HCB
Amituk	1989-2015	14	-6.4*	-6.7*	-5.5*	-7.1*	-14*	-7.3	-4.1*
	Mid-00s-2013	8	-0.41						
Char	1993-2012	12	-5.2	-7.6*	-8.1*	-11*	-14*	+1.8	-4.7*
	Mid-00s-2012	7	+0.2						
Hazen	1990-2015	14	-7*	-6.9*	-6.5*	-10*	-11*	+6.1*	-4.1 *
	Mid-00s-2013	8	-0.08						
Resolute	1997-2015	17	+19*	-3.5	-2.4	-2.3	-7.8*	-3.1	+1.8*
	Mid-00s-2013	9	+9*						

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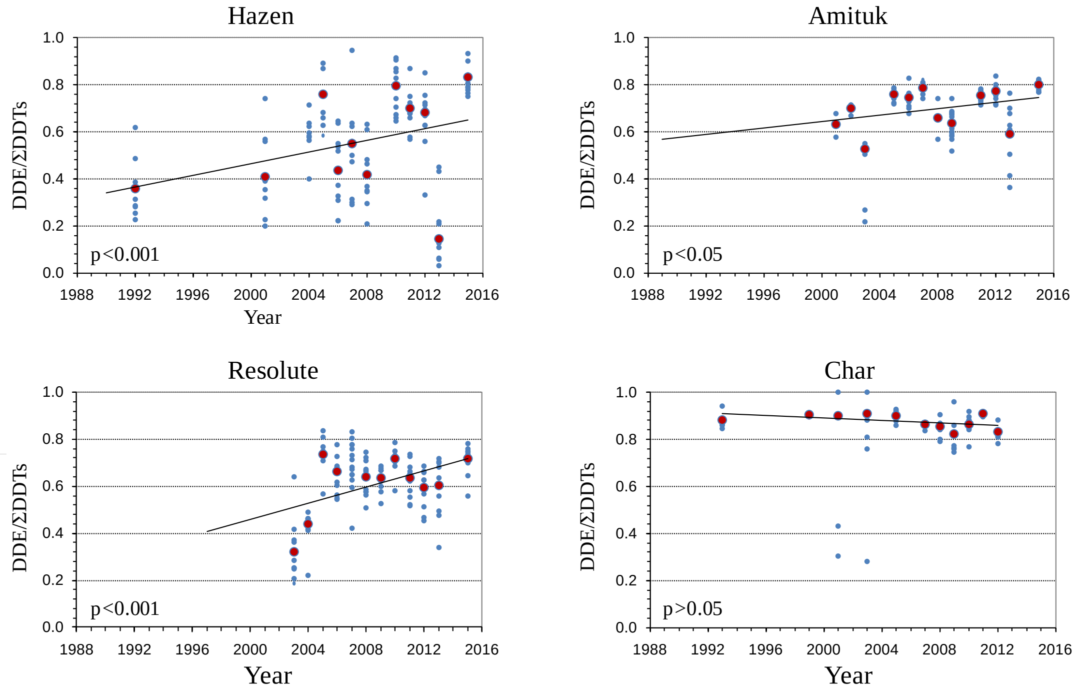

Figure S9. DDE/ΣDDTs ratio versus years of Lake Hazen, Amituk, Resolute and Char lakes

