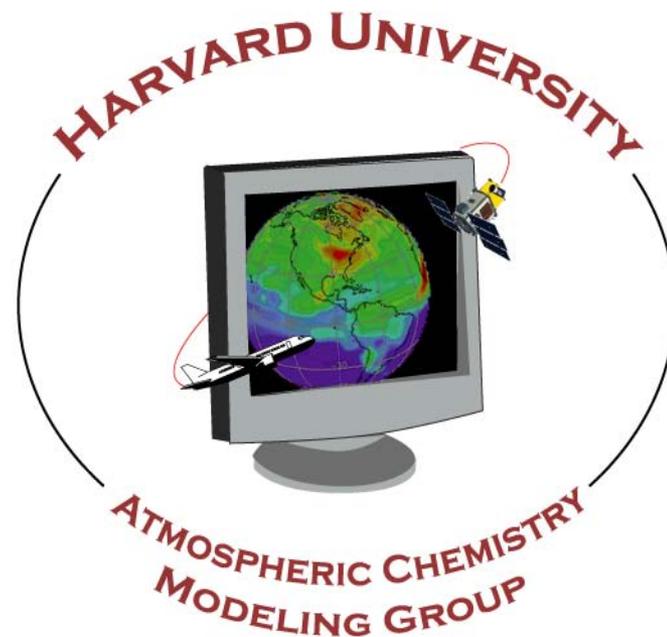
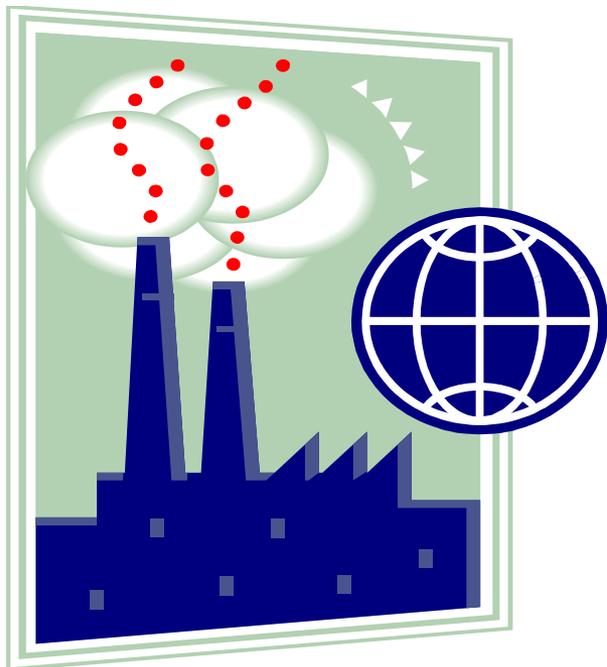


# Linking regional air pollution with global chemistry and climate: The role of background ozone

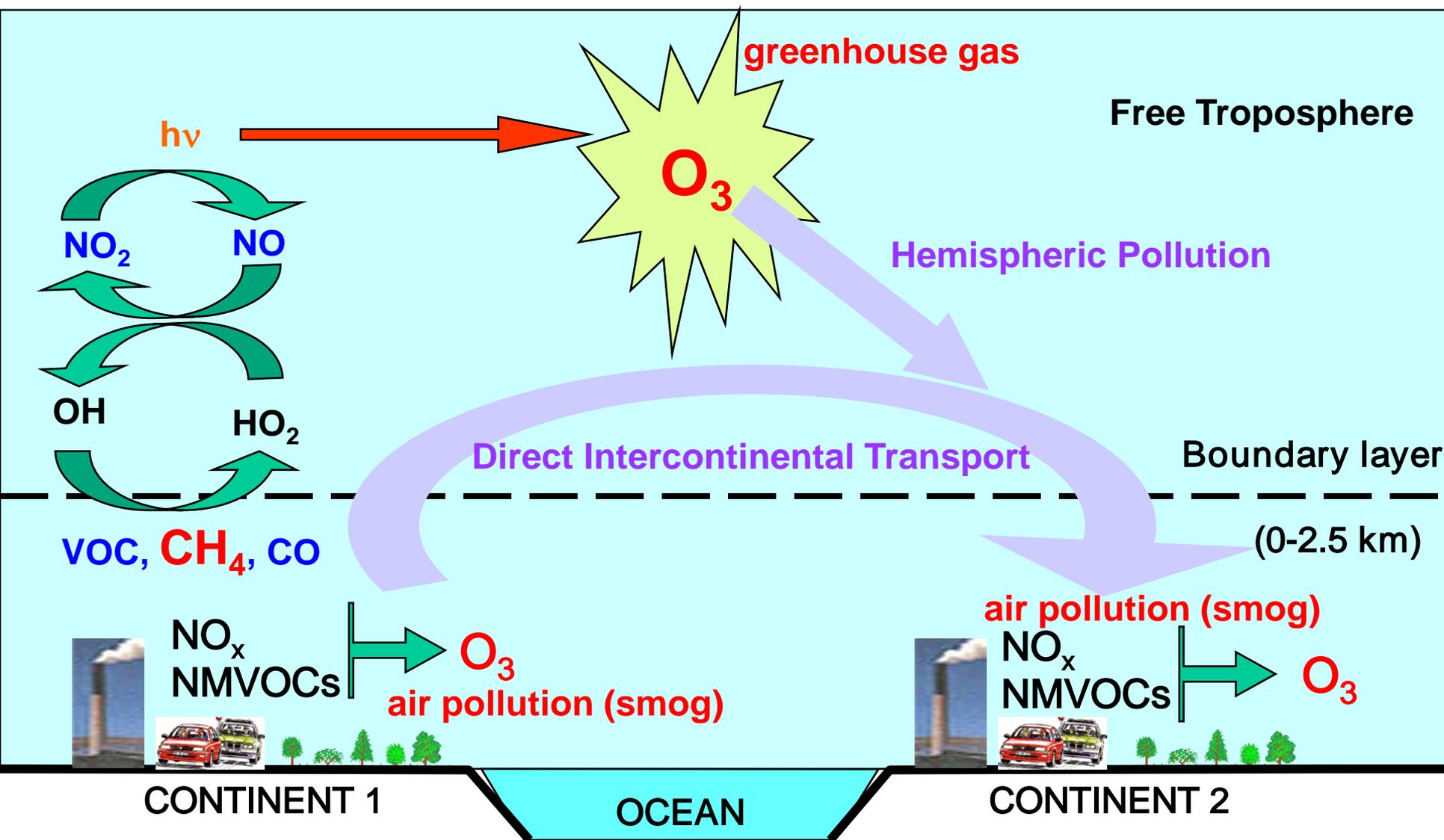


**Arlene M. Fiore**  
**Adviser: Daniel J. Jacob**

**April 22, 2002**

# Tropospheric ozone links air pollution & climate change

(1) primary constituent of smog in surface air [*NRC*, 1991]  
(2) 3<sup>rd</sup> most important greenhouse gas [*IPCC*, 2001]



# Ozone abatement strategies evolve as our understanding of the O<sub>3</sub> problem improves

O<sub>3</sub> smog recognized as an URBAN problem: Los Angeles, Haagen-Smit identifies chemical mechanism

Smog considered REGIONAL problem; role of biogenic VOCs discovered

A GLOBAL perspective: role of intercontinental transport, background

1950s

1980s

Present

Abatement Strategy:

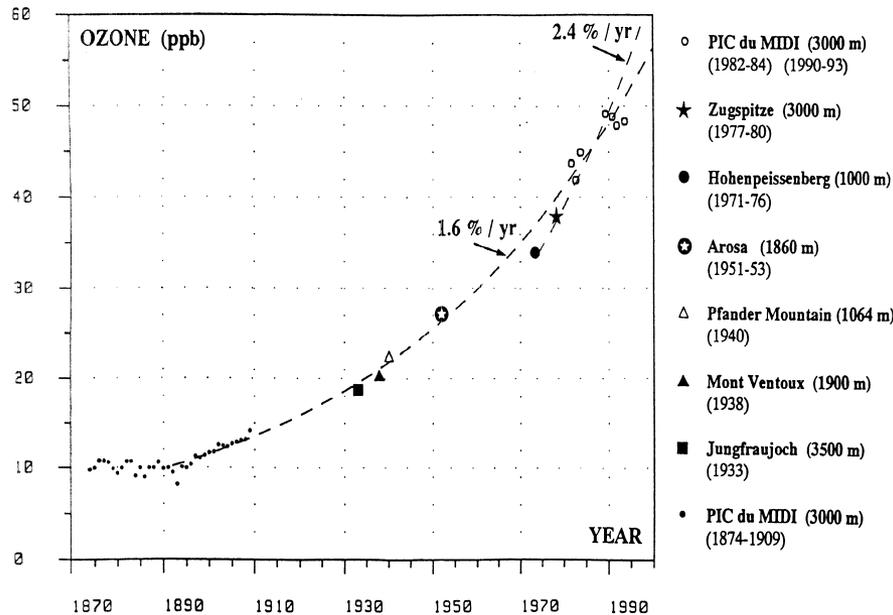
NMVOCs

+ NO<sub>x</sub>

+ CH<sub>4</sub>??

