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Dynamic mercury methylation and demethylation in oligotrophic marine water

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Figure S1: Initial methylation rates of Hg(II) in Tropical Pacific waters.
Methylation rates from the chlorophyll maxima (top) and oxygen minima (bottom) at t 0 h time point. Methylation rates of ^{202}Hg(II) from triplicate bottle incubations (± 1 SD).

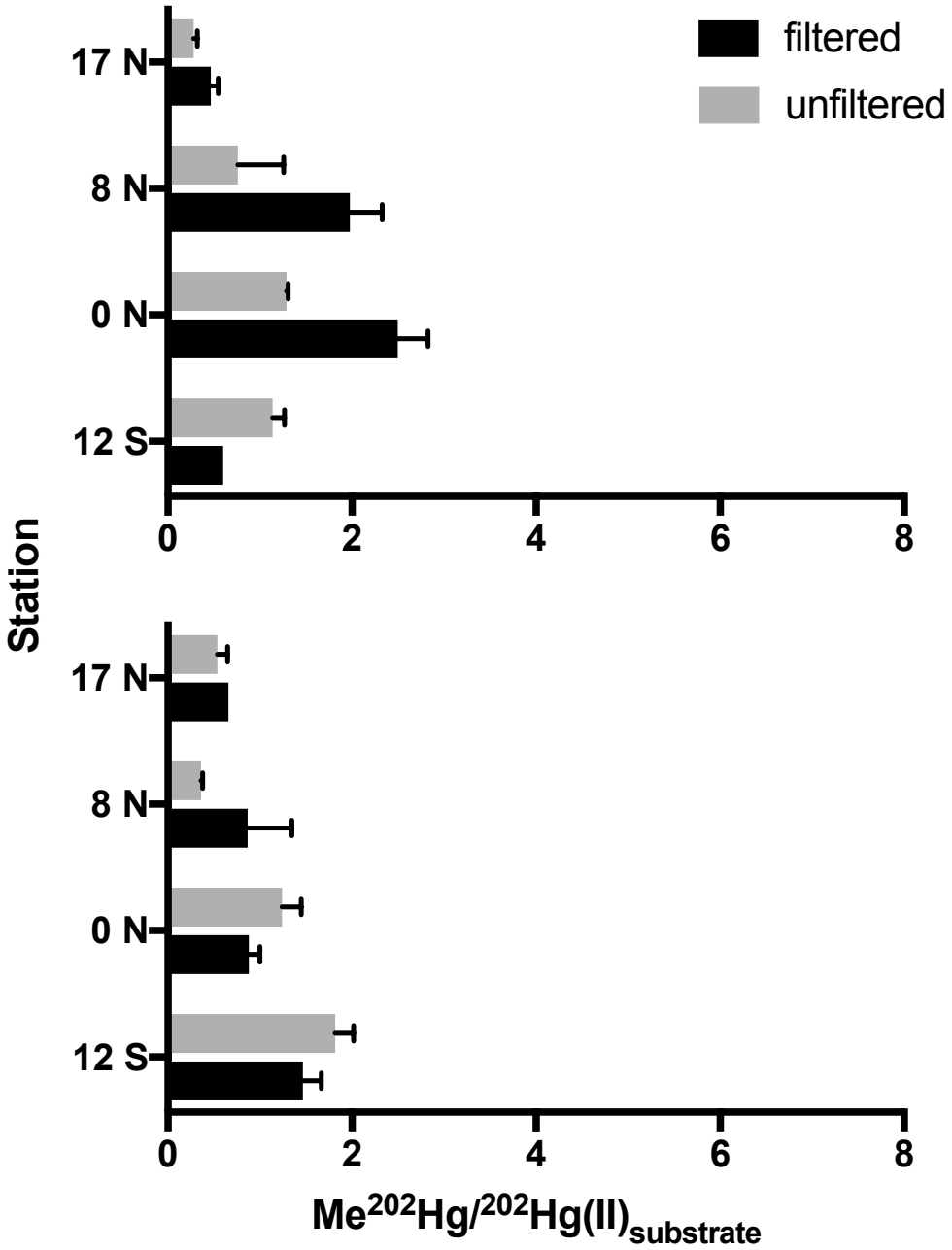


Figure S2: Calculated Hg(II) from full dissolved Hg speciation of Tropical Pacific waters.
Concentrations of Hg(II) as calculated according to Eq. (1) in the main text from full dissolved Hg speciation measurements (THg, Hg(0), MMHg, and DMHg) along the cruise transect.

