Changes in tropospheric chemistry and their impacts on climate: roles of climate change and the stratosphere

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The 4\textsuperscript{th} SPARC General Assembly
Bologna, Italy, 31 Aug. -- 5 Sep. 2008
Radiative Forcing in the 20th century

IPCC-AR4: CCSR/NIES/FRCGC GCM (MIROC)

(a) Radiative Forcing

- LLGHG
- Ozone (troposphere)
- Ozone (stratosphere)
- Aerosol Direct
- Aerosol 1st Indirect
- Aerosol 1st+2nd Indirect
- Volcanic Eruptions
- Solar
- Land Use
- Total

(b) Surface Forcing

- LLGHGs
- Ozone (troposphere)
- Ozone (stratosphere)
- Aerosol Direct
- Aerosol 1st Indirect
- Aerosol 1st+2nd Indirect
- Volcanic Eruptions
- Solar
- Land Use
- Total

Air pollution / Atmospheric Chem.

Takemura et al [2006]
4th SPARC-GA in Bologna 2008  (Sudo K.)  

Changes in tropo. chem and their impacts on climate

The AC&C Initiative

Atmospheric chemistry (Air Quality) ↔ Climate Interaction

Hemispheric Transport of Air Pollution

**WCRP-SPARC/IGBP-IGAC**  
Atmospheric Chemistry & Climate Initiative

**Unifying Thematic Areas**
a) Composition impacts on climate  
b) Climate impacts on chemistry  
c) Climate impacts on surface-level ozone & aerosols  
("air quality")

**AC&C Research Activities**
1) 20 year hindcast for tropospheric gases/aerosols  
2) What controls the distribution of tropospheric aerosols/gases? (Step 1: Focus on 5km to tropopause distribution)  
3) Cloud/aerosol/chemical interactions  
4) Future scenarios: sensitivities & uncertainties

**Cross-Cutting Activities**
1) Emissions Harmonization Committee  
2) Data Center Committee  
3) AC&C Web Page and "E-newsletter"

**Research Implementation Bodies**
- CCMVal (stratospheric chemistry)  
- AeroCom (tropospheric aerosols)  
- "TropChem" (tropospheric gas-phase chem)  
- TF HTAP  
- GEIA  
- Observational Data  
- ACCENT
Contributions to Annual Mean Tropospheric Column O$_3$ (TCO)

**Tropospheric Origin**

- a) TROPO (POLTD + REMOT) \( <\text{MM}=\text{Ann} > \)
- c) POLTD \( <\text{MM}=\text{Ann} > \)

**Stratospheric Origin**

- b) STRAT \( <\text{MM}=\text{Ann} > \)
- d) REMOT \( <\text{MM}=\text{Ann} > \)

*<Production in polluted regions>*

*<Production in remote regions>*

Sudo and Akimoto (2007)
Tropospheric Column Ozone (TCO) seasonal-annual variation