# Historical and recent evidence suggest that human activities are increasing the hemispheric O<sub>3</sub> background

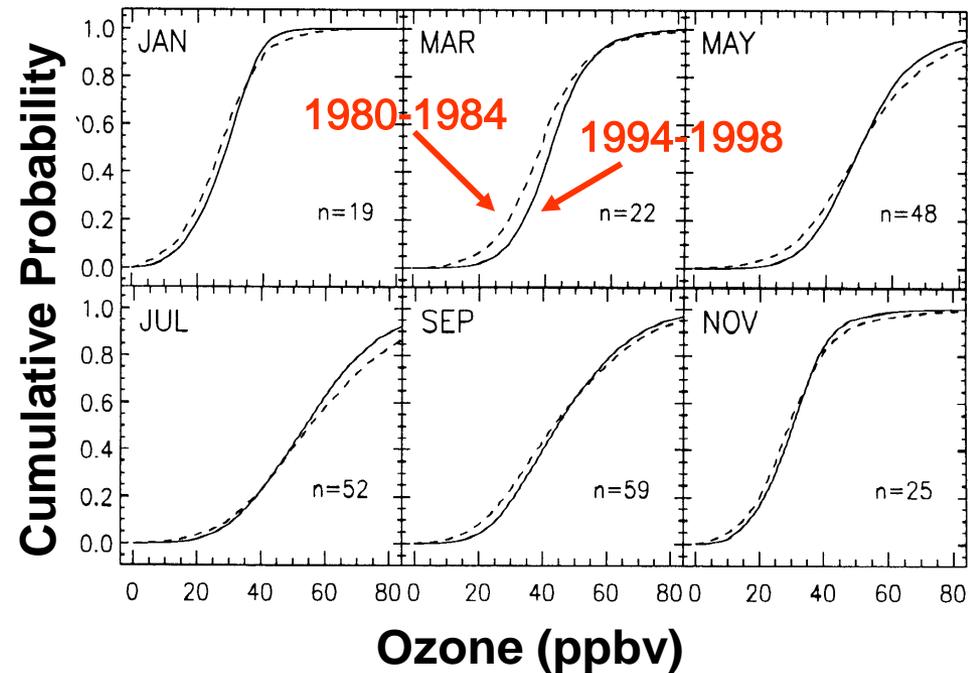
Ozone trend at European mountain sites, 1870-1990 [*Marenco et al., 1994*]



~ 5-fold increase over past century

8-h daily maximum ozone probability distribution at rural U.S. sites [*Lin et al., 2000*]

--- 1980-1984    — 1994-1998



~ 3 ppbv increase over past 20 years

# Need to quantify U.S. background O<sub>3</sub> in surface air for current review of National Ambient Air Quality Standard for O<sub>3</sub> (NAAQS)

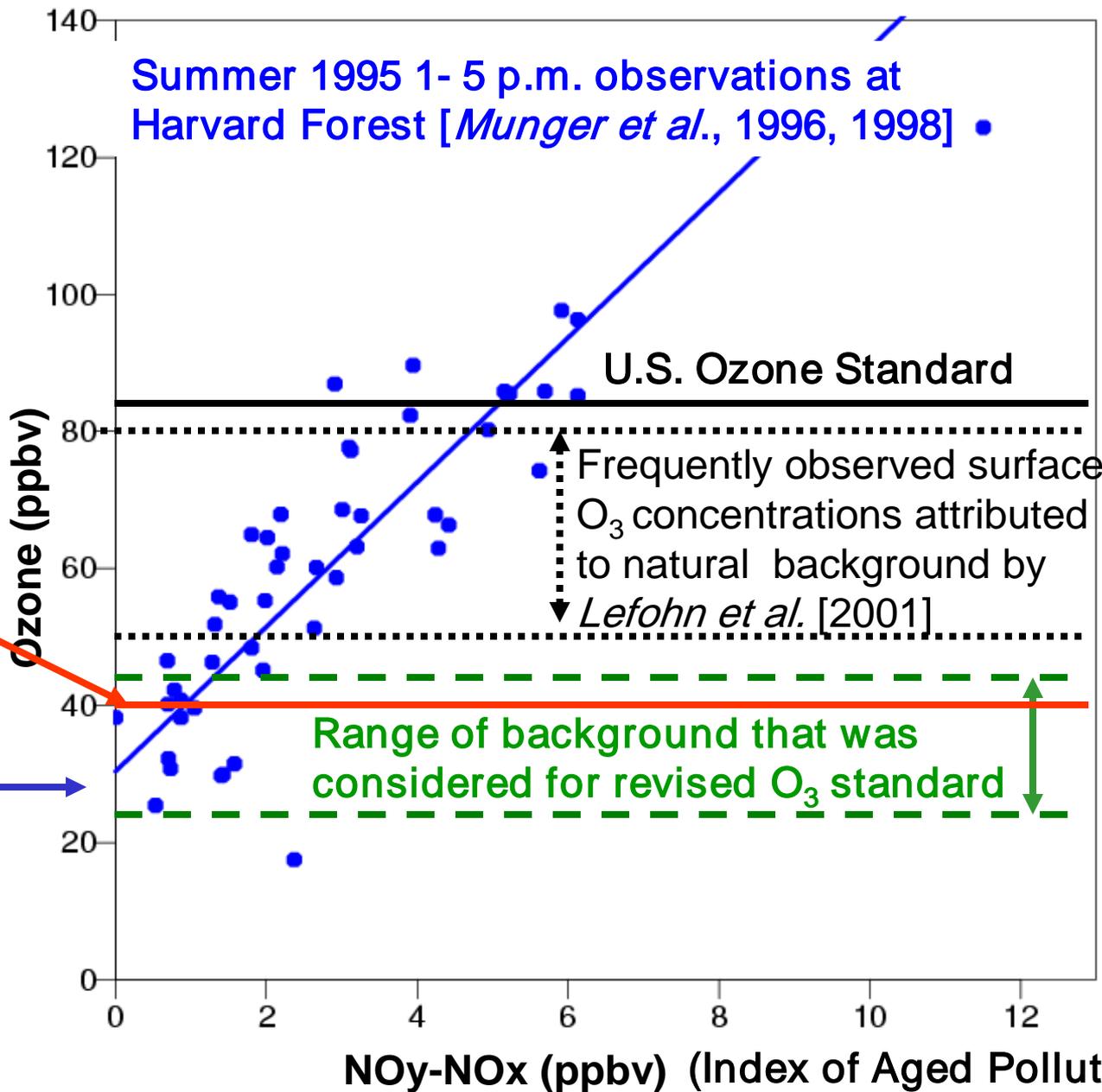
1. What background concentrations should be used to assess the human health risk associated with exposure to O<sub>3</sub>?
2. Is the present (or more stringent future) NAAQS too close to background concentrations?

## “REGULATORY BACKGROUND” DEFINITION:

Ozone concentrations that would exist in the absence of anthrop. emissions from North America [*EPA*, 2003]

25-45 ppbv [*EPA*, 1996]

# An approach for estimating O<sub>3</sub> background from observations



background presently used in EPA risk assessments

Intercept 30 ppb background (clean air)

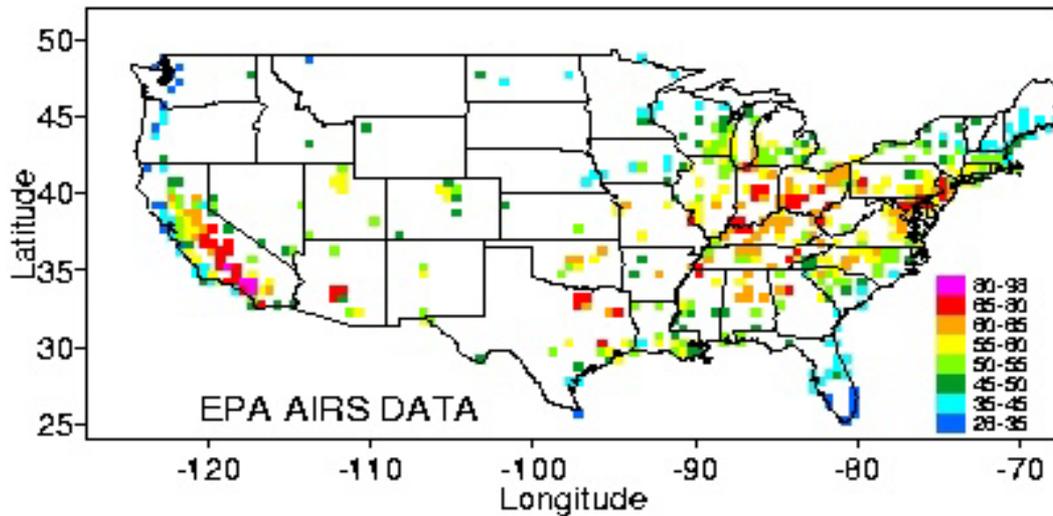
**QUESTION:** What role does background O<sub>3</sub> play in linking regional air quality with global chemistry & climate?

