Supporting Information

for

Sources and Deposition of Polycyclic Aromatic Hydrocarbons to Western U.S. National Parks.

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Lacustrine Catchments. Emerald Lake (Sequoia) is a high-altitude cirque lake (2800 meter above sea level (masl)) with a predominantly granite catchment orientated northwest. Pear Lake (Sequoia) is also a high-altitude cirque lake (2904 masl) located 0.5 km from Emerald Lake; its granite catchment is orientated north. Mills Lake (Rocky) is a high-altitude cirque lake (~3030 masl) located east of the Continental Divide. Lone Pine Lake (Rocky) (~3024 masl) is located approximately 10 km from Mills

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Lake west of the Continental Divide. Hoh Lake (Olympic) is a hanging cirque lake (1384 masl) orientated on the north side of Mt. Olympus. PJ Lake (Olympic) is a hanging cirque lake (1433 masl) orientated on the northeast side of Mt. Olympus, approximately 27 km from PJ Lake. Golden Lake (Rainier) is high-altitude perched lake orientated on the west side of Mt. Rainier. LP19 (Rainier) is an unnamed high-altitude perched lake also orientated on the west side of Mt Rainier, approximately 7.5 km south of Golden Lake. Snyder Lake (Glacier) is a perched cirque lake (1600 masl) located east of the Continental Divide. Oldman Lake (Glacier), a cirque lake (2026 masl), is located approximately 30 km from Snyder Lake west of the Continental Divide. Wonder Lake (Denali) and McLeod Lake (Denali) are a piedmont lakes located approximately 55 km north of Mt. McKinley. Lake Matcharak (Gates) is located in the arctic tundra along the Noatak River south of the central Brooks Range. Burial Lake (Noatak) is located in the arctic and resides in the Foothills of the North Brooks Range approximately 144 km west of Lake Matcharak.

Snowpack Collection and Analysis. Snow sampling was conducted as described previously (1,2). Briefly, season snowpack (50 kg) was collected at the end of the snow accumulation season (March or April) into polytetrafluoroethylene (PTFE) bags from a snow pit. Samples were shipped overnight on dry ice back to the laboratory. Snow samples were shielded from light and were allowed to melt in sealed bags at 22°C overnight prior to extraction using a modified hydrophobic/hydrophilic Speedisk (*1*). Prior to extraction, 1 mL of methanol spiked with isotopically labeled deuterated polycyclic aromatic hydrocarbons (d-PAH) surrogates-ethyl acetate (EA) solution was apportioned amongst the PTFE bags to correct for PAH loss over the entire analytical method. PAHs were isolated and matrix interferences were removed using gel permeation chromatography, followed by silica gel chromatography (*1*). Extracts were concentrated (~200 μL) and spiked with an isotopically labeled d-PAH (internal standard-EA solution. Gas chromatographic mass spectrometry with electron impact ionization (GC/EI-MS) with selective ion monitoring was used for the separation, detection, and quantification of PAHs (*1*).

Lichen Collection and Analysis. In brief, one lichen species was collected within a ~0.01 km^2 sampling area at each site. Lichens were collected from Snyder Lake catchment (*Platismatia glauca*) and Oldman Lake Catchment (*Letharia vulpine*). The lichen samples were collected upon availability at each site by hand and stored in 2-L metalized polyester bags (Kapak Corp, Minneapolis, MN). Once full, the bags were sealed with laboratory tape and double bagged in zipper-locking plastic bags. The samples were stored and shipped overnight to the laboratory in coolers with ice and then stored at -40^{o}C until analysis. The samples were ground using a Büchi Mixer B-400 (Büchi-Mixer B-400, Flawil, Switzerland), packed in 100 mL Accelerated Solvent Extractor (ASE) cells (Dionex, Sunnyvale, CA), spiked with d-PAH surrogates solution, and extracted using pressurized liquid extraction. Lichen samples were extracted twice using an ASE 300 and dichloromethane (100 °C, 1500 psi, 1 cycles of 5 minutes, 75% flush volume). Extract cleanup steps, isotopically-labeled surrogate and internal standard spikes, separation, detection, and quantification (GC/EI-MS) were the same as described above.

Sediment Collection, Dating, and Analysis. Sediment sampling was conducted as described previously (*3*). Briefly, at least two sediment cores were collected from the deepest area of the lake using an Uwitec gravimetric corer and 78 mm polycarbonate core tube. Sediment cores were extruded vertically and sectioned in the field, 0.5 cm intervals for first 10 cm and 1 cm intervals thereafter. Intervals were stored separately in pre-cleaned glass jars. Sediment samples were kept on blue ice and shipped overnight to the laboratory. Sediment cores were identified for SOC analysis based on visual inspection of the sediment core, followed by ^{210}Pb dating. Intervals from within a sediment core were identified based on ^{210}Pb dating and contaminant use profiles. An aliquot from each sediment core interval was analyzed for percent moisture, total organic carbon (TOC), and ^{210}Pb, ^{137}Cs, ^{226}Ra, and ^{241}Am activity. ^{210}Pb, ^{137}Cs, ^{226}Ra, and ^{241}Am were analyzed by direct gamma assay at the Liverpool University Environmental Radioactivity Laboratory, using an Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detector (4). Radiodating of $_{210}$Pb and $_{226}$Ra was determined by the gamma emissions at 46.5 keV and 295 keV, respectively. Complete details of these analyses are provided elsewhere (3,4). The constant rate of supply model, along with measured ^{210}Pb activity, was used to determine the sedimentation rate and average date of each interval (*4-6*). The total and supported ^{210}Pb activity for each core, along with the ^{137}Cs concentrations versus depth are provided in Figure S2 and S15.

The extraction, isolation, and quantification of PAHs in sediment is described elsewhere (*3*). In brief, wet sediment (10-40 g ww) was dried with sodium sulfate and extracted using pressurized liquid extraction. Prior to extraction, samples were spiked with an isotopically labeled recovery surrogates to correct for PAH loss over the analytical method. Extracts were purified using the gel permeation chromatography and silica gel chromatography (*3*). Extract cleanup steps, isotopically-labeled surrogate and internal standard spikes, separation, detection, quantification (GC/EI-MS), recoveries, and WACAP quality assurance objectives have been previously described (3,7). The sediment extracts were analyzed for target PAHs by GC/MS, using electron impact (EI) ionization with selective ion monitoring as described in by Usenko et al (*3*).

Sodium sulfate was used as the laboratory blank and was carried through the entire analytical method (extraction, cleanup, and concentrating), starting at the grinding step, the laboratory blank was spiked with the same quantity of isotopically labeled surrogate and internal standards as samples (*3*). PAH concentrations in sediment were surrogate recovery (concentration calculated relative to surrogate) and laboratory blank corrected.

Sediment Focusing Correction. Focusing factors (FF) were calculated for each sediment core in order to correct for sediment focusing. Focusing factors are used to quantify sedimentation enhancement or reduction at the coring site. The ^{210}Pb inventory was derived by plotting the unsupported ^{210}Pb against the mass sedimentation accumulation rate (8) and the ^{210}Pb atmospheric fallout was modeled from ice cores, soil samples, and atmospheric collectors near sampling sites (9-13). ^{210}Pb atmospheric fallout may vary over short time periods and in mountainous terrain, where precipitation varies greatly with elevation and orientation. However, over the time frame of these sediment cores (<150 years), the ^{210}Pb atmospheric fallout is considered to be fairly constant in a regional context (4,14). The lake sediment core FFs ranged from 0.78 to 4.55 (Table S1). In order to compare the spatial and temporal trends of PAH deposition among different sediment cores, all sediment PAH concentrations were multiplied by the lake sedimentation rate and normalized to the FF. This converted all PAH concentrations in sediment (μg g^{-1} dry wt) to focus-corrected PAH fluxes (μg m^{-2} y^{-1}).

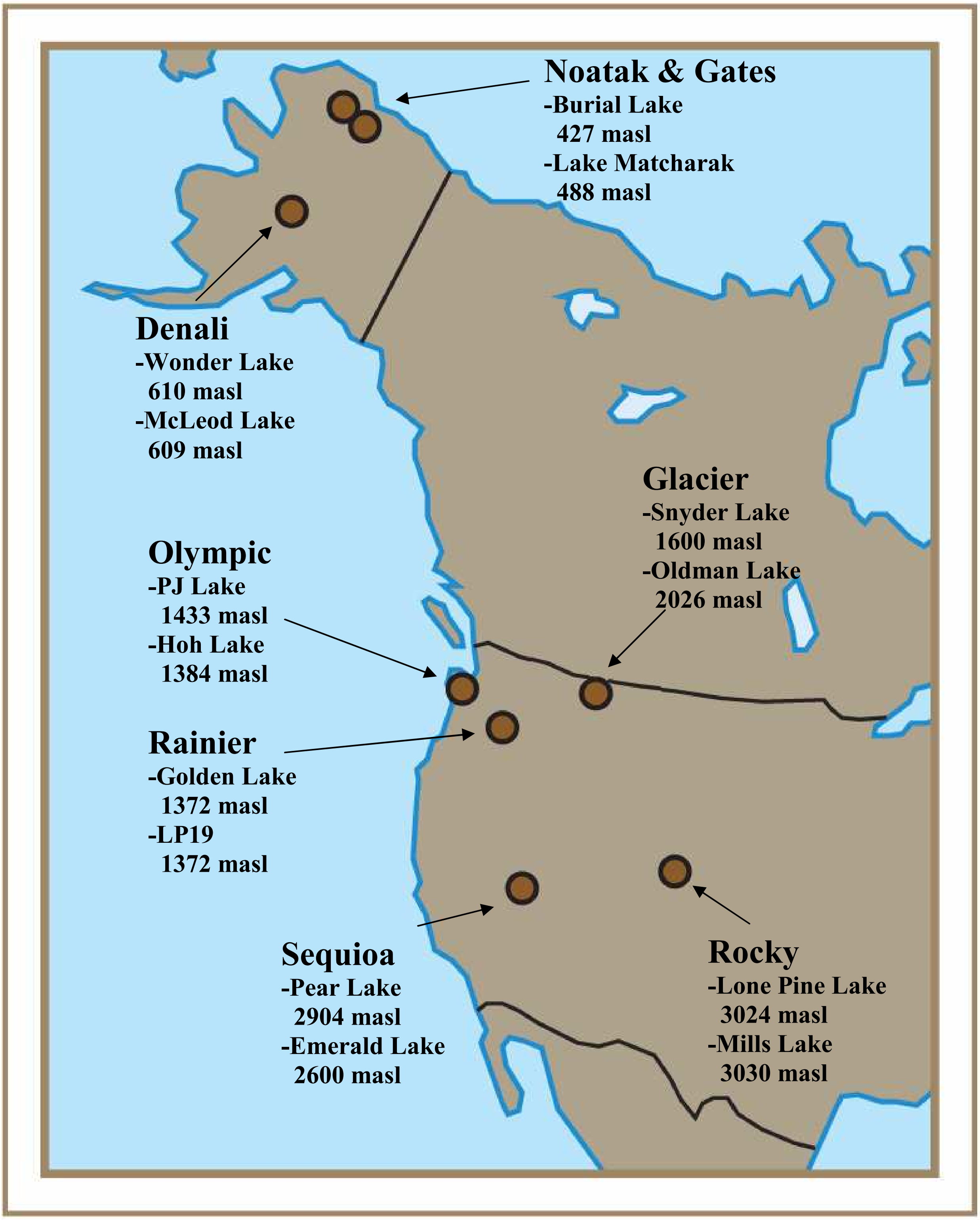

Figure S1. Location of National Parks sampled. Meters above sea level is denoted with masl.

