

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

Kengo Sudo^{1,2}

¹ Graduate School of Environmental Studies, Nagoya University, Japan

² Frontier Research Center for Global Change, JAMSTEC, Japan

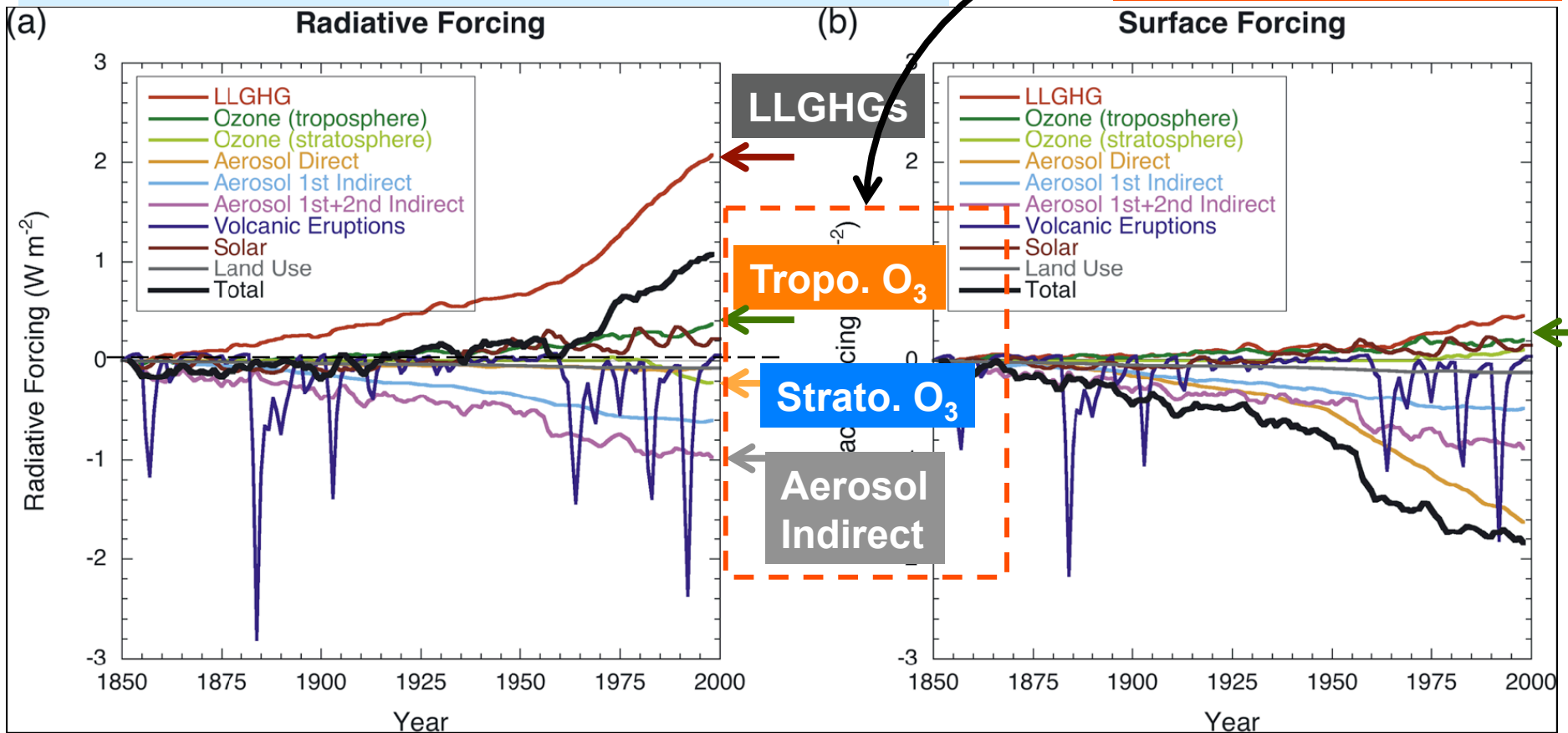
The 4th SPARC General Assembly
Bologna, Italy, 31 Aug. -- 5 Sep. 2008



Radiative Forcing in the 20th century

Air pollution / Atmospheric Chem.

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

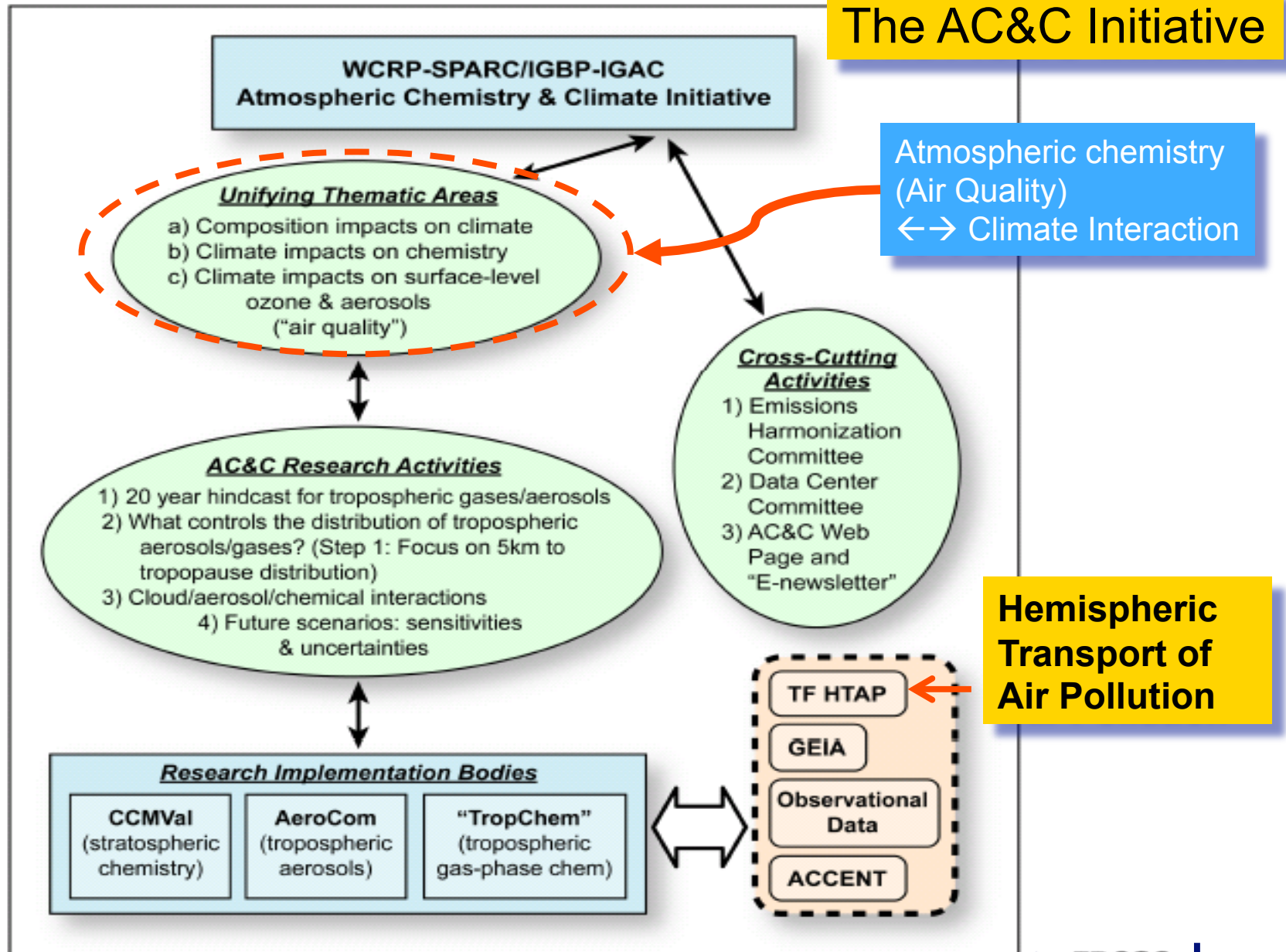


Takemura et al [2006]



FRCGC
Frontier
Research Center
for Global Change

The AC&C Initiative

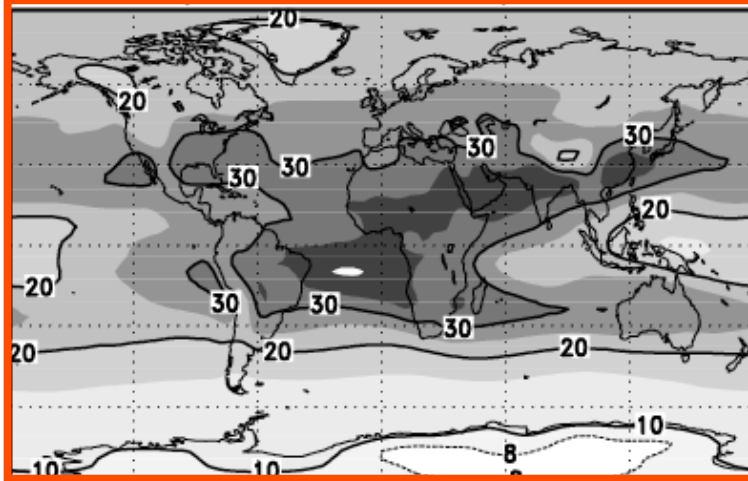


Contributions to Annual Mean Tropospheric Column O₃ (TCO)

