External influences on ozone air quality: Intercontinental transport, changing climate, and stratospheric exchange

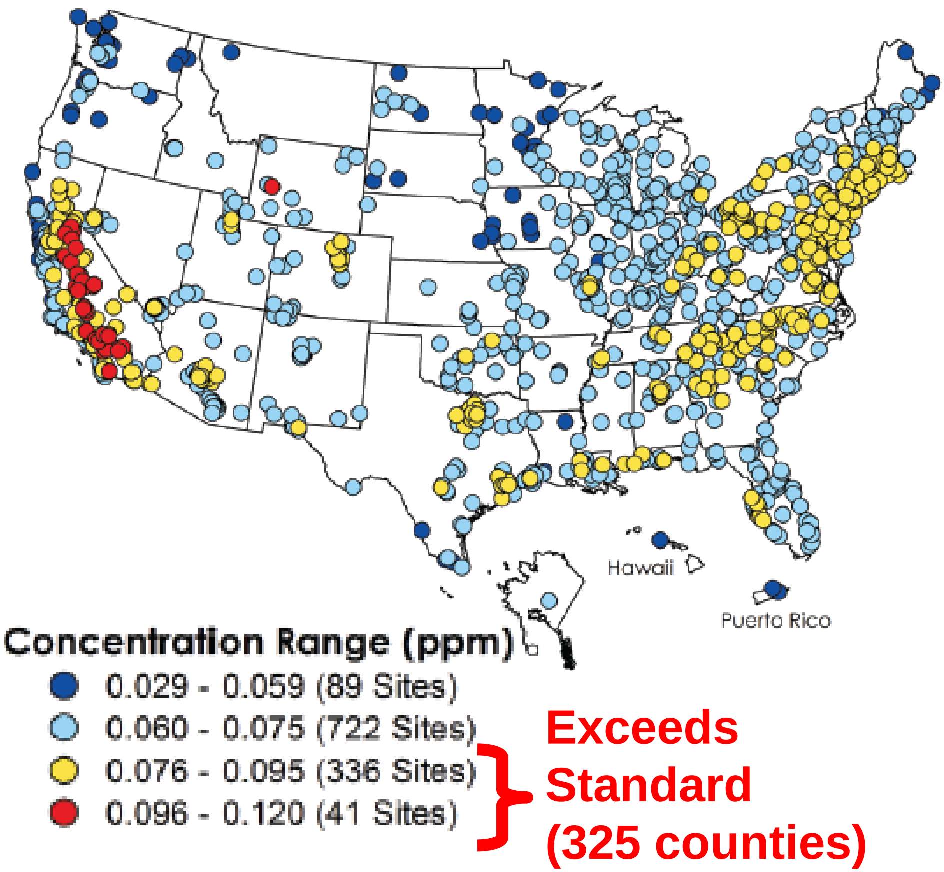
Arlene M. Fiore

Acknowledgments:
Yuanyuan Fang, Larry Horowitz, Hiram Levy II, Vaishali Naik (GFDL)
Emily Fischer, Dan Jaffe, David Reidmiller (U WA)
Meiyun Lin (U WI-Madison), Martin Schultz (ICG, Julich)
Ron Cohen (UC Berkeley), Johannes Stähelin, Shubha Pandey (ETH Zürich)
Numerous collectors of PAN, NO_y and O_3 measurements
GFDL Atmospheric Model Development Team, TF HTAP Modeling Team

Princeton EGGS seminar, May 6, 2010

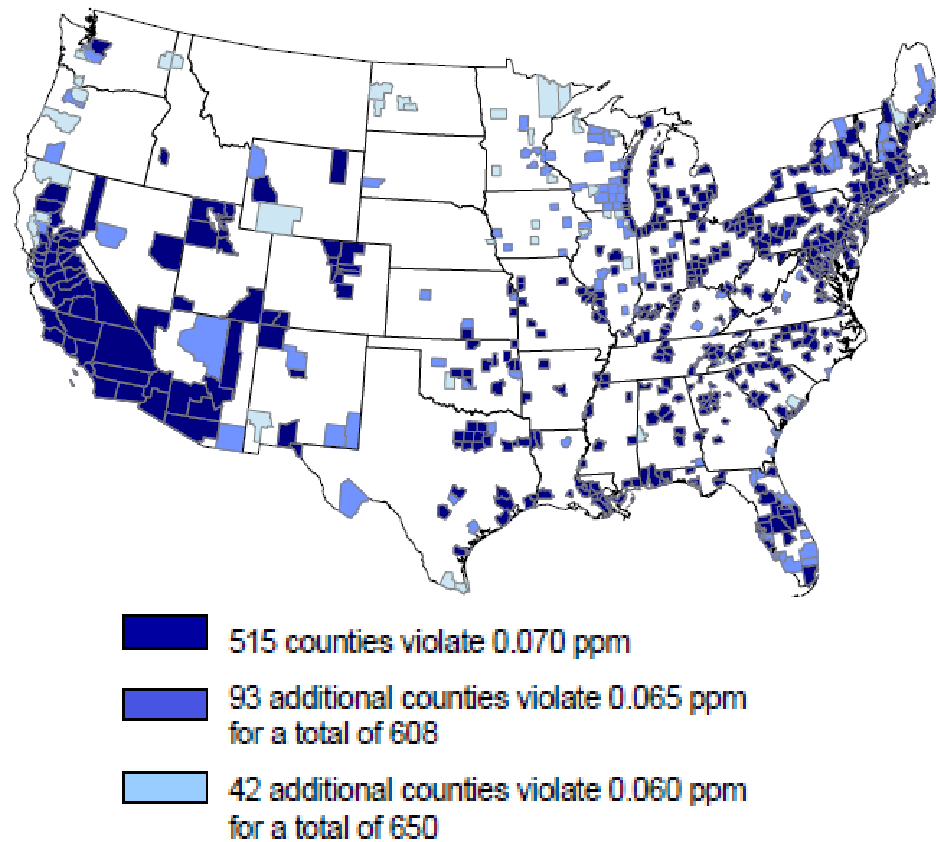
The U.S. ozone smog problem is spatially widespread, affecting ~120 million people [U.S. EPA, 2010]

4th highest daily max 8-hr O_3 in 2008



http://www.epa.gov/air/airtrends/2010/

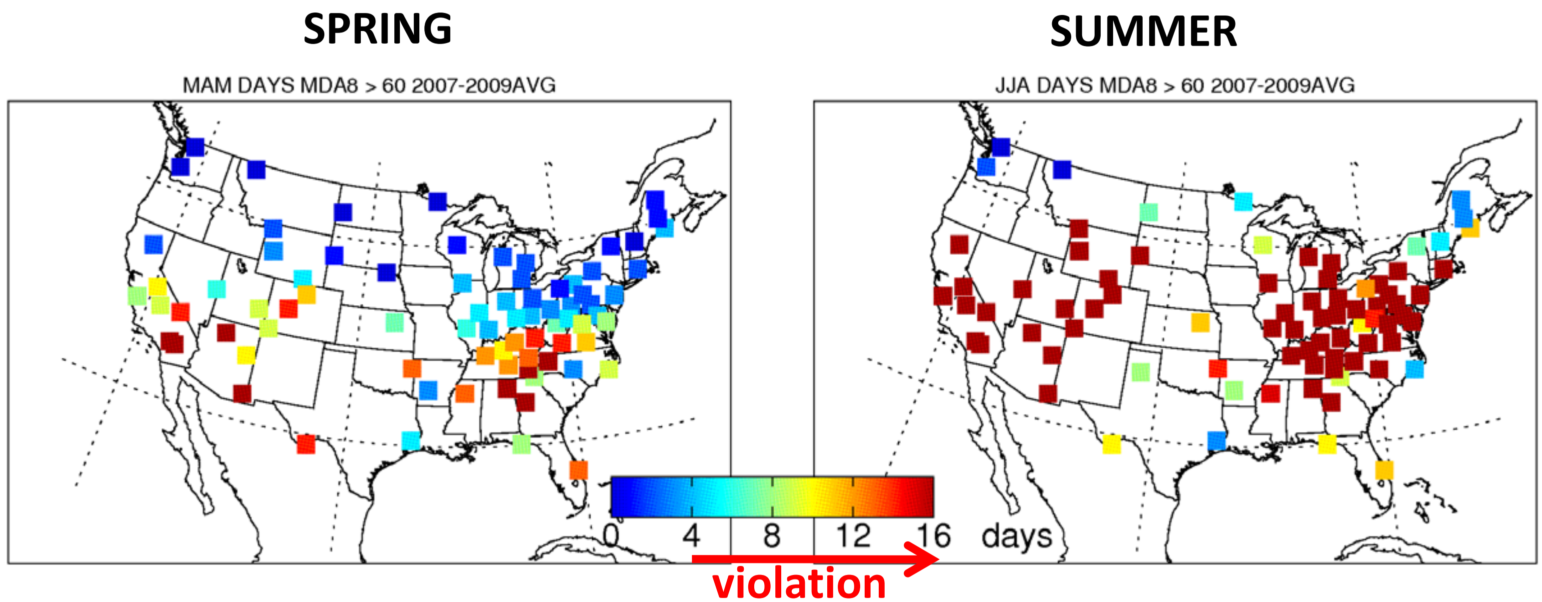
Counties with monitors violating newly proposed primary O_3 standard in range of 0.060-.070 ppm (2006-2008 data)



http://www.epa.gov/air/ozonepollution/pdfs/20100104maps.pdf

Newly proposed standards exceeded in spring in west (and SE) U.S. at "background" sites

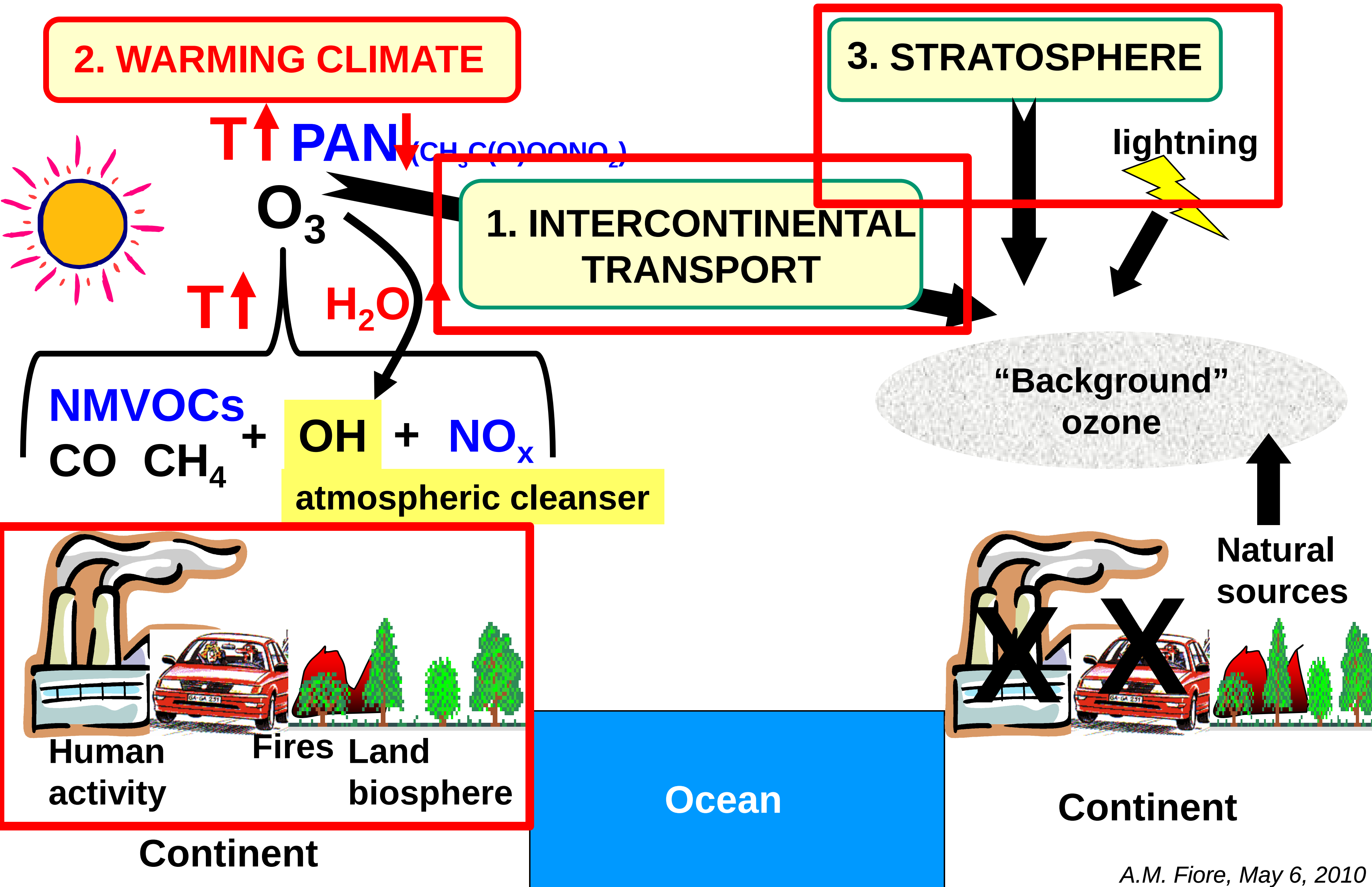
Number of days with daily max 8-hr O_3 > 60 ppb



2007-2009 averages using hourly O_3 from CASTNet database (www.epa.gov/castnet/data.html)