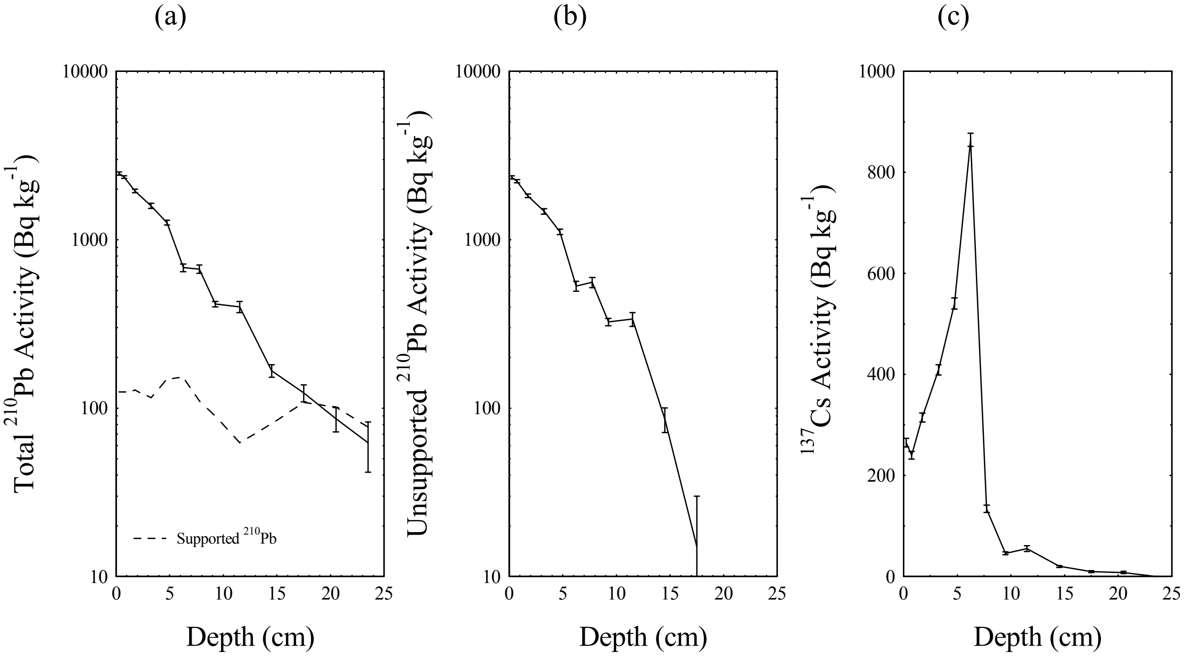

Figure S2: Fallout radionuclides in Pear Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

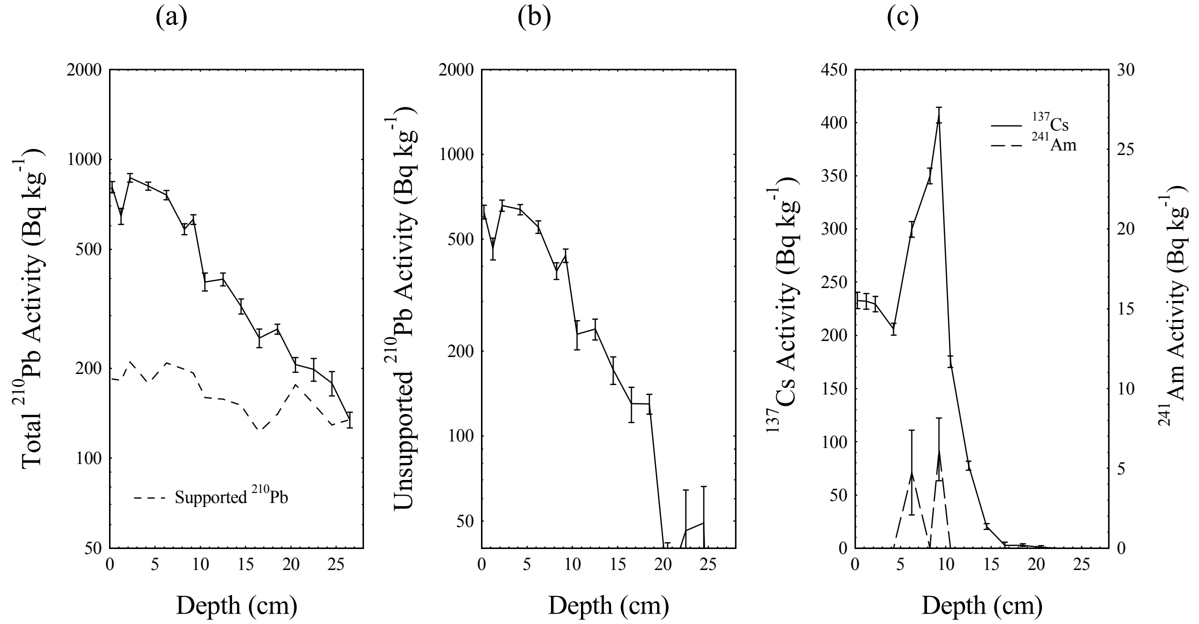

Figure S3: Fallout radionuclides in Emerald Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

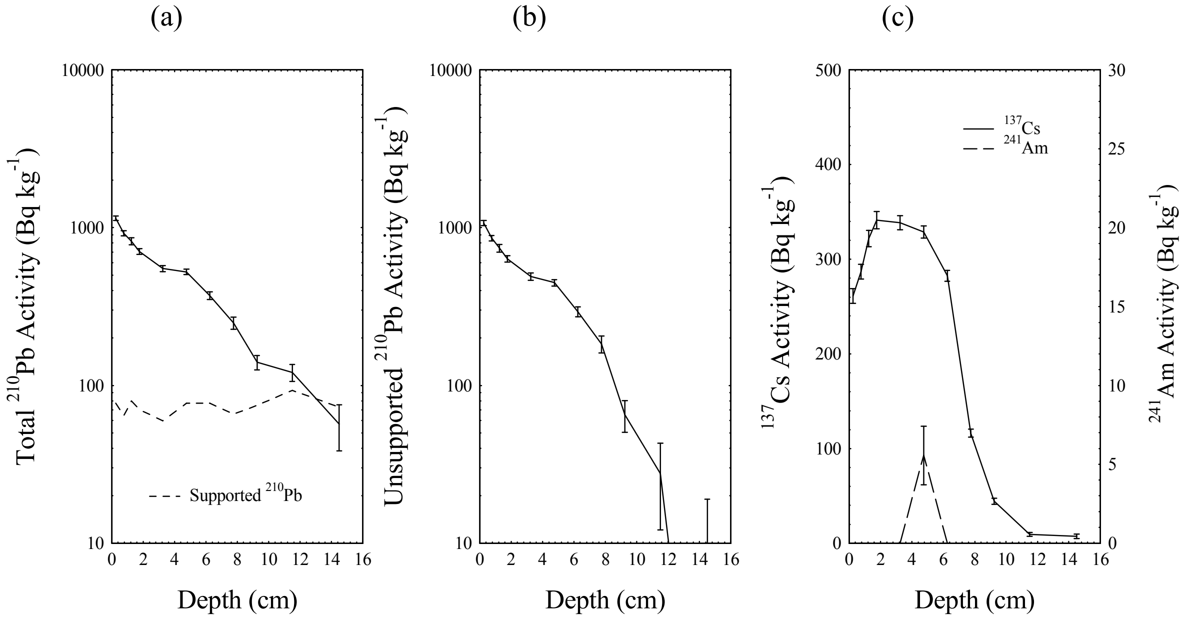

Figure S4: Fallout radionuclides in Lone Pine Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

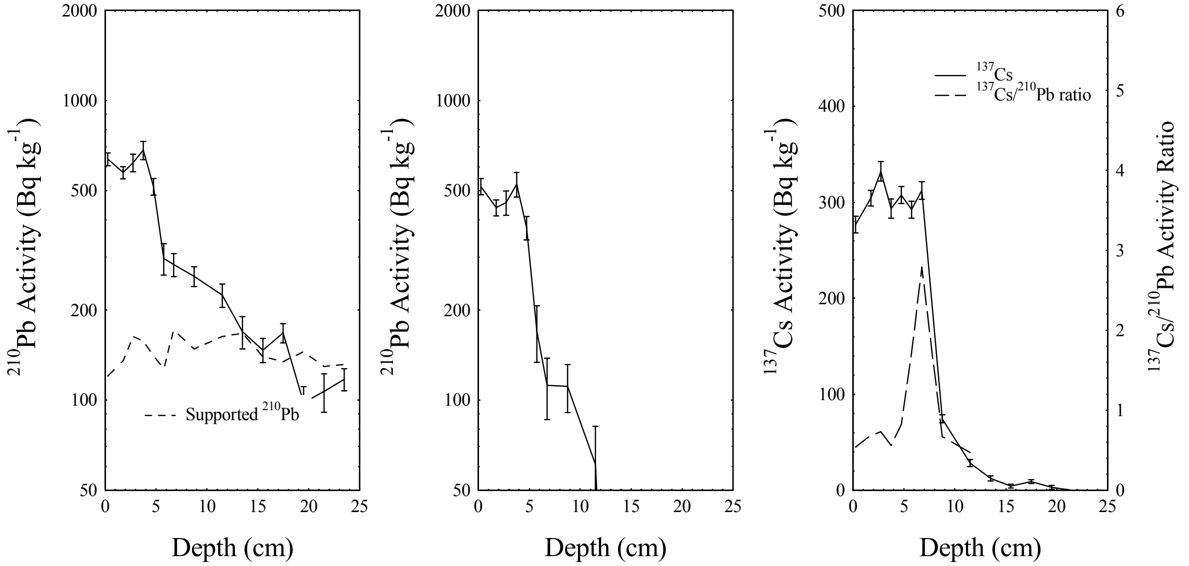

Figure S5: Fallout radionuclides in Mills Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations and ^{137}Cs/^{210}Pb activity ratios versus depth.

