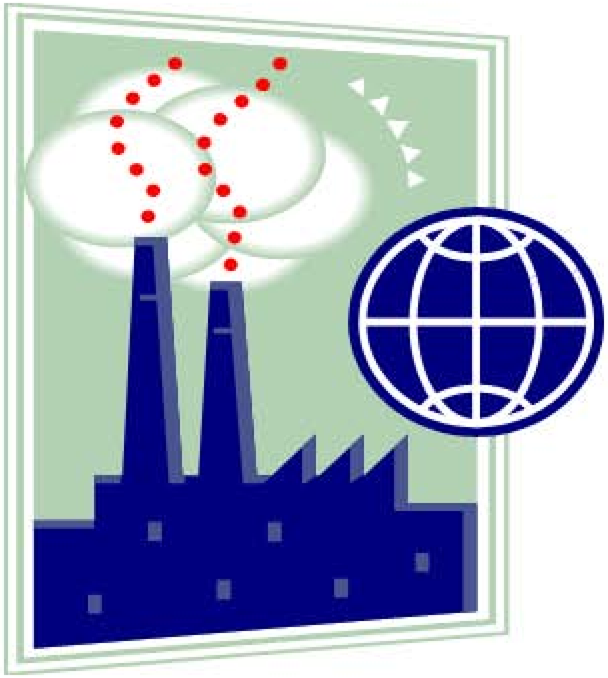

Background Ozone in Surface Air over the United States:

Variability, Climate Linkages, and Policy Implications

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Department of Environmental Sciences Seminar
Rutgers University
March 4, 2005

