Supporting Information

for

Current and Historical Deposition of PBDEs, Pesticides, PCBs, and PAHs to Rocky Mountain National Park, USA

SASCHA USENKO,[†] DIXON H. LANDERS,[§] PETER G. APPLEBY,[⊥]

AND STACI L. SIMONICH [,†,‡]*

[†]Department of Chemistry, [‡]Department of Environmental and Molecular Toxicology, Oregon State University, Corvallis, Oregon 97331-7301
[§]United States Environmental Protection Agency-Western Ecology Division, Corvallis, OR 97330, USA
[⊥]Environmental Radioactivity Research Centre, University of Liverpool, Liverpool L69 3BX, UK

[†] Department of Chemistry

[‡] Department of Environmental and Molecular Toxicology

[§] United States Environmental Protection Agency-Western Ecology Division, Corvallis, OR 97330, USA

[⊥]Environmental Radioactivity Research Centre, University of Liverpool, Liverpool L69 3BX, UK

[*] Corresponding author phone: +541-737-9194: fax: +541-737-0497: e-mail: Staci.Simonich@orst.edu

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Chemicals.

All semi-volatile organic compound (SOC) standards were acquired from the EPA repository or purchased from Chem Services Inc. (West Chester, PA), Restek (Bellefonte, PA), Sigma-Aldrich Corp. (St. Louis, MO), or AccuStandard (New Haven, CT). Standard Reference Material (SRM) #1941b was acquired from the National Institute of standards and Technology (NIST) (Gaithersburg, MD). Solvents were Fisher Scientific (Fairlawn, NJ) Optima grade and the anhydrous sodium sulfate was Mallinckrodt Baker (Phillipsburg, NJ) pesticide grade. Isotopically labeled standards were purchased from CDN Isotopes (Pointe-Claire, Quebec, Canada) or Cambridge Isotope Labs (Andover, MA). The isotopically-labeled recovery surrogates were d_{10}-fluorene, d_{10}-phenanthrene, d_{10}-pyrene, d_{12}-triphenylene, d_{12}-benzo[a]pyrene, d_{12}-benzo[ghi]perylene, d_{14}-EPTC, d_5-atrazine, d_{10}-diazinon, d_7-malathion, d_{10}-parathion, d_8-p,p'-DDE, d_8-p,p'-DDT, d_6-methyl parathion, d_{13}-alachlor, d_{11}-acetochlor, $^{13}C_{12}$-PCB 101 (2,2',4,5,5'-pentachlorobiphenyl), $^{13}C_{12}$-PCB 180 (2,2', 3,4,4',5,5'-heptachlorobiphenyl), d_{10} -chlorpyrifos, $^{13}C_6$-HCB, d_6-γ-HCH, d_4-endosulfan I, d_4-endosulfan II , d_{14}-trifluralin, $^{13}C_{12}$-BDE 28 (2,4,4'-tribromodiphenyl ether), $^{13}C_{12}$-BDE 47 (2,2',4,4'-tetrabromodiphenyl ether), $^{13}C_{12}$-BDE 99 (2,2',4,4',5-tetrabromodiphenyl ether), $^{13}C_{12}$-BDE 100 (2,2',4,4',6-tetrabromodiphenyl ether), $^{13}C_{12}$-BDE 118 (2,3',4,4',5-tetrabromodiphenyl ether), $^{13}C_{12}$-BDE 138 (2,2',3,4,4',5'-tetrabromodiphenyl ether), $^{13}C_{12}$-BDE 153 (2,2',4,4',5,5'-tetrabromodiphenyl ether), and $^{13}C_{12}$-BDE 183 (2,2',3,4,4',5',6-tetrabromodiphenyl ether). The isotopically-labeled internal standards were d_{10}-acenaphthene, d_{12}-benzo[k]fluoranthene, d_{10}-fluoranthene, and $^{13}C_{12}$-PCB 138 (2,2',3,4,4',5'-hexachlorobiphenyl). All standards were stored at 4°C and remade, as needed, to insure stability. The organophosphate standards were stored separately from other chemical classes in ethyl acetate (EA) to minimize degradation.

Sample Collection.

Sediment cores were collected from the deepest point in the lake during the ice-free summer season using a pontoon raft equipped with an Uwitec gravity corer with an 86 mm internal diameter. Vertically-planed sediment cores (25-50 cm in depth), with an intact surface layer, were sectioned in the field with a clean stainless steel blade. The first 10 cm of each core was sectioned into 0.5 cm increments (12-18 g wet wt increment) and the remainder of the core was sectioned into 1.0 cm increments (30-40 g wet wt increment). Each sediment slice was stored in a 250 mL solvent rinsed glass jar. A pre-baked piece of aluminum foil was placed over the mouth of the jar to separate the sample from the cap. Sediment samples were shipped overnight in ~50-L coolers with cold packs to the laboratory where they were stored at 4 ºC for physical and elemental analysis.

Physical and Elemental Analysis.