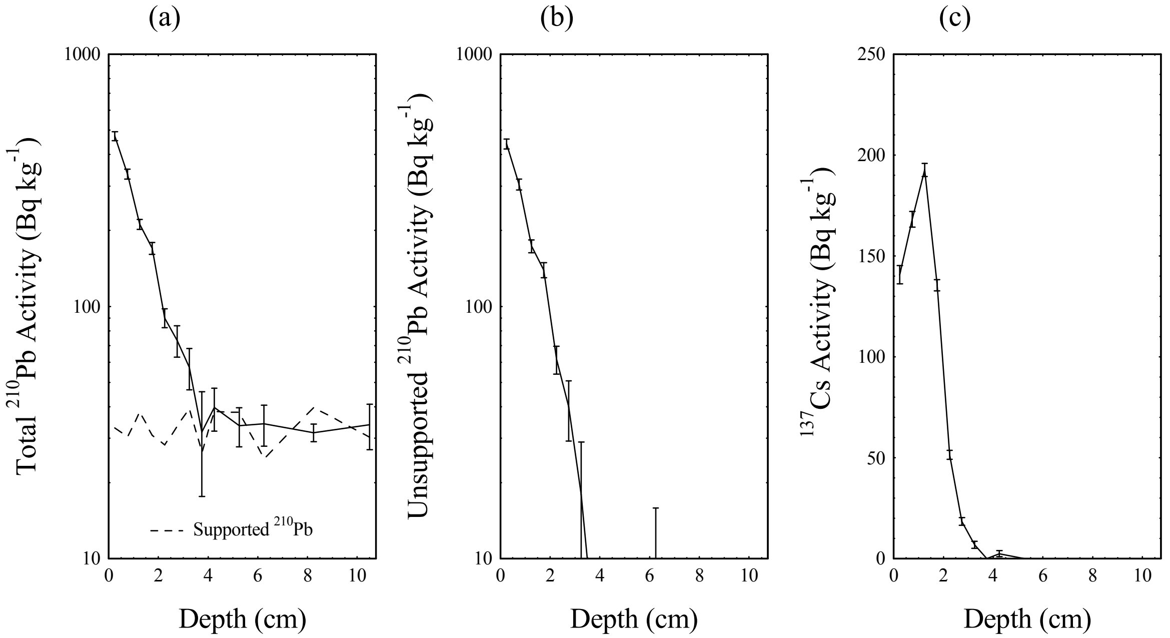

Figure S6: Fallout radionuclides in Burial Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

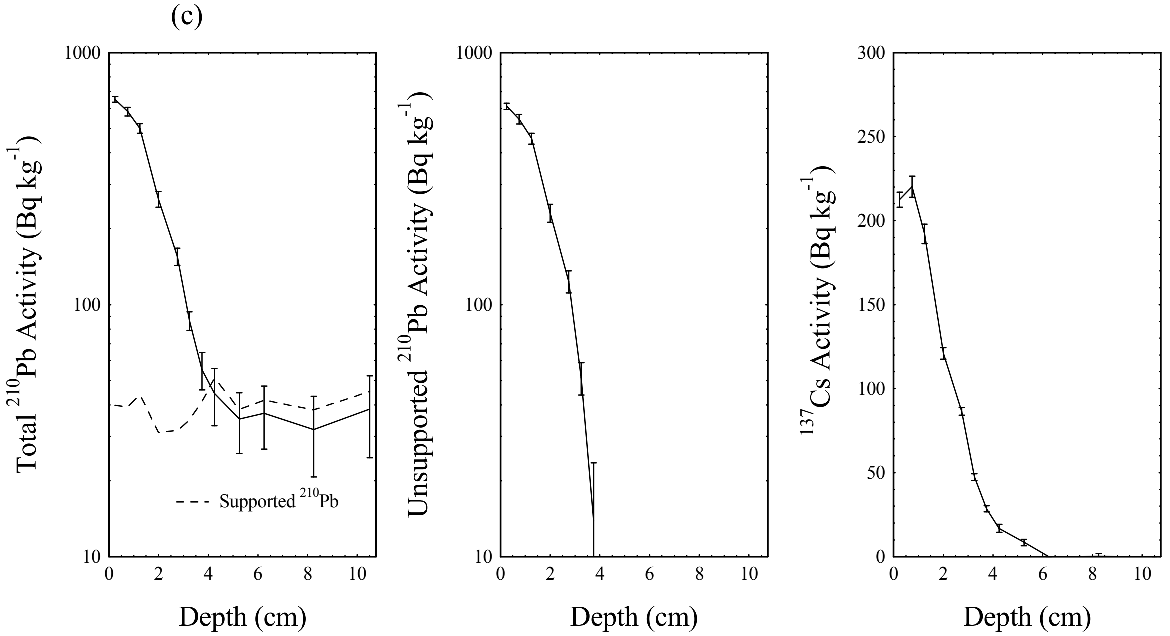

Figure S7: Fallout radionuclides in Wonder Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

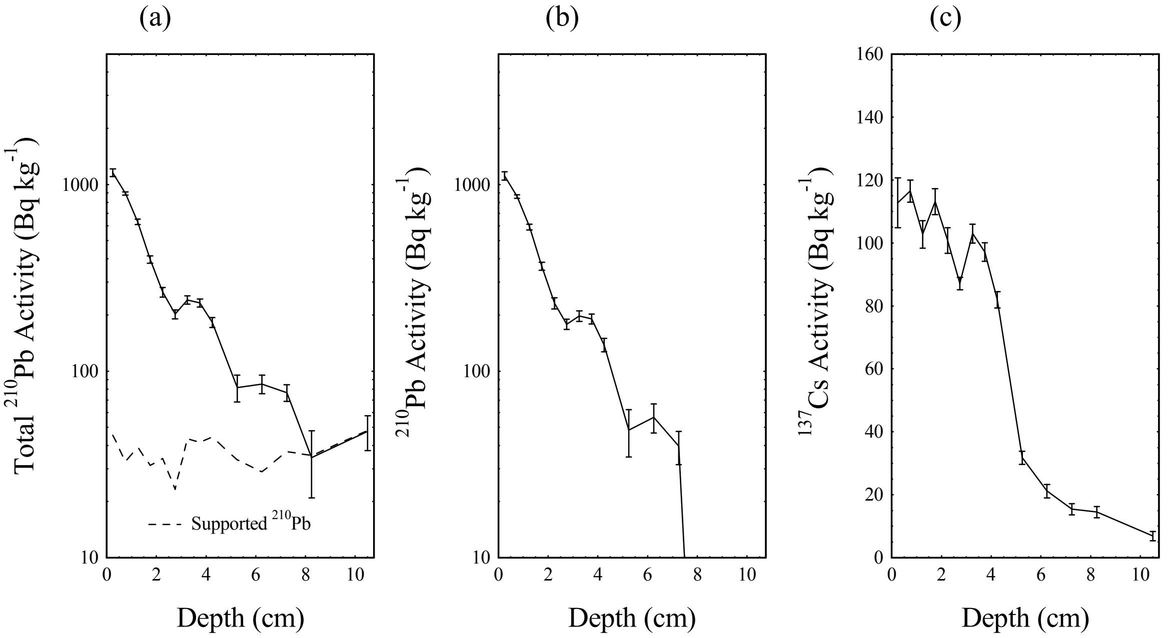

Figure S8: Fallout radionuclides in McLeod Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

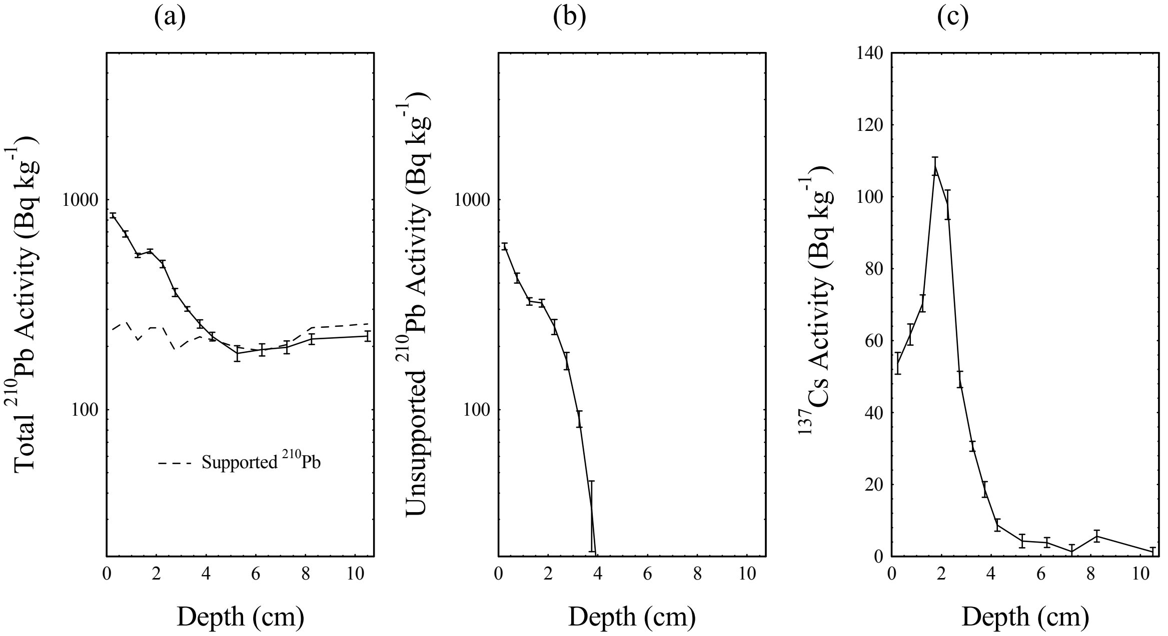

Figure S9: Fallout radionuclides in Matcharak Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

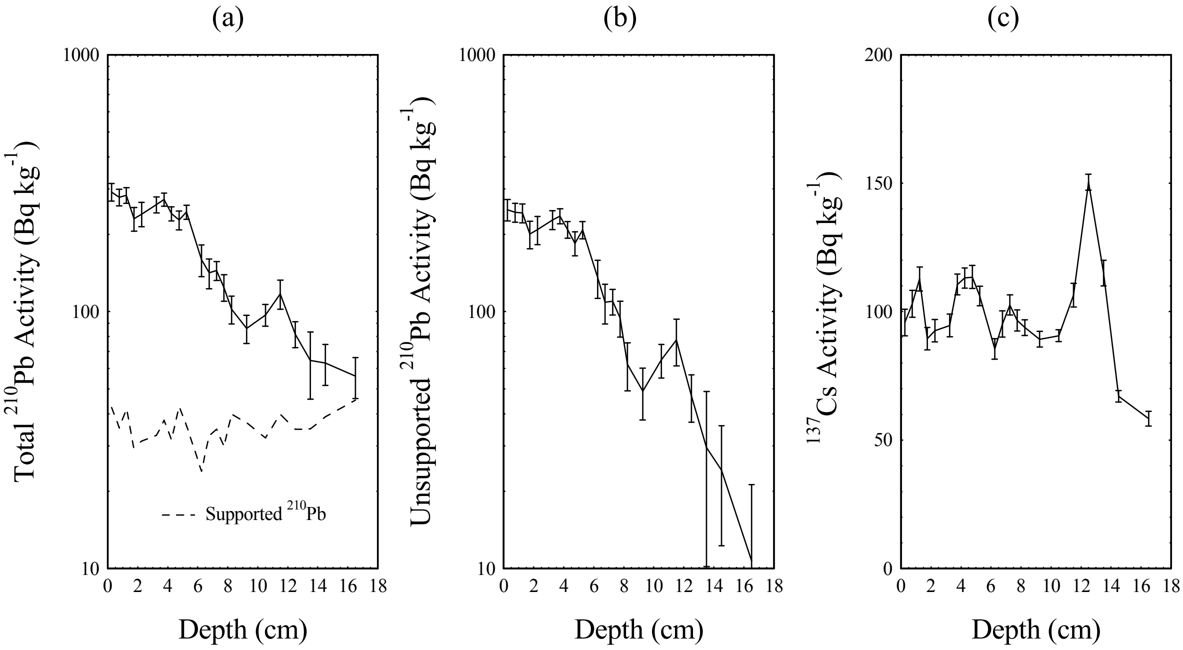

Figure S10: Fallout radionuclides in PJ Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