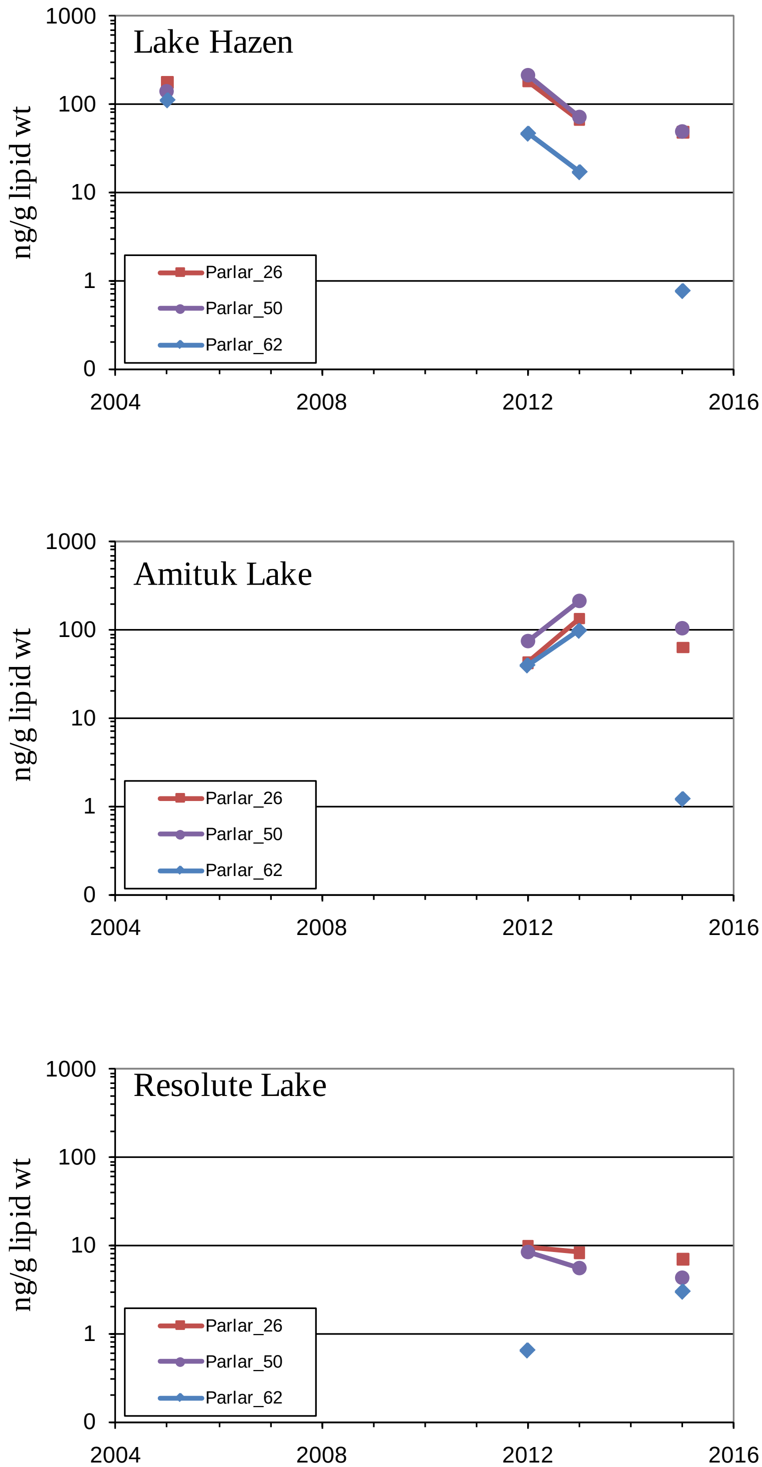

Figure S10. Trends in concentrations (ng/g lipid weight) of individual toxaphene congeners (Parlar 26, 50 and 62) in Arctic char muscle from Hazen Lake (2005 to 2015) and Amituk and Resolute lakes from 2012 to 2015.

ANNEX V. Influence of climatic oscillations on the occurrence of legacy POPs in Arctic char

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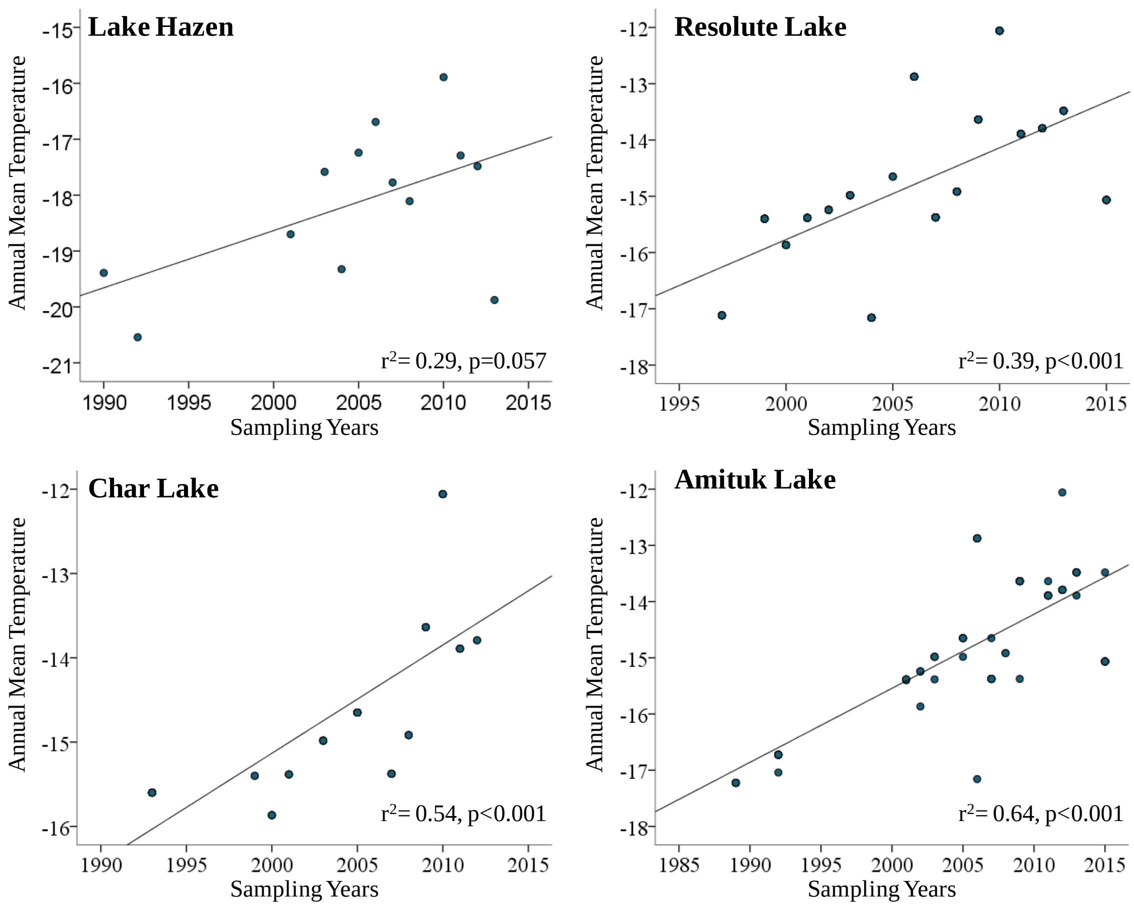

