The Impact of Accounting for 3-D CO₂ Production on Inversion for Natural Fluxes
Using GOSSAT and In Situ Observations

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1. Introduction
Most atmospheric inversions for estimating natural carbon dioxide (CO₂) fluxes have placed CO₂ release from fossil fuel combustion and other sources entirely at the surface. However, a portion of fossil fuel and biospheric carbon emissions (≈1 Pg C yr⁻¹) occurs in the form of reduced carbon species including carbon monoxide (CO), methane (CH₄), and non-methane volatile organic compounds (NMVOCs), which are oxidized to CO₂ downstream of the emissions. Omission of this “chemical pump” can result in a shift in the distribution of the inferred fluxes, e.g., in the global sink from the tropics to the northern extratropics. A few inversion studies using surface CO₂ observations have accounted for the chemical pump, the most thorough analysis being conducted by Suntharalingam et al. [2005]. Nassar et al. [2010] presented a forward model analysis using advanced chemistry model output. We further examine the impact of accounting for the chemical pump on flux inversions, with the added dimension of considering satellite observation-based as well as surface in situ-based inversions. Our hypothesis is that there will be differing regional impacts in GOSSAT and in situ inversions due to differences in horizontal and vertical observational sampling. This study employs a relatively high spatiotemporal resolution Bayesian inversion approach, and atmospheric CO₂ production rates derived from a state-of-the-art NASA chemistry model historical simulation. Early results are presented below.

2. Methods
• Assimilated observations
  - In situ: Individual flask and afternoon-averaged continuous measurements from NOAA ESRL and JMA
  - GOSSAT: ACOS B3.4 retrieval of weighted column-average CO₂, filtered and bias-corrected (figure on the right)
• Prior constraints
  - Net ecosystem production (NEP) and fire fluxes from CASA-GFED v.3 model
  - Ocean fluxes from Takahashi et al. [2009]
  - Fossil CO₂ emissions from CDIAC
  - Atmospheric CO₂ production and surface correction
  - CO oxidation (+ CO₂ production) rates from NASA GEOS-5 GMI chemistry-climate model nudged to MERRA-2 reanalysis meteorology
  - Fossil fuel CO and NMVOCs: As initial estimate, assume a uniform 4.89% of fossil CO₂ as with Nassar et al. [2010]
  - Biomass and biofuel burning: Apply non-CO₂ emission factor from GFED averaged over ecosystem types
  - Biospheric CH₄: TransCom-CH₄, interannually varying emissions [Patra et al., 2011]
  - Biospheric NMVOCs: Initially, apply global NMVOC/CH₄ ratio from Nassar et al. [2010] uniformly to our biospheric CH₄ distribution
• Transport model
  - PCTM, with MERRA meteorology, 2° latitude x 2.5° longitude x 56 layers
  - Inversion technique [Wang et al., 2018, in review, ACP]
  - TransCom-style batch Bayesian inversion synthesis
• Optimize natural fluxes in 108 regions (map on the right) over 8-day intervals, and initial concentrations; high resolution minimizes aggregation error.

3. Forward Model Simulations
3-D CO₂ Production and Surface Correction Budgets

<table>
<thead>
<tr>
<th>Component</th>
<th>Global, Annual Total (Pg C/y)</th>
<th>2009-2010, this work</th>
<th>2006, Nassar et al. [2010]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total chemical production</td>
<td>1.15</td>
<td>1.05</td>
<td></td>
</tr>
<tr>
<td>Total surface correction</td>
<td>0.97</td>
<td>0.83</td>
<td></td>
</tr>
<tr>
<td>Fossil fuel combustion</td>
<td>0.41</td>
<td>0.38</td>
<td></td>
</tr>
<tr>
<td>Biomass and biofuel burning</td>
<td>0.23</td>
<td>0.00</td>
<td></td>
</tr>
<tr>
<td>Biospheric CH₄</td>
<td>0.16</td>
<td>0.28</td>
<td></td>
</tr>
<tr>
<td>Biospheric NMVOCs</td>
<td>0.16</td>
<td>0.16</td>
<td></td>
</tr>
</tbody>
</table>

(Note that Nassar et al. did not apply surface corrections for biomass and biofuel burning since they did not include non-CO₂ carbon emissions for those sources in their baseline CO₂ simulation)

4. Inversion Results

• CO₂ production is greatest where OH oxidant is most abundant, i.e., in the tropics, and secondarily where CO and VOC concentrations are highest.

5. Conclusions and Further Work
• Our analysis of the impact of 3-D CO₂ production and surface correction on flux inversions confirms small but possibly significant shifts in the global sink seen in a previous analysis
• In addition, we find that a GOSSAT inversion may be more sensitive to the ‘chemical pump’ in ocean regions than a surface observation-based inversion
• We will examine in more depth the impact of the chemical pump on posterior fluxes via column vs. surface observations
• We will also test an alternative fossil CO₂ emissions database, CDIAC, in place of CDIAC

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