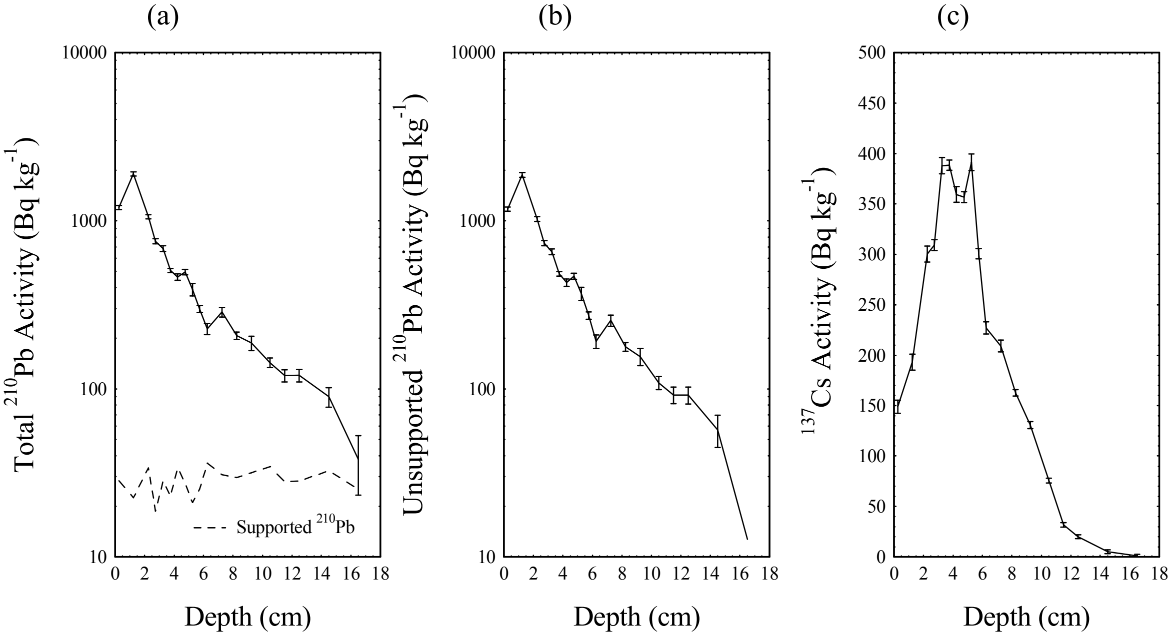

Figure S11: Fallout radionuclides in Hoh Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

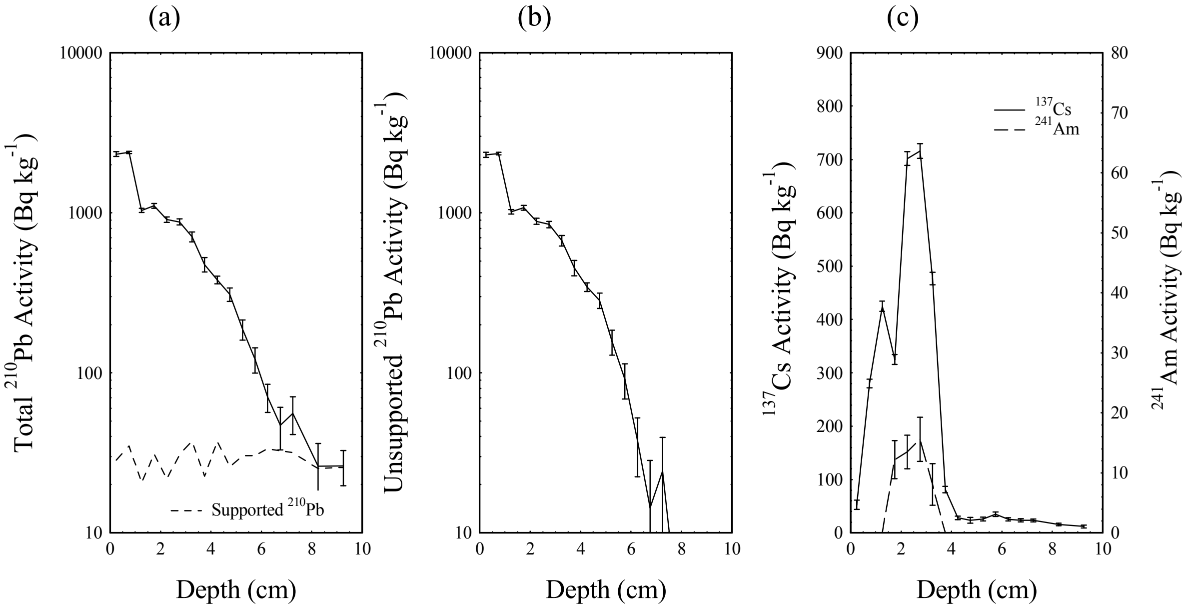

Figure S12: Fallout radionuclides in Golden Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

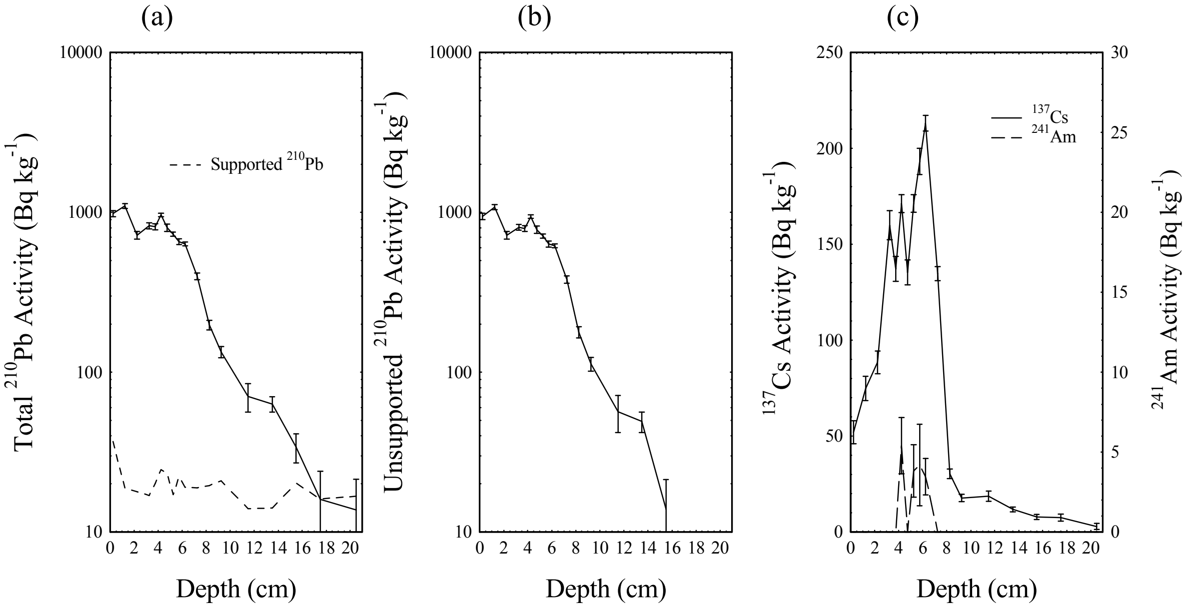

Figure S13: Fallout radionuclides in LP19 Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

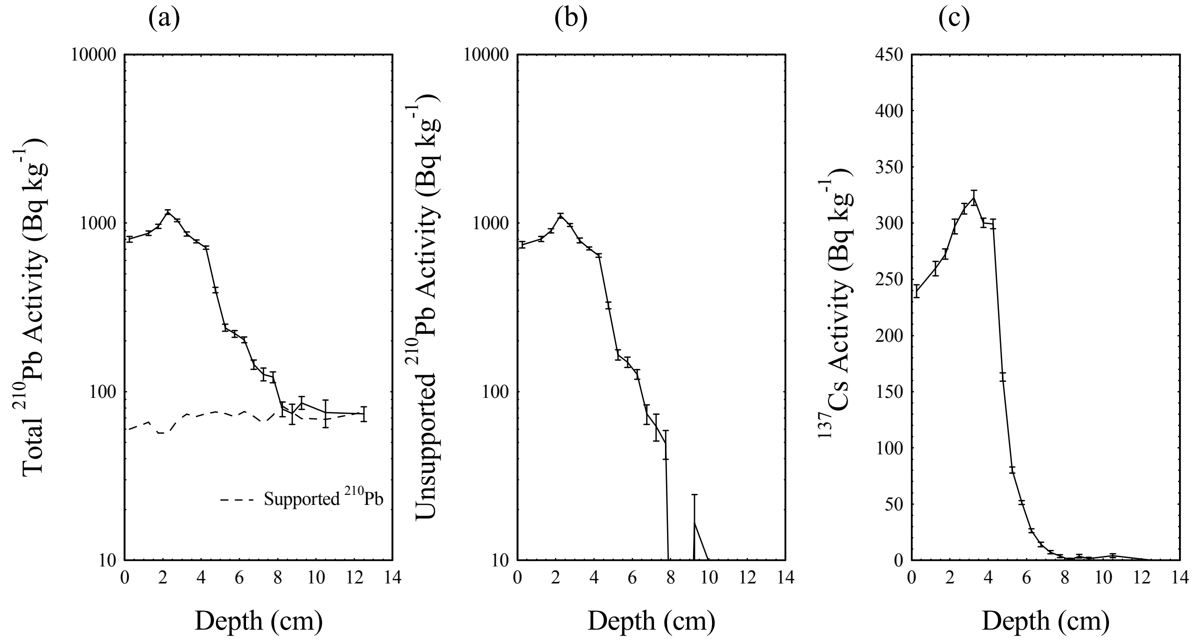

Figure S14: Fallout radionuclides in Oldman Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs concentrations versus depth.

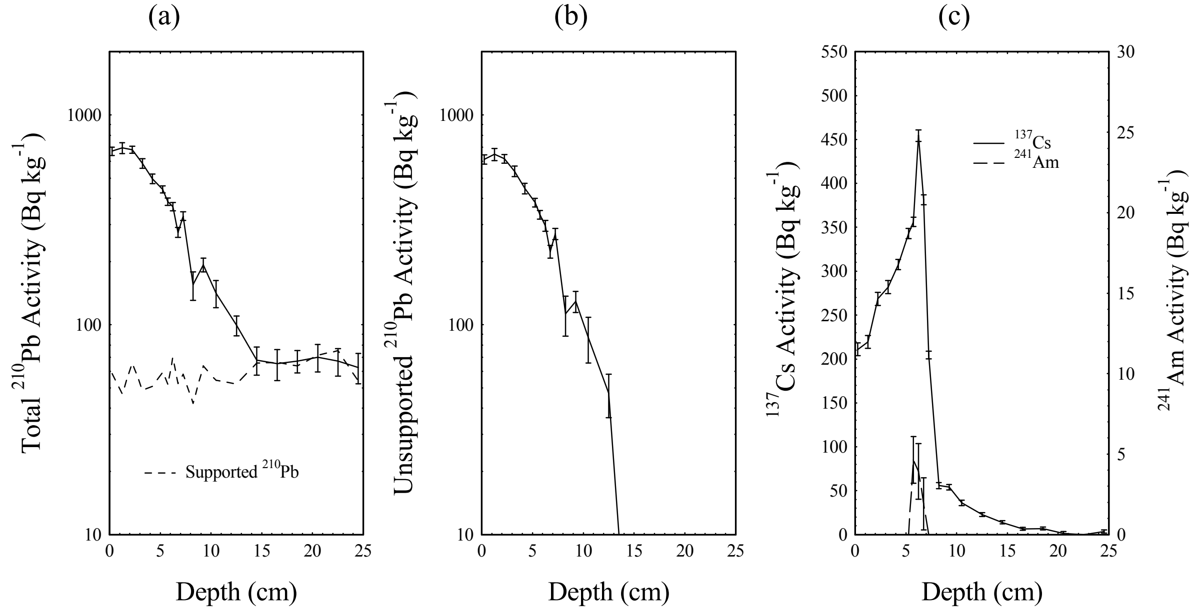

Figure S15: Fallout radionuclides in Snyder Lake core showing (a) total and supported ^{210}Pb, (b) unsupported ^{210}Pb, (c) ^{137}Cs and ^{241}Am concentrations versus depth.

