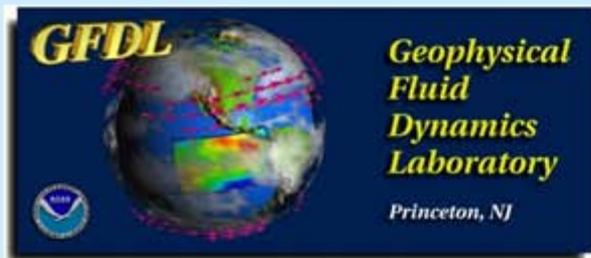


# Recent Changes in Eastern U.S. Forests: Implications for Air Quality

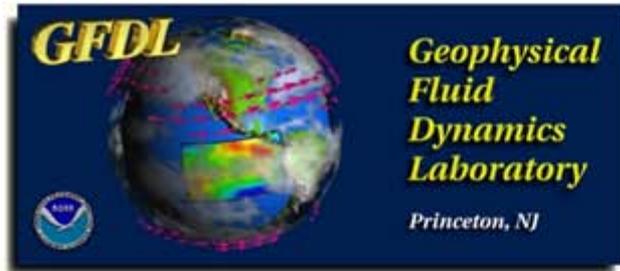
**Arlene M. Fiore**

**Seminar at the  
Center for Sustainability and the Global Environment (SAGE)  
University of Wisconsin-Madison**

**December 8, 2004**



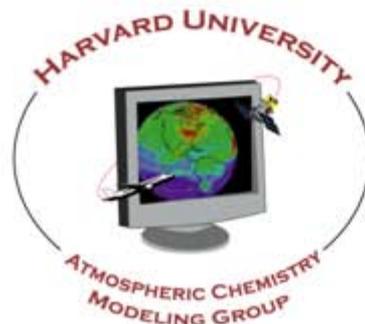
# Acknowledgments



Larry Horowitz  
Chip Levy

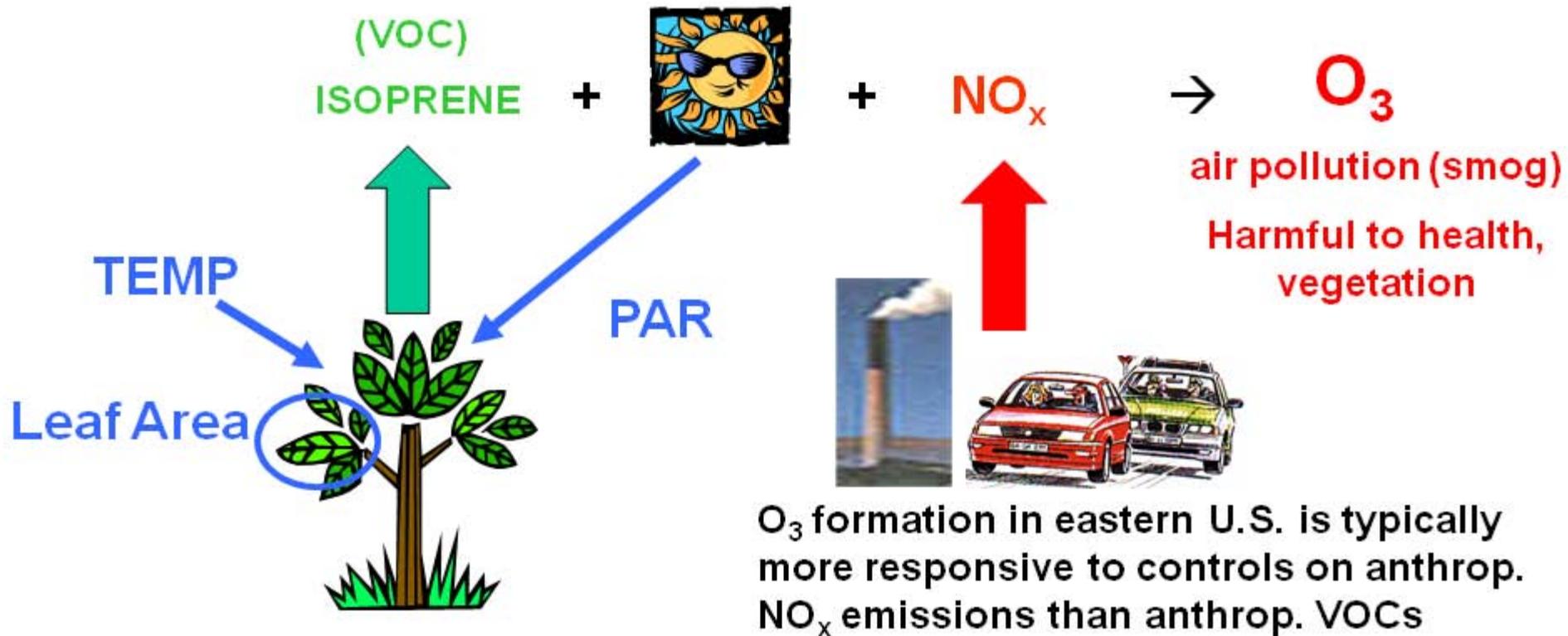


Drew Purves  
Steve Pacala



Mat Evans  
Qinbin Li  
Bob Yantosca  
Yuxuan Wang

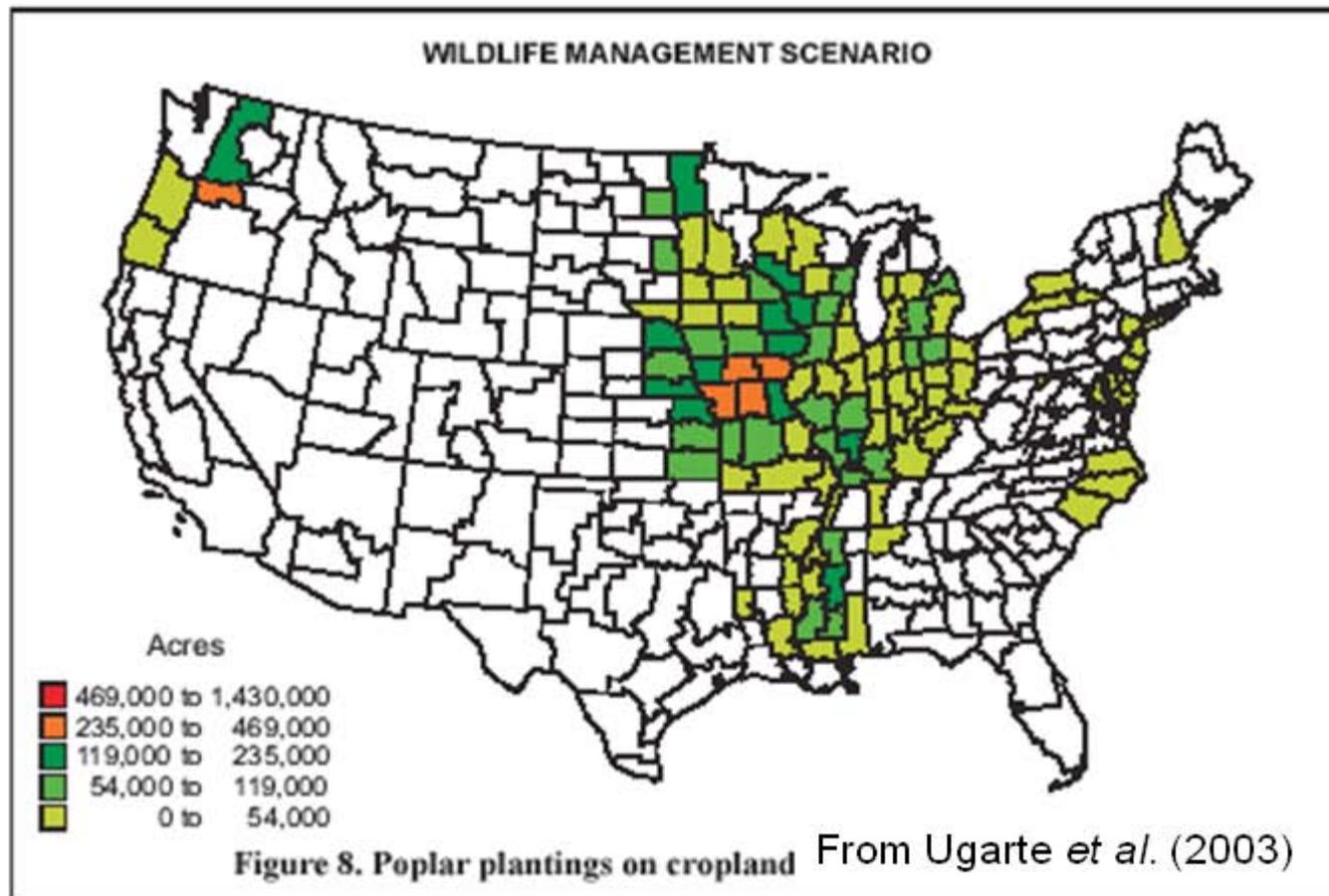
**Isoprene Emissions are generally thought to contribute to  $O_3$  production over the eastern United States [e.g. Trainer et al., 1987; NRC 1991]**



Vegetation changes → Impact on  $O_3$ ?

