Supporting Information for

Tracing mercury Contamination Sources in Sediments Using Mercury Isotopic Compositions

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Total Hg analysis at IGCAS

The digestion procedure required approximately 200 mg dry sample. The sample was placed inside a 25 mL glass tube, 5mL Milli-Q water and 5mL aqua regia (3:1 HCl:HNO_3) were added, covered with a glass marble, and digested at 95 °C for 5min. Subsequently, 1mL BrCl solution was added at 95 °C for another 30 min. After cooling, the sample reacted for an additional 24 h and 0.2 mL $NH_2OH{\cdot}HCl$ solution (25 g of reagent grade $NH_2OH{\cdot}HCl$ is dissolved in 100 mL Milli-Q water) was added to the sample. Then, THg was determined using cold vapor atomic fluorescence spectrometry (CVAFS). Quality control for the THg determinations was addressed with method blanks, blank spikes, matrix spikes, certified reference material (GBW07305), and blind duplicates. The average THg concentration of the geological standard of GBW07305 were 0.10±0.01 mg/kg (N=6), which is comparable with the certified value of 0.10±0.02 mg/kg. The percentage of recoveries of spiked samples ranged from91 to 112% for THg in sediment samples. The relative percentage difference was <6% for THg analysis in sediment samples.

Table S1 Hg isotope composition of sediment in different depth from a core taken at Matou site in Baihua Reservoir and of cinnabar samples collected in Wanshan Hg mine (in ‰)

Depth (cm)	δ^{199} Hg	SD*	δ^{200} Hg	SD	δ^{201} Hg	SD	δ^{202} Hg	SD	Δ^{199} Hg	SD	Δ^{200} Hg	SD	Δ^{201} Hg	SD
1	-0.26	0.03	-0.54	0.04	-0.87	0.05	-1.10	0.07	0.01	0.02	0.01	0.01	-0.04	0.02
2	-0.23	0.03	-0.41	0.04	-0.66	0.05	-0.81	0.07	-0.02	0.02	0.00	0.02	-0.05	0.02
4	-0.21	0.03	-0.41	0.04	-0.66	0.05	-0.85	0.07	0.00	0.03	0.02	0.01	-0.02	0.04
6	-0.23	0.03	-0.44	0.04	-0.70	0.07	-0.87	0.08	-0.01	0.02	0.00	0.01	-0.04	0.02
8	-0.25	0.07	-0.43	0.07	-0.70	0.10	-0.87	0.09	-0.03	0.04	0.00	0.02	-0.05	0.03
10	-0.19	0.03	-0.34	0.04	-0.57	0.05	-0.70	0.07	-0.01	0.02	0.01	0.01	-0.04	0.02
12	-0.18	0.03	-0.31	0.04	-0.52	0.05	-0.62	0.07	-0.03	0.02	-0.01	0.01	-0.06	0.02
14	-0.22	0.05	-0.39	0.03	-0.61	0.06	-0.75	0.06	-0.03	0.04	0.01	0.01	-0.05	0.03
16	-0.23	0.04	-0.38	0.04	-0.63	0.05	-0.77	0.07	-0.03	0.03	0.01	0.02	-0.05	0.02
18	-0.17	0.06	-0.29	0.06	-0.50	0.11	-0.60	0.13	-0.02	0.04	0.01	0.02	-0.05	0.02
20	-0.21	0.03	-0.35	0.04	-0.58	0.05	-0.69	0.07	-0.04	0.02	-0.01	0.02	-0.06	0.02
22	-0.26	0.06	-0.39	0.04	-0.63	0.07	-0.75	0.08	-0.07	0.03	-0.01	0.01	-0.06	0.02
24	-0.23	0.03	-0.42	0.04	-0.70	0.05	-0.87	0.07	-0.01	0.02	0.01	0.01	-0.05	0.02
26	-0.27	0.05	-0.47	0.08	-0.76	0.12	-0.93	0.13	-0.04	0.02	0.00	0.02	-0.06	0.03
28	-0.36	0.08	-0.56	0.07	-0.91	0.08	-1.10	0.06	-0.08	0.07	-0.01	0.04	-0.08	0.03
Hg ore 1	-0.26	0.04	-0.52	0.06	-0.81	0.09	-1.06	0.10	0.01	0.02	0.01	0.01	0.01	0.02
Hg ore 2	-0.21	0.06	-0.50	0.04	-0.80	0.07	-1.06	0.07	0.06	0.04	0.03	0.01	0.01	0.02
Hg ore 3	-0.24	0.03	-0.42	0.04	-0.68	0.05	-0.85	0.07	-0.02	0.03	0.00	0.03	-0.05	0.02

* SD stands for standard deviation

Table S2 Hg isotope composition of sediment in different depth from a core taken at Houwu site in Hongfeng Reservoir, of soil and coal samples (in ‰)

