

# Intercontinental Source-Receptor Relationships for Ozone Pollution



**Arlene M. Fiore**  
**([arlene.fiore@noaa.gov](mailto:arlene.fiore@noaa.gov))**



F. Dentener, O. Wild, C. Cuvelier, M. Schultz, C. Textor, M. Schulz, C. Atherton, D. Bergmann, I. Bey, G. Carmichael, R. Doherty, B. Duncan, G. Faluvegi, G. Folberth, M. Garcia Vivanco, M. Gauss, S. Gong, D. Hauglustaine, P. Hess, T. Holloway, L. Horowitz, I. Isaksen, D. Jacob, J. Jonson, J. Kaminski, T. Keating, A. Lupu, I. MacKenzie, E. Marmer, V. Montanaro, R. Park, K. Pringle, J. Pyle, M. Sanderson, S. Schroeder, D. Shindell, D. Stevenson, S. Szopa, R. Van Dingenen, P. Wind, G. Wojcik, J. West, S. Wu, G. Zeng, A. Zuber



**Task Force on Hemispheric  
Transport of Air Pollution**

40<sup>th</sup> Air Pollution Workshop and Symposium, Raleigh, NC, April 7, 2008

# Evidence of intercontinental transport at northern midlatitudes: 2001 Asian dust event

## Dust leaving the Asian coast in April 2001

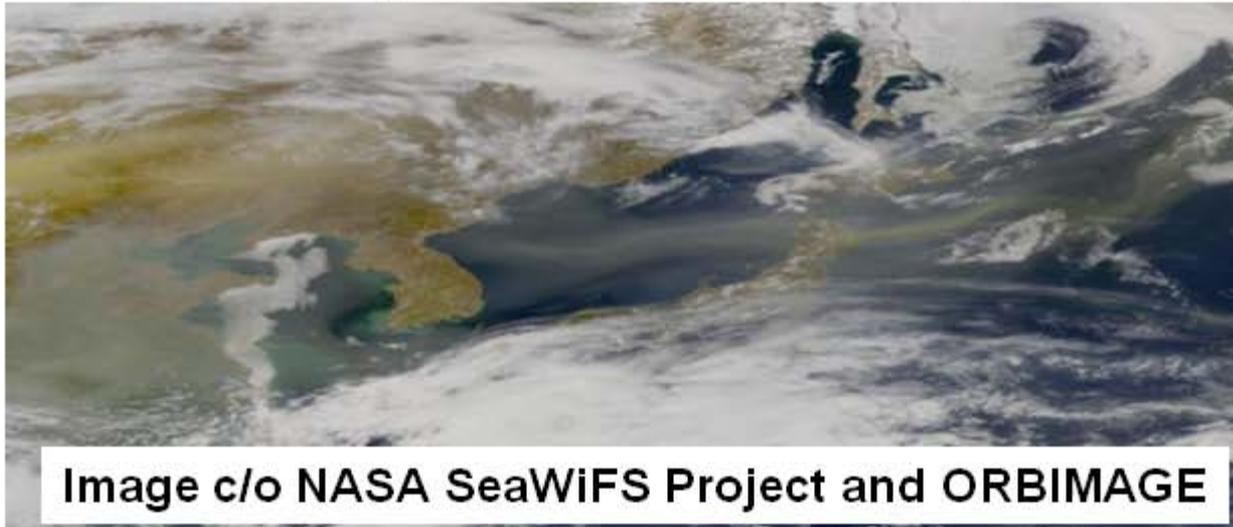


Image c/o NASA SeaWiFS Project and ORBIMAGE

## Reduced Visibility from Transpacific Transport of Asian Dust



Glen Canyon, Arizona

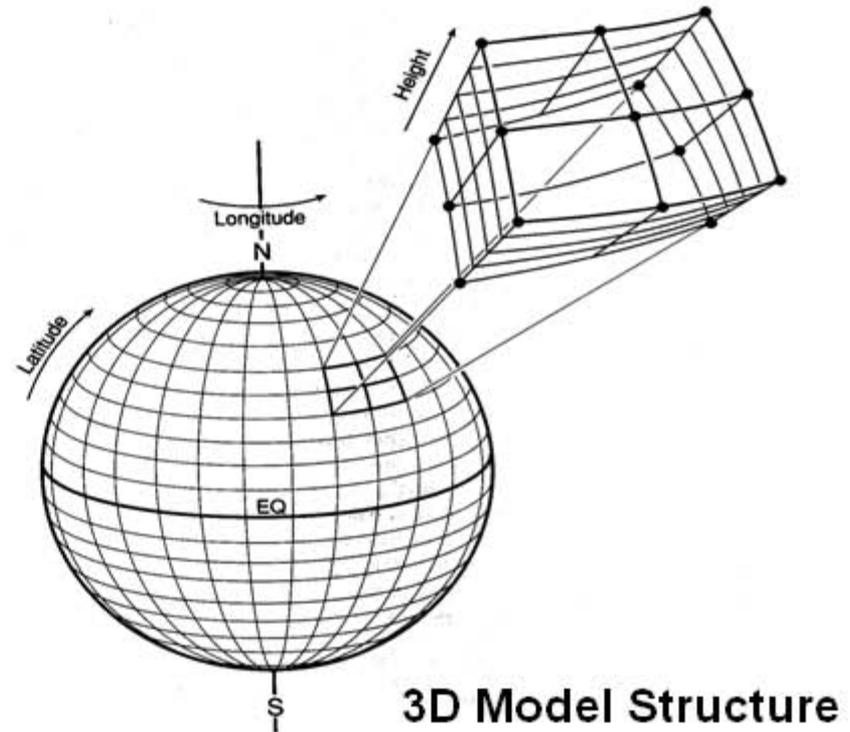
Clear Day

April 16, 2001

# How do we estimate source-receptor (S-R) relationships that describe hemispheric transport of air pollution?



**Measurements at remote sites?**  
(Monitoring site at Yosemite NP)



**Models with anthropogenic emissions reduced in source regions?**

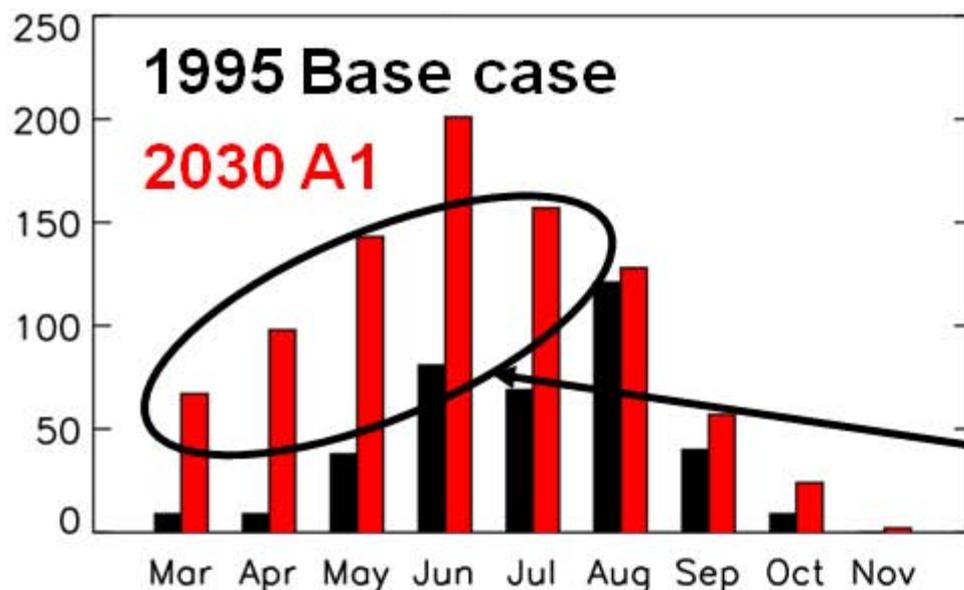
**Difficult to directly measure a region's contribution to pollution over a receptor region, particularly for ozone [e.g. Goldstein et al., 2004]**

# Regional control efforts (even under optimistic scenarios) may be offset by increases in hemispheric ozone pollution

By 2030 under the Current LEgislation (CLE) scenario, “the benefit of European emission control measures is... significantly counterbalanced by increasing global O<sub>3</sub> levels...”