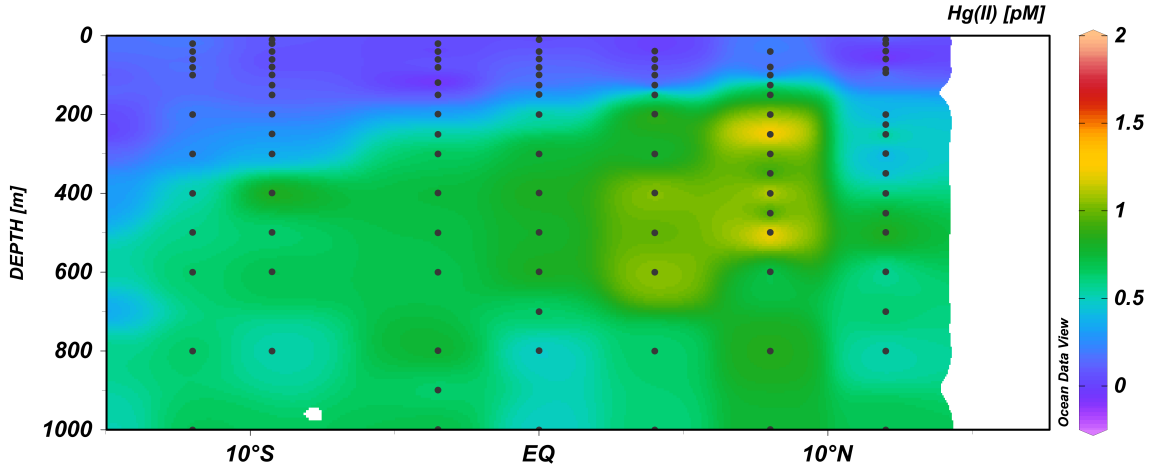


Figure S3: Organic matter remineralization rates from incubation stations. Organic carbon remineralization rates (OCRR) calculated for incubation stations between 17 °N and 12 °S and reported by Munson et al., (2015). All CFC ages provided by John Bullister.

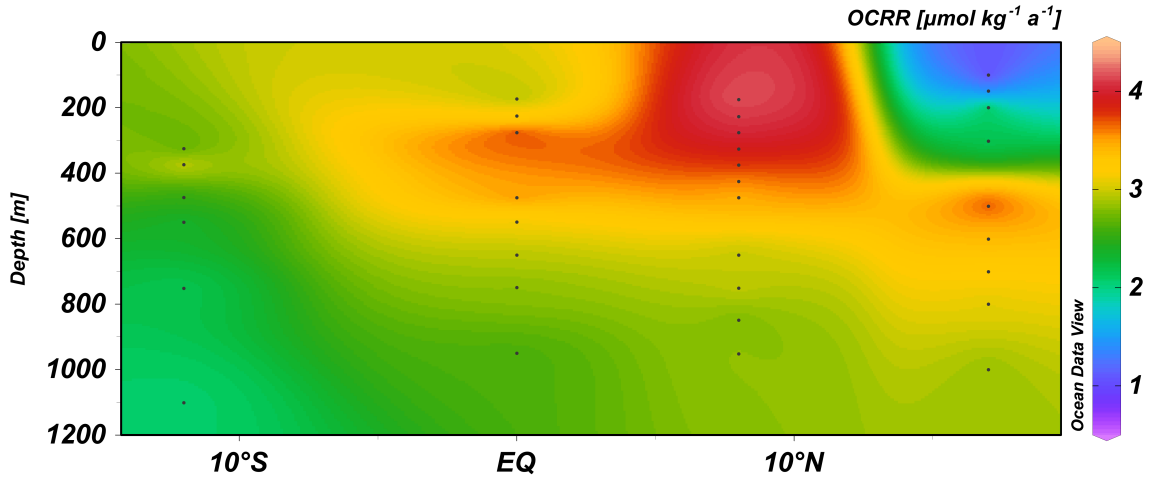
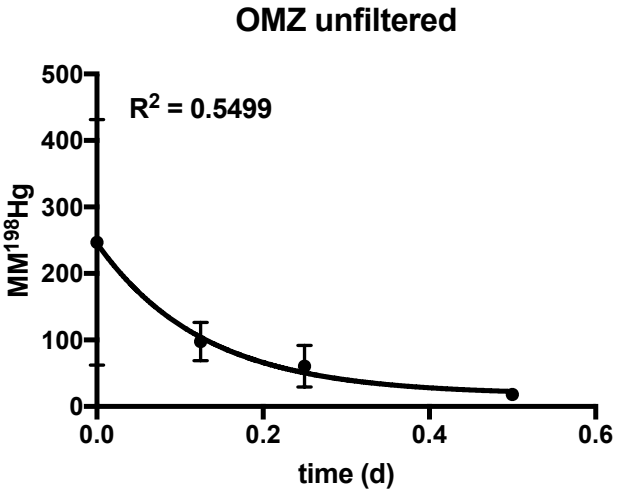