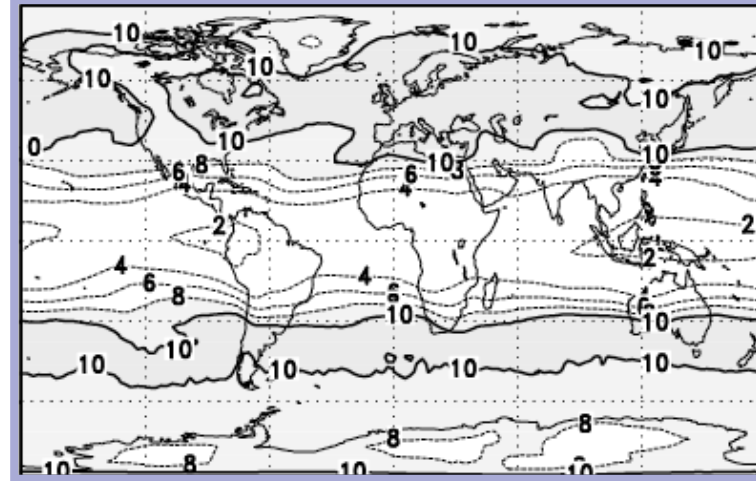
Tropospheric Origin

Stratospheric Origin

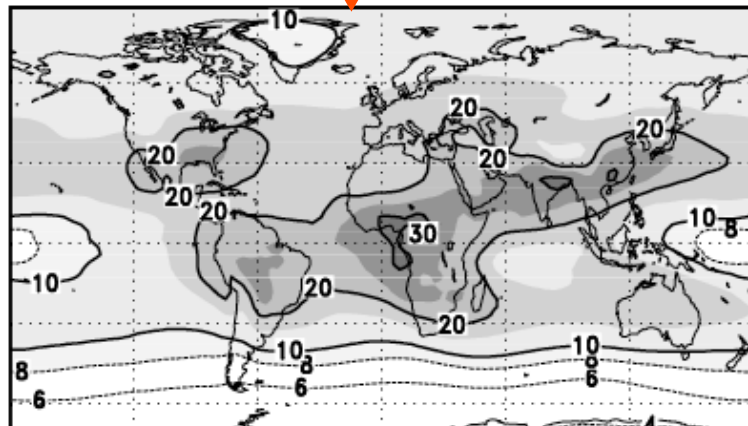
a) TROPO (POLTD+REMOT) <MM=Ann>



b) STRAT <MM=Ann>

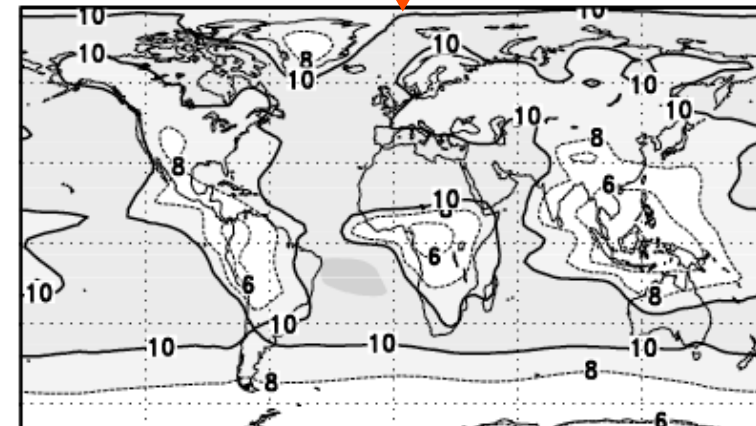


c) POLTD <MM=Ann>

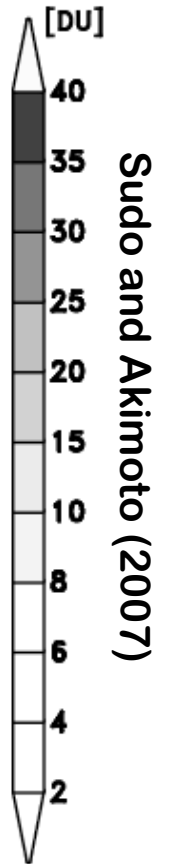


<Production in polluted regions>

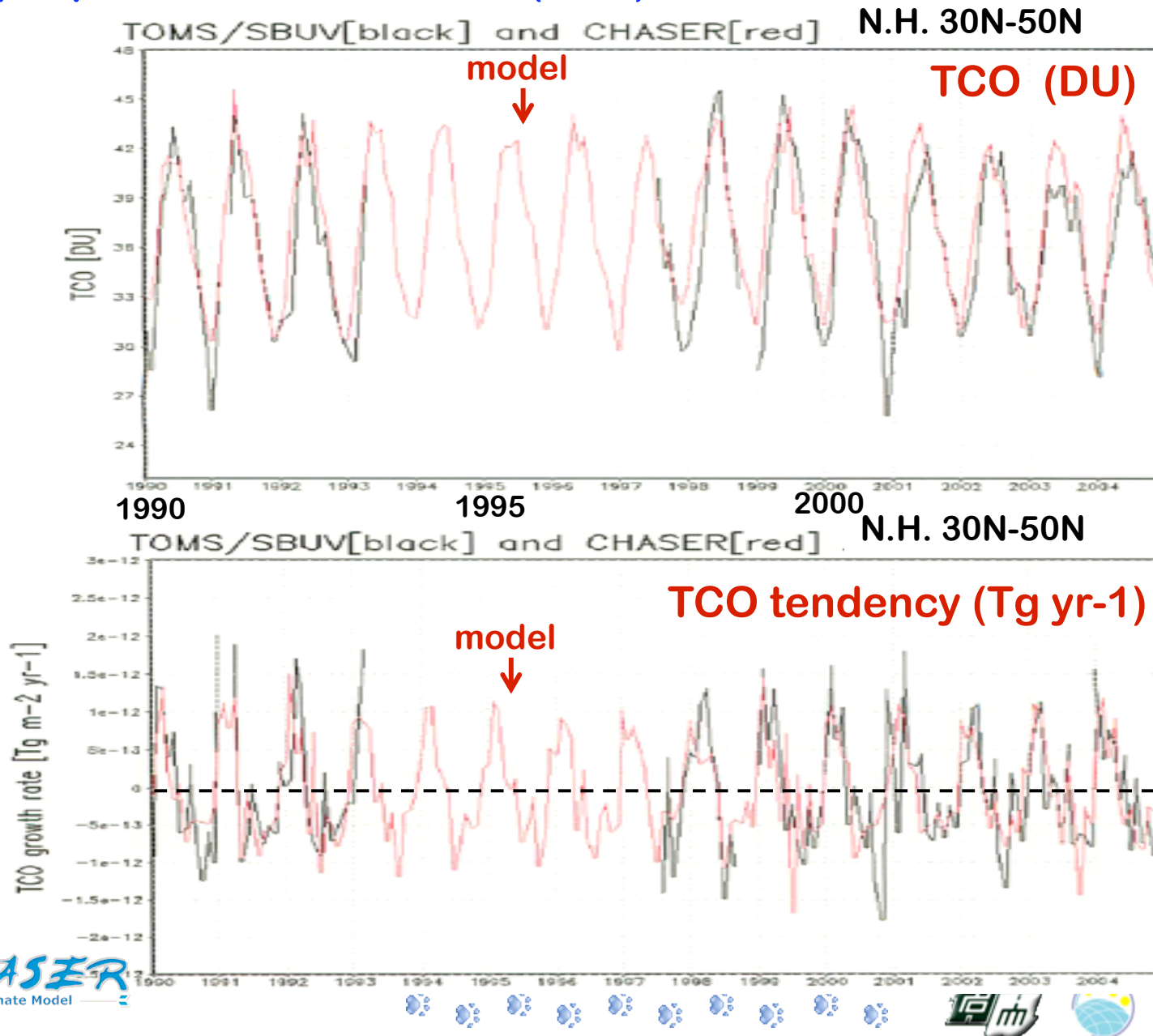
d) REMOT <MM=Ann>



<Production in remote regions>



Tropospheric Column Ozone (TCO) seasonal-annual variation



Global Budgets of Tropospheric O₃ from Distinct Source Regions

Stratospheric origin

Sudo and Akimoto (2007)

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

| Tracer ID ^a | Chem. production | | | | Tropo. Burden (TgO ₃) | | Lifetime(days) | | |
|------------------------|------------------|------------------|------------|------------------|-----------------------------------|--------------|----------------|-------------|----|
| | P ^b | P-L ^c | Deposition | STE ^d | Global (%) | NH | Chemical | Residential | |
| O ₃ -ALL | 4744 | 444.0 | -917 | 472.7 | 344.6 (100.0%) | 187.3 | 26 | 22 | |
| STRAT | 0 | -484.1 | -131 | 615.6 | 77.9 (22.6%) | 38.6 | 45 | 37 | |
| REMOI | 1735 | 338.7 | -257 | -81.7 | 101.0 (29.3%) | 51.1 | 25 | 21 | |
| POLTD | 3010 | 589.5 | -528 | -61.3 | 165.7 (48.1%) | 97.5 | 23 | 20 | |
| PBL | BL-AMN | 162 | 55.4 | -47 | -8.1 | 7.0 (2.0%) | 6.8 | 24 | 16 |
| | BL-AMM | 162 | 29.8 | -30 | -0.3 | 6.6 (1.9%) | 5.5 | 18 | 15 |
| | BL-AMS | 203 | 49.6 | -49 | -0.3 | 7.4 (2.1%) | 0.9 | 18 | 13 |
| | BL-AFN | 166 | 33.1 | -33 | -0.2 | 5.4 (1.6%) | 4.0 | 15 | 12 |
| | BL-AFS | 175 | 38.8 | -38 | -0.3 | 6.6 (1.9%) | 0.9 | 17 | 14 |
| | BL-EUR | 116 | 49.4 | -44 | -5.6 | 4.5 (1.3%) | 4.4 | 27 | 14 |
| | BL-CEU | 76 | 31.3 | -28 | -3.7 | 3.0 (0.9%) | 2.9 | 28 | 14 |
| | BL-MES | 92 | 22.3 | -22 | 0.0 | 3.2 (0.9%) | 2.9 | 17 | 13 |
| | BL-IND | 97 | 20.9 | -20 | -0.8 | 3.9 (1.1%) | 3.4 | 18 | 14 |
| | BL-TLD | 50 | 7.8 | -7 | -0.4 | 2.2 (0.6%) | 1.7 | 18 | 16 |
| | BL-CHN | 128 | 34.6 | -34 | -1.0 | 5.9 (1.7%) | 5.5 | 23 | 17 |
| | BL-JPN | 22 | 6.4 | -6 | 0.0 | 1.1 (0.3%) | 1.1 | 24 | 18 |
| | BL-IDN | 61 | 9.7 | -9 | -0.5 | 2.8 (0.8%) | 0.9 | 19 | 17 |
| | BL-AUS | 88 | 19.7 | -19 | -0.3 | 3.7 (1.1%) | 0.2 | 20 | 15 |
| FT | FT-AMN | 167 | 29.6 | -19 | -10.6 | 11.9 (3.4%) | 10.7 | 29 | 26 |
| | FT-AMS | 295 | 32.9 | -27 | -5.8 | 21.0 (6.1%) | 7.0 | 27 | 26 |
| | FT-AFN | 203 | 21.2 | -19 | -2.2 | 13.4 (3.9%) | 9.1 | 25 | 24 |
| | FT-AFS | 186 | 21.6 | -18 | -3.8 | 13.1 (3.8%) | 2.1 | 27 | 25 |
| | FT-EUR | 60 | 15.9 | -12 | -4.1 | 4.2 (1.2%) | 4.1 | 34 | 25 |
| | FT-ASA | 325 | 39.7 | -32 | -8.0 | 24.9 (7.2%) | 20.5 | 30 | 28 |
| | FT-IDN | 90 | 8.9 | -7 | -2.2 | 6.9 (2.0%) | 2.2 | 29 | 28 |
| | FT-AUS | 86 | 11.0 | -8 | -3.2 | 6.9 (2.0%) | 0.8 | 31 | 29 |



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. O₃ increase and strato. O₃ decrease in the 20th century

2. Long/near term future projection of tropospheric ozone and related species (CH₄ / 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

| | Ctrl | L | LT | LTm | LTS |
|------------------------|------|----|----|-----|-----|
| LLGHGs | PI | PD | PD | PD | PD |
| Tropo. O ₃ | PI | PI | PD | PD* | PD |
| Strato. O ₃ | PI | PI | PI | PI | PD |

(!) LLGHGs = CO₂ + CH₄ + N₂O + CFCs

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

Zonally averaged
O₃ increases



Chemistry-coupled climate model CHASER Sudo et al. [2002a,b]



| | |
|----------------|---|
| 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 | <p>54 chemical species, 152 chemical reactions (gas, liquid, heterogeneous*)</p> <p>(1) O₃-HO_x-NO_x-CO-CH₄, (2) NMHCs oxidation, and (3) SO₂, DMS oxidation</p> <p>* heterogeneous reactions are considered for surface of cloud particles, surface of aerosols, and surface of ice crystals (satellite data are used for stratospheric O₃ and HO_x above 20km)</p> <p>+ Simplified stratospheric chemistry (no-PSCs)</p> |
| Emission | <p>Industry, biomass burning, vegetation/soil/ocean, lightning NO_x (NO_x, CO, C₂H₆, C₂H₄, C₃H₈, C₃H₆, acetone, isoprene, terpenes, SO₂, DMS)</p> <p>Lightning NO_x is parameterized in the GCM convection [Price & Rind, 1992]</p> |
| 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 |

