Supplemental Information

Methylmercury dynamics in upper Sacramento Valley rice fields with low background soil mercury levels

10 pages in total, 3 figures, 7 Tables
Impact of husk on rice grain MeHg and THg concentrations

In this study we measured the concentration of MeHg and THg in rough rice:

\[
[Hg]_R = \frac{Hg_R}{M_R}
\]

Where \([Hg]_R\) is the concentration of either MeHg or THg in rough rice, \(Hg_R\) = mass of the Hg species measured in rough rice, and \(M_R\) is the total mass of rough rice. “Hg” will be used to represent either MeHg or THg throughout these calculations.

Previous studies measured concentrations in brown rice grain after the husk was removed:

\[
[Hg]_G = \frac{Hg_G}{M_G}
\]

Where \([Hg]_G\) is the concentration, \(Hg_G\) is the mass of Hg, and \(M_G\) is the total mass for brown rice grain with the husk removed.

MeHg and THg are present in both the grain and the husk so

\[
Hg_R = Hg_H + Hg_G
\]

Where \(Hg_H\) and \(Hg_G\) are the mass of Hg contained in the husk and grain respectively. Because \(Hg_R\) was measured and rice cannot contain a negative amount of Hg, the value of \(Hg_G\) must fall within a known range:

\[
0 \leq Hg_G \leq Hg_R
\]

Dividing by \(M_G\) to convert to concentration, and substituting Eq. 2 gives:

\[
0 \leq [Hg]_G \leq \frac{Hg_R}{M_G}
\]
The mass of rough rice includes the mass of the husk and the mass of the grain:

\[ M_R = M_H + M_G \]

where \( M_H \) = mass of husk. Grain is 80% of the total mass of rough rice (IRRI, 2016):

\[ M_G = 0.8 \times M_R \]

Substituting Eq into Eq gives:

\[ 0 \leq [\text{Hg}]_G \leq \frac{\text{Hg}_R}{0.8 \times M_R} \]

Finally, substitute Eq.1

\[ 0 \leq [\text{Hg}]_G \leq 1.25 \times [\text{Hg}]_R \]

Therefore, the MeHg and THg concentrations in brown rice in this study could not have been more than 25% higher than the values measured for rough rice. Previous research (Meng et al., 2010) has shown that MeHg concentrations are higher in brown rice than in the husk. If the same holds true in the present study, we can assume the MeHg concentration in brown rice grain is higher than the concentration in rough rice:

\[ [\text{MeHg}]_R \leq [\text{MeHg}]_G \leq 1.25 \times [\text{MeHg}]_R \]

THg concentrations are generally higher in the husk than in brown rice grain (Meng et al., 2010), so the measured THg concentration in rough rice is likely an upper bound on THg concentration in brown rice grain. Because MeHg is included in THg, the lowest possible MeHg concentration is a reasonable lower bound on THg concentration in brown rice:

\[ [\text{MeHg}]_R \leq [\text{THg}]_G \leq [\text{THg}]_R \]
Figure S1. Map showing the Sacramento Valley rice growing area in California. Points show locations of current (squares) and past (circles) field-scale studies of MeHg export from rice systems: 1) Butte (current study) and 2) Yolo (current study) in the primary rice growing region of the Sacramento Valley, and 3) Yolo Bypass (Windham-Myers et al., 2014a) and 4) Cosumnes River Preserve (Eagles-Smith et al., 2014) in the Sacramento-San Joaquin Delta (Delta). Oval indicates area of the landscape scale study (Tanner et al., 2017).
Figure S2. Hydrologic time series and sampling schedule for Butte (left) and Yolo (right) fields. a) and b) Water depth; c) and d) irrigation; e) and f) drainage (excluding drain events at harvest and early season drain at Yolo); and g) and h) precipitation. Soil sampling events are indicated by vertical dotted lines in a) and b). ST = Spring Tillage, HD = Harvest Drain, FT = Fall Tillage, FD = Fallow Drain. Irrigation water and drainage water were sampled on the dates indicated by points in the irrigation (c, d) and drainage (e, f) panels respectively.
Figure S3. Butte field soil MeHg and THg at four depth intervals. Samples were collected prior to draining the field at the end of the growing season (Harvest) and end of the fallow season (Fallow). Data are mean ± SD (n = 4).
Table S1. Comparison of studied fields.