[Szopa et al., GRL, 2006]

U.S. grid-square days with 1-5p.m. O<sub>3</sub> > 70 ppb



IPCC 2030 Scenario	Anthrop. NO <sub>x</sub> emis. Global	U.S.	Methane emis.
A1	+80%	-20%	+30%

GEOS-Chem Model (4°x5°)  
[Fiore et al., GRL, 2002]

longer O<sub>3</sub> season

U.S. air quality degrades despite domestic emissions controls (A1 2030)

**International approach to ozone abatement?**

# Convention on Long-Range Transboundary Air Pollution (CLRTAP)



**Task Force on Hemispheric  
Transport of Air Pollution**

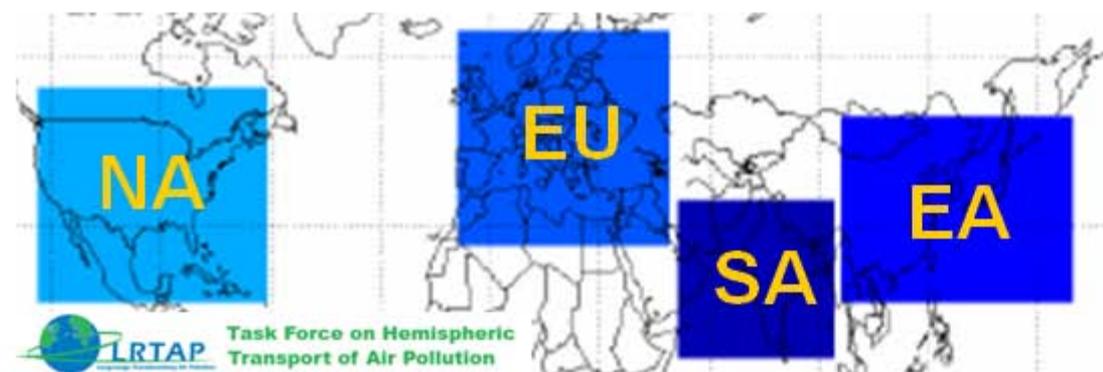
**Co-chairs: Terry Keating (U.S. EPA) and André Zuber (EC)**

**TF HTAP Mission: Develop a fuller understanding of hemispheric transport of air pollution to inform future negotiations under CLRTAP**

# Multi-model assessment involving >25 modeling groups

**OBJECTIVES:** Quantify S-R relationships for HTAP regions and assess uncertainties in these estimates

## HTAP S-R Regions



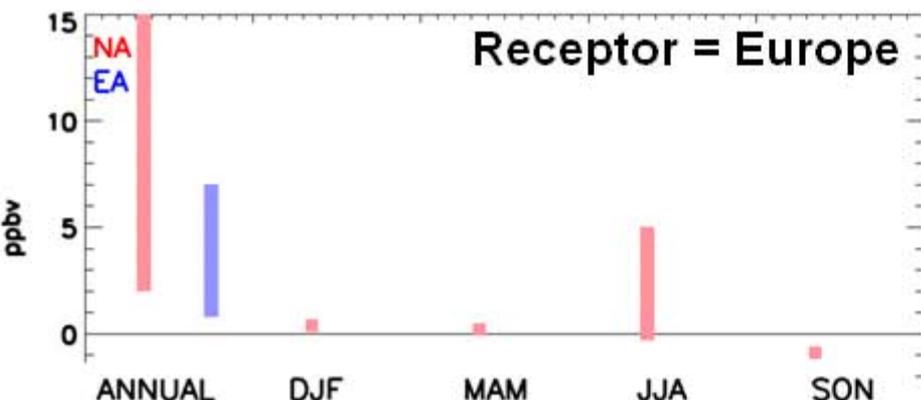
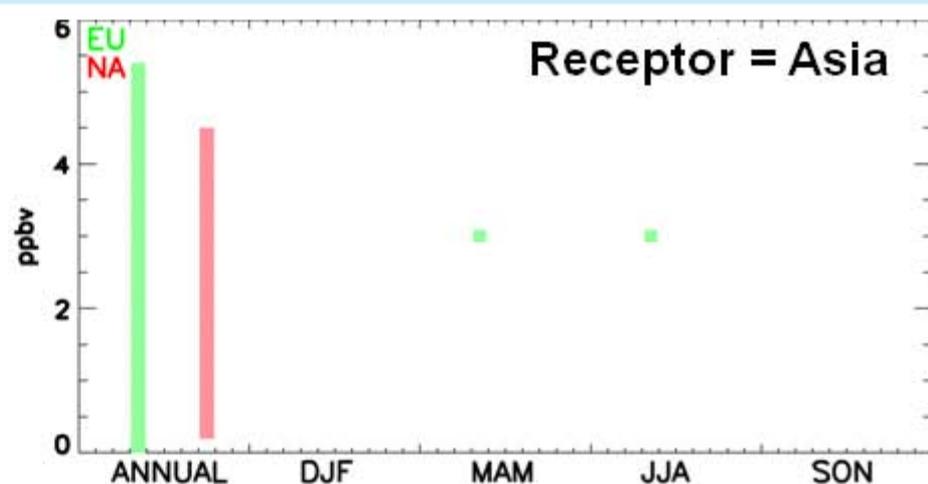
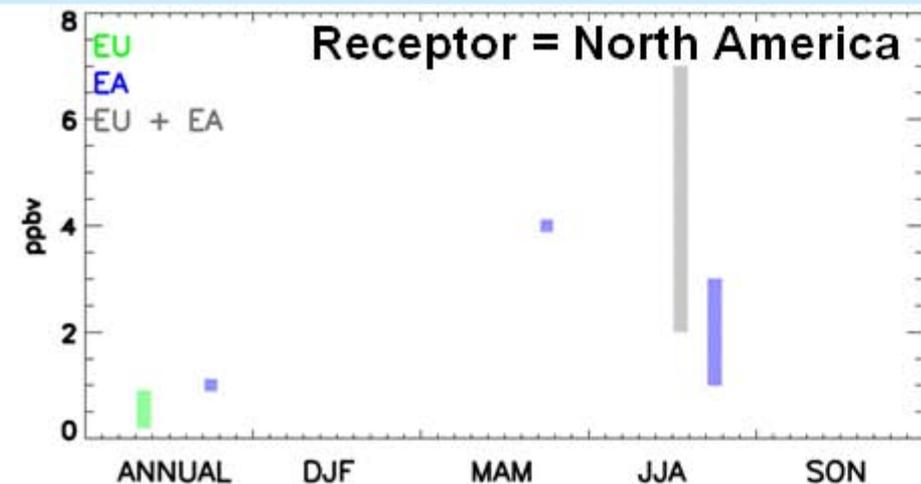
## Focus species:

- Ozone and precursors
- Aerosols and precursors
- Mercury
- Persistent Organic Pollutants
- Idealized Tracers
- Oxidized Nitrogen

**PRODUCTS:** **2007 Interim Report** to inform the review of the 1999 CLRTAP Gothenburg Protocol to abate acidification, eutrophication, and tropospheric ozone ([www.htap.org](http://www.htap.org)).

**2009 Assessment Report** to inform the CLRTAP on hemispheric air pollution and S-R relationships.

# Wide range in literature estimates of mean surface O<sub>3</sub> S-R relationships at northern mid-latitudes



Assessment hindered by different:

- 1) methods
- 2) regional definitions
- 3) reported metrics
- 4) years (meteorology)

→ Adopt a multi-model approach

→ Consistency across models

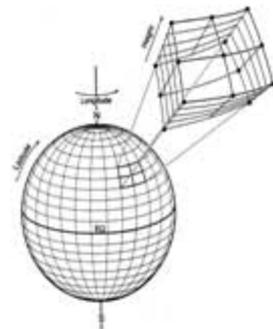
→ Examine all seasons

Estimates are from studies cited in TF HTAP [2007] Ch5, plus new work [Holloway et al., 2008; Duncan et al., 2008; Lin et al., 2008]

# Model Setup for TF HTAP Ozone S-R Study

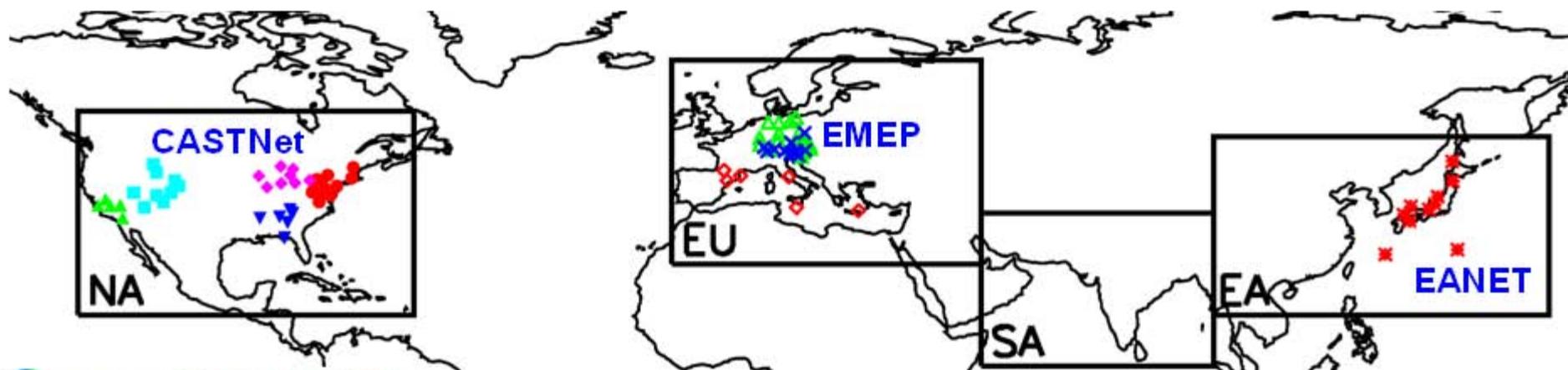
## BASE SIMULATION

- horizontal resolution of  $4^{\circ} \times 5^{\circ}$  or finer
- 2001 meteorology
- each group's best estimate for emissions in 2001
- methane set to a uniform value of 1760 ppb