A.M. Fiore, May 6, 2010

Brief overview of tropospheric ozone and external influences on air quality



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

Dust leaving Asian coast, April 2001

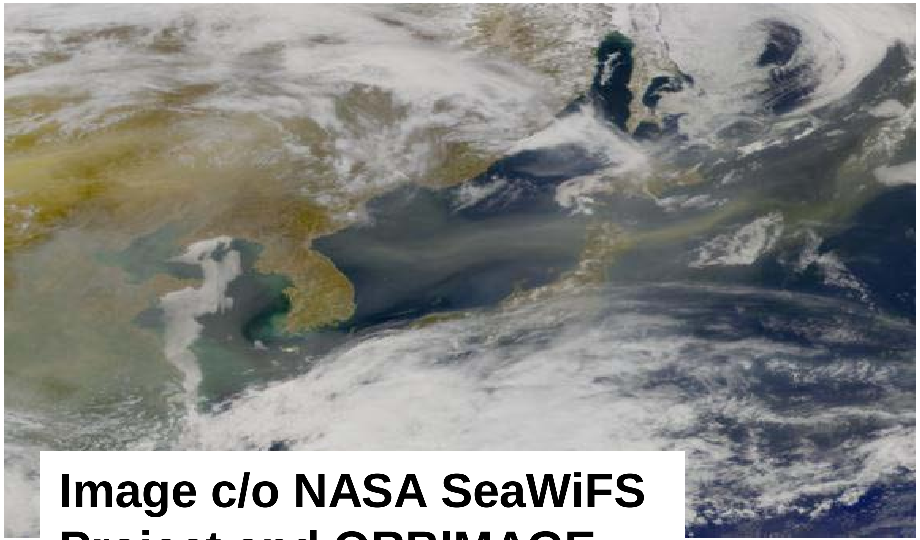
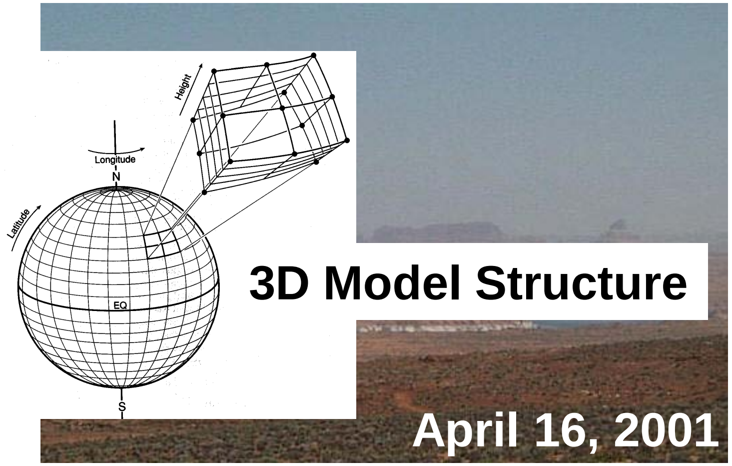


Image c/o NASA SeaWiFS Project and ORBIMAGE

Reduced Visibility from Asian Dust over Glen Canyon, Arizona, USA

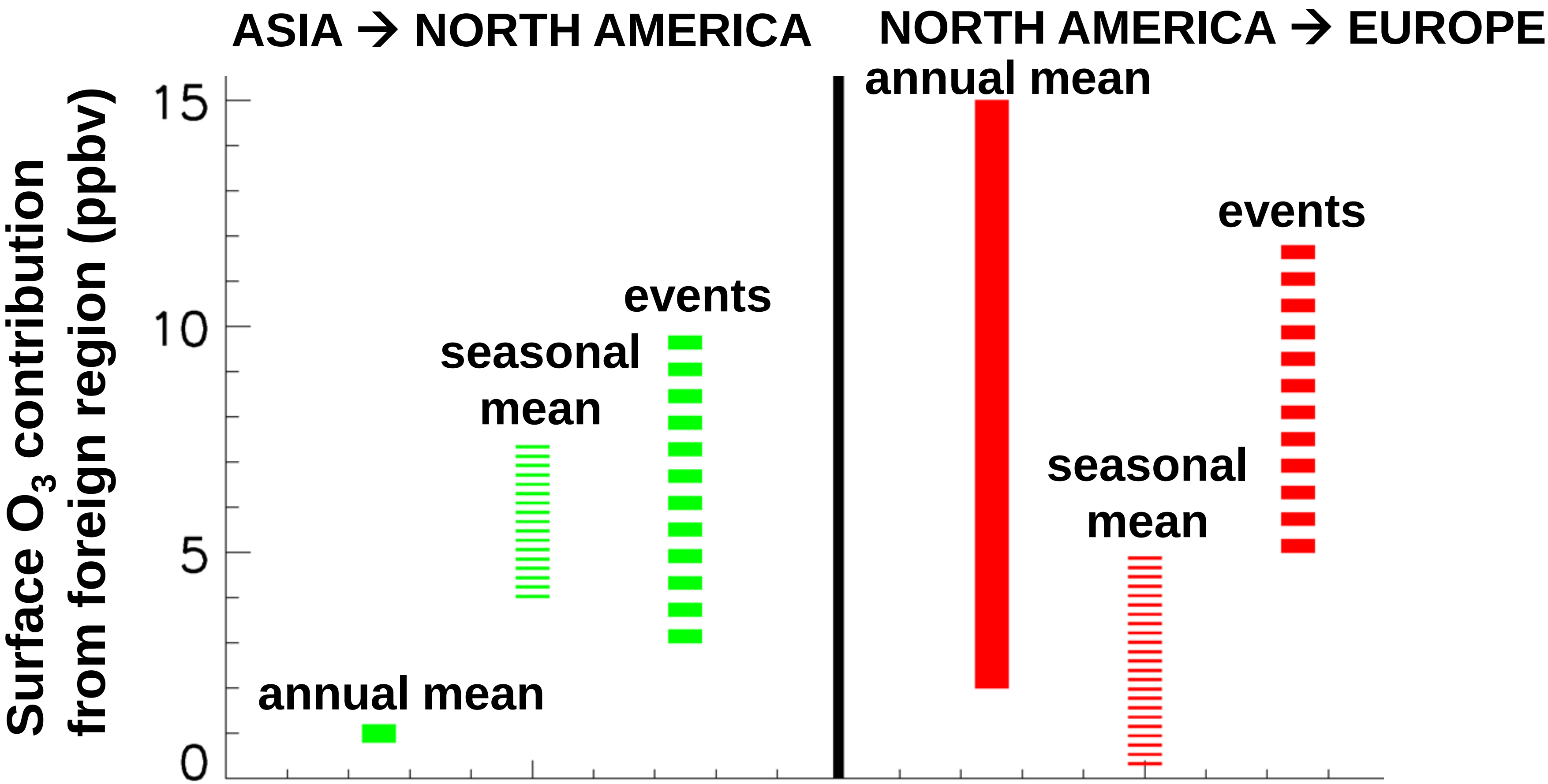


3D Model Structure

Intercontinental ozone transport is difficult (impossible?) to observe directly at surface *[e.g., Derwent et al., 2003; Fiore et al., 2003; Goldstein et al., 2004; Jonson et al., 2005]*

→ Estimates rely on models

Wide range in prior estimates of intercontinental surface ozone source-receptor (S-R) relationships



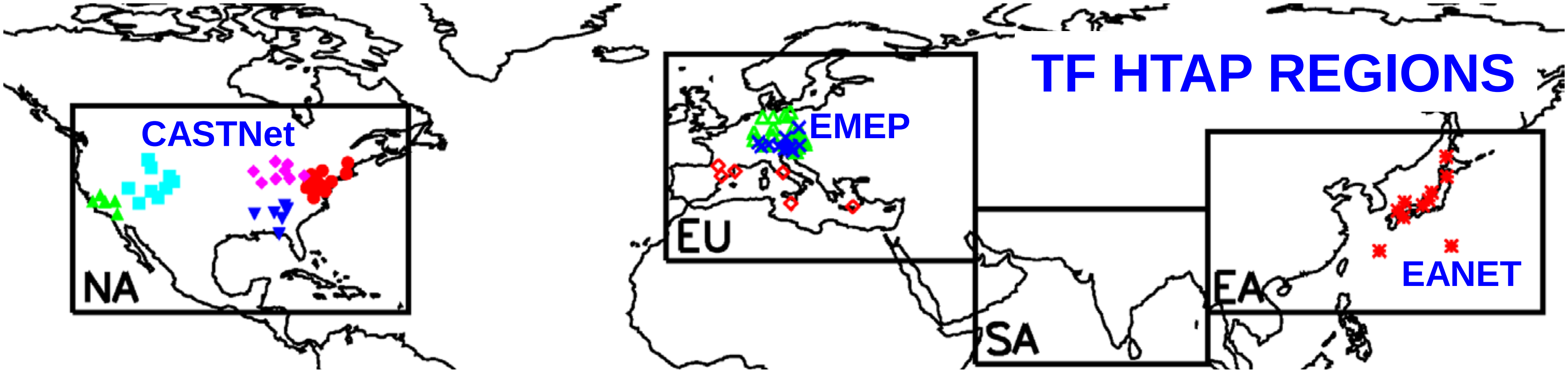
Studies in TF HTAP [2007] + *Holloway et al*., 2008; *Duncan et al*., 2008; *Lin et al*., 2008

Assessment hindered by different (1) methods, (2) reported metrics, (3) meteorological years, (4) regional definitions

Few studies examined all seasons

A.M. Fiore, May 6, 2010

Task Force on Hemispheric Transport of Air Pollution (TF HTAP): Multi-model studies to quantify & assess uncertainties in N. mid-latitude source-receptor relationships to inform CLRTAP



BASE SIMULATION (21 models):

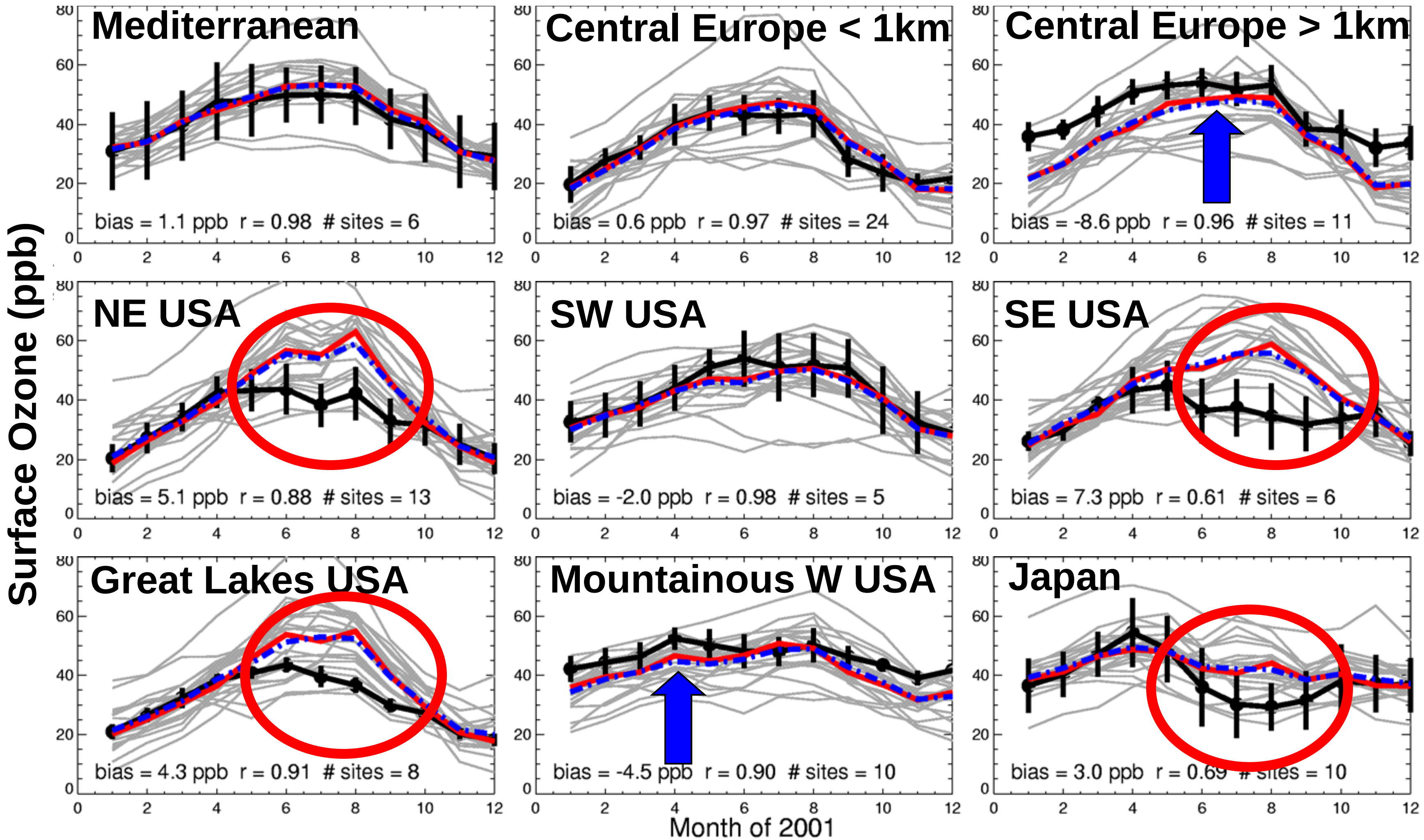
→ horizontal resolution of 5 x5° or finer
→ 2001 meteorology
→ each group's best estimate for 2001 emissions
→ methane set to 1760 ppb