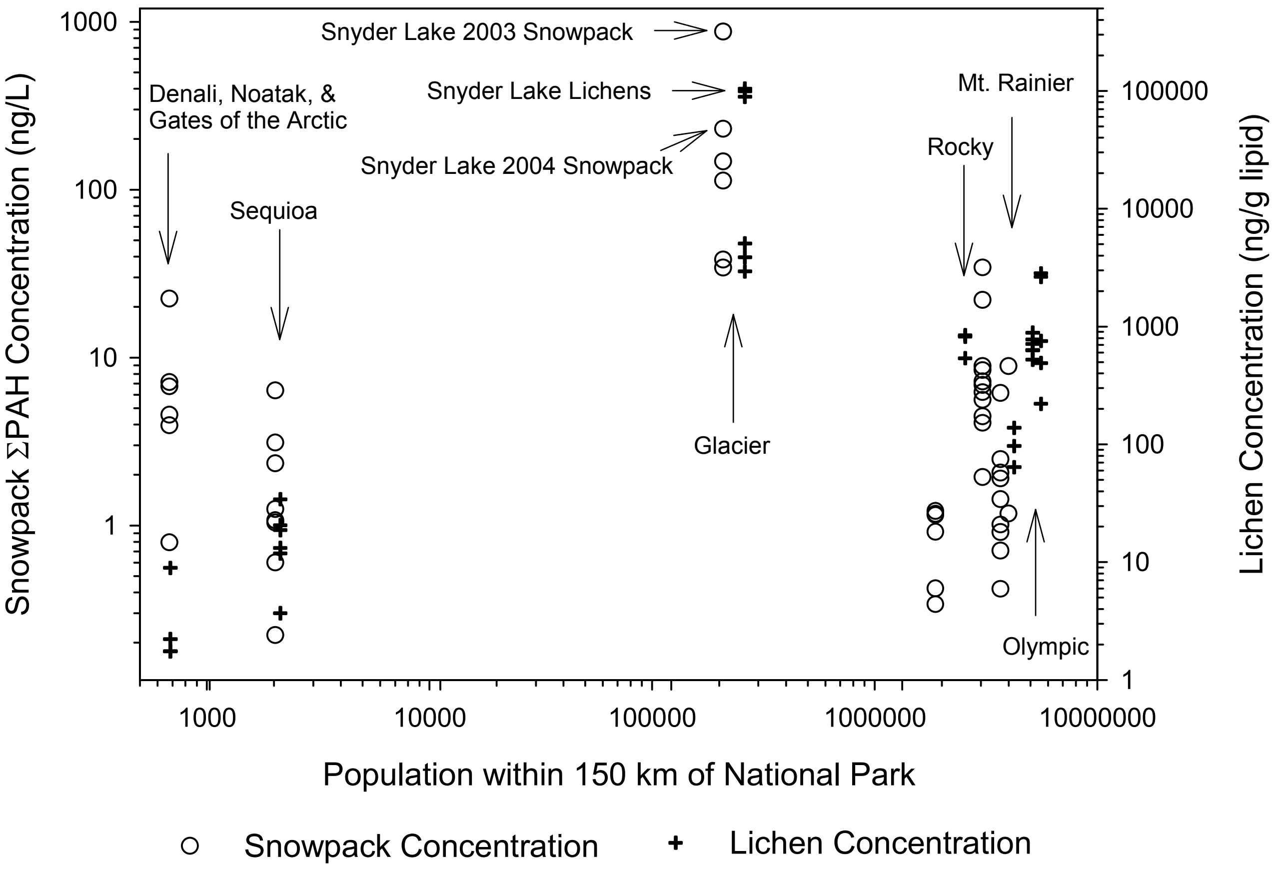

Figure S16: Snowpack and lichen ΣPAH concentrations versus population within 150 km of WACAP national parks.

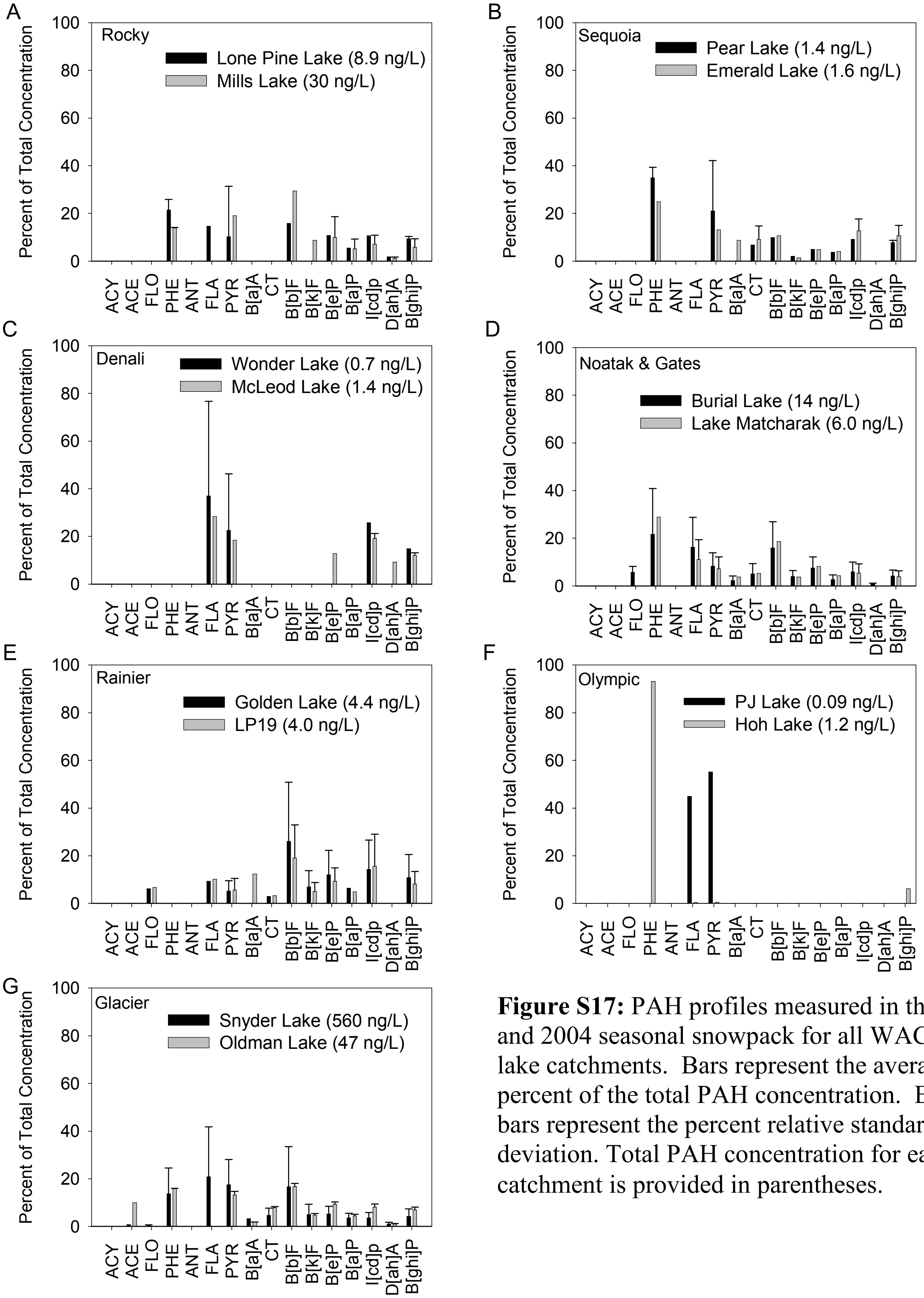

Figure S17: PAH profiles measured in the 2003 and 2004 seasonal snowpack for all WACAP lake catchments. Bars represent the average percent of the total PAH concentration. Error bars represent the percent relative standard deviation. Total PAH concentration for each lake catchment is provided in parentheses.

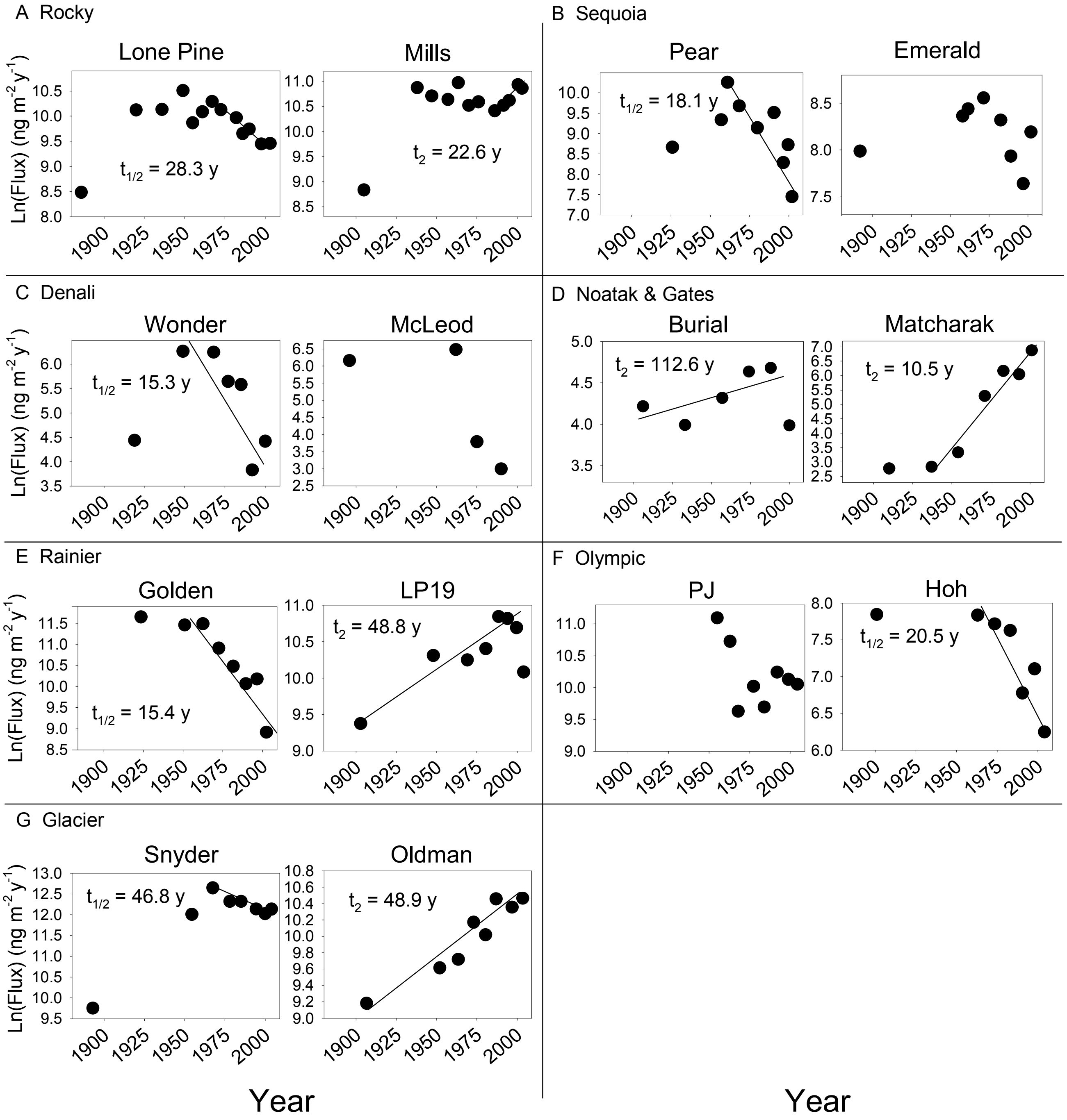

Figure S18. Natural log focus-corrected flux (ng m^{-2} y^{-1}) profiles of PAHs in WACAP sediment cores. Doubling times (t_2) and half-lives ($t_{1/2}$) are given where least squares regressions were statistically significant ($p<0.05$).

