Validation of CH₄ surface emission using forward chemistry-transport model


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Research Institute for Global Change, JAMSTEC
Yokohama, Japan
Introduction

Emission Inventory (EI) for MCF, CH$_4$, CO$_2$, SF$_6$…

Process-based model

Remote-sensing products

Site-based observation

Transport Processes
Meteorology
Radicals (OH, Cl)

Chemistry
Transport Model (CTM)

Model-Observation Comparison of CH$_4$

Validation of Transport (SF$_6$)

Validation of OH (CH$_3$CCl$_3$ - MCF)

Atmospheric Observations (site & satellite)

Multiple Tracers is key for Forward modeling and Validation research
Space-Time scales

Input parameter, e.g., Emission Inventory

Atmospheric Observables, e.g., Concentration

Connecting processes, e.g., Meteorology
ACTM transport validation: inter-hemispheric exchange time ($\tau_{ex}$)

\[
\tau_{ex} = \left[ \frac{\Delta c_{n-s} \left( \frac{E_n}{E_s} + 1 \right)}{\frac{E_n}{E_s}} \right] \left[ \frac{\frac{d c_s}{d t} - \frac{d c_n}{d t}}{E_n} \right]
\]

$E_n = \text{NH emission}$  
$E_s = \text{SH emission}$  
$c_n = \text{NH concentration}$  
$c_s = \text{SH concentration}$

<table>
<thead>
<tr>
<th>$\tau_{ex}$ Estimation Method</th>
<th>ACTM Based Mean±1σ</th>
<th>Observ. Based Mean±1σ</th>
<th>LH96 Mean</th>
<th>LG97 Mean</th>
<th>SD99 Range</th>
<th>BL04 Range</th>
<th>DR07 Range</th>
<th>AA08 Mean</th>
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<tbody>
<tr>
<td>Site based estimates</td>
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<tr>
<td>Case 1</td>
<td>1.75±0.03</td>
<td>1.86±0.16</td>
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<td>Case 2</td>
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<td>Case 3</td>
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<td>Whole Hemisphere Based Estimates</td>
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<td>Case 4</td>
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<td>0.76–1.97</td>
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<td>Case 5</td>
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<td>0.80–1.2</td>
<td>0.78–1.02</td>
<td>0.70</td>
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ACTM framework for CH$_4$ simulation  
(Patra et al., JMSJ, in acceptance)

- CCSR/NIES/FRCGC AGCM-based CTM (ACTM) run at resolution T42 L67 (top 90km)
- NCEP-2 reanalysis meteorology (U,V,T nudged)
- Hadley Center Sea-Surface Temperature & Sea-Ice Cover
- CH$_4$ chemistry (Sander et al., JPL Pub. 06-2, 2006) as:

  \[
  \text{CH}_4 + \text{O}^1\text{D} \rightarrow \text{Products (K}_{\text{O}} \text{D} = 1.5 \times 10^{-10})
  \]

  \[
  \text{CH}_4 + \text{OH} \rightarrow \text{CH}_3 + \text{H}_2\text{O (K}_{\text{OH}} = 2.45 \times 10^{-12} \exp(-1775/T)}
  \]

  \[
  \text{CH}_4 + \text{Cl} \rightarrow \text{CH}_3 + \text{HCl (K}_{\text{Cl}} = 7.3 \times 10^{-12} \exp(-1280/T)}
  \]

  \[
  \text{CH}_4 + \text{h} \nu \rightarrow \text{Products (wavelength dependent; not considered here)}
  \]

- All the radicals are taken from CHASER/STRAT (Sudo et al., Takigawa et al.) models at monthly (or hourly) intervals
The comparison for the recent times raises a couple of questions:
1. Is MCF emission decreasing since 2000 or so. If yes, at what rate and what is present day emission magnitude
2. Is the OH production in CHASER is higher than the real values? If so, at which latitudes, longitudes or heights
# Surface flux types and annual budget of CH$_4$

<table>
<thead>
<tr>
<th>Sources</th>
<th>Range Estim. Reported by IPCC [2001]</th>
<th>A Priori Estimates, Tg CH$_4$/yr</th>
<th>Year</th>
<th>Total emission (E2)</th>
<th>Tropospheric Budget (E2)</th>
<th>Year 2000 (E2)</th>
<th>Top emission country (E2)</th>
<th>Aggr. Emission (E2)</th>
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<td>Total wetlands</td>
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<td>Fossil fuel</td>
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<td>Industrial</td>
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<td>574.8</td>
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</tr>
</tbody>
</table>

*Mikaloff Fletcher et al., GBC, 2004*  
*Patra et al., JMSJ, 2009, in acceptance*
CH$_4$ emission, sinks, and concentration