Figure S11. Mean Annual Temperature (C) versus year of sampling in Hazen, Resolute, Char and Amituk lakes

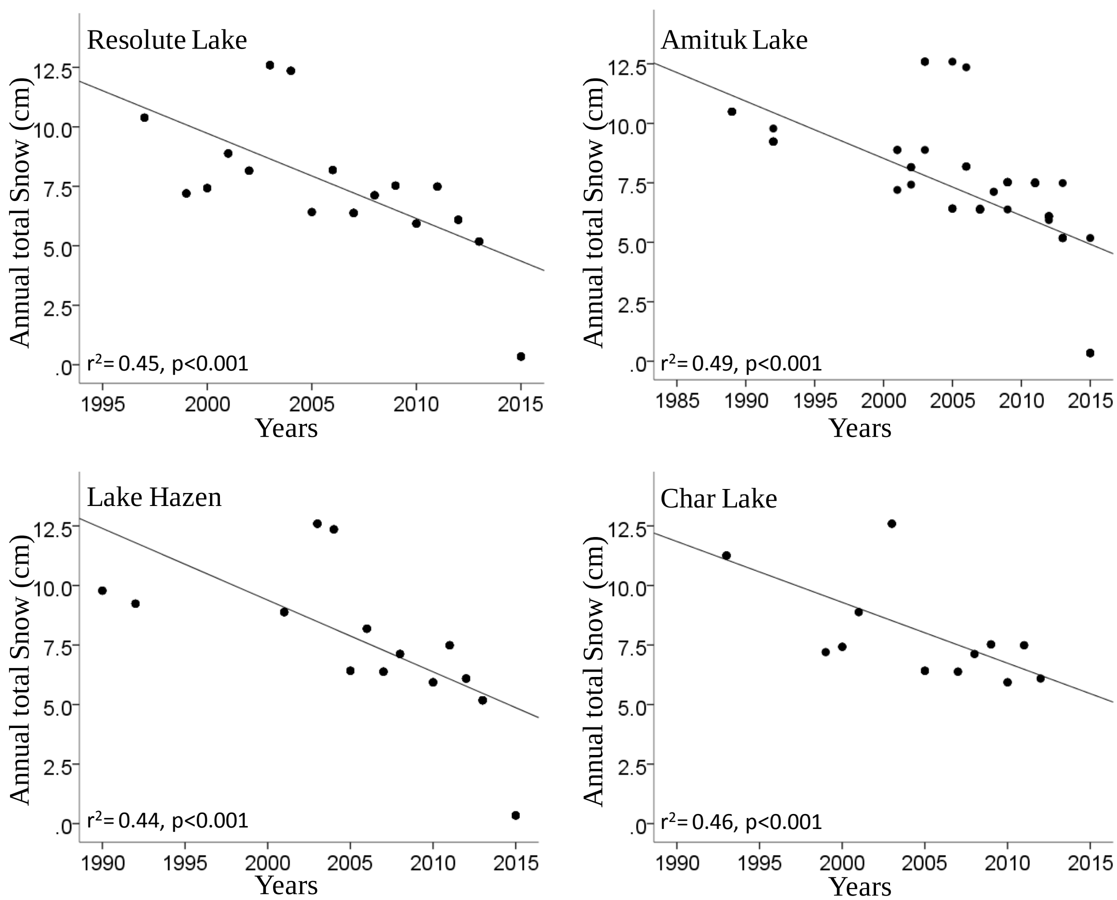

Figure S12. Annual Total Snow (cm) versus Sampling years in Resolute, Amituk, Hazen and Char lakes

