Supporting Information

ISOTOPIC VARIABILITY OF MERCURY IN ORE, MINE-WASTE CALCINE, AND LEACHATES OF MINE-WASTE CALCINE FROM AREAS MINED FOR MERCURY

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Table S-1: δ^{x}Hg values relative to NIST 3133 for all samples of Hg ore, mine-waste calcine and leachates of mine-waste calcine analyzed in this study.

Sample[a]	n	δ^{202}Hg (‰)	2sd[b]	δ^{201}Hg (‰)	2sd[b]	δ^{200}Hg (‰)	2sd[b]	δ^{199}Hg (‰)	2sd[b]	Δ201Hg[c]	2sd	Δ200Hg[c]	2sd	Δ199Hg[c]	2sd
UM-Almadén 1 μg/L	38	-0.61	0.24	-0.46	0.1	-0.3	0.14	-0.14	0.1	-0.01	0.11	0.01	0.06	0.01	0.10
UM-Almadén 2 μg/L	18	-0.58	0.09	-0.45	0.08	-0.28	0.05	-0.16	0.06	-0.01	0.06	0.01	0.03	-0.02	0.05
		Mine-waste calcine-Terlingua													
03MAR1	2	1.04	0.24	0.78	0.10	0.53	0.14	0.23	0.10	-0.01	0.06	0.00	0.03	-0.03	0.05
03MAR2	2	0.76	0.24	0.53	0.10	0.36	0.14	0.14	0.10	-0.04	0.06	-0.02	0.03	-0.05	0.05
03MAR3	2	0.46	0.24	0.37	0.15	0.25	0.14	0.15	0.10	0.03	0.06	0.02	0.03	0.03	0.05
03MSM1	4	0.39	0.24	0.21	0.17	0.12	0.40	-0.07	0.38	-0.08	0.05	-0.08	0.36	-0.17	0.36
03MSM2	2	0.64	0.46	0.34	0.64	0.33	0.25	0.10	0.17	-0.15	0.14	0.01	0.01	-0.06	0.05
03MSM3	3	1.52	0.32	1.02	0.24	0.74	0.19	0.25	0.12	-0.13	0.06	-0.03	0.03	-0.13	0.05
03MSM4	2	0.64	0.24	0.45	0.10	0.34	0.14	0.14	0.10	-0.03	0.06	0.02	0.03	-0.02	0.05
03MSM5	2	0.7	0.24	0.49	0.10	0.33	0.14	0.09	0.10	-0.04	0.06	-0.02	0.03	-0.09	0.05
03SB1	4	-0.46	0.24	-0.20	0.14	-0.18	0.14	0.12	0.10	0.14	0.06	0.05	0.03	0.23	0.05
03SB2	2	-0.47	0.24	-0.32	0.10	-0.24	0.14	-0.06	0.10	0.03	0.06	0.00	0.03	0.05	0.05
03SB3	2	-1.34	0.28	-0.97	0.24	-0.65	0.14	-0.20	0.10	0.04	0.06	0.03	0.03	0.14	0.05
03SB4	2	-0.53	0.24	-0.38	0.10	-0.25	0.14	-0.11	0.10	0.01	0.06	0.01	0.03	0.02	0.05
03SB5	2	-0.6	0.24	-0.42	0.10	-0.30	0.14	-0.09	0.10	0.04	0.06	0.00	0.03	0.07	0.05
03TER1	2	0.08	0.24	0.15	0.23	0.10	0.14	0.12	0.10	0.10	0.11	0.06	0.06	0.10	0.14
03TER2	2	0.31	0.24	0.27	0.10	0.19	0.14	0.12	0.10	0.04	0.11	0.03	0.06	0.04	0.10
03TER3	2	0.61	0.24	0.41	0.10	0.32	0.14	0.13	0.10	-0.05	0.11	0.02	0.06	-0.02	0.10
03TER4	2	0.6	0.24	0.43	0.27	0.27	0.14	0.04	0.11	-0.02	0.18	-0.03	0.04	-0.12	0.14
03TER5	2	0.3	0.24	0.10	0.43	0.11	0.14	0.02	0.10	-0.12	0.32	-0.04	0.06	-0.05	0.10
		Mine-waste calcine -McDermitt													
99MCD1	4	-1.49	0.29	-1.05	0.24	-0.73	0.16	-0.21	0.17	0.07	0.15	0.02	0.08	0.17	0.11
99MCD2	3	-0.22	0.24	-0.12	0.10	-0.12	0.16	-0.07	0.10	0.05	0.15	-0.01	0.07	-0.02	0.10
99MCD3a	2	-0.64	0.24	-0.45	0.10	-0.30	0.14	-0.15	0.10	0.03	0.11	0.02	0.13	0.01	0.10
99MCD3b	2	-0.47	0.24	-0.30	0.12	-0.29	0.14	-0.16	0.14	0.05	0.11	-0.05	0.06	-0.05	0.12
01MCD2	3	0.28	0.24	0.25	0.22	0.16	0.14	0.09	0.18	0.05	0.11	0.02	0.06	0.02	0.13