1   Aliquots of the freeze-dried sediment were used for the analyses of total carbon (TC), total

2 organic carbon (TOC), ^{210}Pb, ^{137}Cs, [226]Ra, and [241]Am. Destructive carbon analysis (TC and TOC) was

3 preformed using flash combustion and a Carlo Erba 1108A CN analyzer. Carbonate was removed,

4 using HCl fumes for 18hrs, prior to combustion for TOC analysis. Freeze-dried sediment sample

5 aliquots were analyzed for ^{210}Pb, [226]Ra, ^{137}Cs, and [241]Am by direct gamma assay at the Liverpool

6 University Environmental Radioactivity Laboratory, using an Ortec HPGe GWL series well-type coaxial

7 low background intrinsic germanium detector (*1*). Radiodating of ^{210}Pb and [226]Ra was determined by the

8 gamma emissions at 46.5 keV and 295 keV, respectively. [137]Cs and [241]Am were measured by their

9 emissions at 662 keV and 59.5 keV, respectively.

10 **SOC Extraction and Isolation.**

11   Samples were allowed to thaw in sealed grass jars in the dark for ~15 minutes and were ground with

12 sodium sulfate (1:15 ratio), that had been baked at 400 ºC for 3 hrs and cooled, to remove excess water.

13 The sample was packed into three or four 66-mL Accelerated Solvent Extractor (ASE) cells (Dionex,

14 Sunnyvale, CA) with each cell containing ~80 g of the sediment/sodium sulfate mixture. Once in the

15 ASE cells, 15 µL of 10 ng/µL isotopically labeled surrogate-ethyl acetate (EA) solution was distributed

16 equally among the tops of the cells containing sample. The sample was extracted using an ASE 300

17 and dichloromethane (DCM) (100 °C, 1500 psi, 3 cycles of 3 minutes, 150% flush volume).

18   The sediment extract was concentrated to 0.5 mL in the TurboVap II (Zymark, Hopkinton,

19 MA) with nitrogen and solvent exchanged to hexane. Polar matrix interferences were removed using a

20 20-g silica solid phase extraction (SPE) cartridge (Varian). Analytes were eluted from the silica SPE

21 using 100 mL DCM:EA. The eluate was then concentrated and solvent exchanged to DCM. Elemental

22 sulfur and the high molecular weight interferences were removed using a Waters Gel Permeation

23 Chromatography Cleanup System (Milford, MA) as previously described (*2*). The target fraction was

24 concentrated to 0.3 mL under a gentle stream of nitrogen and spiked with 15 µL of 10 ng/µL isotopically

25 labeled internal standard-EA solution just prior to gas chromatographic mass spectrometry (GC/MS)

26 injection.

27   Sodium sulfate was used as the laboratory blank and was carried through the entire analytical

28 method (extraction, cleanup, and concentrating), starting at the grinding step, the laboratory blank was

29 spiked with the same quantity of isotopically labeled surrogate and internal standards as mention above.

30 The minimum SOC concentrations in samples to the laboratory blank was of 3:1.

31 **Quantification and Validation.**

32   Quantification was preformed with a surrogate standard calibration curve (4-12 points) and SOC

33 concentrations were calculated relative to surrogates. Sample-specific estimated method detection limits

34 (EDLs), calculated using EPA-method 8280A (*3*), were determined for all target SOCs. A

1   representative sediment sample (0.5-1.0 cm) (not spiked with target analytes) from Waldo Lake was

2   used to calculate EDLs, which ranged from 0.1-200 ng/g dry wt, depending on the SOC (Table S1). The

3   analytical method was validated for efficiency with triplicate spike and recovery experiments using

4   Waldo Lake sediment. These recoveries were corrected for background SOC concentrations in sediment

5   and represent the efficiency of the entire analytical method because the target SOCs were spiked prior to

6   extraction and the isotopically-labeled surrogates were spiked just prior to analysis. Background SOC

7   concentrations measured in Waldo Lake sediment ranges from 0.0 to 5.0% of the spike concentration,

8   expect for phenanthrene which was approximately 40%. The accuracy and precision of the analytical

9   method were determined using NIST SRM 1941b.

10   **PRISM.**

11   The Parameter-elevation Regressions on Independent Slopes Model (PRISM) was designed to

12   estimate the orographic climate parameters (*4*). PRISM provides 2×2 km resolution monthly annual

13   precipitation estimates from 1895 to February 2007. PRISM also provides the average precipitation for

14   a month from 1971-2000 with 800×800 m resolution.

15   **Analysis and Quality Control.**

16   The sediment extracts were analyzed for target SOCs by GC/MS, using both electron impact (EI)

17   ionization and electron capture negative ionization (ECNI) with selective ion monitoring as described in

18   detail by Usenko et al (*2*) and Ackerman et al (*5*). Target SOCs were quantified using the mode of

19   ionization that resulted in the lowest instrumental detection limits (IDLs) (*2*). IDLs ranged from 0.063-

20   6.7 pg/µL for GC/EI-MS and from 0.006-1.1 pg/µL for GC/ECNI-MS.   Target SOCs were identified

21   using the following criteria; GC retention time (± 0.05 min of standard), quantification and confirmation

22   ion ratios ($\pm 20\%$ of standard), and a signal-to-noise ratio of 10:1. SOC concentrations in sediment were

23   surrogate recovery (concentration calculated relative to surrogate) and laboratory blank corrected. All

