Lagrangian model analysis of CARIBIC observations – quantifying transport, chemistry, and mixing

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Motivation

Flights 202 from Frankfurt to Guangzhou on 14/15 Aug 2007

back-trajectories for some CO peaks

Big thanks to KNMI campaign support!
Motivation

→ Interpret observations by modeling the history of observed air masses
→ Learn from deviations between model and observations
**Method**

- Trajectory box model **CAABA** re-simulates 3-D AC-GCM **EMAC** along trajectories.
- No exchange of air masses during trajectory chemistry calculation → no “mixing”

Riede et al, GMD, 2009
Method – single trajectory

- Completely based on modeling
- High consistency between trajectory-box model CAABA and 3-D model EMAC in transport and chemistry
- Use differences between models of a model hierarchy
- Individual for each species
- Dependent on trajectory travel time

![Diagram showing mixing ratio μ over trajectory time forward and flight track with comparison between CAABA/MJT and EMAC models.]

![Bar chart showing differences in ppb and ppt for O3, CO, NOx, and HCHO with transport, chemistry, and mixing contributions.]
CARIBIC flights

CARIBIC flights to Guangzhou and Manila 2005 – 2008
Flight 202: CO

Flight Frankfurt → Guangzhou on 14/15 August 2007

nmol/mol

CO

CARIBIC

EMAC @ flight track

AUG 15
Flight 202: CO

Flight Frankfurt → Guangzhou on 14/15 August 2007

ppbv

240
200
160
120
80
40

processes over the past 8 days before reaching the flight path
Flight 202: CO 8–0.5 days back

8 days back

5 days back

3 days back

2 days back

1 day back

½ day back
Flight 202: CO

8 days back

5 days back

3 days back
Transport FRA–CAN–MNL 5 days

flight track longitudes
- 0–50 Europe
- 50–80 Asian plains
- 80–100 Himalayas
- 100 – 110 China
- 110–130 South China Sea

histogram levels (%)

Oct – Apr

May – Sep
CO transport, chemistry, mixing (5d)

- net effect after 5 days, mapped onto starting points
- East Asia: important source area, chemical sink, net mixing input
O$_3$ transport and mixing

**Transport**

**Mixing**

Start latitude vs. latitude

ppbv vs. latitude

Latitude range: 15°N to 55°N

ppbv range: 0 to 1100 ppbv
NOy chemistry (5d)

- NOy usually “conserved” by almost quantitative NOx $\rightarrow$ HNO$_3$
- in (South) East Asia net source via chemistry $\rightarrow$ NH$_3$
### Lifetimes based on 5-day chemistry

<table>
<thead>
<tr>
<th>Substance</th>
<th>Chemically Active</th>
<th>Average</th>
<th>Reduction</th>
<th>Min</th>
<th>Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt; 600 hPa NO</td>
<td></td>
<td>(median)</td>
<td>by %</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NO</td>
<td>56 sec</td>
<td>30 (4) min</td>
<td>97</td>
<td>20 sec</td>
<td>3 days</td>
</tr>
<tr>
<td>HCHO</td>
<td>4 h</td>
<td>9 (6) h</td>
<td>55</td>
<td>3 h</td>
<td>6 days</td>
</tr>
<tr>
<td>NO2</td>
<td>4 h</td>
<td>18 (14) h</td>
<td>80</td>
<td>3 min</td>
<td>4 days</td>
</tr>
<tr>
<td>CH₃COCH₃</td>
<td>12 days</td>
<td>40 (20) days</td>
<td>70</td>
<td>8 days</td>
<td>3 years</td>
</tr>
<tr>
<td>CO</td>
<td>25 days</td>
<td>5 (3) months</td>
<td>80</td>
<td>12 days</td>
<td>20 years</td>
</tr>
<tr>
<td>O₃</td>
<td>15 days</td>
<td>450 (300) days</td>
<td>97</td>
<td>6 days</td>
<td>20 years</td>
</tr>
</tbody>
</table>

- **Photolysis, HOₓ:**
  - HCHO + hv → CO + H₂
  - HCHO → CO + H₂O₂
  - HCHO + O₃ → CO + H₂O + 2 O₂

- **and NOx chemistry**
  - CO + O₃ → O₂ + CO₂
  - CO + 2 O₂ → O₃ + CO₂

- **Identifying catalytic cycles**
- **Identifying important pathways**

R. Lehmann, J. Atmos. Chem., 2004
OH reactivity (OHR)

\[ \text{OHR} = \sum_i k_{OH} X_i [X_i] = 1/\tau_{OH} \]

<table>
<thead>
<tr>
<th>( \tau_{OH} \text{ (s)} )</th>
<th>OH reactivity (s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Min. 0.14</td>
<td>7.2</td>
</tr>
<tr>
<td>Q1 2.7</td>
<td>0.37</td>
</tr>
<tr>
<td>Median 4.7</td>
<td>0.21</td>
</tr>
<tr>
<td>Ave. 4.6</td>
<td>0.21</td>
</tr>
<tr>
<td>Q3 6.4</td>
<td>0.16</td>
</tr>
<tr>
<td>Max. 13.1</td>
<td>0.08</td>
</tr>
</tbody>
</table>
Typical OHR values

120  Mexico City morning rush hour [Shirley 2006, ACP]
80   Amazon rain forest [Noelscher, ORSUM meeting 2014]
30   stressed Finnish forest [Noelscher 2012, ACP]
25   Mexico City average daytime [Shirley 2006, ACP]
10   Mainz average in summer [Sinha 2008, ACP]
4    Asian pollution outflow, West-Pacific, INTEX-B [Mao 2009, ACP]
2.5  11.5 km above continental US [Xinrong Ren, ORSUM meeting 2014]
OH lifetime and trajectory start

OH reactivity ($s^{-1}$)

Trajectory start altitude (hPa)
OHR and trajectory ascent/descent

- OH reactivity (s⁻¹)
- End-start pressure (hPa)
CAABA in trajectory mode is an analytical tool to interpret 3-D model data from EMAC

additional information about the history of observed air masses

observation-model comparisons → why do differences occur, e.g., relatively coarse model resolution → often overestimated mixing in EMAC

(NOy chemical input above (South) East Asia by NH$_3$)

hot spots of chemistry cause exceptional variations in chemical lifetimes

OH reactivity values very low, but well reproducible, CO plays major role in the troposphere, O$_3$ in stratosphere

highest OH reactivity in ascending air parcels (pressure-related)
Thank you!

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Observations → Trajectories → Qualitative

Trajectories → EMAC → Quantitative Understanding of Data

Quantitative Understanding of Model Deficits

Modeling
Method

- Consistent model hierarchy
- CAABA: no “mixing” (3-D exchange) such as scavenging, or emissions
Method

Riede et al, GMD, 2009
CO mixing and chemistry (5d)

CO in ppbv at start points

CO production small over 5 days, up to 15 ppbv in 5 days for individual trajectories
O₃ transport and chemistry
Guangzhou + Manila 2005-2008

flight altitude – start altitude (hPa)
Guangzhou + Manila 2005-2008

OH lifetime (s)

trajectory starting positions