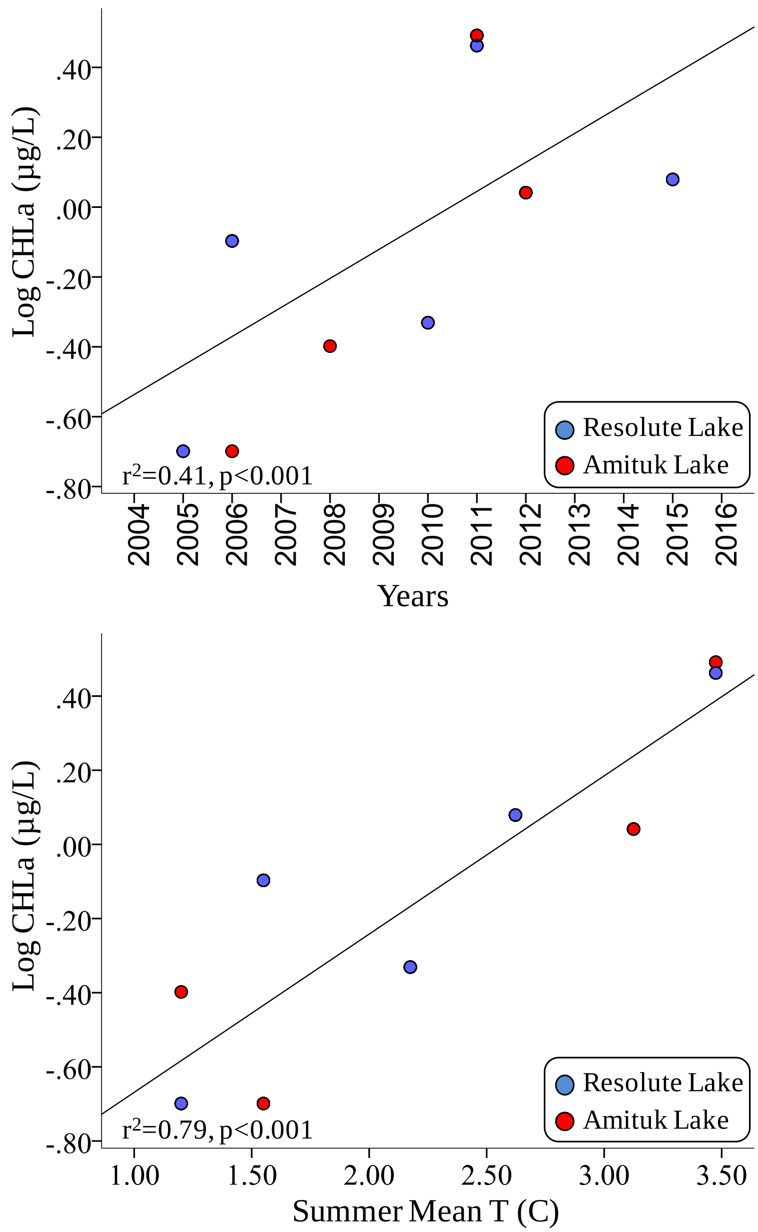

Figure S13. Influence of Year of sampling and Summer Mean Temperature (C) on the Concentrations of Chlorophyll-a in Resolute and Amituk lakes

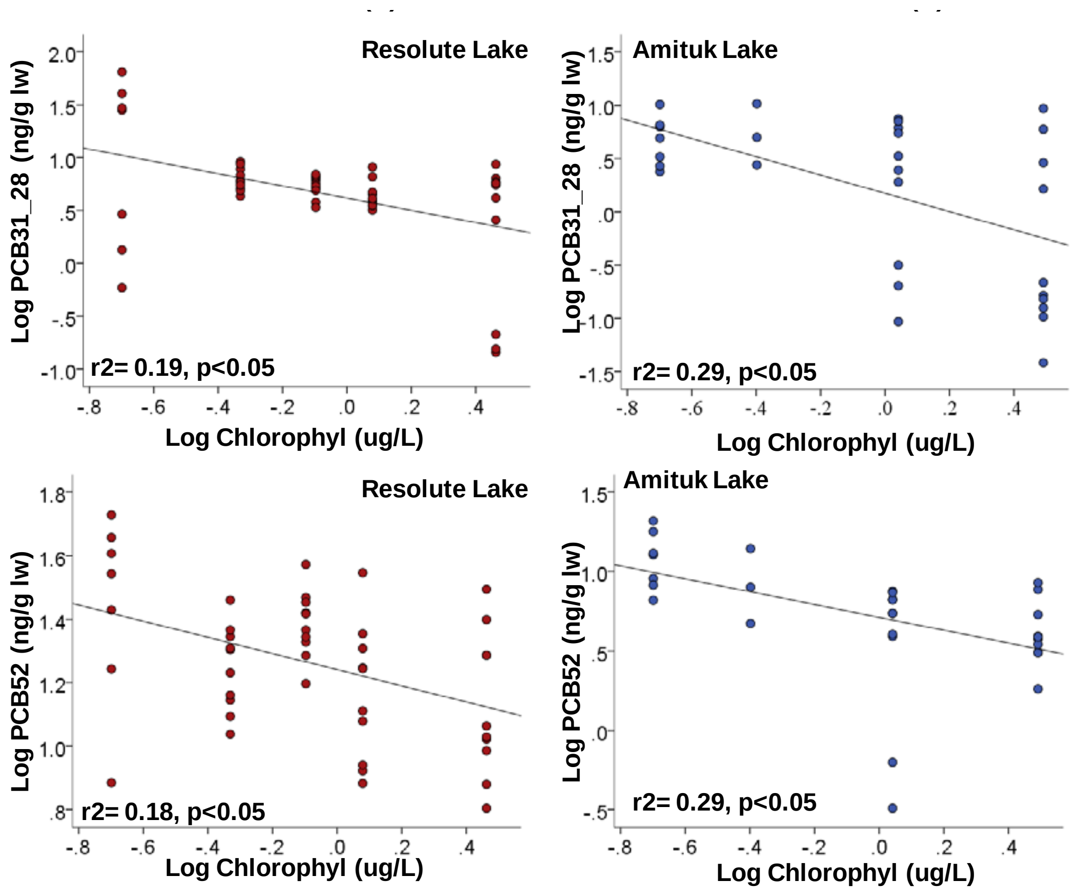

Figure S14. Influence of Chlorophyl-a (μg/L) on the concentrations of PCB31_28, and PCB52 on Arctic char (ng/g lipid wt) at Resolute and Amituk lakes along the temporal series

