Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supporting Information "Tracing the sources and depositional history of mercury to coastal Eastern U.S. lakes"

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Supplemental Methods: age model and mass accumulation rate.

The Constant Rate of Supply (CRS) model is derived explicitly from closed-system radiodecay law, appearing as follows[1,2]:

$$t = \lambda^{-1} \cdot ln\left(\frac{\sum_{\infty}^{0} Pb}{\sum_{\infty}^{i} Pb} \right) \qquad \text{Eq. 1}$$

Here $\sum_{\infty}^{0} Pb$ is the total ^{210}Pb inventory [Bq m^{-2}] summed for all sediment layers from the sediment surface (0) to bottom (∞), and $\sum_{\infty}^{i} Pb$ is summed for all layers below the i^{th} layer to be dated. Each layer inventory is calculated as the measured concentration [Bq kg^{-1}] multiplied by total sediment mass collected for that layer per core cross-sectional area [kg m^{-2}]. The age of the i^{th} layer is given as *t*, and because of the summation this corresponds to the age of the lower layer boundary. The mean age of the layer is thus the average of upper and lower boundary ages.

The Linked Radionuclide aCcumulation (LRC) model is derived from open-system radiodecay law and implemented by a least-squares solution that minimizes the differences between R_{acc} and R_{exp} according to Eq. 2.[3]

$$R_{acc} = \frac{\sum_{i}^{0} Be}{\sum_{i}^{0} Pb} \approx R_{exp} = R_D \frac{\lambda_{Pb} 1 - e^{-\lambda_{Be} t}}{\lambda_{Be} 1 - e^{-\lambda_{Pb} t}}$$

Eq. 2

R_{acc} is the measured accumulative ^{7}Be:^{210}Pb ratio, which for each i^{th} sediment layer is summed for that layer (lower boundary) through all overlying layers to the sediment surface (0). The ratio is thus summed from the bottom up, contrasting to the CRS model which is summed top-down. R_{exp} is the experimental ratio calculated as the quotient of ^{7}Be and ^{210}Pb inventories. The ratio R_D is the empirical flux ratio, which is calculated from the total measured ^{7}Be and ^{210}Pb inventories for the core, where flux (D) = λA and A is total inventory and λ is the corresponding radioactive decay constant. t is the layer age, but in contrast to the CRS model t appears twice and thus precludes an analytical solution to Eq. 2 and is thus solved as unknown using least-squares minimization. As for the CRS model, the LRC is implemented by a summation process and the average layer age is calculated as the average of the upper and lower boundary ages.

For the CRS model, the mass sedimentation rate (r) is derived explicitly from Eq. 2 and implemented as described by Binford et al. (1990), where A is the inventory below the layer and C is the concentration for that layer:

$$r = \lambda^{Pb} \cdot A \cdot C^{-1} \quad \text{Eq. 3}$$

This is approximated (within 1%) as the mass of the interval divided by the difference of upper and lower boundary ages:

$$r = \frac{m}{age_{bottom} - age_{top}}$$

Eq. 4

For the LRC model sedimentation is calculated following Eq. 4 because no analytical solution is available.

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Table 1S: Geographic coordinates, lake and watershed land area and ratio, and attributes of the lakes sampled in this study.

Site	Lake	Location (latitude, longitude)	Lake area (hectares)	Land area (hectares)	Watershed: Lake ratio	Elevation (meters above sea level)	Depth (meters)	Distance to ocean (kilometers)
ANP	Sargent Mountain Pond	44°20.069'N, 68°16.185'W	0.6	2.1	3.6	336	3.5	5.3
ANP	The Bowl	44°20.242'N, 68°11.591'W	4.5	22.9	5.1	126	6	1.4
CCNS	Long Pond	41°56.312'N, 70°0.578'W	15.3	28.9	1.9	1	15	2.1
CCNS	Dyer Pond	41°56.637'N, 70°0.541'W	4.9	13.4	2.7	2	10	1.4

Table 2S is attached as a separate document: Cumulative mass depth, percent loss on ignition (%LOI), radionuclide (lead-210 [^{210}Pb], beryllium-7 [^{7}Be], cesium-137 [^{137}Cs], americium-241 [^{241}Am]) and mercury (Hg) and lead (Pb) concentrations in sections of sediment cores from Sargent Mountain Pond and The Bowl (Acadia National Park, ANS), and Long Pond and Dyer Pond (Cape Cod National Seashore, CCNS). Calculated sediment ages by the Linked Radionuclide aCcumulation (LRC) and Constant Rate of Supply (CRS) model are also given, along with Hg and Pb flux derived from sedimentation rates for LRC and CRS ages.