**TOOL:** GEOS-CHEM 3D Tropospheric Chemistry Model [*Bey et al.*, 2001]  
(uses assim. met.; 20-30  $\sigma$ ; 4°x5° or 2°x2.5° horiz. resn., 24 tracers)

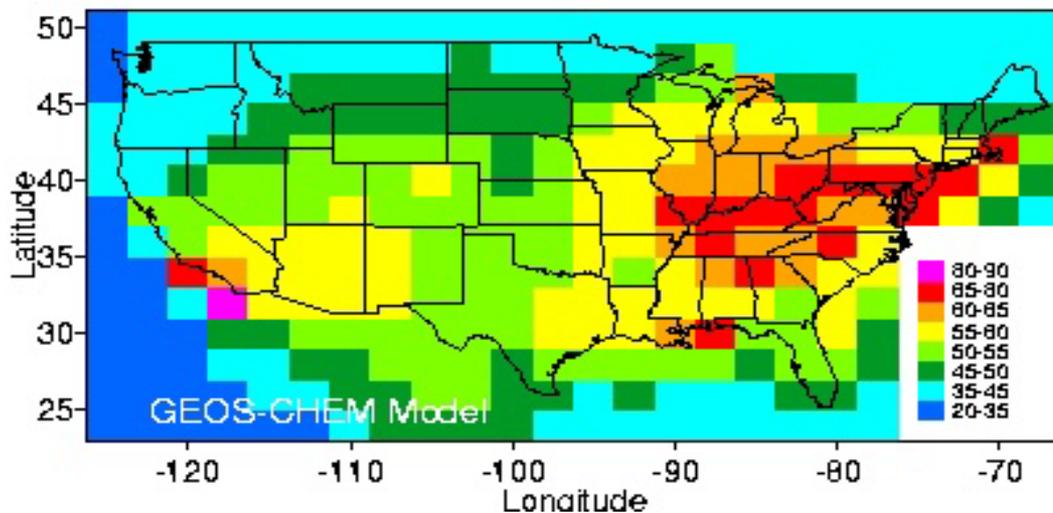
**RESEARCH OBJECTIVES:**

1. Assess whether GEOS-CHEM is a suitable tool for quantifying the O<sub>3</sub> background over the U.S.
  - Standard statistical metrics + EOF analysis
2. Quantify background contribution to average vs. polluted days
3. Identify origin of background and variability of its sources
4. Diagnose source of high-O<sub>3</sub> events at remote U.S. sites in spring
5. Determine combined air quality and climatic implications of various O<sub>3</sub> control strategies
  - Sensitivity simulations & tagged tracers in GEOS-CHEM

# Summer 1995 afternoon (1-5 p.m.) ozone in surface air over the United States



**AIRS  
Observations**



**GEOS-CHEM 2°x2.5°  
r = 0.66, bias=5 ppbv**

**(for 4°x5° resolution  
r = 0.84, bias=2 ppbv)**

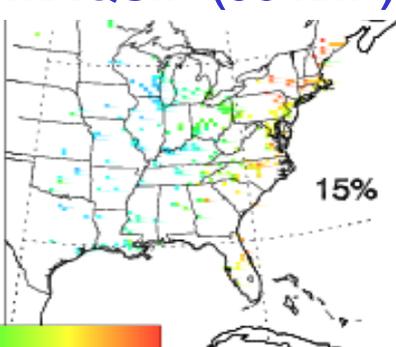
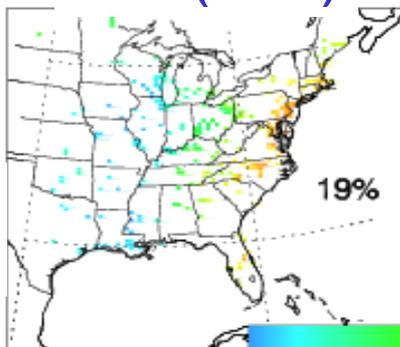
# EOF ANALYSIS: Characterize spatiotemporal variability of surface O<sub>3</sub> (daily 1-5 p.m. mean concentrations in summer 1995 over eastern U.S.)

OBS (AIRS)

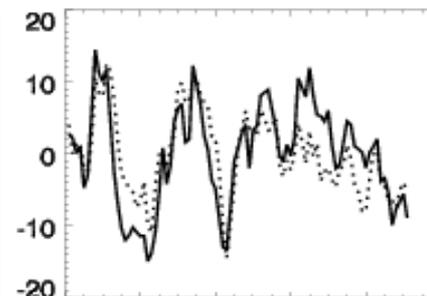
MAQSIP (36 km<sup>2</sup>)

East-west  
EOF

$r^2 = 0.86$   
Slope = 1.0



-0.12 -0.06 0.00 0.06

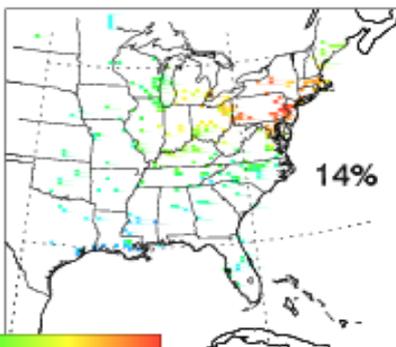
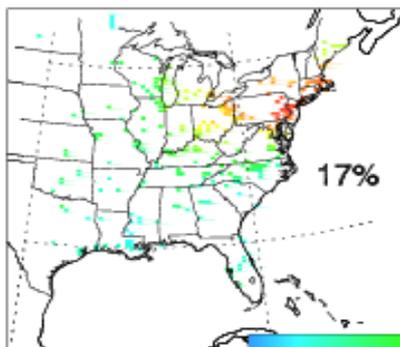


June July August

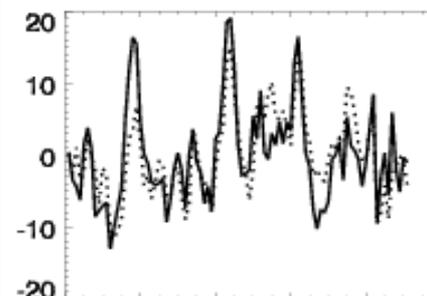
$r^2 = 0.60$   
Slope = 0.9

Midwest-  
Northeast  
EOF

$r^2 = 0.76$   
Slope = 1.0



-0.07 0.00 0.06 0.13

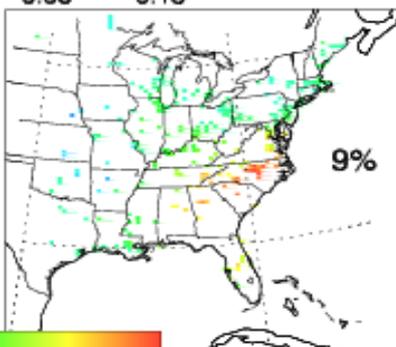
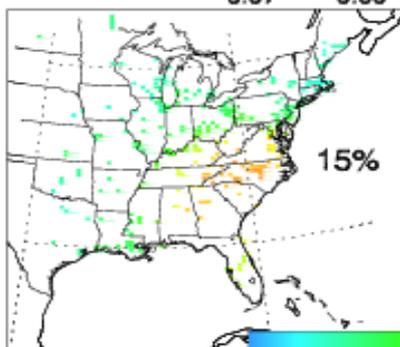