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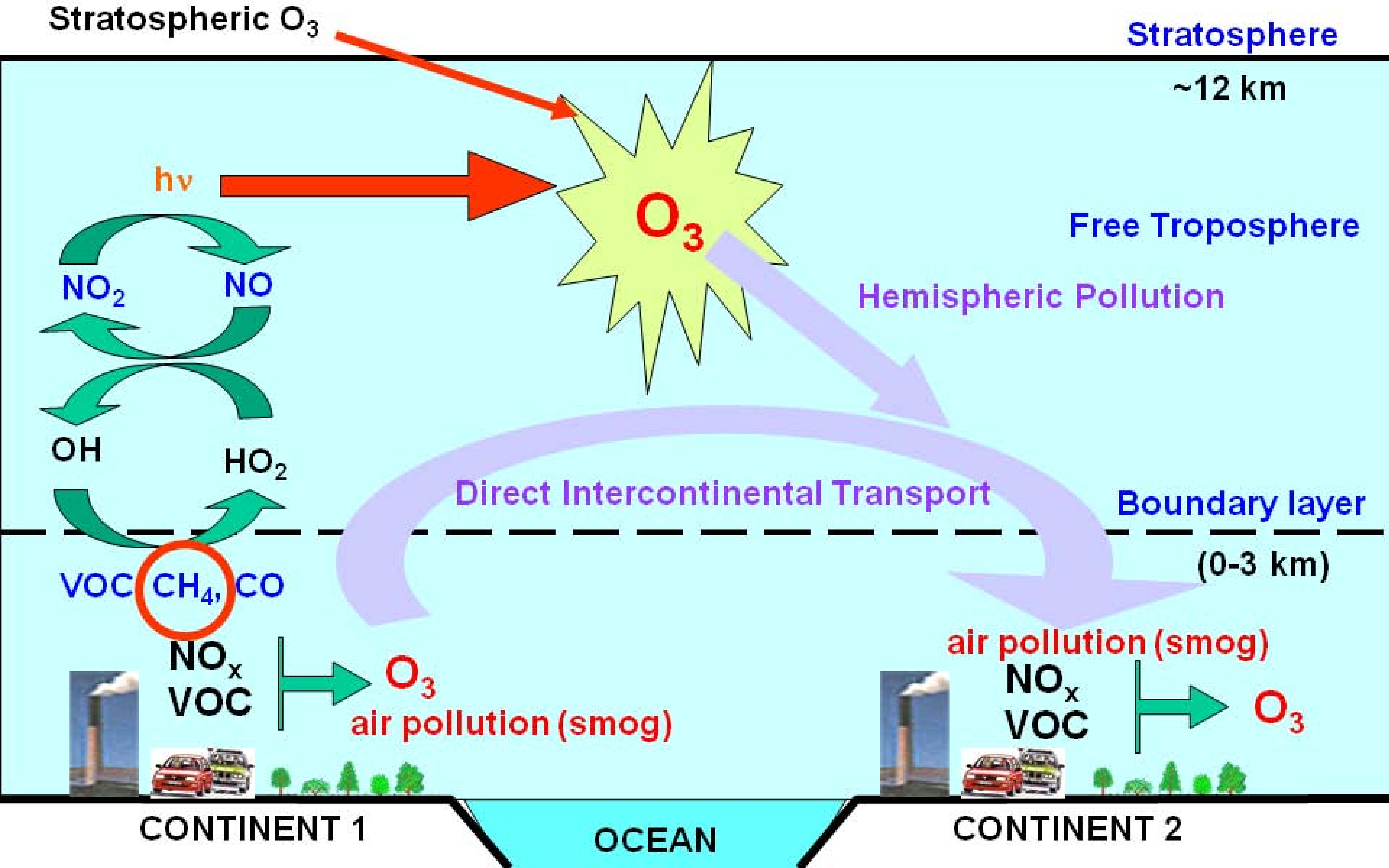
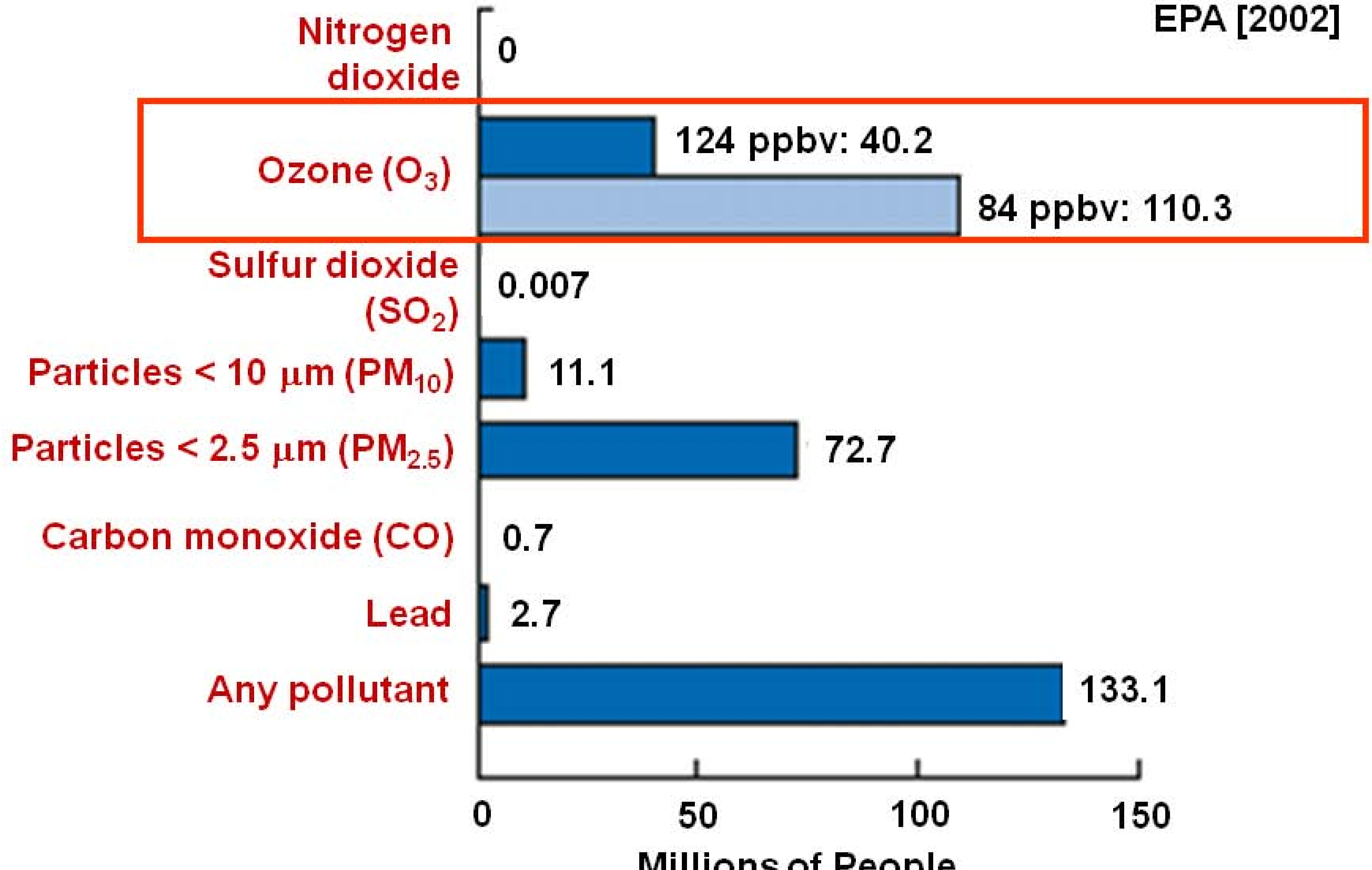
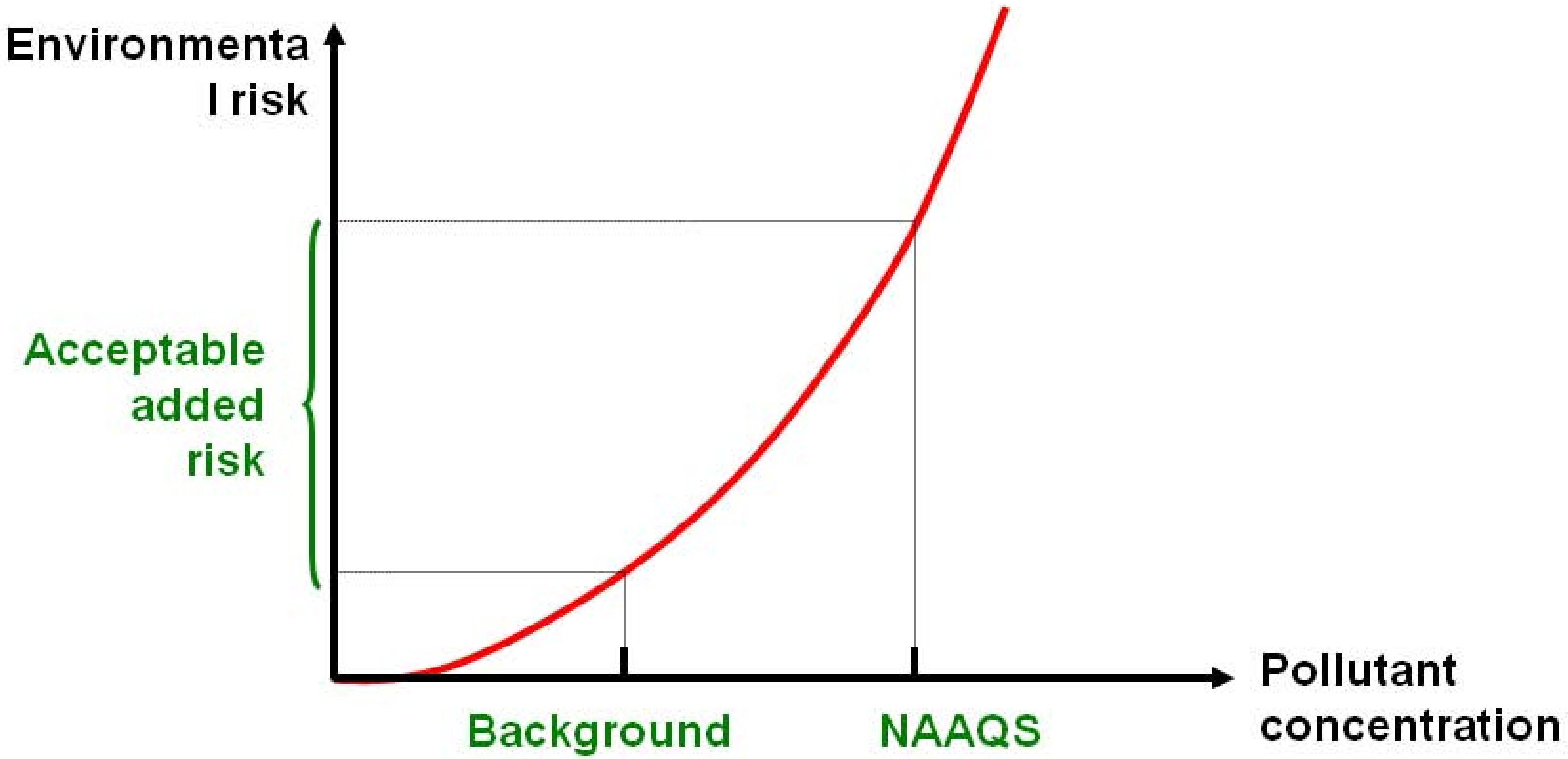
What is the origin of tropospheric ozone?



