Supporting information for "Photo-microbial visible light-induced magnetic mass independent fractionation of mercury in a marine microalga"

K. Kritee[1], Laura C. Motta, Joel D. Blum, Martin Tsz-Ki Tsui and John R. Reinfelder

<u>This SI includes supporting text, SI Tables 1-6, SI Figures 1-3 and SI references</u>

SI Table 1: Summary of Hg(II) reduction experiments
SI Table 2: Summary of MeHg degradation experiments
SI Table 3: Isotope data from all Hg(II) reduction experiments
SI Table 4: Isotope data from all MeHg degradation experiments
SI Table 5: Isotope data for standard reference materials UM-Almáden during 2011-2013
SI Table 6: Calculation of rate of photo-microbial intracellular degradation of MeHg and reduction of Hg(II)

SI Figure 1: Mass dependent fractionation during degradation of MeHg by exudates, growing microalga and intra-cellular processes.
SI Figure 2: Δ^{199}Hg/Δ^{201}Hg ratios for Hg(II) reduction experiments
SI Figure 3: Δ^{199}Hg/Δ^{201}Hg ratios for MeHg degradation experiments

Supporting text

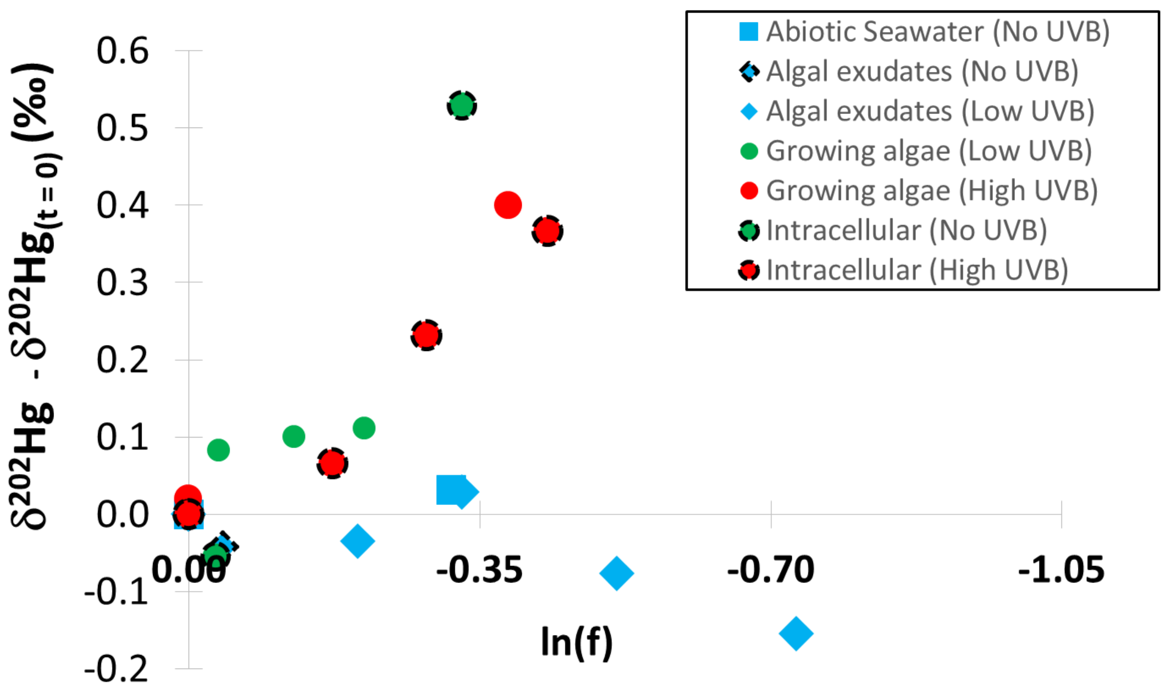
Emission spectra of cool white fluorescent lamp: Cool white fluorescent lamps produce light with sharp peaks corresponding to emissions from mercury and rare earth phosphors in the UV (312 nm, 365 nm) and visible (405 nm, 436 nm, 544 nm, 546 nm, and 611 nm), a broad peak at 485-490 nm, and several small peaks between 578 nm and 693 nm[1]. In contrast, natural sunlight consists of a continuum of finely spaced peaks of UV and visible light[2].