June July August

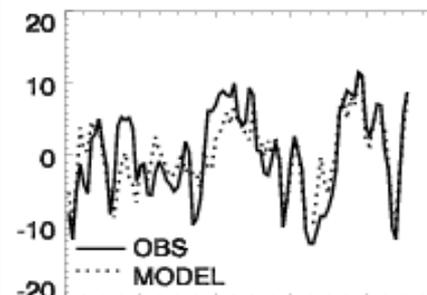
$r^2 = 0.57$   
Slope = 0.8

Southeast  
EOF

$r^2 = 0.80$   
Slope = 1.0



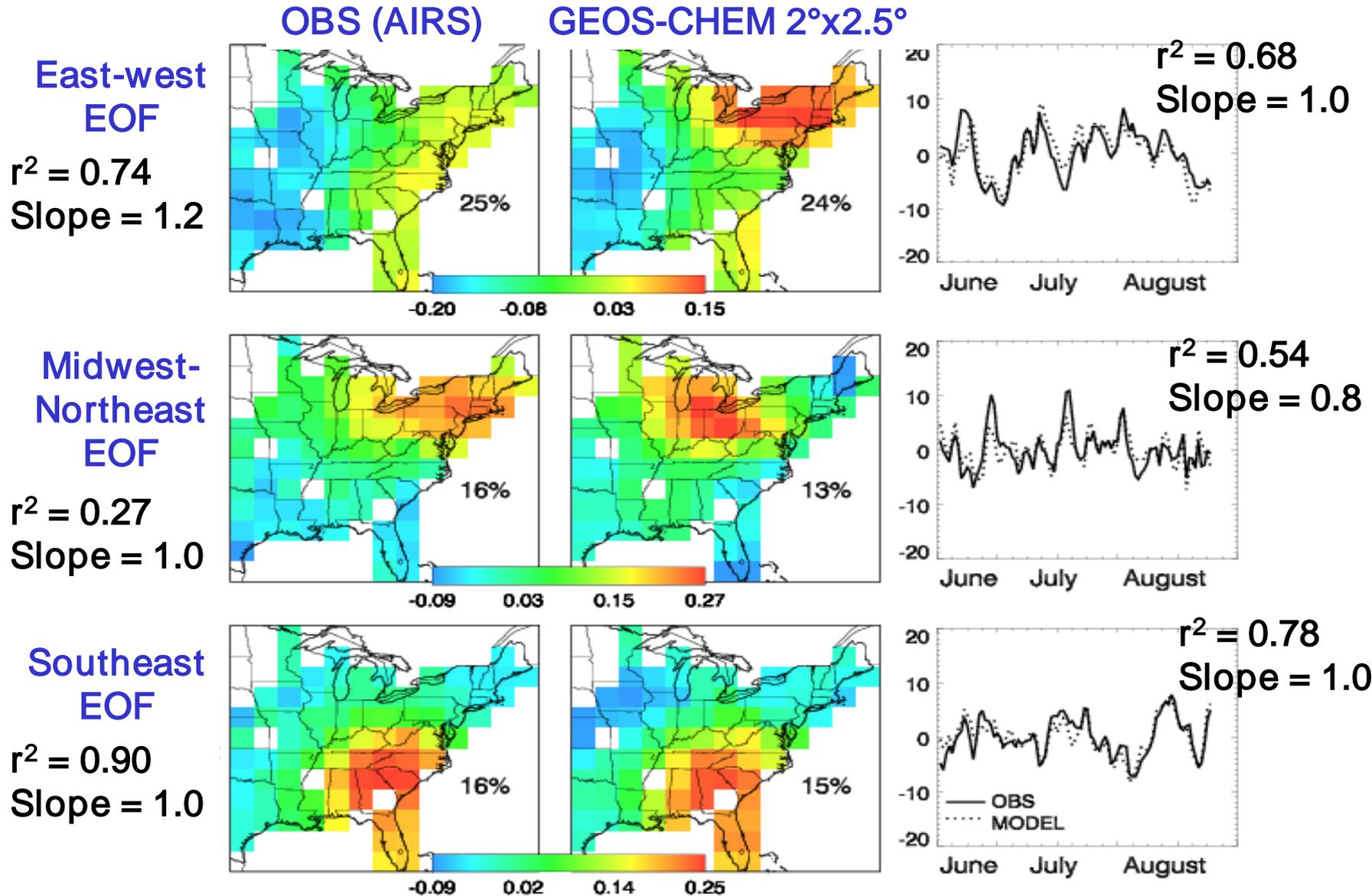
-0.09 0.00 0.08 0.17



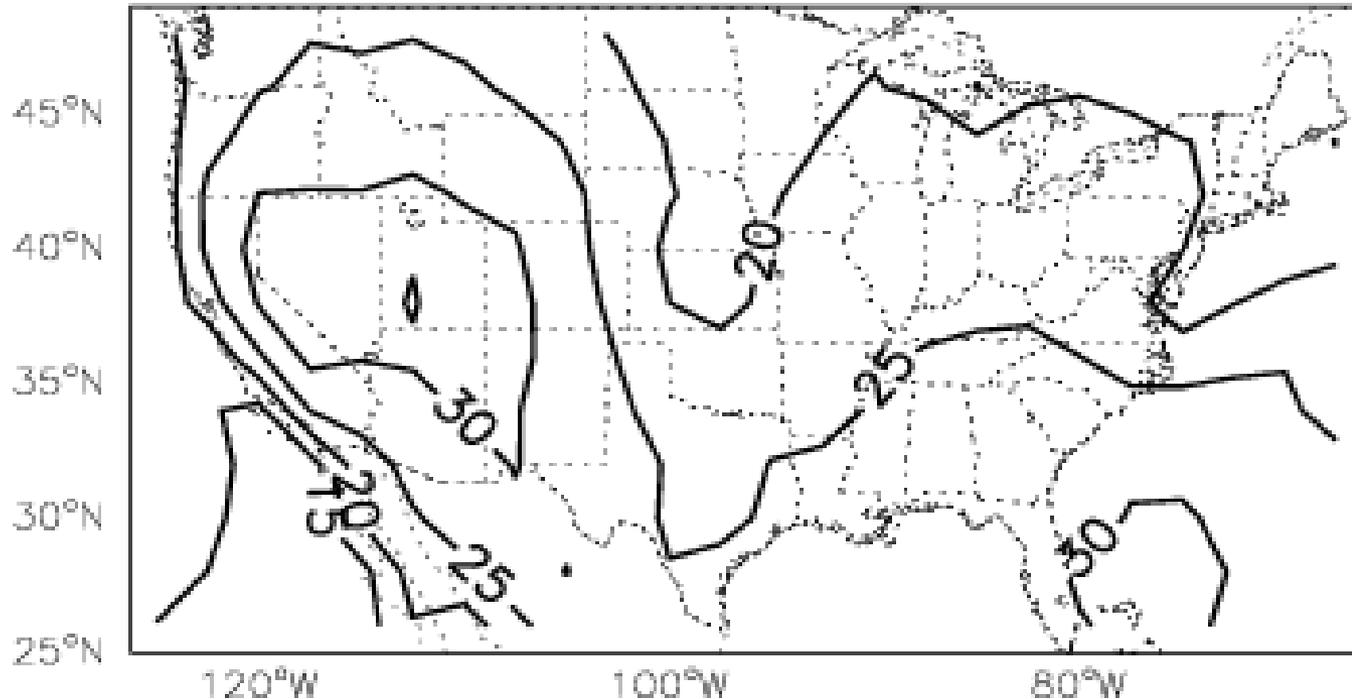
June July August

$r^2 = 0.68$   
Slope = 0.7

# Same fundamental, synoptic-scale processes modulate observed O<sub>3</sub> variability at scale of global model horizontal resolution



## Mean Afternoon Surface Ozone Background (ppbv) in GEOS-CHEM model, Summer 1995



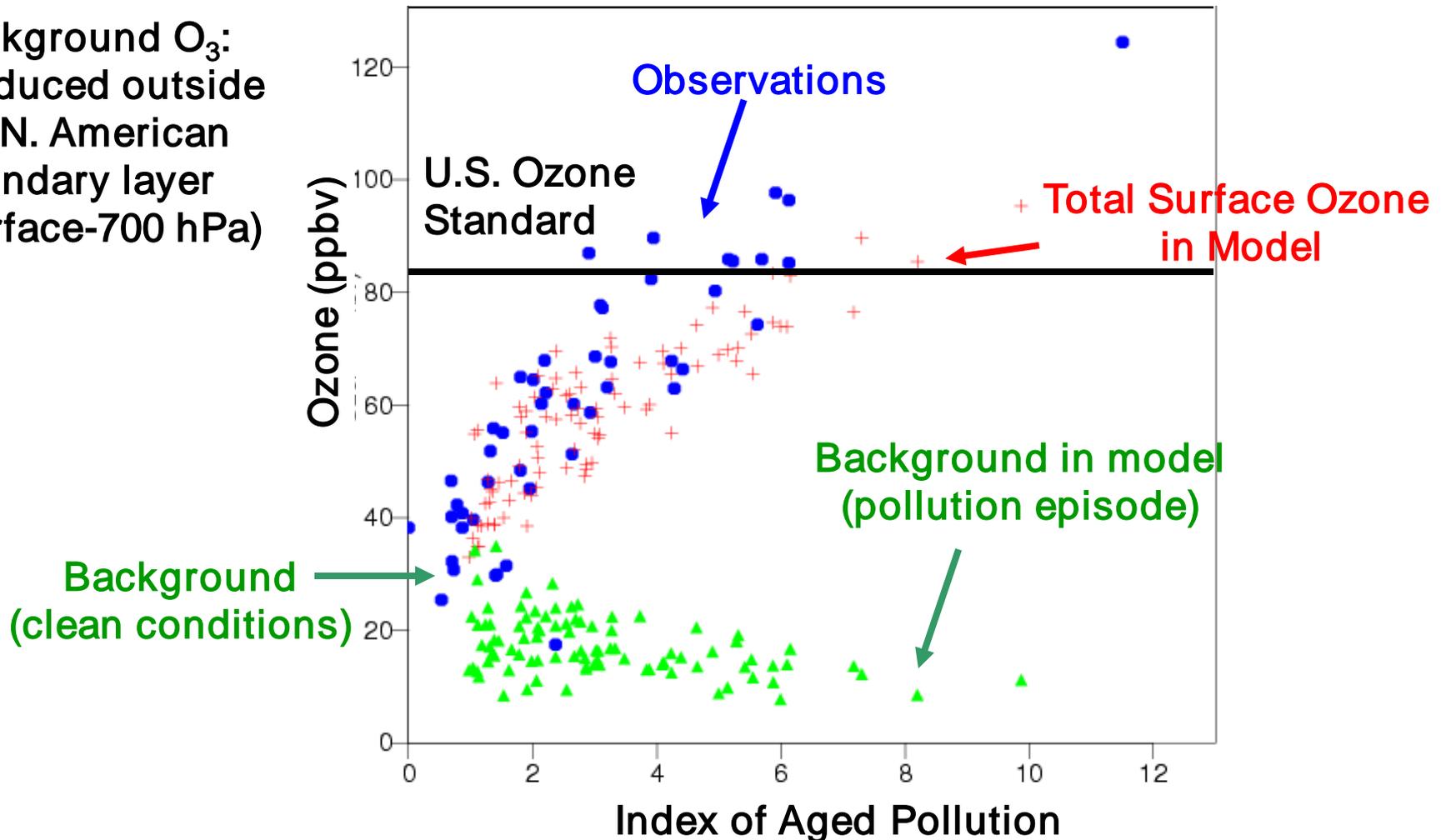
Background is tagged as ozone produced outside the N. American boundary layer (surface-700 hPa)

**What is the contribution of the background to pollution episodes?**

Ozone Background is depleted during regional pollution episodes  
(due to deposition and chemical loss under stagnant conditions)