Depth (cm)	δ^{199}Hg	SD***	δ^{200}Hg	SD	δ^{201}Hg	SD	δ^{202}Hg	SD	Δ^{199}Hg	SD	Δ^{200}Hg	SD	Δ^{201}Hg	SD
1	-0.53	0.03	-1.00	0.04	-1.57	0.05	-2.02	0.07	-0.02	0.02	0.02	0.01	-0.06	0.03
3	-0.51	0.06	-0.89	0.05	-1.43	0.06	-1.79	0.04	-0.06	0.05	0.01	0.03	-0.08	0.04
4	-0.56	0.03	-0.90	0.06	-1.46	0.07	-1.77	0.09	-0.11	0.02	0.00	0.01	-0.13	0.02
5	-0.50	0.03	-0.92	0.04	-1.46	0.05	-1.85	0.07	-0.04	0.02	0.02	0.01	-0.06	0.03
6	-0.52	0.03	-0.85	0.04	-1.40	0.06	-1.73	0.07	-0.08	0.02	0.02	0.01	-0.10	0.02
9	-0.48	0.03	-0.86	0.04	-1.41	0.05	-1.76	0.07	-0.03	0.03	0.02	0.03	-0.08	0.02
11	-0.49	0.03	-0.93	0.04	-1.51	0.05	-1.93	0.07	0.00	0.02	0.04	0.02	-0.06	0.02
14	-0.49	0.03	-0.91	0.04	-1.49	0.05	-1.87	0.07	-0.02	0.02	0.03	0.01	-0.08	0.03
16	-0.50	0.03	-0.83	0.04	-1.53	0.05	-1.95	0.07	-0.01	0.02	0.16	0.01	-0.06	0.02
18	-0.53	0.05	-0.89	0.04	-1.48	0.06	-1.84	0.07	-0.06	0.06	0.03	0.01	-0.09	0.02
20	-0.50	0.03	-0.78	0.07	-1.36	0.09	-1.67	0.12	-0.08	0.02	0.06	0.02	-0.11	0.02
22	-0.55	0.03	-0.83	0.08	-1.42	0.13	-1.70	0.18	-0.12	0.02	0.02	0.01	-0.14	0.02
24	-0.54	0.04	-0.89	0.04	-1.50	0.05	-1.83	0.07	-0.08	0.03	0.03	0.03	-0.13	0.02
26	-0.52	0.04	-0.81	0.04	-1.39	0.05	-1.68	0.07	-0.09	0.05	0.04	0.01	-0.13	0.02
28	-0.61	0.03	-0.92	0.05	-1.51	0.08	-1.80	0.08	-0.16	0.02	-0.02	0.01	-0.16	0.02
30	-0.58	0.03	-0.87	0.04	-1.47	0.05	-1.76	0.07	-0.14	0.02	0.01	0.02	-0.14	0.02
32	-0.54	0.03	-0.86	0.04	-1.48	0.05	-1.75	0.07	-0.10	0.02	0.02	0.02	-0.17	0.03
34	-0.55	0.04	-0.93	0.04	-1.53	0.07	-1.87	0.09	-0.08	0.02	0.01	0.01	-0.13	0.02
39	-0.59	0.03	-0.87	0.04	-1.49	0.05	-1.75	0.07	-0.15	0.02	0.01	0.01	-0.17	0.02
Soil *	-0.59	0.03	-0.98	0.04	-1.64	0.05	-2.04	0.07	-0.08	0.02	0.02	0.06	-0.11	0.01
Guiyang coal 1**	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	-1.37	n.a.	-0.02	n.a.	n.a.	n.a.	0.00	n.a.
Guiyang coal 2**	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	-1.22	n.a.	0.04	n.a.	n.a.	n.a.	0.04	n.a.

* The soil sample was collected from the catchment of Hongfeng Reservoir; * Data from Biswas et al. (Biswas, Blum et al. 2008); *** SD stands for standard deviation

Biswas, A., J. D. Blum, et al. (2008). "Natural Mercury Isotope Variation in Coal Deposits and Organic Soils." Environmental Science & Technology **42**(22): 8303-8309.

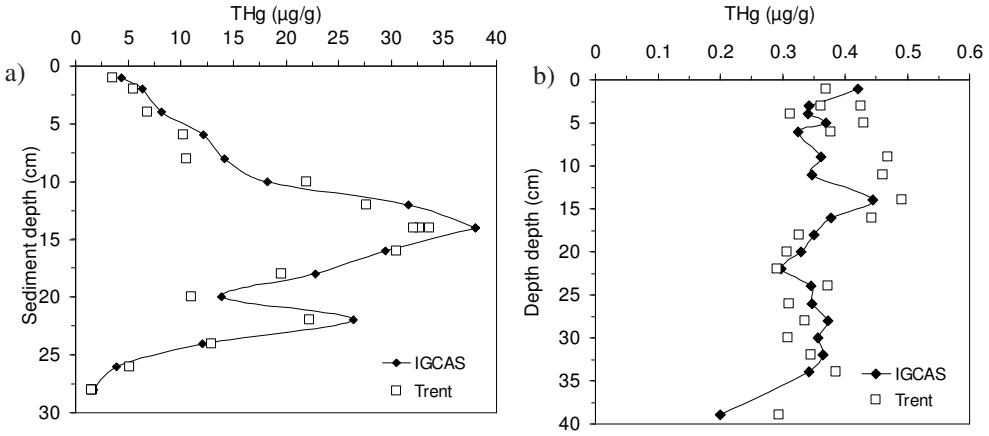

Figure S1 Comparison of THg concentrations in sediment cores in a) Baihua Reservoir and b) in Hongfeng Reservoir. □ Trent: THg measured by using MC-ICP-MC at Trent University;—◆—IGCAS: THg measured by CVAFS at Institute of Geochemistry, Chinese Academy of Sciences