24   WACAP quality assurance objectives were meet (*6*).

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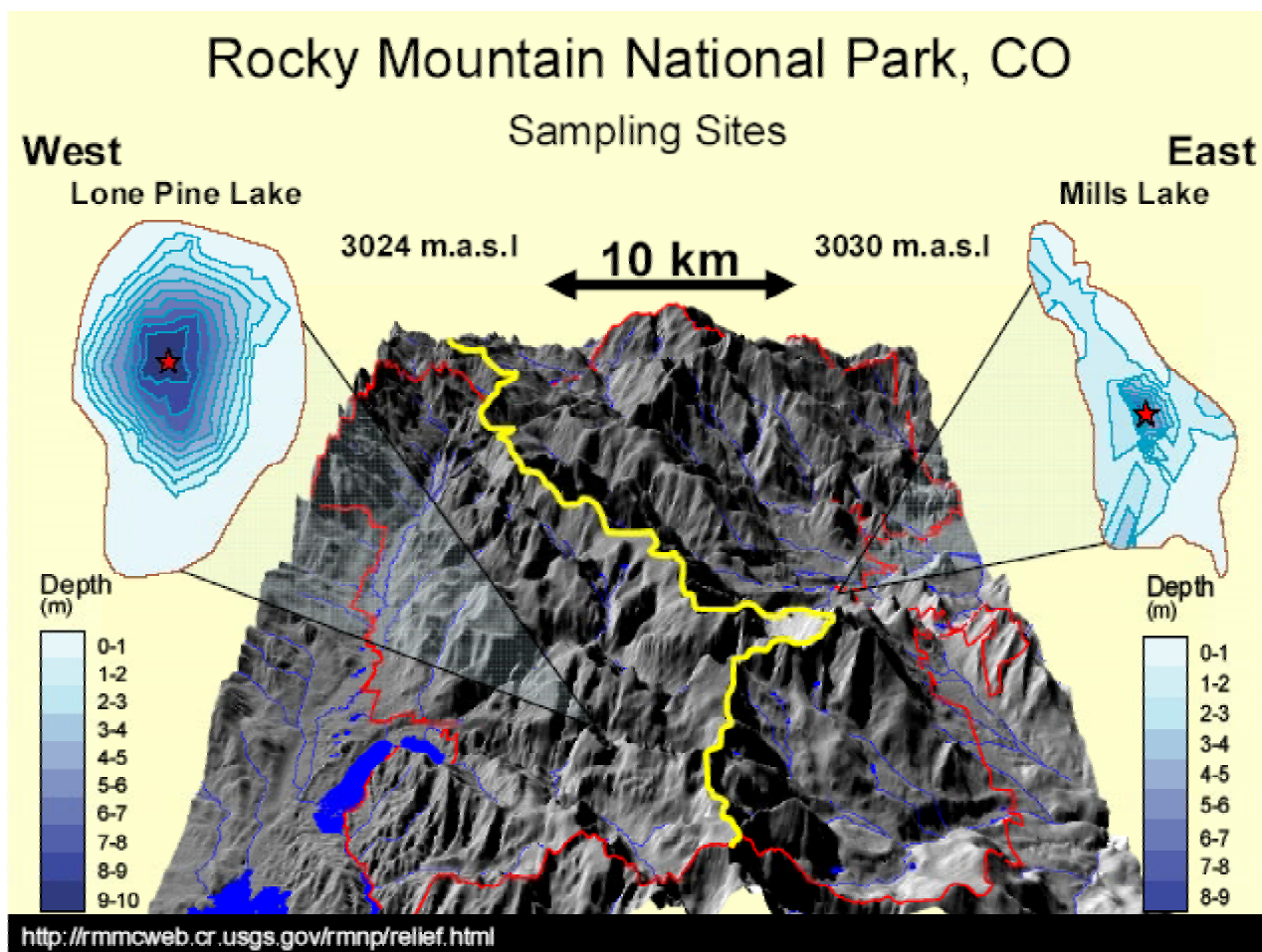
Figure S1. Shaded relief map of Rocky Mountain National Park with lake bathymetry maps for Lone Pine Lake and Mills Lake. Red line represents the park boundary and the yellow line represents the Continental Divide ~ 4150 m.a.s.l. between the two catchments. Stars indicate coring site location with in lake.
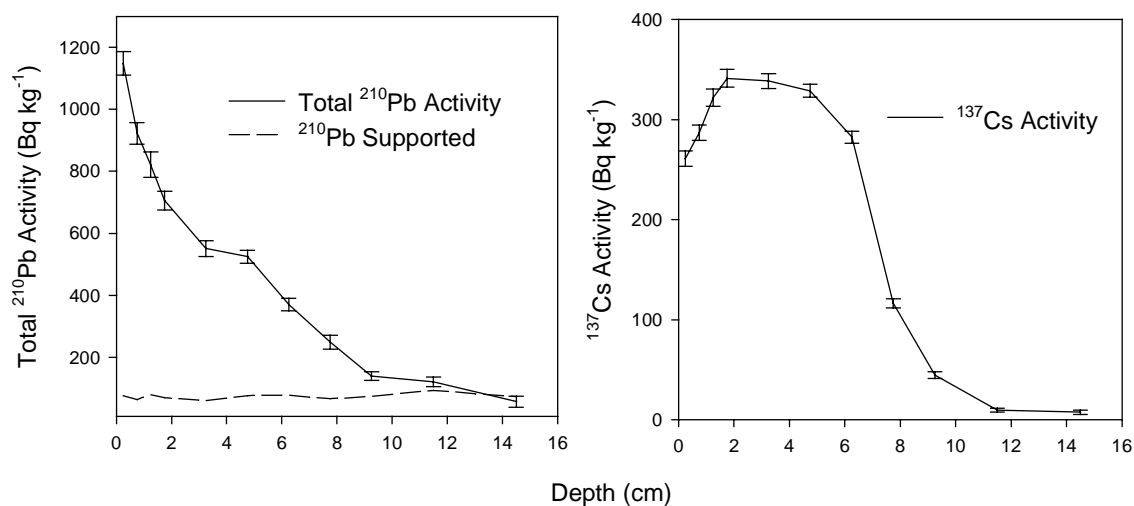
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2 **Figure S2. Fallout radionuclides in Lone Pine Lake (a) total and supported ^{210}Pb activity, (b)**
3 **^{137}Cs concentrations versus depth.**
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6 **Figure S3. Fallout radionuclides in Mills Lake core (a) total and supported ^{210}Pb activity, (b) ^{137}Cs**
7 **concentrations and ^{137}Cs/^{210}Pb activity ratios versus depth.**
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Table S1. Recovery of target SOCs in sediment over the entire analytical method.