Number of People Living in U.S. Counties Violating National Ambient Air Quality Standards (NAAQS) in 2001

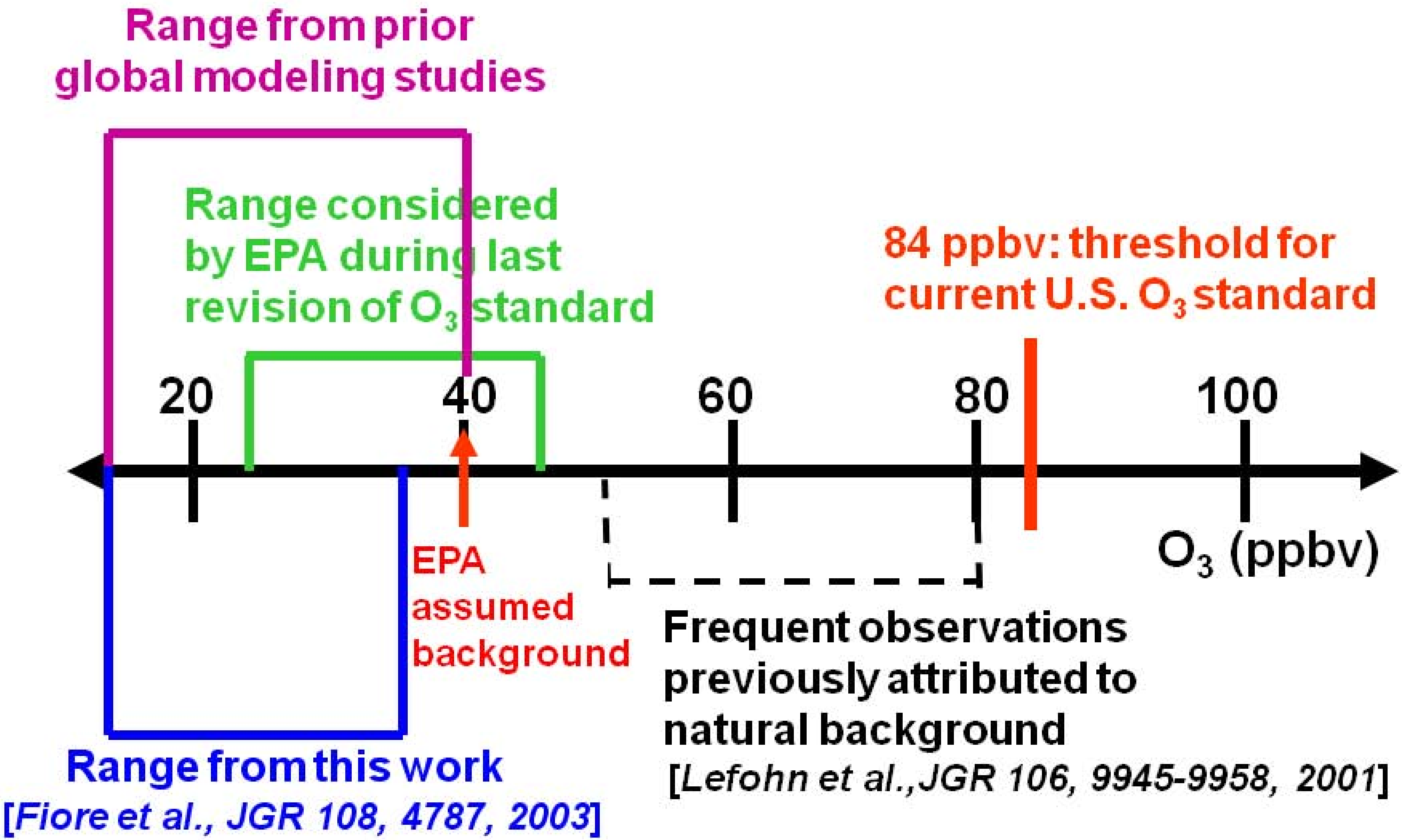


Background Estimates are Used When Setting NAAQS



→Risk assessments account for risks associated with exposure to the increment of ozone above the background (*i.e.*, the risk that can potentially be reduced via North American anthropogenic emission controls)

