

Estimating Intercontinental Source-Receptor Relationships for Ozone Pollution



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

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Example of intercontinental transport at northern midlatitudes: 2001 Asian dust event

Dust leaving the Asian coast in April 2001

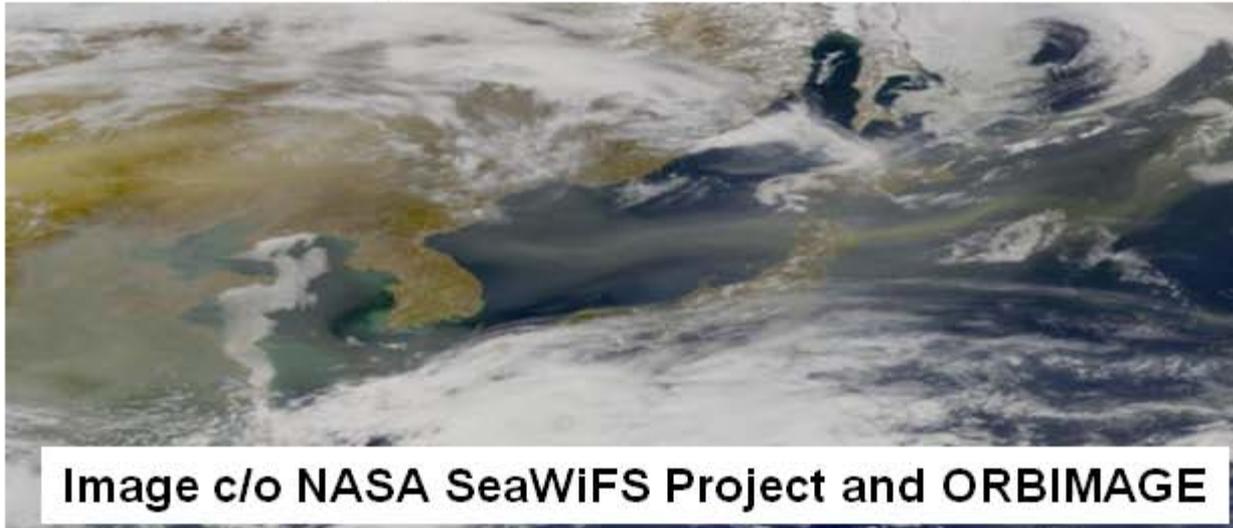


Image c/o NASA SeaWiFS Project and ORBIMAGE

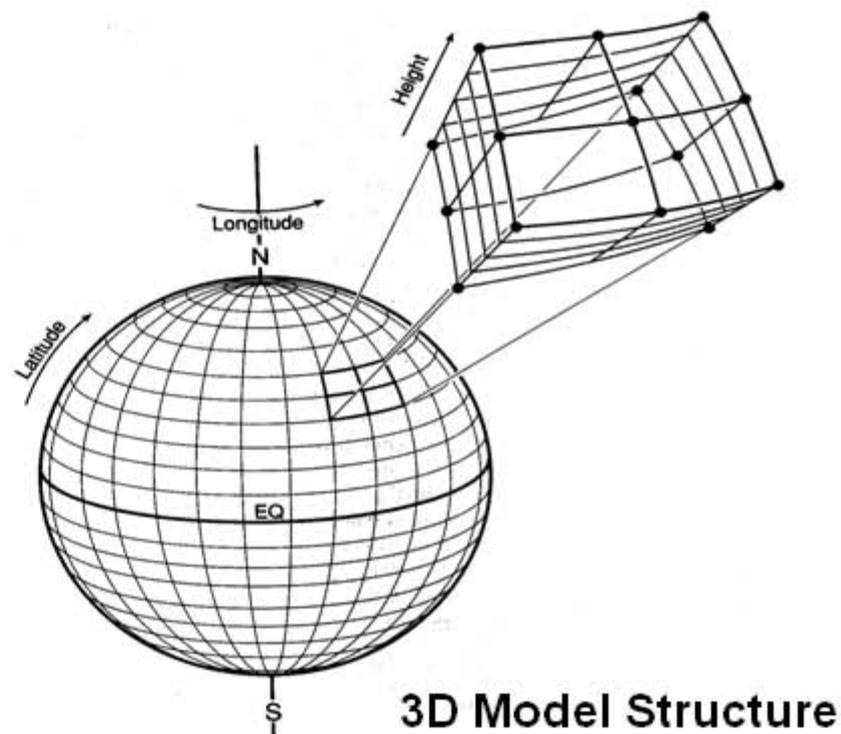
Reduced Visibility from Transpacific Transport of Asian Dust



How do we estimate source-receptor relationships that describe hemispheric transport of air pollution?



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



Models with domestic anthropogenic emissions turned off?

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

Hemispheric Transport of Ozone

Stratospheric O₃

Stratosphere

~12 km

h ν

O₃

Free Troposphere

Hemispheric Pollution

Direct Intercontinental Transport

Boundary layer

(0-3 km)

NO₂

NO

OH

HO₂

VOC CH₄ CO

NO_x

VOC

O₃

air pollution (smog)

air pollution (smog)

NO_x

VOC

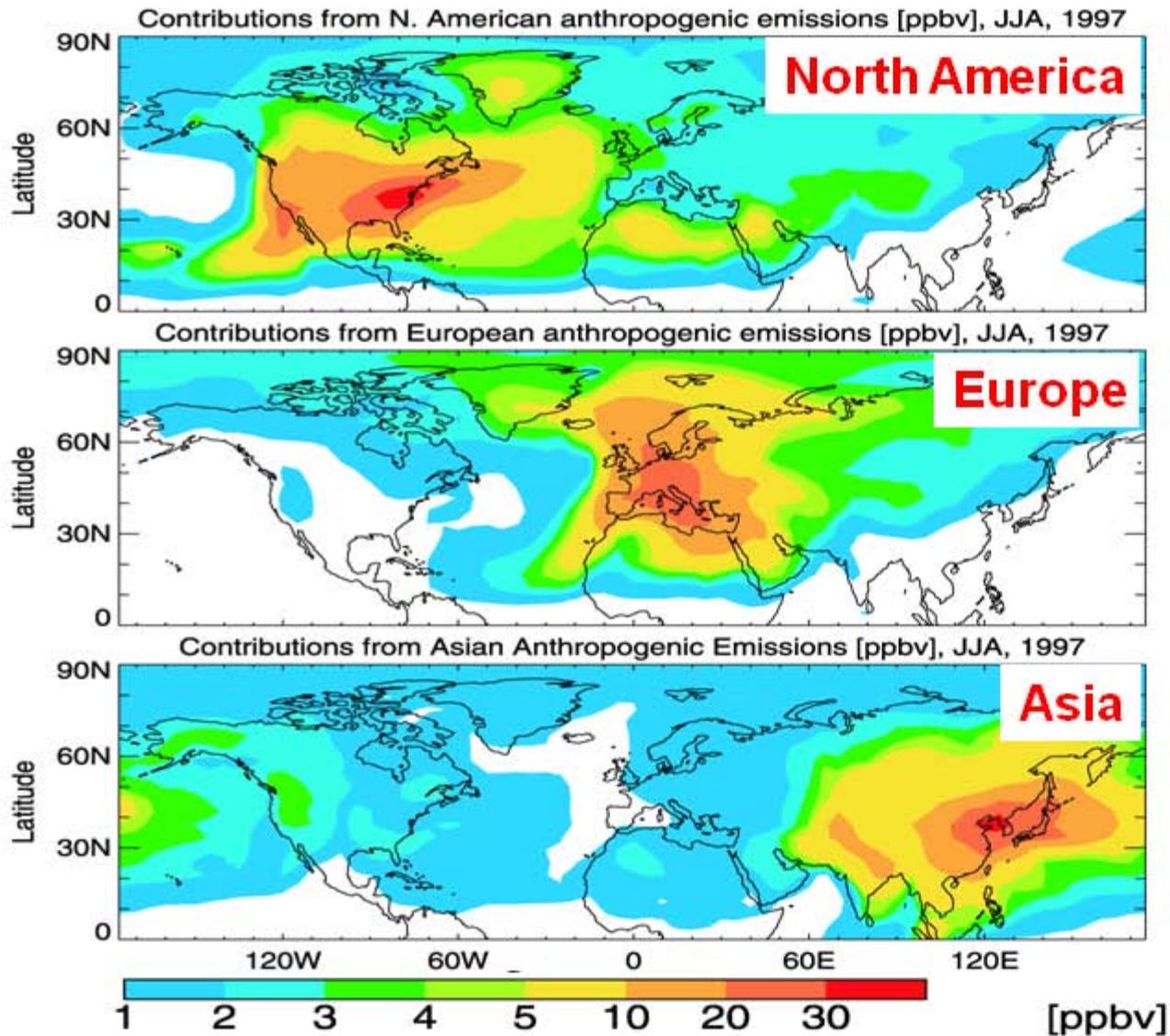
O₃

CONTINENT 1

OCEAN

CONTINENT 2

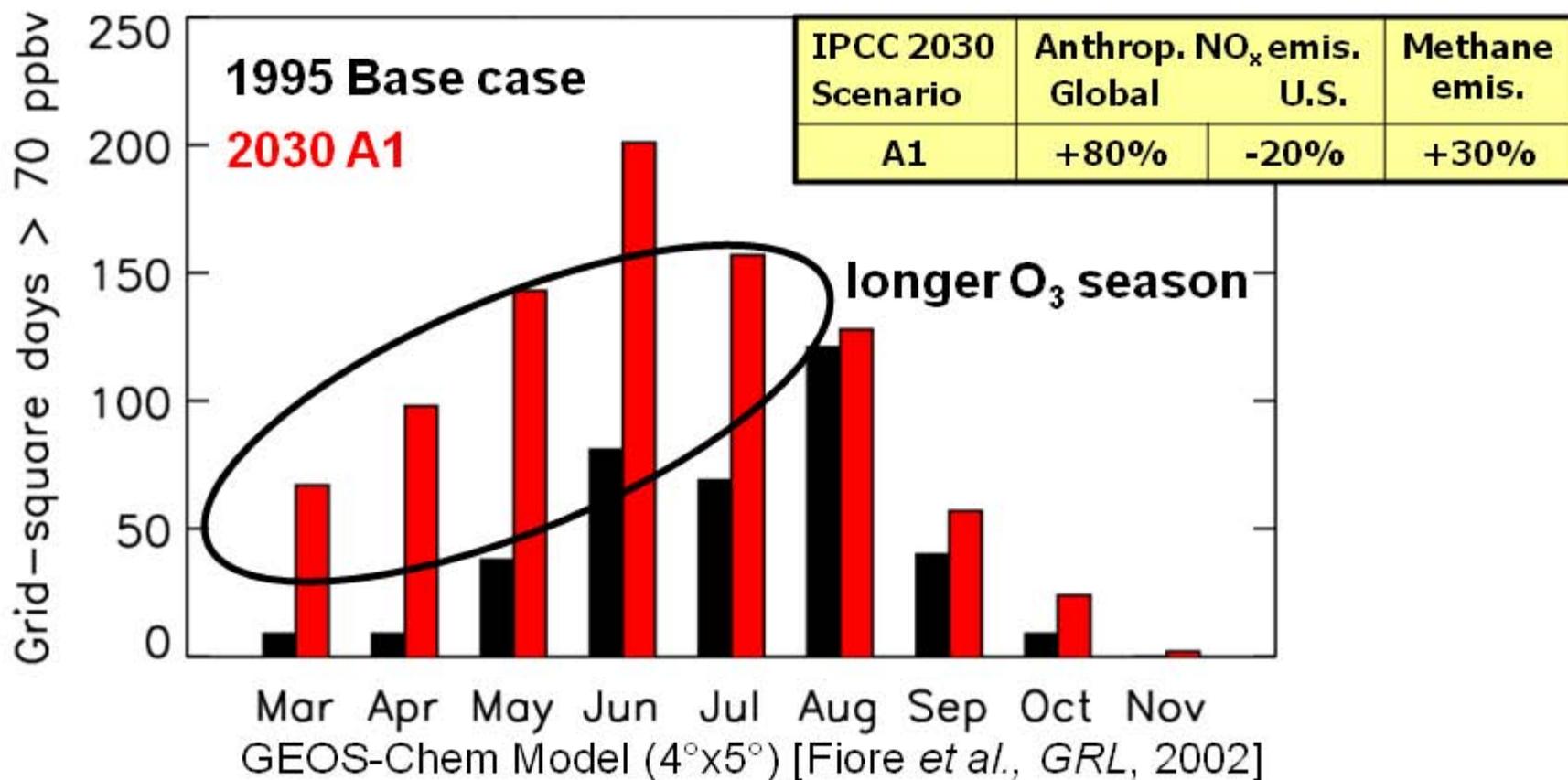
Hemispheric scale contribution of major source regions to NH surface ozone in summer