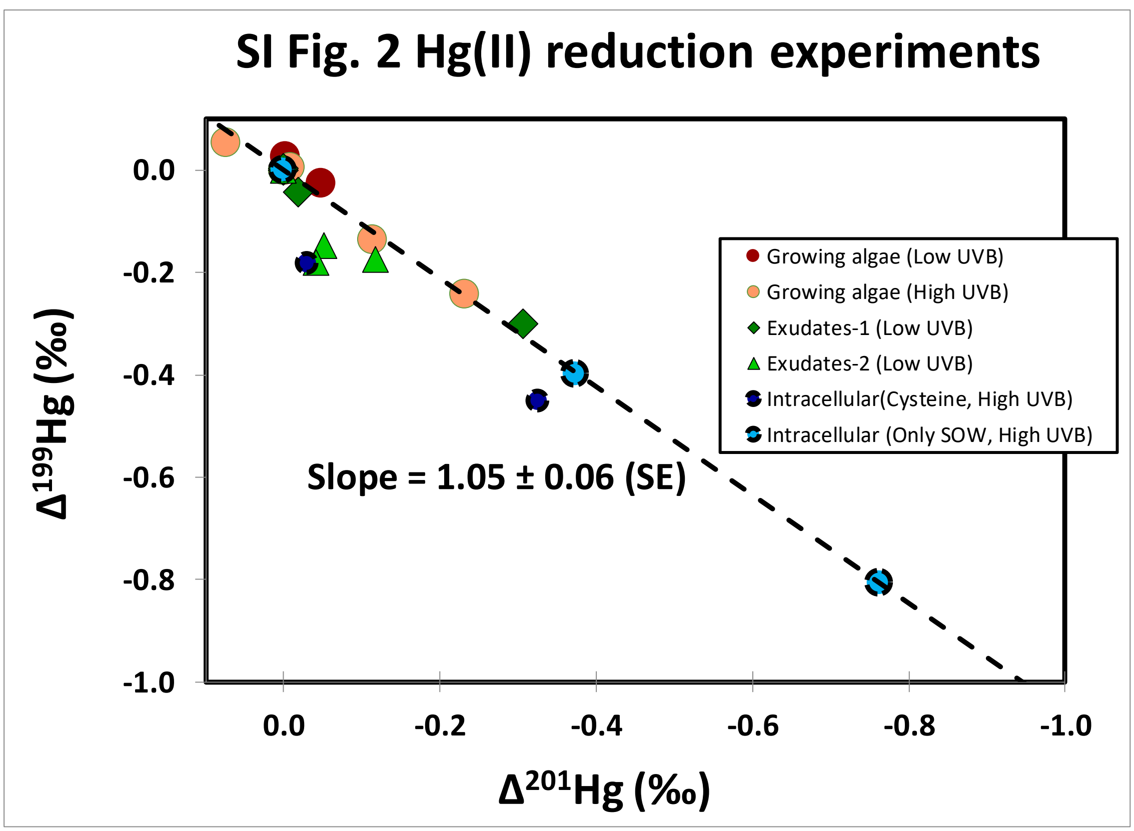
No effect of UV lamp expected or observed in our study: While MIF may be enhanced inside of Hg vapor compact fluorescent lamps[3], we did not observe any MIF of Hg(II) in glass reactors and Hg(II) MIF was of the opposite sign as MeHg in Teflon reactors, which indicates that the chemical form and aqueous speciation of Hg, not the light source, were the most important factors in the generation of MIF. In addition, much of the energy from the Hg vapor lamps, that cells and media were exposed to in our experiments, was concentrated in Hg emission lines at 365.4, 404.7 and 435.8 nm. All of these lines would be attenuated according to the measured spectrally resolved percent transmissions for glass, Teflon, and the Lee filter. Rose et al[4] tested the effect of different regions of the solar spectrum on the expression of MIF caused by the MIE during Hg(II) and MeHg photo-reduction. The experiments indicate that MIF produced during photo-reduction of Hg(II) is significantly influenced by both UVB and UVA radiation. They showed that for MeHg photodemethylation, however, UVB radiation is primarily responsible for the MIF with only minor contributions from UVA. To create their "no UV" experimental conditions in the reactors, Rose et al (2015) covered their reactors with acrylic sheets such that 96% and 98% of incident UV-B and UV-A, respectively, were blocked. Given that incident UV-B was ~2.5 W m^{-2} and UV-A was ~50 W m^{-2} (see Figure 1C and 1D in Rose et al), the power experienced by the contents of the reactor was ~0.1 W m^{-2} for UV-B and ~1 W m^{-2} for UV-A. If all of the UV-A was from wavelength 365.4 nm, it would translate to >3 µmol m^{-2} s^{-1}. In our no UVB (visible plus very low UV-A) experiments, UV-A irradiance was 0.2 µmol m^{-2} s^{-1}, 15 times lower than UVA intensity in the Rose et al experiments.