Figure S4: First order exponential decay of MeHg. Exponential decay of MeHg in incubations of Tropical Pacific water at the 12° S station.



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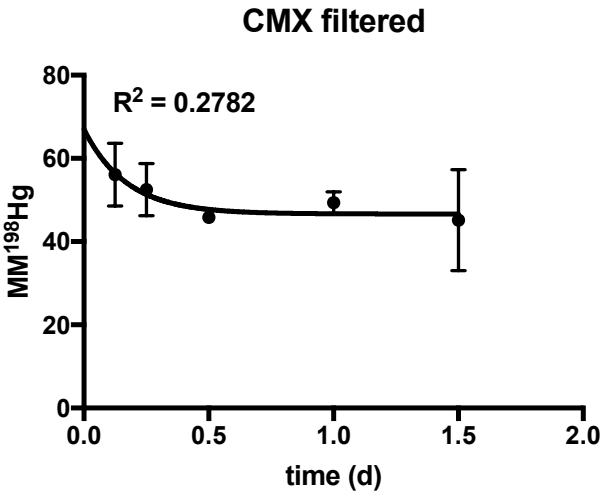

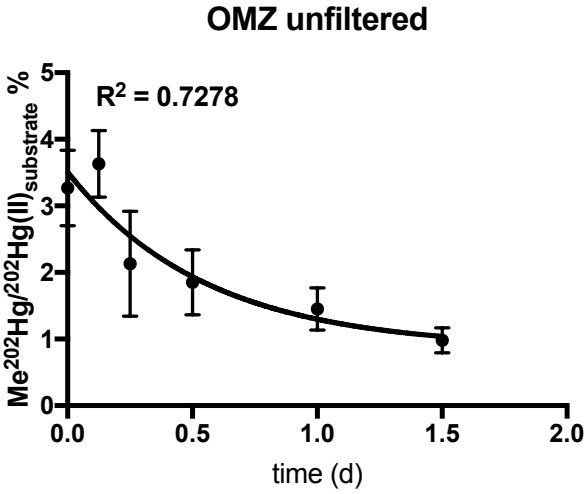

Table S1: Suspended particulate matter total Hg (THg) added as punch treatments to incubations.

Total mercury (Hg) added to amended incubation bottles from filtered particulate matter collection with in situ pumps (14).

Station	Hg added in filter punch (pmol)
17 °N	0.70
8 ° N	1.05
0 °	0.53
12 °S	0.35

Table S2: Methylation and demethylation in Pacific Ocean water traced with isotope enriched spikes of ^{202}Hg(II) and MM^{198}Hg.