Estimated with model simulations that zero out anthropogenic emissions of O_3 precursors within the source region

GEOS-Chem Model
4°x5° horizontal resolution
Li et al., JGR, 2002

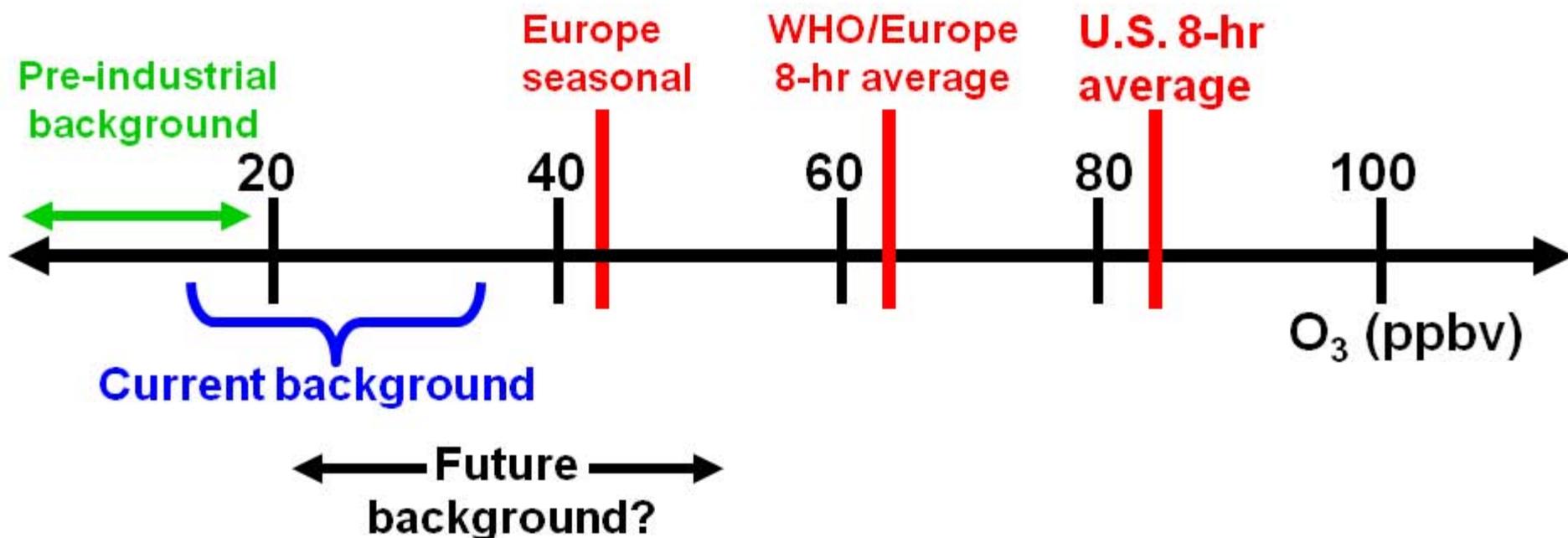
Changes in global anthropogenic emissions affect regional air quality



Rising global emissions may offset U.S. efforts to reduce pollution

Rising background O₃ has implications for attaining air quality standards

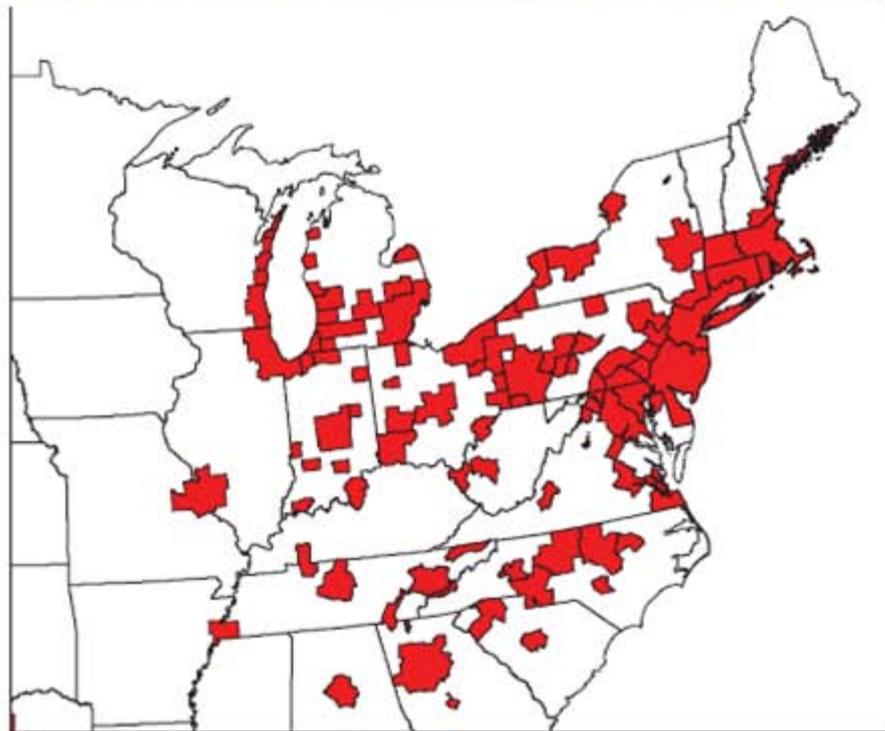
Surface O₃ background appears to be rising
[e.g. *Lin et al.*, 2000; *Jaffe et al.*, 2003, 2005;
Vingarzan, 2004; *EMEP/CCC-Report 1/2005*]



The U.S. smog problem is spatially widespread, affecting >100 million people [U.S. EPA, 2004]

OZONE

Nonattainment Areas (2001-2003 data)

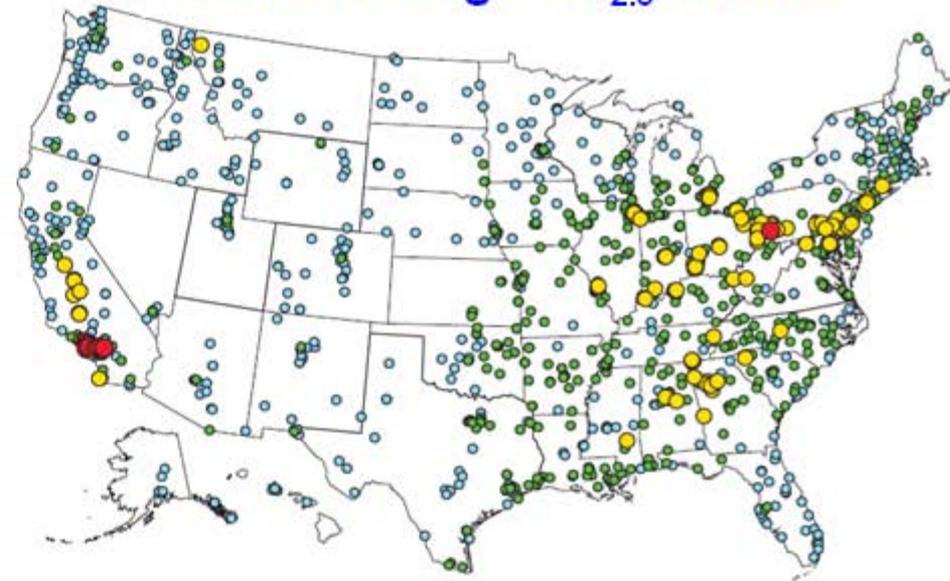


4th highest daily max 8-hr O₃ > 84 ppbv

U.S. EPA, 2006

AEROSOLS (particulate matter)

Annual Average PM_{2.5} in 2003



Concentration range (μg/m³)

■ ≤ 10

■ 10.1 - 15

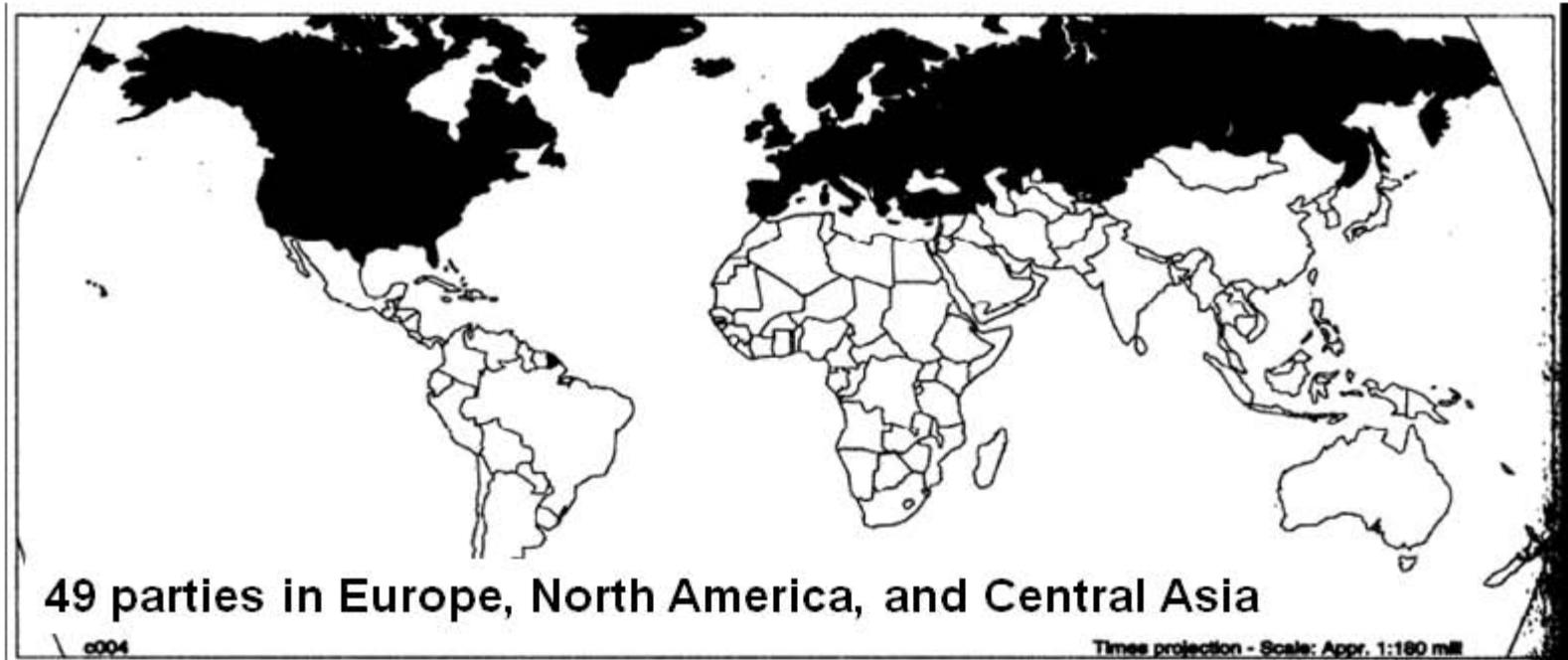
■ 15.1 - 20

■ > 20

} Exceeds standard

U.S. EPA, 2004

Convention on Long-Range Transboundary Air Pollution (CLRTAP)