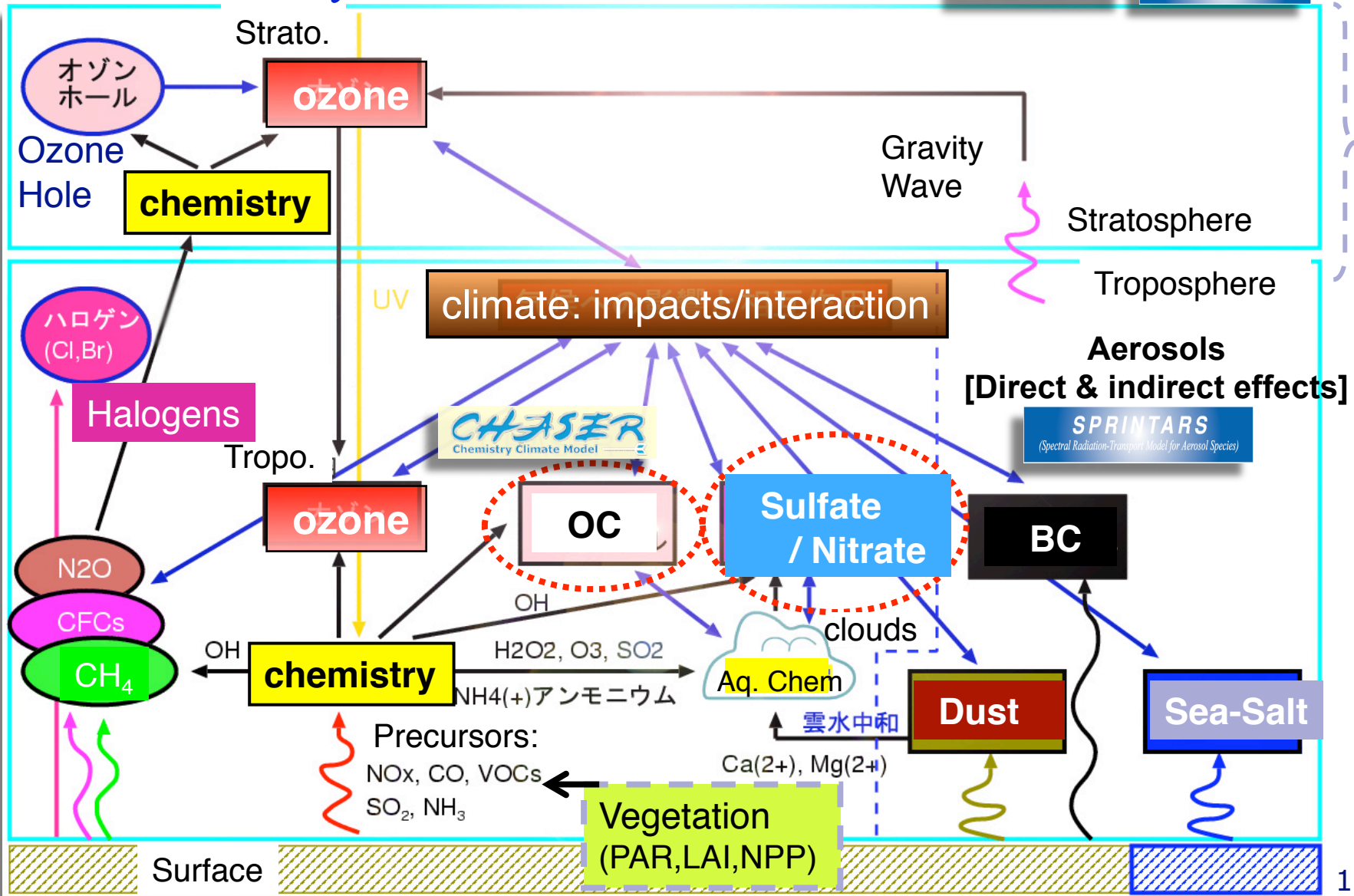


MIROC-SPRINTARS-CHASER: (CCSR/NIES/FRCGC)

Aerosol-Chemistry-Climate Model

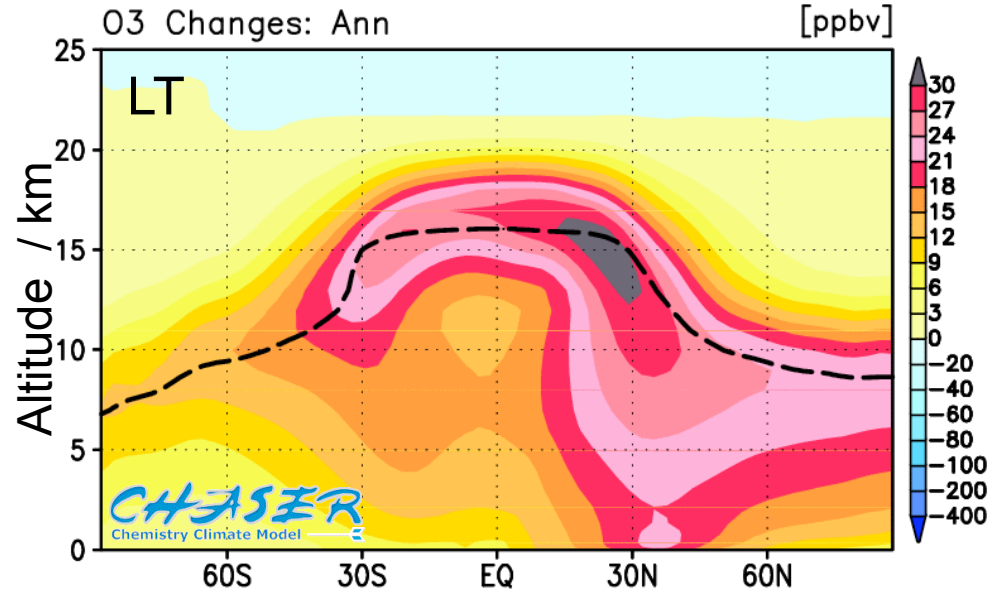
CHASER
Chemistry Climate Model

SPRINTARS
(Spectral Radiation-Transport Model for Aerosol Species)

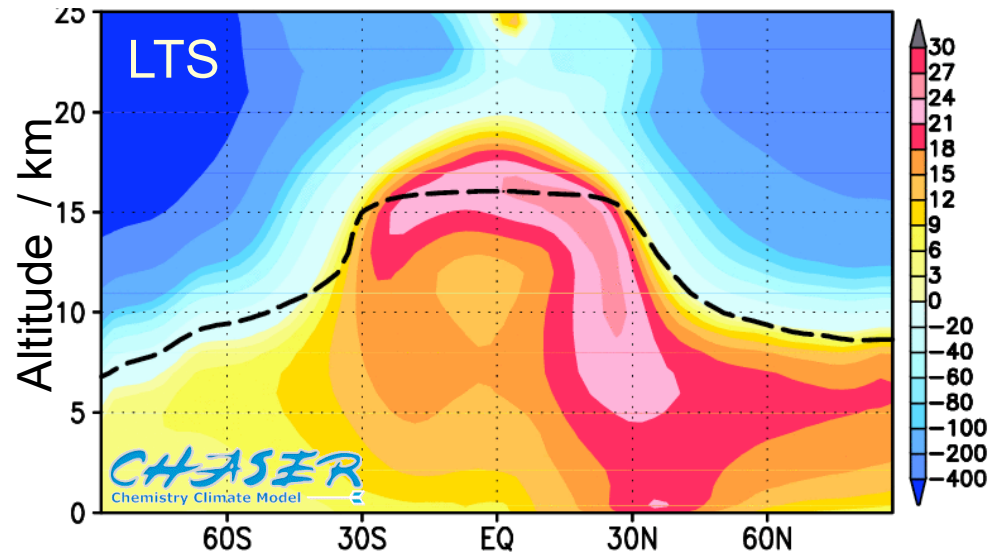


Past Ozone Changes
PI(1850) → Present

Tropo. O₃ Increases
(only due to emissions increase)



Tropo. O₃ Increases
+ Strato. O₃ decreases



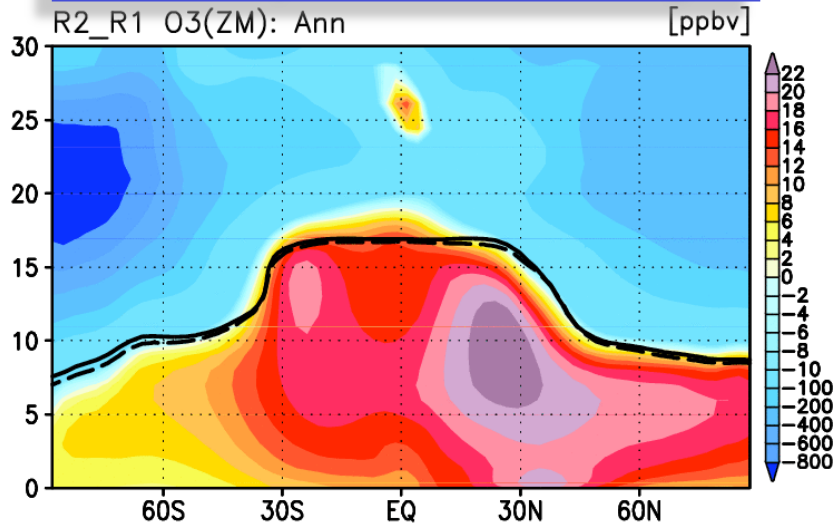
Tropo. Column Ozone Changes (DU)

| | global | NH | SH |
|-----|--------|-------|------|
| LT | +10.2 | +12.4 | +7.9 |
| LTS | +9.2 | +11.4 | +6.9 |

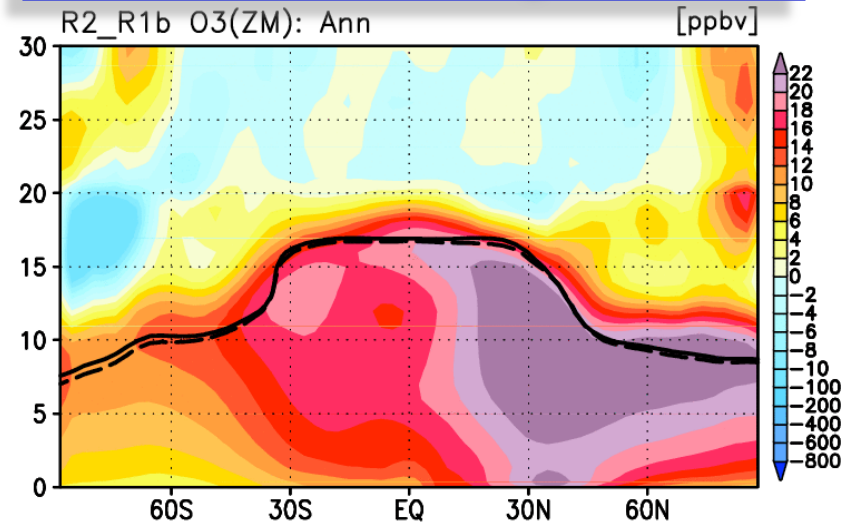
Zonal mean O3 changes: preindustrial → present