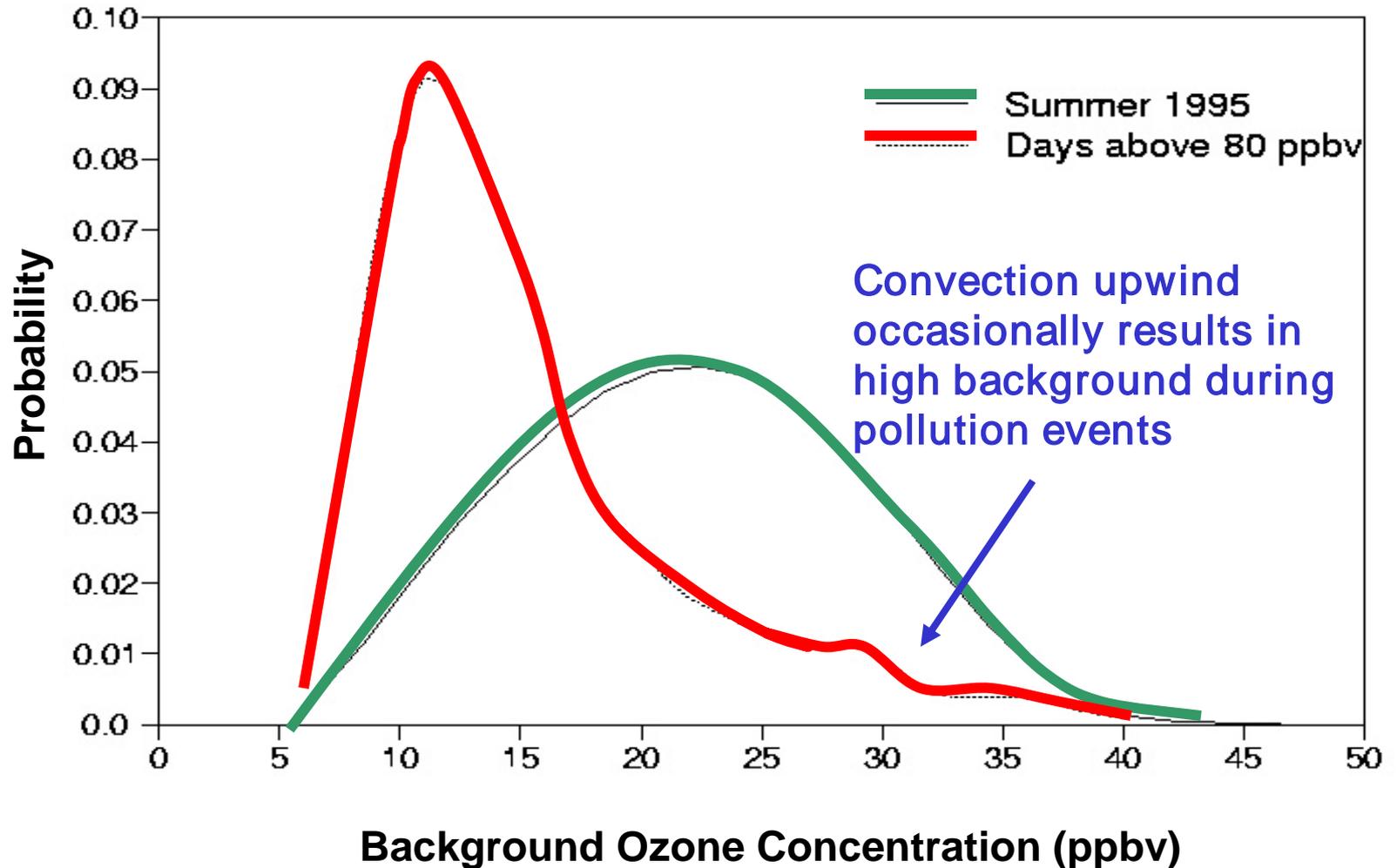
Daily mean afternoon  $O_3$  vs.  $(NO_y - NO_x)$  At Harvard Forest, MA

Background  $O_3$ :  
produced outside  
the N. American  
boundary layer  
(surface-700 hPa)



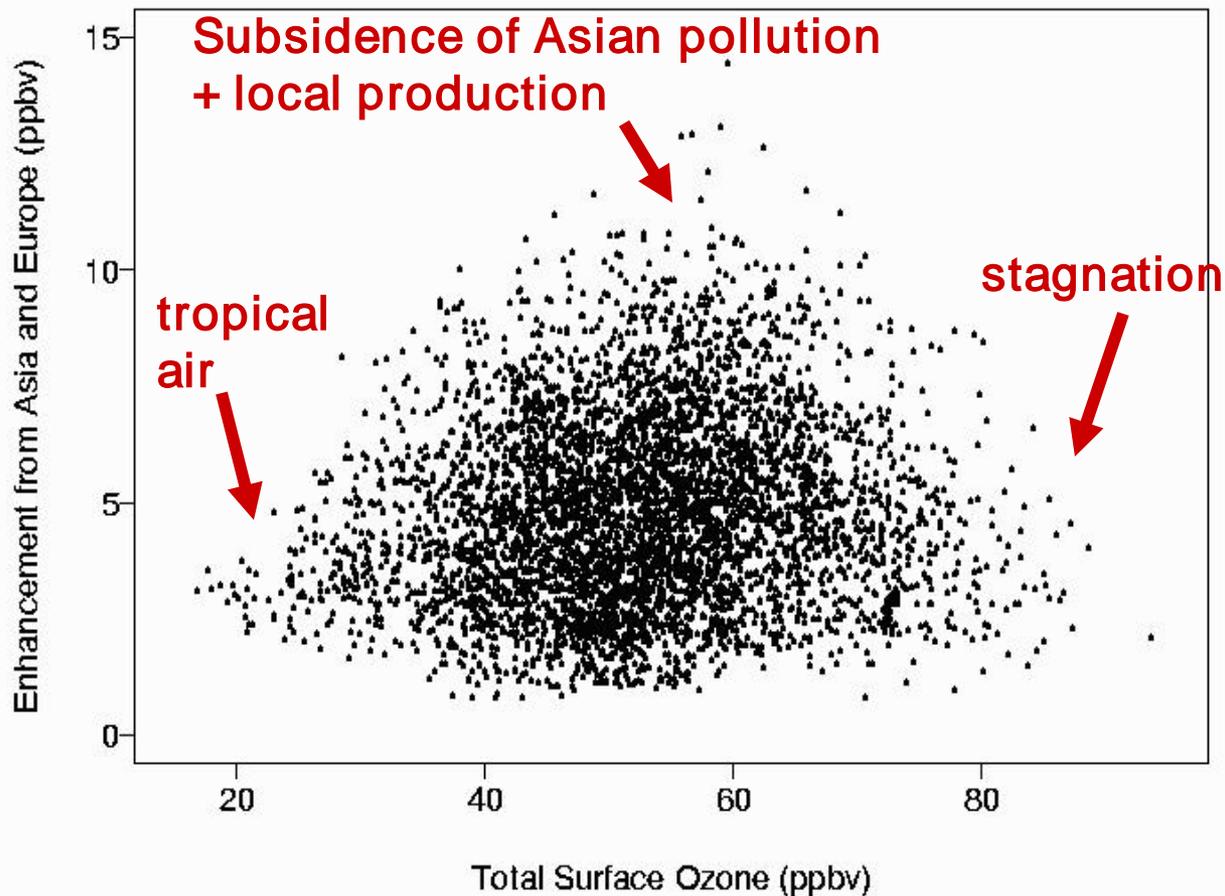
# Frequency Distribution of Afternoon Background Ozone Concentrations in U.S. Surface Air Summer 1995 (GEOS-CHEM model)

summer ensemble vs. pollution events



# Range of Asian/European Pollution Surface Ozone Enhancements Over the U.S. in summer

as determined from a simulation without these emissions ( $4^{\circ}\times 5^{\circ}$ )



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

**MAJOR CONCERN  
IF OZONE STANDARD  
WERE TO DECREASE**

# Sensitivity Simulations for source attribution

## Mar-Oct 2001

Note: Background in the following results is as defined by EPA

- **Standard simulation**.....2x2.5 GEOS-CHEM, 48 sigma levels  
2001
- **Background**.....no anthrop. NO<sub>x</sub>, CO, NMVOC  
emissions from N. America
- **Natural O<sub>3</sub> level**.....no anthrop. NO<sub>x</sub>, CO, NMVOC  
emissions globally; CH<sub>4</sub> = 700 ppbv
- **Stratospheric**.....tagged O<sub>3</sub> tracer simulation