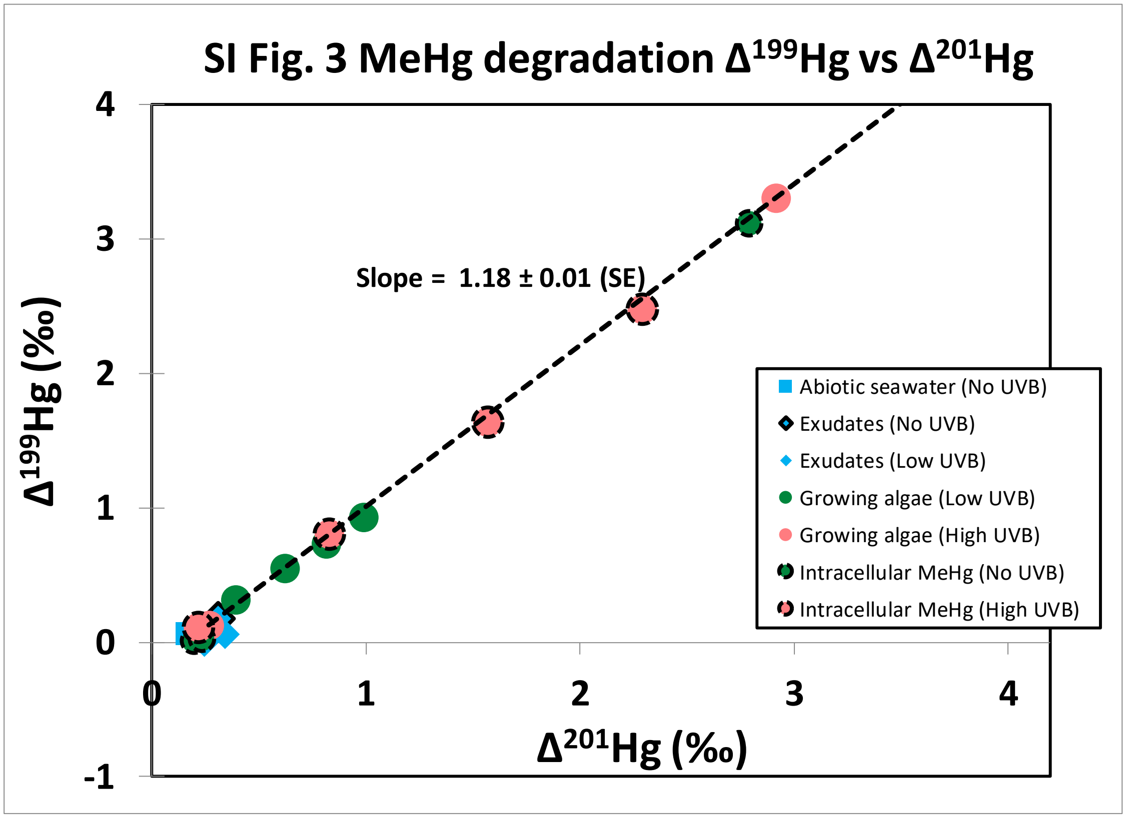
Mass dependent fractionation: We note that in the growing algae experiments with Hg(II) and MeHg have a fluctuating level of exposure of light because of changing number of cells in the reactor. One experiment (abiotic exudates, low UV) apparently had negative MDF with enrichment of lighter isotopes in the reactor. This particular experiment was long (10 days), the associated Rayleigh plot had an R^2 of 0.66 and the mass dependent isotope ratios fluctuated during the experiment with significant negative MDF signal appearing only for the last data-point. Theoretically, negative (or inverse) kinetic mass dependent fractionation happens when back reactions occur (e.g., it is well documented for N stable isotopes during nitrification of nitrite to nitrate). We do not expect significant back reactions of Hg(0) in our study because we were constantly bubbling the reactors to purge out Hg(0). It is possible that some form of ionic mercury was back reacting to form MeHg.

Steps involved in mass independent fractionation during demethylation: As noted in the main text, magnetic isotope effect (MIE)[5, 6] leads to a $\Delta^{199}Hg/\Delta^{201}Hg$ ratio of 1.0 for Hg(II) and 1.2 to 1.3 for MeHg. Given that some of our $\Delta^{199}Hg/\Delta^{201}Hg$ values for our demethylation experiments (SI Table 2) are between 1 and 1.3 (instead of being between 1.2 and 1.3), a small influence of MIF during Hg(II) reduction on demethylation experiments is possible. The fact that the changes in $\Delta^{199}Hg$ for growing algae experiment with low UV for Hg(II) were not highly linear (R^2 <0.3) makes it possible that we have somehow missed seeing high negative MIF during low UVB Hg(II) reduction experiments.