IPCC-AR4:
MIP

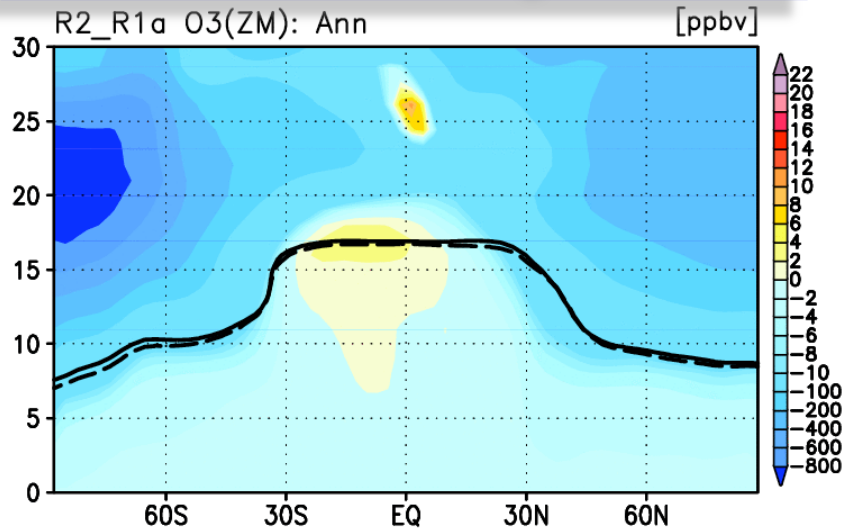
Preind → present: total change



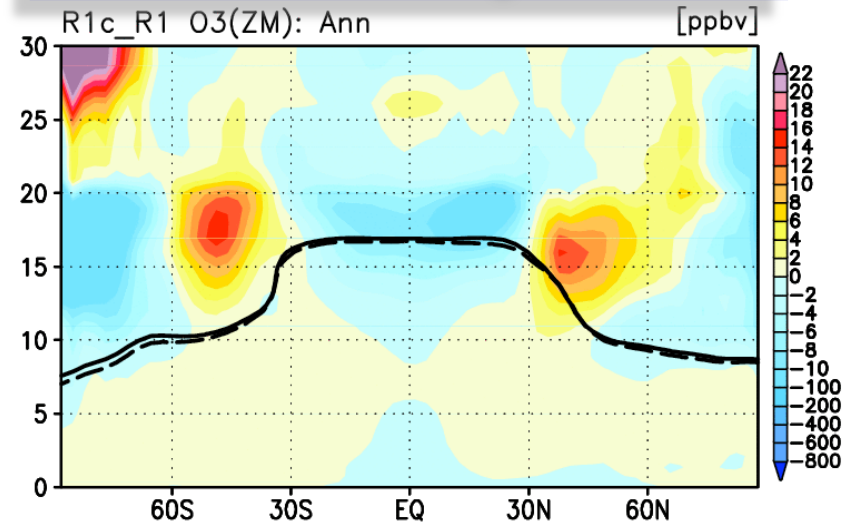
Due to emission change R2-R1b



Due to strato. O3 change R2-R1a

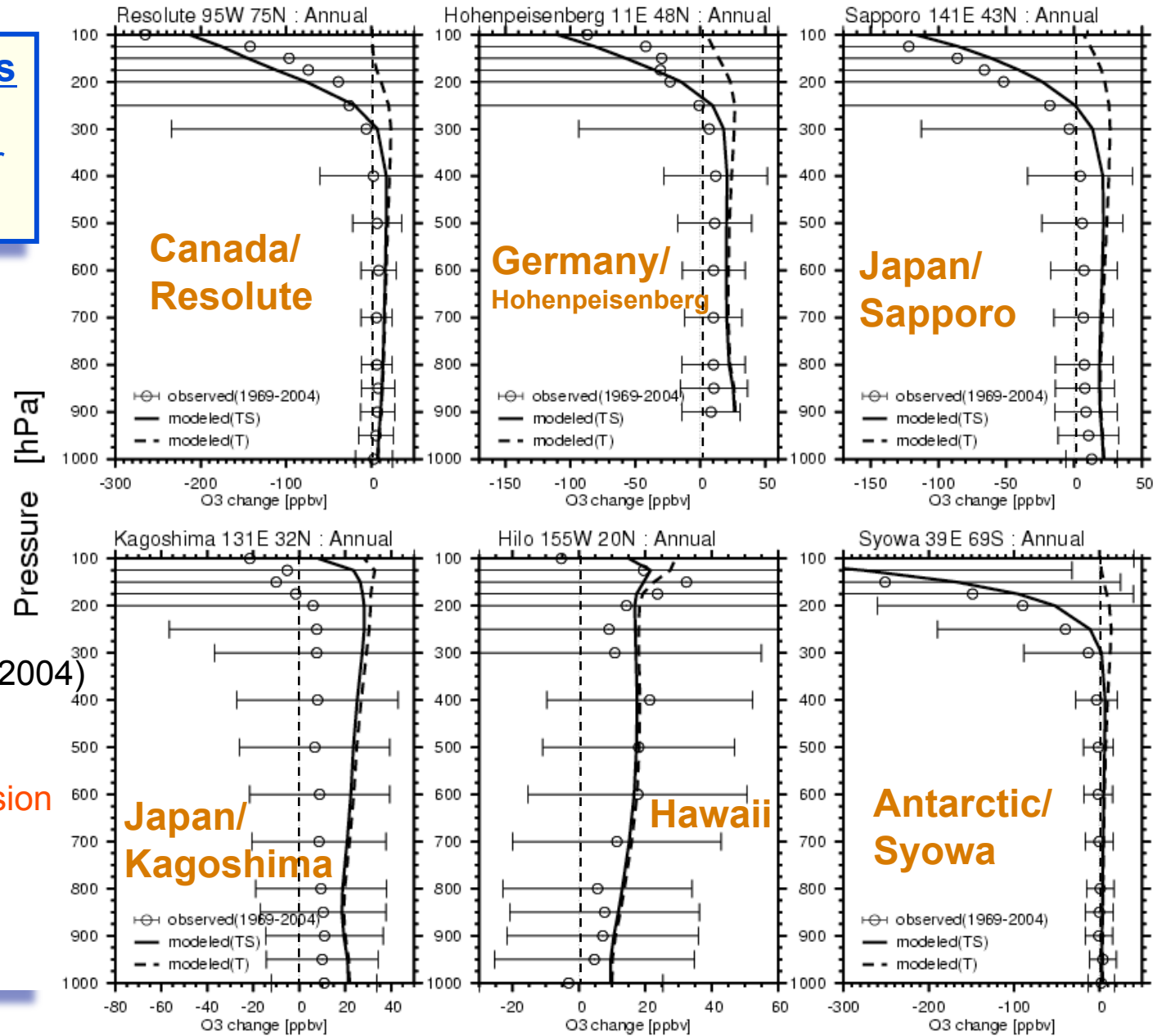


Due to climate change R1c-R1



O₃ Change Profiles

(with sonde obs. for 1970-2004)



⊕ observed (1970-2004)

--- modeled
(only with emission increases)

— modeled
(total changes)

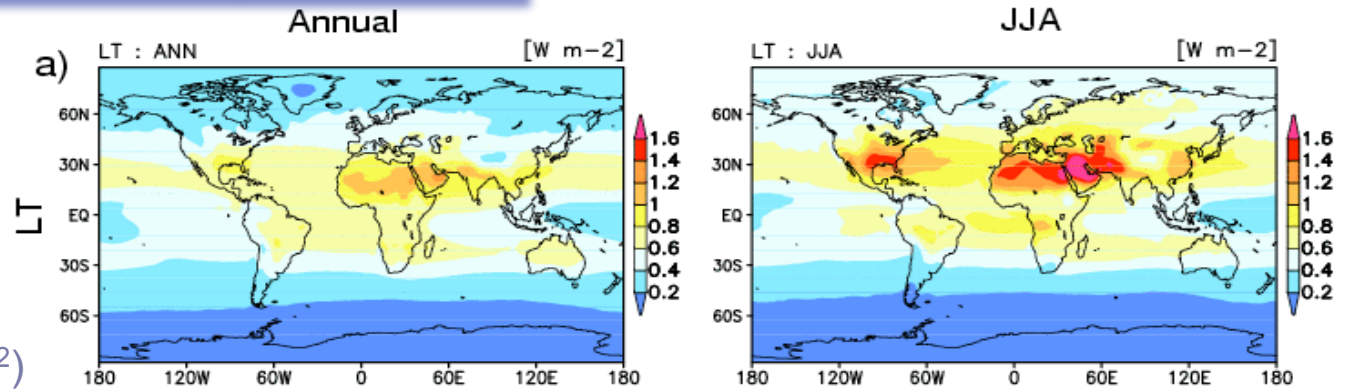
Radiative forcing from tropo. O₃ increases