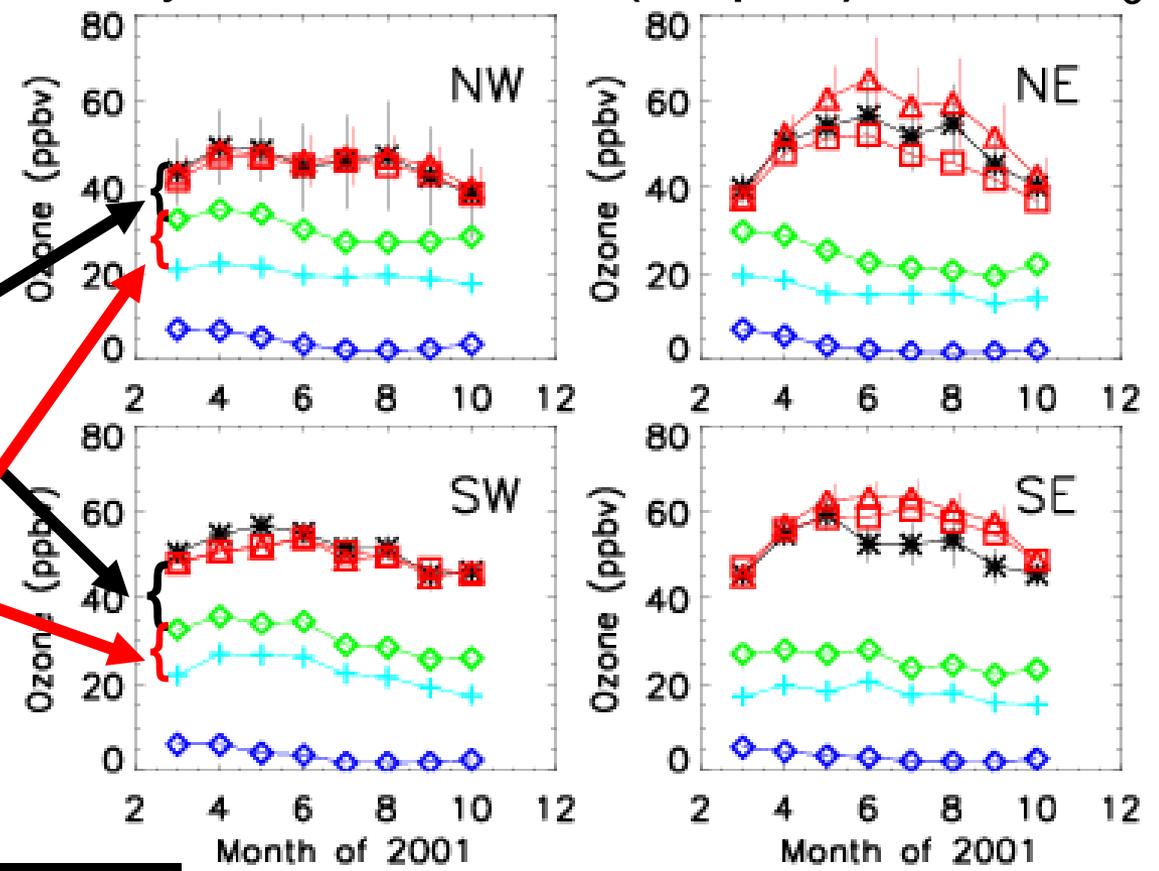
Regional Pollution = **Standard** – **Background**

Hemispheric Pollution = **Background** – **Natural O<sub>3</sub> level**

Use 2001 CASTNet data in conjunction with GEOS-CHEM to investigate how background O<sub>3</sub> varies with season and region

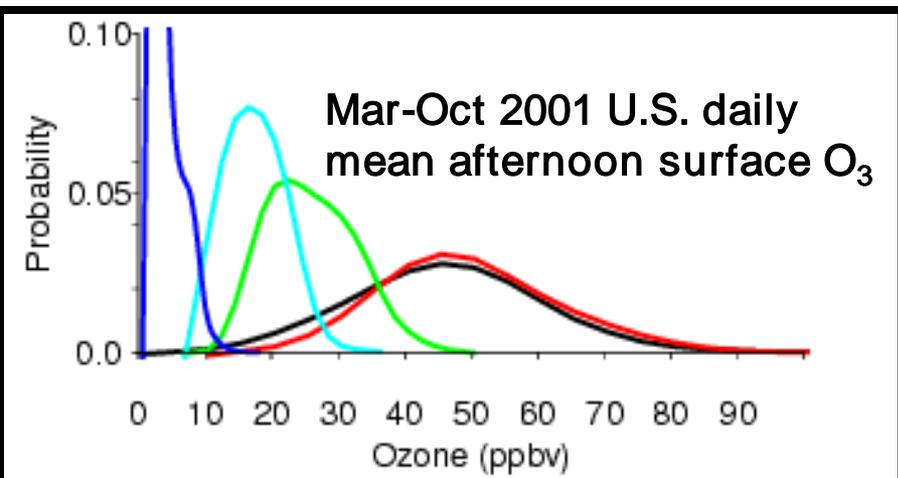
- \* CASTNet sites
- ▲ Model at CASTNet
- Model entire region
- ◆ Background
- + Natural O<sub>3</sub> level
- ◆ Stratospheric

### Monthly mean afternoon (1-5 p.m.) surface O<sub>3</sub>



Regional pollution from N. Am. emis. (8-30 ppbv)

Hemispheric pollution enhancement (5-12 ppbv)



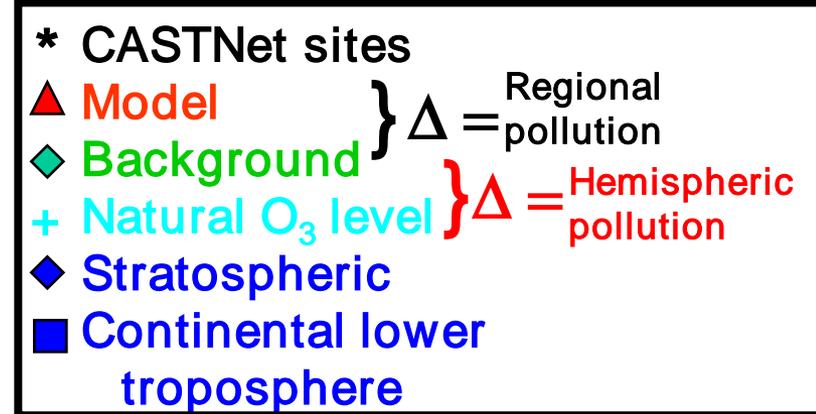
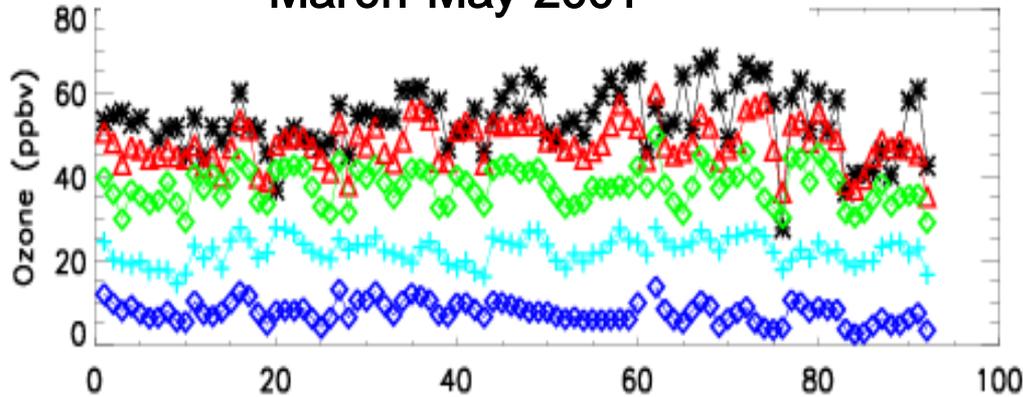
Mean background: 20-35 ppbv  
 Mean natural level: 13-27 ppbv  
 Mean stratosphere: 2-7 ppbv



Background < 50 ppbv  
 Natural level < 40 ppbv  
 Stratosphere < 20 ppbv

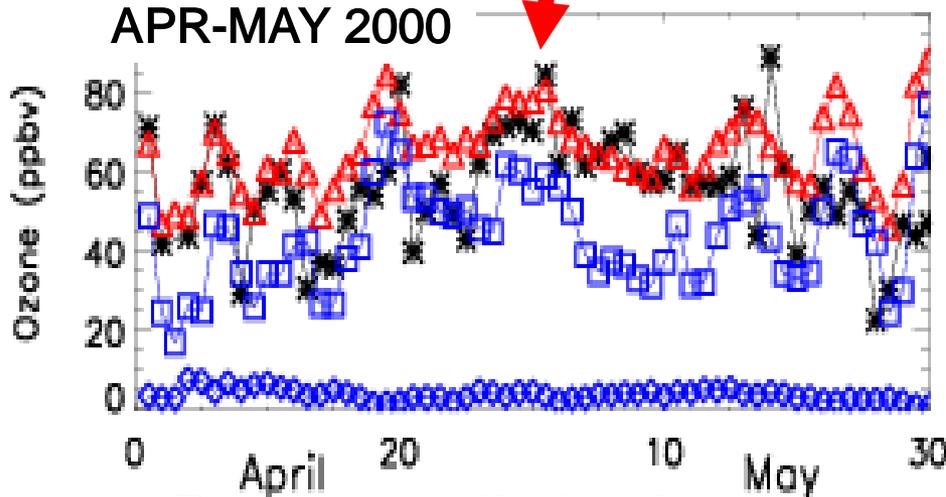
Time series at CASTNet sites where high-O<sub>3</sub> events in spring were previously attributed to stratospheric origin on basis of back-trajectories