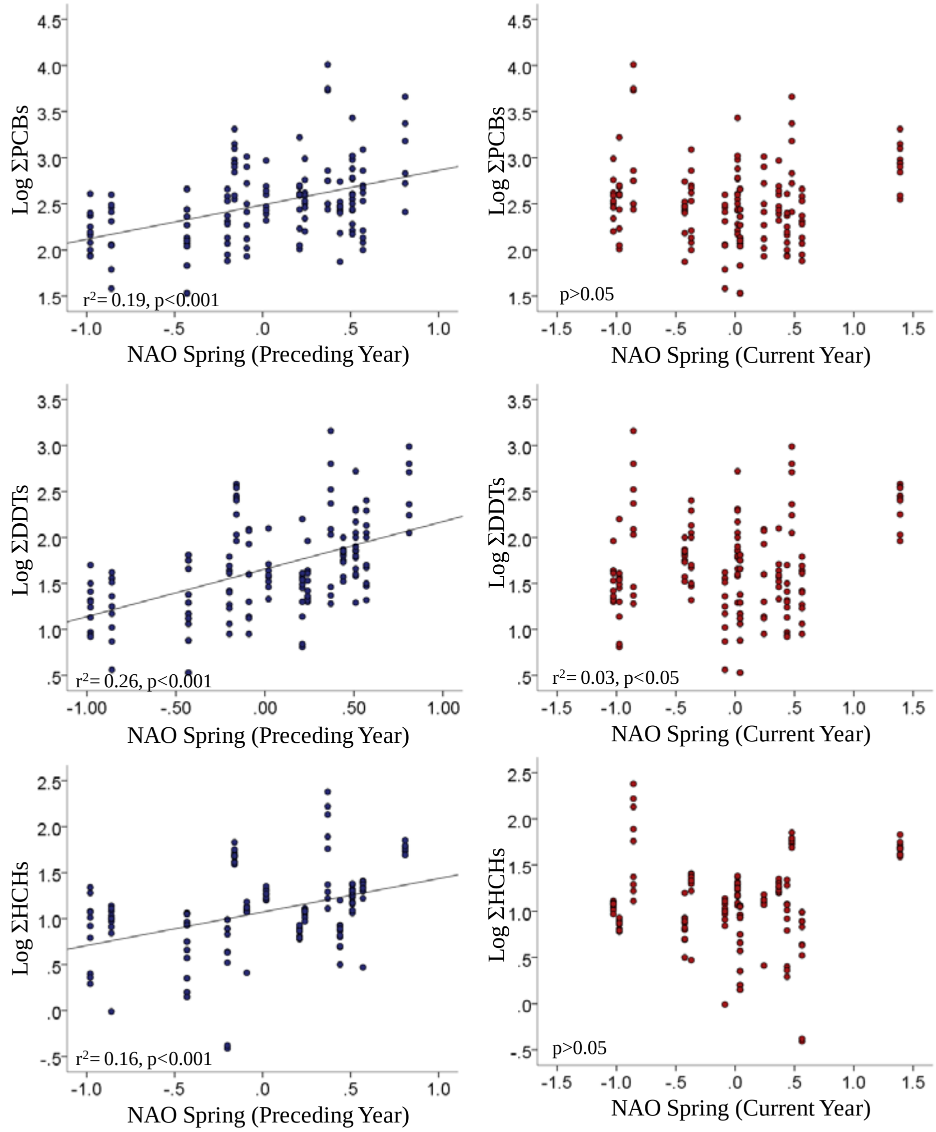

Figure S15. Relationships between the concentrations of ΣPCB, ΣDDTs and ΣHCHs in char from Lake Hazen, and the predictors: NAO in the preceding and current springs