SENSITIVITY SIMULATIONS (13-18 models):

→ -20% regional anthrop. NO_x, CO, NMVOC emissions, individually + all together (=16 simulations)
→ -20% global methane (to 1408 ppb)

TF HTAP, 2007, 2010; Sanderson et al., GRL, 2008; Shindell et al., ACP, 2008; Fiore et al., JGR, 2009, Reidmiller et al. ACP, 2009; Casper Anenberg et al., ES&T, 2009; Jonson et al., ACPD,2010

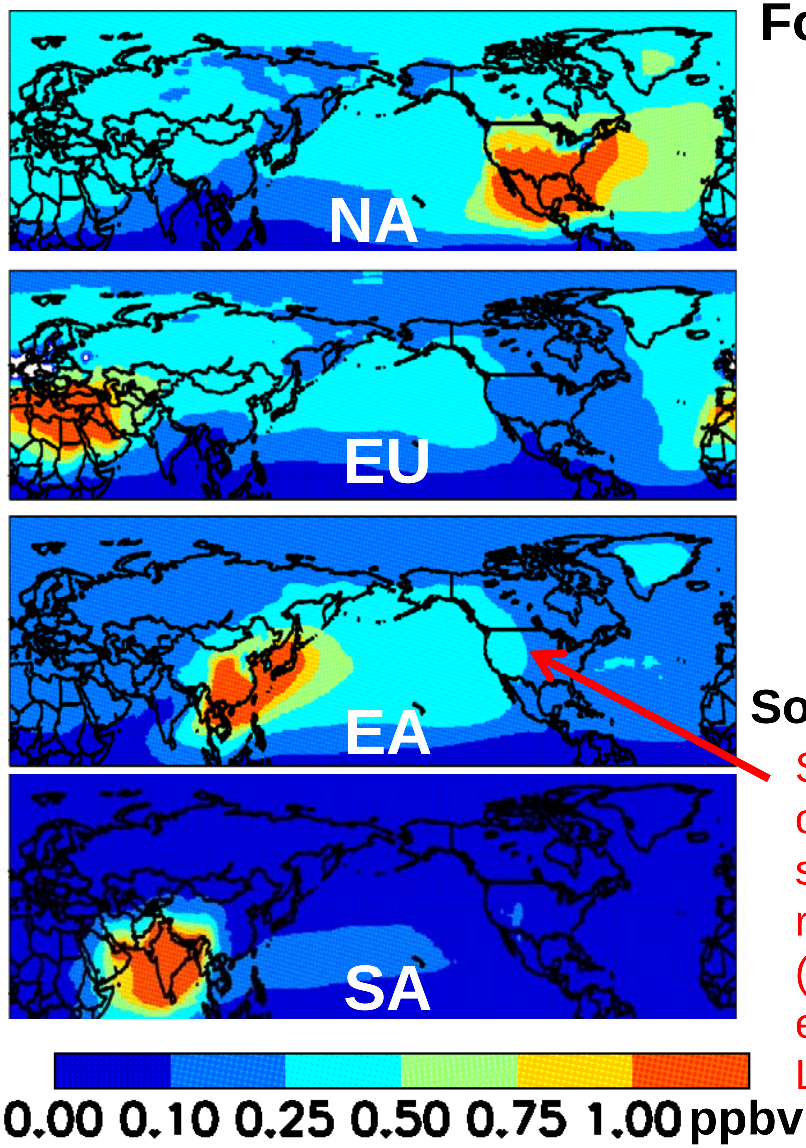
Large inter-model range; multi-model mean generally captures observed monthly mean surface O_3



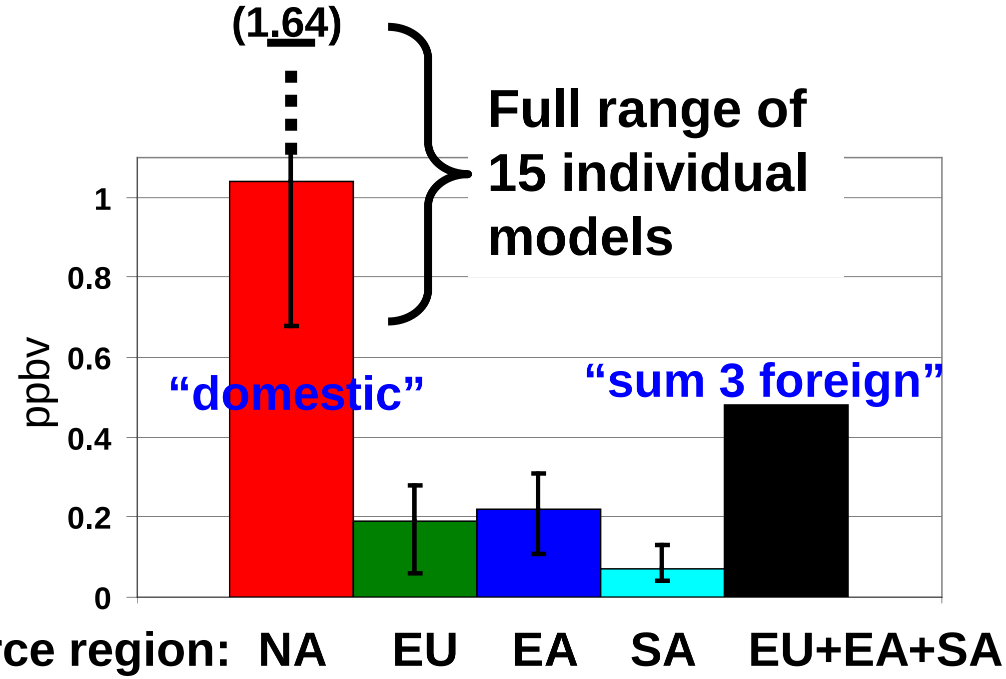
→ **Many models biased low at altitude, high over EUS+Japan in summer**

→ **Good springtime/late fall simulation**

Model ensemble annual mean decrease in surface O_3 from 20% reductions of regional anthrop. O_3 precursors



Foreign vs. "domestic" influence over NA:



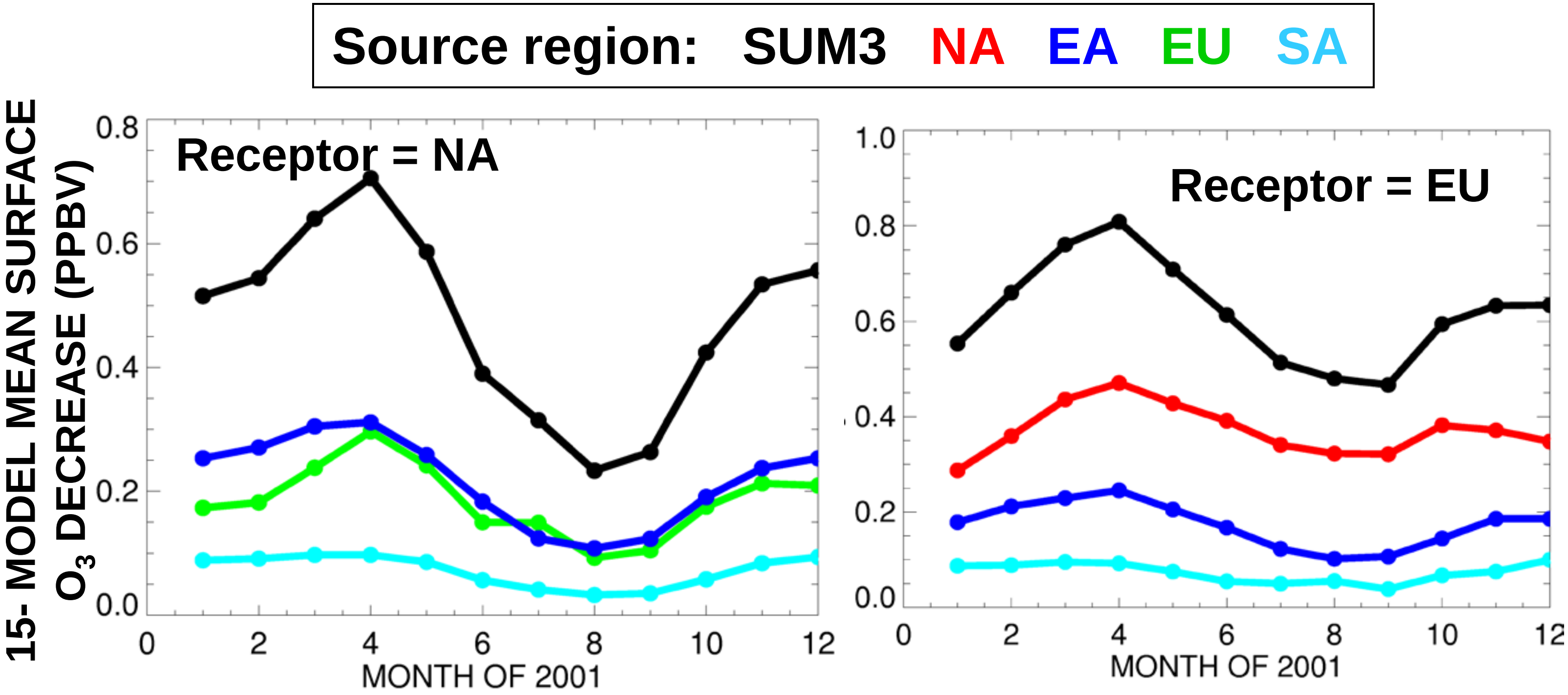
Spatial variability over continental-scale receptor region (NA) (see also Reidmiller et al, 2009; Lin et al., 2010)

$$\frac{\sum \text{3 foreign}}{\text{domestic}} = 0.45$$

"import sensitivity"

Fiore et al., JGR, 2009

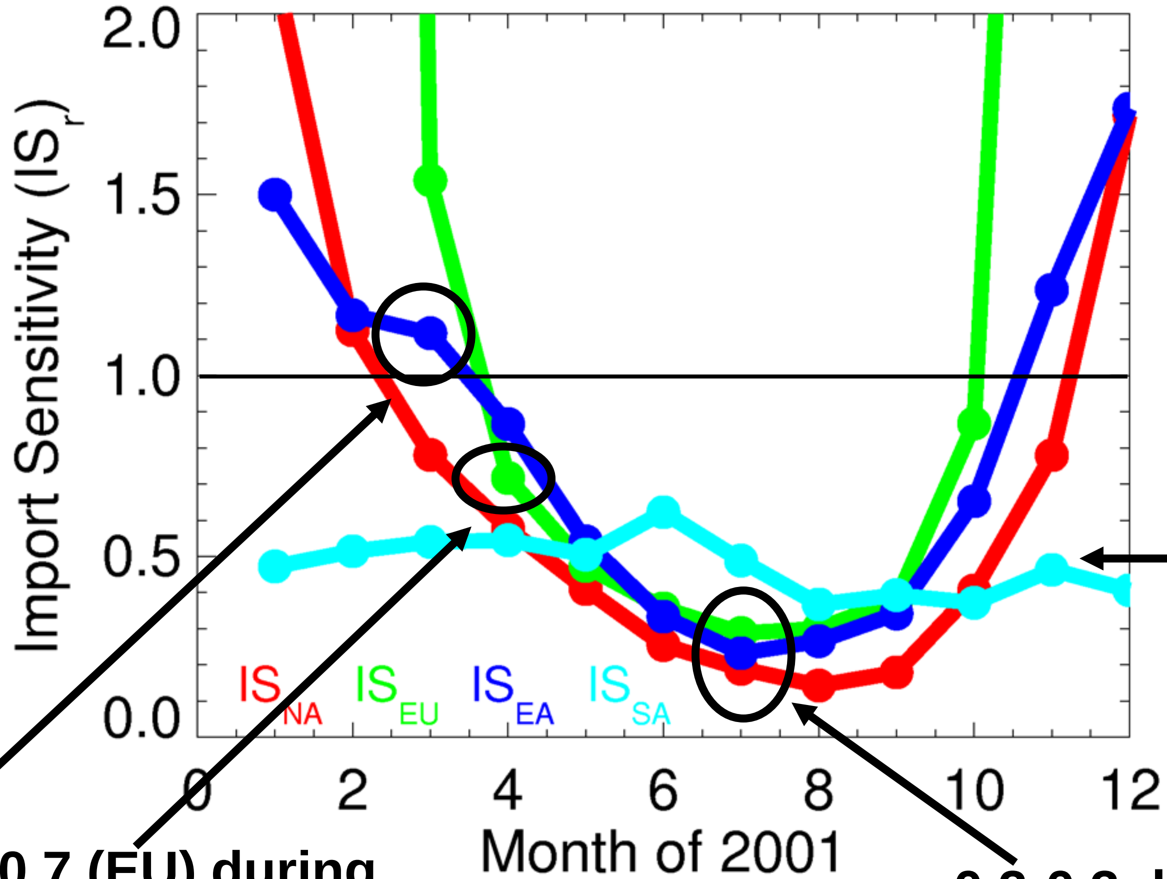
Seasonality of surface ozone response over North American and Europe to -20% foreign anthrop. emissions



1. Spring max due to longer O_3 lifetime, efficient transport [e.g., Wang et al., 1998; Wild and Akimoto, 2001; Stohl et al., 2002; TF HTAP 2007]
2. Response typically smallest to SA emissions (robust across models)
3. Similar response to EU& EA emissions over NA Apr-Nov (varies by model)
4. NA>EA>SA over EU (robust across models)