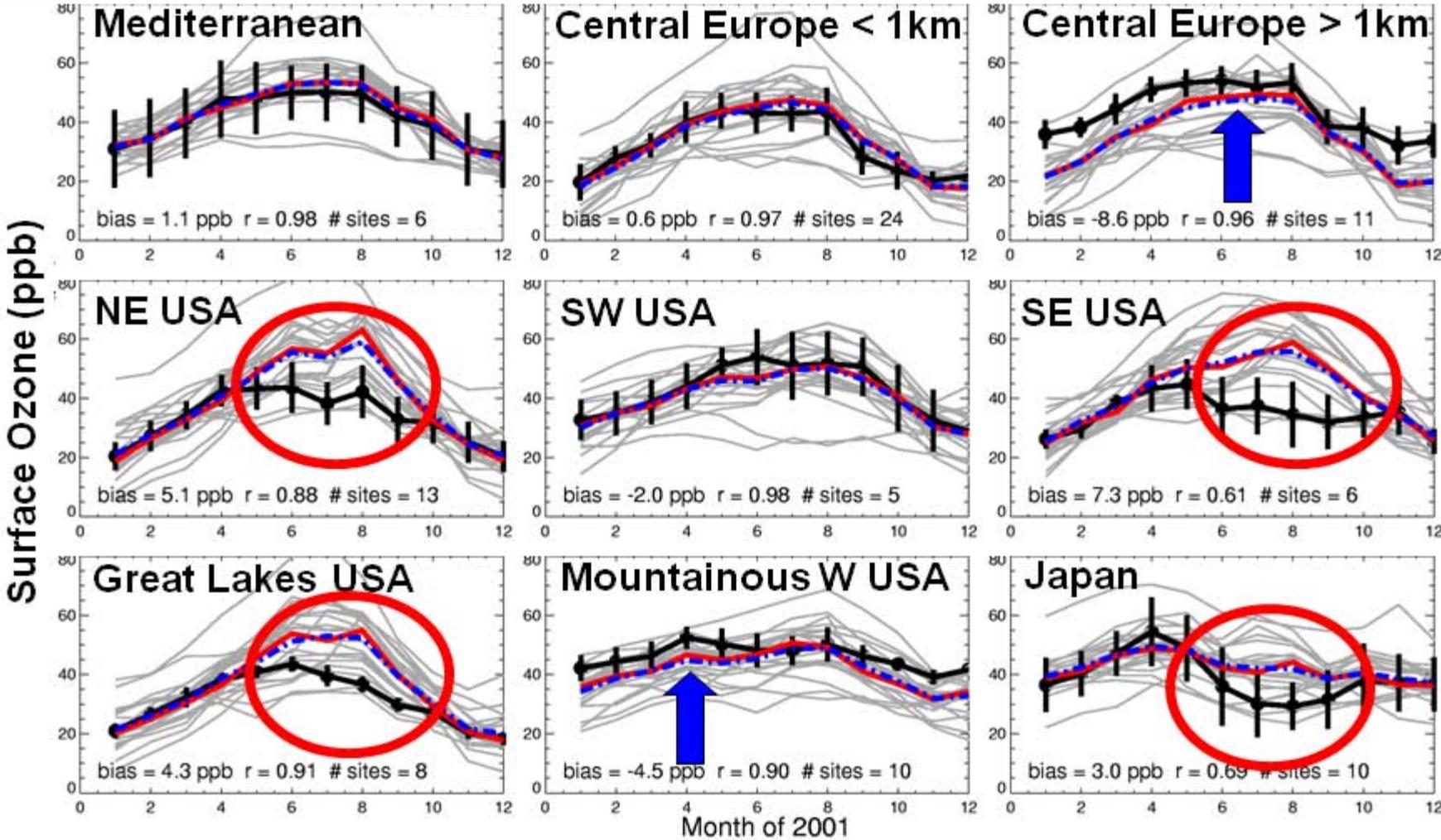


## SENSITIVITY SIMULATIONS (17 total) with sustained 20% decreases in

- regional anthrop.  $\text{NO}_x$ , CO, NMVOC emis. individually (4x3)
- regional anthrop. emis. of all  $\text{O}_3$  and aerosol precursors (4)
- global methane (to 1408 ppb)



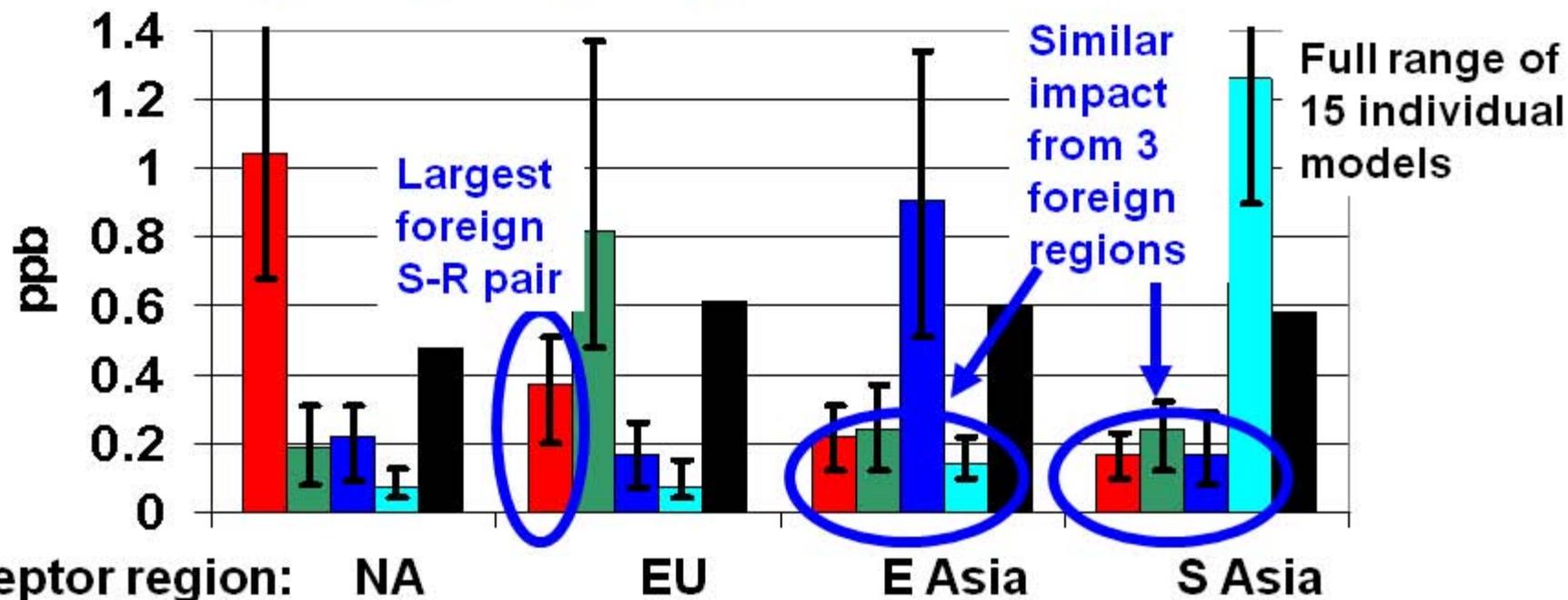
# Large inter-model range; 21-model mean generally captures observed monthly mean surface O<sub>3</sub>



- Many models biased **low at altitude**, **high over EUS+Japan in summer**
- Good springtime/late fall simulation

# Estimates of S-R relationships: Annual mean surface O<sub>3</sub> decrease from 20% reductions in anthropogenic NO<sub>x</sub>+CO+NMVOC emissions