- Climate
- Land-use

## Future scenario for conversion of agricultural lands to poplar plantations for biofuels



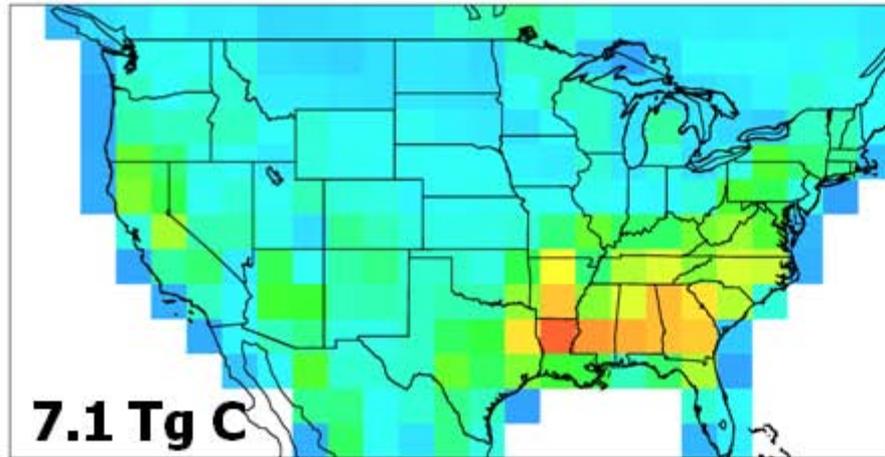
**Poplars are high-isoprene emitting trees**  
→ **land-use decisions have implications for air quality**

*from D. Purves*

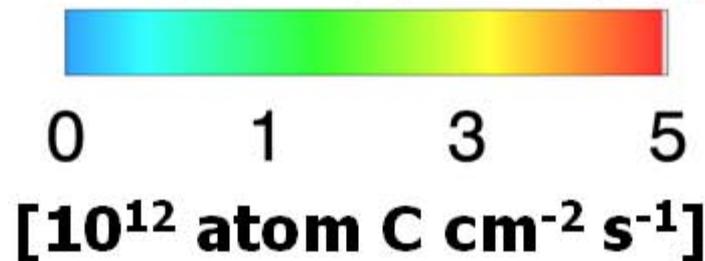
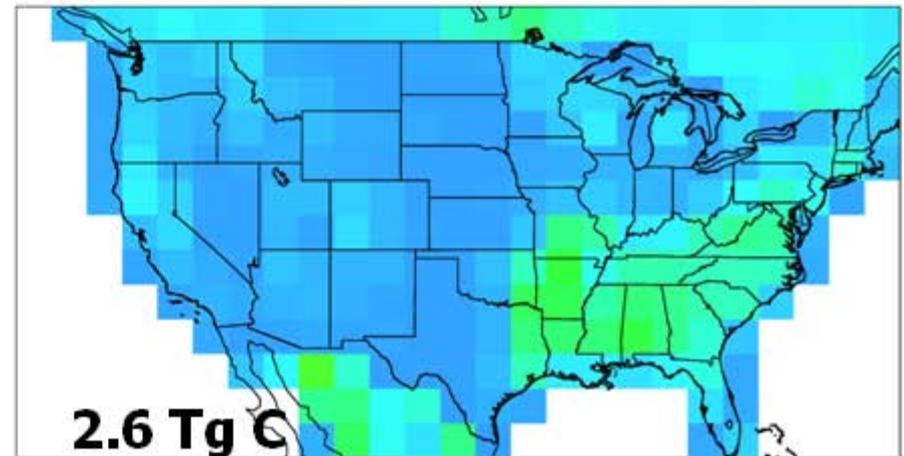
# Isoprene Emission Inventories uncertain by at least a factor of 2

## Isoprene emissions – July 1996

**GEIA: Global Isoprene  
Emission Inventory**



**BEIS2: Regional Isoprene  
Emission Inventory**



[from Paul Palmer]

... and there is new evidence for decadal changes



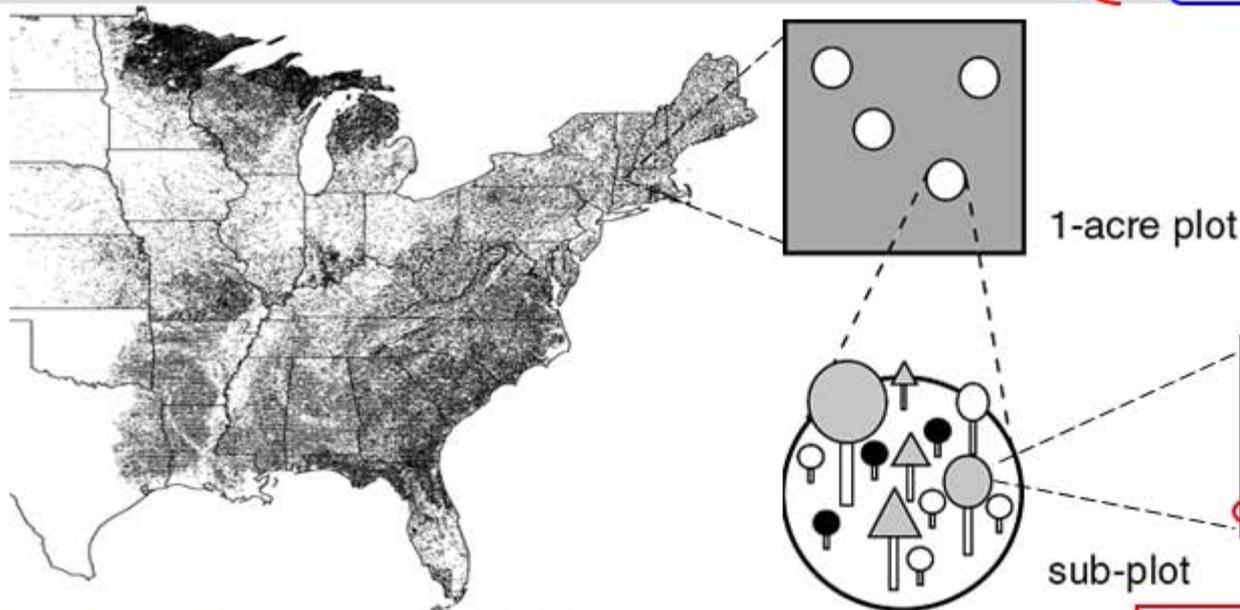
# Purves et al. [2004]: observation-based BVOC emission estimates for mid-1980s and mid-1990s

FIA data: ground surveys and aerial photography

Grid-cell

Plot

Tree



Choice of two different times

Species,  
Stem diameter (1985, 1995)  
Fate (lived, died, harvested)

'Natural' changes vs Logging

**280,000 Re-surveyed plots,  
2.7 million trees in FIA in eastern U.S.**

**→ Estimate recent changes in isoprene and monoterpene emissions**

**→ Purves Base isoprene inventory similar to BEIS2**

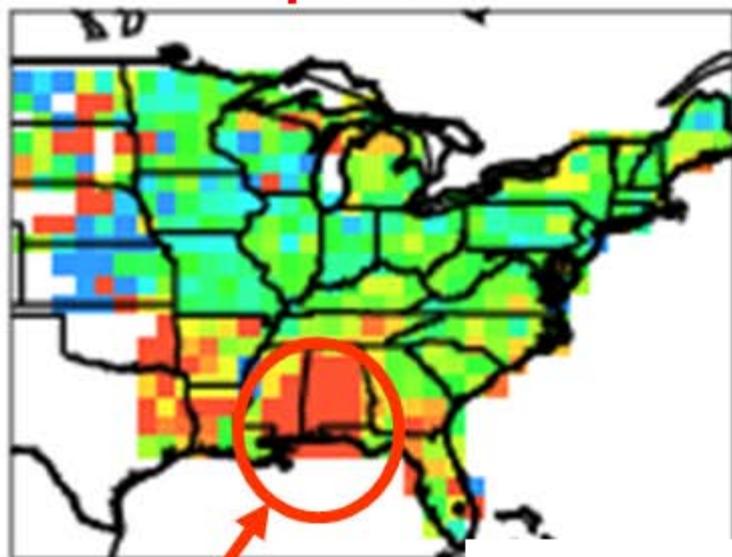
*from D. Purves*

## Recent Changes in Biogenic VOC Emissions

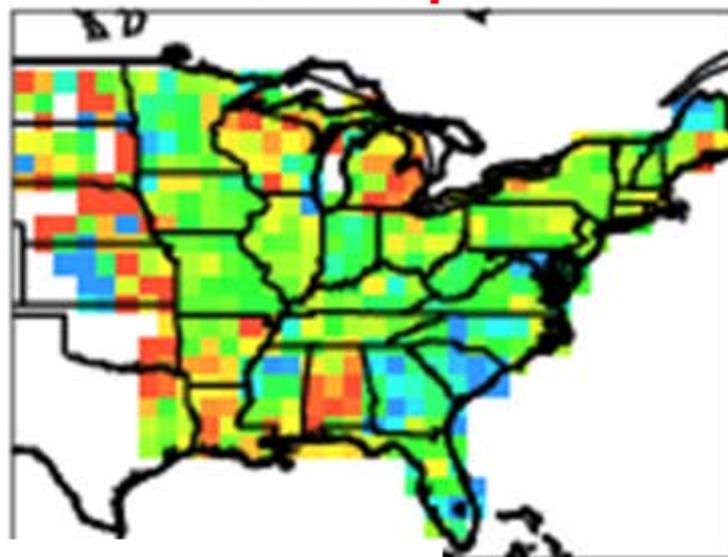
[Purves et al., *Global Change Biology*, 2004]

- Substantial isoprene increases in southeastern USA largely driven by human land-use decisions
- Land-use changes not presently considered in CTMs