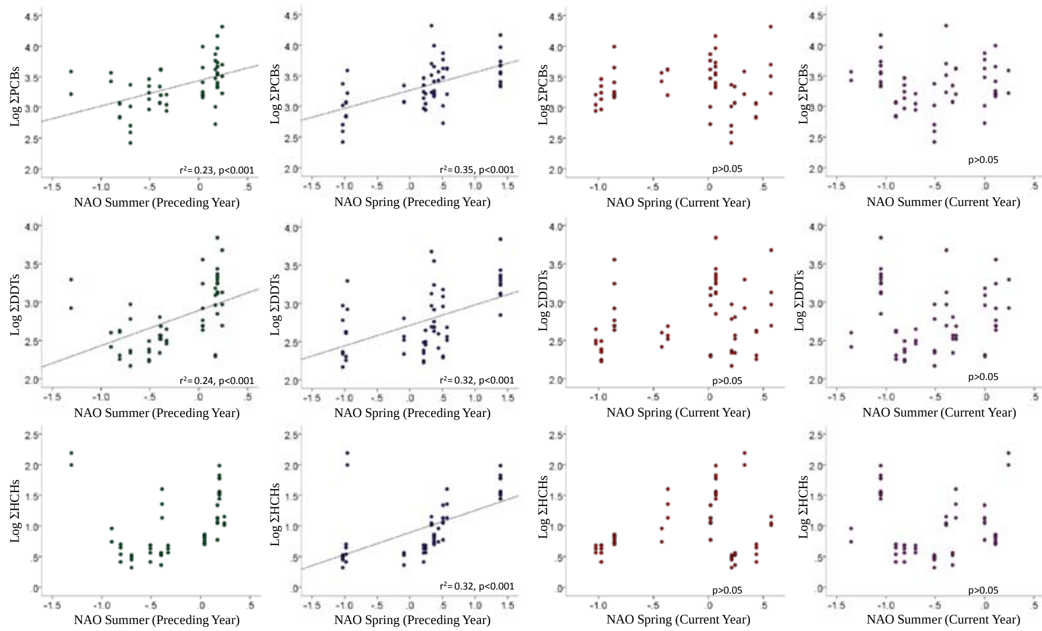

Figure S16. Relationships between the concentrations of ΣPCB, ΣDDTs and ΣHCHs in char from Char Lake, and the predictors: NAO in the preceding and current summers and springs

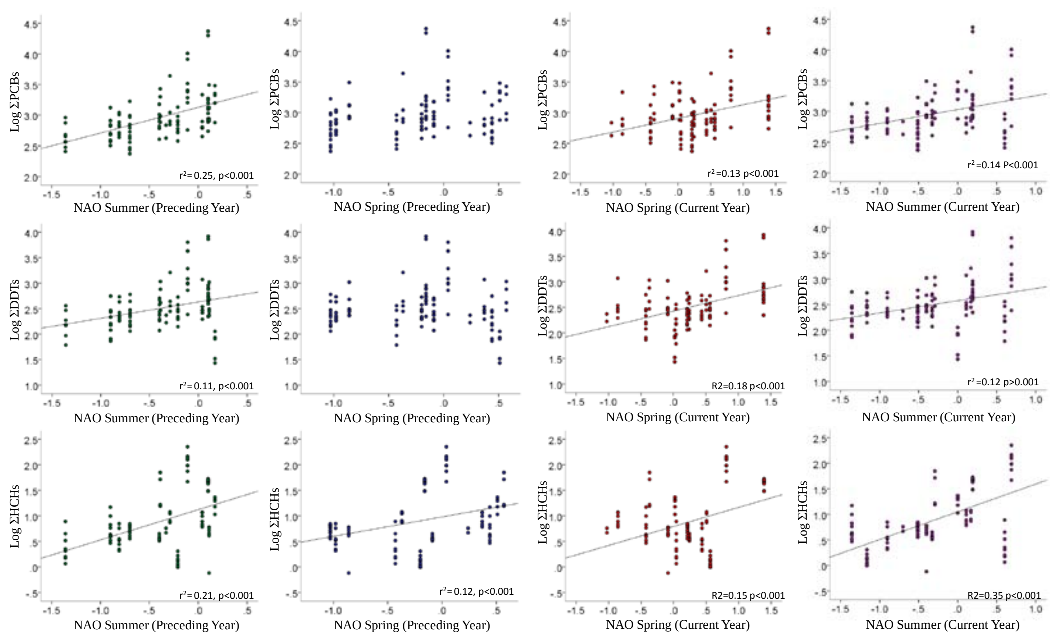

Figure S17. Relationships between the concentrations of ΣPCB, ΣDDTs and ΣHCHs in char from Amituk Lake, and the predictors: NAO in the preceding and current summers and springs

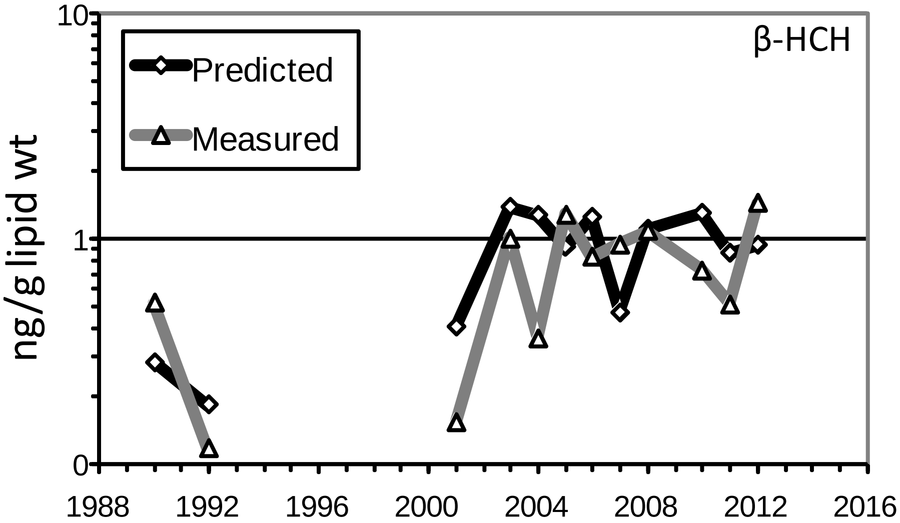

Figure S18. Measured vs predicted concentration of ß-HCH in Lake Hazen.