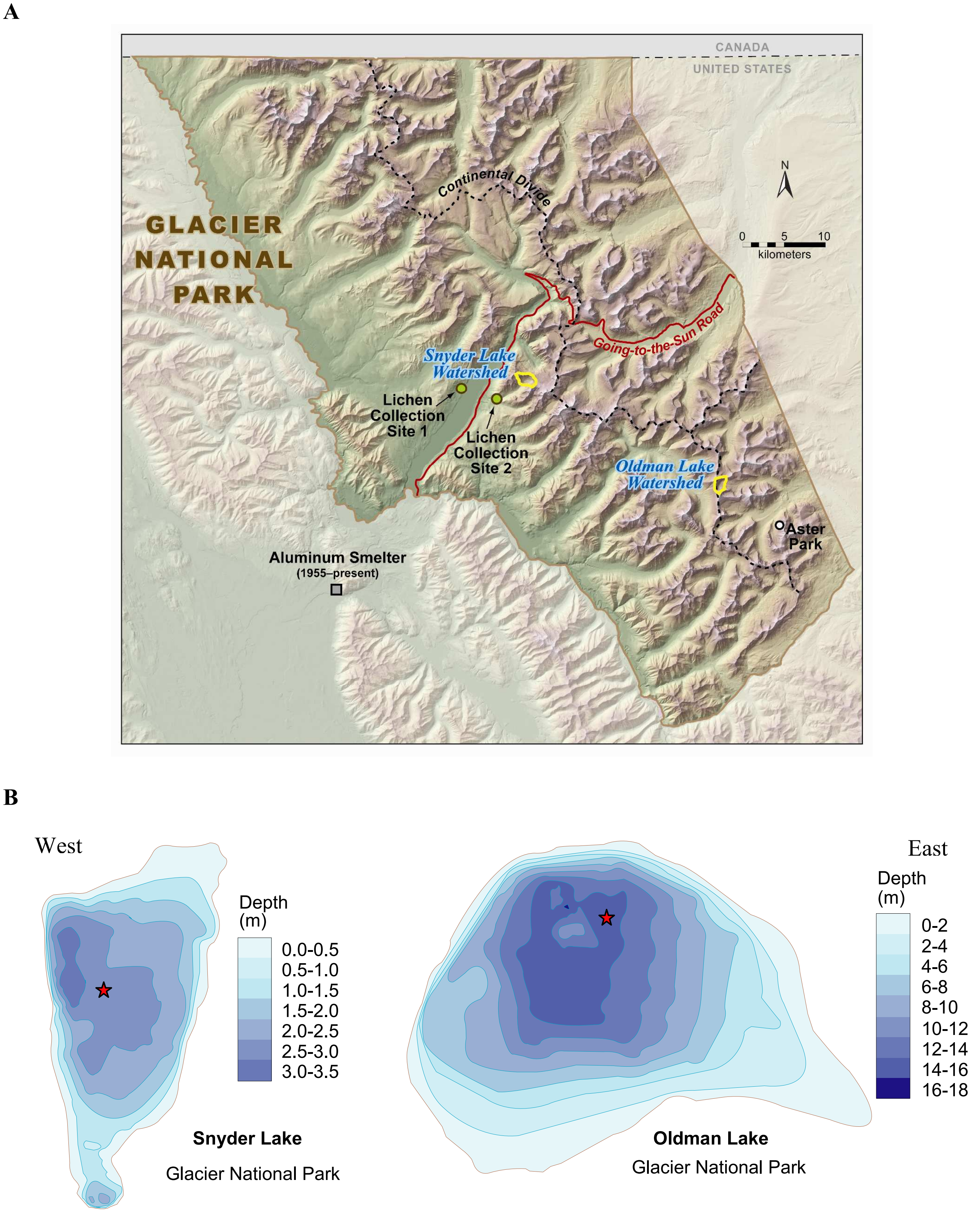

Figure S19. **A)** Map of Glacier National Park, MT. Brown line indicates park boundary and yellow lines indicate lake catchment. **B)** Lake bathymetry maps for Snyder Lake (west) and Oldman Lake (east). Stars indicate coring site location with in lake.

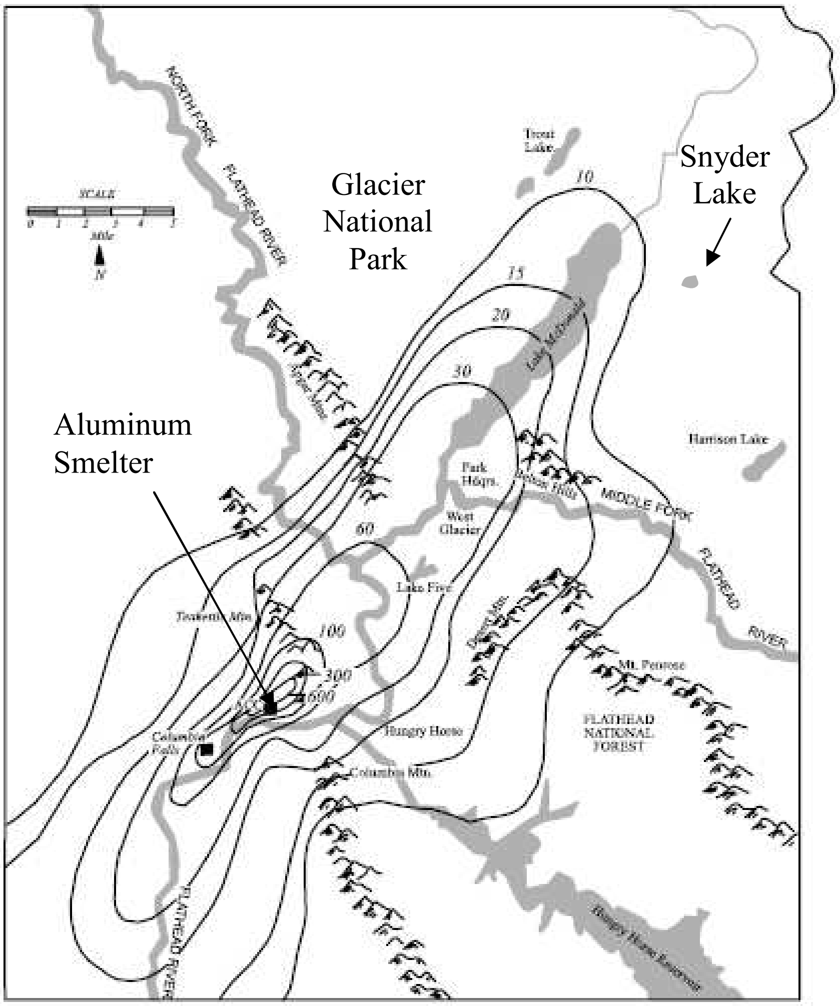

Figure S20: Isolines of fluoride (ppb) measured in foliage samples taken along transects from aluminum smelter in 1970 from reference (15).

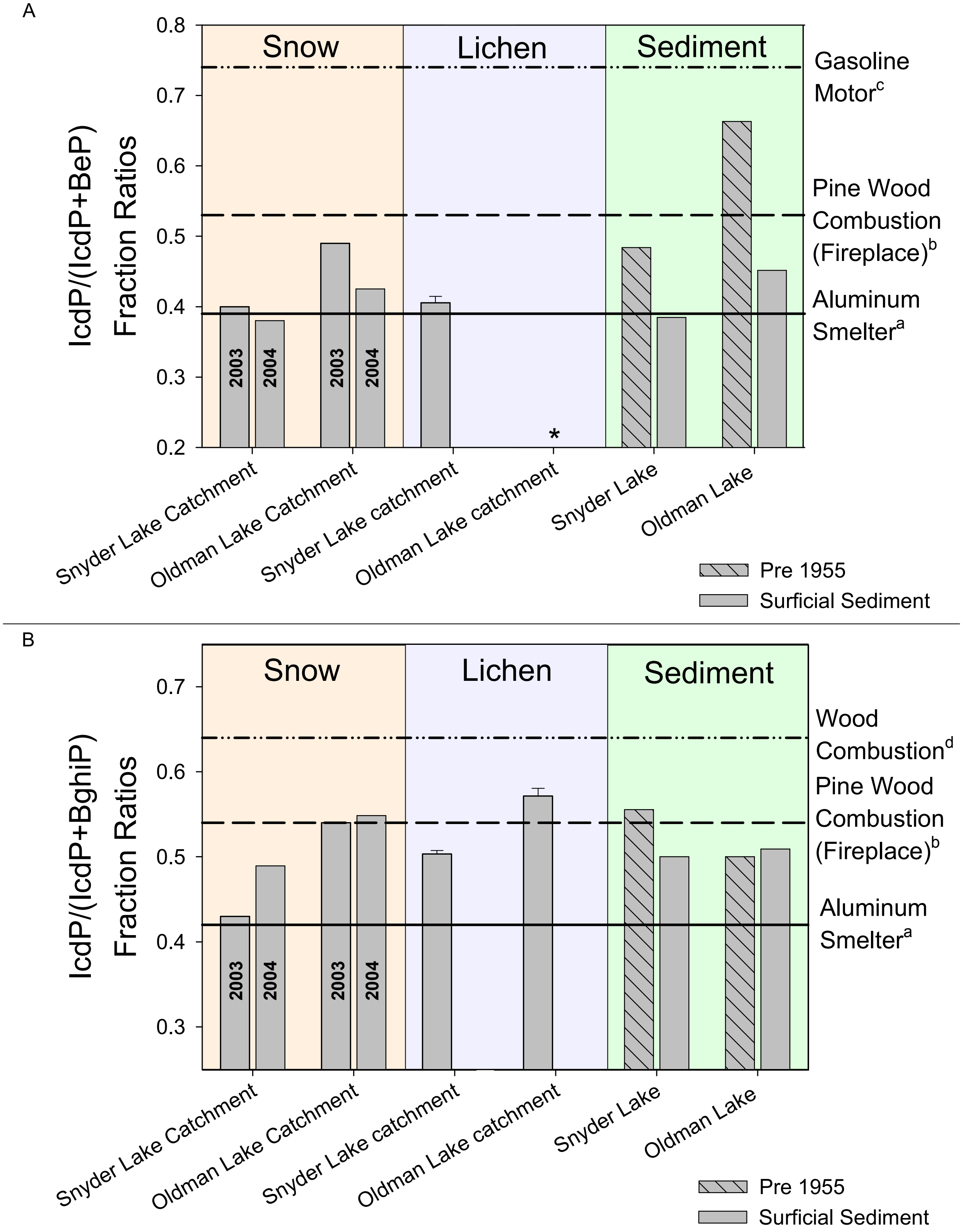

Figure S21: A) IcdP/(IcdP+BeP) and **B)** IcdP/(IcdP+BghiP) fraction ration (average ± standard deviation) calculated from seasonal snowpack, lichen, and pre 1955 and surficial sediment cores intervals for lake catchments in Glacier National Park. [a] reference (16), [b] reference (17), [c] reference (18), and [d] reference (19). * indicates concentration was below detection limits.