Source region: ■ NA ■ EU ■ EA ■ SA ■ sum of 3 foreign regions

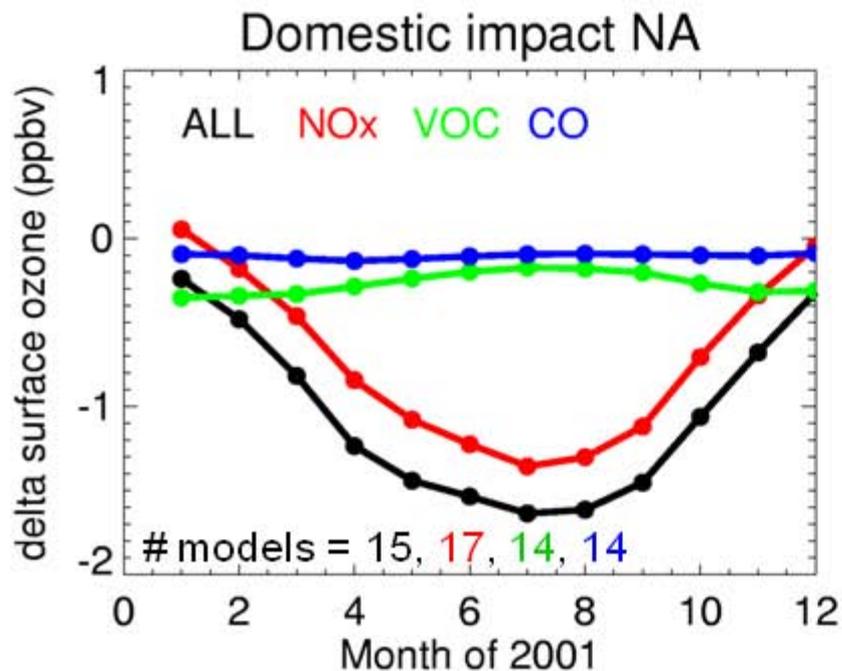


$$\frac{\Delta(-20\% \text{ foreign anthrop. emis.})}{-20\% \text{ domestic anthrop. emis.}} = 0.45-0.75$$

“import sensitivity”

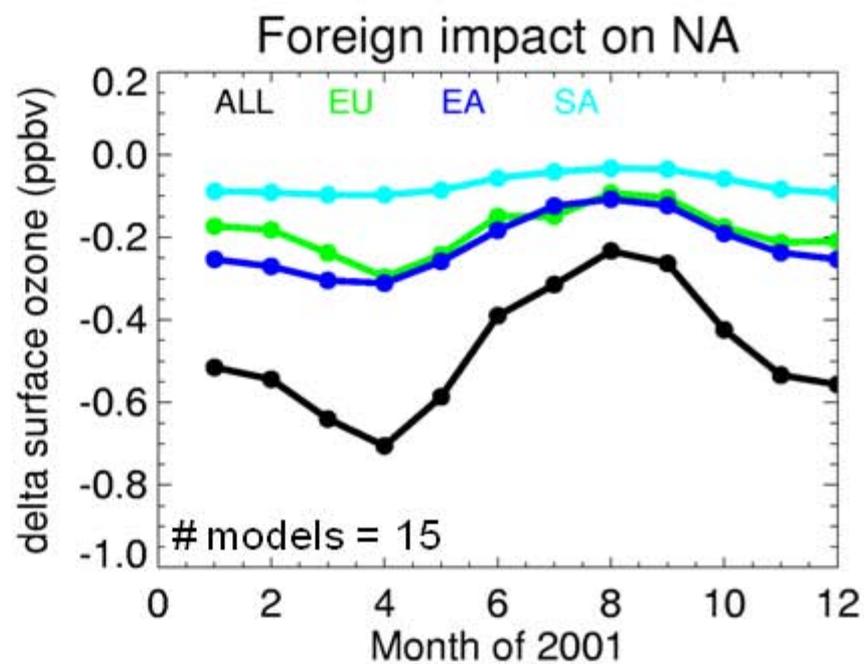
→ But... seasonality in sensitivity to domestic vs. foreign sources?

# Model ensemble mean surface O<sub>3</sub> response over NA to decreases in anthropogenic emissions of O<sub>3</sub> precursors



- Max influence in summer (NO<sub>x</sub>)
- Max NMVOC contribution in winter

-20% anthrop. NO<sub>x</sub>+CO+NMVOC emis.



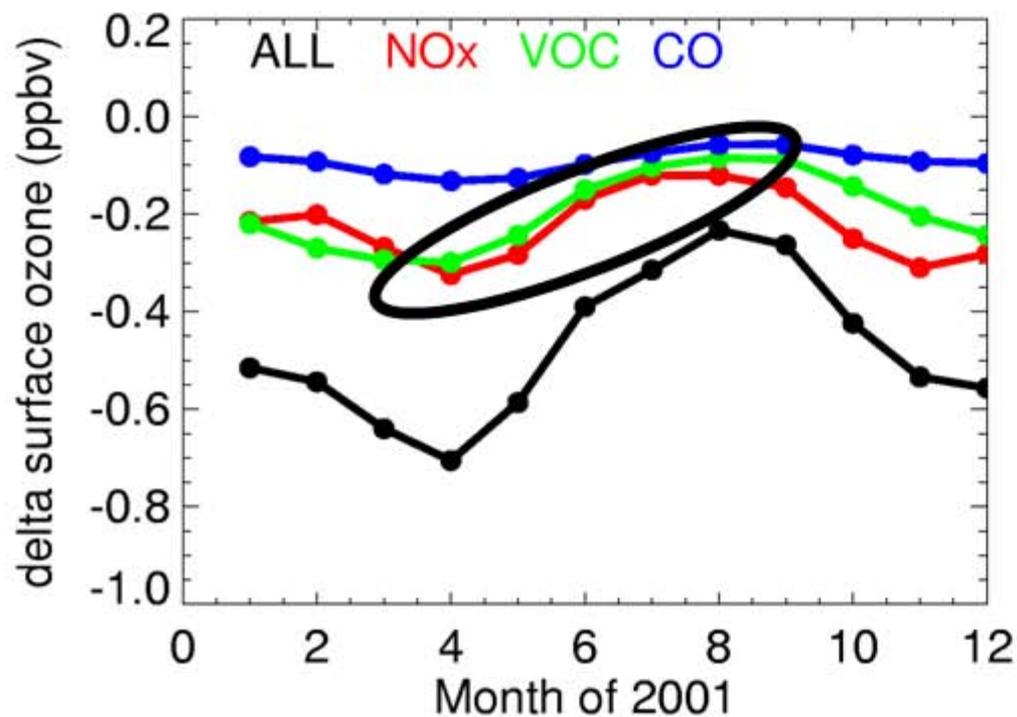
- Max foreign influence spring + late fall
- SA contributes least
- EU + EA contribute similarly Apr-Nov (but depends on model)

→ Import Sensitivity for NA in July ~ 0.2, in April ~ 0.6

# Model ensemble mean surface O<sub>3</sub> response over NA to decreases in anthropogenic emissions of O<sub>3</sub> precursors

## Foreign impact on NA

(sum of O<sub>3</sub> responses to -20% anthrop. emis. in the 3 foreign regions)



Model ensemble mean indicates a comparable influence from foreign NO<sub>x</sub> and NMVOC...

...but relative importance varies across models

Wide range in EU anthrop. NMVOC inventories used in the models

→ large uncertainty in the estimated response of NA O<sub>3</sub> to EU emissions

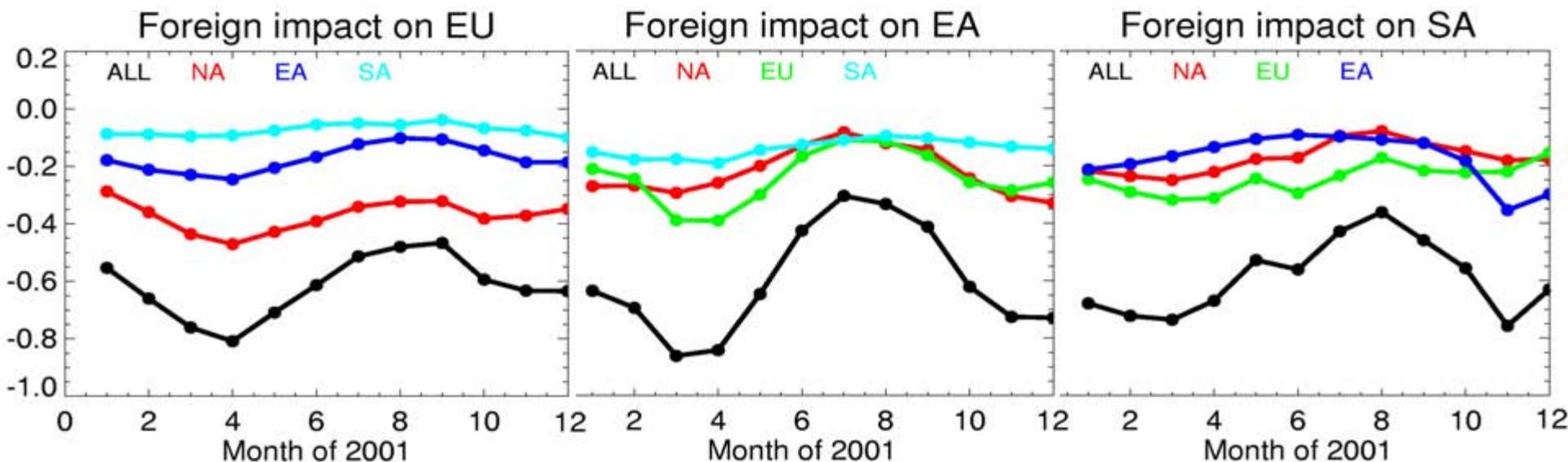
# Summary: Intercontinental S-R relationships for O<sub>3</sub> and conventional O<sub>3</sub> precursors