***ß-HCH predicted concentration estimated using: Log ß-HCH = a + b (year) + c (Log annual Precipitation) + d (NAO spring preceding year)**

Table S14. Influence of year, weight and climatic parameters on the occurrence of ΣPCBs, ΣDDTs and ΣHCHs in Lake Hazen, Amituk, Resolute and Char Lakes.*r^2 is the amount of variation explained by the model. Regression coefficients ± SE are shown for (Weight = Log W; Year = Yr, NAO (summer/spring preceding Yr), Log P= Total Annual Precipitation (snow+rain)

	Time Period	N	Intercept	Year	Log W	NAO	r^2	P
Lake Hazen								
ΣPCBs	1990-2015	129	61±9	-0.029±0.004	0.320±0.105	0.180±0.065	0.40	<0.001
PCB99	1990-2015	129	30±13	-0.015±0.006	0.639±0.127	0.303±0.084	0.35	<0.001
Amituk Lake								
ΣPCBs	1989-2015	96	39±10	-0.018±0.005	0.418±0.111	0.187±0.074	0.41	<0.001
PCB99	1989-2015	96	53±8	-0.026±0.004	0.414±0.094	0.180±0.062	0.59	<0.001
Resolute Lake								
ΣPCBs	1997-2015	165	30±9	-0.014±0.004	0.398±0.130	0.140±0.044	0.18	<0.001
PCB99	1997-2015	165	27±8	-0.013±0.004	0.425±0.112	0.113±0.037	0.19	<0.001
	Time Period	**N**	**Intercept**	**Year**	**Log P**	**NAO**	r^2	**P**
Lake Hazen								
ΣHCHs	1990-2015	147	91±8	-0.045±0.004		0.0122±0.056	0.53	<0.001
Amituk Lake								
ΣHCHs	1989-2015	107	116±6	-0.057±0.003	0.224±0.053	0.164±0.041	0.87	<0.001
Char Lake								
ΣHCHs	1993-2012	55	174±11	-0.086±0.006		-0.425±0.081	0.83	<0.001
Resolute Lake								
ΣHCHs	1997-2015	165	68±7	-0.034±0.003	0.132±0.042	-0.141±0.030	0.55	<0.001
	Time Period	**N**	**Intercept**	**Year**	**Log W**	**NAO**	r^2	**P**
Lake Hazen								
ΣDDTs	1990-2015	129	88±9	-0.043±0.005	0.415±0.115	0.261±0.0.071	0.55	<0.001
Amituk Lake								
ΣDDTs	1989-2015	96	57±10	-0.028±0.005	0.432±0.133	-0.120±0.061	0.33	<0.001
Char Lake								
ΣDDTs	1993-2012							

Resolute Lake								
ΣDDTs	1997-2015	164	29±8	-0.014±0.004	0.327±0.129	-0.094±0.036	0.11	<0.001

Table S15. Results of General Linear Model Analyses and factors affecting the concentration of POPs in Arctic char from Lake Hazen, Amituk, Char and Resolute over the long temporal series *r^2 is the amount of variation explained by the model. Weight = W; Year = Y.