Table S2a: Station 17 °N

					Me^{200}Hg traced from isotope tracers (fM)					
					Me^{198}Hg		^{202}Hg(II)		% ^{202}Hg(II) substrate	
Station	Depth	Filtered/ unfiltered	Timepoint (h)	Treatment	Averages	St Dev	Averages	St Dev	Averages	St Dev
1	CMX	filtered	0	C	31.90	0.30	4.13	0.75	0.61	0.17
1	CMX	filtered	24	C	32.58	4.59	2.32	0.65	0.62	0.17
1	CMX	filtered	0	Co	59.02	23.88	6.62	1.07	0.90	0.18
1	CMX	filtered	24	Co	40.17	9.42	3.58	0.45	0.88	0.08
1	CMX	filtered	0	CoC	38.82	7.65	4.29	3.12	1.16	0.43
1	CMX	filtered	24	CoC	40.92	9.65	4.36	2.15	0.85	0.06
1	CMX	filtered	0	punch	25.97	21.81	2.24	2.66	0.27	0.10
1	CMX	filtered	24	punch	22.29	7.63	1.39	0.07	0.34	0.03
1	CMX	filtered	0	none	57.49	17.67	3.79	2.09	0.47	0.08
1	CMX	filtered	24	none	53.75	3.46	4.88	0.32	0.69	0.04
1	CMX	unfiltered	0	C	31.41	4.70	3.42	0.31	0.79	0.08
1	CMX	unfiltered	24	C	33.67	8.25	2.46	0.34	0.50	0.06
1	CMX	unfiltered	0	Co	58.81	1.39	5.60	0.51	0.77	0.07
1	CMX	unfiltered	24	Co	39.44	2.96	5.14	2.82	0.64	0.09
1	CMX	unfiltered	0	CoC	35.46	3.59	4.30	0.42	1.00	0.29
1	CMX	unfiltered	24	CoC	36.30	5.79	3.72	1.94	0.86	0.02
1	CMX	unfiltered	0	punch	29.48	5.12	2.00	0.28	0.61	0.05
1	CMX	unfiltered	24	punch	32.07	4.74	2.03	0.98	0.47	0.16
1	CMX	unfiltered	0	none	42.82	0.85	2.49	0.85	0.28	0.04
1	CMX	unfiltered	24	none	49.34	4.47	4.64	0.41	0.69	0.03
1	OMZ	filtered	0	C	81.56	7.07	7.10	0.62	0.60	0.02
1	OMZ	filtered	24	C	33.56	5.14	4.21	2.30	1.30	0.57
1	OMZ	filtered	0	Co	111.54	9.21	7.18	1.26	0.53	0.05
1	OMZ	filtered	24	Co	161.68	19.44	13.78	3.50	0.52	0.14
1	OMZ	filtered	0	punch	59.26	7.80	4.33	0.61	0.55	0.14
1	OMZ	filtered	24	punch	16.83	16.77	3.38	5.99	0.20	0.07
1	OMZ	filtered	0	none	122.36	13.06	15.33	2.12	0.66	0.05
1	OMZ	filtered	24	none	113.75	56.60	16.48	4.29	1.10	0.09
1	OMZ	unfiltered	0	C	70.16	13.96	6.71	1.75	0.56	0.06
1	OMZ	unfiltered	24	C	38.71	21.61	5.96	2.78	0.75	0.44
1	OMZ	unfiltered	0	Co	205.83	27.46	12.63	13.20	0.49	0.51
1	OMZ	unfiltered	24	Co	144.14	130.18	16.35	16.42	0.58	0.24
1	OMZ	unfiltered	0	punch	51.34	7.80	4.28	1.78	0.55	0.22
1	OMZ	unfiltered	24	punch	9.27	7.72	0.16	0.20	0.08	0.08
1	OMZ	unfiltered	0	none	149.57	35.19	20.18	12.40	0.54	0.11
1	OMZ	unfiltered	24	none	267.77	29.71	50.68	1.88	0.74	0.06