TCO (DU) model

1990 1995 2000

TCO tendency (Tg yr-1) model

N.H. 30N-50N
### Global Budgets of Tropospheric O₃ from Distinct Source Regions

**Table 3.** Global Budget of Tropospheric O₃ from Distinct Source Regions.

<table>
<thead>
<tr>
<th>Tracer ID</th>
<th>Chem. production</th>
<th>Tropo. Burden (TgO₃)</th>
<th>Lifetime (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pᵇ</td>
<td>P – Lᶜ</td>
<td>Global (%)</td>
</tr>
<tr>
<td>O₃-ALL</td>
<td>4744</td>
<td>444.0</td>
<td>344.6 (100.0%)</td>
</tr>
<tr>
<td>STRAT</td>
<td>0</td>
<td>-484.1</td>
<td>77.9 (22.6%)</td>
</tr>
<tr>
<td>REMO1</td>
<td>1735</td>
<td>338.7</td>
<td>101.0 (29.3%)</td>
</tr>
<tr>
<td>POLTD</td>
<td>3010</td>
<td>589.5</td>
<td>165.7 (48.1%)</td>
</tr>
<tr>
<td>BL-AMN</td>
<td>162</td>
<td>55.4</td>
<td>7.0 (2.0%)</td>
</tr>
<tr>
<td>BL-AMM</td>
<td>162</td>
<td>29.8</td>
<td>6.6 (1.9%)</td>
</tr>
<tr>
<td>BL-AMS</td>
<td>203</td>
<td>49.6</td>
<td>7.4 (2.1%)</td>
</tr>
<tr>
<td>BL-AFN</td>
<td>166</td>
<td>33.1</td>
<td>5.4 (1.6%)</td>
</tr>
<tr>
<td>BL-AFS</td>
<td>175</td>
<td>38.8</td>
<td>6.6 (1.9%)</td>
</tr>
<tr>
<td>BL-EUR</td>
<td>116</td>
<td>49.4</td>
<td>4.5 (1.3%)</td>
</tr>
<tr>
<td>BL-CEU</td>
<td>76</td>
<td>31.3</td>
<td>3.0 (0.9%)</td>
</tr>
<tr>
<td>BL-MES</td>
<td>92</td>
<td>22.3</td>
<td>3.2 (0.9%)</td>
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<tr>
<td>BL-IND</td>
<td>97</td>
<td>20.9</td>
<td>3.9 (1.1%)</td>
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<tr>
<td>BL-TLD</td>
<td>50</td>
<td>7.8</td>
<td>2.2 (0.6%)</td>
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<tr>
<td>BL-CHN</td>
<td>128</td>
<td>34.6</td>
<td>5.9 (1.7%)</td>
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<tr>
<td>BL-JPN</td>
<td>22</td>
<td>6.4</td>
<td>1.1 (0.3%)</td>
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<tr>
<td>BL-IDN</td>
<td>61</td>
<td>9.7</td>
<td>2.8 (0.8%)</td>
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<tr>
<td>BL-AUS</td>
<td>88</td>
<td>19.7</td>
<td>3.7 (1.1%)</td>
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<tr>
<td>FT-AMN</td>
<td>167</td>
<td>29.6</td>
<td>11.9 (3.4%)</td>
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<td>FT-AMS</td>
<td>295</td>
<td>32.9</td>
<td>21.0 (6.1%)</td>
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<tr>
<td>FT-AFN</td>
<td>203</td>
<td>21.2</td>
<td>13.4 (3.9%)</td>
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<td>FT-AFS</td>
<td>186</td>
<td>21.6</td>
<td>13.1 (3.8%)</td>
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<tr>
<td>FT-EUR</td>
<td>60</td>
<td>15.9</td>
<td>4.2 (1.2%)</td>
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<tr>
<td>FT-ASA</td>
<td>325</td>
<td>39.7</td>
<td>24.9 (7.2%)</td>
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<tr>
<td>FT-IDN</td>
<td>90</td>
<td>8.9</td>
<td>6.9 (2.0%)</td>
</tr>
<tr>
<td>FT-AUS</td>
<td>86</td>
<td>11.0</td>
<td>6.9 (2.0%)</td>
</tr>
</tbody>
</table>

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**References:**
- Sudo K. (2007)
Tropospheric chemistry and its climate impacts -- roles of climate change and stratosphere?--

1. The impacts of global ozone changes on climate:
   + Evaluate climate equilibrium response
   + Tropo. O$_3$ increase and strato. O$_3$ decrease in the 20$^{th}$ century

2. Long/near term future projection of tropospheric ozone and related species (CH$_4$ / aerosols):
   + Impacts of climate change
   + Impacts of stratospheric O3 change

3. Summary
Experimental Setup

① Evaluate climate (equilibrium) responses to changes in “Tropo. O₃”, “Strato. O₃”, and “LLGHGs” from preindustrial times to the present.

② Past O₃ changes are reproduced with a global chemistry climate model CHASER (Sudo et al., 2006); stratospheric O₃ changes are expressed as a function of halogen loading as with the SPARC Ozone Trend Estimate for 1980–2000.