Table 3S: Quality assurance and control for standard reference materials analyzed for stable Hg isotopes. 2SD is twice the standard deviation. Standard reference materials are as follows: UM-Almaden (National Institute of Standards and Technology (NIST) SRM 8610), secondary standard for mercury (Hg) isotope analyses; MESS-4 (National Research Council Canada), marine sediment; and NIST 1944, estuarine sediment.

Standard	***n***	**δ^{202}Hg Ave. (‰)**	**δ^{202}Hg 2SD (‰)**	**Δ^{199}Hg Ave. (‰)**	**Δ^{199}Hg 2SD (‰)**	**Δ^{200}Hg Ave. (‰)**	**Δ^{200}Hg 2SD (‰)**	**Δ^{201}Hg Ave. (‰)**	**Δ^{201}Hg 2SD (‰)**	**Δ^{204}Hg Ave. (‰)**	**Δ^{204}Hg 2SD (‰)**
UM-Almaden	20	-0.54	0.07	-0.02	0.05	0.01	0.05	-0.03	0.05	-0.01	0.12
NRCC MESS-4	2	-2.00	0.08	-0.01	0.06	-0.02	0.05	-0.04	0.05	0.03	0.10
NIST 1944	3	-0.41	0.08	-0.01	0.03	0.00	0.01	-0.02	0.07	-0.02	0.09

Table 4S: Hg isotope ratio data for sediment core sections from Sargent Mountain Pond, Acadia National Park (ANP) and Long Pond, Cape Cod National Seashore (CCNS).

Core	Depth (cm)	Calendar Age	δ^{202}Hg (‰)	Δ^{199}Hg (‰)	Δ^{200}Hg (‰)	Δ^{201}Hg (‰)	Δ^{204}Hg (‰)
Sargent	0-0.5	2016	-1.18	-0.15	0.08	-0.20	-0.11
Sargent	1-1.5	2008	-1.05	-0.14	0.11	-0.24	-0.18
Sargent	2-2.5	2001	-1.06	-0.17	0.08	-0.23	-0.12
Sargent	3-3.5	1993	-0.96	-0.12	0.11	-0.20	-0.16
Sargent	4-4.5	1983	-1.01	-0.20	0.08	-0.25	-0.15
Sargent	5-5.5	1973	-0.99	-0.15	0.10	-0.16	-0.15
Sargent	6-6.5	1961	-0.99	-0.12	0.06	-0.15	-0.18
Sargent	7-7.5	1948	-1.01	-0.07	0.09	-0.13	-0.17
Sargent	8-8.5	1933	-1.00	-0.04	0.07	-0.14	-0.19
Sargent	9-9.5	1919	-1.07	-0.09	0.04	-0.14	-0.14
Sargent	10-11	1902	-1.26	-0.12	0.06	-0.18	-0.04
Sargent	12-13	1863	-1.46	-0.33	0.04	-0.42	-0.05
Sargent	14-15	1825	-1.73	-0.49	0.02	-0.53	0.00
Sargent	16-17	1772	-1.83	-0.50	0.03	-0.48	-0.03
Sargent	18-19	1732	-1.75	-0.48	-0.01	-0.51	-0.13
Long	0-0.5	2019	-0.29	-0.10	0.07	-0.18	-0.10
Long	1-1.5	2015	-0.35	-0.07	0.05	-0.17	-0.07
Long	2-2.5	2008	-0.44	-0.10	0.09	-0.12	-0.02
Long	3-3.5	2000	-0.42	-0.08	0.10	-0.11	-0.07
Long	4-4.5	1989	-0.39	-0.05	0.05	-0.17	-0.14
Long	5-5.5	1978	-0.43	-0.06	0.05	-0.13	0.00
Long	6-6.5	1967	-0.38	-0.04	0.07	-0.09	-0.10
Long	7-7.5	1956	-0.41	-0.04	0.07	-0.11	-0.07
Long	8-9	1946	-0.52	-0.01	0.02	-0.16	-0.09
Long	9-10	1938	-0.69	0.00	0.03	-0.20	-0.11
Long	10-11	1931	-0.78	-0.03	0.06	-0.22	-0.08
Long	12-13	1920	-0.92	0.01	0.04	-0.18	-0.06
Long	14-15	1904	-0.81	-0.02	0.08	-0.12	-0.04
Long	16-17	1893	-0.78	0.00	0.09	-0.14	-0.02
Long	18-19	1874	-0.69	0.04	0.08	-0.09	-0.08
Long	20-25	1842	-0.57	-0.02	0.05	-0.23	-0.12
Long	25-27	1814	-0.74	-0.02	0.05	-0.24	-0.08