Table S2b: Station 8 °N

					$Me^{200}Hg$ traced from isotope tracers (fM)					
					$Me^{198}Hg$		$^{202}Hg(II)$		% $^{202}Hg(II)$ substrate	
Station	Depth	Filtered/ unfiltered	Timepoint (h)	Treatment	Averages	St Dev	Averages	St Dev	Averages	St Dev
3	CMX	filtered	0	punch	25.48	3.29	2.35	0.89	2.04	0.63
3	CMX	filtered	24	punch	11.37	2.48	-1.06	0.02	-0.41	1.51
3	CMX	filtered	0	none	52.04	8.43	9.62	9.29	1.98	0.76
3	CMX	filtered	24	none	59.46	6.34	5.71	0.29	1.71	0.29
3	CMX	unfiltered	0	punch	15.32	6.20	0.87	0.91	0.99	0.52
3	CMX	unfiltered	24	punch	12.93	6.00	-2.61	4.09	-0.43	1.17
3	CMX	unfiltered	0	none	34.50	10.08	-0.99	0.89	0.76	0.50
3	CMX	unfiltered	24	none	10.22	5.13	1.52	0.74	0.99	0.12
3	OMZ	filtered	0	punch	-2.29	1.29	-21.28	0.31	-0.54	0.66
3	OMZ	filtered	24	punch	-0.43	0.68	-8.38	2.80	-0.41	0.43
3	OMZ	filtered	0	none	5.03	0.45	8.24	13.89	0.87	0.48
3	OMZ	filtered	24	none	1.09	4.10	11.78	6.81	4.21	1.79
3	OMZ	unfiltered	0	punch	-1.29	1.03	-9.96	7.27	-0.52	0.13
3	OMZ	unfiltered	24	punch	-0.39	0.69	-10.19	1.15	-1.50	0.45
3	OMZ	unfiltered	0	none	13.64	0.90	9.12	0.19	0.36	0.02
3	OMZ	unfiltered	24	none	3.57	4.29	1.13	1.62	0.73	0.82

Table S2c: Station 0 °N

					$Me^{200}Hg$ traced from isotope tracers (fM)					
					$Me^{198}Hg$		$^{202}Hg(II)$		% $^{202}Hg(II)$ substrate	
Station	Depth	Filtered/ unfiltered	Timepoint (h)	Treatment	Averages	St Dev	Averages	St Dev	Averages	St Dev
5	CMX	filtered	0	C	28.32	6.47	28.28	9.81	3.48	0.73
5	CMX	filtered	24	C	25.79	2.25	21.84	3.05	2.53	0.25
5	CMX	filtered	0	Co	21.33	7.18	16.00	1.76	2.32	0.01
5	CMX	filtered	24	Co	37.86	7.69	20.34	6.58	2.64	0.32
5	CMX	filtered	0	CoC	22.11	1.22	11.86	1.25	1.65	0.10
5	CMX	filtered	24	CoC	23.05	0.68	31.04	25.07	2.56	0.33
5	CMX	filtered	0	punch	13.57	1.76	8.58	1.89	1.24	0.02
5	CMX	filtered	24	punch	10.81	0.89	7.77	2.14	0.58	0.10
5	CMX	filtered	0	none	97.71	10.44	55.85	14.87	2.19	0.28
5	CMX	filtered	24	none	116.51	12.02	92.75	3.58	2.15	0.08
5	CMX	unfiltered	0	C	18.80	6.85	4.45	2.04	1.06	0.08
5	CMX	unfiltered	24	C	14.96	3.74	6.15	1.52	1.45	0.46
5	CMX	unfiltered	0	Co	36.08	0.67	11.92	2.58	1.16	0.19
5	CMX	unfiltered	24	Co	42.35	9.41	14.62	2.03	1.36	0.14
5	CMX	unfiltered	0	CoC	6.20	2.05	3.59	1.35	0.72	0.17
5	CMX	unfiltered	24	CoC	22.71	4.89	10.75	2.45	1.66	0.24
5	CMX	unfiltered	0	punch	14.17	7.67	5.93	3.21	0.72	0.00
5	CMX	unfiltered	24	punch	42.24	4.92	20.30	2.07	0.70	0.07
5	CMX	unfiltered	0	none	66.91	13.12	20.33	2.49	0.98	0.03
5	CMX	unfiltered	24	none	71.38	0.01	30.91	7.10	1.54	0.34
5	OMZ	filtered	0	C	5.67	1.54	6.77	1.57	1.19	0.06
5	OMZ	filtered	24	C	5.77	4.43	9.55	8.16	1.10	0.21
5	OMZ	filtered	0	Co	16.60	2.40	15.92	3.46	1.08	0.13
5	OMZ	filtered	24	Co	19.15	1.02	22.14	1.63	1.42	0.00
5	OMZ	filtered	0	CoC	14.74	1.69	15.42	5.62	0.91	0.26
5	OMZ	filtered	24	CoC	13.99	2.15	17.56	2.42	0.25	0.36
5	OMZ	filtered	0	punch	3.30	1.31	4.92	2.46	0.56	0.05
5	OMZ	filtered	24	punch	5.87	0.47	5.76	2.61	1.63	0.07
5	OMZ	filtered	0	none	24.30	0.49	15.30	3.64	0.67	0.08
5	OMZ	filtered	24	none	16.72	1.51	15.75	2.19	0.88	0.19
5	OMZ	unfiltered	0	C	6.88	1.46	7.68	2.20	0.53	0.04
5	OMZ	unfiltered	24	C	12.43	0.44	14.09	0.99	1.11	0.02
5	OMZ	unfiltered	0	Co	14.05	0.80	13.06	0.94	0.77	0.03
5	OMZ	unfiltered	24	Co	4.27	1.60	6.25	0.76	1.47	0.14
5	OMZ	unfiltered	0	CoC	7.34	1.83	12.22	2.04	1.16	0.21
5	OMZ	unfiltered	24	CoC	7.27	0.80	9.48	2.26	1.79	0.12
5	OMZ	unfiltered	0	punch	9.53	2.41	7.36	1.84	0.33	0.25
5	OMZ	unfiltered	24	punch	4.11	1.33	4.73	1.12	0.77	0.30
5	OMZ	unfiltered	0	none	20.22	2.10	19.96	7.42	0.87	0.16
5	OMZ	unfiltered	24	none	22.96	0.85	28.70	2.36	1.49	0.35