Fiore et al., JGR, 2009

Monthly mean import sensitivities (surface O_3 response to foreign vs. domestic emissions)



SA fairly constant ~0.5

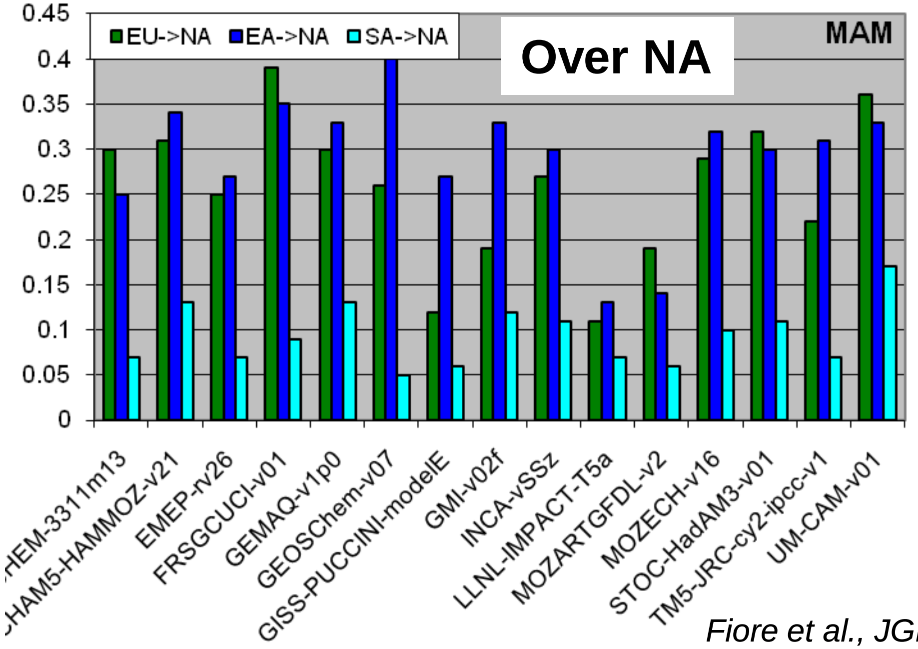
1.1 (EA), 0.7 (EU) during month with max response to foreign emissions

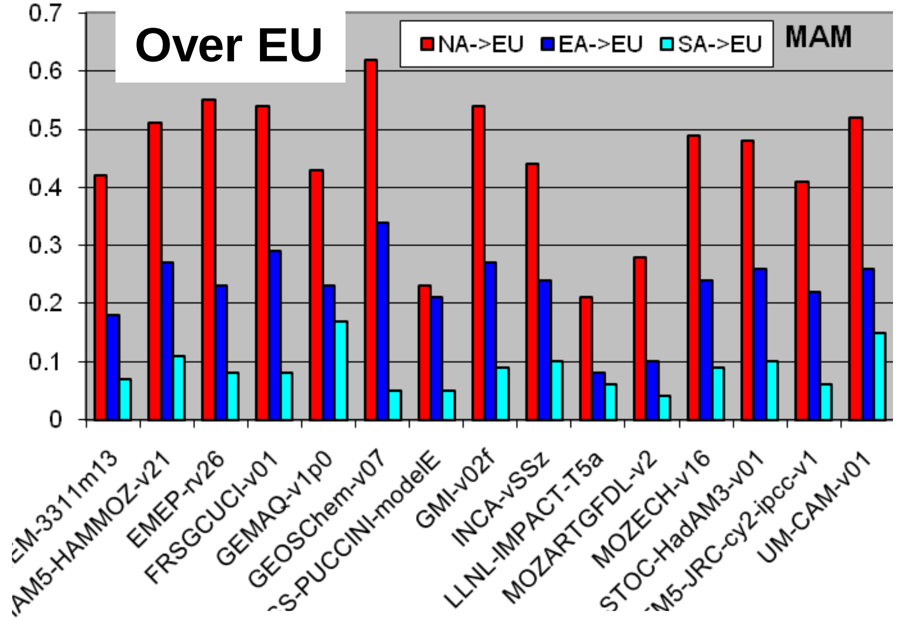
0.2-0.3 during month of max response to domestic emissions

Fiore et al., JGR, 2009

Models differ in estimates of surface O_3 response to foreign emission changes… which are best?

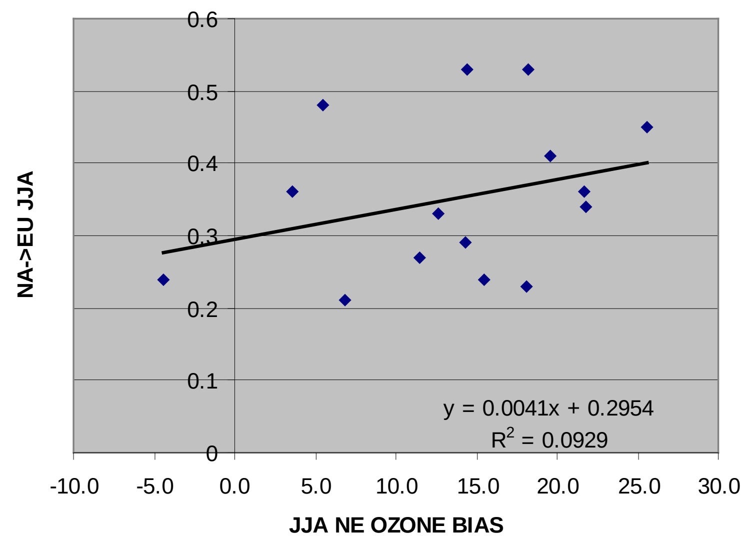
O_3 decrease from -20% foreign emissions



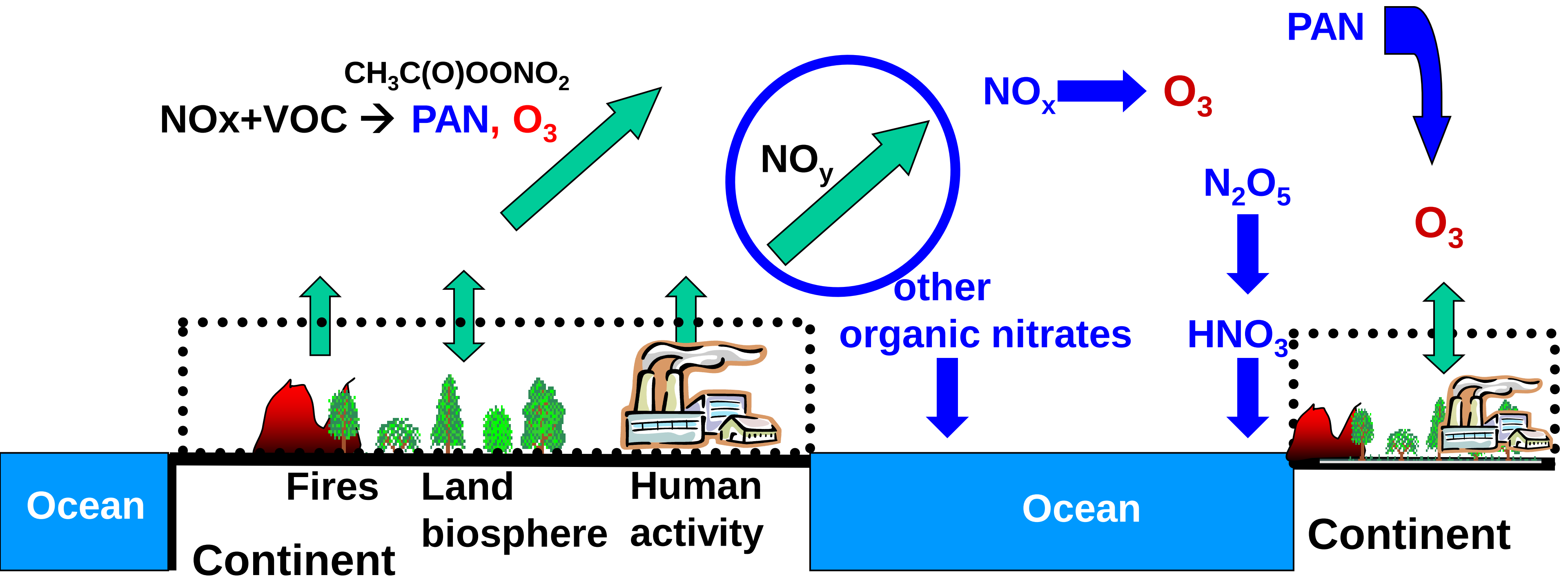


Fiore et al., JGR, 2009

No obvious relationship btw "base-case bias" and magnitude of source-receptor relationship (e.g., NA->EU)



Models likely differ in export of O_3 + precursors, downwind chemistry (PAN [Emmerson and Evans, ACP, 2009]), and transport to receptor region



NO_y partitioning (e.g., PAN vs. HNO_3) influences O_3 formation potential far from source region

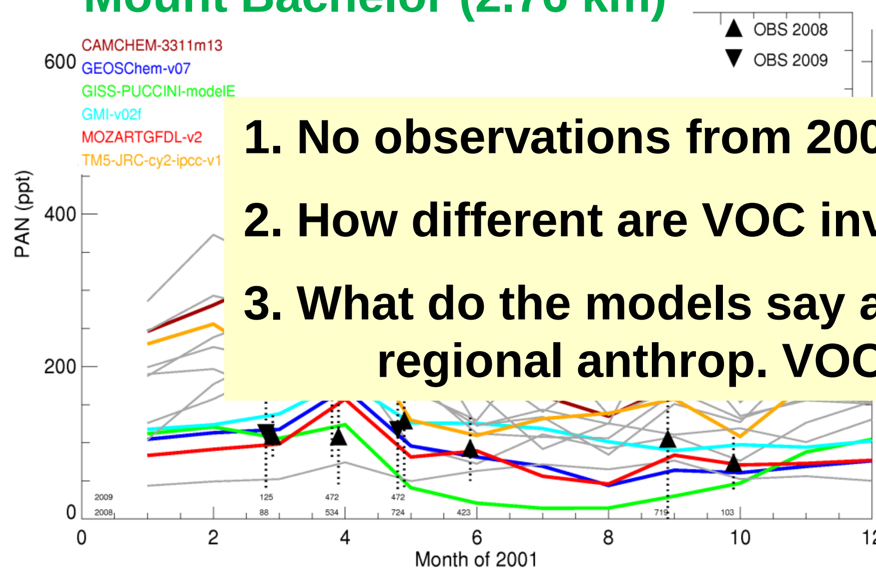
Observational evidence for O_3 production following PAN decomposition in subsiding Asian plumes [*e.g., Heald et al., JGR, 2003; Hudman et al., JGR, 2004; Zhang et al., 2008; Fischer et al., 2010*]

A.M. Fiore, May 6, 2010

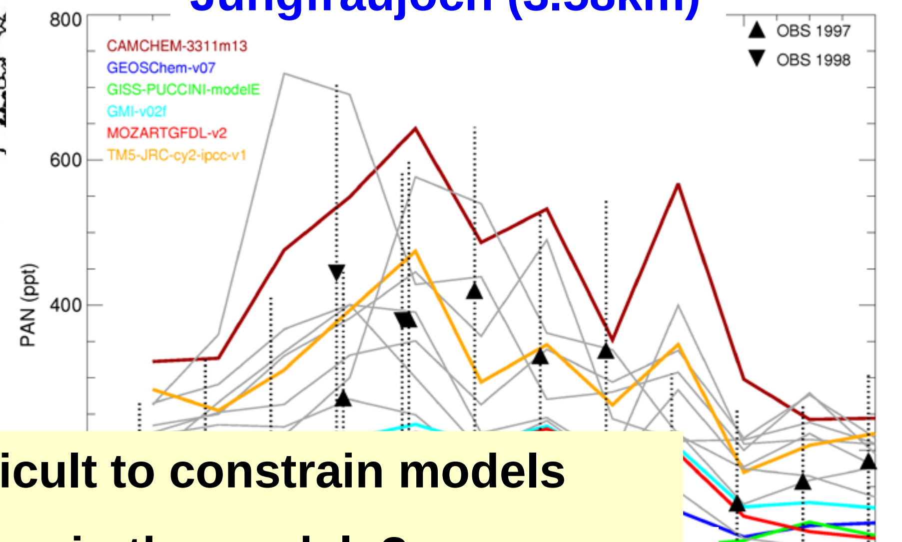
Compare models with PAN measurements at high elevation sites in Europe (3 sites) and WUS (2 sites)

c/o Emily Fischer, U Washington

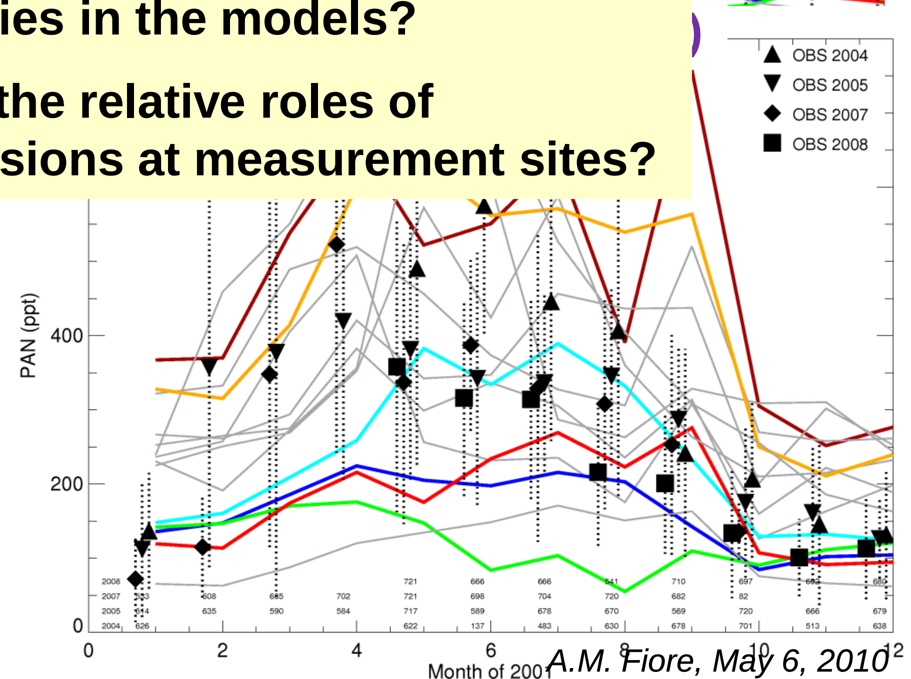
Mount Bachelor (2.76 km)