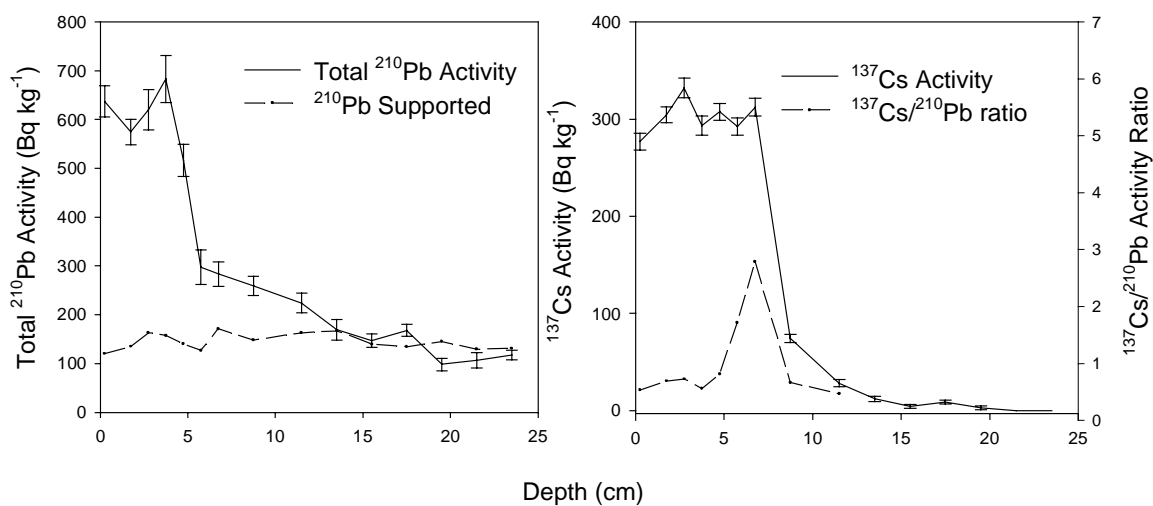|	Waldo Lake[a]		EDL[b]	SRM1941b				Waldo Lake[a]		EDL[b]	SRM1941b		
	Avg. % Rec	% RSD	ng/g dw	ng/g dw	PD[c]	% RSD		Avg. %Rec	%RSD	ng/g dw	ng/g dw	PD[c]	% RSD
Amide Pesticides													
Propachlor	49.8	3.3	7.8				Acetochlor	46.1	9.3	9.3			
Alachlor	53.1	12.2	13.3				Metolachlor	58.6	12.2	14.2			
Organochlorine Pesticides and Metabolites													
HCH, gamma[d]	29.6	9.4	118				Chlordane, cis	45.7	14.7	18.4	0.7	10.0	29.5
HCH, alpha[d]	50.8	9.0	133				p,p'-DDD[f]	60.3	10.2	16.5	5.1	0.0	40.0
HCH, beta[d]	36.2	9.1	176				Nonachlor, trans	46.8	17.2	3.7	0.1	53.2	32.8
HCH, delta[d]	51.8	9.4	59.5				o,p'-DDD[f]	55.6	10.8	4.3	1.1		
Methoxychlor	67.4	14.8	18.6	1.0			Chlordane, trans	46.8	15.2	2.0	0.4	5.8	28.2
Heptachlor epoxide	46.8	13.8	89.4				Nonachlor, cis	53.6	13.0	1.5	0.1	55.4	24.2
Endrin aldehyde	51.8	7.9	19.6				Aldrin	29.0	12.5	83.2			
Endrin	70.4	11.5	205				o,p'-DDT[g]	44.4	12.0	23.6			
Heptachlor	32.5	12.4	111.9				p,p'-DDE[e]	55.9	12.7	3.4	3.1	0.0	15.8
o,p'-DDE[e]	57.7	11.2	11.3				Mirex	56.3	6.3	41.4			
Chlordane, oxy	43.7	14.8	12.2				p,p'-DDT[g]	54.7	13.5	37.9			
Dieldrin	74.0	13.1	115	0.32									
Organochlorine Sulfide Pesticides and Metabolites													
Endosulfan sulfate	61.4	9.6	4.4				Endosulfan II	58.5	10.3	9.0			
Endosulfan I	50.2	13.2	8.1										
Phosphorothioate Pesticides													
Methyl parathion	49.9	5.1	33.0				Parathion	54.0	6.5	15.7			
Malathion	48.3	7.9	65.8				Ethion	60.0	10.4	10.8			
Diazinon	47.9	5.4	5.1				Chlorpyrifos	45.3	9.7	1.2			
Triazine Herbicides and Metabolites													
Simazine	63.2	3.4	58.3				Atrazine	57.6	6.3	9.5			
Cyanazine	136	19.3	171										
Miscellaneous Pesticides													
Metribuzin	43.6	20.6	30.0				Dacthal	55.5	11.5	6.4			
Etridiazole	21.6	13.9	29.1				Trifluralin	32.9	10.8	1.7			
Triallate	41.1	8.6	24.2				Hexachlorobenzene	33.5	8.0	1.0	7.6	24.1	22.8
Polycyclic Aromatic Hydrocarbons													
Acenaphthylene (ACY)	20.9	14.7	13.3	139			Retene	59.2	6.8	9.66			
