Conclusions

- We use 5 different setups for OH abundances in a global CTM, all consistent with constraints on CH$_4$ lifetime and hemispheric [OH] based on MCF observations & constrain the CO sources in the model by using IASI CO columns in a source inversion framework.

- The top-down fluxes are sensitive to the setup: HTAPv2 $\pm 20\%$, biogenic $\pm 15\%$, but comparisons with a wide range of independent CO measurements (FTIR total columns, network surface mixing ratios, aircraft in situ data) $\rightarrow$ the inversion adopting the lowest average OH abundance in NH provides the best match with observations.

- In this best OH setup, the global top-down anthropogenic emissions are by 20% lower than in HTAPv2, by 30% lower in the US (supported by independent studies).

- Results suggest that the NH/SH OH ratio is likely lower ($\approx 0.85$) than the best estimate (0.97) of Patra et al. (2014), and that the global photochemical CH$_4$ lifetime might be longer ($\approx 12.5$ years) than the best estimate of Prather et al. (2012).
Purpose of this study

✓ Direct (incomplete combustion) and indirect (HC oxidation) CO sources
✓ CO + OH is a direct sink and CH₄ + OH is the main source → knowledge of OH fields is crucial!
✓ Modelling studies are largely dependent on the representation of OH in the model
✓ Average OH is overestimated by most models in NH → top-down CO emissions are likely too high
→ assess the global CO budget & account for uncertainties on [OH] based on observations

Methods

✓ Use prescribed OH fields satisfying MCF observations, 5 simulations each with different OH field

<table>
<thead>
<tr>
<th>Name</th>
<th>Setup</th>
<th>Global CH₄ lifetime</th>
<th>Interhemispheric ratio NH/SH OH</th>
<th>OH in NH (10⁵ molec/cm³)</th>
<th>SH OH (10⁵ molec/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>STD</td>
<td>Standard</td>
<td>11.2*</td>
<td>0.97**</td>
<td>9.2</td>
<td>9.5</td>
</tr>
<tr>
<td>NH-H</td>
<td>High OH in NH</td>
<td>9.8</td>
<td>1.10</td>
<td>11.2</td>
<td>10.1</td>
</tr>
<tr>
<td>NH-L</td>
<td>Low OH in NH</td>
<td>12.5</td>
<td>0.85</td>
<td>7.8</td>
<td>9.0</td>
</tr>
<tr>
<td>SH-H</td>
<td>High OH in SH</td>
<td>9.8</td>
<td>0.85</td>
<td>9.8</td>
<td>11.5</td>
</tr>
<tr>
<td>SH-L</td>
<td>Low OH in SH</td>
<td>12.5</td>
<td>1.10</td>
<td>8.8</td>
<td>8.0</td>
</tr>
</tbody>
</table>

* Best estimate for global CH₄ lifetime: 11.2±1.3 years (Prather et al. 2012)
** Models generally fail to reproduce the interhemispheric OH ratio derived from MCF analyses: interhemispheric N/S ratio of 0.97±0.12 (Patra et al. 2014)
Inverting for CO emissions

HTAPv2, Janssens-Maenhout et al. 2015

MEGAN-MOHYCAN
Stavrakou et al. 2014

GFED4s
Giglio et al. 2013

INPUT

Prior emission estimates $G_0(x, t)$

$G_0(x, t) = \sum_{j=1}^{J_2} \Phi_j(x, t)$

$G(x, t) = \sum_{j=1}^{J_2} \exp(f_j) \cdot \Phi_j(x, t)$

New Emission parameters $f$

Forward IMAGESv2

Modeled global concentrations $H_i(f)$

Cost function $J(f)$

$J(f) = \frac{1}{2} \sum (H_i(f) - y_i)^T E^{-1} (H_i(f) - y_i)^T$

$+ \frac{1}{2} (f - f_0)^T B^{-1} (f - f_0)$

Adjoint of IMAGESv2

Calculate $\nabla J(f)$ using the discrete adjoint of IMAGESv2

No minimum

OUTPUT

Improved set of emission parameters $f$ and emission estimates $G(x, t)$

INPUT

Observed global concentrations $y_i$

Updated fluxes


http://acsaf.org/products
Results

IASI

NH-L (using HTAPv2)

NH-L (top-down fluxes)

Anthropogenic Fires Isoprene

Flux updates

NH-H

NH-L

A priori STD NH-H SH-H SH-L

0 50 100 150 200 250 300 350 400 450 500 550

0 50 100 150 200 250 300 350 400 450 500 550

0 50 100 150 200 250 300 350 400 450 500 550

0 50 100 150 200 250 300 350 400 450 500 550

NH-H

NH-L

Anthropogenic Biomass Burning Biogenic

Anthropogenic Biomass Burning Biogenic
How do we evaluate the top-down results?

- **Comparisons with FTIR total columns (16 stations):** lowest bias achieved by NH-L inversion with lowest N/S (0.85) and longest CH$_4$ lifetime (12.5 yrs)

- **Comparisons with in situ (128 sites):** best performance realized by the NH-L simulation, worst by NH-H (larger spread than with FTIR)

- **Aircraft campaigns over US**

✓ Overall, the inversion using the lowest [OH] levels in NH achieves the best agreement simultaneously against all tested datasets of CO measurements

✓ US anthropogenic emissions derived in that inversion are consistent with previous estimates based on aircraft campaigns
Thanks to the (many!) data providers, the GAIA-Clim EU project for funding
and the GEIA organizing committee