Jungfraujoch (3.58km)

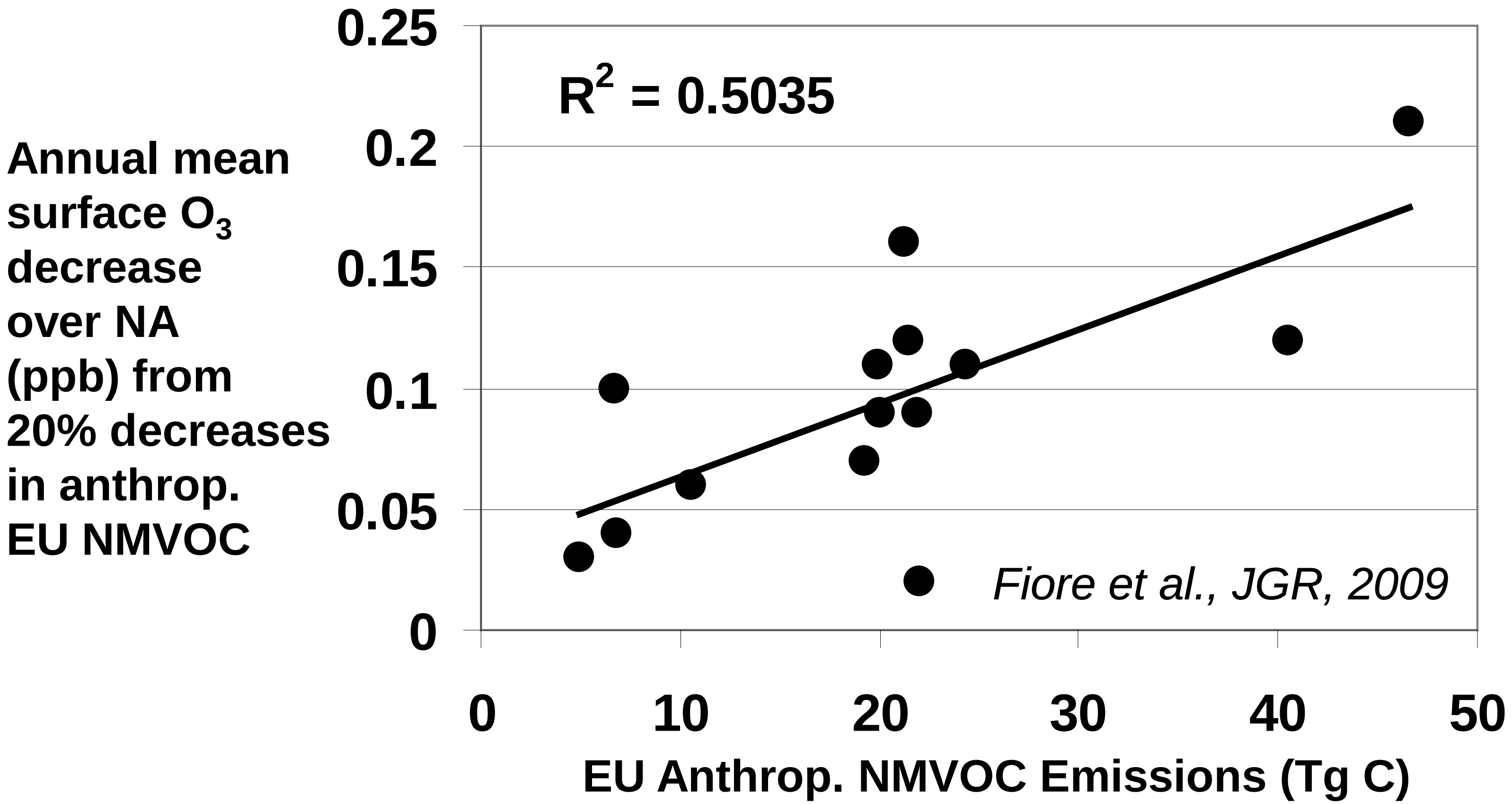


1. No observations from 2001; difficult to constrain models
2. How different are VOC inventories in the models?
3. What do the models say about the relative roles of regional anthrop. VOC emissions at measurement sites?



Model differences in lightning NO_x affect PAN + impact of anthro. emis. [*Fang et al., in review*] in addition to anthrop. sources, chemistry, and transport

Strong sensitivity of exported EU O_3 to large spread in EU NMVOC inventories (anthrop. NO_x fairly similar across models)

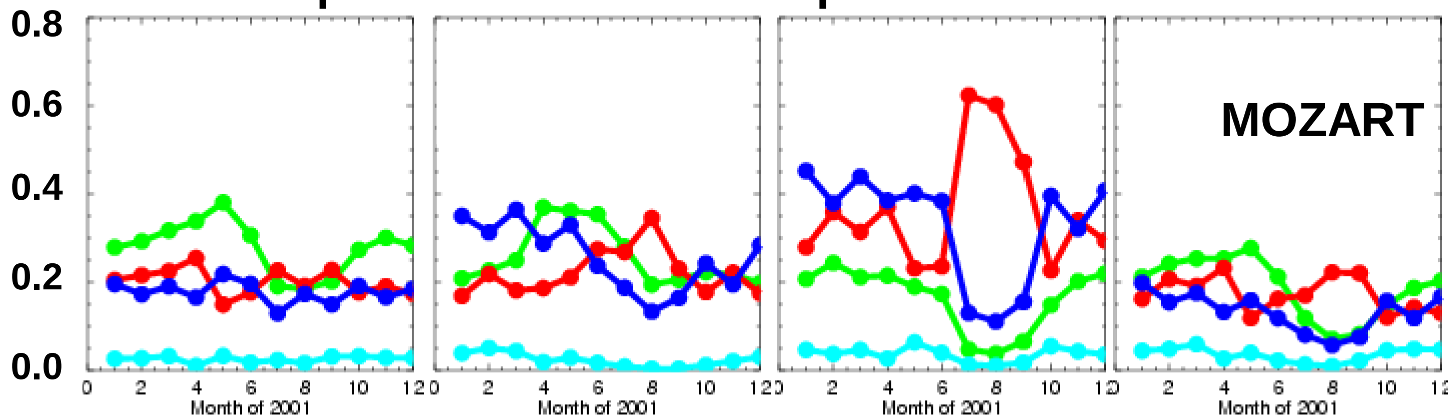


Do the relative contributions from different source regions in the model correlate with NMVOC emissions?

Relative influence of regional O_3 precursors on PAN at Mount Bachelor (OR), as estimated by the HTAP models

4 example HTAP models sampled at Mount Bachelor

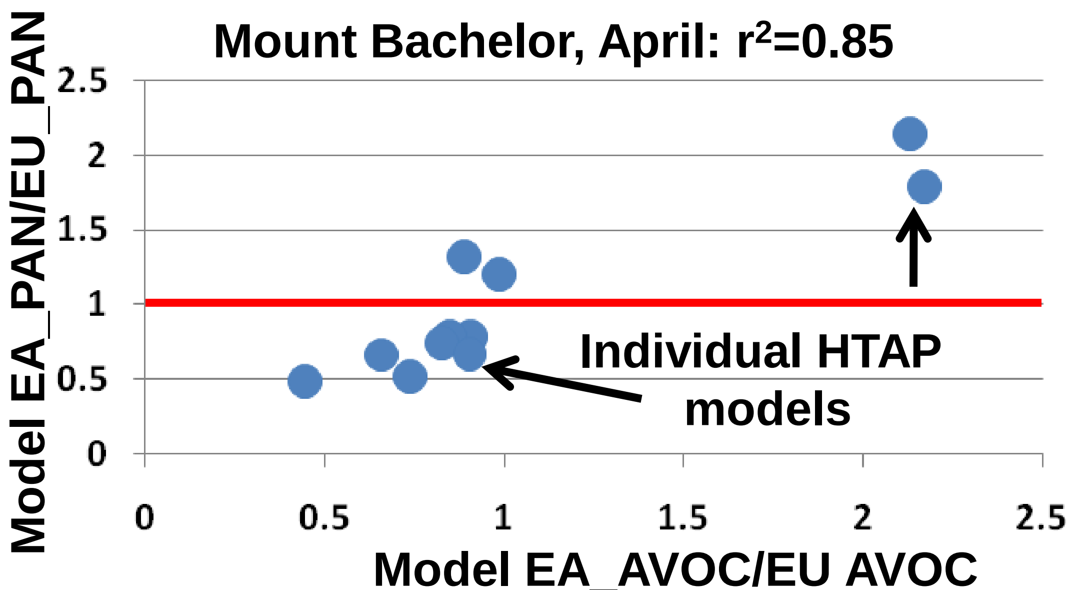
Fraction of total PAN from source region
EU NA EA SA



More EA influence

More EU influence

Mount Bachelor, April: r^2=0.85



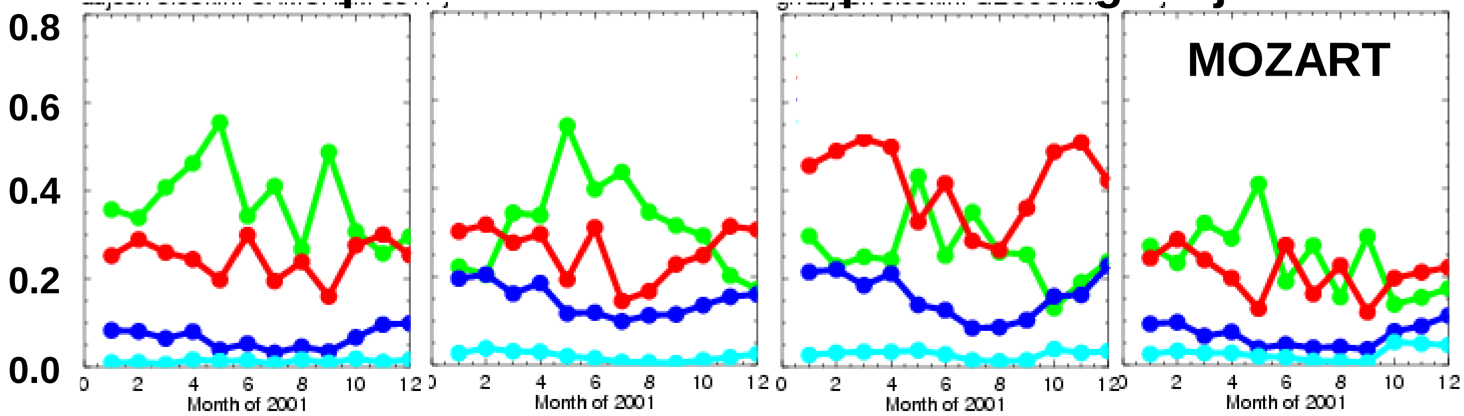
EA/EU PAN influence correlates with EA/EU AVOC emissions

A.M. Fiore, May 6, 2010

Model differences in relative contributions of source regions to PAN at Jungfraujoch, Switzerland

4 example HTAP models sampled at Jungfraujoch

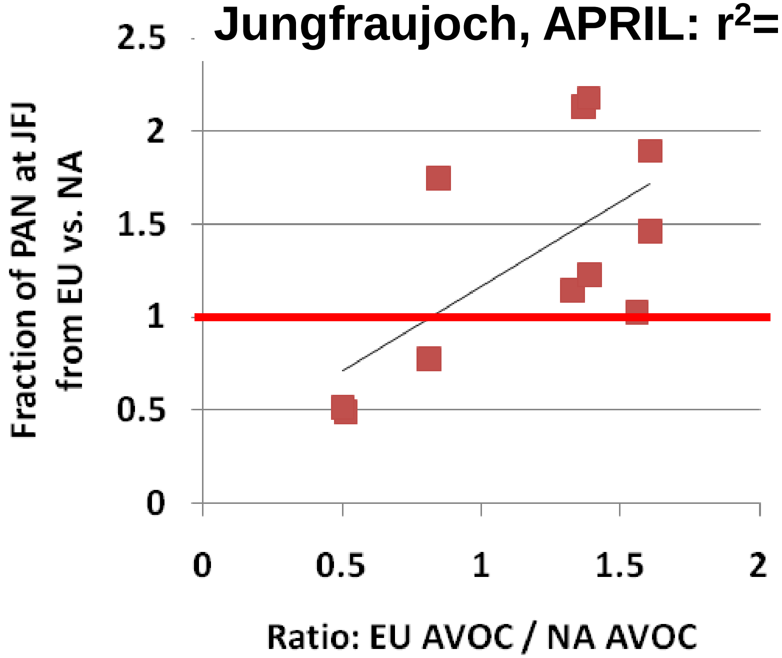
Fraction of total PAN from source region
EU NA EA SA