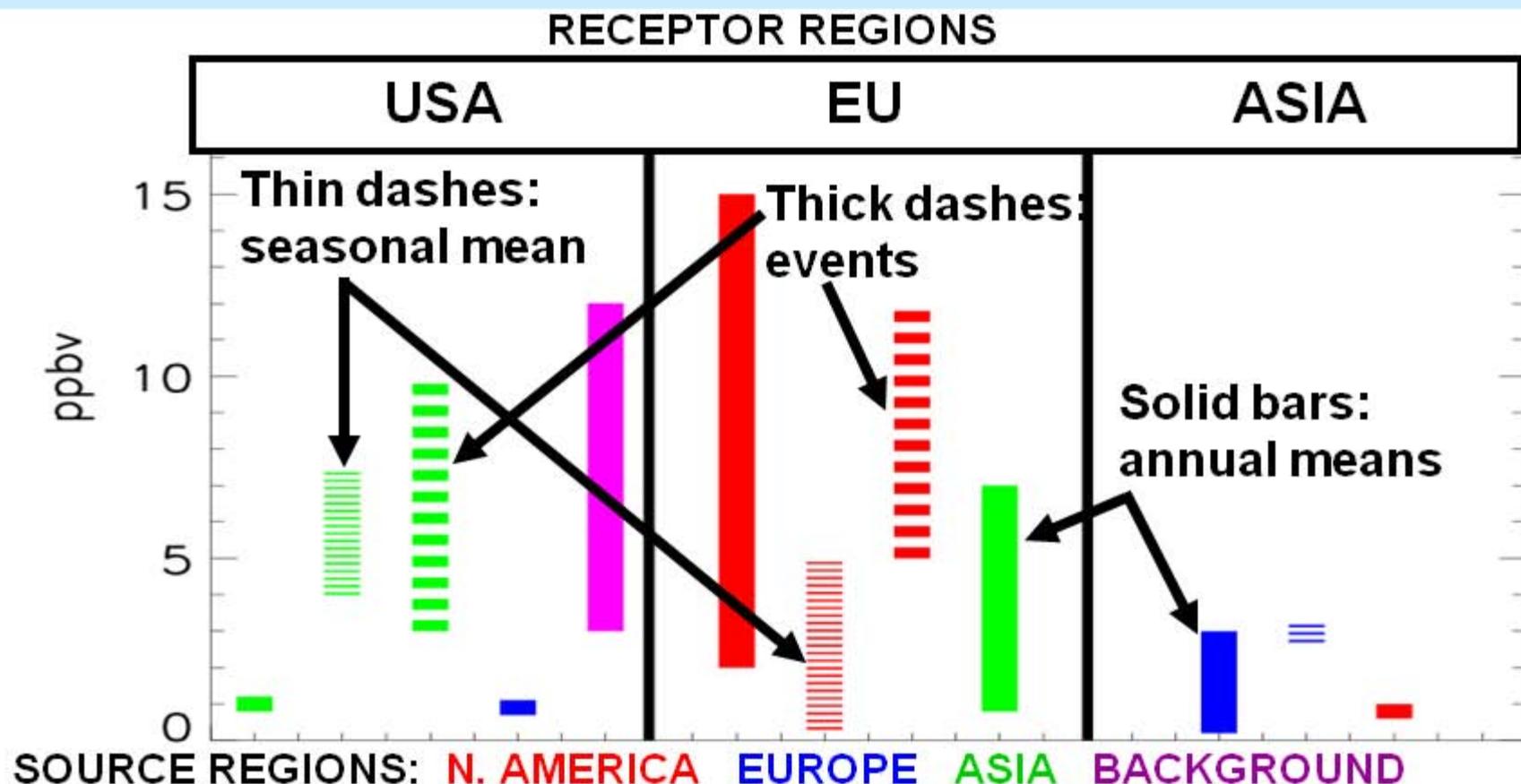


**Task Force on Hemispheric
Transport of Air Pollution**

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

Literature Estimates of Surface O₃ Enhancements at Northern Mid-latitudes from Hemispheric Transport



Estimates are from studies cited in current draft of TF HTAP interim report Ch5, updated from tables in *Holloway et al., ES&T, 2003* and *Fiore et al., EM, 2003*

Difficult to conduct meaningful assessment due to differences in:
1) methods 2) regional definitions 3) reported metrics

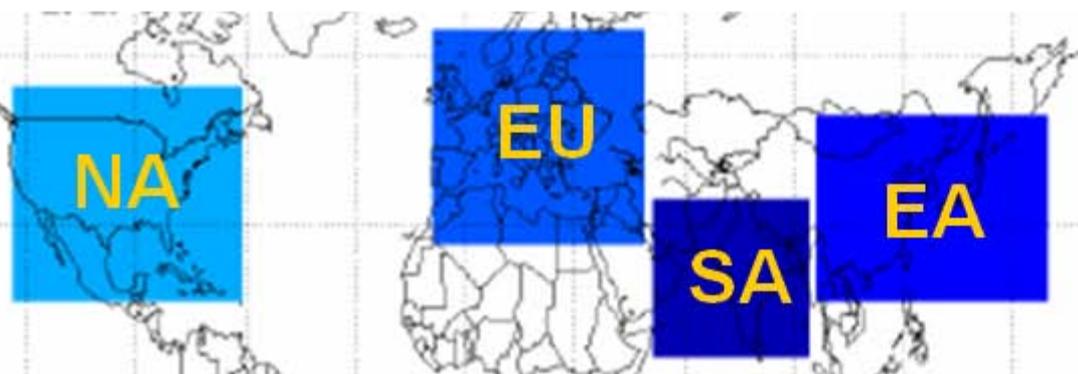
Multi-model assessment involving ~20 modeling groups



Task Force on Hemispheric
Transport of Air Pollution

OBJECTIVES: Quantify source-receptor relationships for HTAP regions and assess uncertainties in these estimates

HTAP Source-Receptor Regions



Focus species:

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

PRODUCTS: **2007 Interim Report** to inform the review of the 1999 CLRTAP Gothenburg Protocol to abate acidification, eutrophication, and tropospheric ozone.

(**draft report available for comment at www.htap.org**)

2009 Assessment Report to inform the CLRTAP on hemispheric air pollution and source-receptor relationships.

14 modeling groups have submitted results for HTAP ozone intercomparison (March, 2007)

Model	Institute
GEOS-Chem (2x2.5)	Harvard University, Cambridge, USA
MOZART-2	GFDL, Princeton, NJ, USA
STOCHEM	Hadley Centre, Met Office, UK
CAMCHEM	NCAR, Boulder, CO, USA
INCA	IPSL, Paris, France
LLNL-IMPACT	LLNL, Livermore, CA, USA
EMEP	Norwegian Met. Inst. Oslo, Norway
OsloCTM2	Oslo University, Norway
FRSGCUCI	NCAS, University of Cambridge, UK
UM-CAM	NCAS, University of Cambridge, UK
TM5-JRC	European Commission, JRC, Italy
MOZECH	Research Centre Juelich, Germany
GEOS-Chem (4x5)	CIEMAT, Madrid, Spain
GEM-AQ	AMDAL/CRESS York University, Canada

Overview of HTAP Model Intercomparison for Ozone



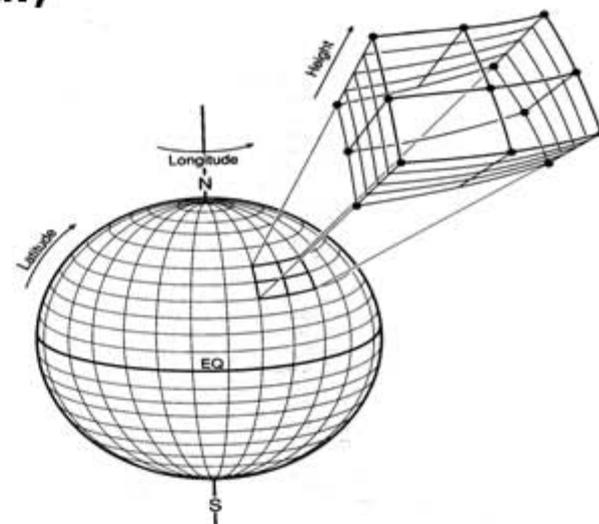
APPROACH: Conduct base simulation with 3-D models

- 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

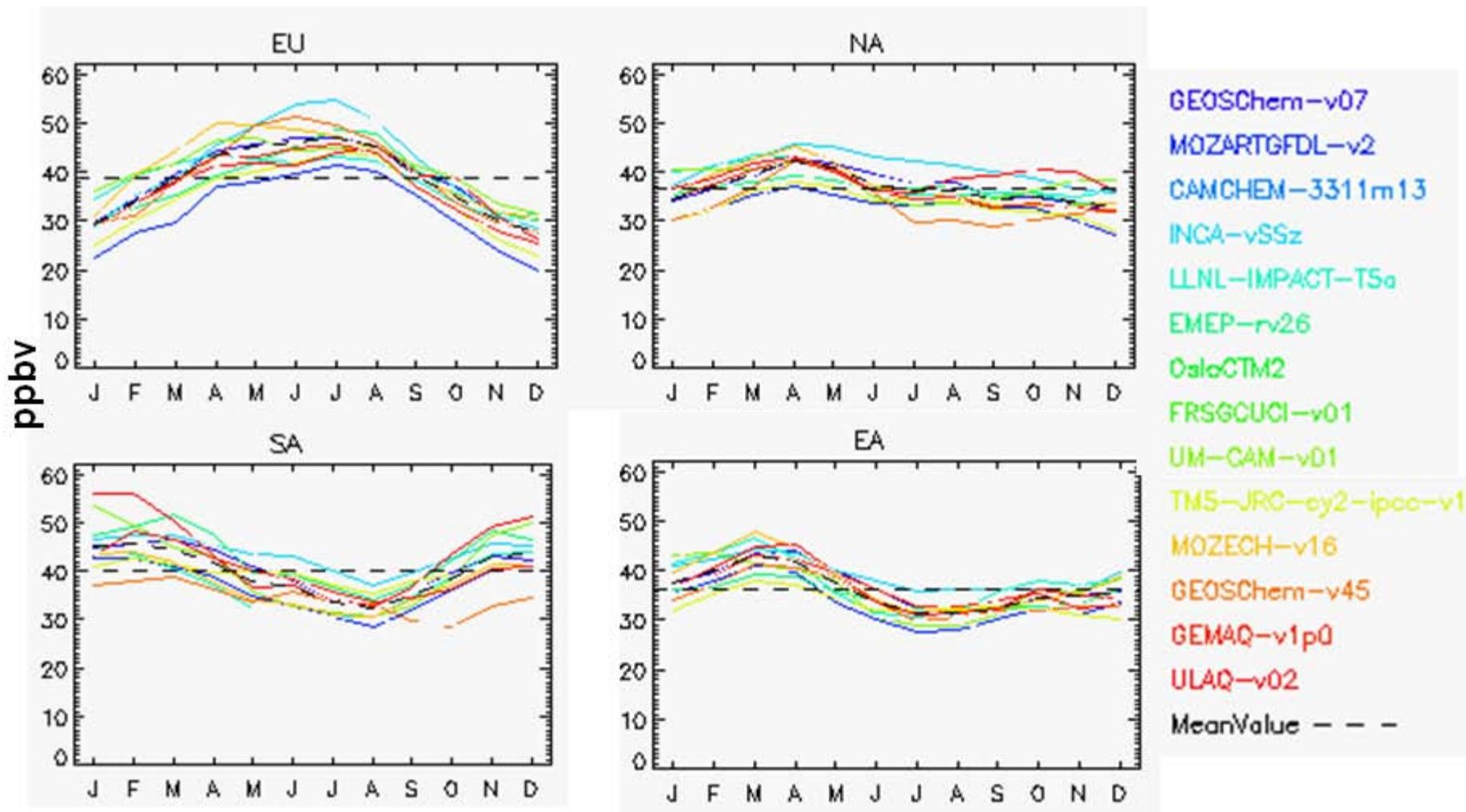
Conduct **sensitivity simulations** (17 total)