Yellowstone NP, Wyoming  
March-May 2001

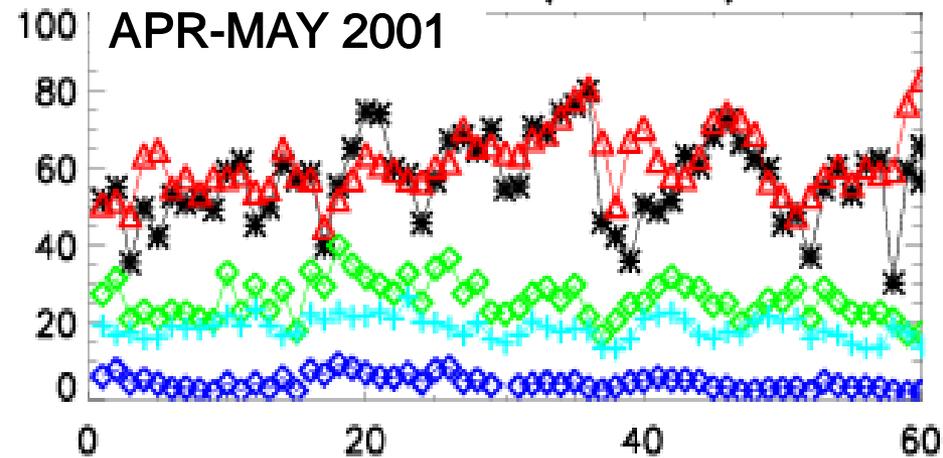


High-O<sub>3</sub> "Haywood County" event in North Carolina

APR-MAY 2000



APR-MAY 2001

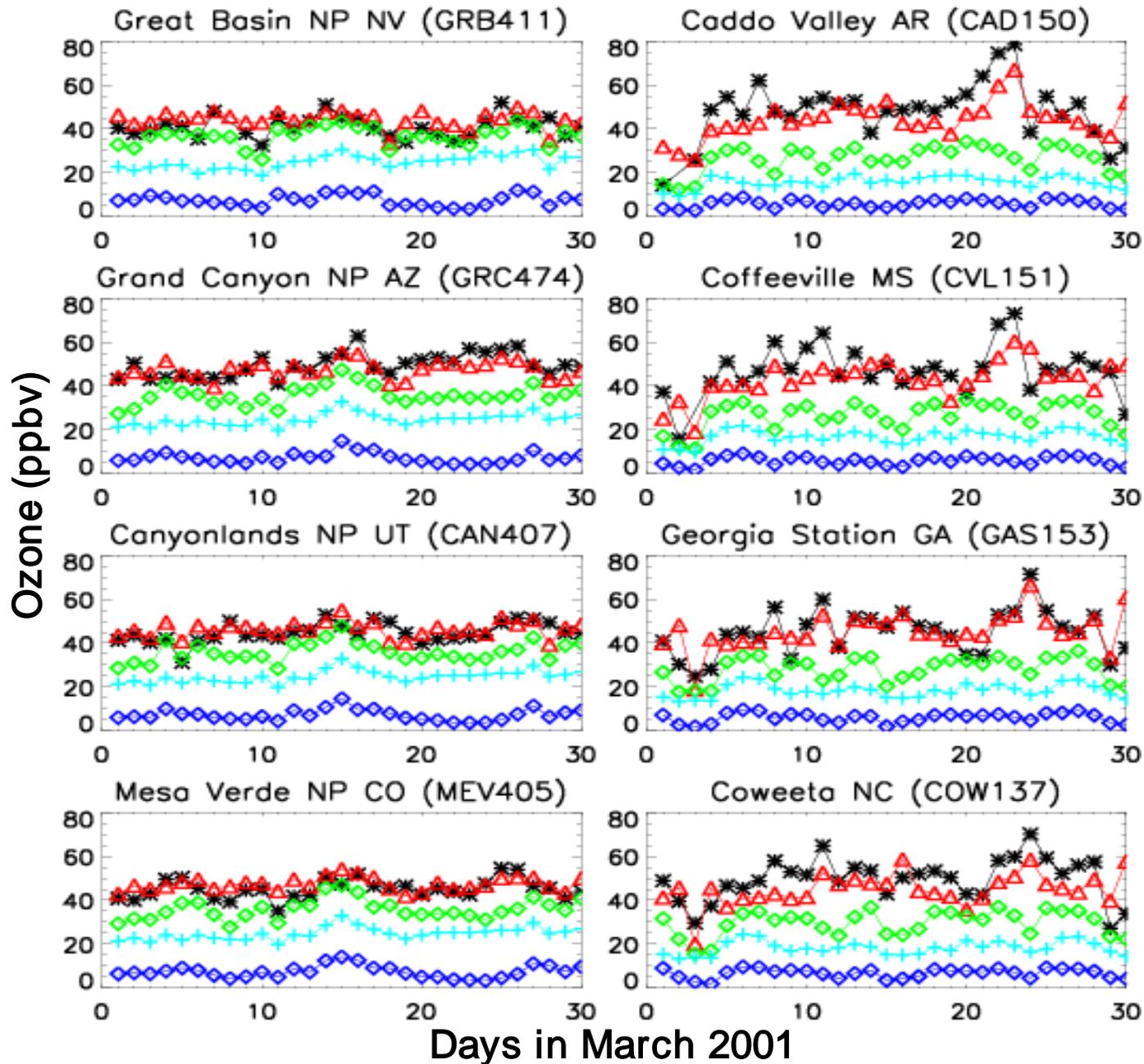


Regional pollution largely controls high-O<sub>3</sub> events in spring;  
 Model explains these events without a large stratospheric influence

- \* CASTNet sites
- ▲ Model
- ◆ Background
- + Natural O<sub>3</sub> level
- ◆ Stratospheric

## West

## Southeast



Background increases with highest observed O<sub>3</sub> at western sites in March

Background decreases with highest observed O<sub>3</sub> at SE sites in March

Days in March 2001

- \* CASTNet sites
- ▲ Model
- ◆ Background
- + Natural O<sub>3</sub> level
- ◆ Stratospheric

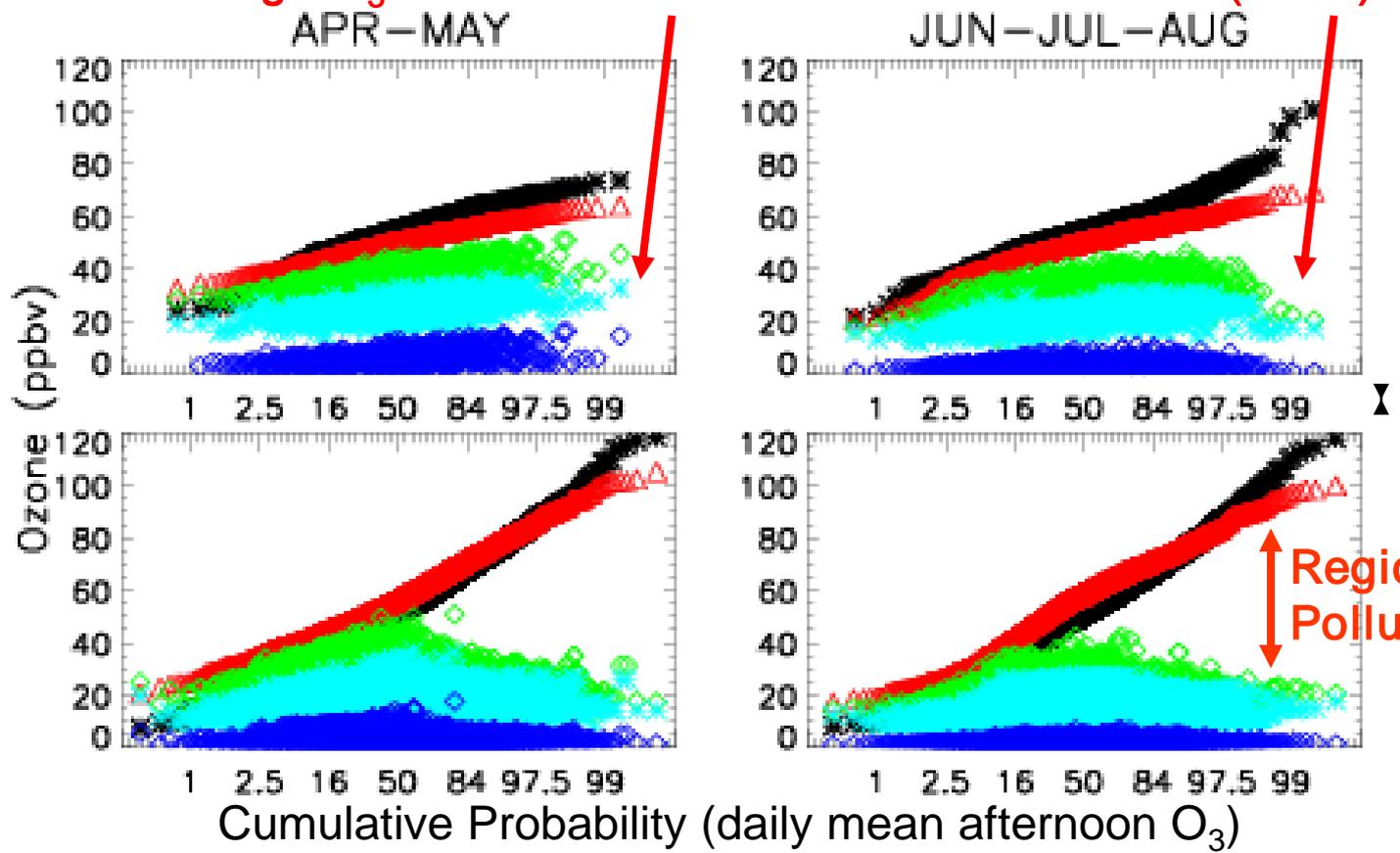
## Background O<sub>3</sub> for risk assessment = f (season, altitude, total O<sub>3</sub> concentration)

Enhancement from N. Amer & hemis. pollution for high O<sub>3</sub> concentrations

Lower background; larger pollution influence in summer (& fall)