Acenaphthene (ACE)	33.5	13.5	11.2	51.6			Benzo[k]fluoranthene (BkF)	68.5	10.0	3.3	206	0.6	21.6
Fluorene (FLO)	25.5	12.7	7.2	59.2	12.7	22.3	Benzo[a]pyrene (BaP)	46.7	9.3	2.1	221	33.6	23.7
Anthracene (ANT)	34.8	8.0	24.6	163	1.5	13.6	Benzo[b]fluoranthene (BbF)	64.8	9.5	4.0	468	0.0	18.7
Phenanthrene (PHE)	26.0	20.0	13.0	383	0.0	18.6	Indeno[1,2,3-cd]pyrene (IcdP)	60.1	9.5	29.0	240	12.9	17.3
Pyrene (PYR)	50.6	5.7	1.0	403	24.0	22.1	Dibenz[a,h]anthracene (DahA)	58.2	9.8	23.7	76.4	25.3	22.9
Fluoranthene (FLA)	50.5	5.1	1.1	443	24.3	20.8	Benzo[e]pyrene (BeP)	64.6	9.1	6.5	285	4.5	22.4
Chrysene/Triphenylene (CT)	59.9	9.2	0.8	171	48.1	22.3	Benzo[ghi]perylene (BghiP)	55.0	11.1	5.1	227	11.3	22.0
Benzo[a]anthracene (BaA)	64.5	10.2	11.4	250	17.8	17.2							
Polychlorinated Biphenyls													
PCB 101	70.7	14.2	129	4.1	13.2	29.9	PCB 118	74.2	11.6	10.2	3.3	17.2	34.5
PCB 138	74.9	11.7	9.7	4.3	12.3	30.4	PCB 187	76.1	13.1	3.9	2.0	0.0	22.1
PCB 153	73.2	11.8	3.5	4.0	21.6	20.1	PCB 183	76.5	13.1	3.7	0.7	23.0	20.4
Polybrominated Diphenyl Ethers													
BDE 7	58.6	3.0	0.2				BDE 85/155	73.0	2.0	1.8			
BDE 8	77.8	2.2	0.1				BDE 99	75.5	2.4	27.2	0.56		
BDE 10	42.7	6.9	0.2				BDE 100	74.1	2.3	9.4	0.85		
BDE 17	78.1	3.6	0.4				BDE 116	72.8	3.2	1.8			
BDE 25	83.3	3.3	0.8				BDE 118	76.5	4.8	15.1			
BDE 28	70.5	4.7	4.1				BDE 119	75.0	2.9	3.3			
BDE 30	70.6	3.9	0.6				BDE 126	69.2	1.7	2.1			
BDE 32	77.2	1.7	0.7				BDE 138	76.0	1.2	3.3			
BDE 35	82.6	3.2	0.7				BDE 153	76.7	1.3	26.0			
BDE 37	80.3	4.0	1.3				BDE 154	84.8	0.8	0.9			
BDE 49	69.4	5.4	1.3				BDE 155	101.6	0.9	15.6			
BDE 47	71.9	4.5	15.6	1.3			BDE 166	72.4	2.0	2.8			
BDE 66	75.2	5.1	0.6				BDE 181	99.9	2.3	5.8			
BDE 71	67.7	4.8	1.3				BDE 183	73.3	2.3	31.3			
BDE 75	70.0	5.4	4.9				BDE 190	104.4	2.1	5.7			
BDE 77	70.5	6.1	0.8										
97.0													
Averages, % RSD, and PD[c]													
average	60.3	8.5	23.7	109	16.8	23.6	max	136	20.6	205	468	55.4	40.0
							min	20.9	0.8	0.1	0.1	0.0	13.6