SI Fig 1 MDF during MeHg Degradation






SI Fig. 3 MeHg degradation Δ199Hg vs Δ201Hg
Slope = 1.18 ± 0.01 (SE)
Δ199Hg (‰)
Δ201Hg (‰)
4
3
2
1
0
-1
0
1
2
3
4
Abiotic seawater (No UVB)
Exudates (No UVB)
Exudates (Low UVB)
Growing algae (Low UVB)
Growing algae (High UVB)
Intracellular MeHg (No UVB)
Intracellular MeHg (High UVB)

Supplementary Table 1: Summary of Hg(II) reduction experiments

Treatments	Reactor type	Initial Cells/ml	Final Cells/ml	Starting [Hg(II)] (ng/ml)	Starting [Hg(II)] (nM)	Initial intra-cellular conc. (nmol/chla)[b]	Length of experiment (days)	Rate constant ($hour^{-1}$)	Rate constant (24 hrs of light) (day^{-1})	Hg reduced by the end (%)	MDF $^{202}\varepsilon$ (‰)	MIF $^{199}\varepsilon$ (‰)	Δ199/ Δ201	Δ199/ δ202	Date
Abiotic marine exudates (DOC) + Hg(II) (Low UVB)	Glass[a]	5.1E+05	N/A	31.2	156	N/A	8	0.0041	0.10	50	1.08	-0.45	0.96	-0.40	April 5 2011
Abiotic marine exudates (DOC) + Hg(II) (Low UVB)	Glass[a]	5.5E+05	N/A	5.1	26	N/A	10	0.0025	0.06	44	1.47	-0.30	1.28	-0.20	Aug 23 2012
Abiotic marine exudates (DOC) + Hg(II) (High UVB)	Teflon	1.4E+06	N/A	1.1	6	N/A	4	0.1003	2.41	97	*Not analyzed for isotopes*				Jan 11 2013
Growing algae + Hg(II) (Low UVB)	Glass[a]	5.1E+05	5.1E+06	35.8	179	N/A	8	0.0031	0.07	46	0.61[c]	(-0.08)[d]	~0[e]	~0[e]	April 5 2011
Growing algae + Hg(II) (High UVB)	Teflon	4.1E+05	2.9E+05	22.1	111	N/A	4	0.0180	0.43	83	0.14[c]	-0.16	1.02	-1.07	Jan 11 2013
Intracellular Hg(II) (Cysteine wash) (High UVB)	Teflon	2.9E+05	1.2E+05	3.9	19	0.85	4	0.0159	0.38	38	0.79	-0.89	1.19	-1.11	Jan 11 2013
Intracellular Hg(II) (Cysteine wash) (High UVB)	Teflon	7.2E+05	5.7E+05	8.0	40	0.69	3	0.0195	0.47	72	*Not analyzed for isotopes*				Feb 6 2013
Intracellular Hg(II) (Ocean-water wash) (High UVB)	Teflon	2.6E+05	2.4E+05	5.8	29	1.41	4	0.0091	0.22	54	0.70	-1.03	1.06	-1.46	Jan 11 2013
Intracellular Hg(II) (Ocean-water wash) (High UVB)	Teflon	1.2E+06	6.7E+05	18.7	94	1.00	3	0.0063	0.15	15	*Not analyzed for isotopes*				Feb 6 2013

[a] *Thick Borosilicate (Also see Table 1 in the main manuscript)*

[b] *chla implies Chlorophyll a. Concentrations of intracellular contents in algal cells in often represented in units of per unit chlororphyll content*

[c] *The range of isotopic enrichments in the growing algae experiments may have resulted from the apparent lack of cell growth in the Teflon ($\delta^{202}\varepsilon = 0.1$) compared with the glass reactor ($\delta^{202}\varepsilon = 0.6‰$).*

[d] *Very poor $R^2 < 0.3$*

[e] *Because of very low MIF.*

Supplementary Table 2: Summary of MeHg degradation experiments

Treatments	Reactor type	Initial Cells/ml	Starting [Hg] (ng/ml)	Starting [Hg] (nM)	Initial intra-cellular conc. (nmol/chla)[c]	Length of experiment (days)	Rate constant ($hour^{-1}$)	Rate constant (24 hrs of light) (day^{-1})	Hg reduced by the end (%)	MDF $^{202}\varepsilon$ (‰)	MIF $^{199}\varepsilon$ (‰)	Δ199/ Δ201	Δ199/ δ202	Date
Abiotic marine exudates + MeHg (Low UVB)	Glass[b]	1.0E+06	10.9	55	N/A	10	0.0030	0.07	52	-0.21[f]	0	N/A	N/A	Dec 12 2012
Abiotic marine exudates + MeHg (No UVB)[a]	Teflon + Lee	2.2E+05	8.1	41	N/A	5	0.0003	0.01	0	N/A[d]	N/A[d]	N/A	N/A	Aug 17 2013
Growing Algae + MeHg (High UVB)	Teflon	7.0E+05	6.1	30	N/A	4	0.0178	0.43	73	1.2[e]	8.3[e]	1.20	8.0	Aug 27 2013
Growing algae + MeHg (Low UVB)	Glass[b]	5.6E+05	7.7	38	N/A	10	0.0010	0.02	19	0.23[f]	3.5	1.09	4.3[f]	Dec 12 2012
Intracellular MeHg (Ocean-water wash) (High UVB)	Teflon	1.5E+05	3.1	16	1.27	4	0.0057	0.14	35	0.9	5.6	1.14	6.2	Aug 26 2013
Intracellular MeHg (Ocean-water wash) (No UVB)[a]	Teflon + Lee	1.6E+05	2.8	14	1.08	4	0.0037	0.09	28	1.7	9.8	1.20	5.5	Aug 26 2013
Abiotic artificial Seawater + MeHg (No UVB)[a]	Teflon + Lee	N/A	15.4	77	N/A	5	0.0020	0.05	27	0.1	0	N/A	N/A	Aug 17 2013
Abiotic (High UVB, Low DOC, Science 2007)	Quartz	N/A	66	330	N/A	0.3	0.0313	0.75	20	1.4	3.3	1.36	2.4	
Abiotic (High UVB, High DOC, Science 2007)	Quartz	N/A	93	465	N/A	0.3	0.0321	0.77	20	1.6	7.8	1.36	4.8	