- **Identified robust estimates + key areas of uncertainty**
  - Model ensemble mean overestimate vs. obs in summer EUS and Japan
  - Robust: NA→EU largest; SA→others smallest
  - Uncertainty in relative roles of EU/EA → NA
  - Uncertainty in exported EU O<sub>3</sub> from EU AVOC emissions inventories

**Import sensitivities in other regions?**

**Comparison with prior literature?**

# Surface O<sub>3</sub> response to decreases in foreign anthropogenic emissions of O<sub>3</sub> precursors



## Robust conclusions:

→ NA > EA > SA on EU ozone

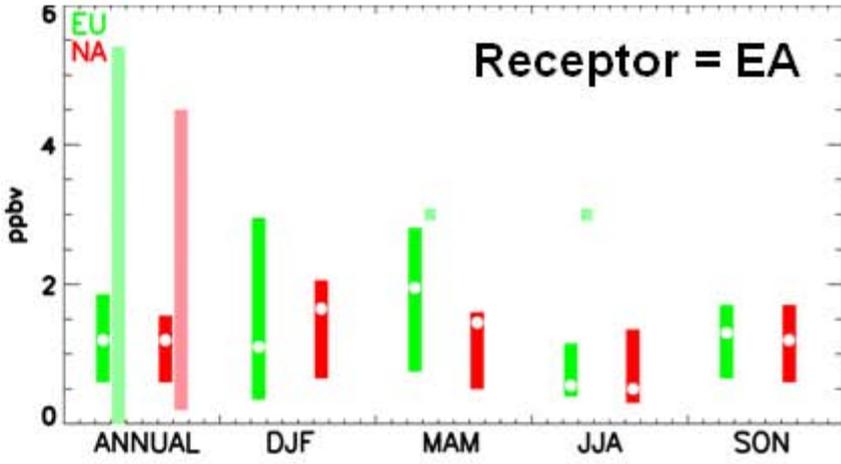
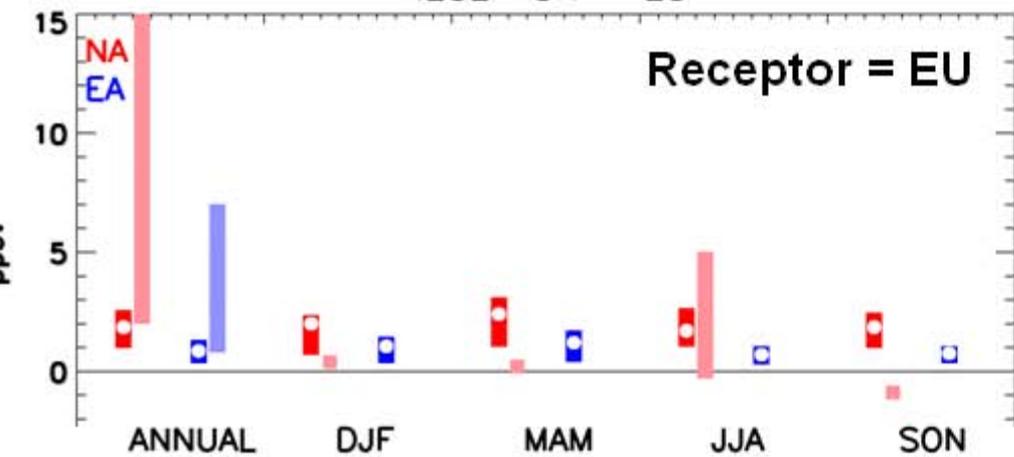
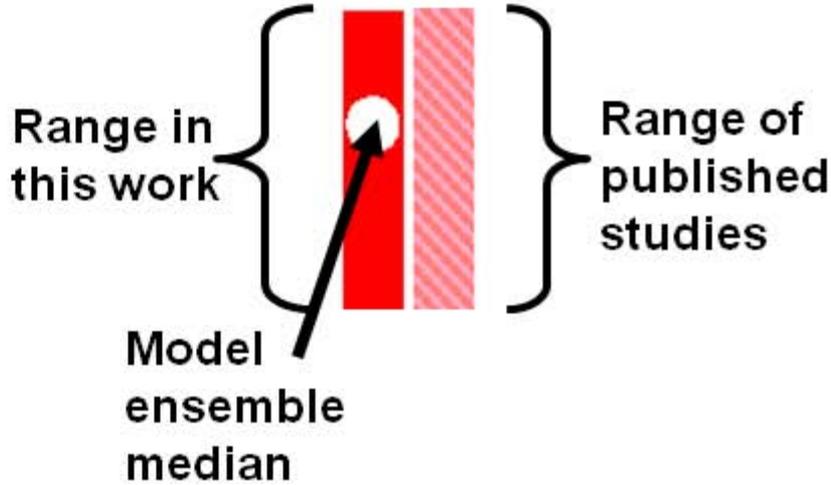
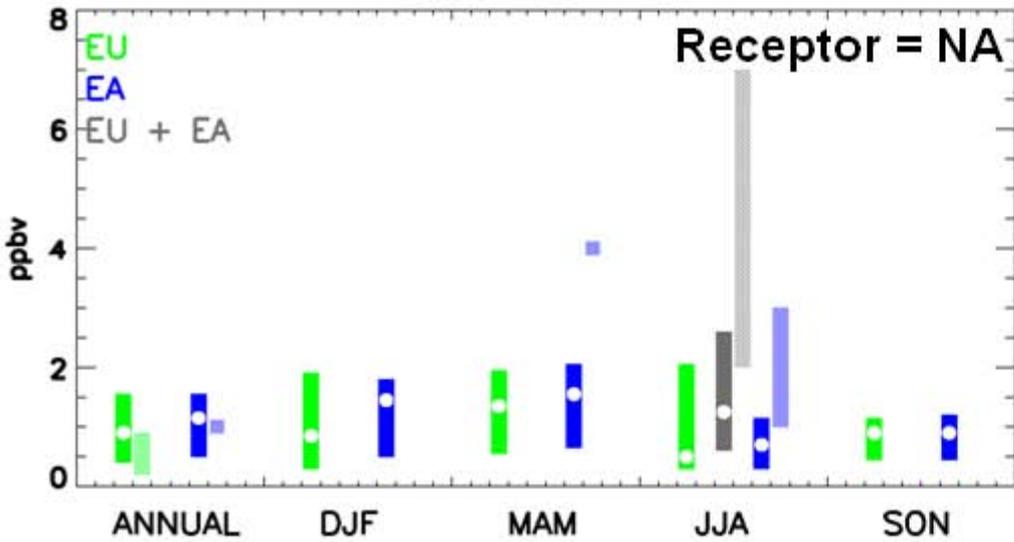
→ NA and EU often > SA or EA on each other;  
dominant contributor varies across models

→ Ozone over foreign regions generally least sensitive to SA emissions

Import sensitivities during month of max. domestic or foreign sensitivity:  
EU: 0.7 (APR) 0.3 (JUL) EA: 1.1 (MAR) 0.2 (JUL) SA: 0.5 (NOV)