Table S2d: Station 12 °S

					$Me^{200}Hg$ traced from isotope tracers (fM)					
					$Me^{198}Hg$		$^{202}Hg(II)$		% $^{202}Hg(II)$ substrate	
Station	Depth	Filtered/ unfiltered	Timepoint (h)	Treatment	Averages	St Dev	Averages	St Dev	Averages	St Dev
9	CMX	filtered	0	none	46.92	1.30	4.69	0.66	0.60	0.01
9	CMX	filtered	3	none	56.11	7.56	6.80	1.51	0.79	0.00
9	CMX	filtered	6	none	52.49	6.30	6.55	1.33	0.91	0.12
9	CMX	filtered	12	none	45.81	0.76	3.68	0.15	0.36	0.11
9	CMX	filtered	24	none	49.35	2.62	7.04	3.02	0.80	0.28
9	CMX	filtered	36	none	45.15	12.18	3.02	0.87	0.36	0.21
9	CMX	unfiltered	0	C	47.46	15.91	9.74	7.91	0.78	0.20
9	CMX	unfiltered	3	C	57.45	10.53	9.95	3.11	0.72	0.02
9	CMX	unfiltered	6	C	43.05	1.54	7.27	3.16	0.87	0.16
9	CMX	unfiltered	12	C	40.92	3.33	5.83	0.95	0.60	0.16
9	CMX	unfiltered	24	C	25.90	6.14	4.36	0.44	0.55	0.05
9	CMX	unfiltered	0	Co	37.79	6.64	4.11	0.65	0.57	0.07
9	CMX	unfiltered	3	Co	59.06	1.69	9.99	2.00	0.89	0.19
9	CMX	unfiltered	6	Co	36.49	3.48	8.32	4.46	0.92	0.27
9	CMX	unfiltered	12	Co	22.48	4.50	2.86	0.13	0.36	0.04
9	CMX	unfiltered	24	Co	48.84	17.13	4.55	3.58	0.39	0.41
9	CMX	unfiltered	36	Co	20.61	6.43	3.15	0.30	0.55	0.07
9	CMX	unfiltered	0	CoC	35.08	0.51	3.77	0.64	0.58	0.13
9	CMX	unfiltered	3	CoC	44.52	6.18	6.85	0.68	0.71	0.07
9	CMX	unfiltered	6	CoC	47.02	5.46	6.00	0.63	0.55	0.21
9	CMX	unfiltered	12	CoC	31.45	34.19	13.90	18.00	1.14	0.71
9	CMX	unfiltered	24	CoC	23.91	4.58	3.86	0.60	0.47	0.17
9	CMX	unfiltered	36	CoC	36.80	0.35	7.04	0.31	0.89	0.14
9	CMX	unfiltered	0	punch	55.76	9.84	8.45	1.22	1.06	0.16
9	CMX	unfiltered	3	punch	31.54	3.05	3.51	1.15	0.55	0.18
9	CMX	unfiltered	6	punch	37.59	4.96	3.08	1.23	0.37	0.14
9	CMX	unfiltered	12	punch	54.08	38.78	4.05	2.43	0.53	0.02
9	CMX	unfiltered	24	punch	17.01	14.48	2.29	1.36	0.40	0.17
9	CMX	unfiltered	36	punch	40.51	29.12	2.79	0.93	0.49	0.37
9	CMX	unfiltered	0	none	52.98	6.65	7.95	1.14	1.14	0.13
9	CMX	unfiltered	3	none	55.61	3.80	9.33	1.13	1.10	0.12
9	CMX	unfiltered	6	none	57.84	1.38	10.81	2.92	1.55	0.37
9	CMX	unfiltered	12	none	52.16	2.08	5.71	2.83	0.72	0.20
9	CMX	unfiltered	24	none	59.34	15.51	9.81	2.81	0.84	0.05
9	CMX	unfiltered	36	none	54.04	25.29	7.53	3.26	0.95	0.16
9	OMZ	filtered	0	none	44.04	42.69	14.51	6.47	1.47	0.18
9	OMZ	filtered	3	none	49.02	25.89	12.16	7.46	1.32	0.35
9	OMZ	filtered	6	none	55.94	14.91	18.84	4.71	2.12	0.05
9	OMZ	filtered	12	none	57.87	65.16	19.97	24.30	2.15	0.43
9	OMZ	filtered	24	none	42.98	1.77	12.47	1.82	2.66	0.70
9	OMZ	filtered	36	none	26.60	5.65	9.72	3.33	2.32	0.23
9	OMZ	unfiltered	0	C	58.93	18.23	26.42	11.78	2.37	0.74
9	OMZ	unfiltered	3	C	57.29	12.74	42.43	14.12	5.75	0.48