20% decreases in:

- anthrop. emis. in HTAP regions for NO_x , CO, NMVOC individually
- anthrop. emis. of all O_3 and aerosol precursors in HTAP regions
- global CH_4



Seasonal cycles in simulated surface O₃ over the HTAP regions: Results from individual models



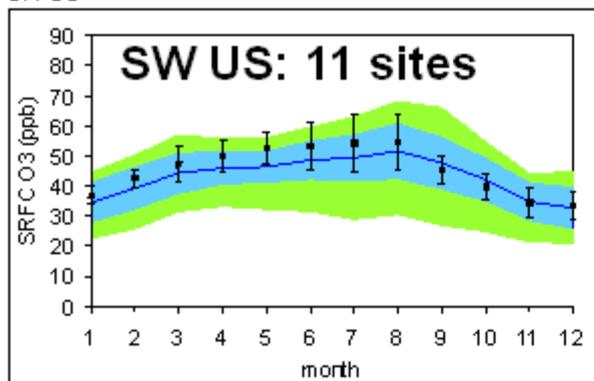
Model range often spans ~15 ppbv

Recent ACCENT multi-model surface O₃ vs. observations in HTAP regions: NA and EU

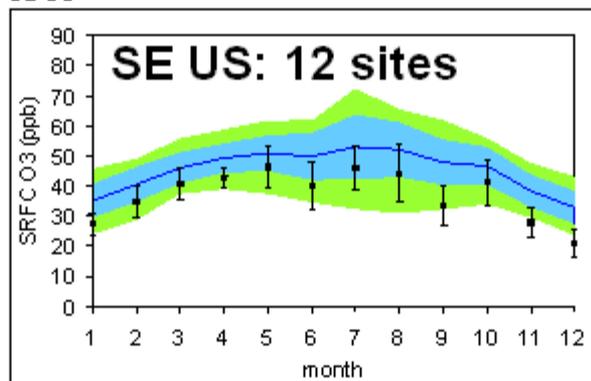
■ OBS Multi-model — mean ■ standard deviation ■ range