[a] Recoveries validated at 26 ng/g wet wt and were corrected for background concentrations of SOCs in sediment. [b] Sample-specific estimated method detection limits. [c] Percent Difference from SRM 1941b certified values n=5. [d] Hexachlorocyclohexane. [e] Dichlorodiphenyldichloroethylene. [f] Dichlorodiphenyldichloroethane. [g] Dichlorodiphenyltrichloroethane.

Table S2. Physical and Chemical Limnological Characteristics of Mills Lake and Lone Pine Lake.
* indicate data from the parameter-elevation regressions on independent slopes model (average
annual total precipitation from 1971-2000, 800×800 m).

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

Table S3. Focus-corrected concentrations and fluxes in sediment from Lone Pine Lake (west) and Mills Lake (east) sediment cores. * indicate below method detection limit and nm indicates no measurement was preformed.

	ΣEndosulfan		Dacthal		ΣDDEs + ΣDDDs		ΣChlordane	
	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)
Lone Pine Lake	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$
Date								
2003	0.57	86	0.034	5.1	2.2	340	0.13	20
1997.5	0.42	67	0.024	3.8	2.5	400	0.13	22
1990	0.47	80	0.029	4.9	4.4	750	0.18	32
1986	0.24	41	0.025	4.3	3.2	550	0.15	26
1982	0.34	57	0.021	3.6	5.2	870	0.20	33
1972.5	0.19	29	0.025	3.7	4.9	730	0.16	24
1967	0.20	31	0.029	4.4	5.5	820	0.18	26
1961	0.15	21	0.022	3.1	5.1	710	0.14	19
1955	0.11	15	0.030	4.0	4.6	590	0.13	17
1949	0.093	12	*	*	7.2	940	0.11	14
1936	0.051	6.1	*	*	5.6	670	0.08	9.4
1920	*	*	*	*	3.3	400	0.03	4.1
1870	*	*	*	*	*	*	*	*
Mills Lake								
Date								
2004	1.7	500	0.068	20	7.0	2100	0.38	110
2000	1.2	360	0.065	19	6.5	2000	0.32	95
1996	1.2	340	0.030	8.6	6.3	1800	0.40	110
1990.5	1.3	310	0.042	10	10	2400	0.43	110
1987.5	0.85	170	0.041	8.1	7.0	1401	0.34	67
1974	0.79	150	0.047	8.8	9.8	1880	0.36	68
1970	0.38	90	0.035	8.4	7.1	1600	0.27	63
1963.5	0.24	77	0.018	5.9	10	3300	0.24	77
1953.5	0.073	23	*	*	8.6	2700	0.12	36
1947	0.045	11	*	*	4.1	1000	0.04	11
1938	0.040	8.7	*	*	1.8	410	0.02	3.7
1905	*	*	*	*	*	*	*	*

	Dieldrin		ΣPAHs		ΣPCBs		ΣPBDEs	
	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)	Conc.(FF)	Flux (FF)
Lone Pine Lake	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$	$ng\,g^{-1}$ dw	$ng\,m^{-2}\,y^{-1}$
Date								
2003	0.12	19	85	13000	0.19	28	1.2	160
1997.5	0.064	10	80	13000	0.22	36	0.45	59
1990	0.075	12	99	17000	0.31	53	nm	nm
1986	0.052	8.9	90	16000	0.24	41	nm	nm
1982	0.070	12	130	21000	0.27	46	1.7	250
1972.5	0.050	7.5	170	25000	0.24	35	nm	nm
1967	0.086	13	190	29000	0.28	41	nm	nm
1961	*	*	170	24000	0.24	33	*	*
1955	*	*	150	19000	0.089	11	nm	nm
1949	*	*	280	37000	0.22	29	nm	nm
1936	*	*	210	25000	0.17	20	*	*
1920	*	*	210	25000	0.061	7.3	*	*
1870	*	*	40	4800	*	*	*	*
Mills Lake								
Date								
2004	0.23	66	180	52000	0.74	210	9.6	2900
2000	0.10	30	190	56000	0.47	140	4.9	1400
1996	0.059	17	150	41000	0.50	140	nm	nm
1990.5	0.18	43	160	37000	0.49	120	2.4	560
1987.5	0.088	18	170	33000	0.41	81	nm	nm
1974	0.095	17	210	40000	0.60	110	0.30	57
1970	0.061	15	160	37000	0.40	97	*	*
1963.5	*	*	180	58000	0.37	120	nm	nm
1953.5	*	*	140	42000	0.10	30	nm	nm
1947	*	*	180	45000	0.064	16	*	*
1938	*	*	240	52000	0.027	5.9	nm	nm
1905	*	*	31	6900	0.050	11	*	*

Table S4. Statistically significant (p<0.05) doubling times and half-lives of current and historic use SOCs in Mills Lake and Lone Pine Lake sediment cores. Half-lives and doubling times were calculated from the natural log focus-corrected flux vs. year and the standard deviation was estimated from the slope of the linear regression. Linear regressions were calculated from the time of U.S. introduction to U.S. restriction and/or U.S. restriction to 2003. ns indicates the linear regression was not statistically significant (p>0.05). [a] reference (7), [b] reference (8), and [c] reference (9).

	Current-use SOCs				Historic-use SOCs		
	ΣEndosulfans	Dacthal	ΣPAHs	ΣPBDEs	ΣChlordane	ΣDDDs + ΣDDEs	ΣPCBs
Mills Lake							
Doubling Time	12.8 ± 0.005	30.7 ± 0.02	22.6 ± 0.01	5.4 ± 0.001	10.9 ± 0.02	ns	8.3 ± 0.01, 13.6 ± 0.01
Half-life	ns	ns	ns	ns	ns	ns	ns
Lone Pine Lake							
Doubling Time	19.1 ± 0.004	ns	ns	ns	27.5 ± 0.01	46.3 ± 0.01	ns
Half-life	ns	ns	28.3 ± 0.01	ns	28.5 ± 0.01	16.0 ± 0.01	ns
Great Lakes							
Doubling Time				17[a], 11[b], 6.4[b]			
Half-life							
Lakes across the U.S.							
Half-life						14.9[c]	19.7[c]