Isoprene



Monoterpenes



-20 -10 0 +10 +20 +30

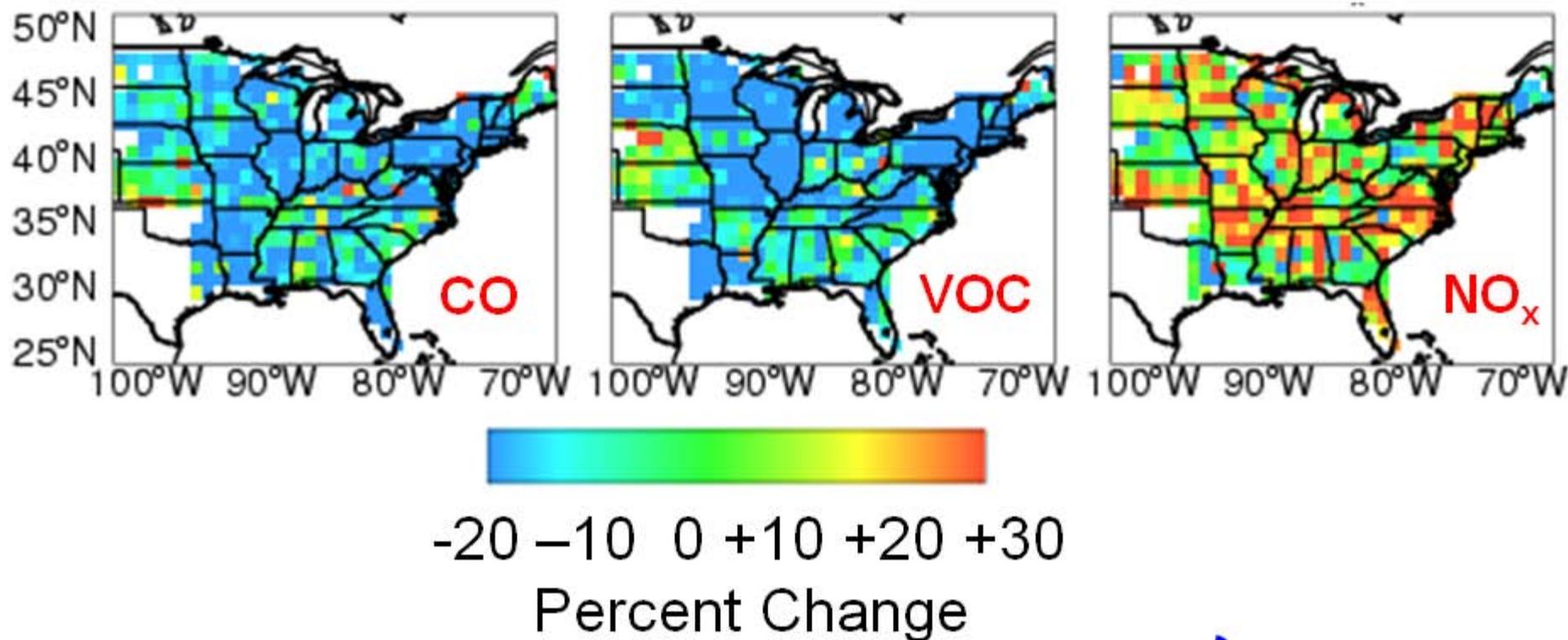
Percent Change mid-1980s to mid-1990s

Sweetgum  
Invasion of  
Pine plantations

Trends in anthropogenic precursors?

## Trends in Anthropogenic Emissions: 1985 to 1995

from US EPA national emissions inventory database  
(<http://www.epa.gov/air/data/neidb.html>)



- Large decreases in CO and VOC Emissions
- Some local increases in NO<sub>x</sub>
- Higher biogenic VOCs

Net effect  
On O<sub>3</sub>?

## Approach: Insights from two chemical transport models

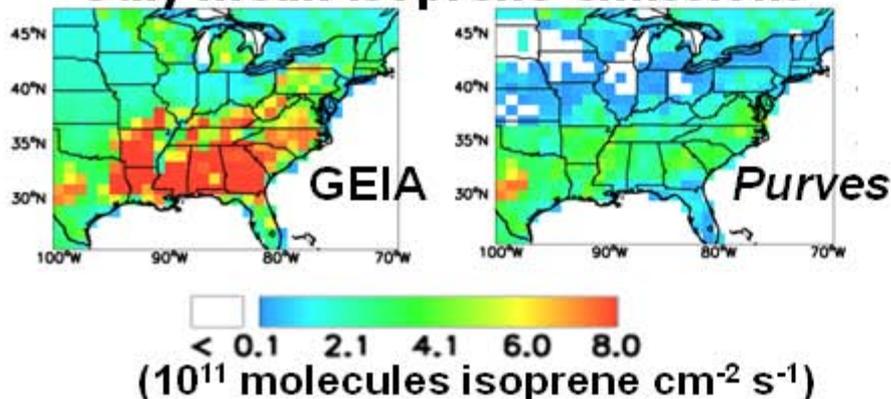


### GEOS-CHEM

**1. Quantify O<sub>3</sub> response to reported biogenic and anthropogenic emissions changes**

**2. Determine sensitivity to uncertainties in isoprene emissions**

#### July mean isoprene emissions



### MOZART-2

**1. Test whether results are model-dependent**

**2. Determine sensitivity to uncertainties in isoprene-NO<sub>x</sub>-O<sub>3</sub> chemistry**

→ Isoprene + NO<sub>x</sub> may lock up NO<sub>x</sub> (as isoprene nitrates), suppressing O<sub>3</sub> formation

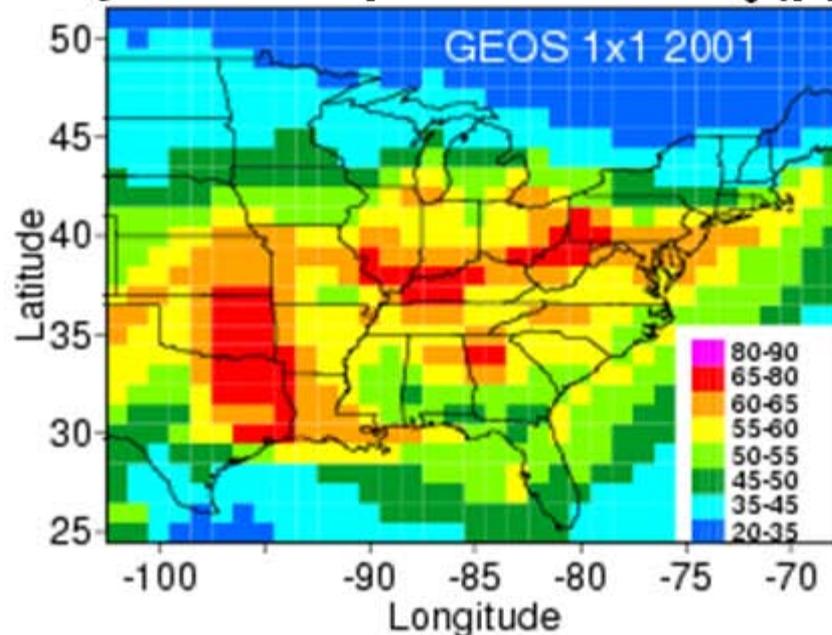
→ Assumed in GEOS-CHEM but not in MOZART-2

## Tool #1: GEOS-CHEM tropospheric chemistry model [*Bey et al., 2001*]

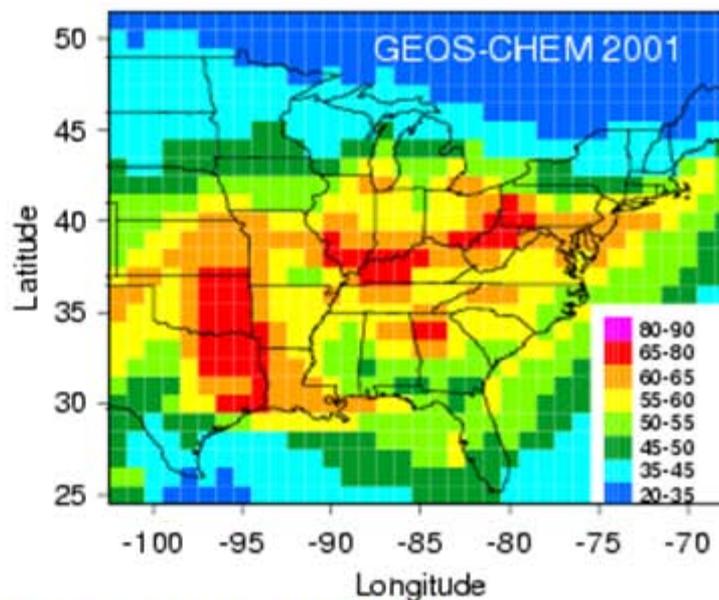
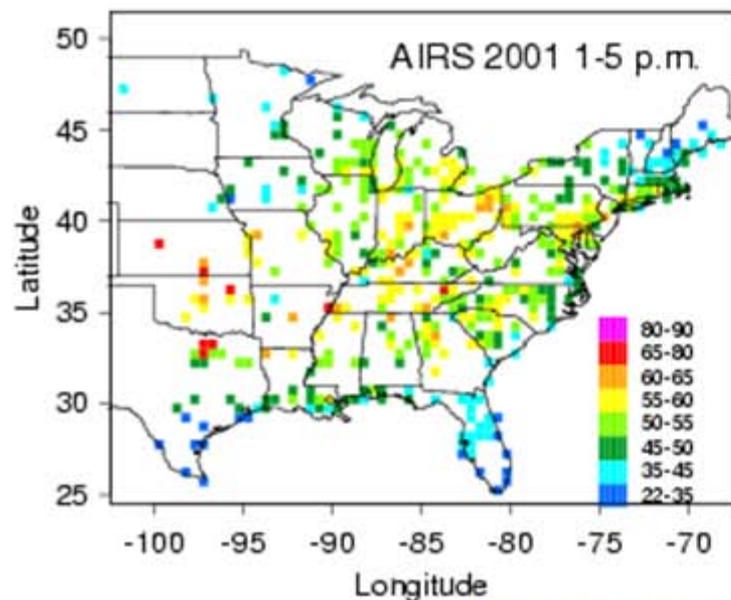


- Uses assimilated meteorology: GEOS-3  $1^\circ \times 1^\circ$  fields for 2001
- 48 vertical levels ( 9 below 2 km)
- RegridDED to  $4^\circ \times 5^\circ$  for global spinup and boundary conditions for **nested  $1^\circ \times 1^\circ$  over North America** [*Wang et al., 2004; Li et al., 2004*]
- 31 tracers;  $\text{NO}_x$ -CO-hydrocarbon- $\text{O}_3$  chemistry coupled to aerosols
- GEIA isoprene emission algorithms [*Guenther et al., 1995*]
- v. 5-07-08 (<http://www-as.harvard.edu/chemistry/trop/geos/index.html>)