12 elevated sites; all in west

58 surface sites

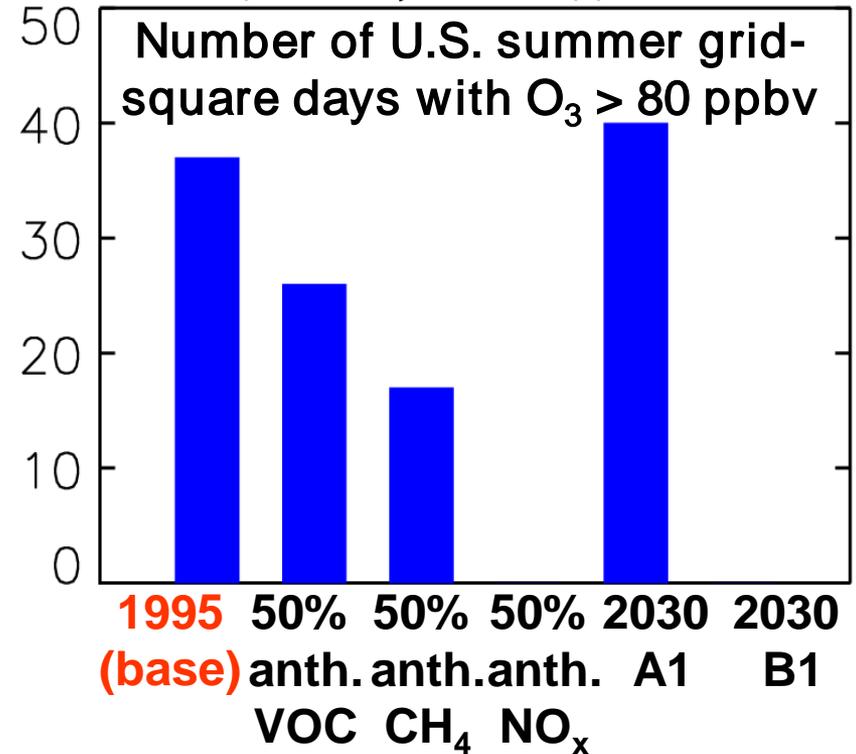
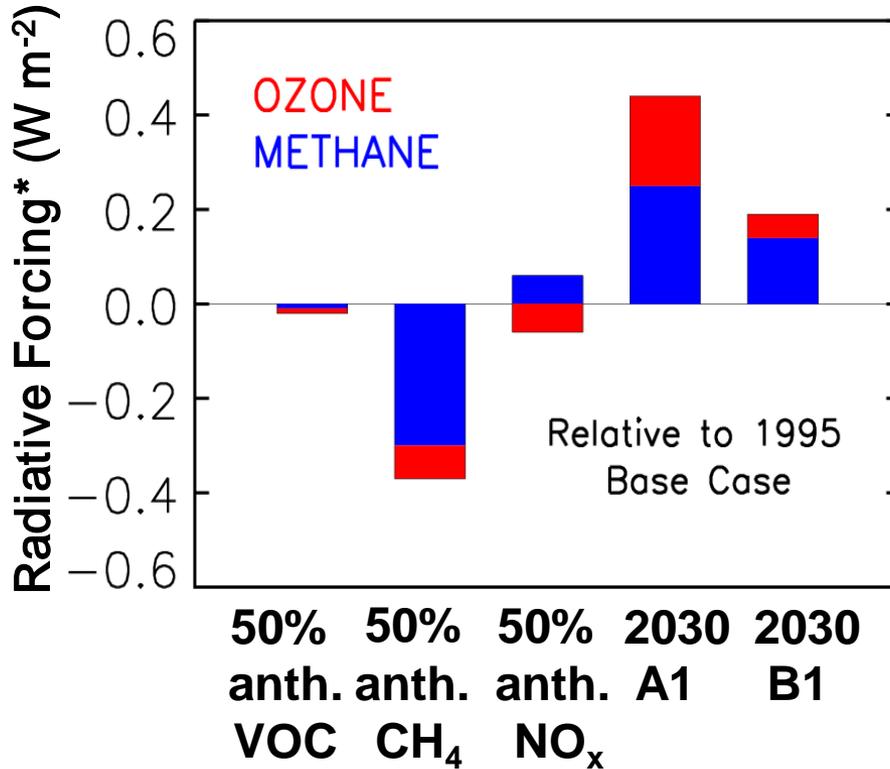


Lower background at surface sites; Maximum contribution at the center of the O<sub>3</sub> distribution



Using average background for pollution episodes underestimates risk to human health!

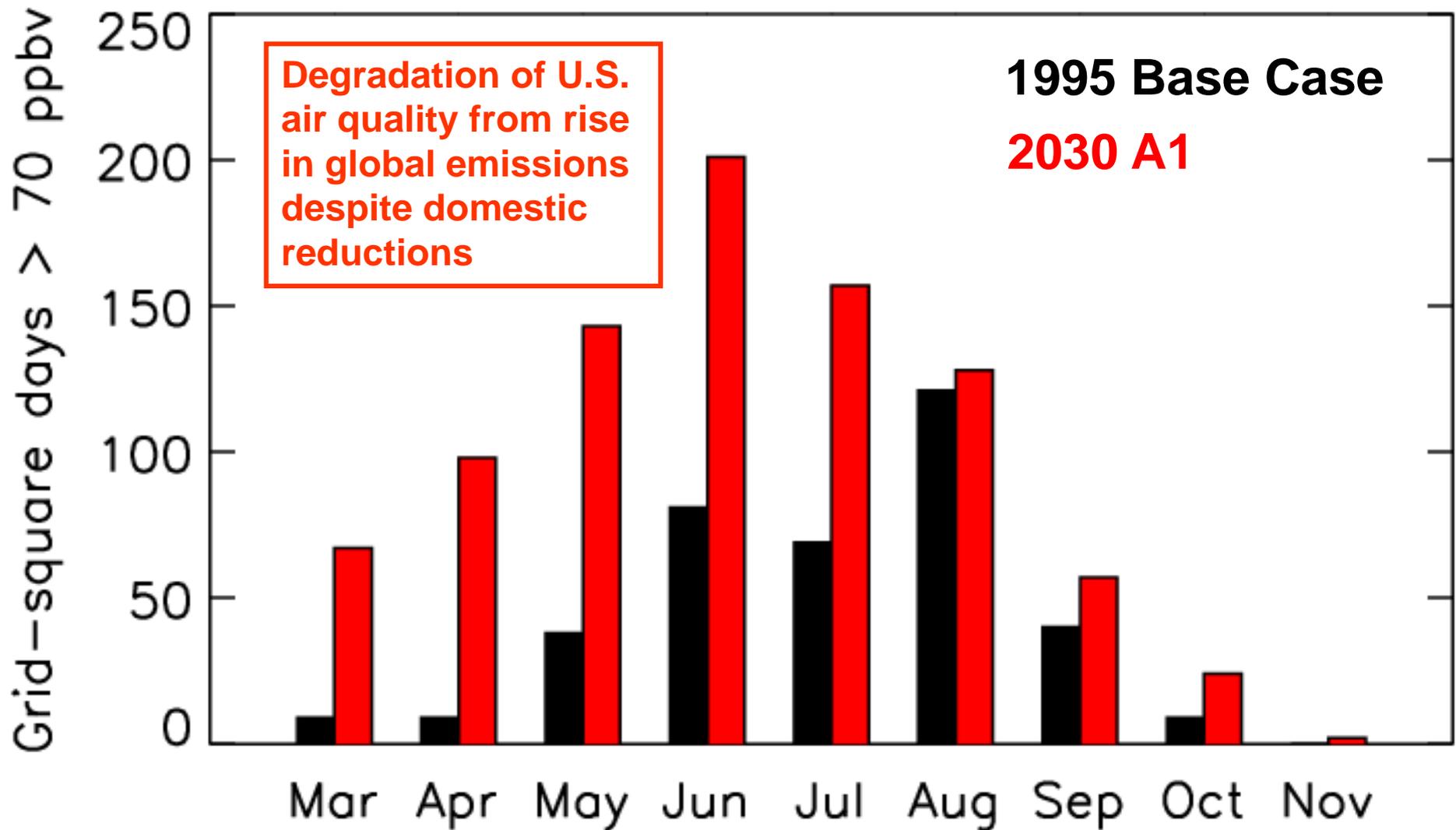
# Air Quality-Climate Linkage: Impacts of future changes in global anthropogenic emissions (GEOS-CHEM; 4 x5 )



IPCC scenario	Anthrop. NO <sub>x</sub> emissions (2030 vs. present)		Methane emissions (2030 vs. present)
	Global	U.S.	
A1	+80%	-20%	+30%
B1	-5%	-50%	+12%

CH<sub>4</sub> links air quality & climate via background O<sub>3</sub>

## Rising emissions from developing countries lengthen the O<sub>3</sub> pollution season in the United States



## CONCLUSIONS and their implications for policy

1. Global models can adequately simulate the synoptic conditions important for resolving background and high-O<sub>3</sub> conditions
2. Background O<sub>3</sub> varies with season, site elevation, and total surface O<sub>3</sub> concentrations
  - highest at high-altitude western U.S. sites in spring
  - lower at surface sites and in summer
  - depleted during polluted conditions

→ health risk from O<sub>3</sub> underestimated in present EPA risk assessments
3. High-O<sub>3</sub> events at remote U.S. sites in spring previously attributed to a natural stratospheric source are explained largely by regional pollution
  - these events should not be used to challenge legitimacy of O<sub>3</sub> NAAQS
4. Hemispheric pollution enhances U.S. background; may rise in future
  - international agreements to reduce hemispheric background would improve U.S. air quality & facilitate compliance w/ more stringent standards
5. Methane emission controls decrease hemispheric background & greenhouse warming
  - co-benefits for air quality & climate change mitigation objectives