[a] *No UVB experiments also had very low UVA exposure (see Table 1)*

[b] *Thick Borosilicate (Also see Table 1 in the main manuscript)*

[c] *chla implies Chlorophyll a. Concentrations of intracellular contents in algal cells in often represented in units of per unit chlororphyll content*

[d] *Only two data points and negligible reduction*

[e] *Anomalously, there was no MDF or MIF between 0 and 54 hours. The t = 0 data point was not included in the calculation of enrichment factors*

[f] *R^2 is poor (0.66) for Abiotic marine exudates and Very poor (0.33) for growing algae (Low UV-B)*

Supplementary Table 3: Isotope data from all Hg(II) reduction experiments

Time (hrs)	[Hg] (ppb)	f (fraction Hg remaining)	δ^{202}Hg (‰)	δ^{204}Hg (‰)	δ^{201}Hg (‰)	δ^{200}Hg (‰)	δ^{199}Hg (‰)	Δ^{199}Hg (‰)	Δ^{201}Hg (‰)	Δ^{204}Hg (‰)	Δ^{200}Hg (‰)
Abiotic marine exudates (DOC) + Hg(II) (Glass: Low UVB) (April 5, 2011)											
0	31.21										
(Just after light) 20.5	31.13	1.00	-0.91	-1.36	-0.67	-0.48	-0.20	0.03	0.01	0.00	-0.03
27.5	30.01	0.96									
45.5	27.66	0.89	-0.72	-1.09	-0.53	-0.37	-0.15	0.04	0.01	-0.02	-0.01
101	22.06	0.71	-0.48	-0.63	-0.36	-0.22	-0.13	-0.01	0.00	0.08	0.02
143	18.13	0.58									
189	15.73	0.51	-0.15	-0.20	-0.41	-0.06	-0.31	-0.27	-0.29	0.02	0.02
Growing Algae + Hg(II) (Glass: Low UVB) (April 5, 2011)											
0	35.81										
(Just after light) 20.5	33.93	1.00	-0.91	-1.31	-0.68	-0.48	-0.23	0.00	0.01	0.05	-0.02
27.5	30.93	0.91									
45.5	29.36	0.87	-0.71	-1.08	-0.53	-0.32	-0.15	0.02	0.00	-0.02	0.03
101	24.63	0.73									
189	19.47	0.57	-0.54	-0.75	-0.45	-0.26	-0.16	-0.03	-0.04	0.06	0.01
Abiotic marine exudates (DOC) + Hg(II) (Glass: Low UVB) (August 23, 2012)											
(Just after light)0	5.12	1.000	-0.36	-0.52	-0.31	-0.15	-0.04	0.05	-0.05	0.01	0.03
172.9	2.96	0.578	0.40	0.62	0.20	0.20	0.00	-0.10	-0.10	0.03	0.00
220.9	2.88	0.563	0.50	0.77	0.29	0.27	-0.01	-0.13	-0.09	0.02	0.02
244.9	2.86	0.559	0.54	0.70	0.24	0.26	0.01	-0.13	-0.16	-0.10	-0.01
Abiotic marine exudates (DOC) + Hg(II) (Teflon: High UVB) (Jan 11, 2013)											
0.00	1.10										
(Just after light)15.66	0.72										
21.25	0.21	*Not analyzed for isotopic composition*									
39.22	0.06										
43.97	0.03										
Growing Algae + Hg(II) (Teflon: High UVB) (Jan 11, 2013)											
0.00	22.14		-0.54	-0.81	-0.46	-0.27	-0.16	-0.02	-0.06	0.00	0.00
(Just before light)15.6	15.45	1.00	-0.55	-0.84	-0.40	-0.26	-0.11	0.03	0.02	-0.01	0.01
21.2	13.12	0.85	-0.60	-0.82	-0.51	-0.29	-0.17	-0.02	-0.06	0.07	0.01
39.2	6.92	0.45	-0.44	-0.67	-0.50	-0.22	-0.27	-0.16	-0.17	-0.01	0.00
90.4	3.84	0.25	-0.33	-0.49	-0.53	-0.20	-0.35	-0.26	-0.29	0.00	-0.03
Intracellular Hg(II) (Cysteine + SOW washed cells; Teflon: High UVB) (Jan 11, 2013)											
0.00	3.86	1	-0.60	-0.90	-0.54	-0.30	-0.11	0.04	-0.08	0.00	0.01
(Just before light)15.7	3.73	*Not analyzed*									
21.25	3.38	0.88	-0.53	-0.80	-0.51	-0.27	-0.28	-0.14	-0.11	-0.01	-0.01
44.00	2.37	0.62	-0.23	-0.42	-0.58	-0.15	-0.47	-0.41	-0.41	-0.08	-0.03
Intracellular Hg(II) (SOW washed cells; Teflon: High UVB) (Jan 11, 2013)											
0	5.79	1	-0.71	-1.05	-0.57	-0.35	-0.16	0.02	-0.04	0.01	0.01
(Just before light) 15.9	5.32	*Not analyzed*									
39.25	3.97	0.68	-0.40	-0.69	-0.72	-0.21	-0.48	-0.38	-0.42	-0.09	-0.01
90.55	2.65	0.46	-0.16	-0.31	-0.92	-0.08	-0.83	-0.79	-0.80	-0.08	0.00
Intracellular Hg(II) (Cysteine + SOW washed cells; Teflon: High UVB) (Feb 6, 2013)											
(Just before light) 0	8.02										
15.90	4.88	*Not analyzed for isotopic composition*									
27.8	4.75										
74.0	1.79										
Intracellular Hg(II) (SOW washed cells; Teflon: High UVB) (Feb 6, 2013)											
(Just before light) 0	18.72										
15.90	17.35										
27.83	16.03	*Not analyzed for isotopic composition*									
46.25	13.32										
74.00	12.13										