(a) CH$_4$ flux (g-CH$_4$/m$^2$/mon): Jan. 2000
(b) CH$_4$ flux (g-CH$_4$/m$^2$/mon): Jul. 2000
(c) OH column (x1e9 molec/cm$^2$): Jan. 2000
(d) OH column
(e) CH$_4$ mixing ratio (ppb): Jan. 2000
(f) CH$_4$ mixing ratio

(b) Precipitation (shaded) and Winds: January
CH₄ lifetime and budgets

Instantaneous CH₄ Lifetime

\[ \text{Instantaneous CH}_4 \text{ Lifetime} = \frac{1}{[K_{OH} \cdot OH + K_{O}^{1}D \cdot O^{1}D + K_{Cl} \cdot Cl]} \]

(useful for understanding the dominance of dynamics vs. chemistry on variability)

Atmospheric Lifetime = burden/loss

(Prather et al., IPCC, 2001)

CH₄ L. T. = 4999 Tg/580 Tg yr⁻¹ = 8.62 years

<table>
<thead>
<tr>
<th>Estimates</th>
<th>Atmospheric Lifetime</th>
<th>References</th>
</tr>
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<tbody>
<tr>
<td>IPCC TAR</td>
<td>8.4</td>
<td>Prather et al.</td>
</tr>
<tr>
<td>IPCC FAR (prescribed)</td>
<td>8.67±1.32(m#26)</td>
<td>Stevenson et al., JGR, 2006</td>
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<tr>
<td></td>
<td>8.45±0.38 (m#12)</td>
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<tr>
<td>This work (full model)</td>
<td>8.62</td>
<td>Patra et al., JMSJ, 2009</td>
</tr>
</tbody>
</table>
CH$_4$ Measurement Sites – can we track emissions?
(~50 used here; >100s are in operation in 2007)

Contributing Institutes: 1. NOAA/ESRL, 2. FEA, Germany, 3. JMA, Japan, 4. EC, Canada, 5. NIWA
CH₄ Latitudinal gradients: seasonal and longitudinal variations

Seasonal cycle in the extra tropics/high latitudes is controlled by chemical loss and surface emission.
CH$_4$ Seasonal Cycles in the tropics – produced by chemical loss and dynamics (monsoon)
Preliminary comparison with AIRS/Aqua satellite retrievals

AIRS Precision: ~20 ppb
AIRS – ACTM = 20 ppb

E1: 80 Tg-CH₄ Rice emis
E2: 40 Tg-CH₄ Rice emis

Source: NOAA/NESDIS/STAR

80°E-110°E, 20°N -35°N; 250-350 mb
CH4 Synoptic variations arising from synoptic meteorology and concentration gradients.

(a) CGO  (b) MLO  (c) YON  (d) AMY  (e) NGL  (f) BRW

2002                         |                             2003                         |                             2004
GSN  CDL  AMY  BRW  THD  ALT  FRD  ZSF  RYO  NGL  SMO  DEU  MNM  YON  MLO  COI  RPB  ZGT  SCH  MHD  IZO  CGO  HAT

Measure of Synoptic peak shape / phase

Correlation (r)

Measure of Synoptic peak height

NSD (unitless)

Ideal~1.0

(σ_{obs}/σ_{mod})
CH$_4$ diurnal cycles: caused by local emission and PBL cycle
Conclusions

• ACTM CH₄ simulations have been optimized for a combinations of fluxes, radicals and transport
  – Examining seasonal cycles of IHGs at a ‘set’ of stations helped to prepare flux-types combinations
  – Very low (~5ppb) CH₄ longitudinal gradients between CRZ/CGO/EIC can be tracked
  – Seasonal cycle, synoptic variability and diurnal cycle; all needed to be checked for testing bottom-up fluxes

• The ACTM-AIRS comparison in the upper troposphere region reveal
  – A reversal of CH₄ seasonal cycle at about 400-300 mb height
  – Role of CH₄ transport by Asian monsoon during high surface emission period

• Both transport and chemistry components in ACTM have been validated independently using two different chemical tracers, SF₆ and MCF, respectively
Outlook

• Reduction in radical (e.g., OH) uncertainty by independent methodology
  – Can the MCF emission be available at more confidence? e.g., global/country total emission inventory for the users?
  – Time dependent gridded distribution of emissions

• Updating chemical (e.g., CH₄) emission distribution and strength
  – A concerted effort?, e.g., EDGAR gives annual mean emission due to rice cultivation
  – IAVs in wetland or other natural emissions? Validation strategy

• Increased use of remote sensing data for long-lived gases
  – Some issues persist with the product quality; more validation and comparison would help probing the causes
  – More dedicated satellite sensors, e.g., GOSAT, new-OCO?