Hg minerals-Terlingua															
Calomel-USGS	2	-0.75	0.24	-0.50	0.10	-0.36	0.14	-0.09	0.10	0.07	0.11	0.01	0.06	0.10	0.10
Calomel-CSM	3	-2.05	0.31	-1.38	0.28	-0.98	0.18	-0.22	0.10	0.17	0.11	0.05	0.03	0.30	0.05
Montroydite-USGS	2	1.05	0.24	0.81	0.22	0.49	0.16	0.19	0.10	0.02	0.11	-0.04	0.07	-0.08	0.10
Montroydite-CSM	2	1.39	0.24	0.99	0.18	0.66	0.16	0.26	0.11	-0.05	0.11	-0.04	0.06	-0.09	0.10
Metacinnabar	1	-0.07		-0.10		-0.01		-0.05		-0.05		0.03		-0.03	
Cinnabar-CSM	3	-1.65	0.25	-1.16	0.33	-0.82	0.17	-0.23	0.12	0.09	0.15	0.01	0.04	0.19	0.06
Cinnabar-CSM	1	-1.6		-1.06		-0.75		-0.18		0.14		0.06		0.22	
Cinnabar-USGS	2	-1.72	0.24	-1.19	0.10	-0.82	0.14	-0.25	0.10	0.10	0.03	0.04	0.09	0.19	0.12
Terlinguaite/Kleinite-USGS	3	-2.7	0.28	-1.80	0.35	-1.29	0.14	-0.33	0.10	0.23	0.15	0.07	0.03	0.36	0.05
Terlinguaite-CSM	2	-2.15	0.33	-1.53	0.22	-1.09	0.18	-0.27	0.10	0.16	0.03	0.03	0.01	0.29	0.06
Kleinite-CSM	3	-2.15	0.33	-1.44	0.29	-1.05	0.16	-0.25	0.10	0.17	0.03	0.03	0.00	0.29	0.08
Cinnabar-McDermitt															
MCD100	3	-0.70	0.24	-0.54	0.16	-0.36	0.14	-0.16	0.10	-0.01	0.16	0.00	0.03	0.01	0.07
MCD101	2	-0.64	0.24	-0.39	0.10	-0.30	0.14	-0.14	0.10	0.09	0.11	0.03	0.06	0.02	0.10
MCD102a	2	-0.52	0.25	-0.32	0.10	-0.21	0.14	-0.13	0.17	0.07	0.13	0.05	0.01	0.00	0.11
MCD102b	2	-0.56	0.24	-0.44	0.10	-0.26	0.14	-0.18	0.13	-0.01	0.04	0.02	0.02	-0.04	0.15
MCD102c	2	-0.50	0.24	-0.36	0.10	-0.25	0.14	-0.15	0.10	0.01	0.11	0.00	0.06	-0.03	0.10
MCD102d	2	-0.42	0.24	-0.41	0.11	-0.24	0.14	-0.11	0.10	0.01	0.50	-0.03	0.07	0.00	0.11
MCD102e	2	-0.61	0.24	-0.45	0.12	-0.28	0.14	-0.12	0.10	0.01	0.11	0.03	0.06	0.04	0.10
MCD102f	4	-0.69	0.26	-0.46	0.17	-0.36	0.14	-0.18	0.10	0.06	0.18	-0.01	0.04	-0.01	0.06
MCD103a	3	-0.58	0.24	-0.39	0.10	-0.31	0.14	-0.12	0.12	0.04	0.11	-0.02	0.06	0.03	0.10
MCD103b	2	-0.60	0.24	-0.48	0.27	-0.26	0.14	-0.15	0.10	-0.03	0.11	0.04	0.06	0.00	0.10
MCD104	3	-0.61	0.24	-0.43	0.10	-0.31	0.14	-0.13	0.10	0.03	0.11	0.00	0.06	0.02	0.10
Leachates-Terlingua															
03MAR1	2	2.09	0.34	1.59	0.24	1.06	0.25	0.50	0.18	0.02	0.06	0.01	0.07	-0.02	0.10
03MAR2	2	0.82	0.09	0.62	0.08	0.40	0.05	0.26	0.06	0.00	0.06	-0.01	0.03	0.05	0.05
03MAR3	2	0.20	0.09	0.20	0.08	0.12	0.05	0.17	0.06	0.05	0.06	0.02	0.03	0.12	0.05
03MSM5	2	1.11	0.20	0.88	0.12	0.54	0.10	0.47	0.16	0.04	0.06	-0.02	0.03	0.19	0.11
03SB1	2	0.71	0.18	0.66	0.20	0.39	0.13	0.30	0.06	0.13	0.06	0.03	0.04	0.18	0.05
03SB4	2	-0.17	0.09	-0.12	0.08	-0.08	0.05	-0.02	0.06	0.00	0.06	0.01	0.03	0.03	0.08
03TER1a	2	0.54	0.35	0.43	0.34	0.32	0.13	0.24	0.10	0.04	0.13	0.03	0.03	0.10	0.05