# Range of estimated S-R relationships narrows from that in the literature with consistently applied HTAP approach

Assume linearity, scale response to -20% to 100% to estimate total contribution



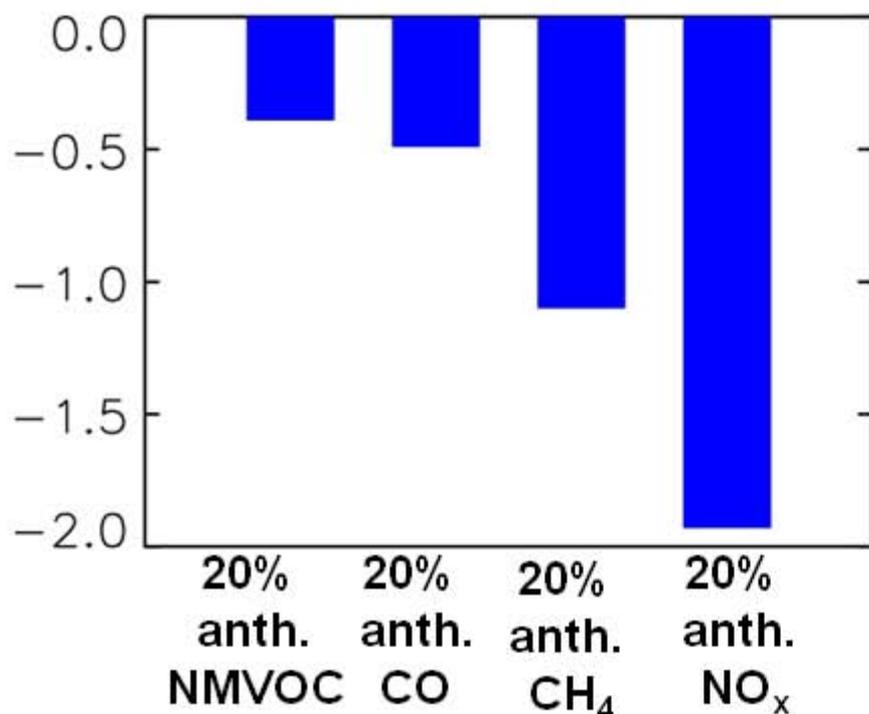
## Summary: Intercontinental S-R relationships for O<sub>3</sub> and conventional O<sub>3</sub> precursors

- **Identified robust estimates + key areas of uncertainty**
  - Uncertainty in exported EU O<sub>3</sub> from EU AVOC emissions inventories
  - Robust: NA→EU largest; SA→others smallest
  - Uncertainty in relative roles of EU/EA → NA; EU/NA→EA/SA
  - Model ensemble mean overestimate vs. obs in summer EUS and Japan
- **Estimated “import sensitivities”**
  - 0.2-0.3 July EA/NA/EU (max domestic production)
  - 0.6-0.9 April EA/NA/EU (max foreign influence)
  - 0.5 November SA (max foreign+domestic influence)
- **Narrowed range of estimates from that in the literature**

How does role of CH<sub>4</sub> compare with NO<sub>x</sub>+CO+NMVOC?

# Globally reducing methane decreases surface ozone everywhere

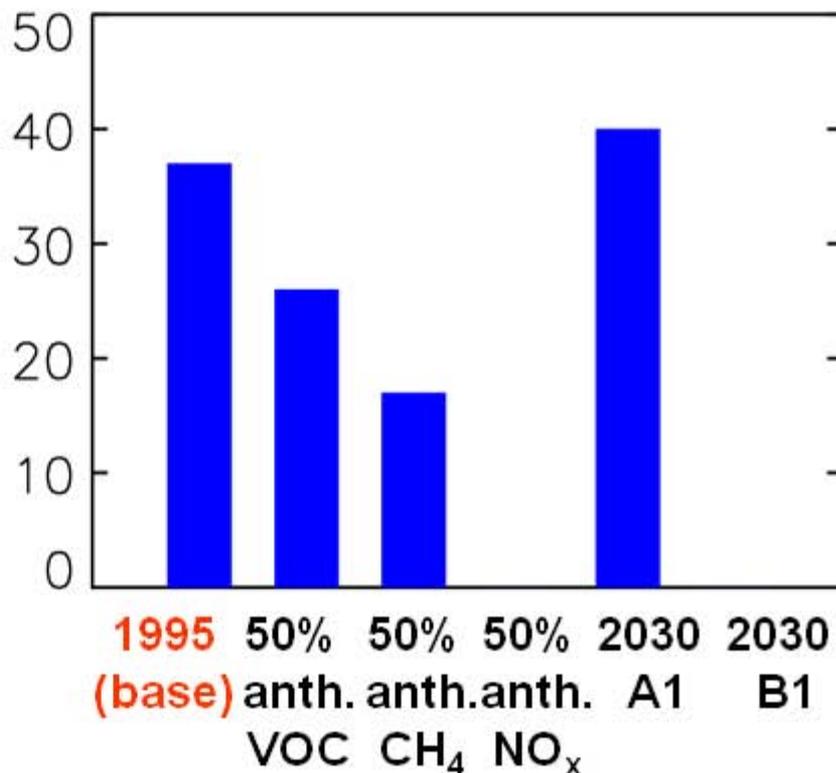
Change in population-weighted mean 8-hr daily max surface O<sub>3</sub> in 3-month "O<sub>3</sub> season" (ppbv)



Steady-state results from MOZART CTM

*West et al., GRL, 2007*

Number of U.S. summer grid-square days with O<sub>3</sub> > 80 ppbv



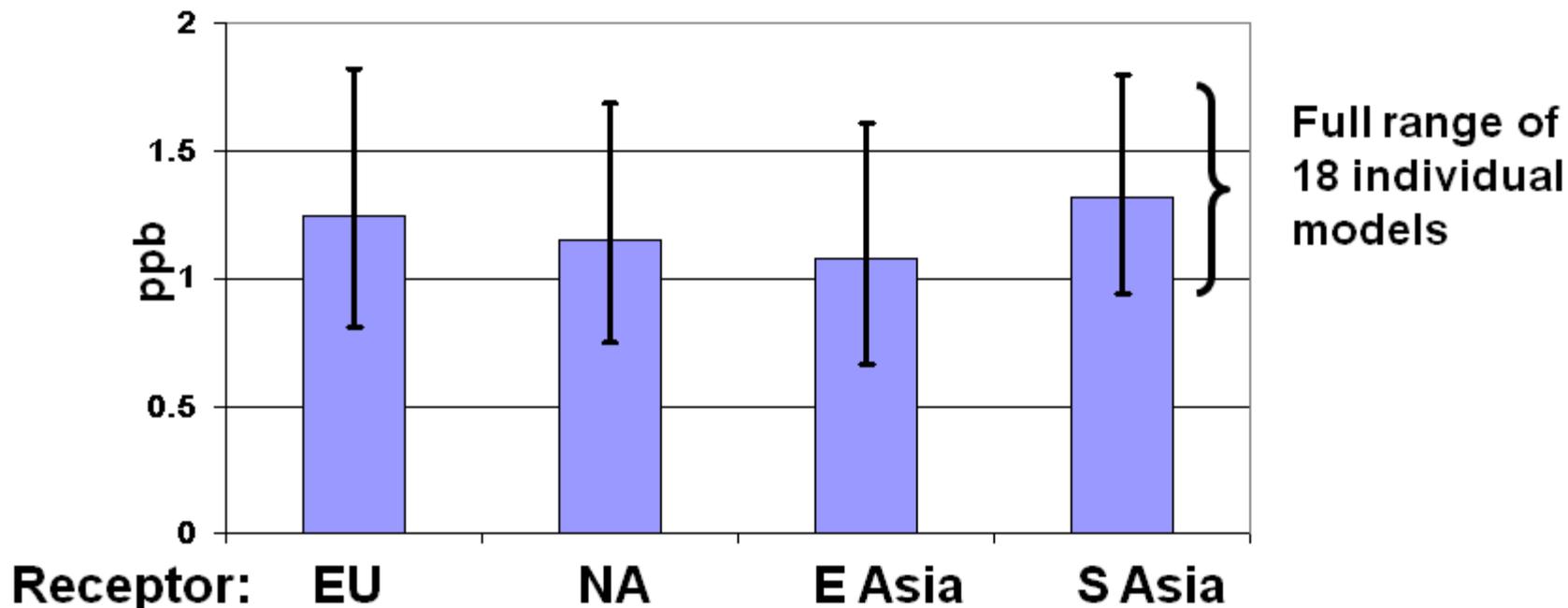
GEOS-Chem results

*Fiore et al., GRL, 2002*

→ Examine role of CH<sub>4</sub> vs. NO<sub>x</sub>+CO+NMVOC in multi-model context