LT: tropo. O₃ increases

→ +0.49 W m⁻²

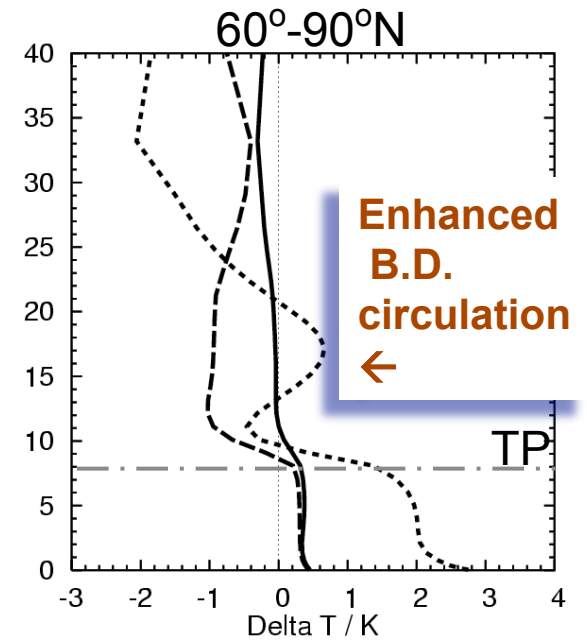
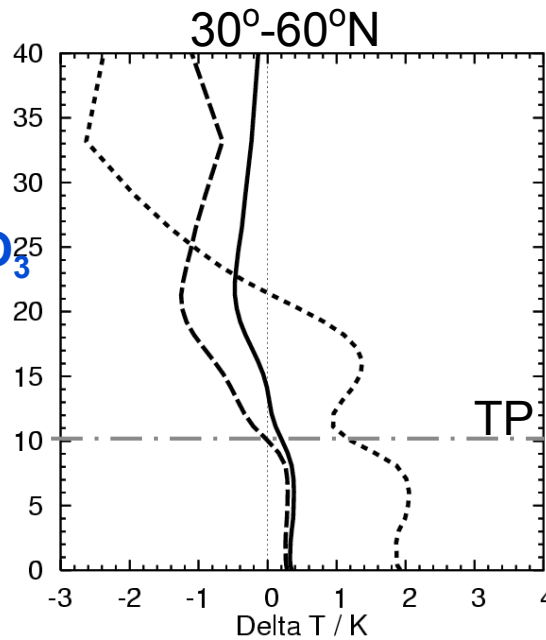
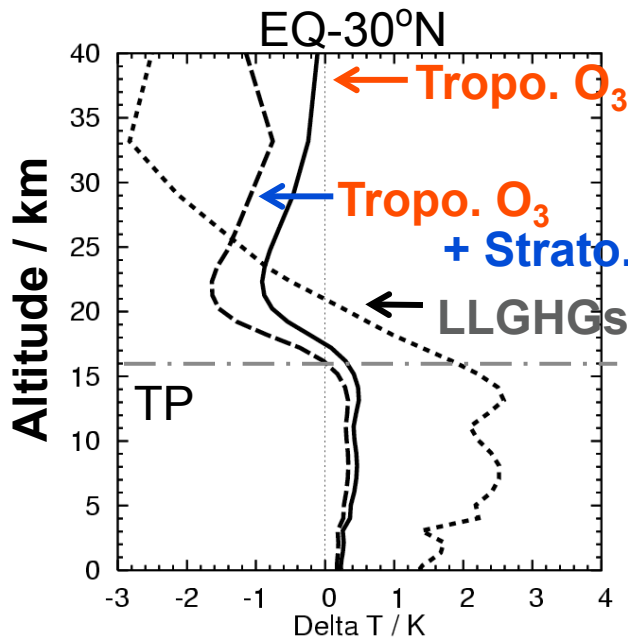
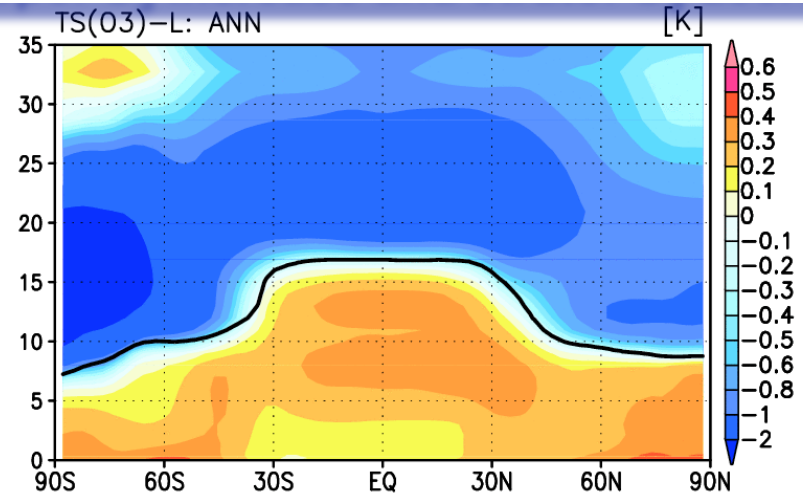
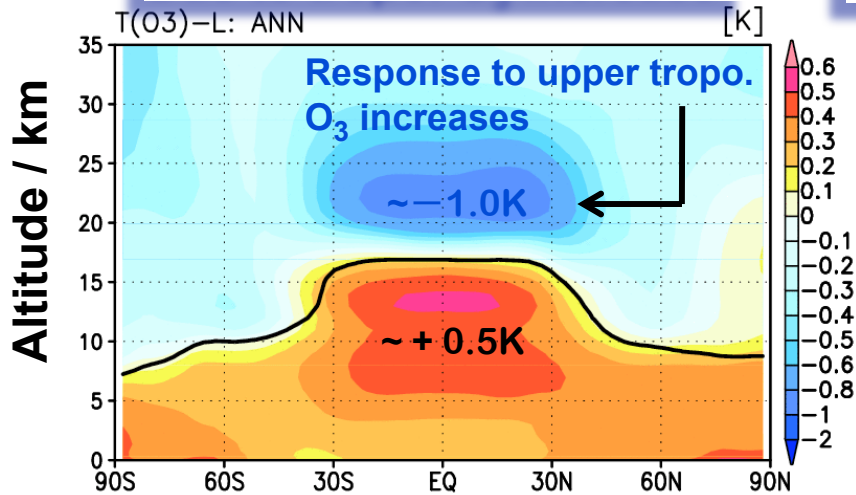
(LLGHGs → +2.38 W m⁻²)



Zonal mean temperature changes

Due to tropo. O₃ increase

tropo. O₃ increase and strato. O₃ decrease

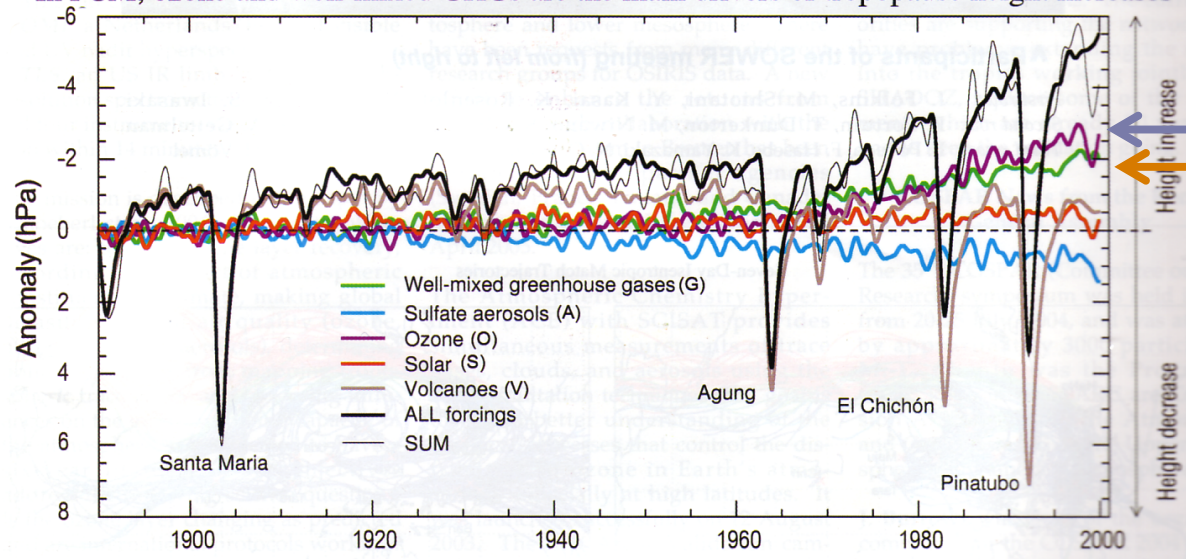


Rising tropopause ?

Global average tropopause pressure changes (hPa)

| | annual | January | July |
|------------|--------|---------|-------|
| LLGHGs | -4.45 | -4.05 | -4.75 |
| Tropo. O3 | -1.31 | -1.11 | -1.22 |
| Strato. O3 | -2.09 | -2.70 | -1.53 |
| Total | -7.86 | -7.86 | -7.40 |

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



LLGHGs

Ozone

Santer et al. (2003)
Science, 301, 479-



Impacts of tropo. O₃ increase on surface temperature

Climate Response

| | Global | N.H. | S.H. |
|------------------------------|-----------------|-----------------|-----------------|
| Tropo. O₃ | +0.28 °C | +0.31 °C | +0.25 °C |
| Strato. O₃ | -0.04 °C | -0.04 °C | -0.04 °C |
| Net O₃ | +0.24 °C | +0.2 °C | +0.21 °C |
| LLGHG | +2.29 °C | +1.78 °C | +2.80 °C |

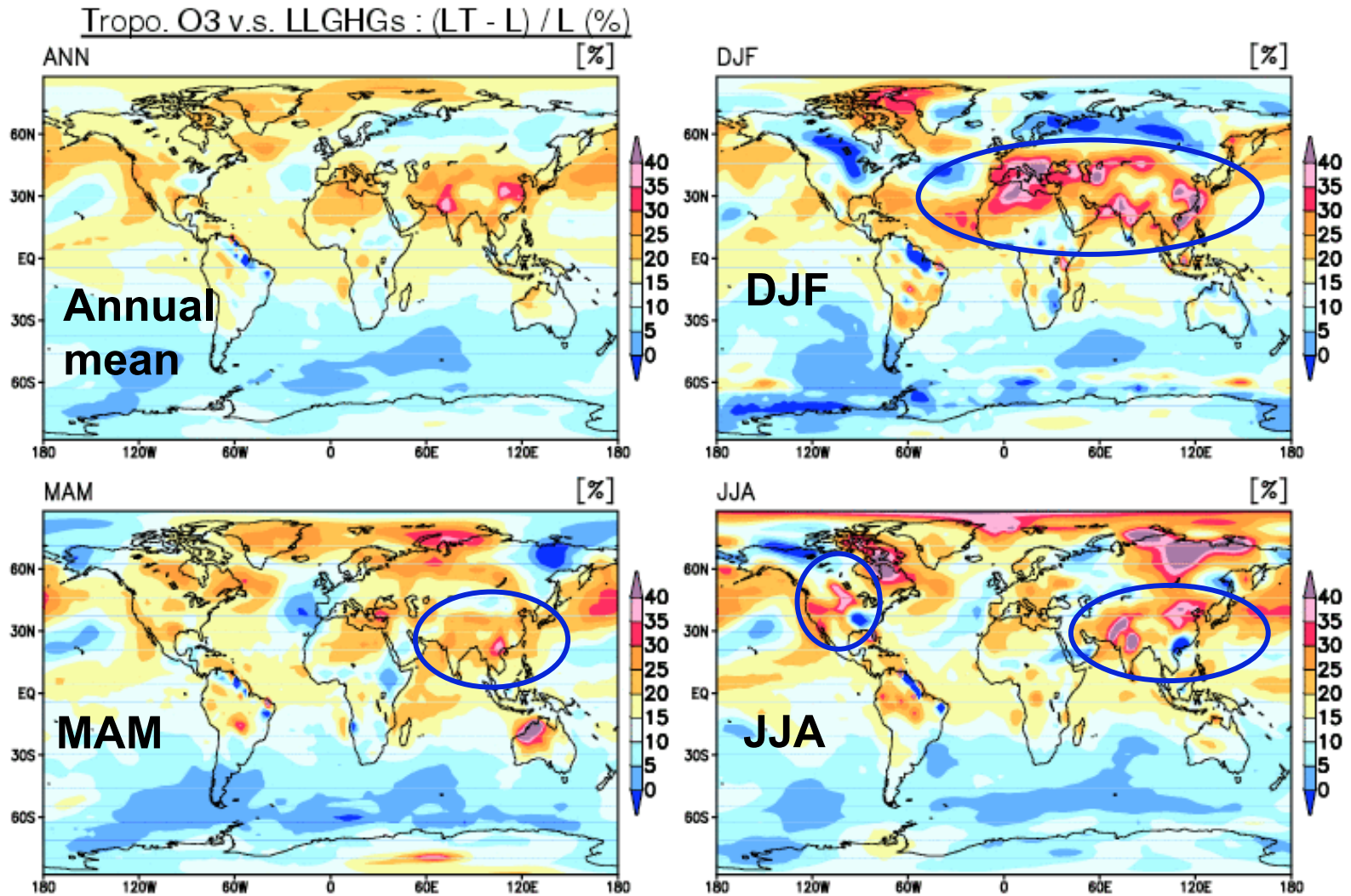
Climate sensitivity to tropospheric O₃ change
= 0.57 K m² W⁻¹

~ 0.6 K m² W⁻¹ [Mickley et al., 2004]

Reduced long-wave absorption (~30%)
& decreased O₃ input to the troposphere (~70%)



Impacts of tropo. O₃ increase on surface T : (tropo. O₃) / (LLGHGs) x 100 (%)



Future Projection of O₃/CH₄/Aerosols (Chemistry/Climate interaction)

Run Scenarios

| | Exp1 | Exp2 | Exp3 |
|------------------------------|---------|---------|--------|
| Emission | Future | Future | Future |
| Climate | Present | Future | Future |
| Stratospheric O ₃ | Present | Present | Future |

- Emission & climate: Using IPCC SRES-A2, A1, B1 scenarios.
- Runs from 2000 to 2100.
- Simplified stratospheric ozone changes.