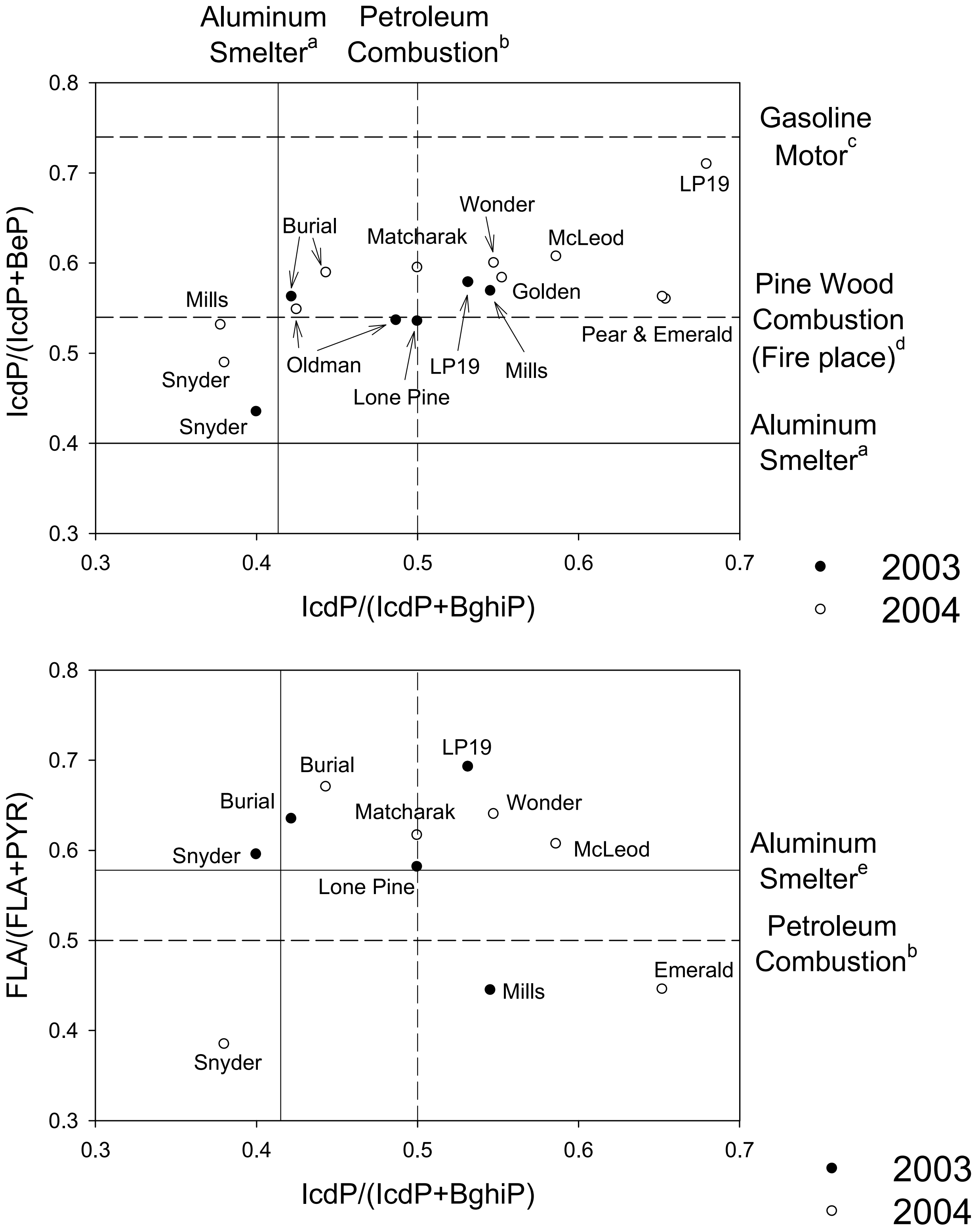

Figure S22: Ratios of Ind/(Ind+BeP), IcdP/(IcdPd+BghiP), and FLA/(FLA+PYR) calculated from all 2003 and 2004 season snowpack samples. Dashed lines (——) indicate the PAH ratio of a specific PAH source. Solid lines (——) indicate the PAH ratio from an aluminum smelter using Söderberg technology. [a] reference (16), [b] reference (19), [c] reference (17), [d] reference (18), and [e] reference (20).

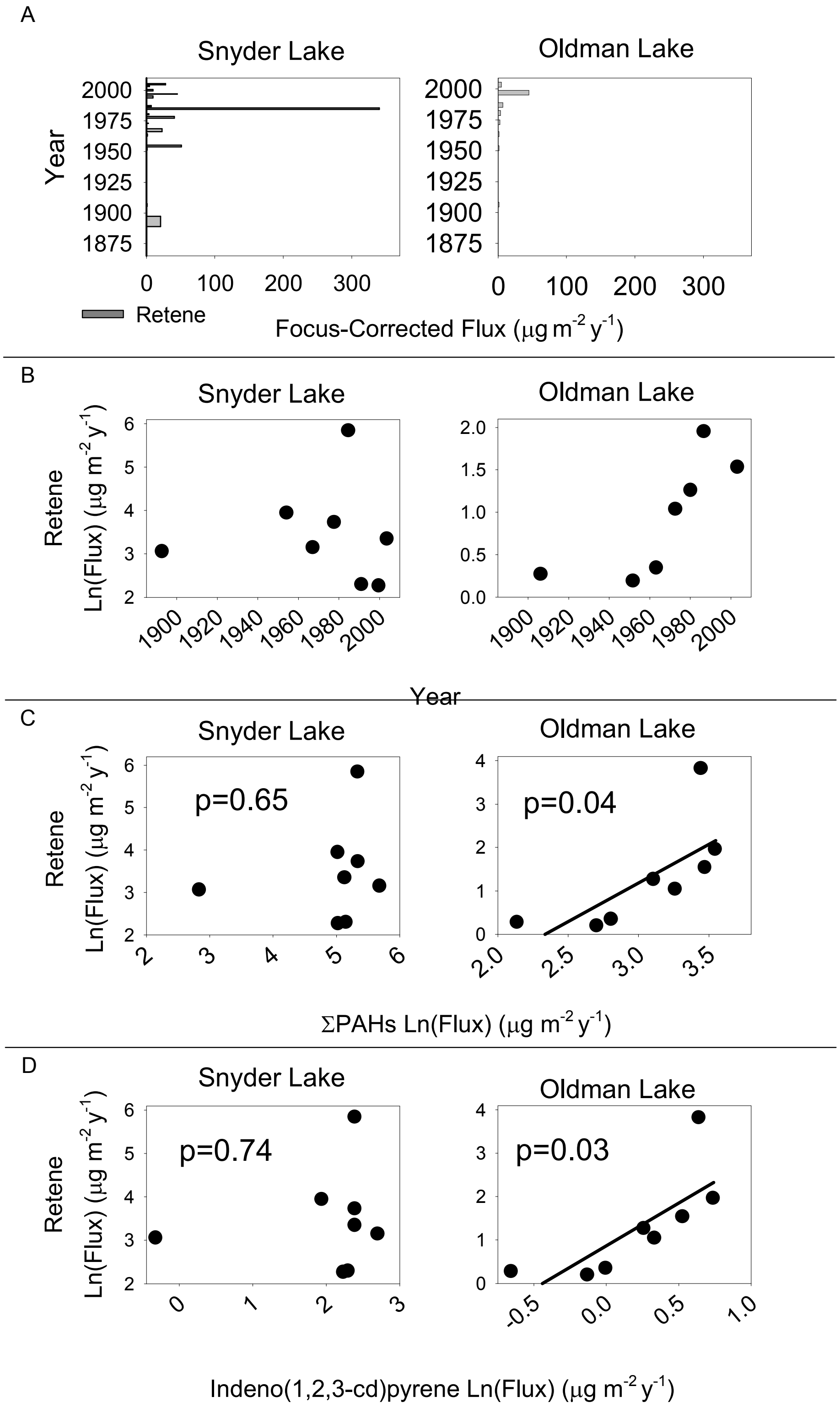

Figure S23: A) Focus-corrected flux (μg m^{-2} y^{-1}) profiles of retene in Oldman Lake (east) and Snyder Lake (west) sediment core. **B)** Natural log focus-corrected flux (μg m^{-2} y^{-1}) profiles of retene in Oldman Lake and Snyder Lake sediment core. **C)** Natural log focus-corrected flux (μg m^{-2} y^{-1}) profiles of retene versus ΣPAHs in Oldman Lake and Snyder Lake sediment core. **D)** Natural log focus-corrected flux (μg m^{-2} y^{-1}) profiles of retene versus indeno(1,2,3-cd)pyrene in Oldman Lake and Snyder Lake sediment core.