# Surface ozone decreases similarly in all HTAP regions when global methane is reduced

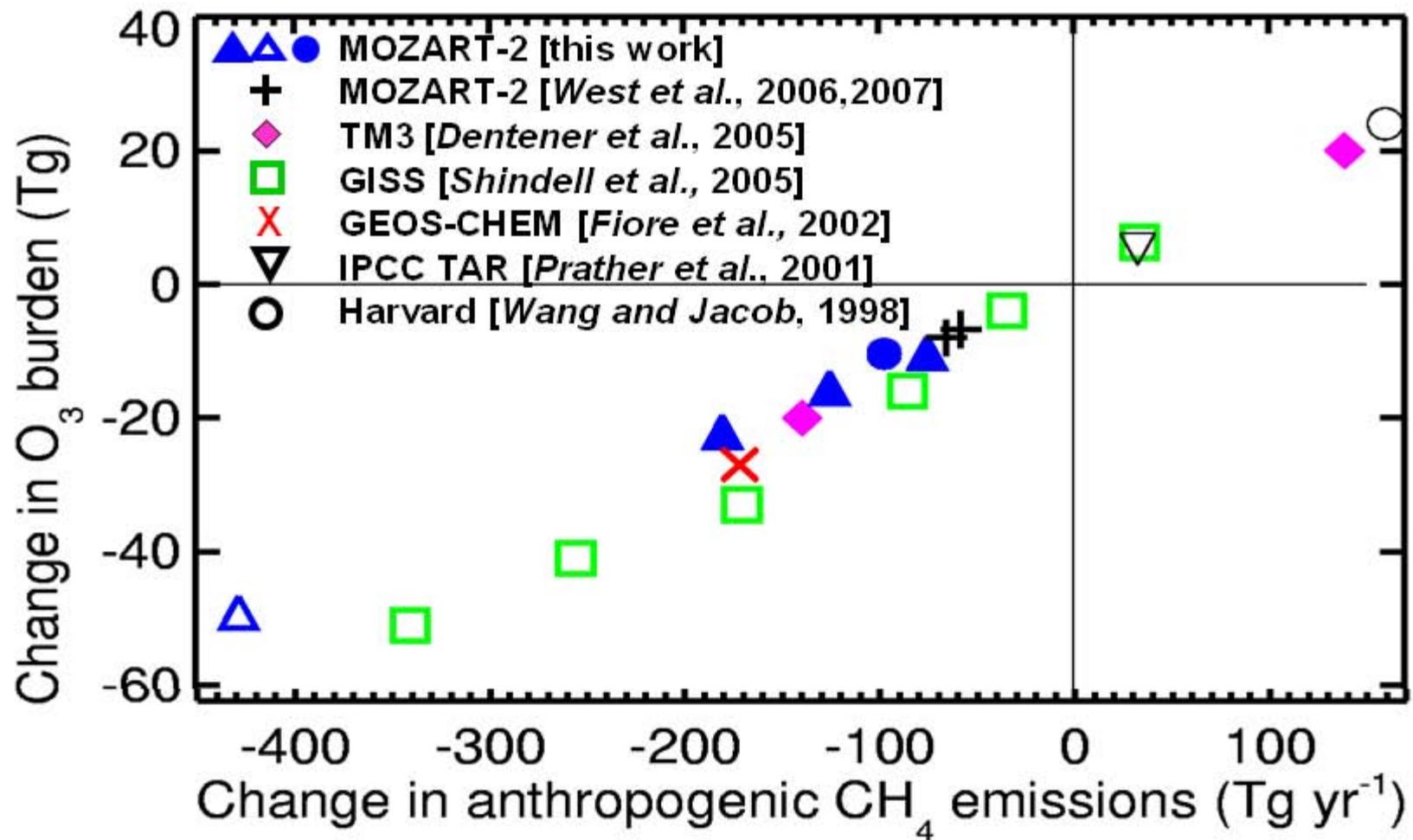
## ANNUAL MEAN OZONE DECREASE FROM 20% DECREASE IN GLOBAL METHANE



- ~1 ppbv O<sub>3</sub> decrease over all NH receptor regions
- Consistent with prior studies

Can we scale these results to 20% reductions in regional anthrop. CH<sub>4</sub> emissions to compare with conventional O<sub>3</sub> precursors?

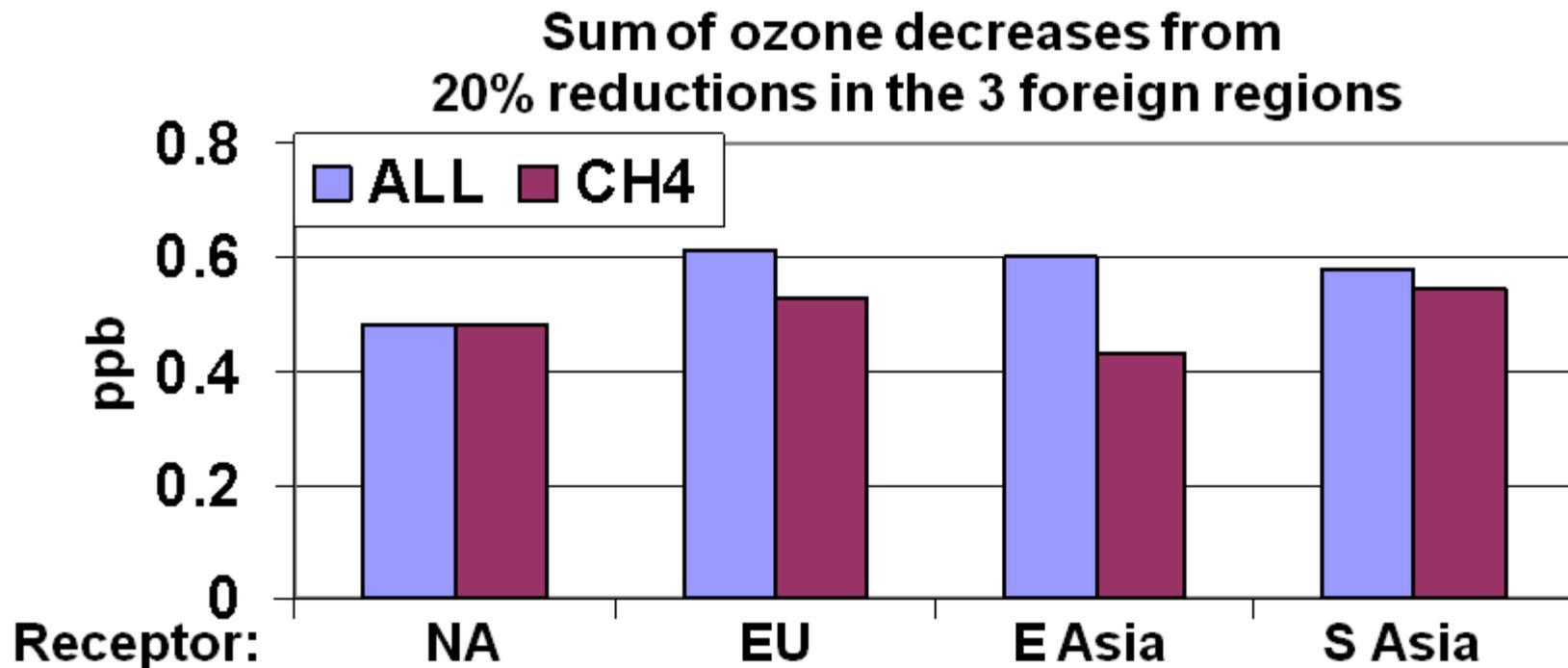
# Tropospheric O<sub>3</sub> responds approximately linearly to anthropogenic CH<sub>4</sub> emission changes across models



Anthropogenic CH<sub>4</sub> contributes ~50 Tg (~15%) to tropospheric O<sub>3</sub> burden  
~5 ppbv to surface O<sub>3</sub>

# Comparable annual mean O<sub>3</sub> decrease from 20% reductions in foreign CH<sub>4</sub> emissions and NO<sub>x</sub>+NMVOC+CO

Use CH<sub>4</sub> simulation + EDGAR 3.2 FT2000 CH<sub>4</sub> emissions [Olivier et al., 2005] to estimate O<sub>3</sub> response to -20% regional anthrop. CH<sub>4</sub> emissions