9	OMZ	unfiltered	6	C	43.77	18.65	17.22	6.86	2.45	0.52
9	OMZ	unfiltered	12	C	47.41	24.67	9.93	2.86	1.12	0.15
9	OMZ	unfiltered	24	C	54.08	13.70	12.03	2.81	1.15	0.11
9	OMZ	unfiltered	36	C	43.40	17.18	9.76	5.65	1.21	0.28
9	OMZ	unfiltered	0	Co	39.42	13.75	16.31	0.81	1.60	0.01
9	OMZ	unfiltered	3	Co	31.52	16.43	7.54	1.33	0.89	0.08
9	OMZ	unfiltered	6	Co	51.94	42.06	20.01	11.13	1.94	1.47
9	OMZ	unfiltered	12	Co	40.65	8.32	8.77	0.64	1.15	0.09
9	OMZ	unfiltered	24	Co	56.44	15.69	10.72	0.97	1.22	0.23
9	OMZ	unfiltered	36	Co	59.33	3.19	16.72	1.08	1.53	0.12
9	OMZ	unfiltered	0	CoC	53.72	18.57	33.54	3.00	3.27	0.57
9	OMZ	unfiltered	3	CoC	53.41	3.84	38.04	7.99	3.63	4.78
9	OMZ	unfiltered	6	CoC	29.83	1.95	28.73	3.26	2.13	0.79
9	OMZ	unfiltered	12	CoC	60.27	52.16	18.51	18.38	1.85	0.49
9	OMZ	unfiltered	24	CoC	88.73	11.99	18.61	1.17	1.45	0.32
9	OMZ	unfiltered	36	CoC	59.29	9.24	10.76	2.52	0.98	0.19
9	OMZ	unfiltered	0	punch	1.26	0.80	3.90	2.03	0.65	0.43
9	OMZ	unfiltered	3	punch	23.15	3.68	13.43	0.72	1.99	0.24
9	OMZ	unfiltered	6	punch	29.44	3.99	10.39	0.74	1.54	0.75
9	OMZ	unfiltered	12	punch	25.02	0.20	4.98	0.28	0.76	0.17
9	OMZ	unfiltered	24	punch	26.63	1.87	5.39	0.97	0.74	0.17
9	OMZ	unfiltered	36	punch	38.11	5.30	7.48	1.25	0.79	0.11
9	OMZ	unfiltered	0	none	246.74	184.70	30.90	16.89	1.82	1.85
9	OMZ	unfiltered	3	none	97.51	28.76	25.12	5.18	2.73	0.17
9	OMZ	unfiltered	6	none	60.80	31.45	21.20	8.58	2.71	0.70
9	OMZ	unfiltered	12	none	18.05	4.71	3.45	0.94	0.66	0.29
9	OMZ	unfiltered	24	none	65.95	16.53	13.68	4.96	1.30	0.21
9	OMZ	unfiltered	36	none	52.56	15.60	9.75	3.24	0.98	0.23
9	175	filtered	0	none	45.44	5.76	3.17	0.54	0.27	0.06
9	175	filtered	24	none	37.28	3.91	3.25	1.07	0.24	0.06
9	175	unfiltered	0	none	49.58	10.89	3.72	1.65	0.31	0.11
9	175	unfiltered	24	none	84.61	10.66	8.35	4.58	0.57	0.04