Supplementary Table 4: Isotope data from all MeHg(II) reduction experiments

Time* (hrs)	[Hg] (ppb)	f (fraction Hg remaining)	$\delta^{202}Hg$ (‰)	$\delta^{204}Hg$ (‰)	$\delta^{201}Hg$ (‰)	$\delta^{200}Hg$ (‰)	$\delta^{199}Hg$ (‰)	$\Delta^{199}Hg$ (‰)	$\Delta^{201}Hg$ (‰)	$\Delta^{204}Hg$ (‰)	$\Delta^{200}Hg$ (‰)
Intracellular MeHg (Teflon with Lee Filter) No UVB and very low UVA (Aug 26, 2013)											
0.0	2.761	1.00	-1.10	-1.71	-0.83	-0.49	-0.11	0.01	0.00	-0.11	0.04
24.4	2.847	1.03									
48.7	2.672	0.97	-1.15	-1.80	-0.83	-0.64	-0.26	0.03	0.04	-0.08	-0.06
94.0	1.987	0.72	-0.57	-0.84	2.16	-0.30	2.97	3.11	2.59	0.01	-0.01
Intracellular MeHg (Teflon) High UVB (Aug 26, 2013)											
0.0	3.114	1.00	-1.05	-1.71	-0.77	-0.51	-0.15	0.11	0.02	-0.14	0.02
24.4	2.621	0.84	-0.99	-1.50	-0.11	-0.48	0.55	0.80	0.63	-0.03	0.02
48.7	2.339	0.75	-0.82	-1.31	0.75	-0.40	1.43	1.64	1.37	-0.08	0.01
74.2	2.023	0.65	-0.68	-0.88	1.58	-0.31	2.31	2.48	2.09	0.14	0.03
Growing Algae with MeHg (Teflon) High UVB (Aug 27, 2013)**											
0	6.091		-1.12	-1.69	-0.73	-0.49	-0.10	0.18	0.11	-0.02	0.07
30.8	5.744										
54.2	2.45	1.00	-1.10	-1.53	-0.76	-0.59	-0.15	0.13	0.07	0.11	-0.03
79.2	1.667	0.68	-0.72	-1.07	2.18	-0.36	3.13	3.31	2.72	0.00	0.00
Growing Algae with MeHg (Glass) Low UVB (Dec 12, 2012)											
0.0	7.69	1.00	-1.02	-1.48	-0.73	-0.55	-0.17	0.08	0.03	0.03	-0.04
81.5	7.41	0.96	-0.93	-1.41	-0.51	-0.46	0.08	0.32	0.19	-0.01	0.01
150.0	6.77	0.88	-0.92	-1.30	-0.27	-0.41	0.32	0.55	0.42	0.07	0.05
196.8	6.30	0.82	-0.98	-1.39	-0.12	-0.50	0.49	0.73	0.61	0.08	0.03
236.8	6.22	0.81	-0.91	-1.27	0.11	-0.44	0.70	0.93	0.79	0.09	0.00
Abiotic marine exudates (DOC) + MeHg (Glass) Low UVB (Dec 12, 2012)											
0.0	10.94	1.00	-0.88	-1.35	-0.59	-0.45	0.06	0.06	0.07	-0.04	-0.01
81.5	8.93	0.82	-0.91	-1.37	-0.54	-0.48	-0.17	0.06	0.14	-0.01	-0.02
150.0	7.87	0.72	-0.85	-1.35	-0.59	-0.49	-0.22	0.00	0.05	-0.09	-0.07
174.8	6.54	0.60	-0.95	-1.43	-0.68	-0.44	-0.13	0.11	0.04	0.00	0.03
236.8	5.27	0.48	-1.03	-1.55	-0.70	-0.53	-0.18	0.08	0.09	0.01	0.00
Abiotic marine exudates (DOC) + MeHg (Teflon + Lee Filter) No UVB (Aug 17, 2013)											
0	8.13	1.00	-1.10	-1.69	-0.73	-0.49	-0.10	0.18	0.11	-0.02	0.07
118.8	7.80	0.96	-1.14	-1.71	-0.81	-0.58	-0.22	0.07	0.05	-0.01	0.00
Abiotic Artificial Seawater + MeHg (Teflon + Lee Filter) No UVB (Aug 17, 2013)											
0	15.38	1.00	-1.06	-1.60	-0.83	-0.53	-0.21	0.06	-0.03	-0.02	0.00
8.3	12.76	0.83									
118.8	11.22	0.73	-1.03	-1.58	-0.75	-0.62	-0.21	0.05	0.03	-0.05	-0.10