# Conclusions: Intercontinental S-R Relationships for O<sub>3</sub>

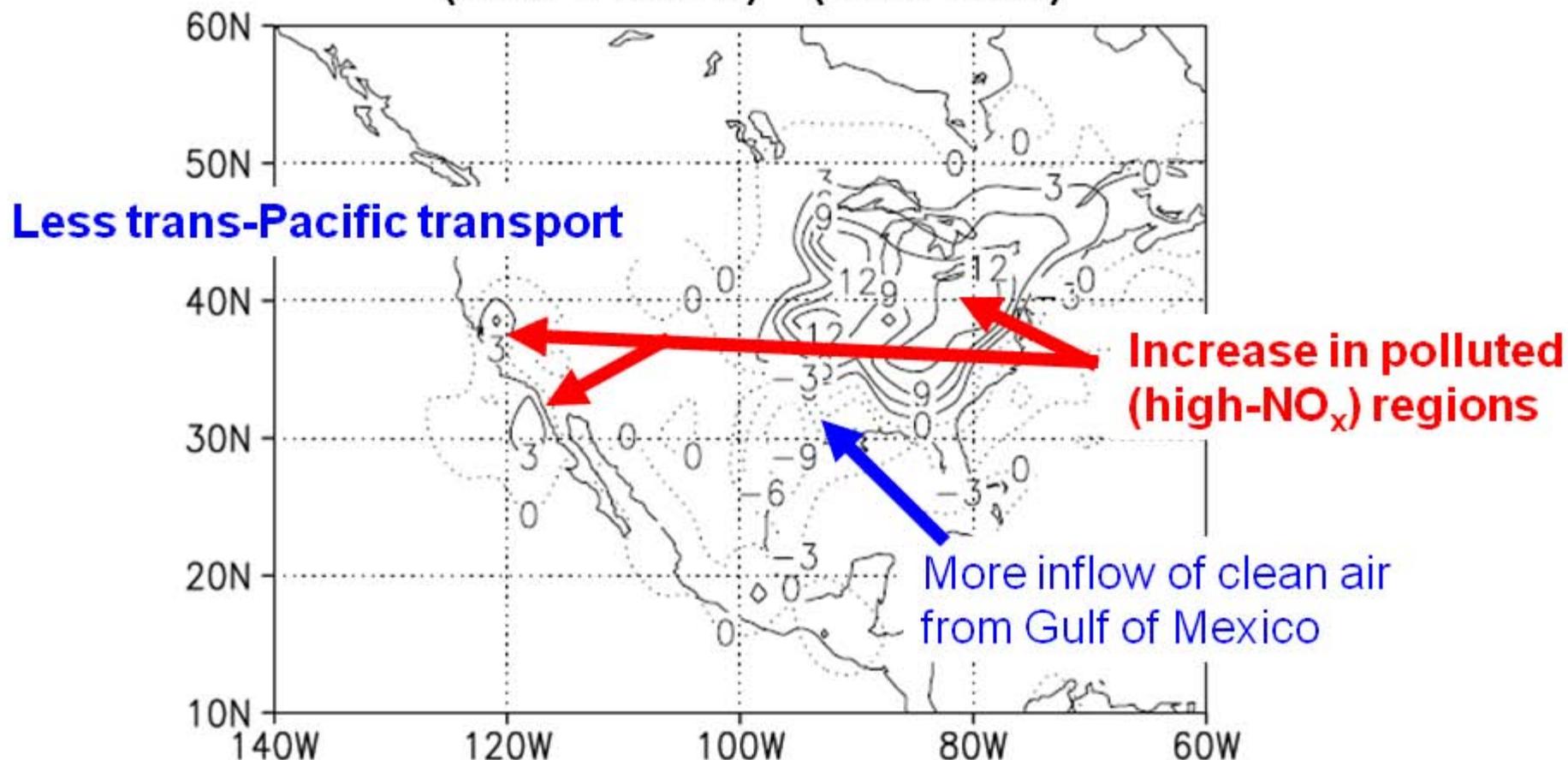
- Identified robust estimates + key areas of uncertainty
  - Model ensemble mean overestimate vs. obs in summer EUS and Japan
  - Robust: NA→EU largest; SA→others smallest
  - Uncertainty in relative roles of EU/EA → NA
  - Uncertainty in exported EU O<sub>3</sub> from EU AVOC emissions inventories
- Estimated “import sensitivities”
  - 0.2-0.3 July EA/NA/EU (max domestic production)
  - 0.6-0.9 April EA/NA/EU (max foreign influence)
  - 0.5 November SA (max foreign+domestic influence)
- Narrowed range of estimates from that in the literature under HTAP setup
- Combined reductions in regional anthropogenic CH<sub>4</sub> + conventional O<sub>3</sub> precursors roughly double the O<sub>3</sub> decrease over foreign regions

**How might climate change affect hemispheric transport of air pollution?**

**Are our S-R relationships consistent with rising Asian emissions driving a 0.1-0.5 ppb yr<sup>-1</sup> surface O<sub>3</sub> increase (as derived from observations)?**

# Global background ozone may decrease in a warmer, more humid climate

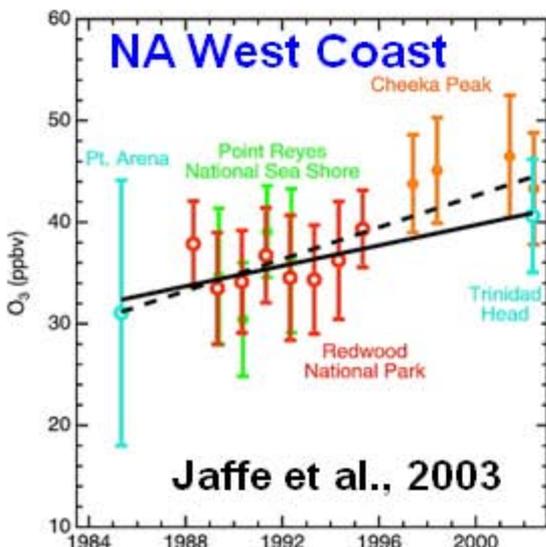
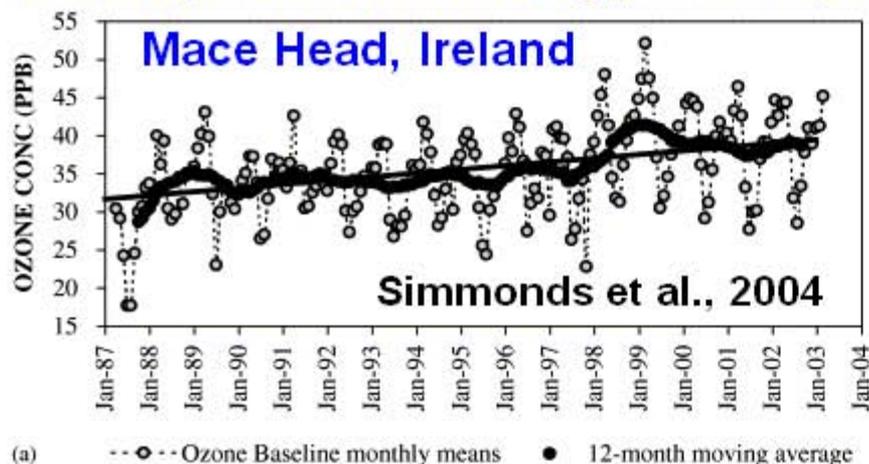
Mean annual change in number of days where daily max 8-hr  $O_3$  > 80 ppbv (2090-2100 A1) – (1990-2000)



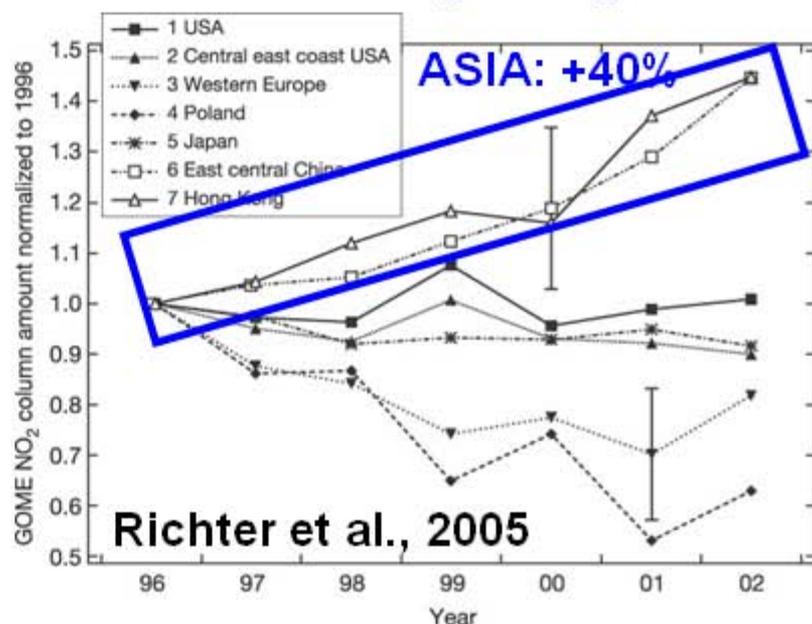
MOZART-2 global tropospheric chemistry model with meteorology from NCAR climate model [Murazaki and Hess, *J. Geophys. Res.*, 2006]

# Apply S-R relationships to address hypothesis of rising background O<sub>3</sub> driven by increasing Asian emissions

OBSERVED: +0.1-0.5 ppb yr<sup>-1</sup>  
in background surface O<sub>3</sub> [TF HTAP, 2007]



SPACE-BASED NO<sub>2</sub> → NO<sub>x</sub> EMISSIONS



Assuming +10% yr<sup>-1</sup> Asian emissions,  
our results imply an O<sub>3</sub> increase  
over NA and EU that falls at the low end  
of observed rise at the most

OUR CAVEATS:

- assumes SA+EA, + other emissions follow NO<sub>x</sub>
- continental-avg vs. "west coast" obs

Fig 3.6 from TF HTAP [2007]