A

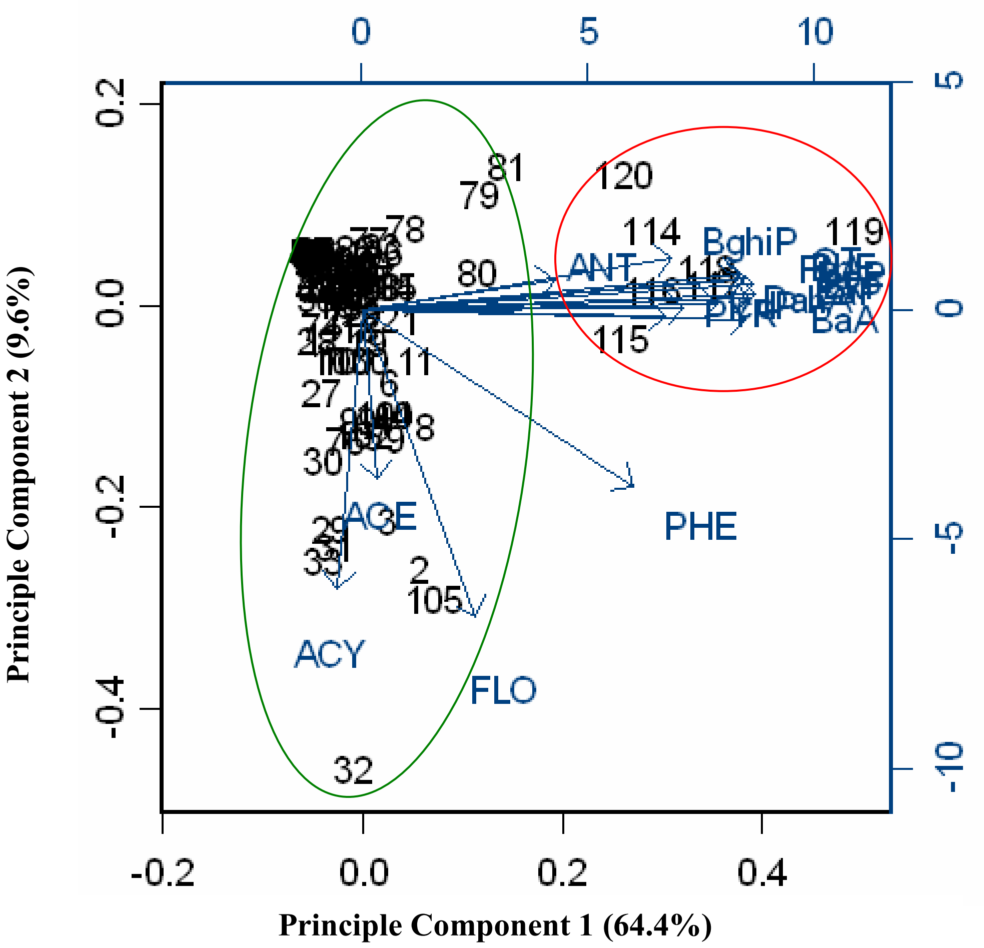

Figure S24: Individual PAH flux profiles in all WACAP lake sediment cores and intervals were analyzed by PCA using S-Plus 7.0 (Insightful, Seattle, WA). Results from the PCA showed that the total variance accounted for by PC1 and PC2. The circle on the right indicates a cluster that contains all Snyder Lake sediment intervals that dated after 1955. The oval on the left indicates a second cluster that contains all sediment intervals from the other WACAP catchments including a sediment interval from Snyder Lake dated at 1893. The arrows indicate the direction and magnitude of the individual PAH loadings.

Table S1: Physical and chemical limnological characteristics of each lake sites. * indicates the data was obtained from the parameter-elevation regressions on independent slopes model (PRISM) (average annual total precipitation from 1971-2000, 800×800 m). Sampling site's physical characteristics were determined from USGS 15' topographic quadrangles, mapping datum WGS84, and on-site measurements.

Sequoia National Park	Pear Lake	Emerald Lake	Pear/Emerald
Catchment Characteristics			
Latitude (dd)	36.6	36.58	1.00
Longitude (dd)	118.67	118.67	1.00
Elevation (masl)	2904	2800	1.04
Lake Volume (m^3)	578000	160000	3.61
Lake Surface Area (m^2)	73294	25342	2.89
Catchment Area (m^2)	1555595	1149318	1.35
Hydraulic Residence Time (d)	96.0	34.5	2.78
*Average Annual Precipitation (cm)	74.9	86.1	0.87
*Average Annual Max Temp (°C)	10.9	11.6	0.94
Focusing Factor	2.22	3.73	0.60
Limnological Characteristics (2003)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	0.82	0.94	0.87
Total Nitrogen ($mg \cdot L^{-1}$)	0.111	0.168	0.66
Total Phosphorus ($\mu g \cdot L^{-1}$)	0.59	1.47	0.40
Chlorophyll a ($\mu g \cdot L^{-1}$)	0.64	0.62	1.03
Turbidity (NTU)	0.232	0.259	0.90
Specific Conductivity ($\mu S \cdot cm^{-1}$)	4.02	5.42	0.74
pH	6.10	6.22	0.98

Rocky Mountain National Park	Lone Pine Lake	Mills Lake	Mills/Lone Pine
Catchment Characteristics			
Latitude (dd)	40.22	40.29	1.00
Longitude (dd)	105.73	105.64	1.00
Elevation (masl)	3024	3030	1.00
Lake Volume (m^3)	128325	78251	0.61
Lake Surface Area (m^2)	49134.9	61148	1.24
Catchment Area (m^2)	21144492	15093297	0.71
Hydraulic Residence Time (d)	4.3	3.3	0.77
*Average Annual Precipitation (cm)	97.6	107.1	1.10
*Average Annual Max Temp (°C)	7.7	7.4	0.96
Focusing Factor	1.87	1.48	0.79
Limnological Characteristics (2003)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	1.73	1.55	0.90
Total Nitrogen ($mg \cdot L^{-1}$)	0.17	0.38	2.24
Total Phosphorus ($\mu g \cdot L^{-1}$)	2.7	2.8	1.04
Chlorophyll a ($\mu g \cdot L^{-1}$)	2.0	2.1	1.05
Turbidity (NTU)	0.3	0.6	2.00
Specific Conductivity ($\mu S \cdot cm^{-1}$)	14.0	11.9	0.85
pH	6.67	6.05	0.91

Mt. Rainier National Park	Golden Lake	LP19	Golden/LP19
Catchment Characteristics			
Latitude (dd)	46.89	46.82	1.00
Longitude (dd)	121.90	121.89	1.00
Elevation (masl)	1372	1372	1.00
Lake Volume (m^3)	689578	99879	6.90
Lake Surface Area (m^2)	66104	18441	3.58
Catchment Area (m^2)	3914345	1591387	2.46
Hydraulic Residence Time (d)	24.8	9.1	2.73
*Average Annual Precipitation (cm)	228.4	231.2	0.99
*Average Annual Max Temp (°C)	8.9	8.2	1.09
Focusing Factor	1.00	1.50	0.67
Limnological Characteristics (2005)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	1.88	1.37	1.37
Total Nitrogen ($mg \cdot L^{-1}$)	0.069	0.074	0.93
Total Phosphorus ($\mu g \cdot L^{-1}$)	0.6	0.92	0.65
Chlorophyll a ($\mu g \cdot L^{-1}$)	0.35	0.6	0.58
Turbidity (NTU)	0.52	0.31	1.65
Specific Conductivity ($\mu S \cdot cm^{-1}$)	10.08	10.72	0.94
pH	6.47	6.63	0.98

Olympic National Park	Hoh Lake	PJ Lake	Hoh/PJ
Catchment Characteristics			
Latitude (dd)	47.90	47.95	1.00
Longitude (dd)	123.79	123.42	1.00
Elevation (masl)	1384	1433	0.97
Lake Volume (m^3)	396198	19099	20.74
Lake Surface Area (m^2)	76595	7551	10.14
Catchment Area (m^2)	2318604	3828864	0.61
Hydraulic Residence Time (d)	40.5	1.1	36.82
*Average Annual Precipitation (cm)	451.6	225.4	2.00
*Average Annual Max Temp (°C)	10.5	8.8	1.19
Focusing Factor	3.10	0.78	3.97
Limnological Characteristics (2005)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	0.74	1.05	0.70
Total Nitrogen ($mg \cdot L^{-1}$)	0.058	0.091	0.64
Total Phosphorus ($\mu g \cdot L^{-1}$)	1.16	2.78	0.42
Chlorophyll a ($\mu g \cdot L^{-1}$)	0.83	1.77	0.47
Turbidity (NTU)	0.39	0.36	1.06
Specific Conductivity ($\mu S \cdot cm^{-1}$)	63.69	127.40	0.50
pH	7.52	8.14	0.92

Table S1 (Continued): Physical and Chemical Limnological Characteristics of Lake sites. * indicates the data was obtained from the parameter-elevation regressions on independent slopes model (average annual total precipitation from 1971-2000, 800×800 m). Sampling site's physical characteristics were determined from USGS 15' topographic quadrangles, mapping datum WGS84, and on-site measurements.

Glacier National Park	Snyder Lake	Oldman Lake	Sndyer/Oldman
Catchment Characteristics			
Latitude (dd)	48.62	48.5	1.00
Longitude (dd)	113.79	113.46	1.00
Elevation (masl)	1600	2026	0.79
Lake Volume (m^3)	38298	1266063	0.03
Lake Surface Area (m^2)	25629	181755	0.14
Catchment Area (m^2)	4012230	2653727	1.51
Hydraulic Residence Time (d)	1.8	92.0	0.02
*Average Annual Precipitation (cm)	155.9	82.5	1.89
*Average Annual Max Temp (°C)	8.6	9.0	0.96
Focusing Factor	1.37	4.55	0.30
Limnological Characteristics (2005)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	0.65	0.70	0.93
Total Nitrogen ($mg \cdot L^{-1}$)	0.095	0.065	1.46
Total Phosphorus ($\mu g \cdot L^{-1}$)	2.67	0.55	4.85
Chlorophyll a ($\mu g \cdot L^{-1}$)	4.73	0.77	6.14
Turbidity (NTU)	0.64	0.35	1.82
Specific Conductivity ($\mu S \cdot cm^{-1}$)	16.80	159.10	0.11
pH	6.42	8.24	0.78

Denali National Park	Wonder Lake	McLeod Lake	Wonder/Mcleod
Catchment Characteristics			
Latitude (dd)	63.48	63.38	1.00
Longitude (dd)	150.88	151.07	1.00
Elevation (masl)	610	609	1.00
Lake Volume (m^3)	77653853	1847704	42.03
Lake Surface Area (m^2)	2656472	358512	7.41
Catchment Area (m^2)	28965859	1722319	16.82
Hydraulic Residence Time (d)	2740	2170	1.26
*Average Annual Precipitation (cm)	66	70	0.94
*Average Annual Max Temp (°C)	3.1	3.2	0.97
Focusing Factor	3.49	2.60	1.34
Limnological Characteristics (2004)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	2.10	2.25	0.93
Total Nitrogen ($mg \cdot L^{-1}$)	0.105	0.131	0.80
Total Phosphorus ($\mu g \cdot L^{-1}$)	0.5	0.104	4.81
Chlorophyll a ($\mu g \cdot L^{-1}$)	0.49	0.61	0.80
Turbidity (NTU)	0.34	0.29	1.18
Specific Conductivity ($\mu S \cdot cm^{-1}$)	190.10	8.41	22.61
pH	8.18	7.24	1.13

Gates of the Arctic National Park and Noatak National Preserve	Matcharak Lake	Burial Lake	Matcharak/Burial
Catchment Characteristics			
Latitude (dd)	67.75	68.43	0.99
Longitude (dd)	156.21	159.18	0.98
Elevation (masl)	488	427	1.14
Lake Volume (m^3)	21889008	5297945	4.13
Lake Surface Area (m^2)	3006999	654630	4.59
Catchment Area (m^2)	20650752	1837879	11.24
Hydraulic Residence Time (d)	4050	88440	0.05
*Average Annual Precipitation (cm)	43	39	1.10
*Average Annual Max Temp (°C)	-3.6	-4.7	0.77
Focusing Factor	1.25	0.88	1.42
Limnological Characteristics (2004)			
Primary Productivity	Oligotrophic	Oligotrophic	
Dissolved Organic Carbon ($mg \cdot L^{-1}$)	4.71	3.32	1.42
Total Nitrogen ($mg \cdot L^{-1}$)	0.284	0.233	1.22
Total Phosphorus ($\mu g \cdot L^{-1}$)	1.09	9.06	0.12
Chlorophyll a ($\mu g \cdot L^{-1}$)	0.96	0.81	1.19
Turbidity (NTU)	0.35	0.32	1.11
Specific Conductivity ($\mu S \cdot cm^{-1}$)	248.10	35.08	7.07
pH	8.31	7.57	1.10

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