CHASER

SPRINTARS

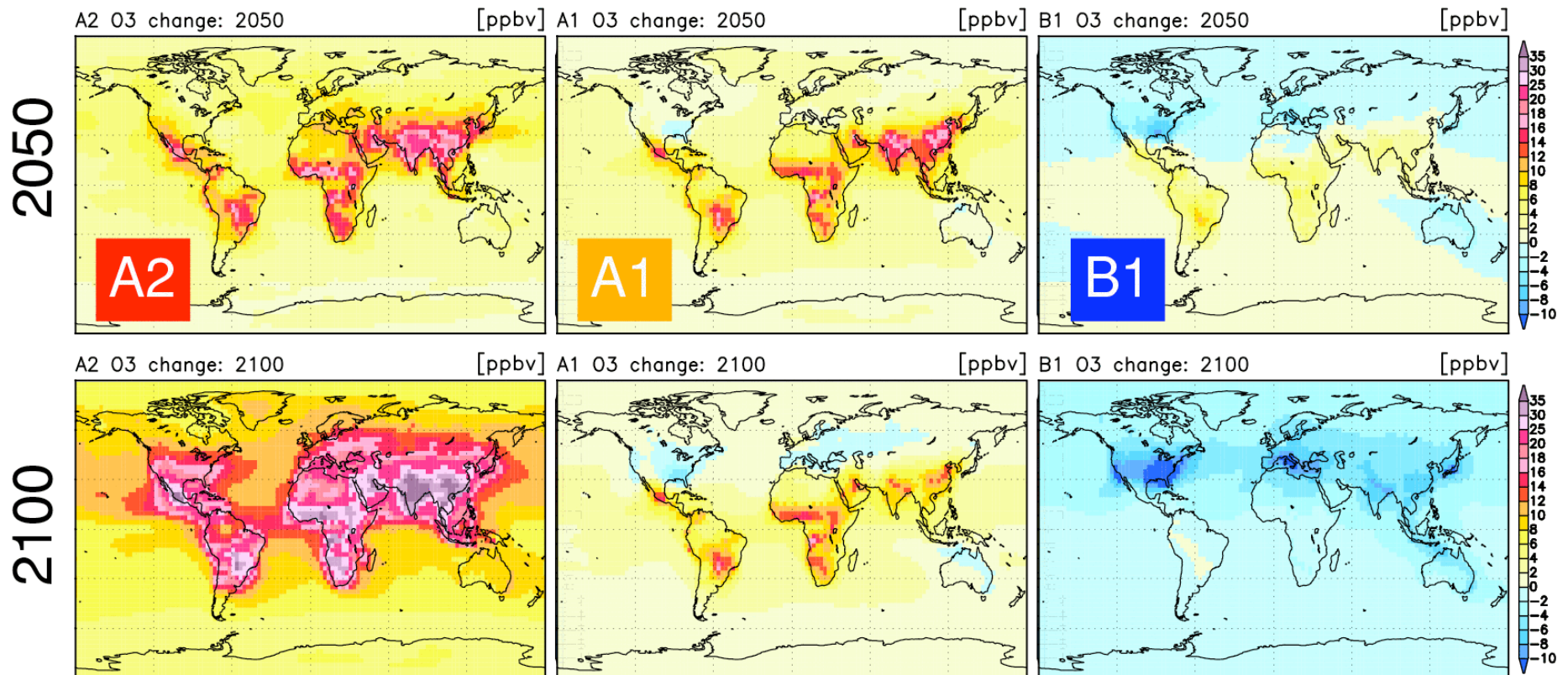
(Spectral Radiation-Transport Model for Aerosol Species)



FRCGC
Frontier
Research Center
for Global Change

Future Simulation of O₃/ CH₄/ Aerosols

Emission Induced Changes in Surface Ozone ΔO_3 (2050/2100)



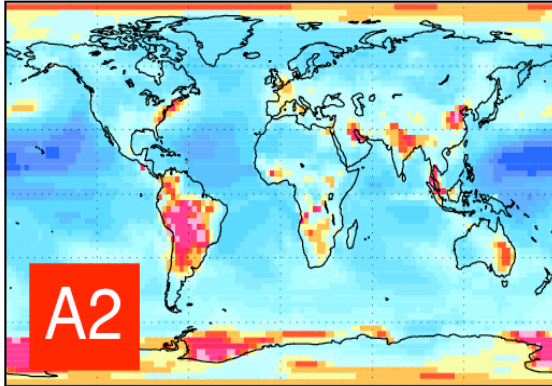
✓ Ozone decreases in the US and Europe for A1/B1



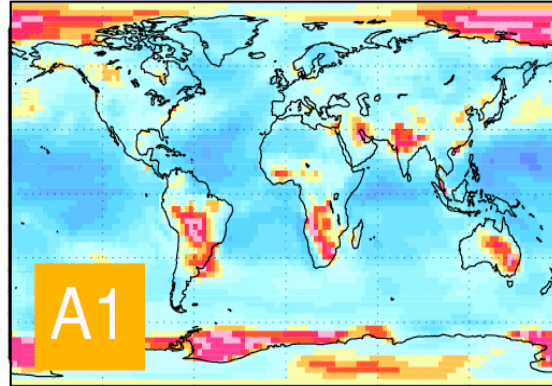
Future Simulation of O₃/ CH₄/ Aerosols

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

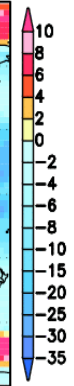
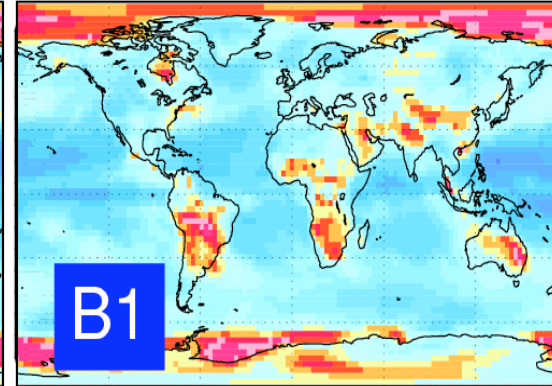
A2 Surface O₃ warming impact(%): 2100



A1 Surface O₃ warming impact(%): 2100



B1 Surface O₃ warming impact(%): 2100



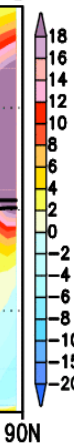
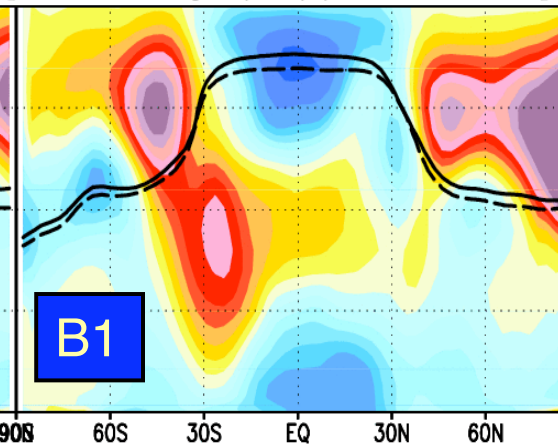
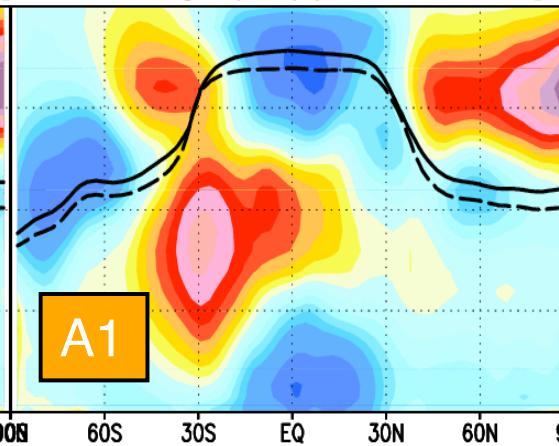
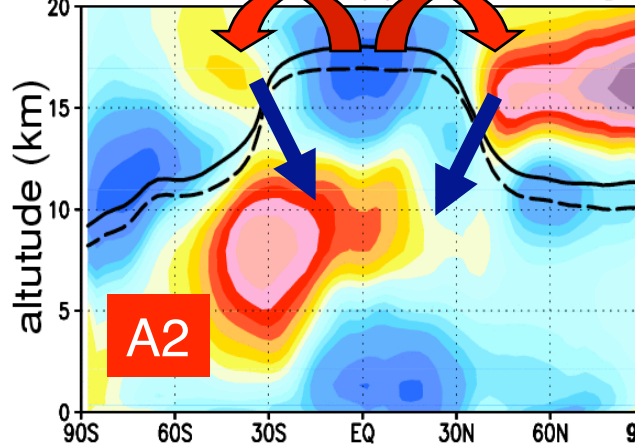
Impacts(%) of Climate Change on Zonal Mean Ozone (2100)