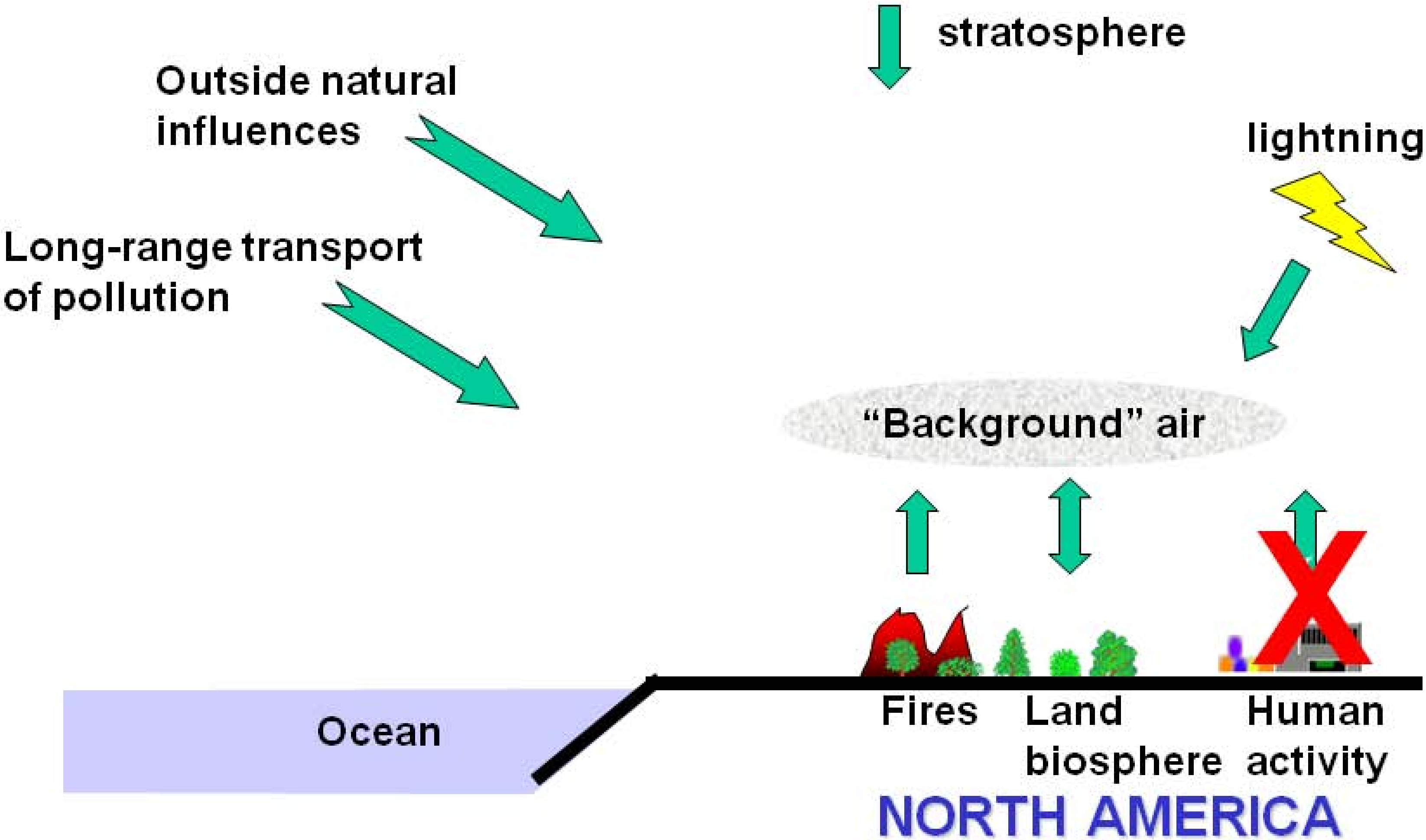
Range of "background O_3" estimates in U.S. surface air



The U.S. EPA considers background levels when setting the NAAQS

"POLICY RELEVANT BACKGROUND" OZONE:

Ozone concentrations that would exist in the absence of anthropogenic emissions from North America [*EPA CD*, 2005]



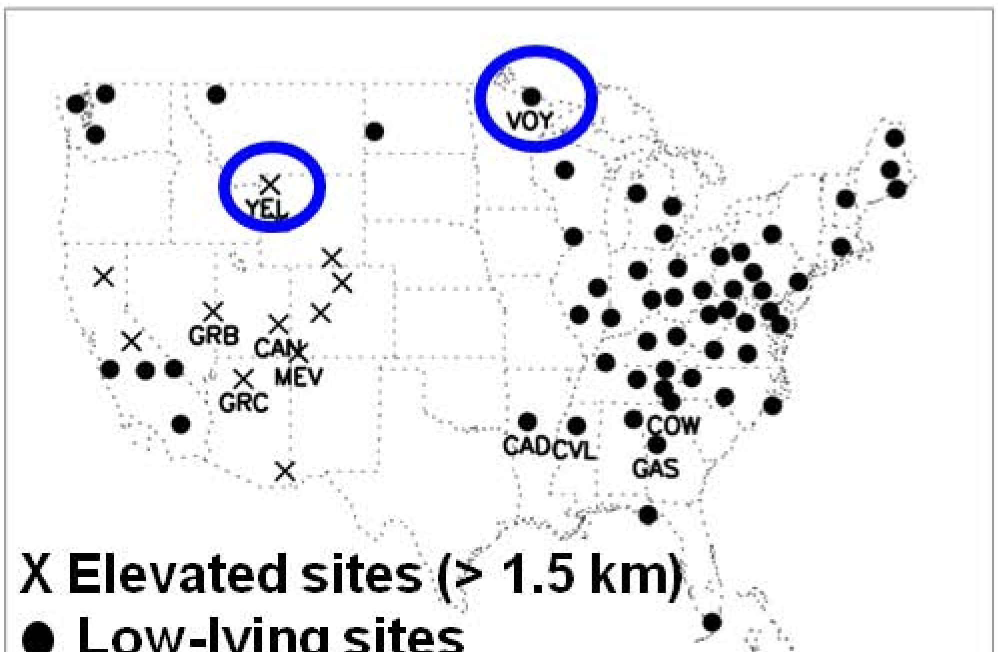
Policy Relevant Background is not directly observable
→ Must be estimated with models

APPROACH: Use 2001 CASTNet data in conjunction with GEOS-CHEM to

1. quantify background O_3 and its various sources
2. diagnose origin of springtime high-O_3 events at remote U.S. sites, previously attributed to natural, stratospheric influence

Observations:

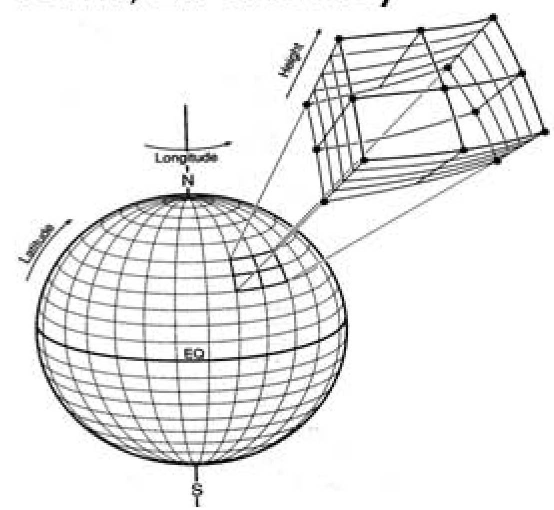
CASTNet Stations
(EPA, Nat'l Park Service)



MODEL:

GEOS-CHEM 3D Tropospheric Chemistry Model

[*Bey et al.*, 2001] (uses assimilated meteorology; 48 σ; 4°x5° or 2°x2.5° horiz. resn., 24 tracers)