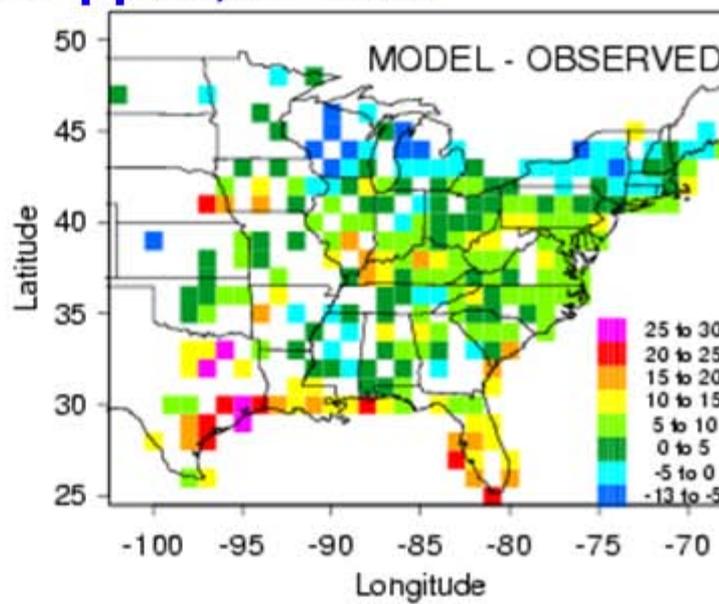
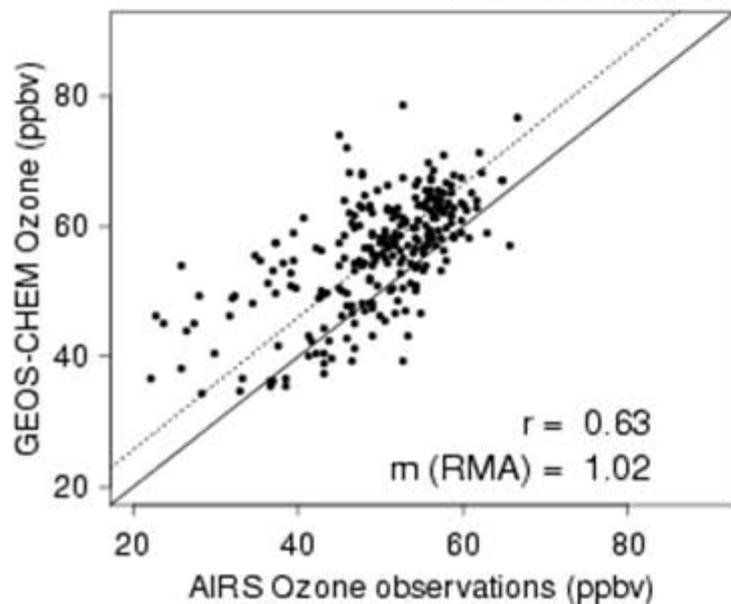
### July 2001 1-5 p.m. Surface $\text{O}_3$ (ppbv)



# GEOS-CHEM Evaluation: July 2001 1-5 p.m. Surface O<sub>3</sub> (ppbv)

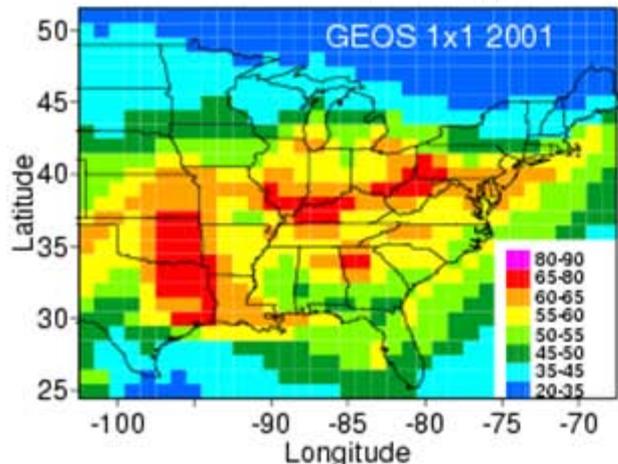


Mean Bias =  $6 \pm 7$  ppbv;  $r^2 = 0.40$



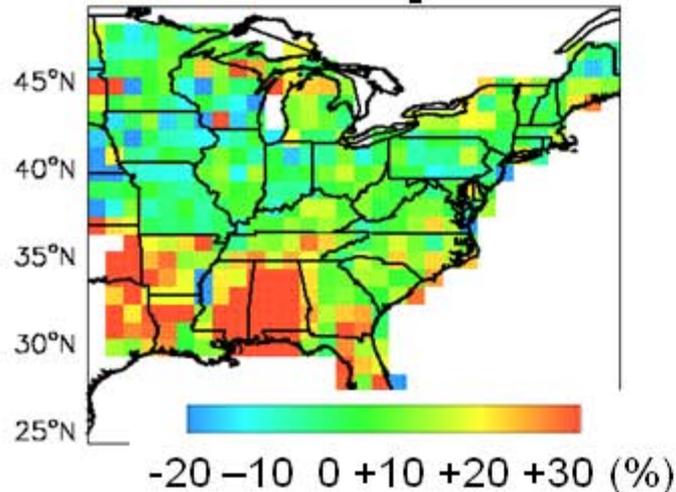
# Increases in isoprene emissions reduce surface O<sub>3</sub> in Southeastern US

GEOS-CHEM July 1-5 p.m. O<sub>3</sub>

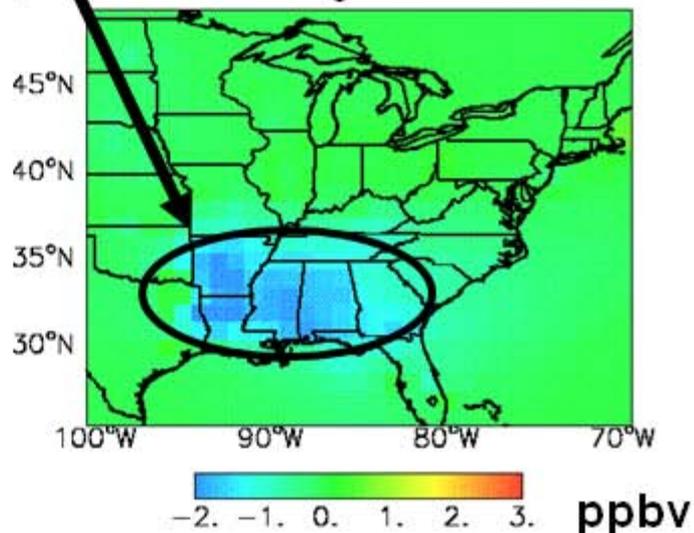


+

Isoprene emission changes from mid-80s to mid-90s [Purves et al., 2004]

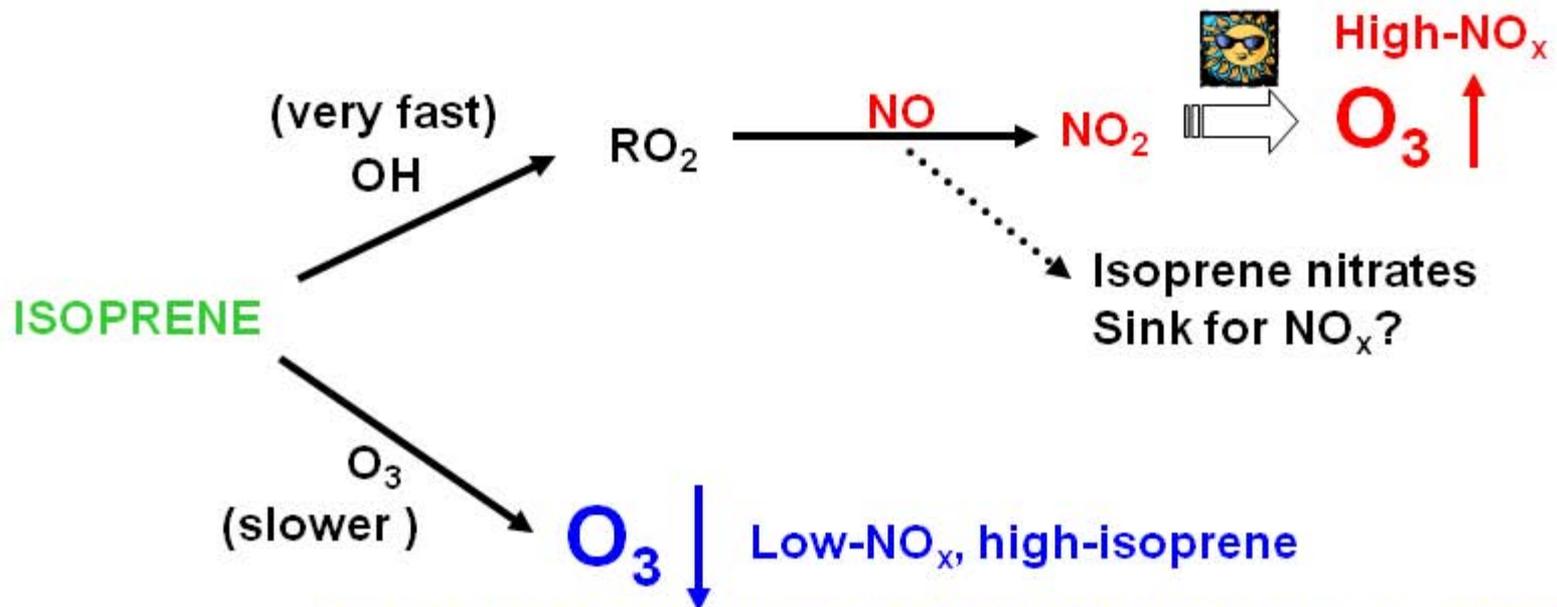
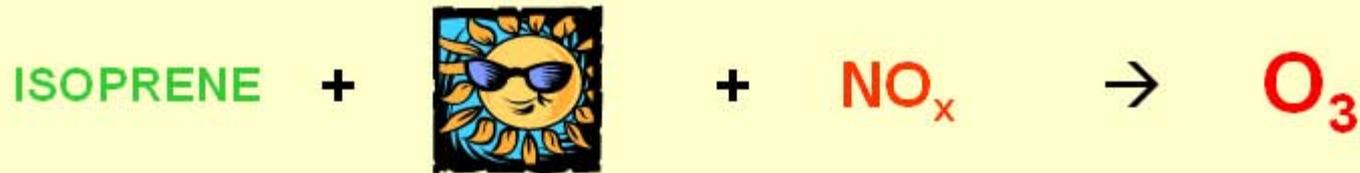


Change in July 1-5 p.m. surface O<sub>3</sub>



Complicated Chemistry...