Table 5S: Lead (Pb) isotope ratio data for sediment core sections from Sargent Mountain Pond, Acadia National Park (ANP) and Long Pond, Cape Cod National Seashore (CCNS).

Sample Name	Depth (cm)	Calendar Age	$^{206}Pb/^{207}Pb$
Sargent	0-0.5	2016	1.198
Sargent	0.5-1	2011	1.200
Sargent	1-1.5	2008	1.194
Sargent	1.5-2	2005	1.200
Sargent	2-2.5	2001	1.200
Sargent	2.5-3	1997	1.201
Sargent	3-3.5	1993	1.200
Sargent	3.5-4	1988	1.203
Sargent	4-4.5	1983	1.204
Sargent	4.5-5	1978	1.201
Sargent	5-5.5	1973	1.201
Sargent	5.5-6	1968	1.197
Sargent	6-6.5	1961	1.195
Sargent	6.5-7	1955	1.195
Sargent	7-7.5	1948	1.198
Sargent	7.5-8	1941	1.199
Sargent	8-8.5	1933	1.200
Sargent	8-5.9	1926	1.200
Sargent	9-9.5	1919	1.202
Sargent	9.5-10	1912	1.209
Sargent	10-11	1902	1.220
Sargent	11-12	1882	1.233
Sargent	12-13	1863	1.246
Sargent	13-14	1842	1.225
Sargent	14-15	1825	1.223
Sargent	15-16	1799	1.223
Sargent	16-17	1772	1.224
Long	0-0.5	2019	1.212
Long	0.5-1	2018	1.209
Long	1-1.5	2015	1.210
Long	1.5-2	2012	1.210
Long	2-2.5	2008	1.210
Long	2.5-3	2004	1.210
Long	3-3.5	2000	1.210
Long	3.5-4	1995	1.209
Long	4-4.5	1989	1.210
Long	4.5-5	1983	1.211

Long	5-5.5	1978	1.213
Long	5.5-6	1972	1.210
Long	6-6.5	1967	1.210
Long	6.5-7	1961	1.218
Long	7.5-8	1956	1.222
Long	9-10	1946	1.227
Long	10-11	1938	1.227
Long	11-12	1931	1.222
Long	12-13	1925	1.218
Long	13-14	1914	1.219
Long	14-15	1904	1.217
Long	15-16	1897	1.215
Long	16-17	1893	1.216
Long	17-18	1884	1.223
Long	18-19	1874	1.230
Long	19-20	1866	1.223
Long	20-25	1842	1.219
Long	25-30	1814	1.215

Figure 1S: Location of sampling sites: Acadia National Park (ANP) and Cape Cod National Seashore (CCNS), and Hg emissions (kg yr^{-1}) in the northeastern region of the United States,[4] scaled by to represent the magnitude of annual emissions from each source.