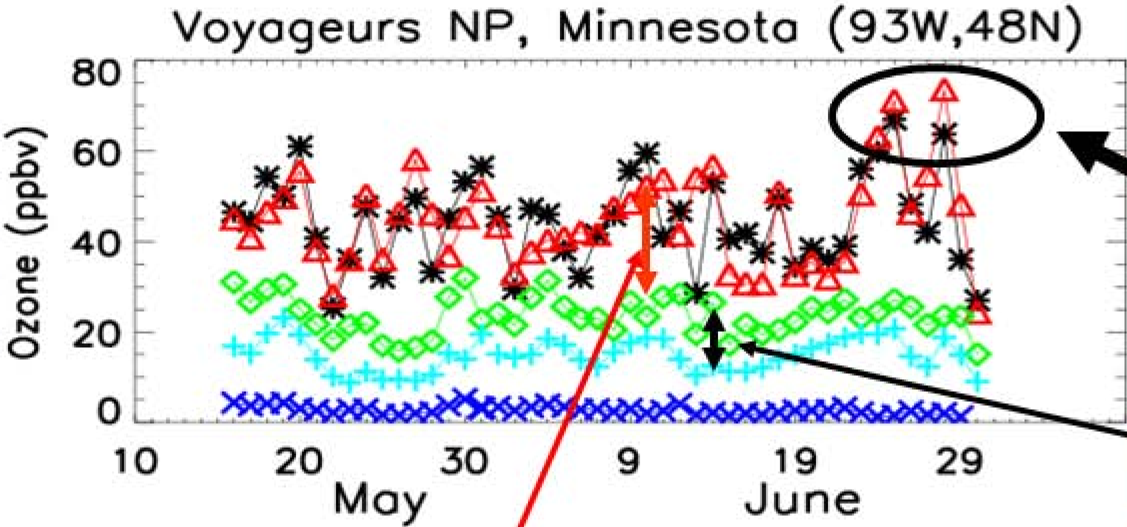


Case Studies: Ozone Time Series at Sites used in *Lefohn et al.* [2001]

* CASTNet observations ▲ Model Base Case (2001) X Stratospheric influence

◆ Background (no N. Amer. Anthrop emissions; present-day CH_4)

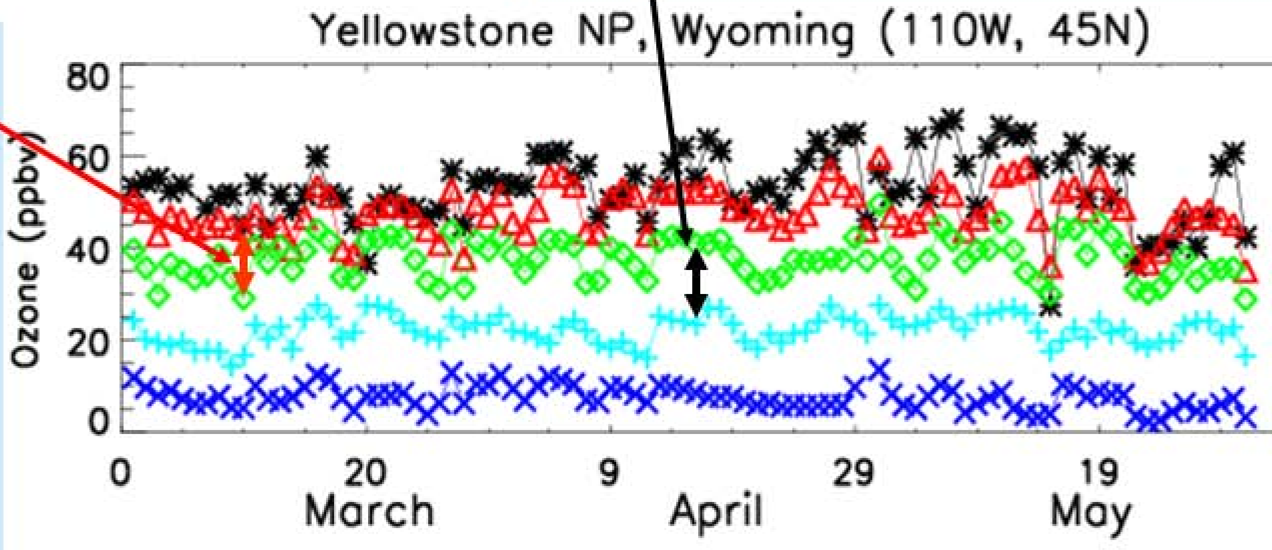
+ Natural O_3 level (no global anthrop. Emissions; preindustrial CH_4)



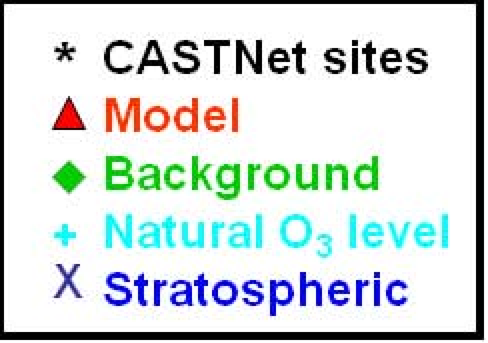
High-O_3 events dominated by regional pollution