③ Run CCSR/NIES/FRCGC climate model (AGCM + simplified ocean model) for 50 years (30 years for analysis) X 6 ensembles.

### Run Scenarios

<table>
<thead>
<tr>
<th></th>
<th>Ctrl</th>
<th>L</th>
<th>LT</th>
<th>LTm</th>
<th>LTS</th>
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</thead>
<tbody>
<tr>
<td><strong>LLGHGs</strong></td>
<td>PI</td>
<td>PD</td>
<td>PD</td>
<td>PD</td>
<td>PD</td>
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<tr>
<td><strong>Tropo. O₃</strong></td>
<td>PI</td>
<td>PI</td>
<td>PD</td>
<td>PD*</td>
<td>PD</td>
</tr>
<tr>
<td><strong>Strato. O₃</strong></td>
<td>PI</td>
<td>PI</td>
<td>PI</td>
<td>PI</td>
<td>PD</td>
</tr>
</tbody>
</table>

(!) LLGHGs = CO₂ + CH₄ + N₂O + CFCs

(!) PI: preindustrial ~1850, PD: present day ~2000

Zonally averaged O₃ increases
### Chemistry-coupled climate model CHASER

**Base model**
CCSR/NIES/FRCGC GCM (5.7b)

**Resolution**
horizontal: T42(2.8°x2.8°), vertical: 32 layers (surface~40km)

**Transport**
Grid scale (flux-form semi-Lagrangian)  
Sub-grid scale (convection, vertical diffusion)

**Chemistry**
54 chemical species, 152 chemical reactions (gas, liquid, heterogeneous*)

- (1) \(O_3\)-HO\(_x\)-NO\(_x\)-CO-CH\(_4\)  
- (2) NMHCs oxidation, and  
- (3) \(SO_2\), DMS oxidation  

* heterogeneous reactions are considered for surface of cloud particles, sulfate, and sea-salt aerosols

- + Simplified stratospheric chemistry (no-PSCs)

**Emission**
Industry, biomass burning, vegetation/soil/ocean, lightning NOx  
(\(NO_x\), CO, \(C_2H_6\), \(C_2H_4\), \(C_3H_8\), \(C_3H_6\), acetone, isoprene, terpenes, \(SO_2\), DMS)

Lightning NOx is parameterized in the GCM convection[Price & Rind, 1992]

**Dry deposition**
Function of vegetation type, temperature, solar flux, snow cover  
[Wesely, 1989]

**Wet deposition**
Rain-out (in-cloud), wash-out (below-cloud), ice-sedimentation
MIROC-SPRINTARS-CHASER: (CCSR/NIES/FRCGC)
Aerosol-Chemistry-Climate Model

Changes in tropo. chem and their impacts on climate

- Ozone Hole
- Halogens (Cl, Br)
- CH₄
- CFCs
- N₂O
- Precursors: NOₓ, CO, VOCs
- SO₂, NH₃
- Surface
- Stratosphere
- Troposphere
- Ozone chemistry
- Gravity Wave
- Aerosols [Direct & indirect effects]
- Sulfate / Nitrate
- OC
- BC
- Dust
- Sea-Salt
- Vegetation (PAR, LAI, NPP)
- Stratosphere
- Troposphere
- UV
**Past Ozone Changes**

PI(1850) → Present

**Tropo. O₃ Increases**  
(only due to emissions increase)

---

**Tropo. O₃ Increases**  
+ Strato. O₃ decreases

---

**Tropo. Column Ozone Changes (DU)**

<table>
<thead>
<tr>
<th></th>
<th>global</th>
<th>NH</th>
<th>SH</th>
</tr>
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<tbody>
<tr>
<td>LT</td>
<td>+10.2</td>
<td>+12.4</td>
<td>+7.9</td>
</tr>
<tr>
<td>LTS</td>
<td>+9.2</td>
<td>+11.4</td>
<td>+6.9</td>
</tr>
</tbody>
</table>
Zonal mean O3 changes: preindustrial → present