Matlab scripts to transform .txt output from Thermo Element 2 data files peak integration

chromcrunch.m

```
%function chromcrunch
%this version meant for use with an ICP method that looks for Hg-198, -199, -200 and -202.

% chl july 2012

%launch user interface to get the file names for data to process.
%downloaded from matlab exchange 12/2011
filelist=uigetfile('*.txt');

% get ready to write file later
filename = 'icpresults.txt';
fid = fopen(filename, 'w');

%loop to process each of the selected files
for w=1:length(filelist)

%import the data from an icp txt file. creates a structure with the first 5 header lines
%stored in field "textdata" and the counts for each isotope in the field "data".
%filename
   A=importdata(filelist{w},'\t',8);

%start writing the output file line-by-line. first with the filename and isotope...does it 7
%times for each of the 7 isotopes. Then, extracts the isotopes from the "data" field and
%prints them to the file, isotope by isotope %in the transposed direction (from left to
%right,
%instead of top to bottom.
   for i=1:7
      fprintf(fid, '%s', filelist{w});
      if i==1
         fprintf(fid, '\t%d', 196);
      elseif i==2
         fprintf(fid, '\t%d', 198);
      elseif i==3
         fprintf(fid, '\t%d', 199);
      elseif i==4
         fprintf(fid, '\t%d', 200);
      elseif i==5
         fprintf(fid, '\t%d', 201);
      elseif i==6
         fprintf(fid, '\t%d', 202);
```

```
155             else
                    fprintf(fid, '\t%d', 204);
                end

                x=i+1;
160             fprintf(fid, '\t%d', A.data(:,x)');
                fprintf(fid, '\n');
            end

        end
165
        %when done, closes the file.
        fclose(fid);
```

Matlab script for the integration of Hg0, MMHg, and HgII peaks from Thermo Element 2 data output following use of chromcrunch.m

chromint.m

```
%chromint
%chl july 2012

clear all;

%set the start and endpoints for integration here
srtpts=[50 125 325];
endpts=[120 300 450];

%determine peak widths
wdth=endpts-srtpts+1;

%import the combined results file
filename=uigetfile('*.txt');
A=importdata(filename);

% get ready to write file later
filename = 'icpintegrations.txt';
fid = fopen(filename, 'w');

fprintf(fid, 'Sample \t Isotope \t Hg(0) Area \t MMHg Area \t Hg(II) Area \t\n');

%smooth the data before integration
Asmooth=[];
windowSize=5;
for j=1:length(A.textdata)
   Asmooth(j,:)=filter(ones(1,windowSize)/windowSize,1,A.data(j,2:end));
end

for i=1:length(A.textdata)
   fprintf(fid, '%s', A.textdata{i});
   fprintf(fid, '\t%d', A.data(i,1));

   hgoarea=sum(Asmooth(i,srtpts(1):endpts(1)))-mean([Asmooth(i,srtpts(1))
Asmooth(i,endpts(1))])*wdth(1);
   mmhgarea=sum(Asmooth(i,srtpts(2):endpts(2)))-mean([Asmooth(i,srtpts(2))
Asmooth(i,endpts(2))])*wdth(2);
   hg2area=sum(Asmooth(i,srtpts(3):endpts(3)))-mean([Asmooth(i,srtpts(3))
Asmooth(i,endpts(3))])*wdth(3);

   fprintf(fid, '\t%d', hgoarea);
```

```
215         fprintf(fid, '\t%d', mmhgarea);
            fprintf(fid, '\t%d', hg2area);

            fprintf(fid, '\n');

220     end
```