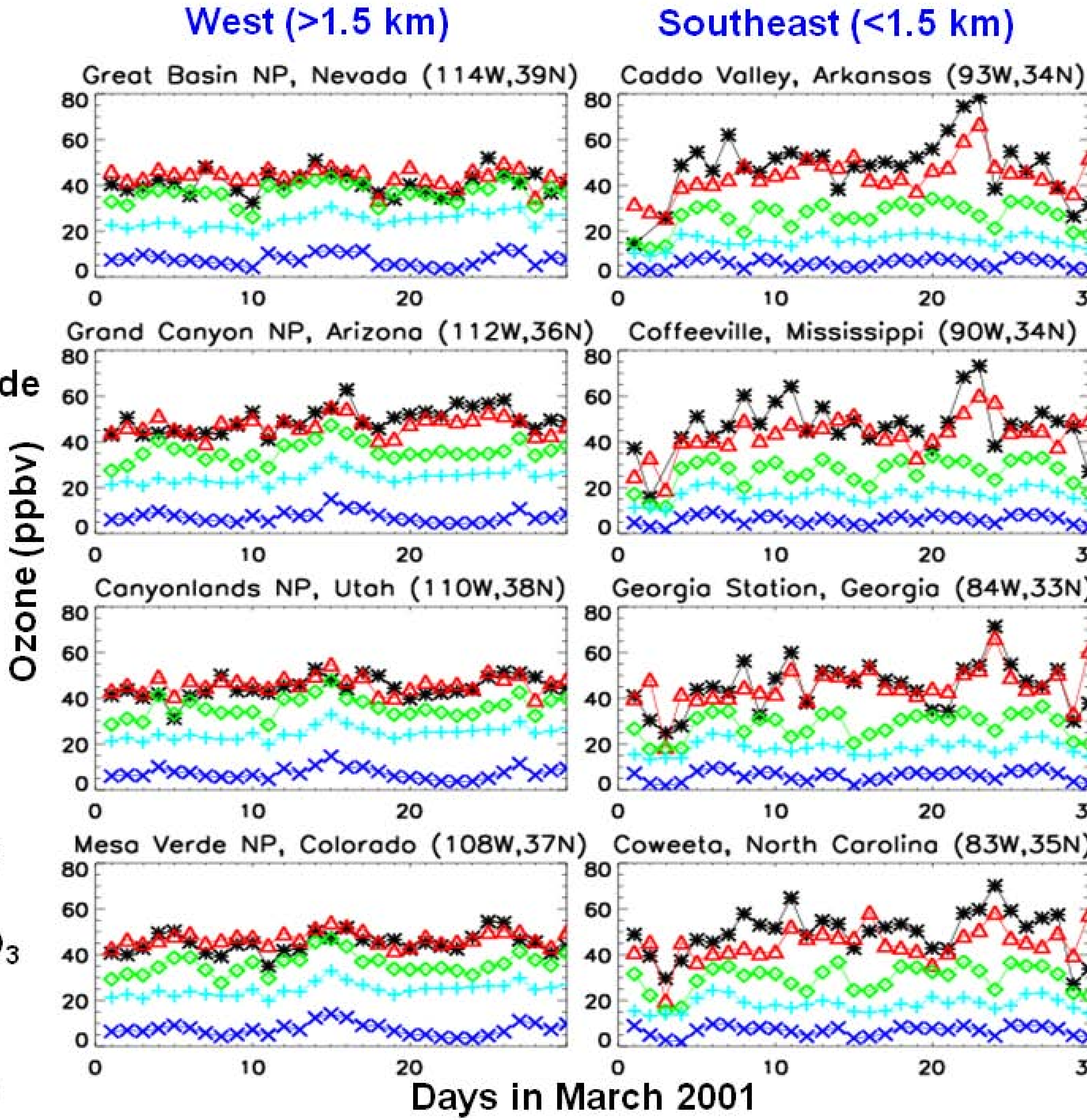
hemispheric pollution

regional pollution



Background at high-altitude site (2.5 km) not representative of contribution at low-lying sites





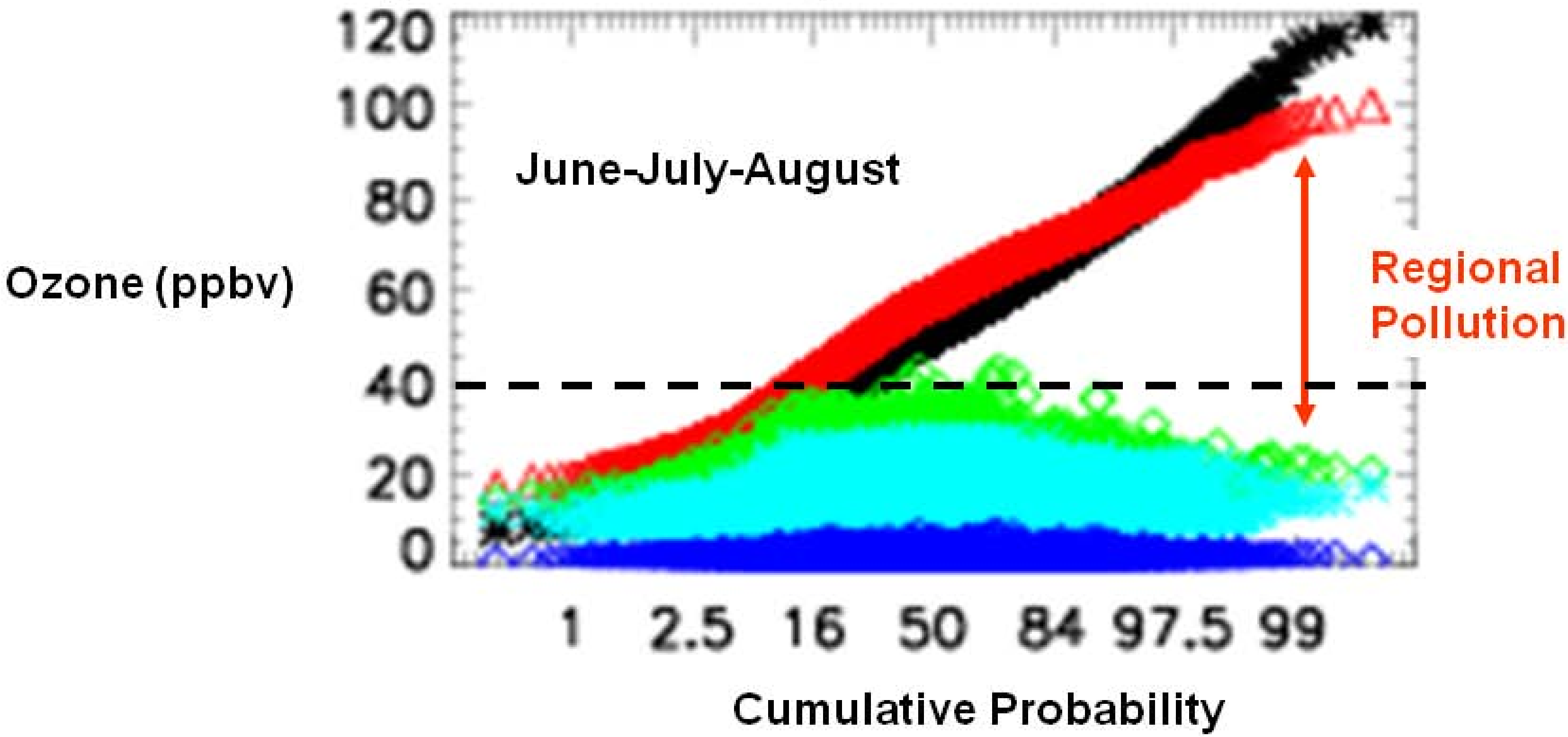
Background O_3 higher at high-altitude western sites