03TER1b	2	0.64	0.35	0.49	0.34	0.33	0.17	0.19	0.10	0.01	0.08	0.01	0.03	0.03	0.05
		Leachates-McDermitt													
99MCD1a	2	-1.43	0.20	-0.96	0.16	-0.70	0.12	-0.32	0.07	0.11	0.06	0.02	0.03	0.04	0.13
99MCD1b	2	-1.49	0.08	-0.99	0.10	-0.71	0.04	-0.06	0.32	0.12	0.06	0.04	0.03	0.32	0.33

a. MAR=Mariposa mine, MSM=Mariscal mine, SB=Study Butte mine, TER=Terlingua mine, MCD=McDermitt mine.

b. The 2sd error for each sample is calculated from replicate analyses of the sample. Where the calculated 2sd was smaller than that of the replicate analyses of the secondary standard UM-Almadén, the value for UM-Almadén is used for the uncertainty of the δ^{x}Hg.

c. The Δ^{x}Hg values were calculated using the approximated formulas published in Blum and Bergquist, 2007.

Table S-2. Isotopic composition of Hg in cinnabar samples from the McDermitt mine relative to NIST 3133. Samples MCD102a-f represent replicate digestions of cinnabar from a single sample of ore. Each digested sample was analyzed for isotopic composition and the average of n isotopic measurements reported.

Sample	n	δ^{202}Hg (‰)	Description
Replicate digestions, single ore			
MCD102a	2	-0.52±0.24	Cinnabar in vein quartz
MCD102b	2	-0.56±0.09	Cinnabar in vein quartz
MCD102c	2	-0.50±0.18	Cinnabar in vein quartz
MCD102d	2	-0.42±0.11	Cinnabar in vein quartz
MCD102e	2	-0.60±0.09	Cinnabar in vein quartz
MCD102f	2	-0.69±0.25	Cinnabar in vein quartz
average	14	-0.57±0.25	
Separate ore samples			
MCD100	3	-0.70±0.16	Cinnabar in sinter[a]
MCD101	2	-0.64±0.09	Cinnabar in sinter[a]
MCD102	14	-0.57±0.26	Cinnabar in vein quartz
MCD103	5	-0.59±0.09	Cinnabar in vein quartz
MCD104	3	-0.61±0.12	Cinnabar in sinter
average	27	-0.60±0.20	

Table S-3: Total and leachable Hg in mine-waste calcine samples from the Terlingua District and McDermitt mine.