# Complications to Chemistry: Isoprene may also decrease surface $O_3$ in low- $NO_x$ , high isoprene settings

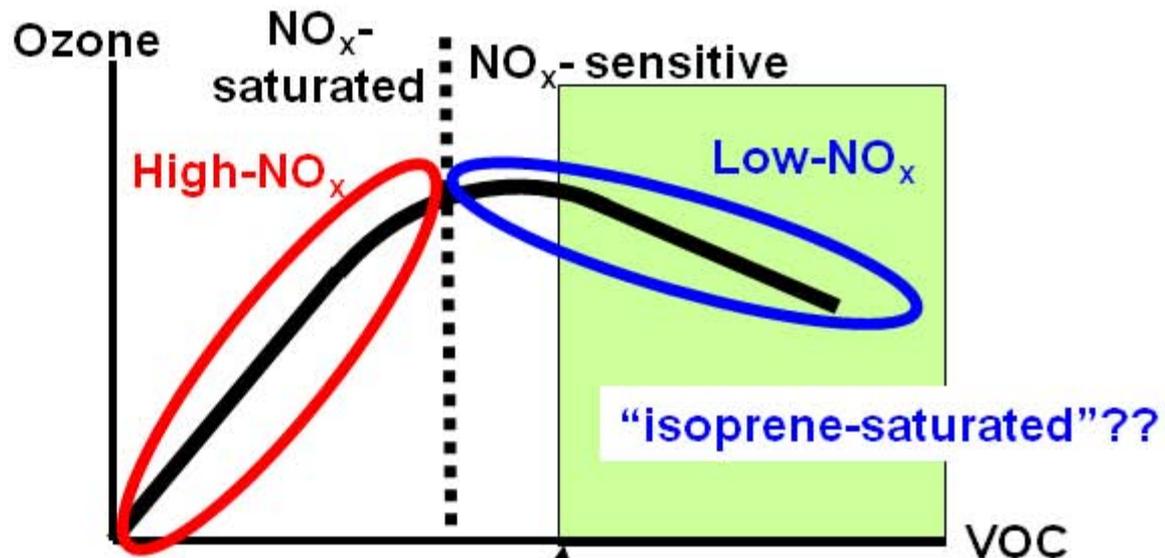
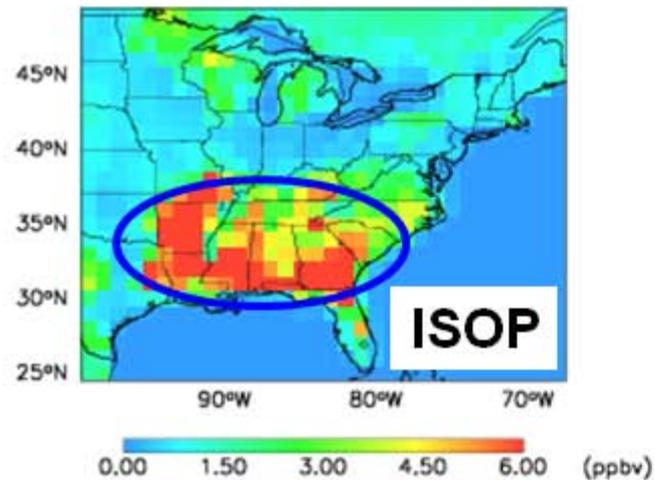
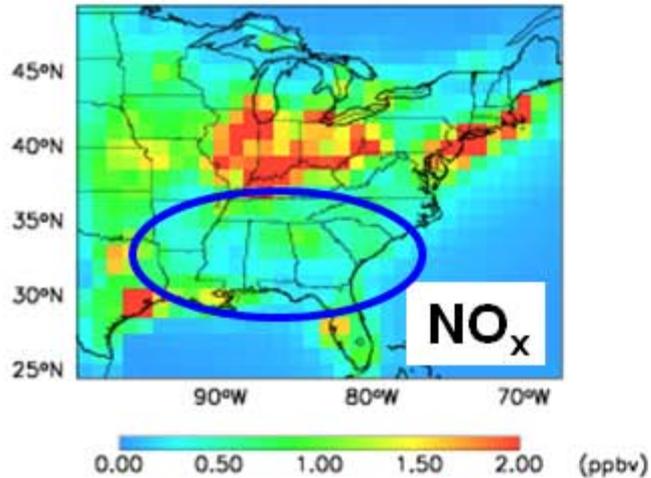


Thought to occur in pre-industrial [Mickley et al., 2001];  
and present-day tropical regions [von Kuhlmann et al., 2004]

Isoprene does react directly with  $O_3$  in our SE US GEIA simulation:  
 $O_3$ +biogenics (10d) comparable to  $O_3$ + $HO_x$  (16d),  $O_3$ + $h\nu$  → OH (11d)

# Increasing Isoprene Decreases O<sub>3</sub> in Low-NO<sub>x</sub>, High-isoprene regions

GEOS-CHEM base-case  
July 1-5 p.m. mean

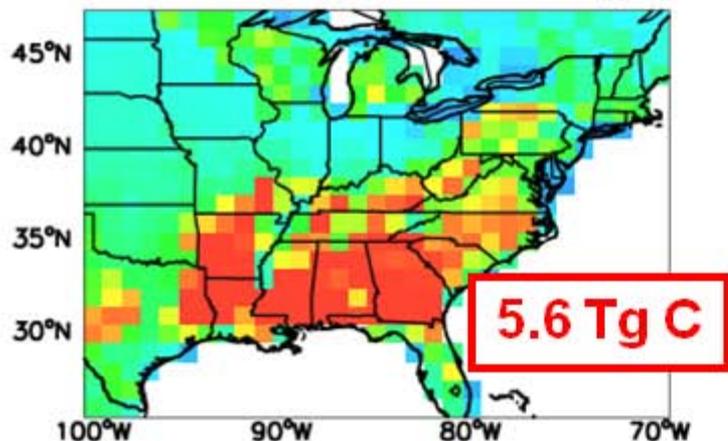


Other model studies indicate SE US is near "maximum VOC capacity point", beyond which VOCs suppress O<sub>3</sub> formation [Kang et al., 2003].

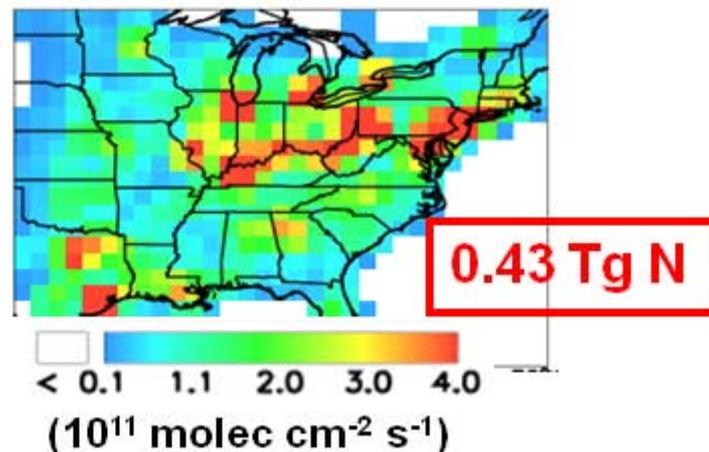
# Choice of isoprene inventory critical for predicting base-case O<sub>3</sub>

GEIA: global inventory

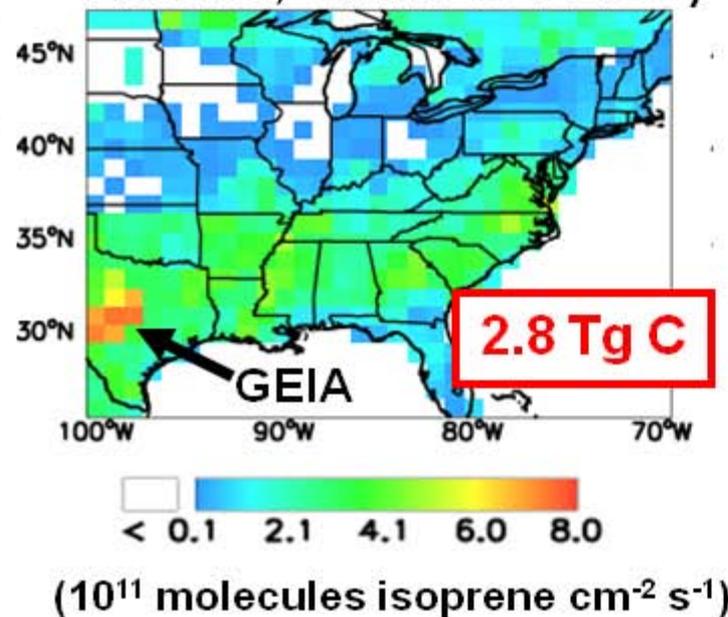
July isoprene emissions



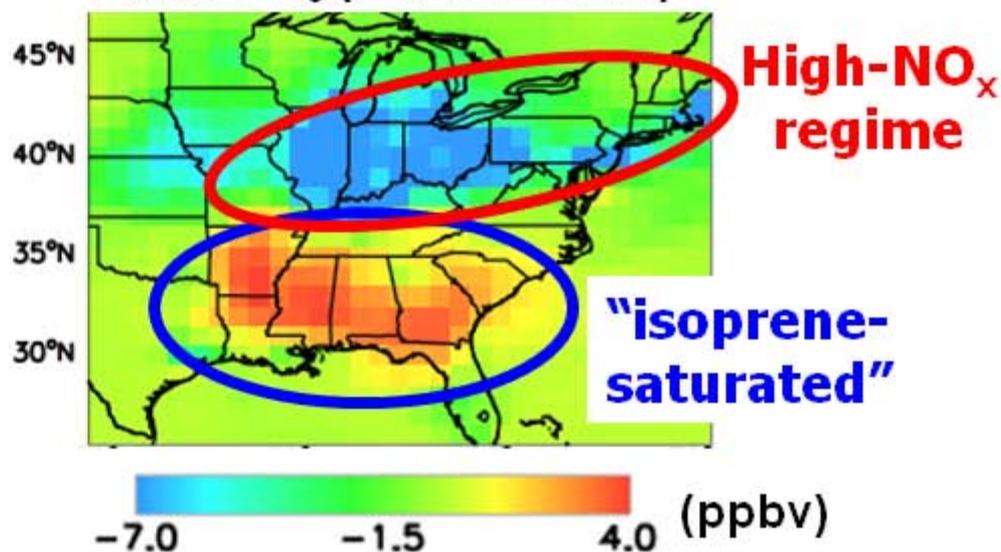
July Anthrop. NO<sub>x</sub> emissions



Purves et al., [2004] (based on FIA data; similar to BEIS-2)