A2 O₃ warming impact(%): 2100

[%]A1 O₃ warming impact(%): 2100

[%]B1 O₃ warming impact(%): 2100

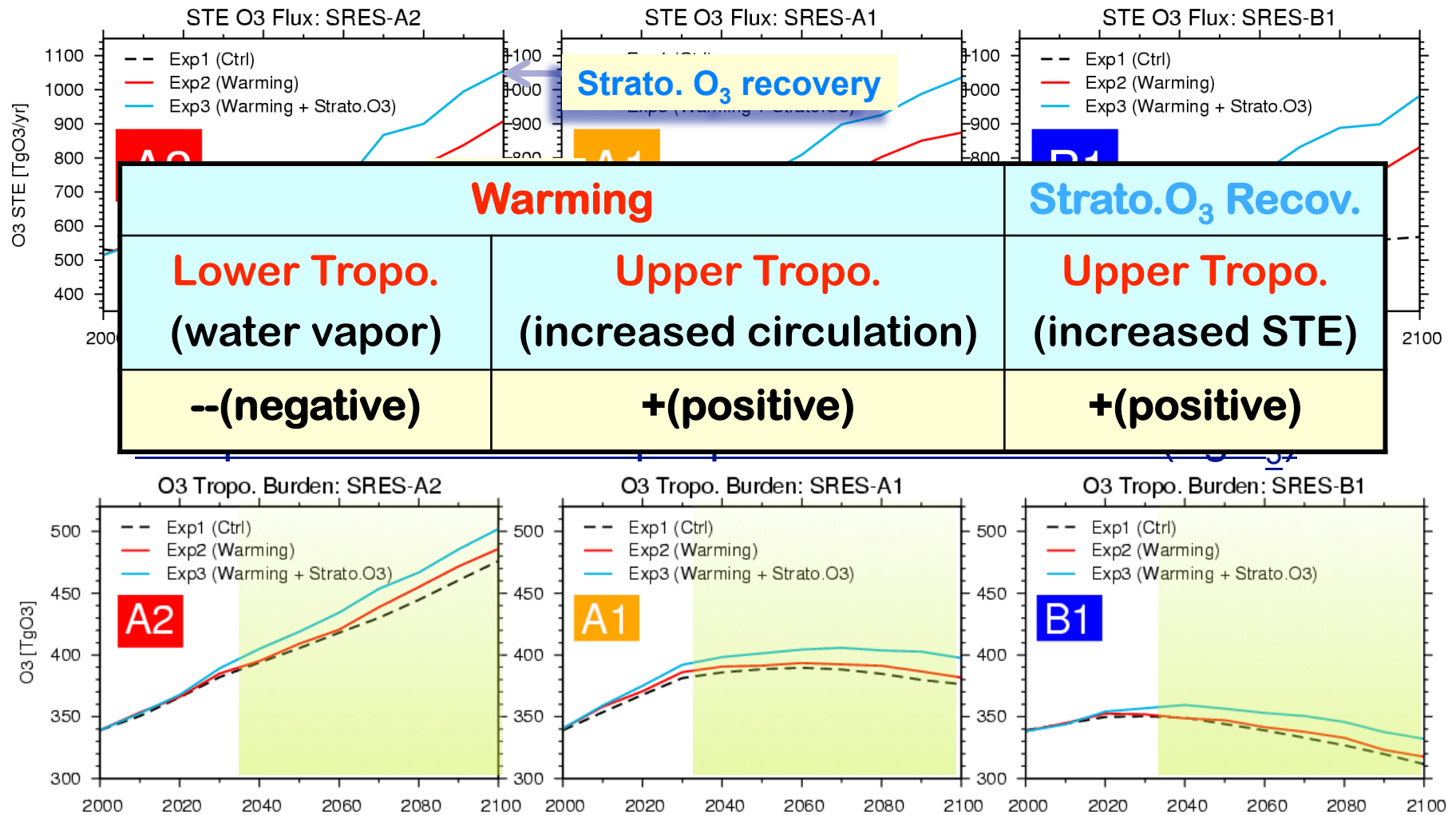
[%]



Future Simulation of O₃/ CH₄/ Aerosols



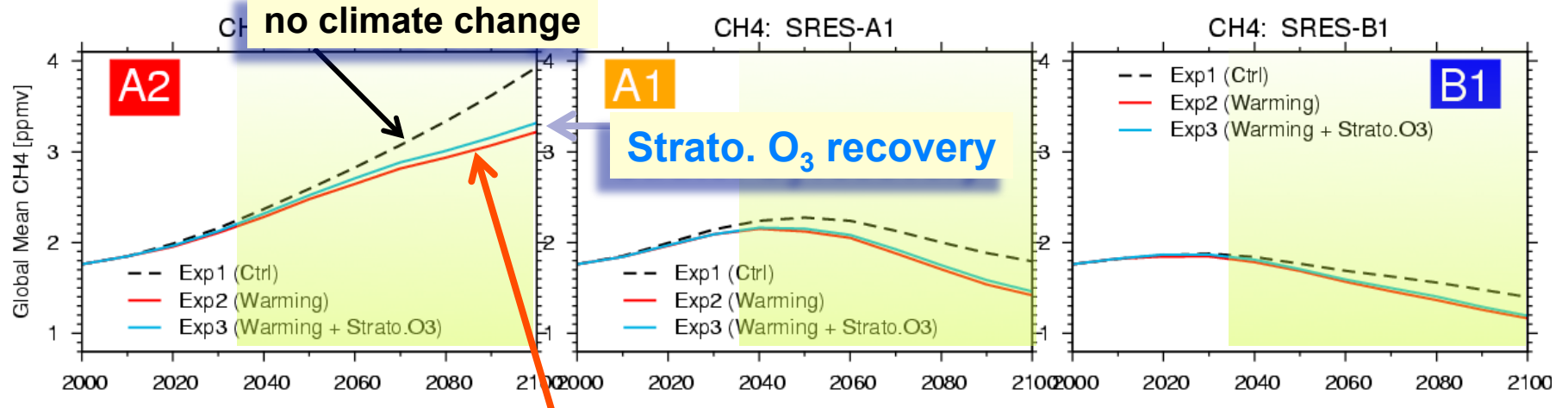
Temporal Evolution : Ozone Strato./Tropo. Exchange (TgO₃/yr)



Future Simulation of O₃/ CH₄/ Aerosols



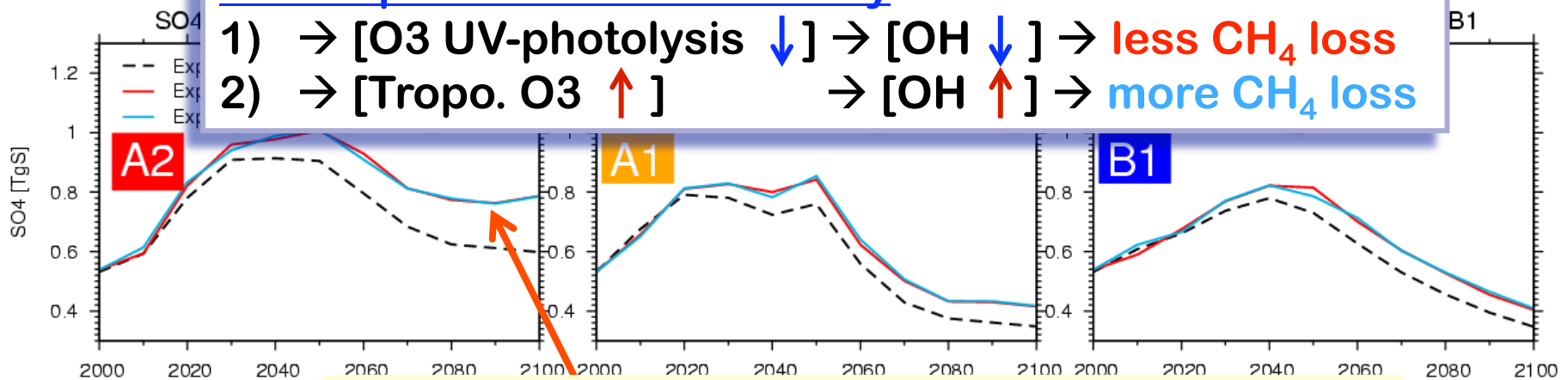
Temporal Evolution : Global Mean Methane CH₄ (ppmv) and SO₄



Climate change: Faster destruction of CH₄ (enhanced OH due to water vapor increase)

Stratospheric Ozone Recovery

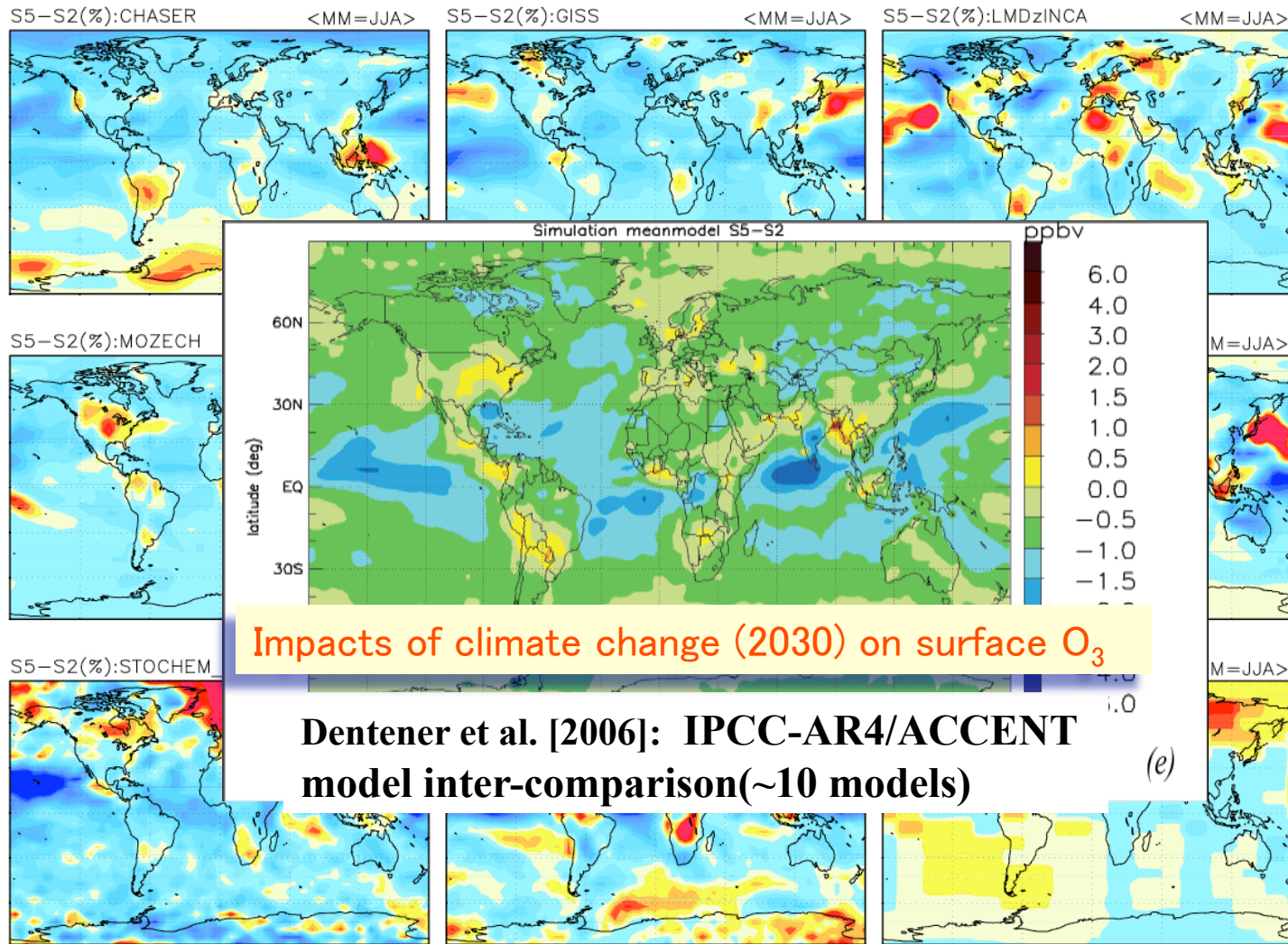
- 1) → [O₃ UV-photolysis ↓] → [OH ↓] → less CH₄ loss
- 2) → [Tropo. O₃ ↑] → [OH ↑] → more CH₄ loss



Climate change: Faster oxidation of SO₂ (increased H₂O₂, etc)

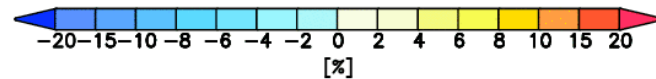
IPCC-AR4 Model inter-comparison

Climate change impacts (%) on Surface O₃ in 2030 for JJA



Impacts of climate change (2030) on surface O₃

Dentener et al. [2006]: IPCC-AR4/ACCENT model inter-comparison (~10 models)