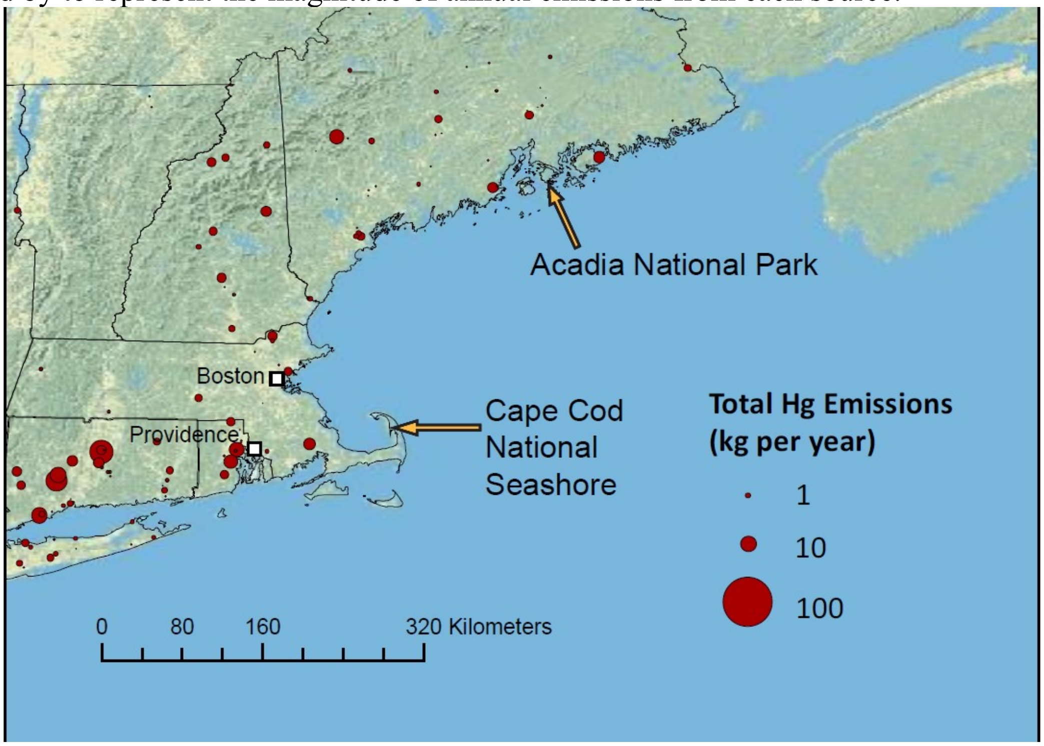


Figure 3S: Mass accumulation rates estimated from the Constant Rate of Supply (CRS) and Linked Radionuclide aCcumulation (LRC) age models.

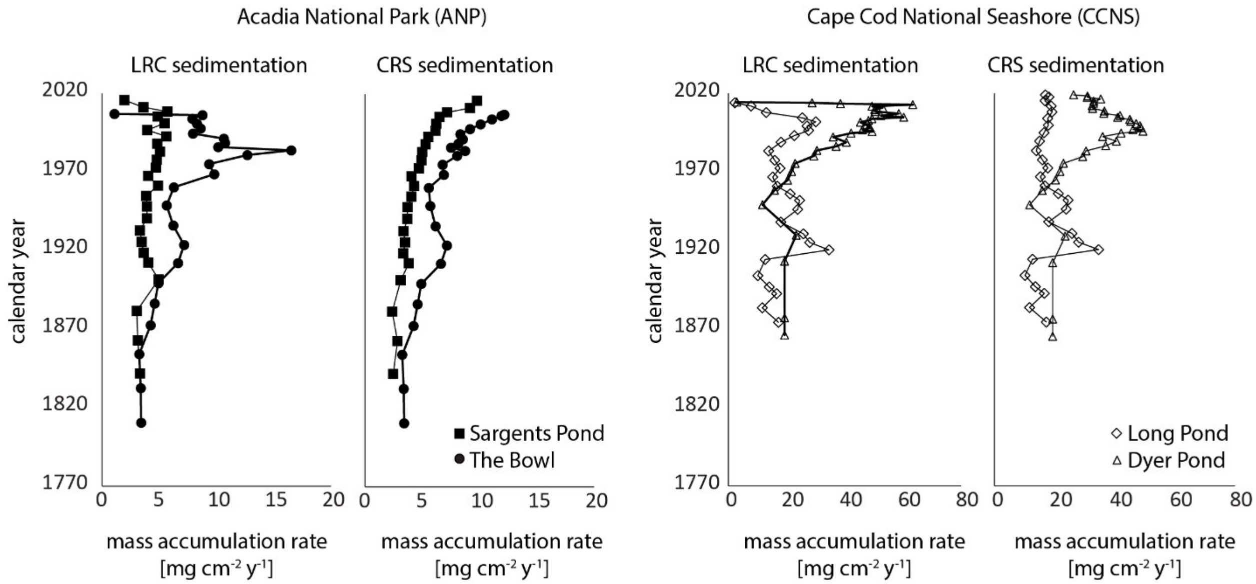


Figure 4S Concentrations and flux rates of Hg and Pb vs. calendar year based on the LRC model for a) Sargent Mountain Pond, ANP; b) The Bowl, ANP; c) Long Pond, CCNS; and d) Dyer Pond, CCNS; and concentrations and flux rates of Hg and Pb vs. calendar year based on the CRS model for e) Sargent Mountain Pond, ANP; f) The Bowl, ANP; g) Long Pond, CCNS; and h) Dyer Pond, CCNS. Dotted lines mark 1850 and 1990, defined here as the beginnings of the industrial and modern eras.