Background O_3 lower at low-lying southeastern sites; decreases with highest observed O_3

Fiore et al., JGR, 2003

Background O_3 even lower under polluted conditions

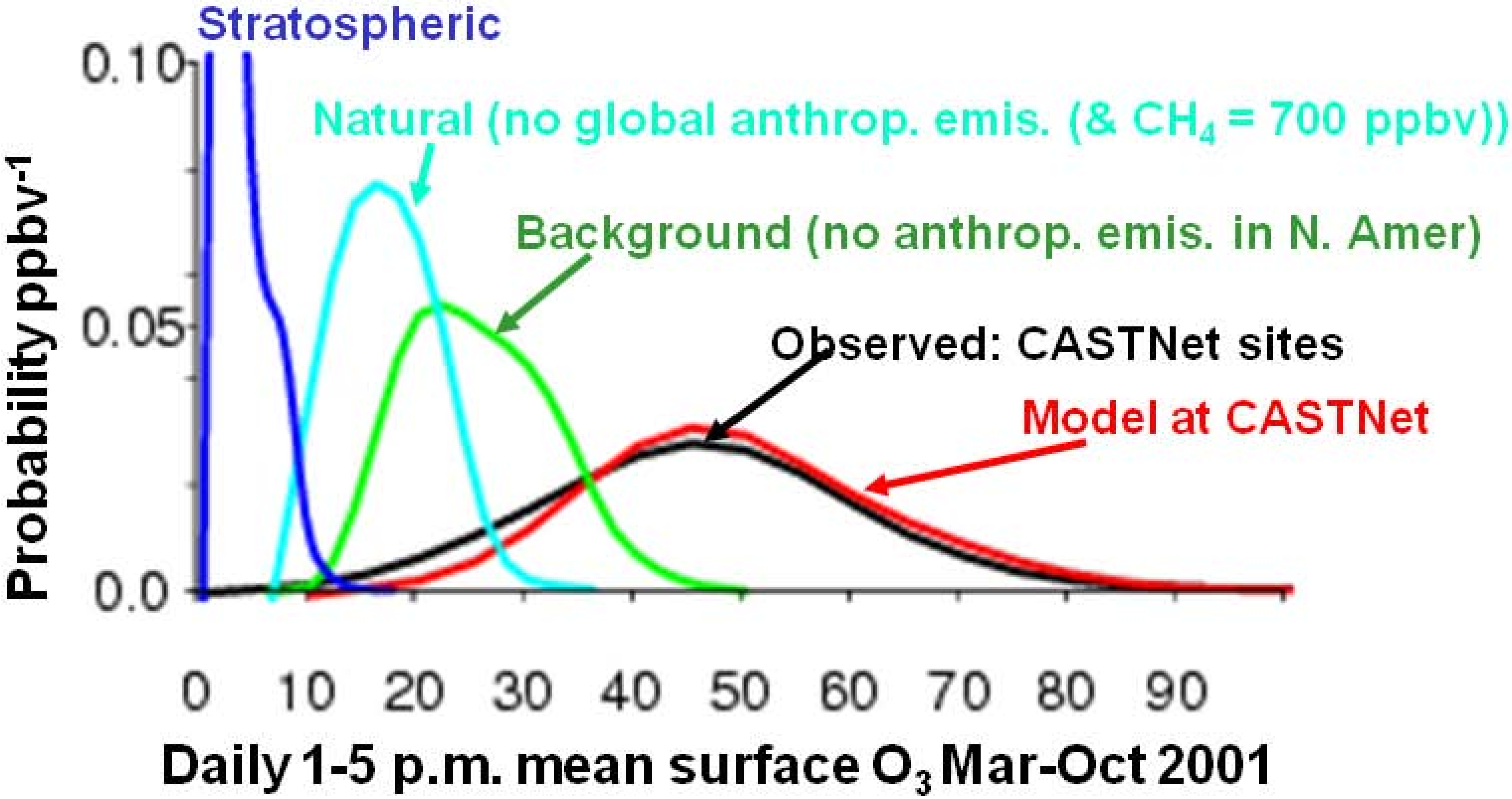
Daily mean afternoon O_3 at 58 low-elevation U.S. CASTNet sites



Background on polluted days well below 40 ppbv assumed by EPA
→ health risks underestimated with current (1996) approach

Fiore et al., JGR, 2003

Compiling Results from all (71) CASTNet sites: Natural vs. Anthropogenic Contributions