*Light was turned on right before time = 0 minute for all experiments.

** There was no fractionation between 0 and 54 hours.

Supplementary Table 5: Isotope data for standard reference materials UM-Almáden during 2011-2013

Session date	Session Name (for internal use)	[Hg] (ppb)	Replicates	δ204	1SD	δ202	1SD	δ201	1SD	δ200	1SD	δ199	1SD	Δ204	1SD	Δ201	1SD	Δ200	1SD	Δ199	1SD
5-Apr-11	Hg_MT_Kritee_05Apr11	5.0	6	-0.90	0.06	-0.59	0.03	-0.49	0.03	-0.28	0.01	-0.16	0.06	-0.01	0.06	-0.04	0.03	0.01	0.01	-0.01	0.06
26-Apr-11	Hg_MT_Kritee_26Apr11	5.0	7	-0.80	0.06	-0.57	0.03	-0.47	0.04	-0.29	0.04	-0.17	0.04	0.05	0.05	-0.04	0.03	-0.01	0.03	-0.03	0.03
7-Jun-11	Hg_MT_Kritee_07Jun11	5.0	7	-0.89	0.09	-0.55	0.04	-0.45	0.04	-0.26	0.03	-0.16	0.03	-0.06	0.07	-0.04	0.02	0.02	0.02	-0.03	0.02
21-Jul-11	Hg_MWJ_Kritee_21Jul11	4.0	5	-0.83	0.13	-0.56	0.06	-0.47	0.11	-0.28	0.06	-0.18	0.04	0.01	0.06	-0.04	0.07	0.00	0.05	-0.03	0.02
6-Oct-11	Hg_MT_Kritee_06Oct11	4.5	7	-0.89	0.05	-0.55	0.02	-0.45	0.04	-0.28	0.04	-0.17	0.03	-0.06	0.04	-0.03	0.03	0.00	0.03	-0.03	0.03
10-Nov-11	Hg_MT_Kritee_10Nov11	5.0	5	-0.85	0.05	-0.57	0.01	-0.46	0.05	-0.27	0.03	-0.17	0.04	0.00	0.05	-0.03	0.04	0.01	0.03	-0.02	0.04
23-Feb-12	Hg_MT_Kritee7th_23Feb12	5.0	5	-0.85	0.08	-0.56	0.03	-0.43	0.04	-0.25	0.03	-0.16	0.05	-0.02	0.04	-0.01	0.03	0.03	0.02	-0.02	0.04
10-May-12	Hg_MT_Kritee8th_10May12	5.0	6	-0.90	0.10	-0.62	0.05	-0.51	0.05	-0.32	0.04	-0.19	0.03	0.03	0.04	-0.04	0.04	-0.01	0.02	-0.03	0.04
4-Jun-12	Hg_MT_HB_Kritee_Steelhead2008_04Jun12	5.0	5	-0.91	0.03	-0.61	0.03	-0.49	0.03	-0.29	0.02	-0.18	0.02	0.00	0.02	-0.03	0.03	0.01	0.02	-0.02	0.02
11-Oct-12	Hg_MT_Laura_Rutgers_23_samples_11Oct1	5.0	4	-0.82	0.10	-0.59	0.06	-0.49	0.04	-0.29	0.05	-0.15	0.06	0.06	0.03	-0.05	0.02	0.01	0.02	0.00	0.06
19-Dec-12	Hg_MT_SJ-sed._MEF-UMBS-bugs_19Dec12	5.0	5	-0.91	0.06	-0.62	0.04	-0.50	0.06	-0.31	0.05	-0.19	0.04	0.01	0.04	-0.03	0.04	0.00	0.04	-0.04	0.04
21-Jan-13	2013Jan21_MT_HgKMnO4_Rutgers	5.0	5	-0.78	0.11	-0.60	0.11	-0.50	0.13	-0.28	0.11	-0.20	0.11	0.12	0.14	-0.05	0.08	0.02	0.06	-0.05	0.09
24-Jan-13	2013Jan24_MT_HgKMnO4_Rutgers	5.0	5	-0.80	0.21	-0.48	0.10	-0.39	0.11	-0.25	0.11	-0.08	0.07	-0.08	0.24	-0.03	0.09	-0.01	0.09	0.04	0.07