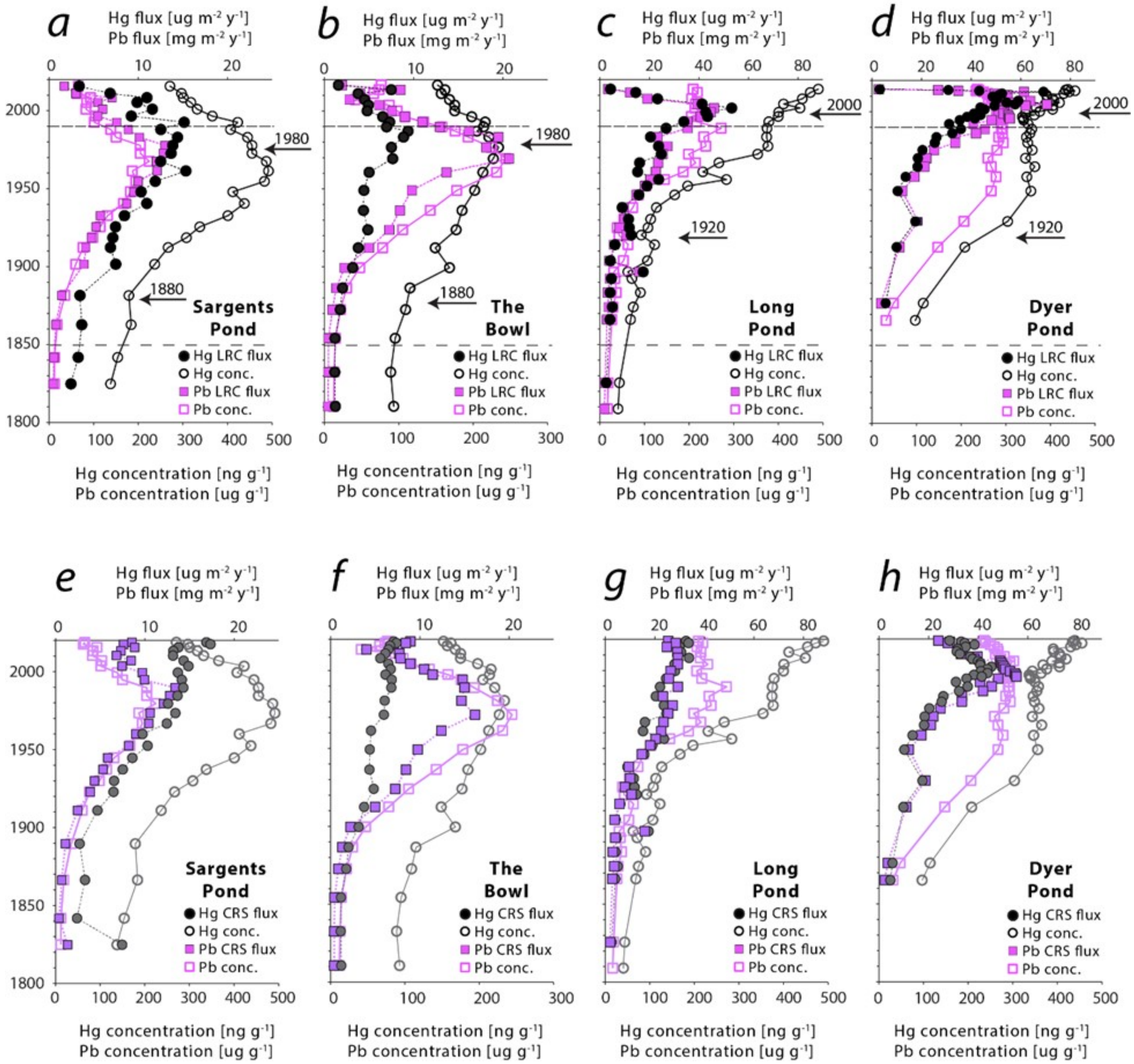


Figure 5S. Hg flux via precipitation from Mercury Deposition Network (MDN) data collected at ANP (Site ME98, blue circles) and CCNS (Site MA01, orange circles). Coefficients of determination (R^2) are calculated for each site. Data were accessed from the National Atmospheric Deposition Program data repository (https://nadp.slh.wisc.edu/networks/mercury-deposition-network/).