North America

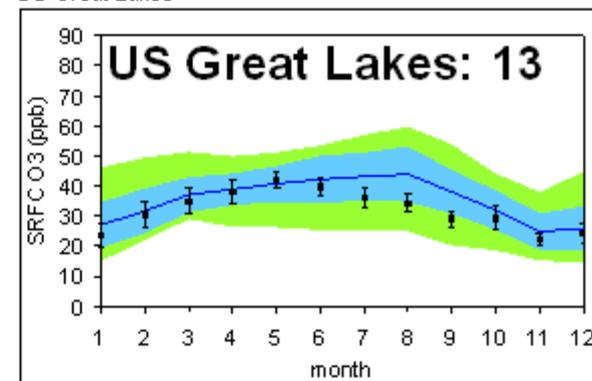
SW US



SE US

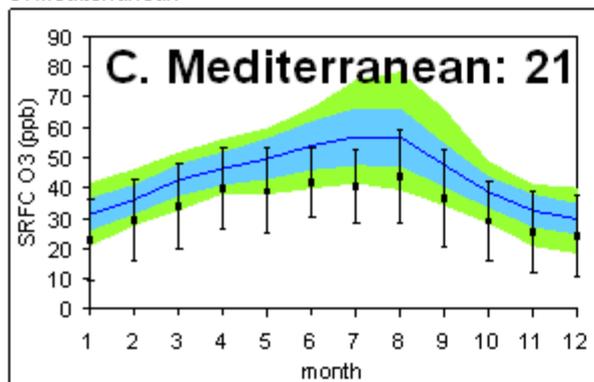


US Great Lakes

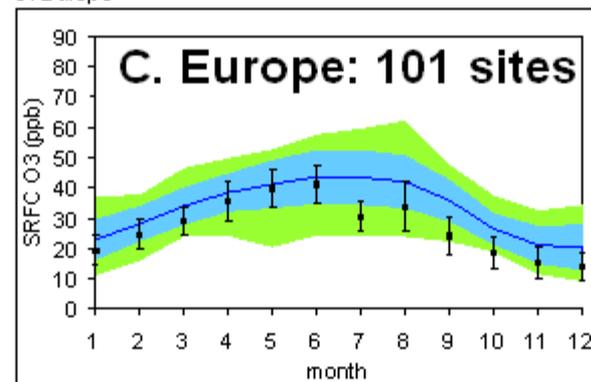


Europe and North Africa

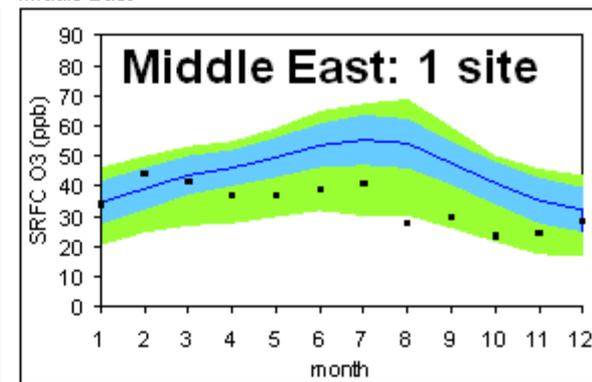
C. Mediterranean



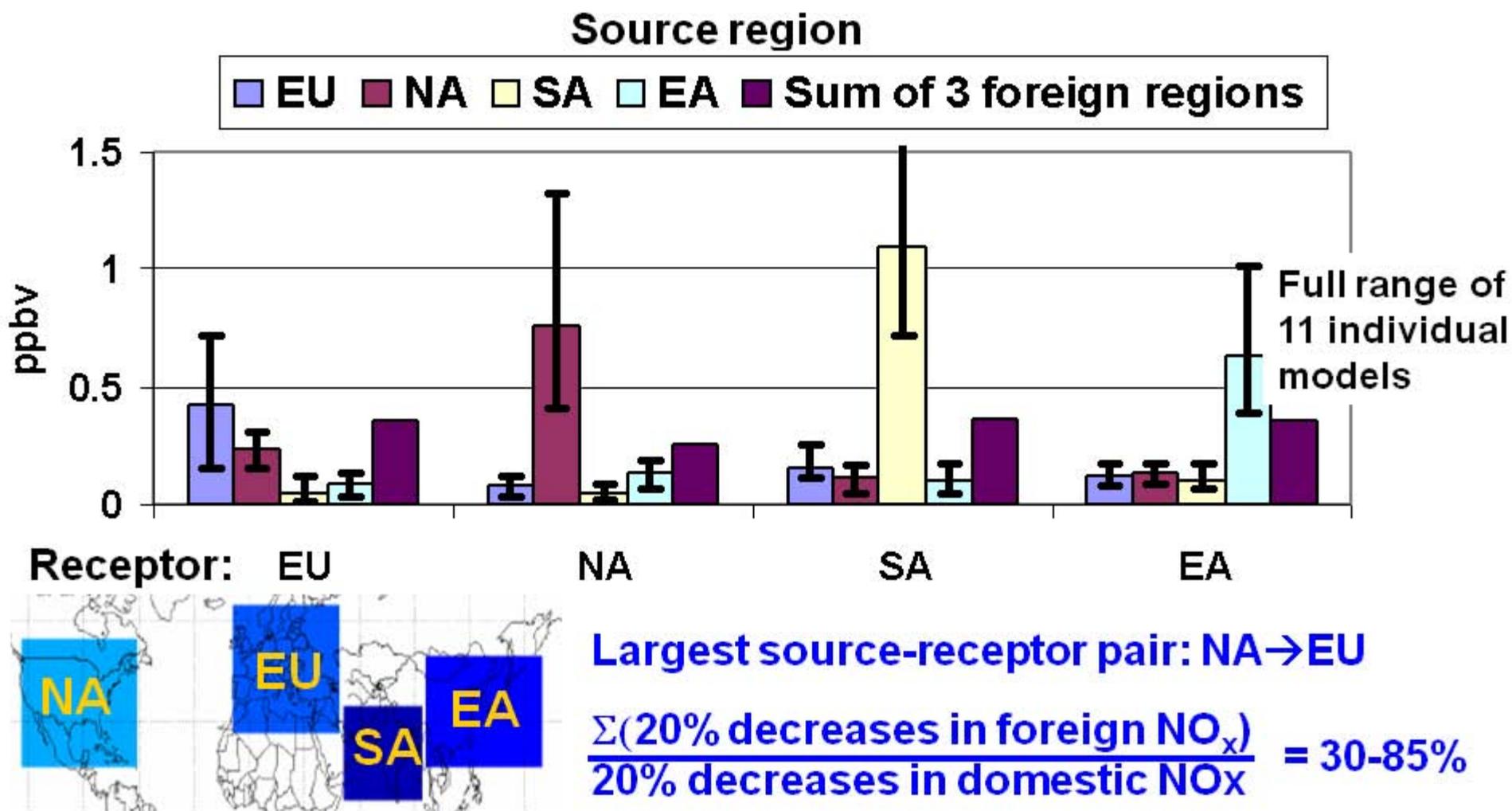
C. Europe



Middle East

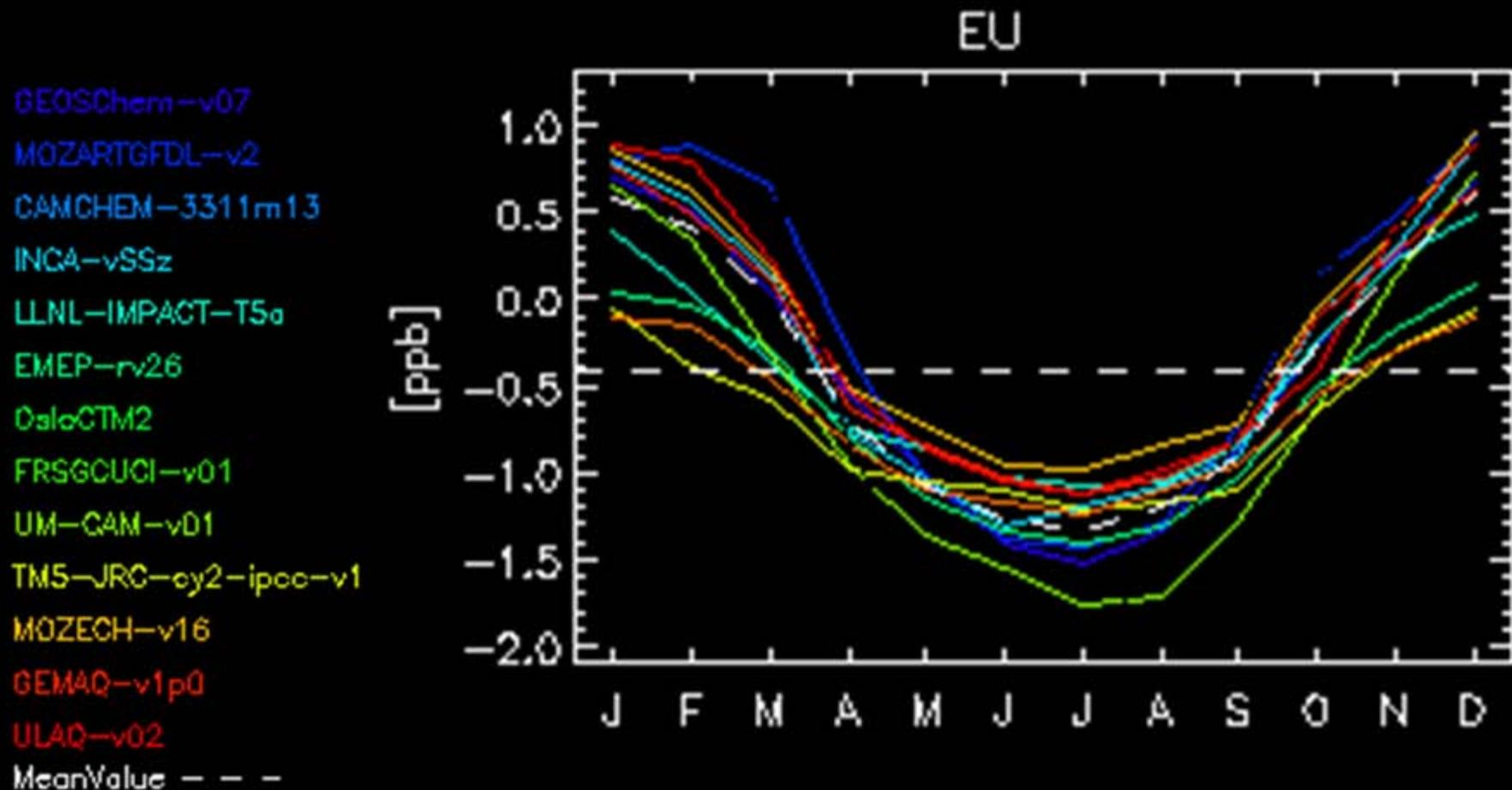


Estimates of S-R relationships: Annual mean surface O₃ decrease from 20% reductions in anthropogenic NO_x emissions



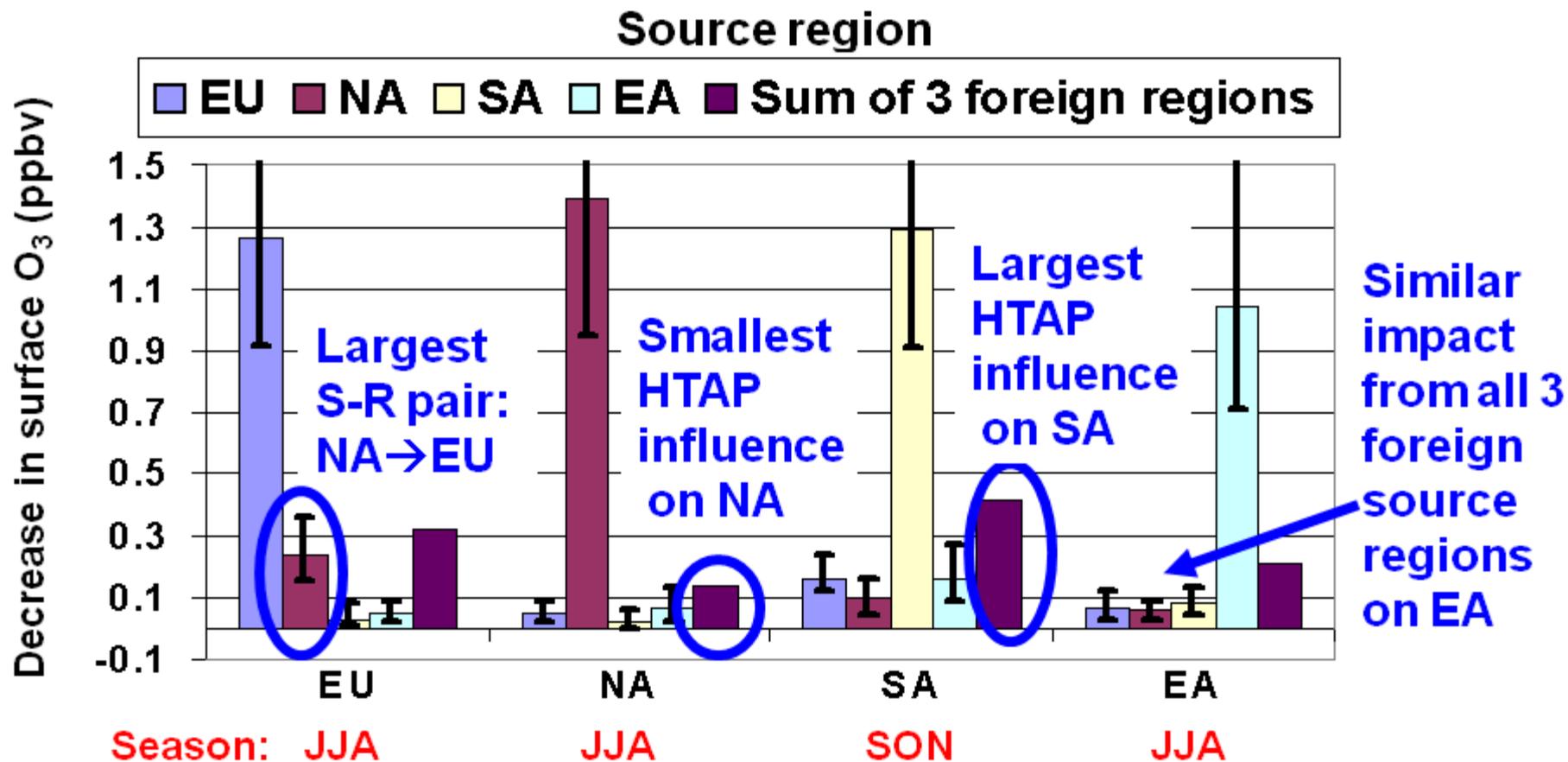
→ But... seasonality in sensitivity to domestic vs. foreign NO_x sources?

Monthly mean surface O₃ change in EU from 20% reductions of domestic NO_x emissions



→ Large seasonality masked by annual mean statistic

Estimates of S-R relationships: Smaller relative contributions from foreign NO_x sources during season of max domestic ozone production



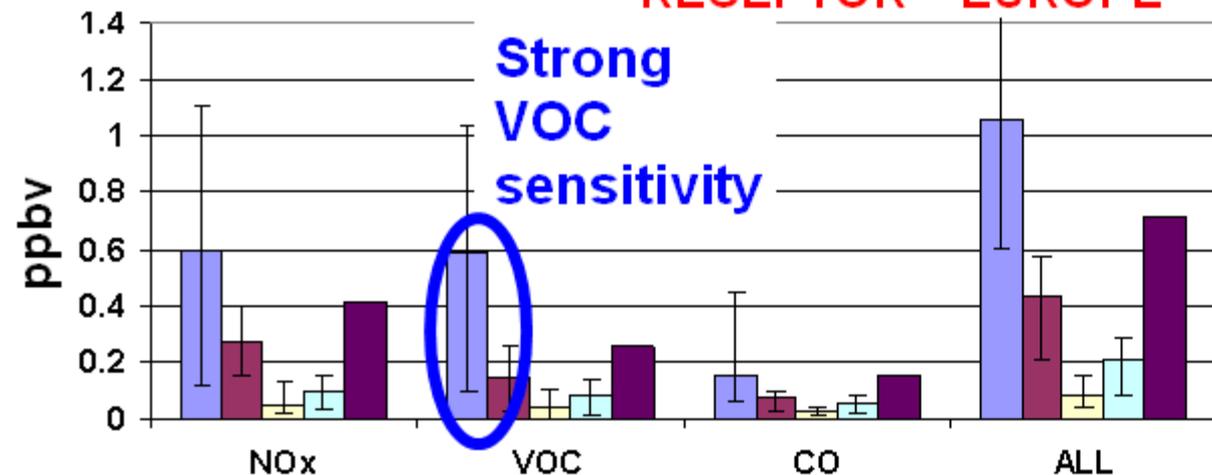
$$\frac{\Sigma(20\% \text{ decreases in foreign NO}_x)}{20\% \text{ decreases in domestic NO}_x} = 10\text{-}30\%$$

- Examine springtime when intercontinental transport typically peaks
- Compare role of NO_x emissions with other O₃ precursors

Estimates of S-R relationships: Springtime surface O₃ decrease in HTAP receptor regions from 20% reductions of O₃ precursors

■ EU
 ■ NA
 ■ SA
 ■ EA
 ■ NA+SA+EA
 SOURCE REGIONS

RECEPTOR = EUROPE

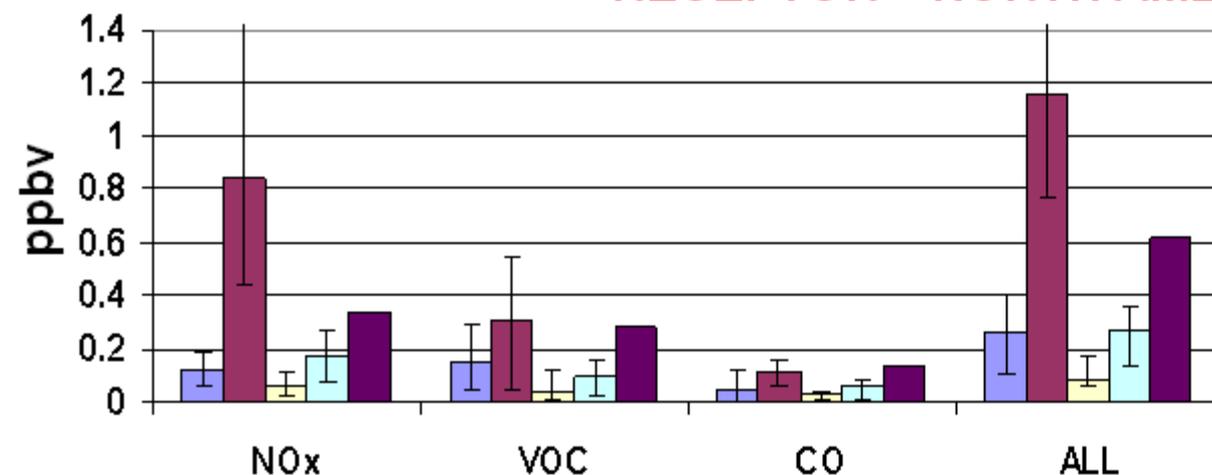


Multi-model means indicate:

NA contributes most

$$\frac{\sum(-20\% \text{ foreign emis.})}{-20\% \text{ domestic emis.}} \approx 70\%$$

RECEPTOR = NORTH AMERICA



EA/EU contribute similarly

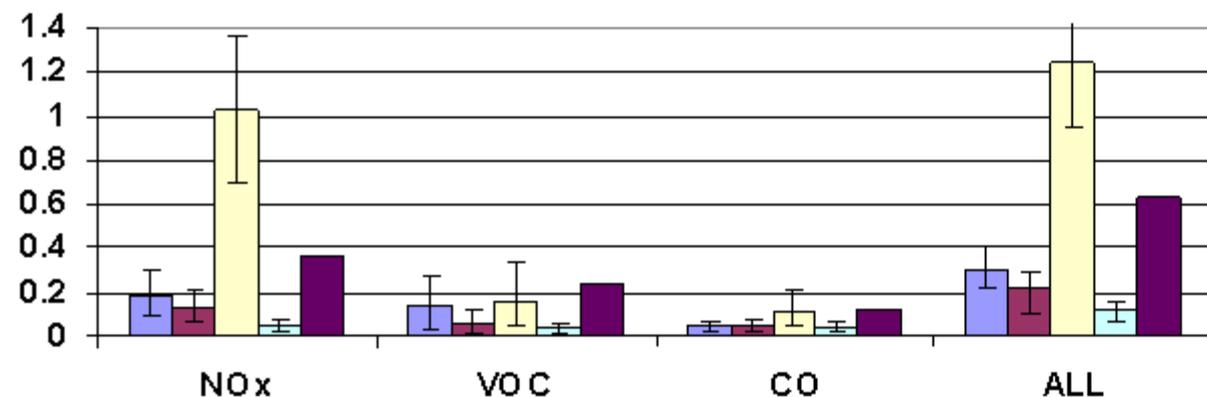
EU VOC at least as important as NO_x

$$\frac{\sum(-20\% \text{ foreign emis.})}{-20\% \text{ domestic emis.}} \approx 50\%$$

Estimates of S-R relationships: Springtime mean surface O₃ decrease in HTAP receptor regions from 20% reductions of O₃ precursors

■ EU
 ■ NA
 ■ SA
 ■ EA
 ■ NA+SA+EA
 SOURCE REGIONS

RECEPTOR = SOUTH ASIA



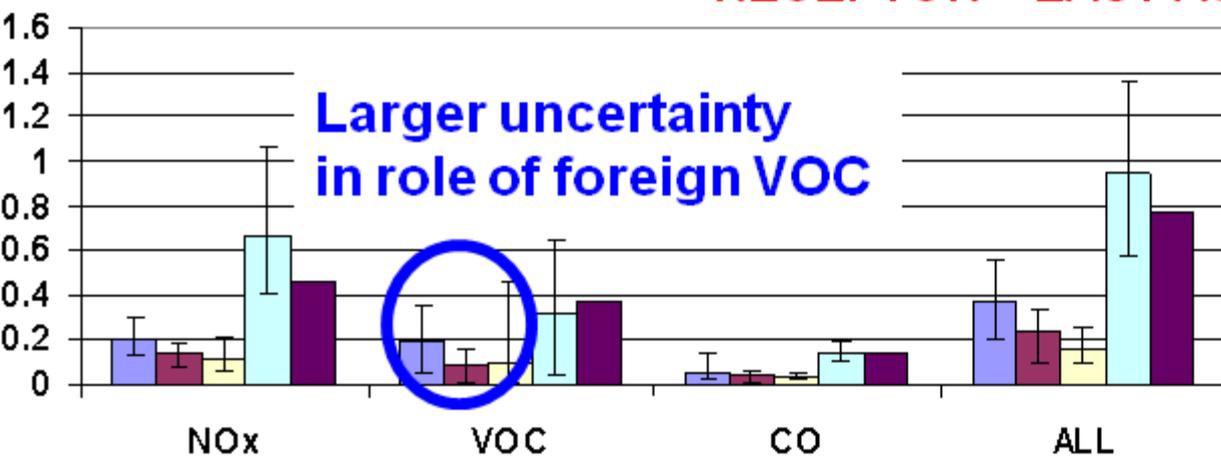
Multi-model means indicate:

EU contributes most

NA > EA

$$\frac{\sum(-20\% \text{ foreign emis.})}{-20\% \text{ domestic emis.}} \approx 50\%$$

RECEPTOR = EAST ASIA



Larger uncertainty
in role of foreign VOC

EU and NA NO_x contribute similarly but stronger EU VOC influence

EU and NA > SA

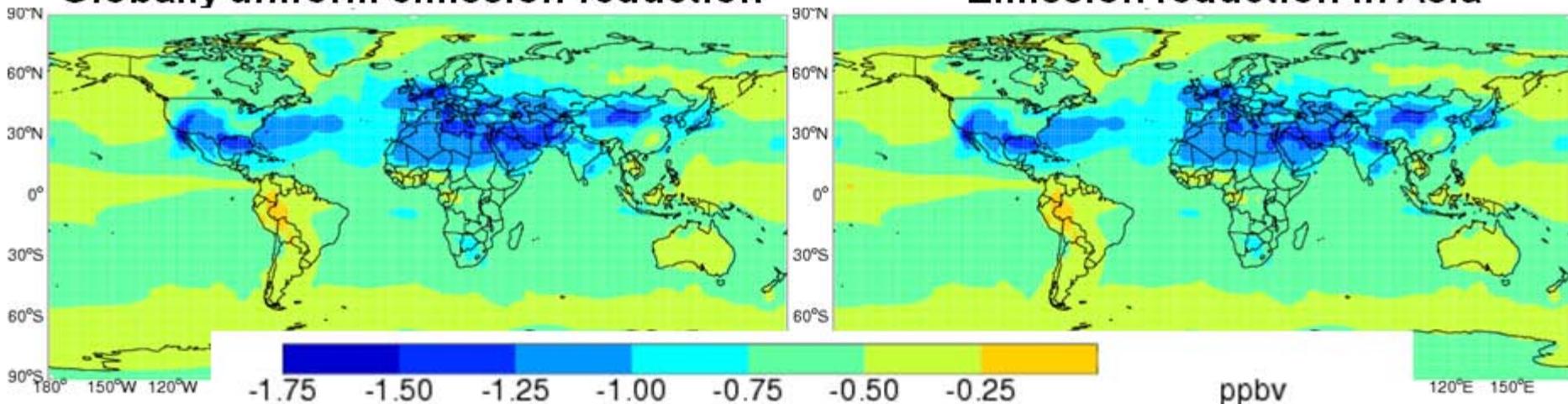
$$\frac{\sum(-20\% \text{ foreign emis.})}{-20\% \text{ domestic emis.}} \approx 80\%$$

Global surface ozone response to CH₄ emission controls is independent of source location

Change in July surface O₃ from 30% decrease in anthropogenic CH₄ emissions

Globally uniform emission reduction

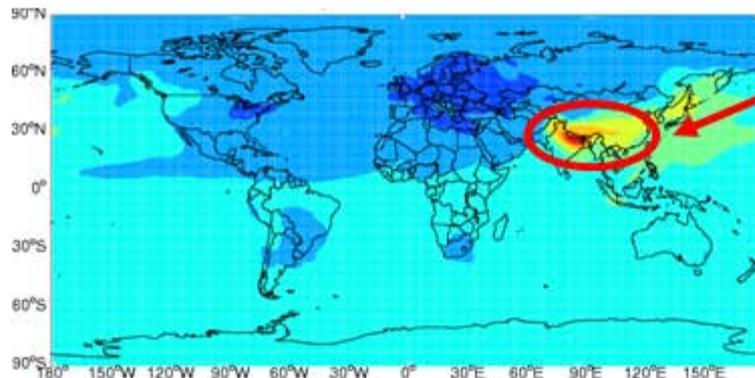
Emission reduction in Asia



Percentage

Difference:

$$\frac{\Delta\text{Asia} - \Delta\text{uniform}}{\Delta\text{Asia}}$$



Enhanced effect in source region

<10% other NH source regions

<5% rest of NH

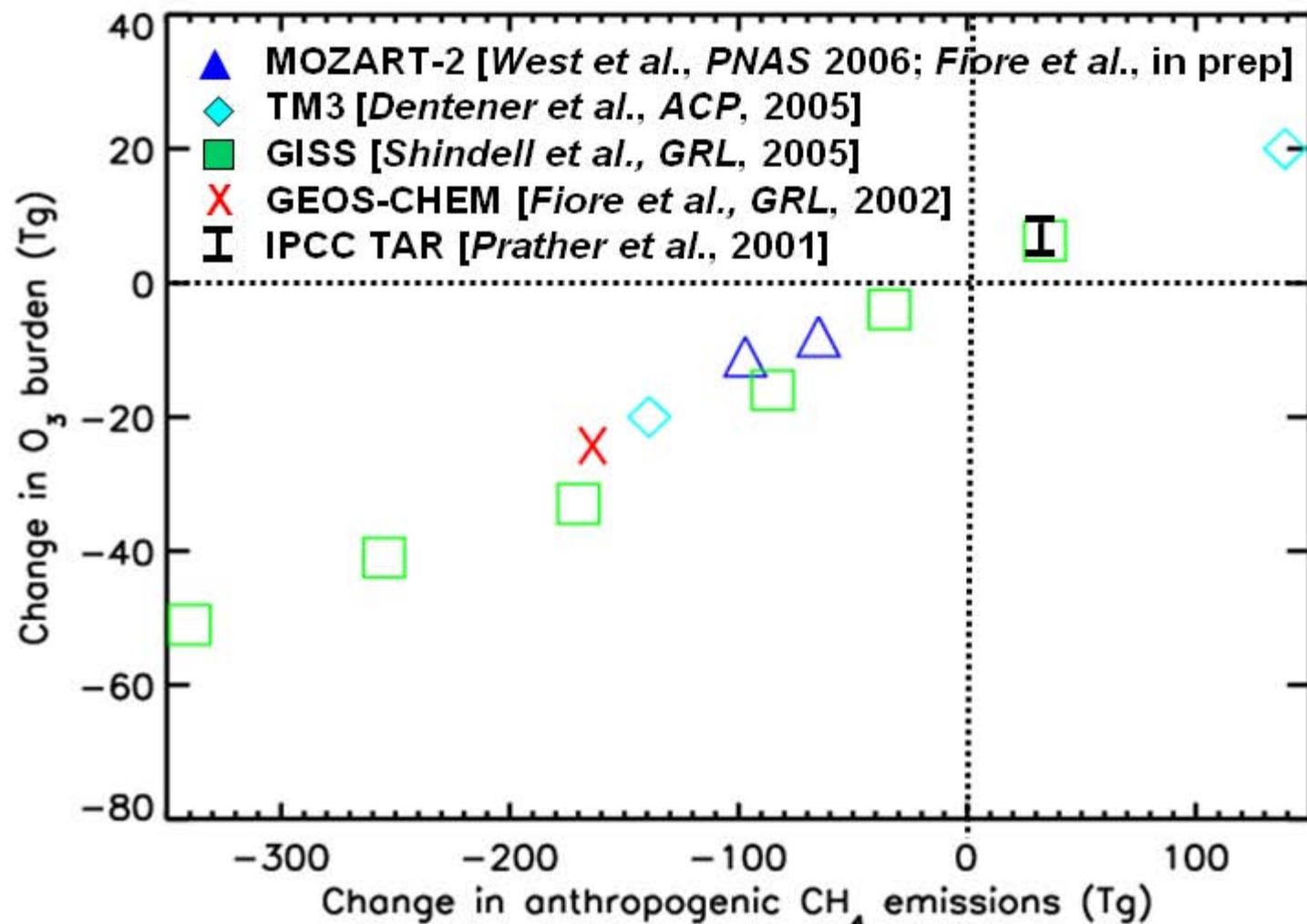
<1% most of SH



1.9°x1.9°

→ Target cheapest controls worldwide

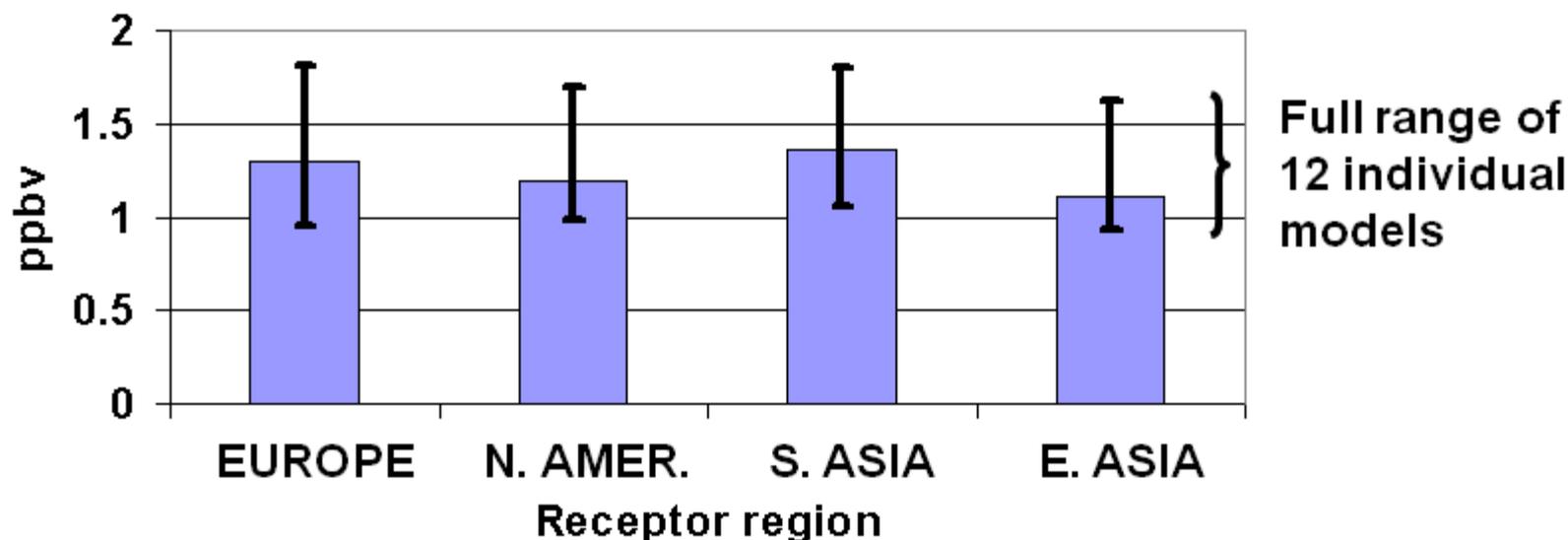
Tropospheric O₃ responds approximately linearly to anthropogenic CH₄ emission changes across models



Anthropogenic CH₄ contributes ~50 Tg (~15%) to tropospheric O₃ burden
~5 ppbv to surface O₃

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 O₃ decrease over all NH receptor regions

→ Consistent with prior studies

What causes the range across individual models?

-- examine ozone production efficiency; NO_x:VOC ratios

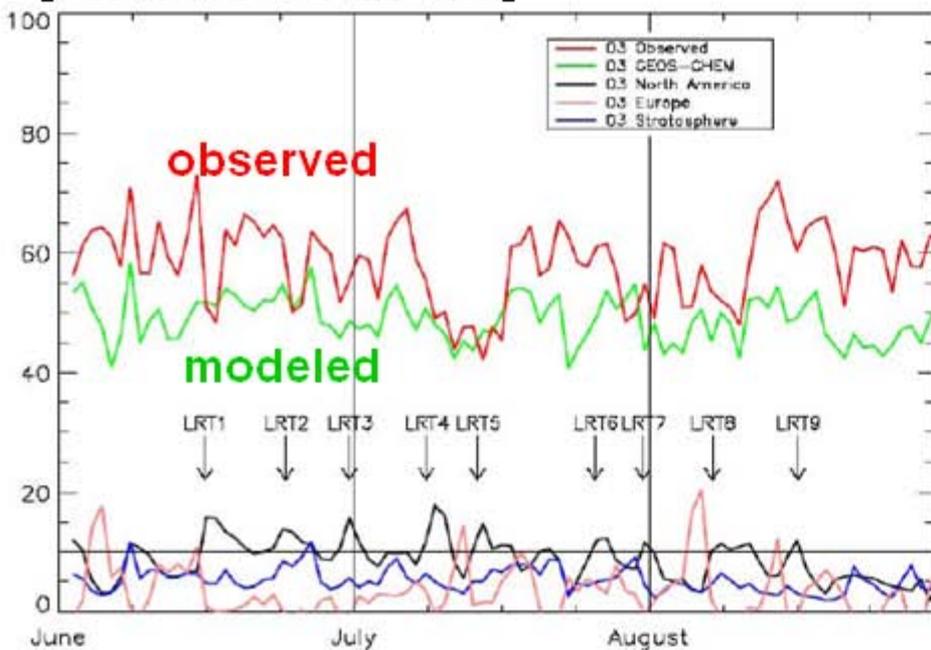
Some remaining questions...

(to be examined as part of the TF HTAP project)

- Which processes contribute most to the inter-model range in S-R relationships?
 - Insights from idealized tracer experiments
 - Normalize response to emission perturbations to “remove” uncertainty due to differences in emission inventories
- How can we best test models of hemispheric transport of air pollution?
 - Need to test **processes** (export, chemical evolution, mixing into surface air)
- Does the response to 20% decreases in emissions scale to reductions of different magnitude?
 - Yes for CH₄; simulations underway for NO_x (expected to be more non-linear) and combined O₃ precursor reductions
- What is the contribution of hemispheric transport to polluted vs. “average” vs. clean conditions)?
 - Analysis underway to compare range of model responses at different places in the overall O₃ distribution
- How will climate change affect hemispheric transport of air pollution?
 - Expected to influence production, loss, transport

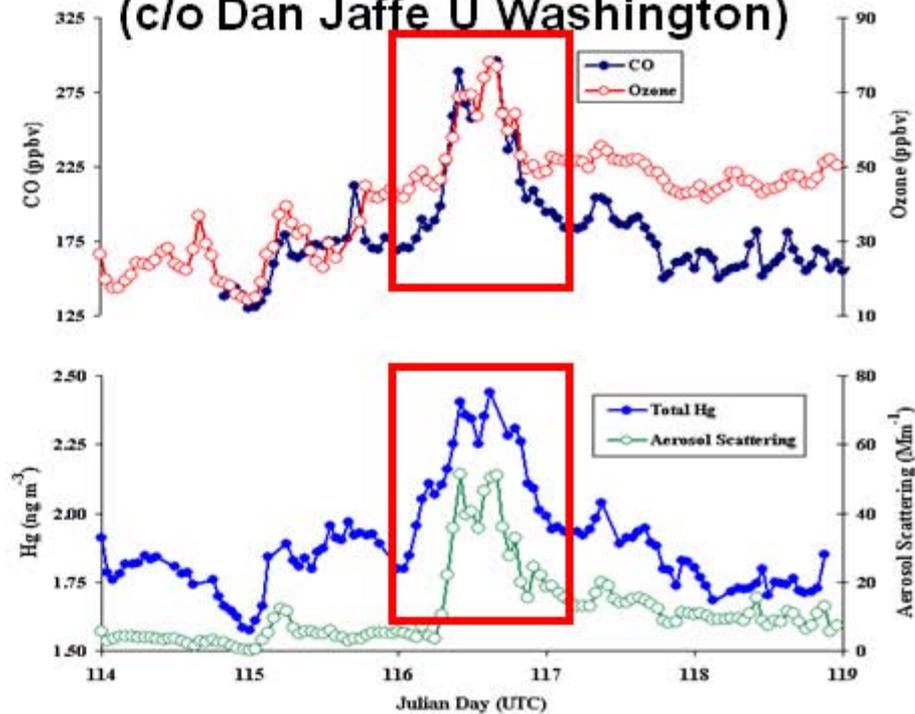
Example observations at high-altitude sites for testing long-range transport in models

Ozone (ppb) at Jungfrauoch, Switzerland
June-August 2000 (3.5 km)
[Guerova et al., 2006]



O₃ from **North America**
Europe
Stratosphere

Mt. Bachelor Observatory in
central Oregon – April 25, 2004
(c/o Dan Jaffe U Washington)