25-Apr-13	2013Apr25_MT_LM_Rutgers	5.0	5	-0.86	0.05	-0.55	0.03	-0.47	0.04	-0.25	0.02	-0.14	0.02	-0.04	0.01	-0.06	0.03	0.03	0.02	0.00	0.03
22-May-13	2013May22_MT_Rutgers_Experimental_Mo	5.0	6	-0.89	0.07	-0.60	0.02	-0.48	0.04	-0.31	0.02	-0.16	0.01	0.00	0.07	-0.03	0.03	-0.01	0.02	-0.01	0.02
24-May-13	2013May22_MT_Rutgers_NJ_Sed_Janssen	5.0	7	-0.85	0.04	-0.56	0.06	-0.47	0.06	-0.28	0.04	-0.17	0.03	-0.01	0.08	-0.04	0.03	0.00	0.02	-0.03	0.02
Total Sessions				Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD	Avg	2SD
16				-0.86	0.08	-0.57	0.07	-0.47	0.06	-0.28	0.04	-0.16	0.05	0.00	0.10	-0.04	0.02	0.01	0.02	-0.02	0.04

Supplementary Table 6: Calculation of rate of photo-microbial intracellular degradation of MeHg and reduction of Hg(II)

	Rate constant (day^{-1})	[Hgp]* (pM)	Intracellular** Hg (%)	Intracellular Hg (pM)	Hg loss (pM day^{-1})	Hg loss (pM $year^{-1}$)	Hg reduction rate (pmol m^{-2} d^{-1})	Hg reduction rate (nmol m^{-2} y^{-1})	Hg reduction rate (Mmol $year^{-1}$)
Intracellular Hg(II) reduction (Upper range)	0.19	0.04	9	0.0036	0.000684	0.2	36.9	13.5	**4.9**
Intracellular Hg(II) reduction (Lower range)	0.234	0.04	9	0.0036	0.0008424	0.3	45.5	16.6	**6.0**
Intracellular MeHg reduction	0.045	0.003	63	0.00189	8.439E-05	0.0	**4.6**	1.7	0.6

* Estimate of concentration of particulate Hg(II) from Soerensen et al, 2010 and particulate MeHg from Hammerschmidt and Bowman (2012)

** % of total particulate that in inside the cells (vs on the cell surface) from Mason et al (1996)

SI References

1. Elvidge, C. D.; D.M. Keith; B.T. Tuttle; Baugh, K. E., *Sensors* 2010, *10*, 3961-3988.
2. ASTM Standard Tables for Reference Solar Spectral Irradiances: Direct Normal and Hemispherical on 37° Tilted Surface G173-03. *American Society for Testing and Materials: 2012*.
3. Mead, C.; Lyons, J. R.; Johnson, T. M.; Anbar, A. D., *Environ. Sci. Technol.* 2013, *47* (6), 2542-2547.
4. Rose, C. H.; Ghosh, S.; Blum, J. D.; Bergquist, B. A., *Chem. Geol.* 2015, *405* (0), 102-111.
5. Bergquist, B. A.; Blum, J. D., *Science* 2007, *318* (5849), 417-420.
6. Zheng, W.; Hintelmann, H., *J. Phys. Chem. A* 2010, *114* (12), 4246-4253.