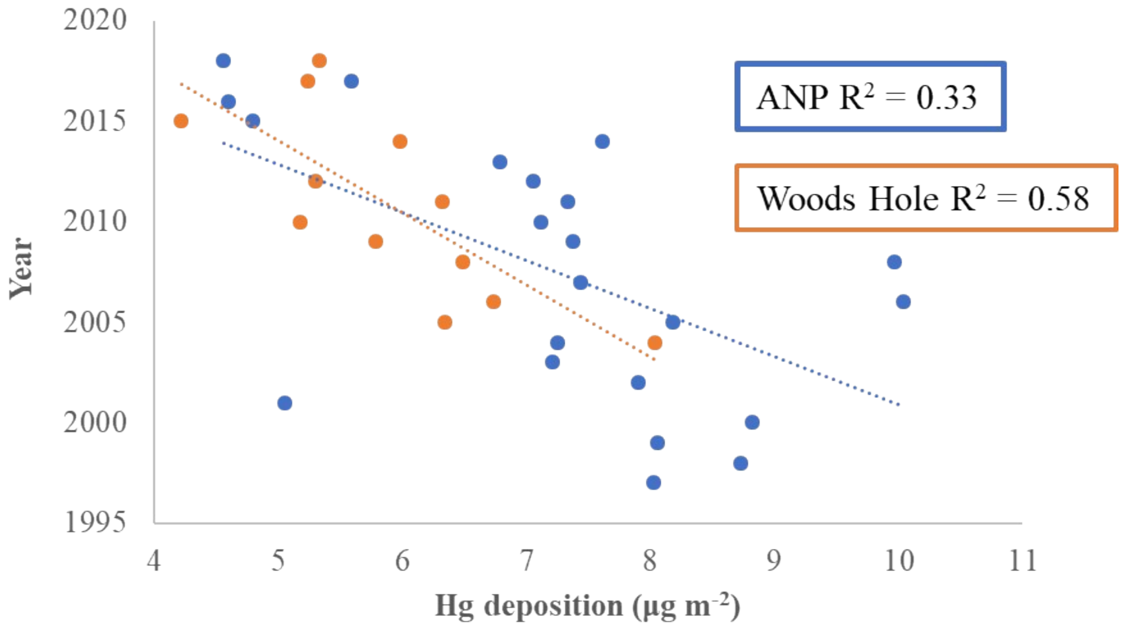


Figure 6S Two endmember mixing plots of δ^{202}Hg and Δ^{199}Hg vs. 1/total Hg (THg) for Sargent Mountain Pond, Acadia National Park (ANP) (blue circles) and Long Pond, Cape Cod National Seashore (CCNS) (orange circles). Open circles denote pre-1850 sediment and closed circles denote post-1850 sediment. Dotted trendlines represent a significant correlation. Brackets show uncertainty in isotope values (2σ).