Table S5. Selected SOC ratios (Mills Lake/Lone Pine Lake) in snow (concentrations, flux, and load), lake water (concentrations), and paired sediment intervals (focus-corrected flux). * indicates where a significant difference (p<0.05) exists between Lone Pine Lake and Mills Lake, nd indicates a non detect, and nm indicates not measured in snow. ** indicates data from the parameter-elevation regressions on independent slopes model (1971-2003, 2×2 km).

|	Snow			Lake Water	Sediment Ratio				
	2003	2003	2003	2003	2003 ± 1	1990 ± 1	1987 ± 1	1974 ± 1	1954 ± 1
SOCs	Conc.	Flux	Load	Conc.	Flux	Flux	Flux	Flux	Flux
ΣEndosulfan	1.4*	3.2*	2.3*	4.0*	6.4*	4.1*	3.2*	5.2*	1.5*
Dacthal	1.1	2.5*	1.8*	1.5*	3.8*	2.0*	1.9*	2.4*	nd
ΣDDEs + ΣDDDs	nd	nd	nd	nd	6.1*	3.2*	2.5*	2.5*	4.5*
Trans-Chlordane	1.3	3.0*	2.1*	nd	6.0*	3.2*	2.3*	2.4*	2.7*
Dieldrin	1.6*	4.3*	3.1*	nd	3.5*	3.4*	2.0*	2.3*	nd
ΣPAHs	0.95	2.2*	1.6*	nd	4.1*	2.2*	2.1*	1.6*	2.2*
ΣPCBs	nd	nd	nd	nd	6.5*	2.2	2.0	3.2*	2.6*
ΣPBDEs	nm	nm	nm	nm	24*	3.1*	nd	nd	nd
Precipitation**									
April-Sept. (Summer)					0.90	1.0	0.97	1.0	0.96
Oct-March (Winter)					1.1	0.97	1.0	0.95	0.91