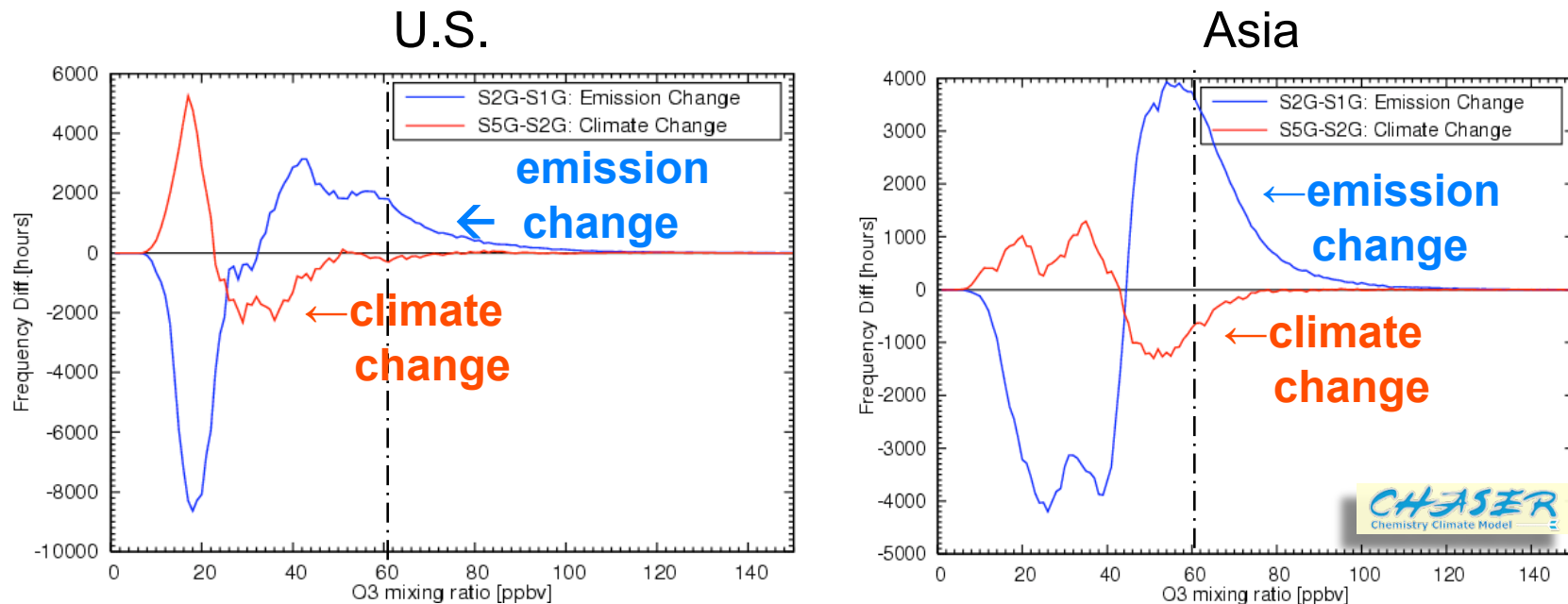


IPCC-AR4 Model inter-comparison



Does Climate Change Mitigate/Amplify Air Pollution ?

Changes in frequency of surf. O₃ level at 2030



IPCC-AR4 Model inter-comparison

Does Climate Change Mitigate/Amplify Air Pollution ?

Impact of climate change on ozone environmental standards for 2030

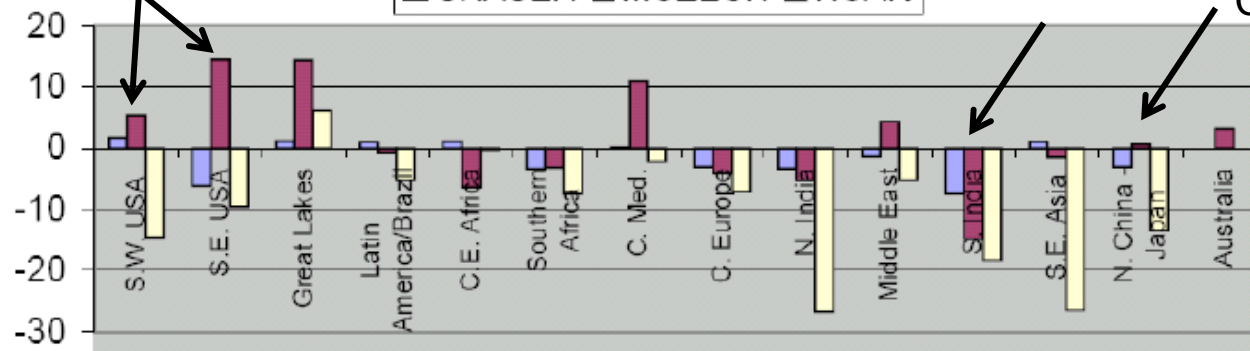
S.W & S.E. US

EU60 S5-S2

CHASER MOZECH NCAR

S-India

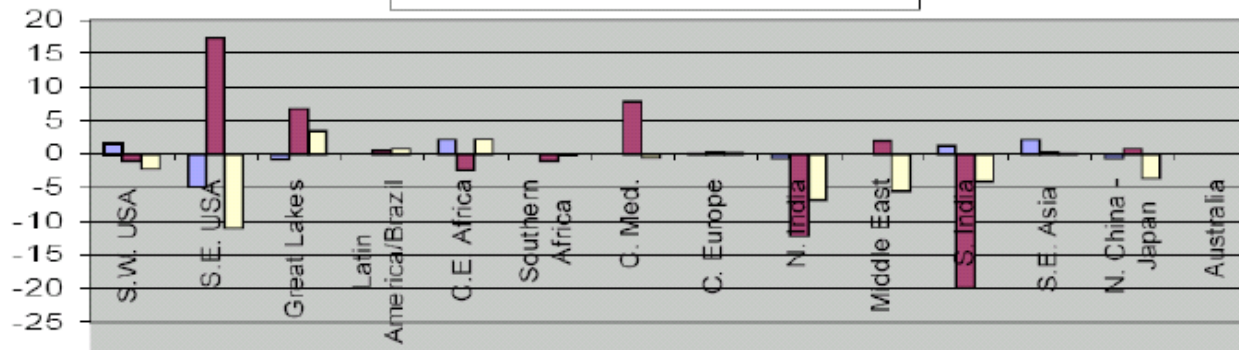
China & Japan



USEPA80 S5-S2

CHASER MOZECH NCAR

Days of exceedance



Ellingsen et al. (2007)



Summary & Conclusions

The past O₃ changes:

- The past changes in **tropospheric** / **stratospheric** O₃ and **LLGHGs** differently affect temperature profiles with different mechanisms.
- Tropospheric O₃ 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 O₃ cools the surface by **reducing long-wave absorption and decreasing O₃ input to the troposphere.**

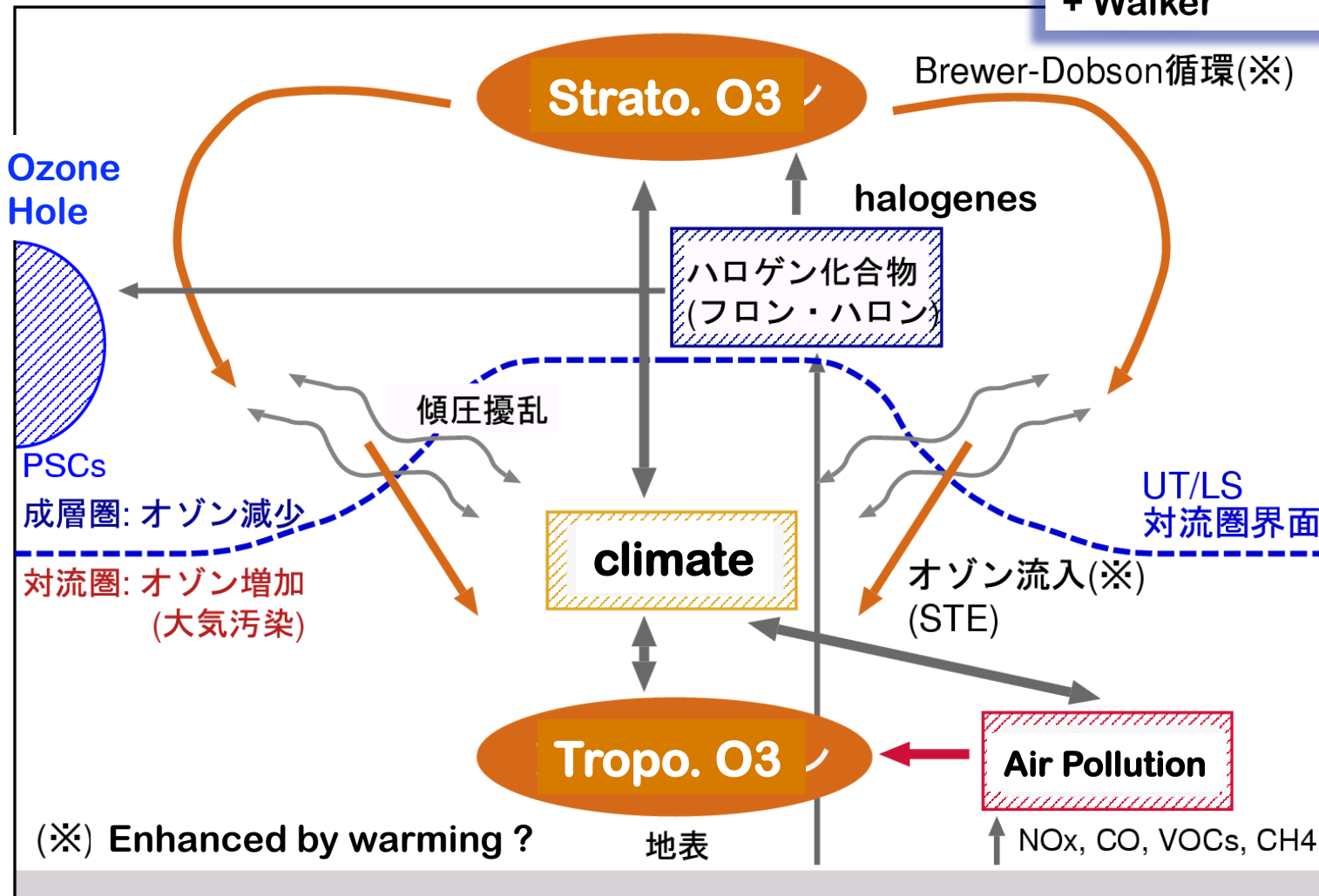
Future perspectives:

- Future climate change is likely to **modulate atmospheric chemistry**.
+ circulation (B.D. & Hadley), lightning NO_x, water vapor, ...
- Future climate change **may affect air quality**, but that process has not been clarified yet.



Circulation changes

- + Brewer-Dobson
- + Hadley
- + Walker



Future Plans ...

✓ AC&C related:

- AC&C Hindcast Experiments (1980-2009):
 - + trends in O₃, CH₄, related species and aerosols
 - + impacts of emissions, climate change, dynamical variation (ENSO/AO..)
- Future simulations toward IPCC-AR5:
 - + global O₃ 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