	Time Period	N	Equation	r^2	p
Lake Hazen					
ΣPCBs	1990-2015	128	70 - 0.034Y + 0.355 Log W	0.36	<0.001
PCB99	1990-2015	128	63 - 0.032Y + 0.515 Log W	0.28	<0.001
PCB153	1990-2015	128	66 - 0.033Y + 0.689 Log W	0.30	<0.001
ΣDDTs	1990-2015	128	100 - 0.050Y + 0.466 Log W	0.50	<0.001
p,p'-DDE	1990-2015	128	53 - 0.027Y + 0.697 Log W	0.23	<0.001
ΣHCHs	1990-2015	147	98 - 0.048Y	0.52	<0.001
α-HCH	1990-2015	147	103- 0.051Y	0.49	<0.001
γ-HCH	1990-2015	147	93 - 0.046Y	0.35	<0.001
HCB	1990-2015	147	33 - 0.016Y	0.12	<0.001
Toxaphene	1990-2015	129	47 - 0.023Y + 0.511 Log W	0.23	<0.001
Amituk Lake					
ΣPCBs	1989-2015	96	54 - 0.026Y + 0.421 Log W	0.37	<0.001
PCB99	1989-2015	96	68 - 0.034Y + 0.417 Log W	0.55	<0.001
PCB153	1989-2015	96	37 - 0.018Y + 0.491 Log W	0.27	<0.001
ΣDDTs	1989-2015	96	53 - 0.026Y + 0.452 Log W	0.31	<0.001
p,p'-DDE	1989-2015	96	38 - 0.018Y + 0.480 Log W	0.18	<0.001
ΣHCHs	1989-2015	107	131 - 0.065Y	0.84	<0.001
α-HCH	1989-2015	107	130- 0.064Y	0.71	<0.001
γ-HCH	1989-2015	107	151 - 0.075Y	0.76	<0.001
HCB	1989-2015	96	82 - 0.016Y + 0.175 Log W	0.28	<0.001
Toxaphene	1990-2015	94	72 - 0.035Y + 0.356 Log W	0.48	<0.001
Char Lake					
ΣPCBs	1993-2012	55	72 - 0.034Y	0.31	<0.001
PCB99	1993-2012	55	79- 0.039Y	0.44	<0.001
PCB153	1993-2012	55	81 - 0.039Y	0.39	<0.001
ΣDDTs	1993-2012	55	99 - 0.048Y	0.51	<0.001
p,p'-DDE	1993-2012	55	97- 0.047Y	0.43	<0.001
ΣHCHs	1993-2012	55	136 - 0.067Y	0.74	<0.001
α-HCH	1993-2012	55	136 - 0.068Y	0.79	<0.001
γ-HCH	1993-2012	55	122- 0.061Y	0.56	<0.001
HCB	1993-2012	55	47 - 0.022Y	0.34	<0.001
Toxaphene	1993-2012	51	65 - 0.031 Log W	0.38	<0.001
Resolute Lake					

ΣPCBs	1997-2015	166	41 - 0.091Y + 0.358 Log W	0.12	<0.001
PCB99	1997-2015	166	36 - 0.018Y + 0.392 Log W	0.14	<0.001
PCB153	1997-2015	166	33 - 0.016Y + 0.482 Log W	0.12	<0.001
ΣDDTs	1997-2015	166	27 - 0.013Y + 0.318 Log W	0.07	<0.001
p,p'-DDE	1997-2015				
ΣHCHs	1997-2015	165	68 - 0.034Y	0.47	<0.001
α-HCH	1997-2015	165	79- 0.039Y	0.55	<0.001
γ-HCH	1997-2015	165	105 - 0.052Y	0.36	<0.001
HCB	1997-2015				
Toxaphene	2003-2015	117	-150+ 0.075Y+0.402 Log W	0.51	<0.001

Table S16. Increments of r^2 (in %) when applying equation [1] (which consider climatic parameters) and equation [2] (which does not consider climatic parameters) in the long temporal series of POPs in Arctic char

% increment r^2 calculated as= ((r^2 Equation [1] - r^2 Equation [2])/ r^2 Equation [2])*100

	Time Period	**N**	**Equation [1]**	**r^2**	**Equation [2]**	**r^2**	**% Increment r^2**
Lake Hazen							
ΣPCBs	1990-2015	129	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.40	Y=a+b (Year)+c (Log W)	0.36	**11**
PCB99	1990-2015	129	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.35	Y=a+b (Year)+c (Log W)	0.28	**25**
Amituk Lake							
ΣPCBs	1989-2015	96	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.41	Y=a+b (Year)+c (Log W)	0.37	**11**
PCB99	1989-2015	96	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.59	Y=a+b (Year)+c (Log W)	0.55	**7**
Resolute Lake							
ΣPCBs	1997-2015	165	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.18	Y=a+b (Year)+c (Log W)	0.12	**50**
PCB99	1997-2015	165	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.19	Y=a+b (Year)+c (Log W)	0.14	**36**
	Time Period	**N**	**Equation [1]**	**r^2**	**Equation [2]**	**r^2**	**% Increment r^2**
Lake Hazen							
ΣHCHs	1990-2015	147	Y=a+b (Year)+ b (Log NAO)	0.53	Y=a+b (Year)	0.52	**2**
Amituk Lake							
ΣHCHs	1989-2015	107	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.87	Y=a+b (Year)	0.84	**4**
Char Lake							
ΣHCHs	1993-2012	55	Y=a+b (Year)+ b (Log NAO)	0.83	Y=a+b (Year)	0.74	**12**

Resolute Lake							
ΣHCHs	1997-2015	165	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.55	Y=a+b (Year)	0.47	**17**
	Time Period	**N**	**Equation [1]**	r^2	**Equation [2]**	r^2	**% Increment** r^2
Lake Hazen							
ΣDDTs	1990-2015	129	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.55	Y=a+b (Year)+c (Log W)	0.50	**10**
Amituk Lake							
ΣDDTs	1989-2015	96	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.33	Y=a+b (Year)+c (Log W)	0.31	**6**
Char Lake							
ΣDDTs	1993-2012				Y=a+b (Year)	0.51	
Resolute Lake							
ΣDDTs	1997-2015	164	Y=a+b (Year)+c (Log W or Log P) + d (Log NAO)	0.11	Y=a+b (Year)+c (Log W)	0.07	**57**

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