Annual					1.1	1.0	0.96	0.94	0.92
%TOC					1.1	1.3	1.2	1.1	1.0
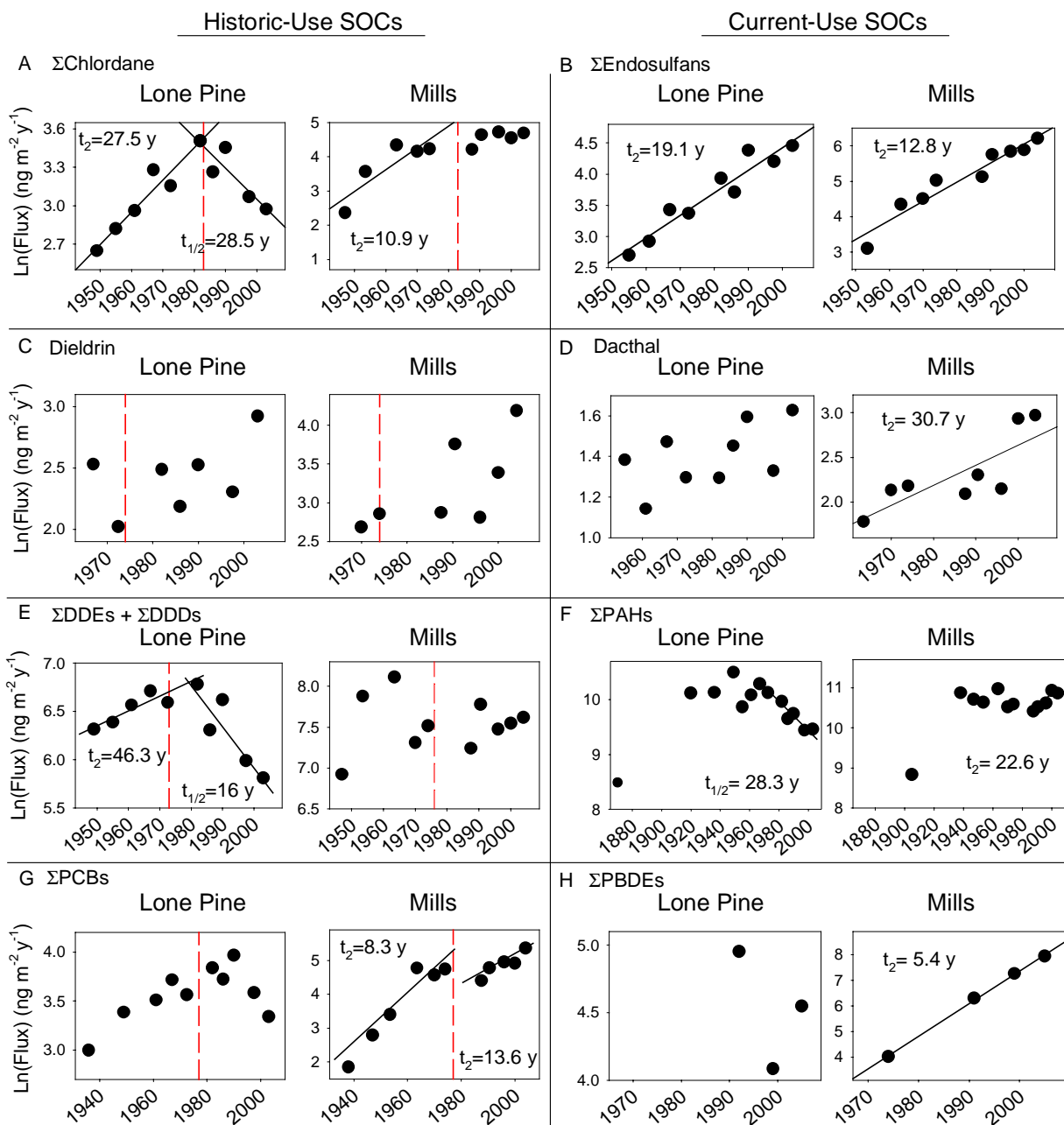
1
2 **Figure S4. Natural log focus-corrected flux (ng m^{-2} y^{-1}) profiles of current and historic-use SOCs in**
3 **Lone Pine Lake (west) and Mills Lake (east) sediment cores. Doubling times (t_2) and half-lives**
4 **($t_{1/2}$) are given where linear regression lines are statistically significant ($p<0.05$). Dashed lines (─**
5 **─) indicate U.S. restriction date. ΣChlordane represents the sum of TC, TN, and CN. Plots start**
6 **at U.S. registration year. Linear regressions were calculated from the time of U.S. introduction to**
7 **U.S. restriction and/or U.S. restriction to 2003.**
8
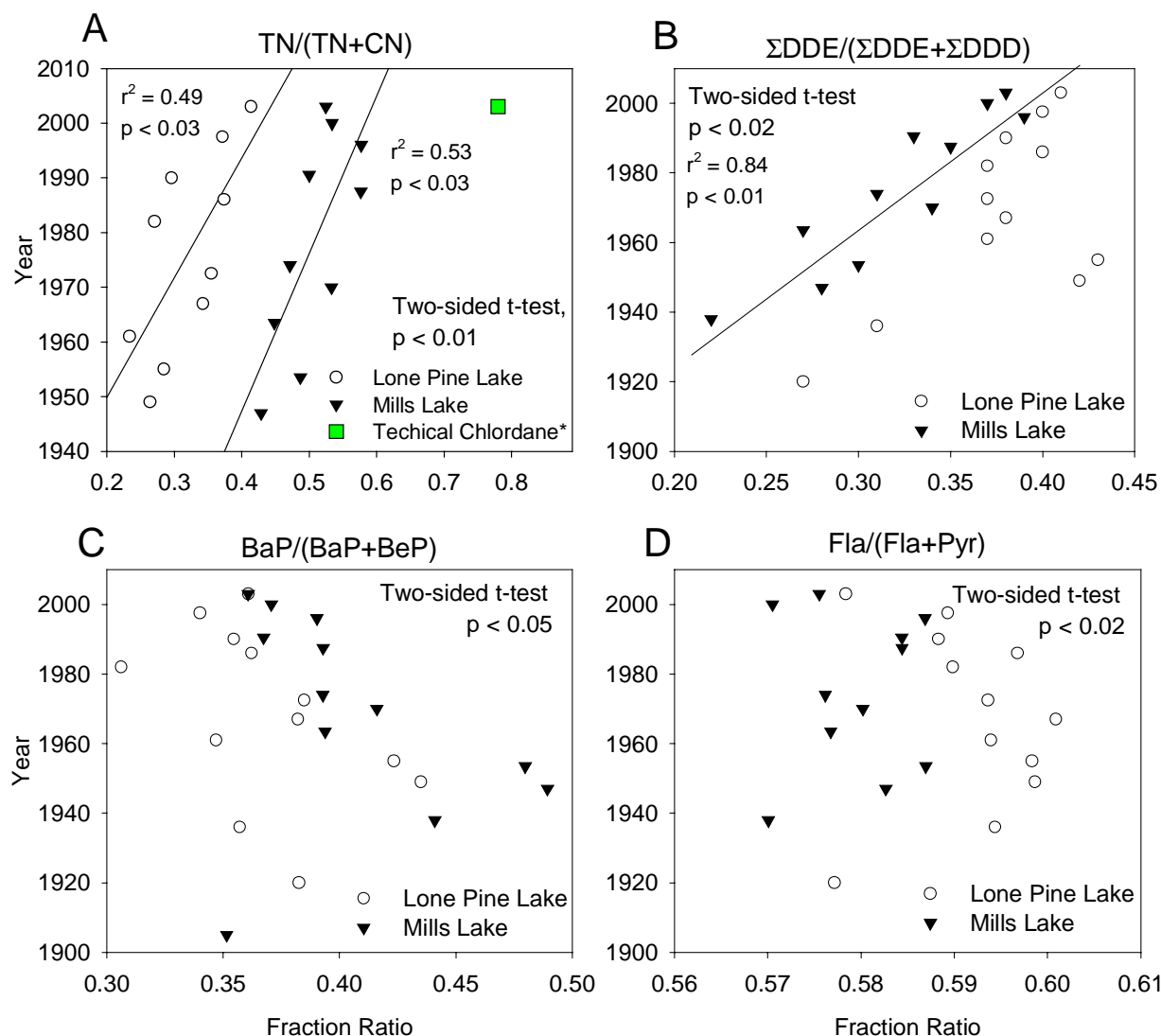
Figure S5. The ratio of TN/(TN+CN), ΣDDE/(ΣDDE+ΣDDD), BaP/(BaP+BeP), and FLA/(FLA+PYR) in sediment from Mills Lake and Lone Pine Lake. Only linear regressions that were statistical significant (p<0.05) are shown. Ratios of contaminants were significantly different between the lakes. * reference (10).

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