More EU influence

More NA influence

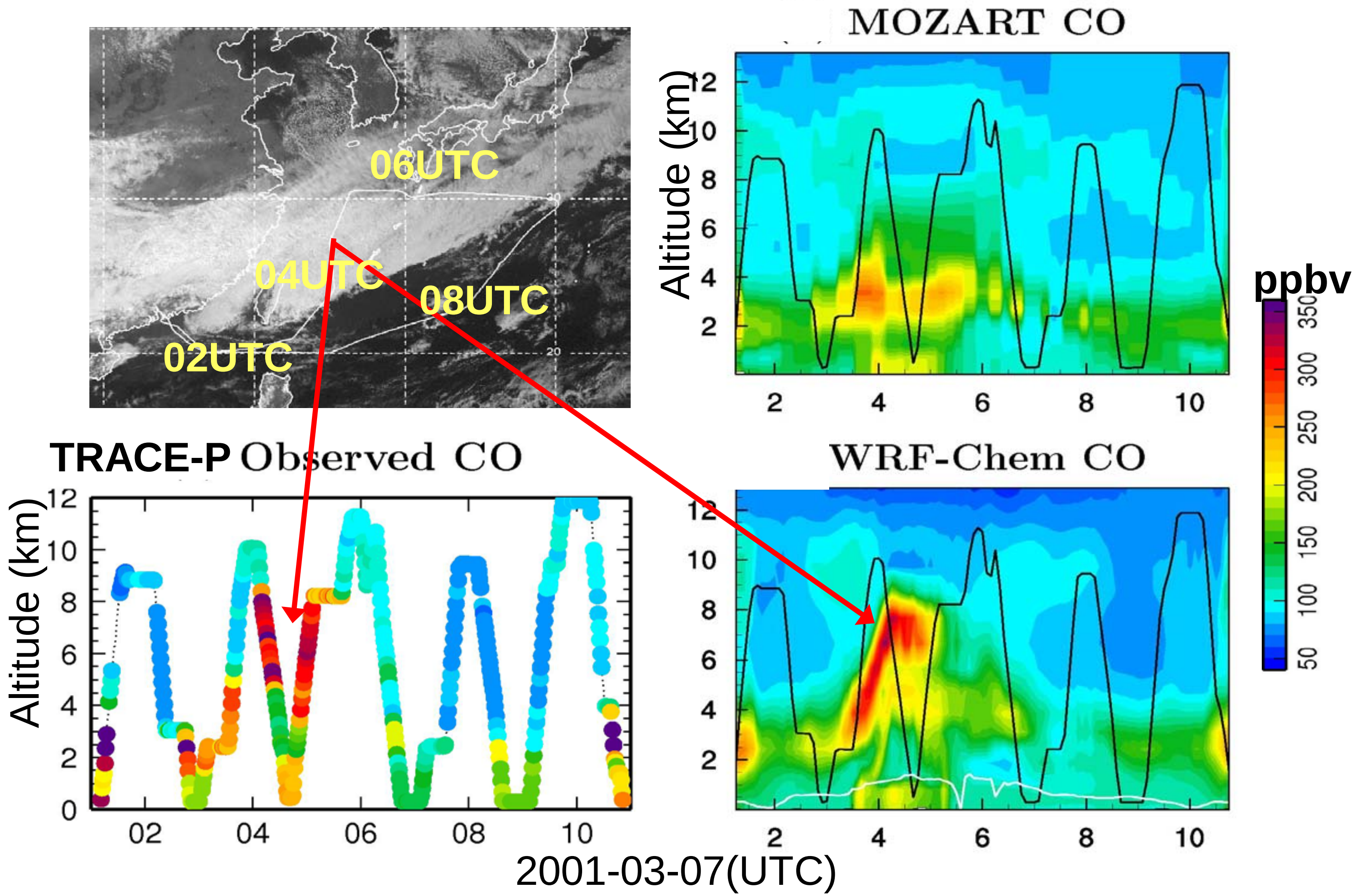
Jungfraujoch, APRIL: r^2=0.40



Wide range of EU NMVOC inventory contributes to model discrepancies

A.M. Fiore, May 6, 2010

Major uncertainties from sub-grid processes: Deep convection at the leading edge of the convergence band and associated pollutant export missing in global model [Lin et al., ACP, in press]



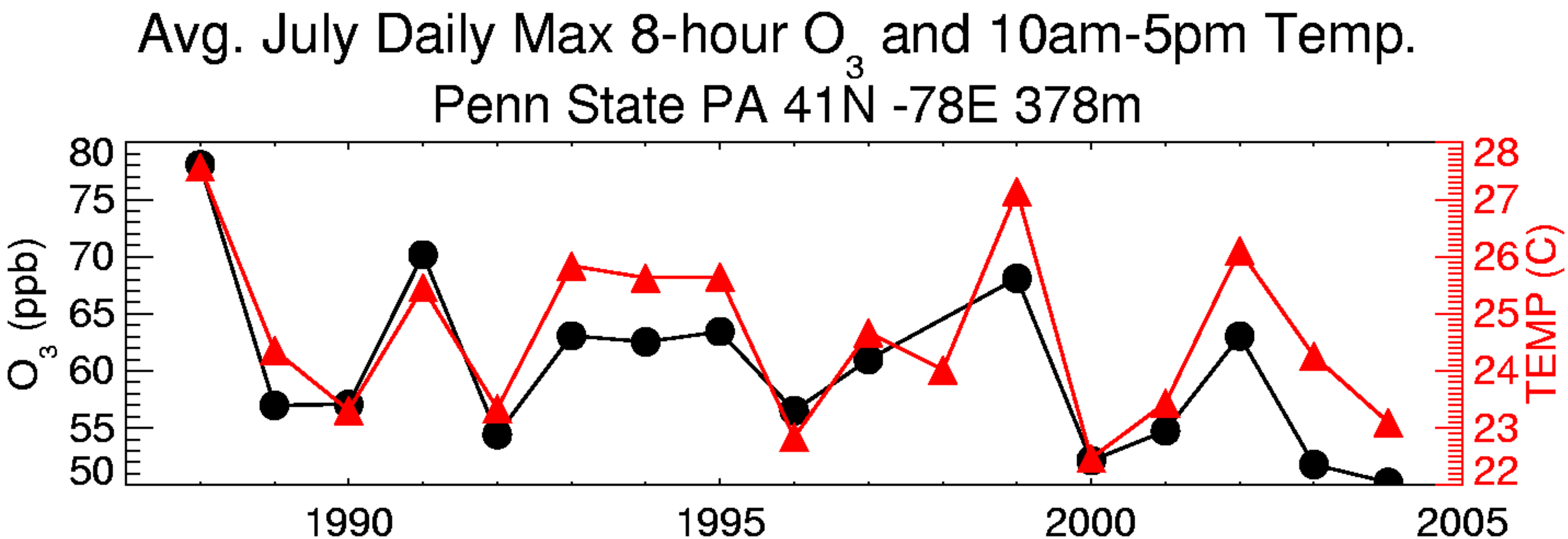
c/o Meiyun Lin, UW-Madison

How well do models represent mixing of free trop. air to the surface?

Summary of external influence on air quality #1: Hemispheric Transport of O_3

- Benchmark for future: Robust estimates + key areas of uncertainty
- "Import Sensitivities" (Δ O_3 from anthrop. emis. in the 3 foreign vs. domestic regions): 0.5-1.1 during month of max response to foreign emis; 0.2-0.3 during month of max response to domestic emissions
- Variability of O_3 response to emission changes within large HTAP regions
- Potential for PAN, NOy, other species, to help constrain model O_3 response to emission changes
- Role of "missing" processes (e.g., mesoscale)

External influence on air quality #2: Changing climate



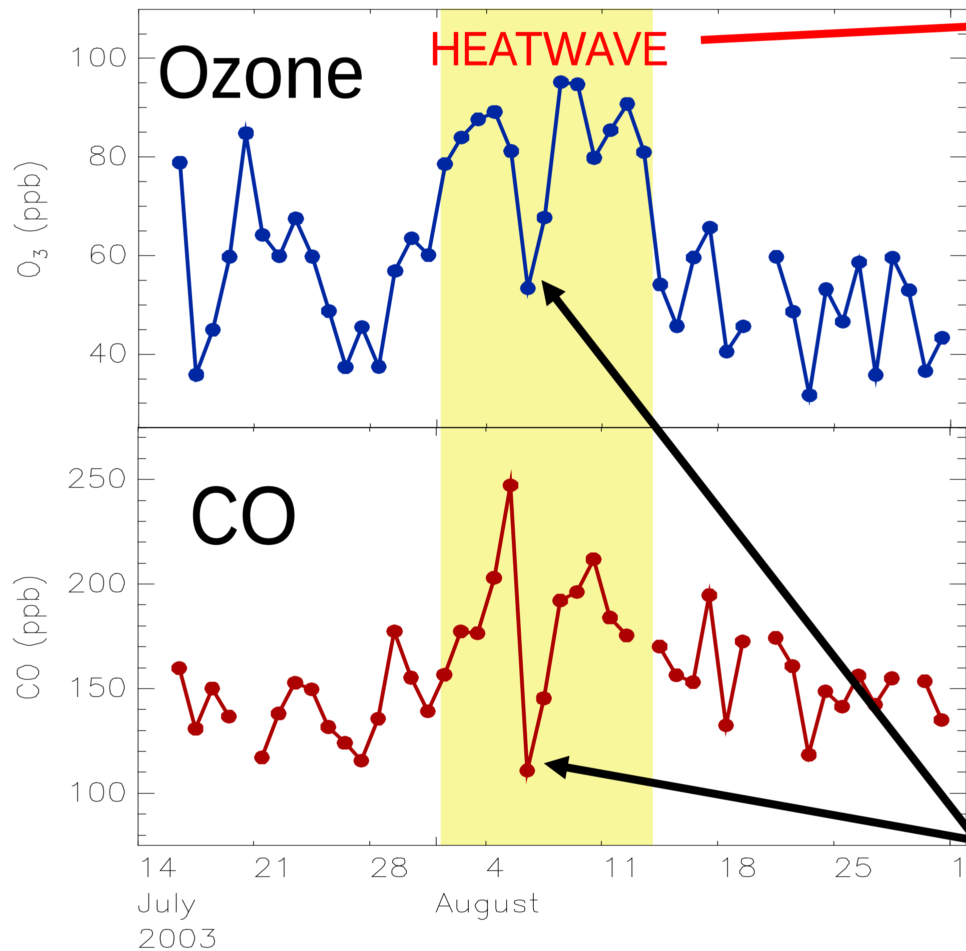
Observations c/o Jenise Swall and Steve Howard, U.S. EPA

Strong relationship between weather and pollution implies that changes in climate will influence air quality

Pollution build-up during 2003 European heatwave

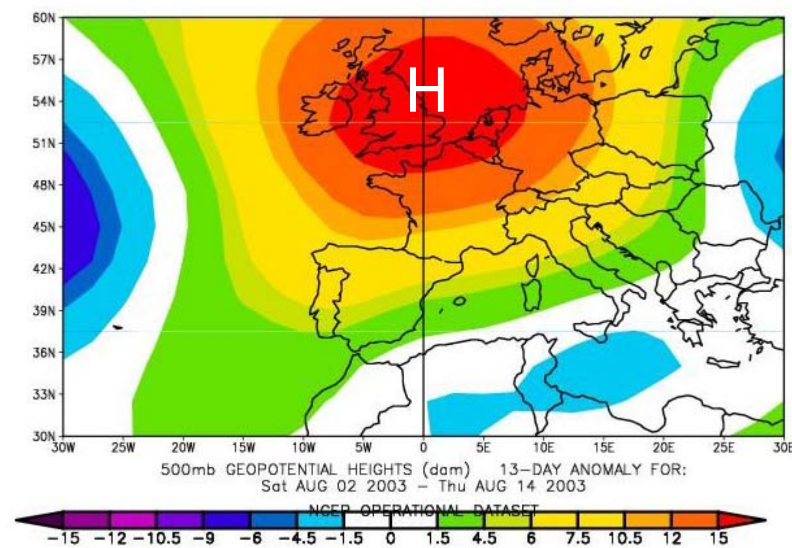
CO and O_3 from airborne observations (MOZAIC)

Above Frankfurt (850 hPa; ~160 vertical profiles



Stagnant high pressure system over Europe

(500 hPa geopotential anomaly relative to 1979-1995 for 2-14 August, NCEP)

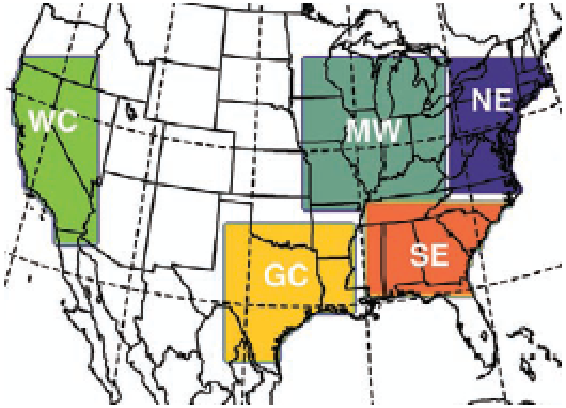


Ventilation
(low-pressure system)