Difference in July 1-5 p.m. surface O<sub>3</sub> (Purves-GEIA)

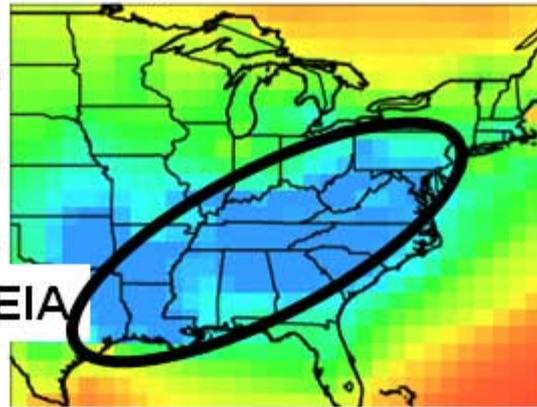
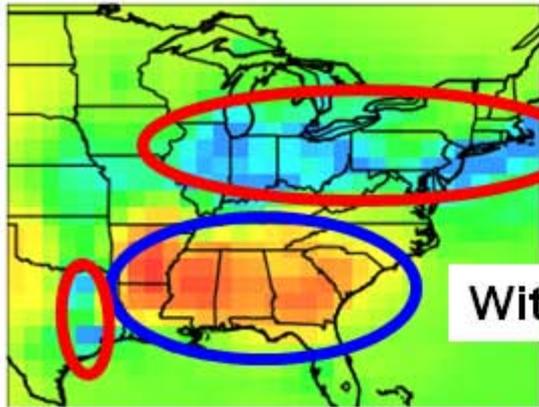


# Identify O<sub>3</sub> chemistry regime with precursor emissions reductions

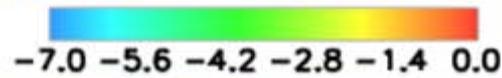
Change in July O<sub>3</sub> (ppbv; 1-5 p.m.)

Isoprene reduced 25%

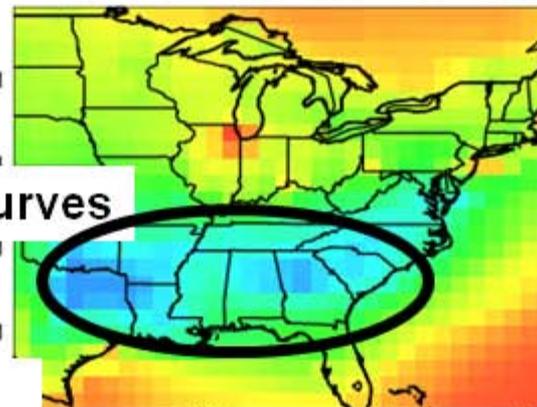
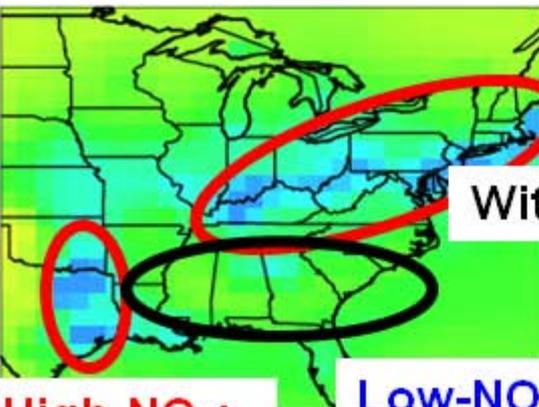
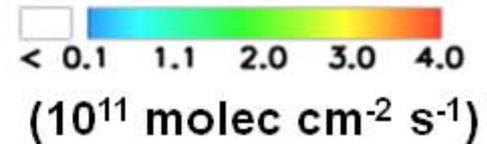
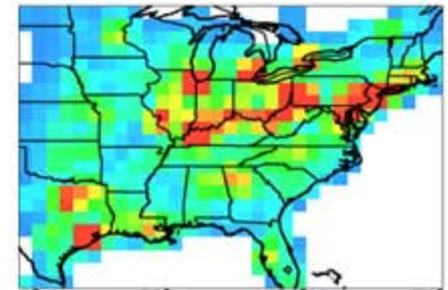
NO<sub>x</sub> reduced 25%



With GEIA



July Anthropogenic NO<sub>x</sub> Emissions



With Purves

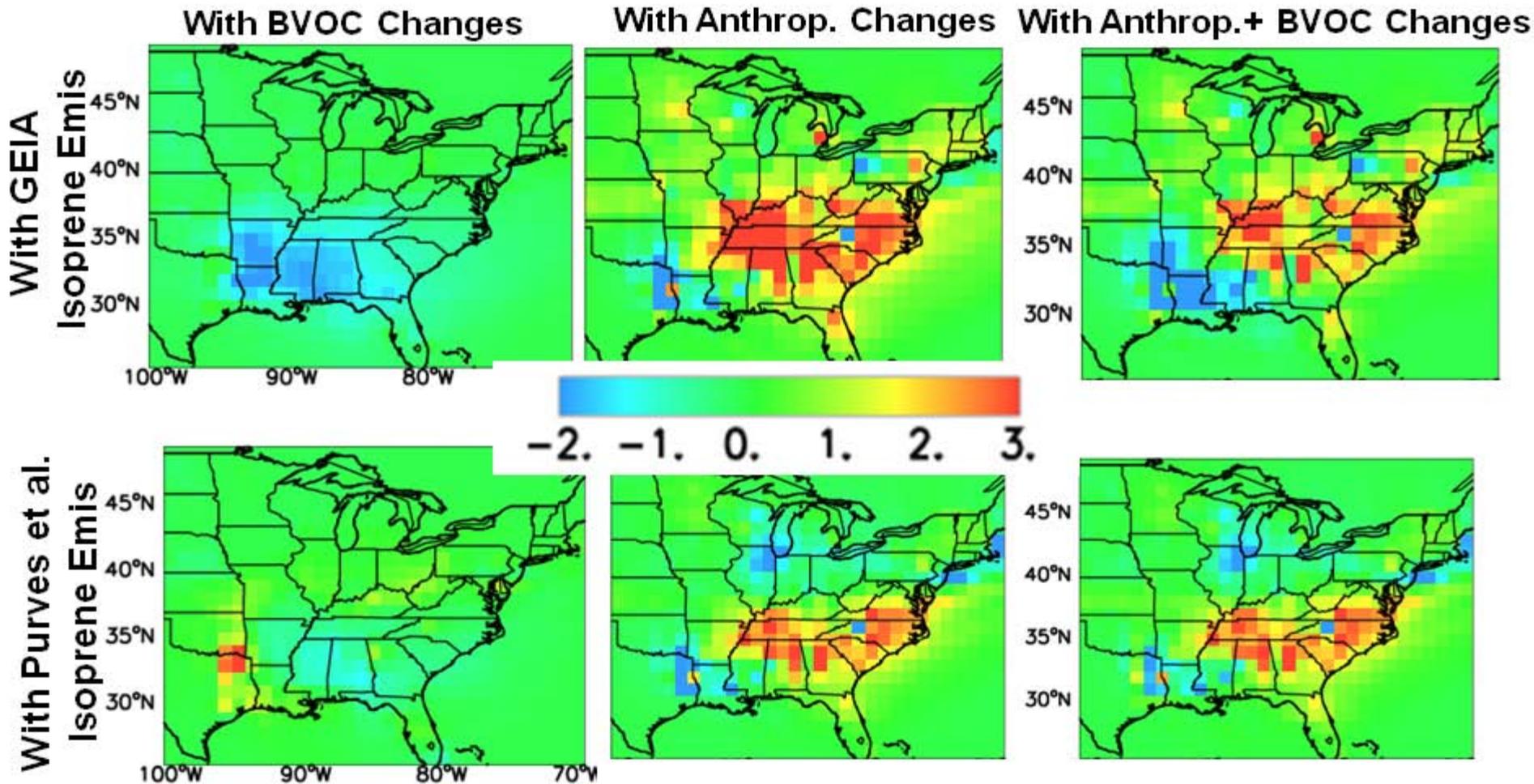
High-NO<sub>x</sub>:  
O<sub>3</sub> ↓ as isop ↓

Low-NO<sub>x</sub>,  
high isop:  
O<sub>3</sub> ↑ as isop ↓

strongly  
NO<sub>x</sub>-sensitive

**Choice of isoprene inventory also critical for predicting O<sub>3</sub> response to changes in isoprene and anthropogenic NO<sub>x</sub> emissions**