<table>
<thead>
<tr>
<th>Study period</th>
<th>Butte</th>
<th>Yolo</th>
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<tr>
<td>Field size (ha)</td>
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<td>48.6</td>
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<tr>
<td>Irrigation water source</td>
<td>Lake Oroville</td>
<td>Sacramento River and recycled</td>
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Growing season management

<table>
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<tr>
<th>Rice Variety</th>
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<th>M206</th>
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<tr>
<td>Planting date</td>
<td>May 16th</td>
<td>May 9th</td>
</tr>
<tr>
<td>Planting method</td>
<td>Water seeding</td>
<td>Water seeding</td>
</tr>
<tr>
<td>Early growing season</td>
<td>No outflow until July 10th</td>
<td>Early season drain May 12th</td>
</tr>
<tr>
<td>Maintenance flow period</td>
<td>July 10th – August 27th</td>
<td>July 1st – August 28th</td>
</tr>
<tr>
<td>Harvest date</td>
<td>September 23rd</td>
<td>September 21st</td>
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Fallow season management

<table>
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<tr>
<th>Straw management</th>
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</tr>
</thead>
<tbody>
<tr>
<td>Winter flood</td>
<td>October 28th – January 15th</td>
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</tbody>
</table>

Soil characteristics

<table>
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<tr>
<th></th>
<th>Butte</th>
<th>Yolo</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg ± SD (ng g⁻¹)</td>
<td>26 ± 6</td>
<td>56 ± 4</td>
</tr>
<tr>
<td>LOI ± SD (%)†</td>
<td>7 ± 1.4</td>
<td>8.6 ± 0.8</td>
</tr>
<tr>
<td>Clay (%)</td>
<td>50</td>
<td>58</td>
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</table>

† LOI = loss on ignition, a measure of organic matter content.
Table S2. Certified reference material analyses.

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<th>Analyte</th>
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<th>CRM Type</th>
<th>Certified Value†</th>
<th>Measured Value†</th>
<th>% Recovery</th>
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<td>0.616</td>
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<td>SQC-1238</td>
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<td>IAEA 140 Fucus</td>
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<td>27.3</td>
<td>91</td>
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† units are ng g⁻¹ for soil and plants, ng L⁻¹ for water
Table S3 Lab duplicates for MeHg and THg analyses.

<table>
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<tr>
<th>Analyte</th>
<th>Matrix</th>
<th>AVG†</th>
<th>DEV†</th>
<th>%DEV</th>
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<td>&lt;MDL</td>
<td>&lt;MDL</td>
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<td>0.049</td>
<td>0.006</td>
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<td>MeHg</td>
<td>Soil</td>
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<td>MeHg</td>
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<td>&lt;RL</td>
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† units are ng g⁻¹ for soil and plants, ng L⁻¹ for water
‡ MDL = 0.06, RL = 0.11 ng g⁻¹ for MeHg in plant tissues
<table>
<thead>
<tr>
<th>Analyte</th>
<th>Matrix</th>
<th>Non-Spiked Value†</th>
<th>Theoretical Value†</th>
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<th>% Recovery</th>
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</tr>
<tr>
<td>MeHg</td>
<td>Soil</td>
<td>0.03</td>
<td>0.93</td>
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<tr>
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</table>

† units are ng g⁻¹ for soil and plants, ng L⁻¹ for water
Table S5. MeHg and THg concentrations in field blank QA samples.

<table>
<thead>
<tr>
<th>Date</th>
<th>Field</th>
<th>Season</th>
<th>THg (ng L(^{-1}))</th>
<th>MeHg (ng L(^{-1}))</th>
</tr>
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<tbody>
<tr>
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<td>Butte</td>
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<td>&lt;0.1</td>
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</tr>
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<td>growing</td>
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<td>&lt;0.007</td>
</tr>
<tr>
<td>8/27/2014</td>
<td>Butte</td>
<td>growing</td>
<td>&lt;0.1</td>
<td>&lt;0.007</td>
</tr>
<tr>
<td>11/10/2014</td>
<td>Butte</td>
<td>fallow</td>
<td>0.3(^\dagger)</td>
<td>0.054(^\dagger)</td>
</tr>
<tr>
<td>12/28/2015</td>
<td>Yolo</td>
<td>fallow</td>
<td>&lt;0.1</td>
<td>&lt;0.007</td>
</tr>
<tr>
<td>2/17/2016</td>
<td>Yolo</td>
<td>fallow</td>
<td>&lt;0.1</td>
<td>&lt;0.007</td>
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</table>

\(^\dagger\) Outlet samples collected at the same time were > 10x higher than blank, and were replicated with low RPD (see Table S3). Inlet THg was 3.7 times higher than blank, whereas inlet MeHg was 0.011, (below the reporting limit of 0.02 ng L\(^{-1}\)).

Table S6. Relative percent deviation (RPD) of MeHg and THg in field duplicate samples.