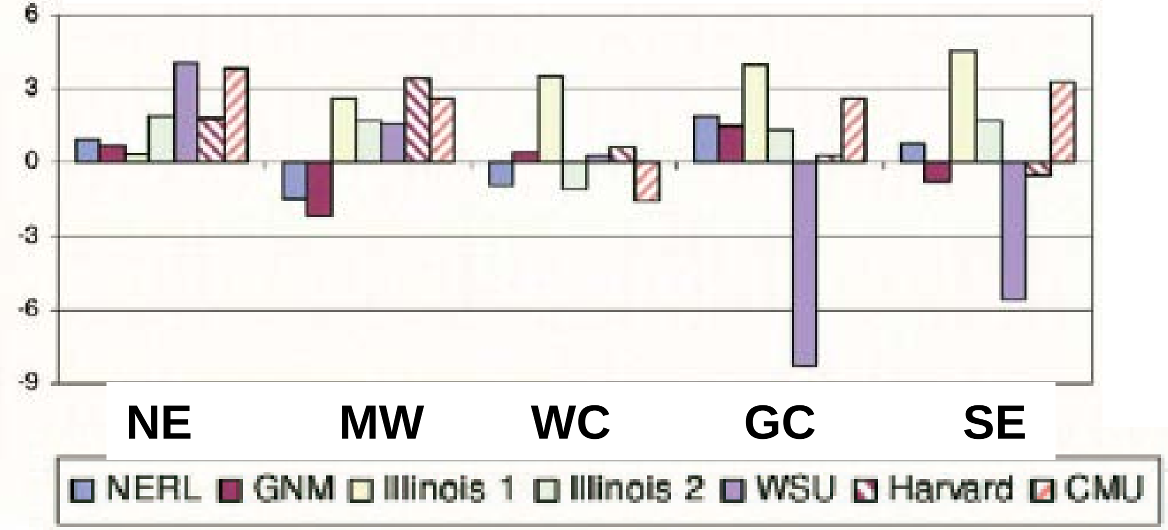
Model estimates of climate change impact on U.S. surface ozone [*Weaver et al., BAMS, 2009*]

ROBUST FINDINGS:

1. Increased summer O_3 (2-8 ppb) over large U.S. regions
2. Increases are largest during peak pollution events



a) Modeled changes in summer mean of daily max 8-hour O_3 (ppb; future – present)



KEY UNCERTAINTIES:

1. Regional patterns of change in meteorological drivers
2. Isoprene emissions and oxidation chemistry
3. Climate signal vs. interannual variability
4. Future trajectory of anthrop. emissions (not shown here)

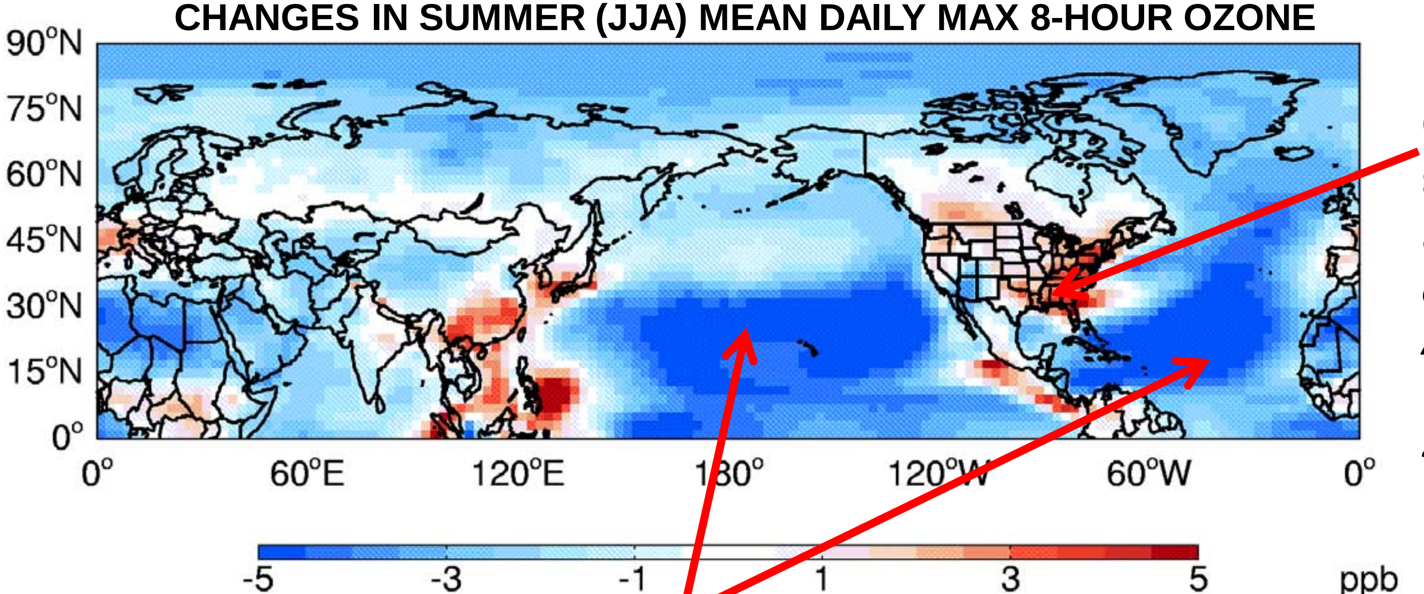
Summertime surface O_3 changes in a warmer climate in the new GFDL chemistry-climate model (AM3)

20-year simulations with annually-invariant emissions of ozone and aerosol precursors

Present Day Simulation ("1990s"): observed SSTs + sea ice (1981-2000 mean)

Future Simulation ("A1B 2090s"): observed SSTs + sea ice + average 2081-2100 changes from 19 IPCC AR-4 models

CHANGES IN SUMMER (JJA) MEAN DAILY MAX 8-HOUR OZONE



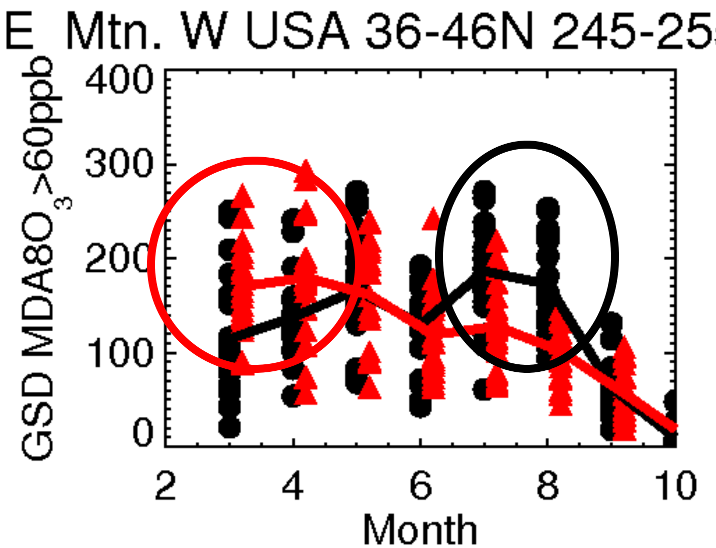
Previously noted degradation of summertime EUS O_3 air quality ***e.g., reviews of Jacob and Winner, Atmos. Environ. 2009 and Weaver et al., BAMS, 2009***

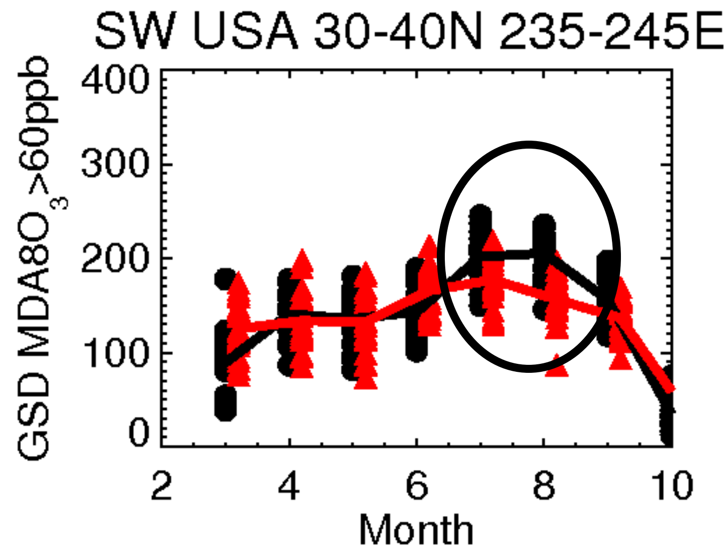
Previously noted decrease of lower troposphere background O_3
e.g., Johnson et al., GRL, 2001; Stevenson et al., JGR, 2006

Preliminary future climate simulations suggest more days with O_3 > 60 ppb in western US in spring

FUTURE: ▲ individual years — 20-yr mean (climate change only)

PRESENT: ● individual years — 20-yr mean





Increase in background? Strat. O_3?

More local destruction or smaller imported background?

How does "background" ozone in WUS respond to climate change?

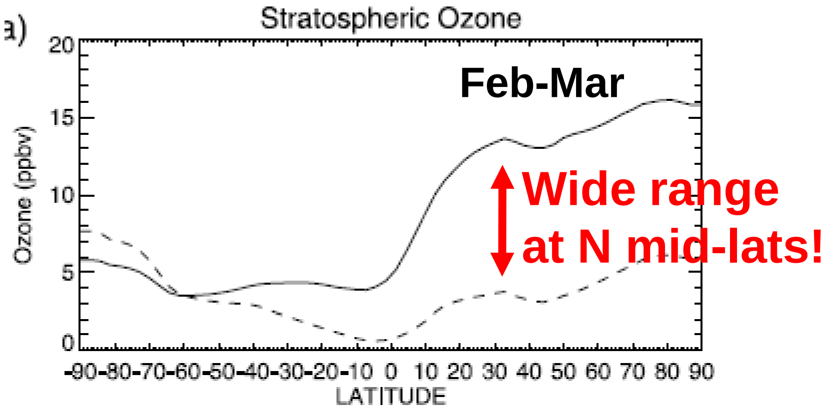
Warmer climate may increase strat-to-trop influence at northern mid-latitudes [*e.g., Collins et al., 2003; Zeng and Pyle, 2003; Hegglin and Shepherd, 2009; Li et al., 2009*]

A.M. Fiore, May 6, 2010

External influence #3: Stratospheric O_3 in surface air… Uncertain and controversial

Models differ in approaches and estimates for strat. O_3 in surface air *[e.g., Roelofs and Lelieveld, 1997; Wang et al., 1998; Emmons et al., 2003; Lamarque et al., 2005]*

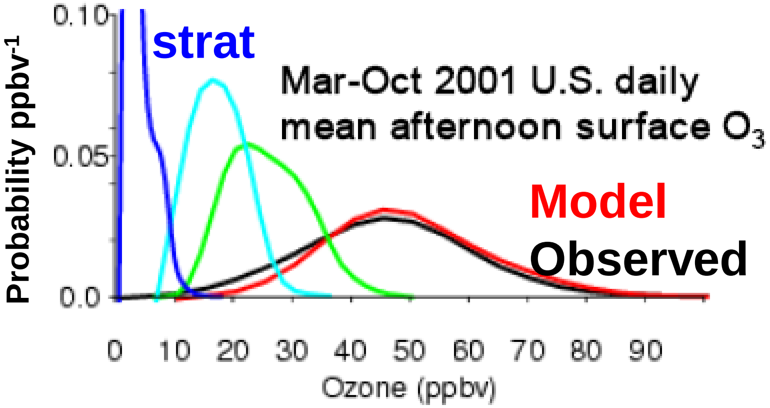
Hess and Lamarque, JGR, 2007: 2 approaches to estimate O_3-strat contribution in surface air



VIEW #2: *Langford et al., GRL, 2009*

Observations of direct strat. influence on surface O_3 at Front Range of CO Rocky Mtns in **1999;** could lead to O_3 standard exceedances