# Change in Mean July Surface O<sub>3</sub> (ppbv; 1-5 p.m.) reflecting 1980s to 1990s emissions changes



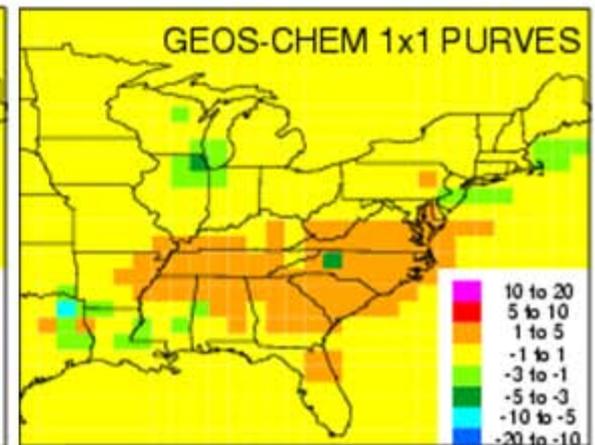
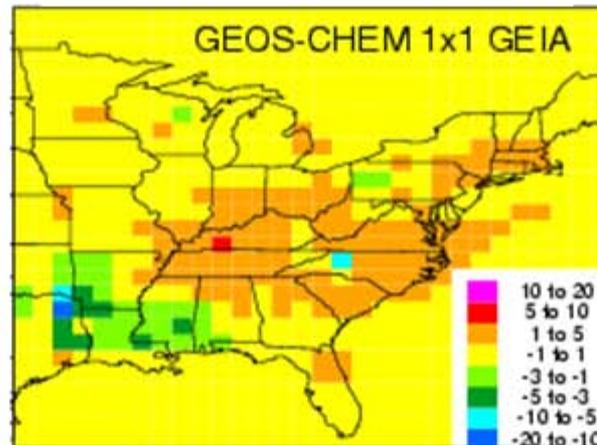
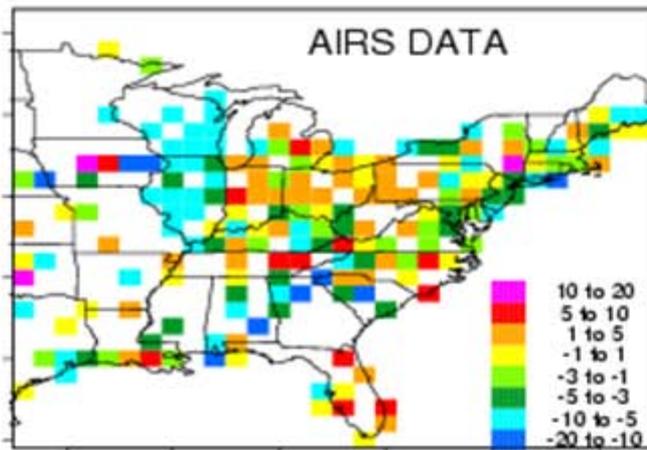
**Changes in Anthropogenic NO<sub>x</sub> emissions dominate O<sub>3</sub> response**  
**But response depends upon choice of isoprene emission inventory**  
**Comparison with observed changes? Impact on high-O<sub>3</sub> events?**

# Model vs. Obs.: Change in July O<sub>3</sub> 1980s to 1990s (ppbv; 1-5 p.m.)

## Obs: EPA AIRS

## GEOS-CHEM: GEIA

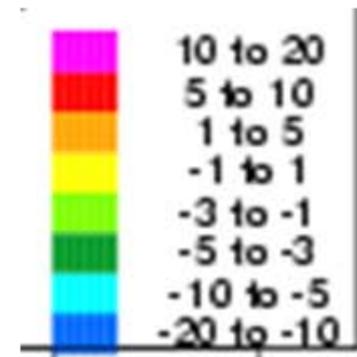
## GEOS-CHEM: Purves



(1993-1997) – (1983-1987)

Poor correlation ( $r^2 \sim 0$ ) between observed and simulated changes

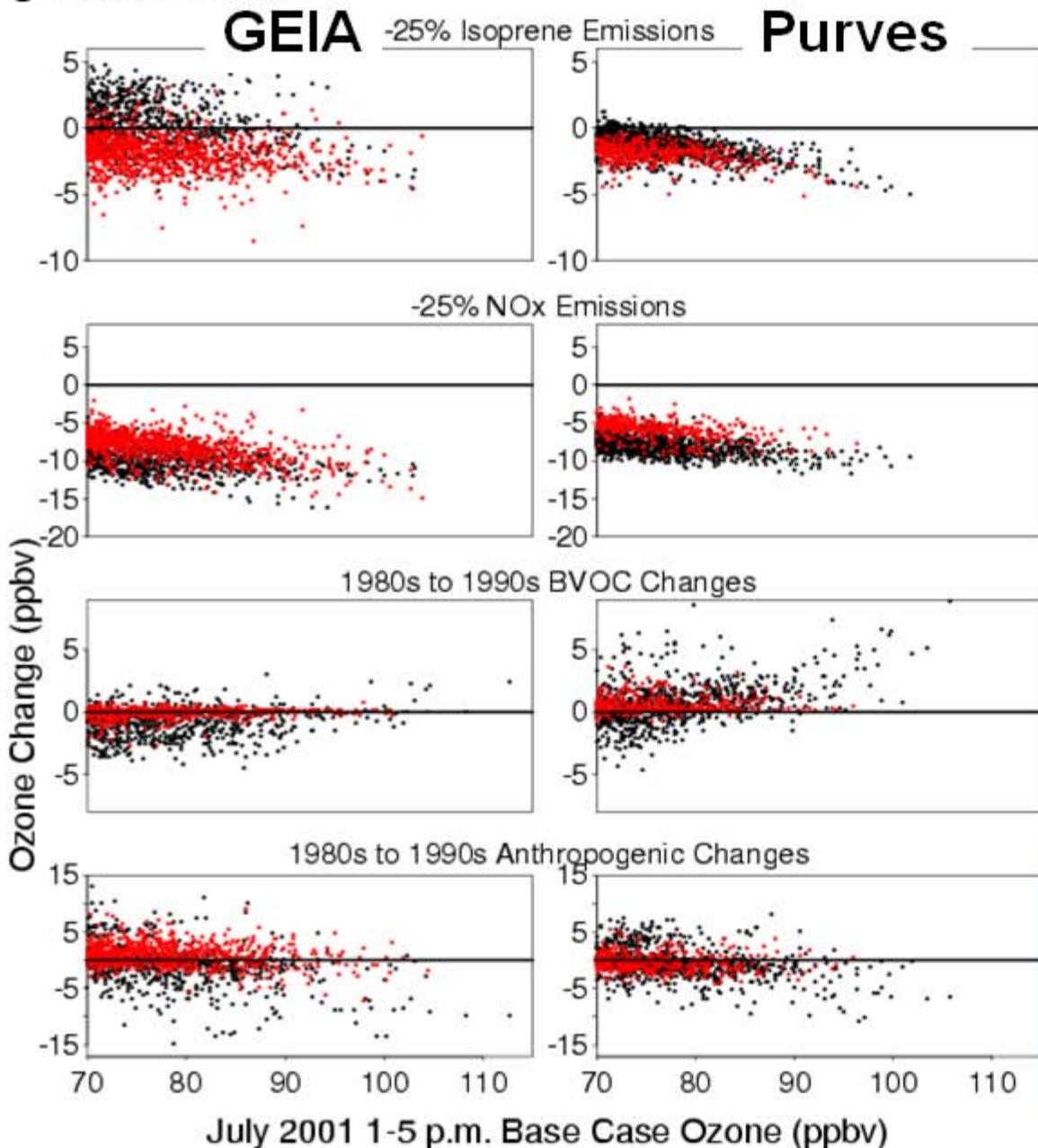
Observed changes in O<sub>3</sub> are not explained by regional emission changes alone...



● **Northeast**

● **Southeast**

## Impact of Sensitivity Simulations on High-O<sub>3</sub> Events:



▪ decrease with isoprene except for GEIA SE

▪ decrease with NO<sub>x</sub>, larger response with GEIA

▪ dominated by anthrop. (NO<sub>x</sub>) emissions but BVOC changes may offset for most extreme events

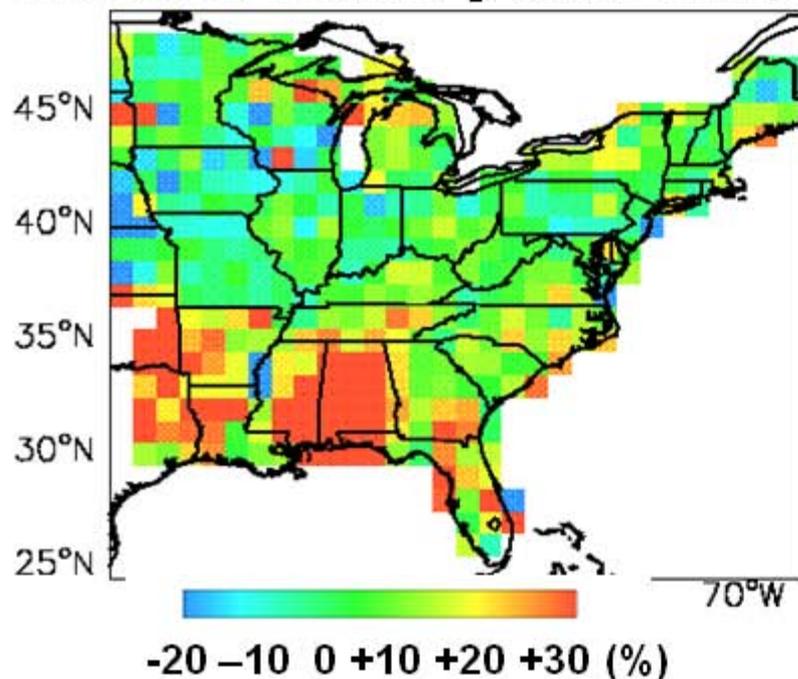
**Unclear whether recent BVOC emission changes mitigated/exacerbated high-O<sub>3</sub> events**