Preind → present: total change

Due to emission change R2-R1b

Due to strato. O3 change R2-R1a

Due to climate change R1c-R1

IPCC-AR4: MIP
O$_3$ Change Profiles
(with sonde obs. for 1970-2004)

- observed (1970-2004)
- modeled (only with emission increases)
- modeled (total changes)

Changes in tropo. chem and their impacts on climate
Radiative forcing from tropo. O$_3$ increases

LT: tropo. O$_3$ increases

→ +0.49 W m$^{-2}$

(LLGHGs → +2.38 W m$^{-2}$)
Zonal mean temperature changes

Due to tropo. O\textsubscript{3} increase

**Response to upper tropo. O\textsubscript{3} increases**

\[ \sim -1.0 K \]

\[ \sim +0.5 K \]

**tropo. O\textsubscript{3} increase and strato. O\textsubscript{3} decrease**

**Enhanced B.D. circulation**
Rising tropopause?

Global average tropopause pressure changes (hPa)

<table>
<thead>
<tr>
<th></th>
<th>annual</th>
<th>January</th>
<th>July</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLGHGs</td>
<td>−4.45</td>
<td>−4.05</td>
<td>−4.75</td>
</tr>
<tr>
<td>Tropo. O3</td>
<td>−1.31</td>
<td>−1.11</td>
<td>−1.22</td>
</tr>
<tr>
<td>Strato. O3</td>
<td>−2.09</td>
<td>−2.70</td>
<td>−1.53</td>
</tr>
<tr>
<td>Total</td>
<td>−7.86</td>
<td>−7.86</td>
<td>−7.40</td>
</tr>
</tbody>
</table>

In PCM, ozone and well-mixed GHGs are the main drivers of tropopause height increases.

Santer et al. (2003)
*Science, 301, 479-*
Impacts of tropo. O$_3$ increase on surface temperature

### Climate Response

<table>
<thead>
<tr>
<th></th>
<th>Global</th>
<th>N.H.</th>
<th>S.H.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tropo. O$_3$</td>
<td>+0.28 °C</td>
<td>+0.31 °C</td>
<td>+0.25 °C</td>
</tr>
<tr>
<td>Strato. O$_3$</td>
<td>-0.04 °C</td>
<td>-0.04 °C</td>
<td>-0.04 °C</td>
</tr>
<tr>
<td>Net O$_3$</td>
<td>+0.24 °C</td>
<td>+0.2 °C</td>
<td>+0.21 °C</td>
</tr>
<tr>
<td>LLGHG</td>
<td>+2.29 °C</td>
<td>+1.78 °C</td>
<td>+2.80 °C</td>
</tr>
</tbody>
</table>

Climate sensitivity to tropospheric O$_3$ change  
= 0.57 K m$^2$ W$^{-1}$

~ 0.6 K m$^2$ W$^{-1}$ [Mickley et al., 2004]

Reduced long-wave absorption (~30%)  
& decreased O$_3$ input to the troposphere (~70%)
Impacts of tropo. O$_3$ increase on surface $T$ : \( \frac{\text{tropo. O}_3}{\text{LLGHGs}} \times 100 \% \)
### Future Projection of O$_3$/CH$_4$/Aerosols

(Chemistry/Climate interaction)

<table>
<thead>
<tr>
<th>Run Scenarios</th>
<th>Exp1</th>
<th>Exp2</th>
<th>Exp3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emission</td>
<td>Future</td>
<td>Future</td>
<td>Future</td>
</tr>
<tr>
<td>Climate</td>
<td>Present</td>
<td>Future</td>
<td>Future</td>
</tr>
<tr>
<td>Stratospheric O$_3$</td>
<td>Present</td>
<td>Present</td>
<td>Future</td>
</tr>
</tbody>
</table>

- Runs from 2000 to 2100.
- Simplified stratospheric ozone changes.
Future Simulation of $\text{O}_3$/CH$_4$/Aerosols