VIEW #1: *Fiore et al., JGR, 2003*

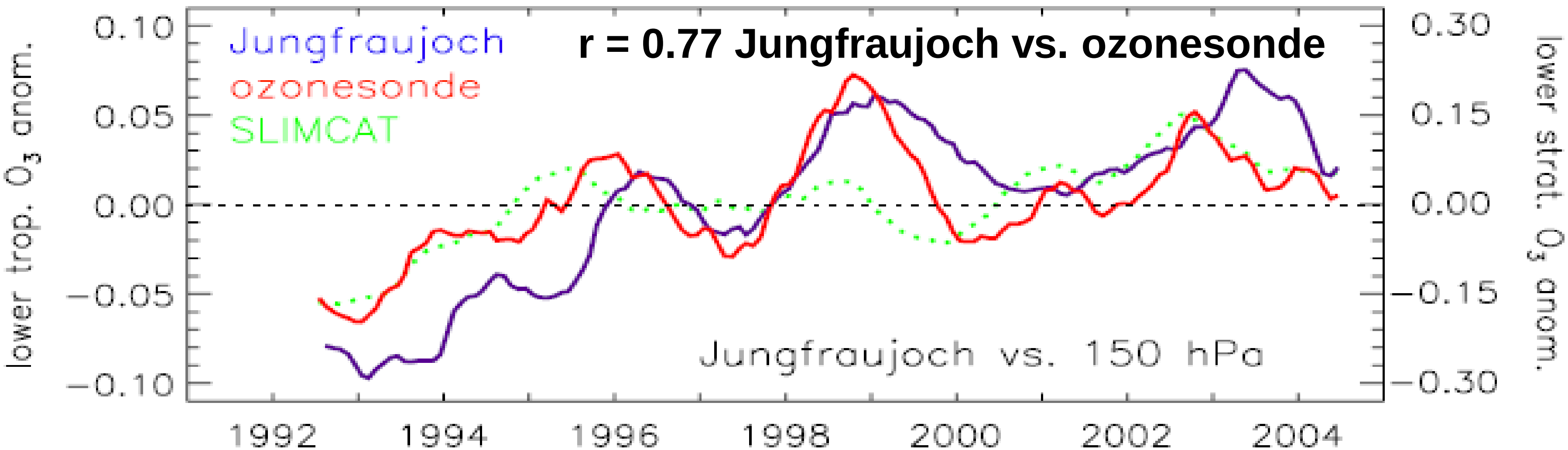


1. Prior interpretation strat. source in WUS [*Lefohn et al.*, 2001]) underestimated role of regional (+hemispheric) pollution
2. Direct strat. intrusions to surface are rare and should not compromise O_3 standard attainment (GEOS-Chem model year **2001**)

→Important to determine frequency of direct strat intrusions
→Focus in next few slides on variability

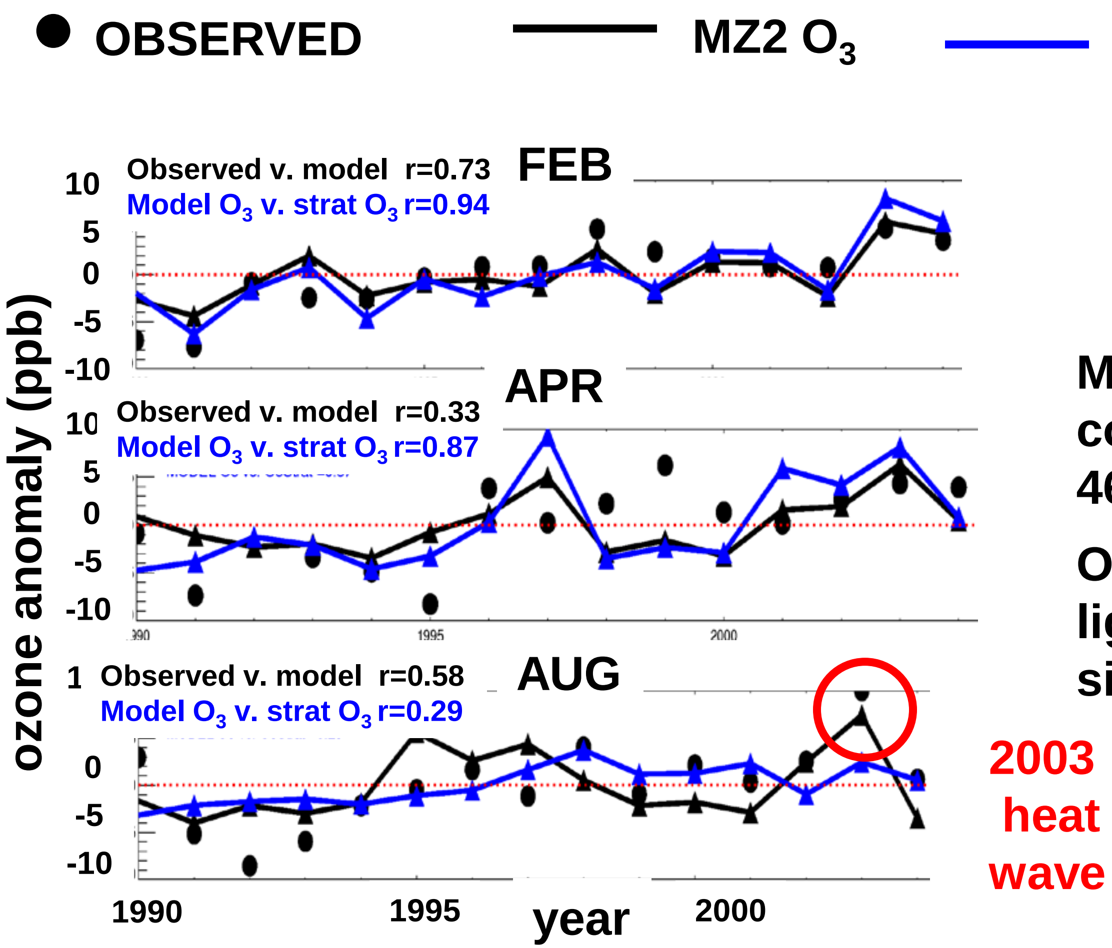
A.M. Fiore, May 6, 2010

Observations suggest influence of stratosphere on lower troposheric ozone over Europe in winter-spring



Ordóñez et al., GRL, 2007

MOZART-2 anomalies in O_3 and in strat. O_3 tracer are correlated in winter-early spring at Jungfraujoch

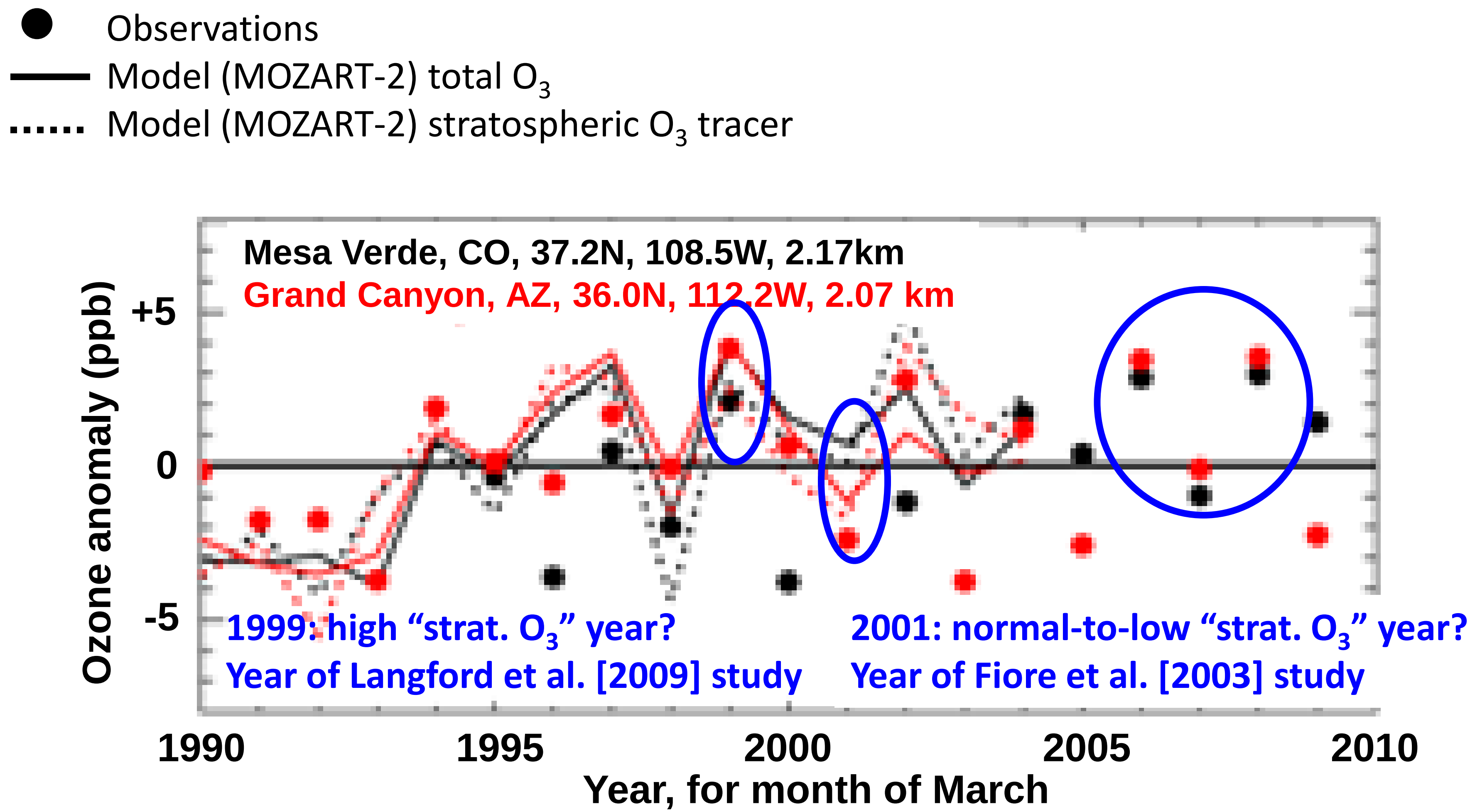


Model is sampled at grid cell containing Jungfraujoch site: 46.55°N, 7.98°E, 3.58km

Only meteorology (and lightning NO_x) vary in this simulation ***[Fiore et al., 2006]***

A.M. Fiore, May 6, 2010

Model indicates a role for stratospheric influence on interannual variability in O_3 at WUS sites in March



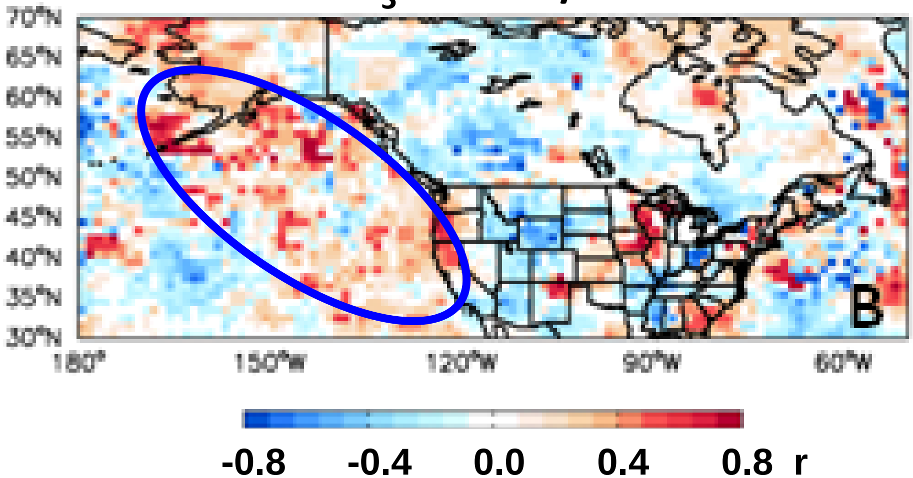
Can we exploit times when stratospheric influence dominates observed variability to determine "indicators" of enhanced strat O_3 contribution?

Potential for developing space-based "indicator" for day-to-day variability in strat O_3 influence at WUS sites?

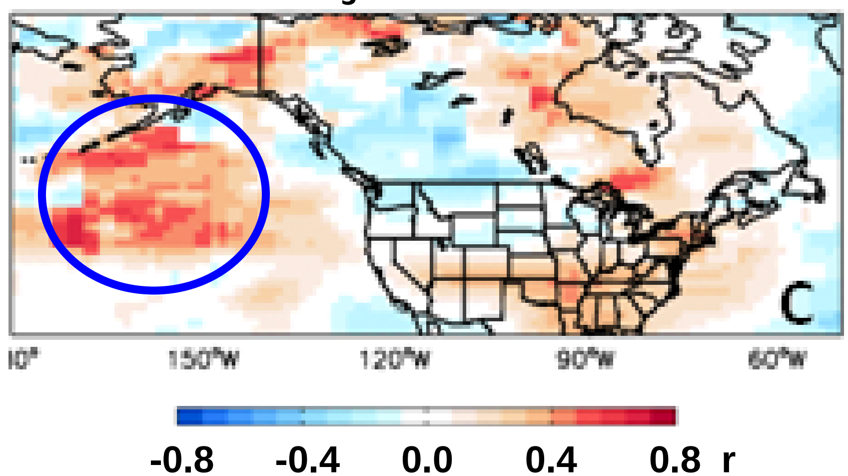
OMI/MLS products: trop. column O_3 (TCO) and strat. column O_3 (SCO) [*Ziemke et al.*, 2006].

Correlate daily anomalies in March in TCO and SCO with those at Mesa Verde CASTNET site, with ground site lagged (8 day lag shown below)

MEV O_3 vs. OMI/MLS TCO



MEV O_3 vs. MLS SCO



Signal of strat influence in upwind trop for WUS? or simply free trop signal?

"strat source region" for WUS?

A.M. Fiore, May 6, 2010

Concluding thoughts…
External influences on surface O_3 at N. mid-latitudes

- Intercontinental transport occurs year-round; peaks in spring
  → Uncertainties in spatiotemporal variability, role of sub-grid processes
  → Need better constraints on response to emissions perturbations

- Warming climate expected to degrade air quality in polluted regions
  → Uncertainties in regional climate response and isoprene-NO_x chemistry
  → Competing influences on tropospheric background ozone

- Stratospheric O_3 influence peaks in early spring, at high altitude sites
  → Uncertainties in contribution to surface air and variability
  → Need better process understanding on daily to decadal time scales

➢ **Implications for attaining ever-tightening air quality standards**

➢ **Potential insights from long-term *in situ* obs, satellite, models into role of meteorology/climate versus emissions on observed variability and trends**