<table>
<thead>
<tr>
<th>Date</th>
<th>Field</th>
<th>Season</th>
<th>Site</th>
<th>MeHg mean (ng L(^{-1}))</th>
<th>MeHg RPD</th>
<th>THg mean (ng L(^{-1}))</th>
<th>THg RPD</th>
</tr>
</thead>
<tbody>
<tr>
<td>5/16/2014</td>
<td>Butte</td>
<td>growing</td>
<td>canal</td>
<td>0.025</td>
<td>56.0</td>
<td>0.94</td>
<td>3.2</td>
</tr>
<tr>
<td>5/16/2014</td>
<td>Butte</td>
<td>growing</td>
<td>inlet</td>
<td>0.023</td>
<td>31.1</td>
<td>0.67</td>
<td>1.5</td>
</tr>
<tr>
<td>6/1/2014</td>
<td>Butte</td>
<td>growing</td>
<td>inlet</td>
<td>0.021</td>
<td>28.6</td>
<td>0.66</td>
<td>15.2</td>
</tr>
<tr>
<td>6/6/2014</td>
<td>Butte</td>
<td>growing</td>
<td>outlet</td>
<td>0.093</td>
<td>20.5</td>
<td>2.6</td>
<td>0.0</td>
</tr>
<tr>
<td>7/10/2014</td>
<td>Butte</td>
<td>growing</td>
<td>outlet</td>
<td>0.051</td>
<td>7.8</td>
<td>1.13</td>
<td>4.4</td>
</tr>
<tr>
<td>8/12/2014</td>
<td>Field 3</td>
<td>growing</td>
<td>outlet</td>
<td>0.020</td>
<td>0.0</td>
<td>0.19</td>
<td>0.0</td>
</tr>
<tr>
<td>8/28/2014</td>
<td>Butte</td>
<td>growing</td>
<td>outlet</td>
<td>0.0035(^\dagger)</td>
<td>28.6</td>
<td>0.07(^\dagger)</td>
<td>28.6</td>
</tr>
<tr>
<td>11/10/2014</td>
<td>Butte</td>
<td>fallow</td>
<td>outlet</td>
<td>0.86</td>
<td>3.7</td>
<td>8.8</td>
<td>0.5</td>
</tr>
<tr>
<td>1/4/2016</td>
<td>Yolo</td>
<td>fallow</td>
<td>outlet</td>
<td>0.18</td>
<td>5.4</td>
<td>17</td>
<td>10.8</td>
</tr>
</tbody>
</table>

\(^\dagger\) Concentration below reporting limit
Table S7. Mean ± SD soil MeHg and THg concentrations and pools in the plow layer (top 15 cm) by Field and sampling event.

<table>
<thead>
<tr>
<th>Field</th>
<th>Event</th>
<th>MeHg (ng g⁻¹)</th>
<th>THg (ng g⁻¹)</th>
<th>%MeHg †</th>
<th>MeHg (ng m⁻²)</th>
<th>THg (ng m⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Butte</td>
<td>Spring Tillage (May)</td>
<td>0.17 ± 0.05</td>
<td>22 ± 1.4</td>
<td>0.76 ± 0.25</td>
<td>22,000 ± 7,600</td>
<td>2,900,000 ± 270,000</td>
</tr>
<tr>
<td>Butte</td>
<td>Harvest Drain (August)</td>
<td>0.19 ± 0.06</td>
<td>28 ± 5.2</td>
<td>0.73 ± 0.26</td>
<td>24,000 ± 4,600</td>
<td>3,600,000 ± 950,000</td>
</tr>
<tr>
<td>Butte</td>
<td>Fall Tillage (October)</td>
<td>0.21 ± 0.01</td>
<td>28 ± 13</td>
<td>0.84 ± 0.25</td>
<td>27,000 ± 4,200</td>
<td>3,700,000 ± 2,100,000</td>
</tr>
<tr>
<td>Butte</td>
<td>Fallow Drain (February)</td>
<td>0.16 ± 0.04</td>
<td>24 ± 2.6</td>
<td>0.74 ± 0.15</td>
<td>23,000 ± 1,200</td>
<td>3,100,000 ± 500,000</td>
</tr>
<tr>
<td>Yolo</td>
<td>Spring Tillage (May)</td>
<td>0.49 ± 0.09</td>
<td>57 ± 1.2</td>
<td>0.87 ± 0.16</td>
<td>56,000 ± 11,000</td>
<td>6,400,000 ± 250,000</td>
</tr>
<tr>
<td>Yolo</td>
<td>Harvest Drain (August)</td>
<td>1.0 ± 0.22</td>
<td>57 ± 2.0</td>
<td>1.8 ± 0.36</td>
<td>120,000 ± 23,000</td>
<td>6,500,000 ± 190,000</td>
</tr>
<tr>
<td>Yolo</td>
<td>Fall Tillage (October)</td>
<td>0.69 ± 0.19</td>
<td>55 ± 8.0</td>
<td>1.2 ± 0.3</td>
<td>78,000 ± 22,000</td>
<td>6,300,000 ± 880,000</td>
</tr>
<tr>
<td>Yolo</td>
<td>Fallow Drain (February)</td>
<td>0.73 ± 0.33</td>
<td>57 ± 4.8</td>
<td>1.3 ± 0.6</td>
<td>84,000 ± 40,000</td>
<td>6,400,000 ± 490,000</td>
</tr>
</tbody>
</table>

† %MeHg = (MeHg / THg) *100