## Tool #2: MOZART-2 tropospheric chemistry model [*Horowitz et al., 2003*]

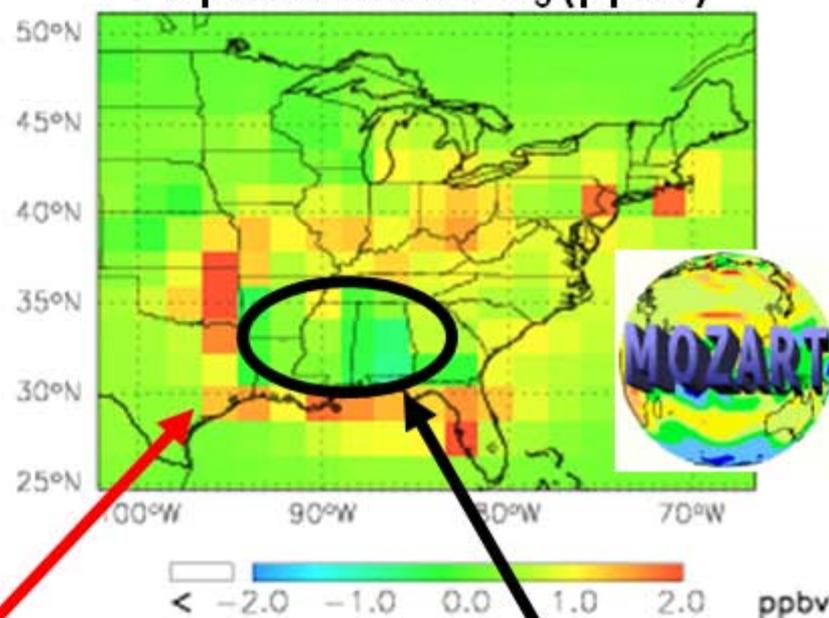
- assimilated meteorology: NCEP T62 ( $\sim 1.9^\circ$ ) 2001 ; 28 vertical levels
- GEIA isoprene inventory [*Guenther et al., 1995*]

### Does MOZART-2 also predict decreases in $O_3$ resulting from increases in isoprene emissions?

Isoprene emission changes from mid-80s to mid-90s [*Purves et al., 2004*]



MOZART-2: Change in July 1-5 p.m. surface  $O_3$  (ppbv)



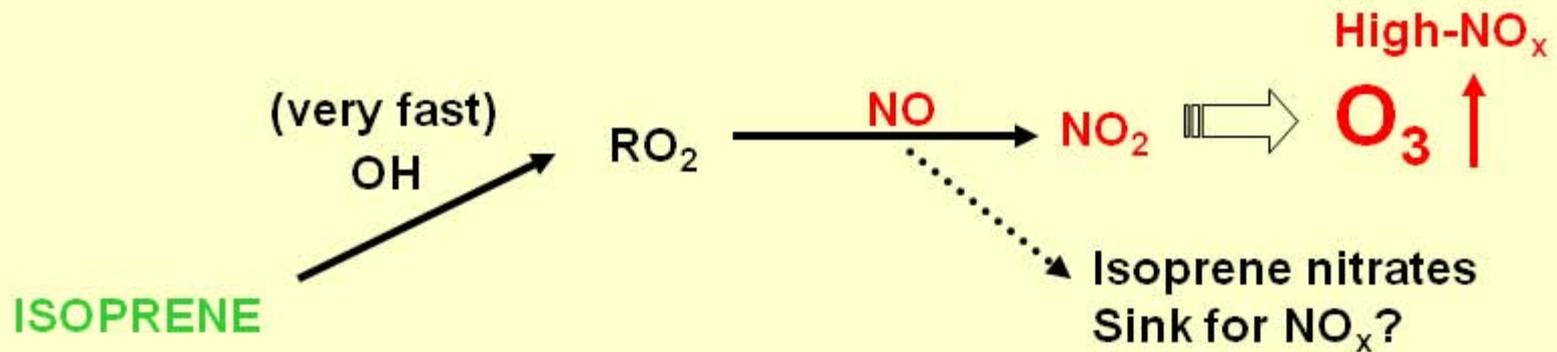
**$O_3$  increases  
(1-2 ppbv)**

**Little change  
( $NO_x$ -sensitive)**

**Isoprene nitrates are not a  $NO_x$  sink in MOZART-2:  $O_3$  Sensitivity?**

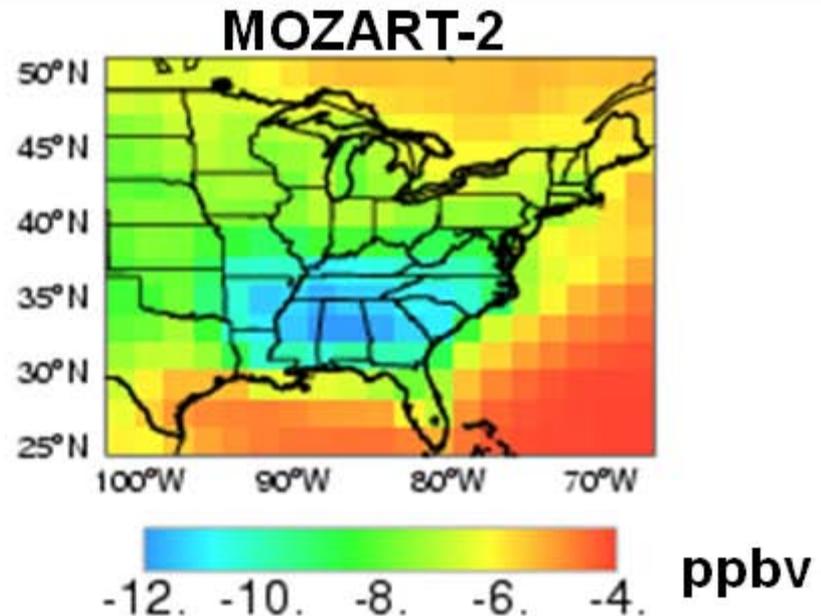


# What is the $O_3$ sensitivity to the uncertain fate of organic isoprene nitrates?



Change in July mean 1-5 p.m. surface  $O_3$  when isoprene nitrates act as a  $NO_x$  sink

→ 6-12 ppbv impact!



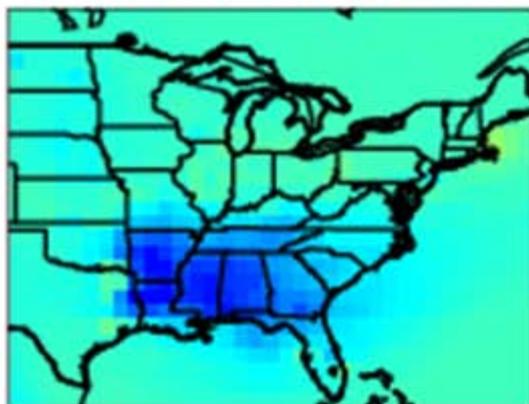
Revisit  $O_3$  response to isoprene changes with this assumption

**Chemical uncertainty: MOZART-2 shows similar results to GEOS-CHEM  
if isoprene nitrates are a  $\text{NO}_x$  sink**

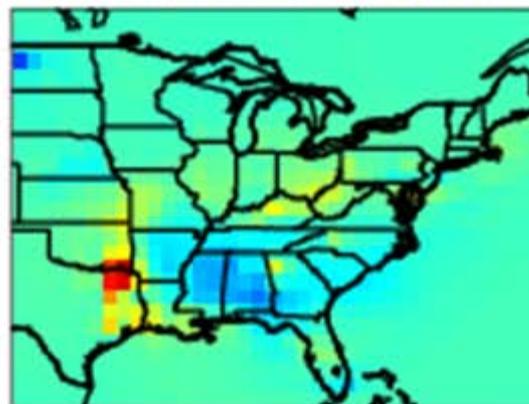
Change in July 1-5 p.m. surface  $\text{O}_3$  (ppbv)  
(due to isoprene emissions changes from mid-1980s to mid-1990s)

With 12% yield of isoprene nitrates

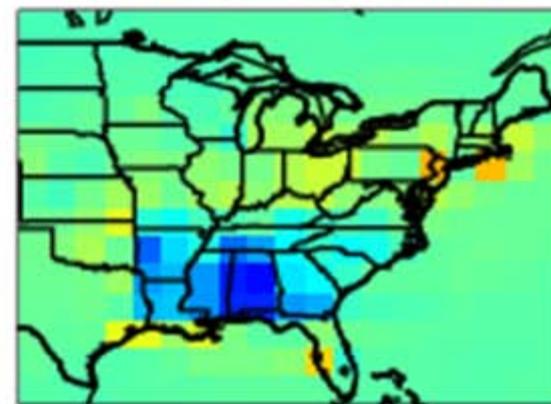
**GEOS-CHEM: GEIA**



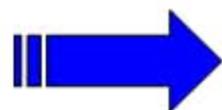
**GEOS-CHEM: Purves**



**MOZART-2: GEIA**



-2. -1. 0. 1. 2. 3. ppbv



**Understanding fate of isop. nitrates essential for  
predicting sign of response to changes in  
isoprene emissions**

## Conclusions and Remaining Challenges

- Better constrained isoprene emissions are needed to quantify:
  1. isoprene contribution to E. U.S. surface O<sub>3</sub>
  2. how O<sub>3</sub> responds to both anthrop. and biogenic emission changes (deserves consideration in planning biofuel plantations)
  - Utility of satellite CH<sub>2</sub>O columns?
  - New inventories (MEGAN, BEIS-3) more accurate?
  - Aircraft observations from recent ICARTT campaign?
- Recent isoprene increases may have reduced surface O<sub>3</sub> in the SE
  - Does this regime actually exist?
  - Fate of organic nitrates produced during isoprene oxidation?
  - Results consistent in MOZART-2 and GEOS-CHEM
- Reported regional emission changes from 1980s to 1990s alone do not explain observed O<sub>3</sub> trends
  - Are anthropogenic emissions inventories sufficient to support trend studies? (*Parrish et al., JGR 2002*)
  - Decadal shifts in meteorology?
  - Changing global O<sub>3</sub> background?