Emission Induced Changes in Surface Ozone $\Delta\text{O}_3$ (2050/2100)

- Ozone decreases in the US and Europe for A1/B1
Changes in tropo. chem and their impacts on climate

Future Simulation of O$_3$/ CH$_4$/ Aerosols

Impacts(%) of Climate Change on Surface Ozone (2100)

Impacts(%) of Climate Change on Zonal Mean Ozone (2100)
Future Simulation of O₃/ CH₄/ Aerosols

Temporal Evolution: Ozone Strato./Tropo. Exchange (TgO₃/yr)

- **Warming**
  - Lower Tropo.
    - (water vapor)
    - --(negative)
  - Upper Tropo.
    - (increased circulation)
    - +(positive)
  - Upper Tropo.
    - (increased STE)
    - +(positive)

- **Strato. O₃ Recov.**
  - (no climate change)
  - Strato. O₃ recovery

- **Changes in tropo. chem and their impacts on climate**

STE O₃ Flux: SRES-A2
STE O₃ Flux: SRES-A1
STE O₃ Flux: SRES-B1

O₃ STE and burden over time for different scenarios.
Future Simulation of O$_3$/CH$_4$/Aerosols

Temporal Evolution: Global Mean Methane CH$_4$ (ppmv) and SO$_4$

- **Strato. O$_3$ recovery**
  1) $\rightarrow$ [O$_3$ UV-photolysis $\downarrow$] $\rightarrow$ [OH $\downarrow$] $\rightarrow$ less CH$_4$ loss
  2) $\rightarrow$ [Tropo. O$_3$ $\uparrow$] $\rightarrow$ [OH $\uparrow$] $\rightarrow$ more CH$_4$ loss

Climate change: Faster destruction of CH$_4$ (enhanced OH due to water vapor increase)

Climate change: Faster oxidation of SO$_2$ (increased H$_2$O$_2$, etc)
Changes in tropo. chem and their impacts on climate

IPCC-AR4 Model inter-comparison

Climate change impacts (%) on Surface O$_3$ in 2030 for JJA

Dentener et al. [2006]: IPCC-AR4/ACCENT model inter-comparison (~10 models)
Does Climate Change Mitigate/Amplify Air Pollution?

Changes in frequency of surf. O$_3$ level at 2030
Does Climate Change Mitigate/Amplify Air Pollution?

Impact of climate change on ozone environmental standards for 2030

Ellingsen et al. (2007)
The past O3 changes:

- The past changes in tropospheric / stratospheric O$_3$ and LLGHGs differently affect temperature profiles with different mechanisms.
- Tropospheric O$_3$ increase causes surface warming of ~0.31 °C in NH; especially large impacts are calculated in Asia (~30-40% of the LLGHGs impacts)
- Decreased stratospheric O$_3$ cools the surface by reducing long-wave absorption and decreasing O$_3$ input to the troposphere.

Future perspectives:

- Future climate change is likely to modulate atmospheric chemistry. + circulation (B.D. & Hadley), lightning NOx, water vapor, ...
- Future climate change may affect air quality, but that process has not been clarified yet.
Changes in tropo. chem and their impacts on climate

- **Ozone Hole**
- **Strato. O3**
- **Tropo. O3**
- **Air Pollution**
- **climate**

Circulation changes:
- + Brewer-Dobson
- + Hadley
- + Walker

Enhanced by warming?

- **Halogenes**
- **PSCs**
- **UT/LS**

(※) Enhanced by warming?
Future Plans …

✓ AC&C related:
    + trends in \( \text{O}_3 \), \( \text{CH}_4 \), related species and aerosols
    + impacts of emissions, climate change, dynamical variation
      (ENSO/AO..)
  • Future simulations toward IPCC-AR5:
    + global \( \text{O}_3 \) fields for new 4 scenarios

✓ Earth-System perspectives (long-term prediction?):

✓ Atmospheric chemistry and climate studies need more tight coupling/fusion between modeling and observation