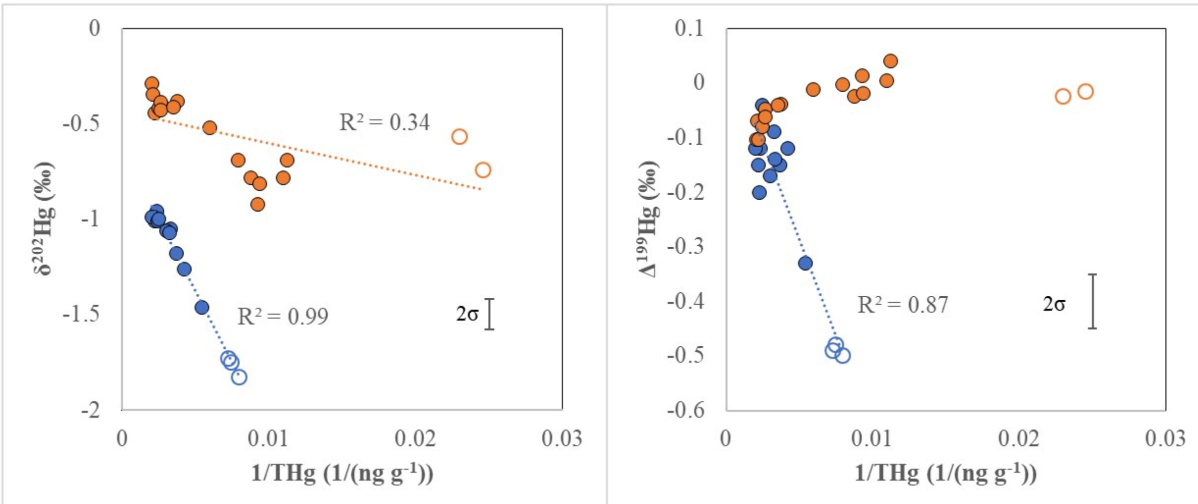
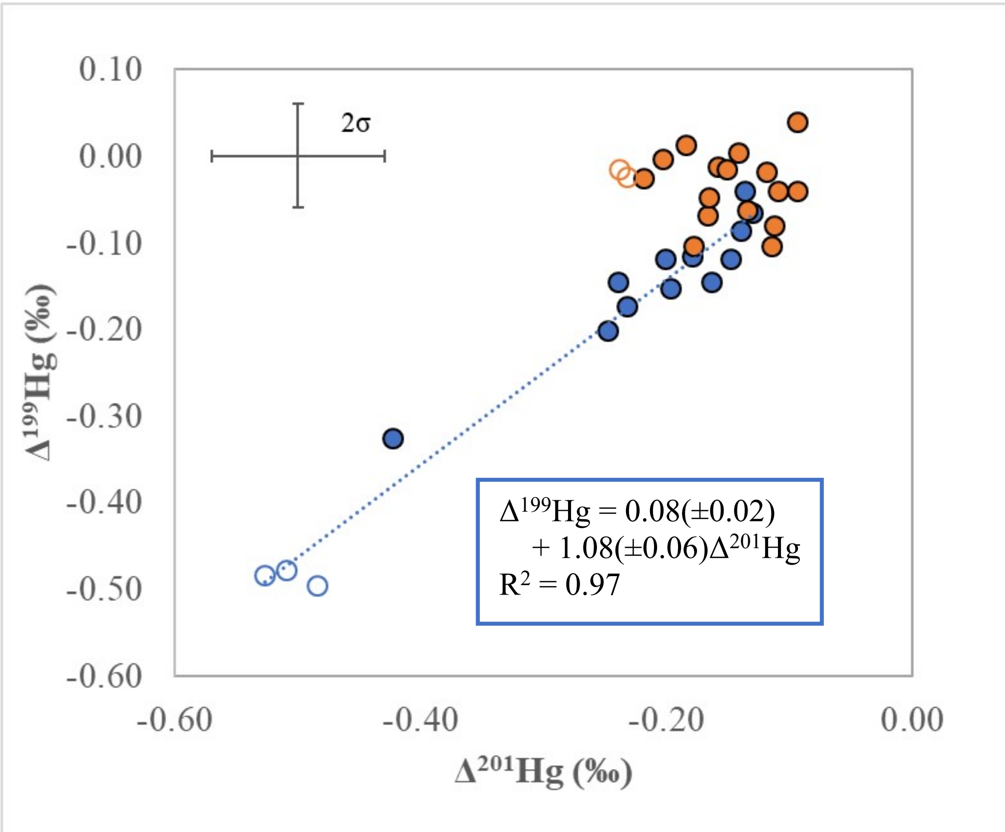


Figure 7S Isotope biplot of $\Delta^{199}Hg$ vs $\Delta^{201}Hg$ for Sargent Mountain Pond, Acadia National Park (ANP) (blue circles) and Long Pond, Cape Cod National Seashore (CCNS) (orange circles) sediments. Open circles denote pre-1850 sediment and closed circles denote post-1850 sediment. Dotted trendlines represent a significant York regression. Brackets show uncertainty in isotope values (2σ).



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