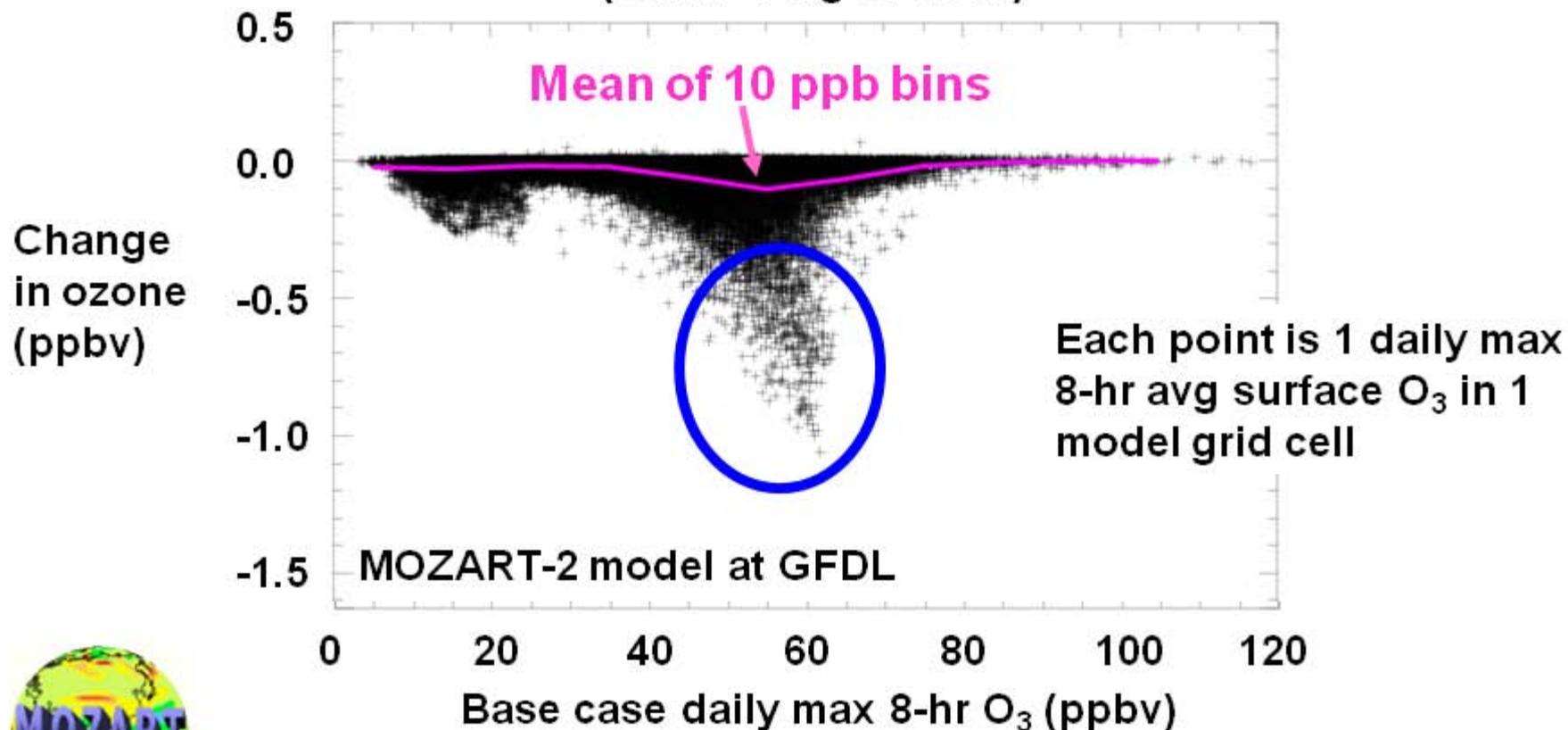


Asian industrial plume diagnosed via back-trajectories, a 3-D model, Hg/CO signature [Jaffe et al., 2005; Weiss-Penzias et al., 2006]

→ Insights into model capability at representing specific processes by integrating analysis of data from model, aircraft, satellite, sondes, surface sites?

Maximum intercontinental influence occurs near the center of the overall O₃ distribution

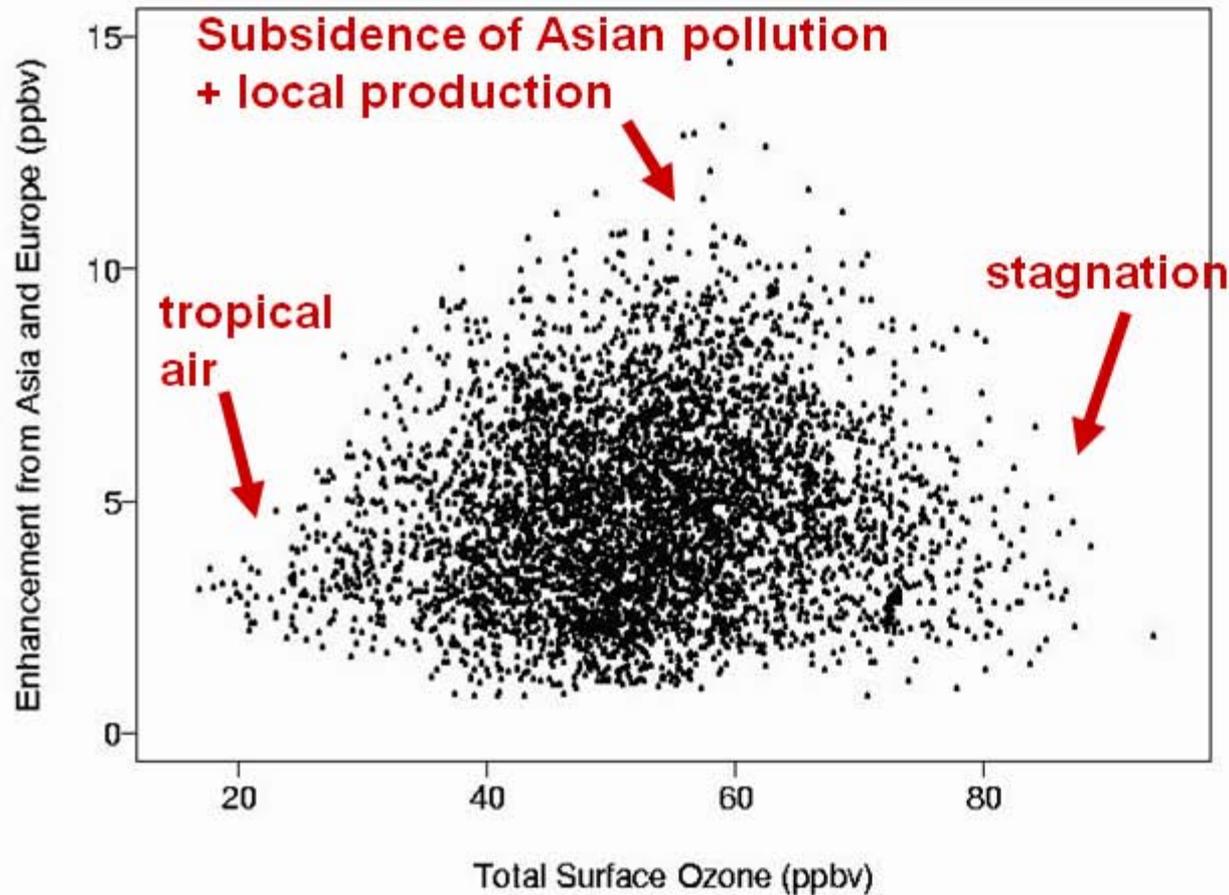
Impact of a 20% decrease in NA anthrop. NO_x to EA surface O₃
(Jun 1 - Aug 31 2001)



→ Is this result robust across the HTAP models?

Consistent results from earlier study: Enhancements from Asian/European pollution over the U.S. in summer

as determined from a simulation without these emissions (GEOS-Chem 4°x5°)

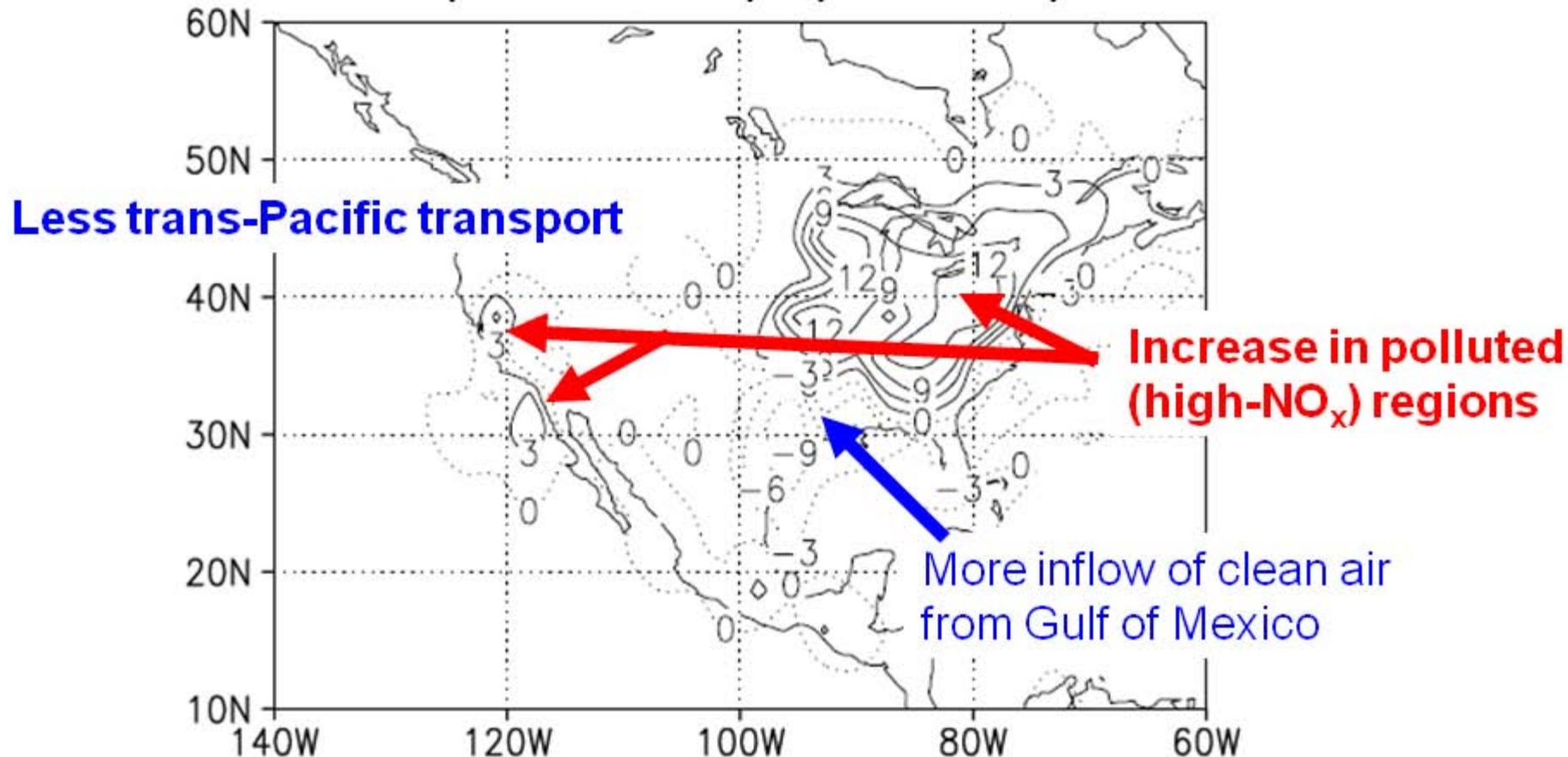


Max Asian/European pollution enhancements (up to 14 ppbv) occur at intermediate ozone levels (50-70 ppbv)

→ Major concern if national ozone standard were to decrease

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]

Conclusions: Intercontinental Source-Receptor Relationships for Ozone

- Range of estimates narrows from that in the literature under HTAP setup
- Multi-model mean results indicate that $\Sigma(-20\% \text{ foreign NO}_x)/(-20\% \text{ domestic NO}_x)$ ranges from
 - 10-30% during the season of maximum local O₃ production
 - 35-70% in spring
- Larger multi-model mean sensitivity to 20% decreases in NO_x+NMVOC+CO (in spring, peak HTAP season):
 $\Sigma(-20\% \text{ foreign O}_3 \text{ precursors})/(-20\% \text{ domestic O}_3 \text{ precursors}) = 50-80\%$
 - 0.6-0.8 ppbv surface O₃ response to the sum of the 3 foreign source regions
 - 0.9-1.2 ppbv response to domestic emissions
 - EU more sensitive to domestic NMVOC emissions than other HTAP regions
- Smaller contribution from HTAP to high-O₃ events than to mean conditions
- Robust model response of a ~1 ppbv (1.0-1.8) decrease in annual mean surface O₃ over HTAP regions when global CH₄ is reduced by 20%
 - Response scales linearly to perturbations of different magnitudes
 - Response independent of location → target cheapest controls worldwide