sample	total Hg (μg g^{-1})	leachable[a] Hg (μg L^{-1})	leachable fraction of total Hg	leachate pH
Mine-waste calcine –Terlingua District				
03MAR1	170	64.1	7.54E-03	7.7
03MAR2	190	5	5.26E-04	7.4
03MAR3	35	0.28	1.60E-04	7.2
03MSM1	6.9	<0.1	<2.90E-04	7.3
03MSM2	31	<0.1	<6.45E-05	7.1
03MSM3	44	<0.1	<4.55E-05	7.1
03MSM4	110	<0.1	<1.82E-05	7.1
03MSM5	150	<0.1	<1.33E-05	7.3
03SB1	5900	102	3.46E-04	7.4
03SB2	12	0.32	5.33E-04	7.1
03SB3	480	<0.1	<4.17E-06	7.3
03SB4	3000	0.5	3.33E-06	3.8
03SB5	35	0.1	5.71E-05	7.3
03TER1	19000	1118	1.18E-03	6.1
03TER2	14	1.04	1.49E-03	7.4
03TER3	16	0.23	2.88E-04	7.1
03TER4	170	0.1	1.18E-05	7.4
03Ter5	4.1	0.1	4.88E-04	7.1
Mine-waste calcine -McDermitt				
99MCD1	1200	21	3.50E-04	8.8
99MCD2	43	0.2	9.30E-05	4.3
99MCD3a	1400	0.2	2.86E-06	3.2
99MCD3b	1400	0.2	2.86E-06	3.2
01MCD2	200	NA	NA	4.3

a. A synthetic rainwater leach (US EPA method 1312) was performed on all calcines using 100g of waste material and 2L of synthetic rainwater. Leachates were filtered by 0.45 mm filter prior to analysis for total Hg in leachate.

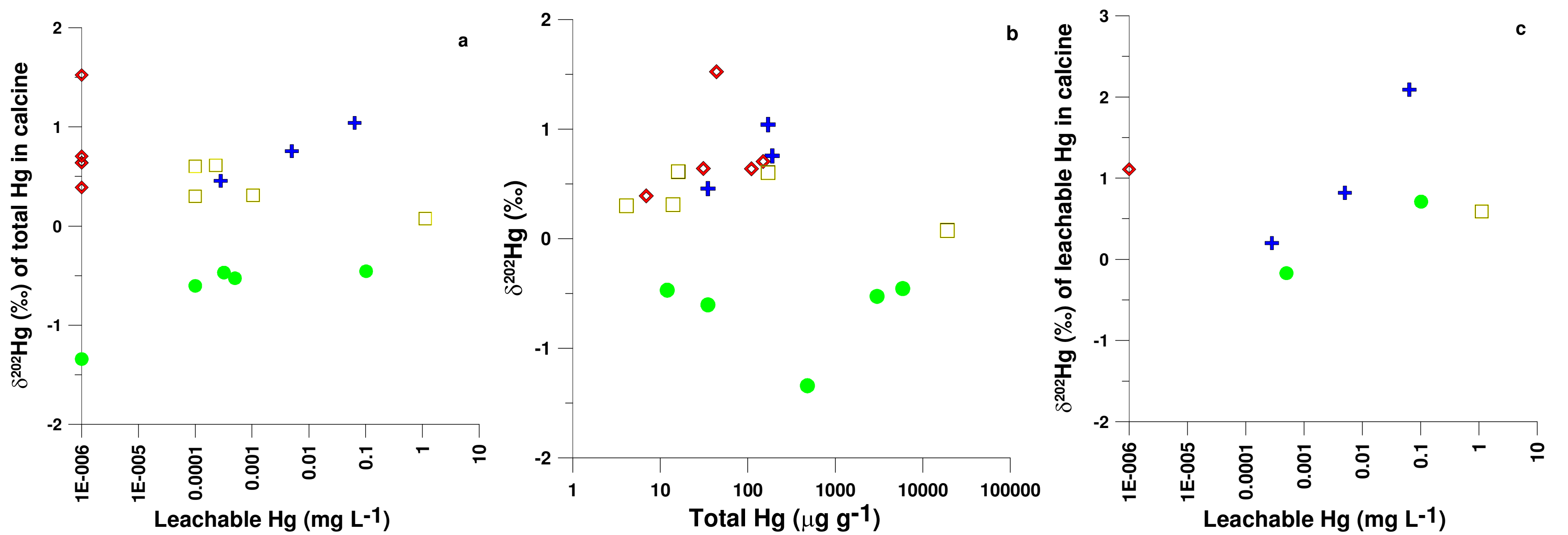

Figure S-1: Variation of Hg isotopic composition in calcines relative to synthetic rainwater leachable Hg concentration (a) and Hg total concentration (b) and variation of Hg isotopic composition in calcine leachates relative to synthetic rainwater leachable Hg concentration (c). Data are plotted by mine:+-Mariposa,◊-Mariscal, ●-Study Butte, □-Terlingua, ▲-McDermitt.