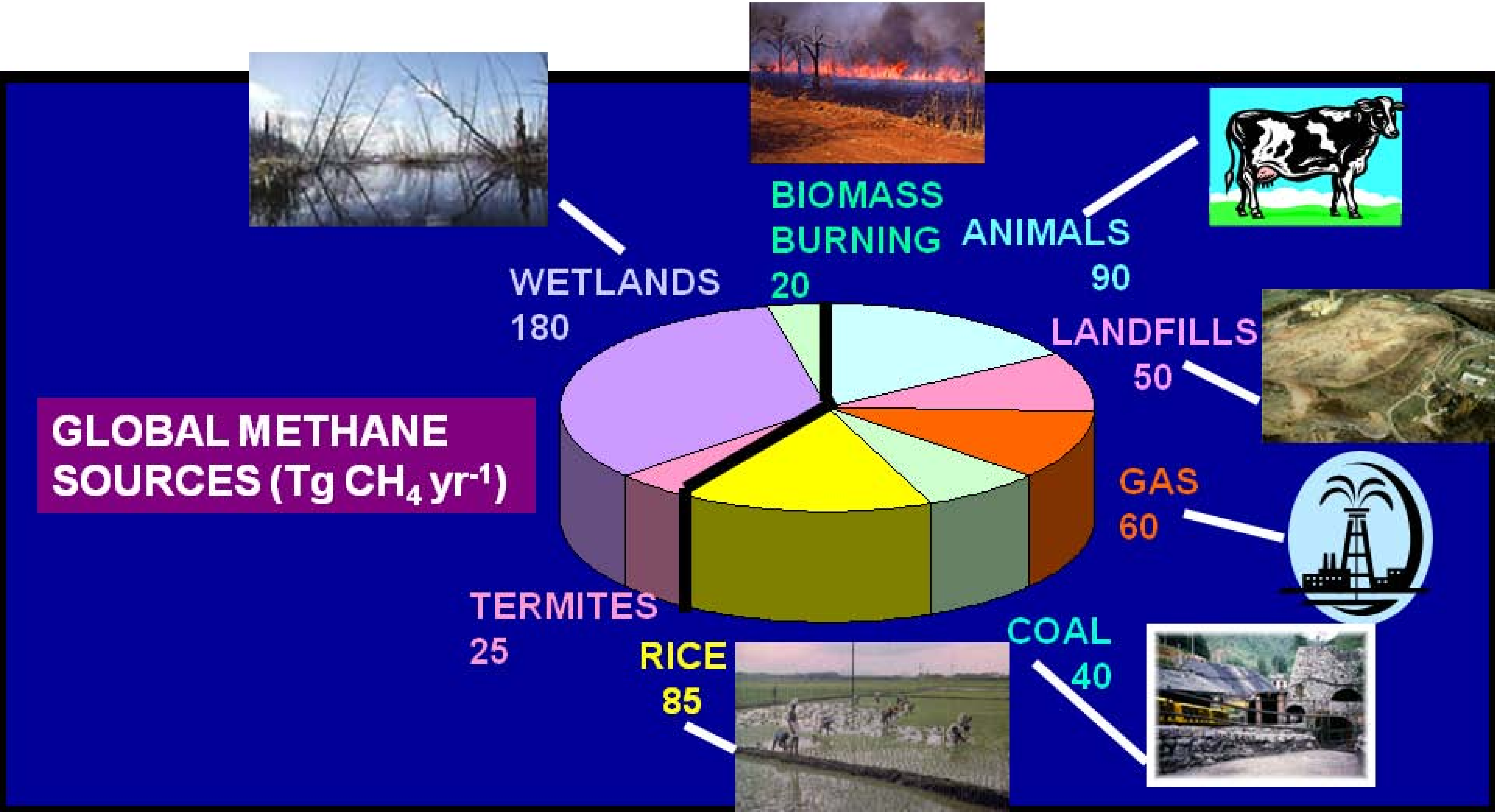
Typical ozone values in U.S. surface air:

Background 15-35 ppbv; Natural 10-25 ppbv; Stratosphere < 20 ppbv

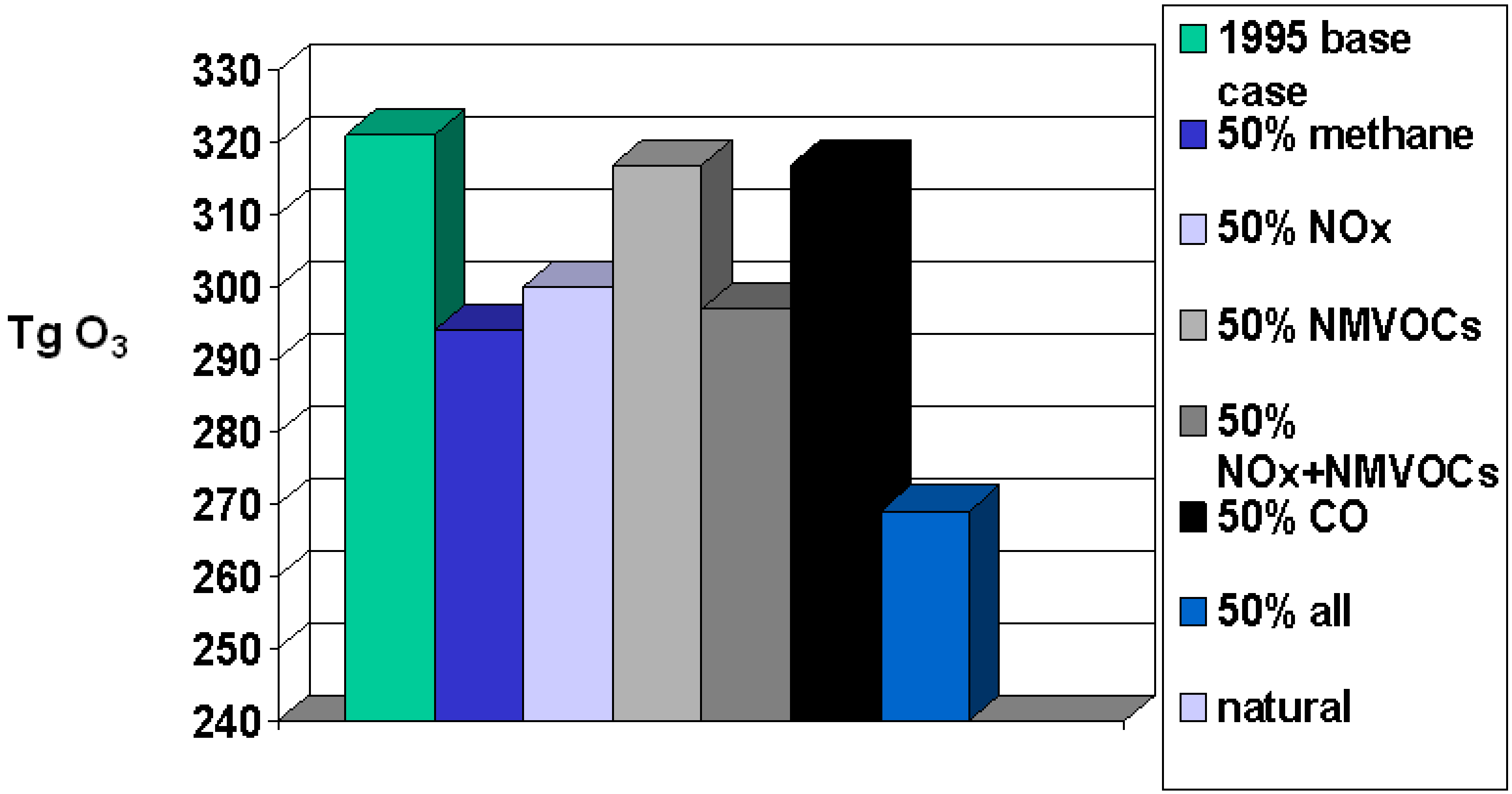
Anthropogenic methane enhances "background" above "natural"

Fiore et al., JGR, 2003

More than half of global methane emissions are influenced by human activities



Anthropogenic Methane Emissions Enhance the Tropospheric Ozone Background

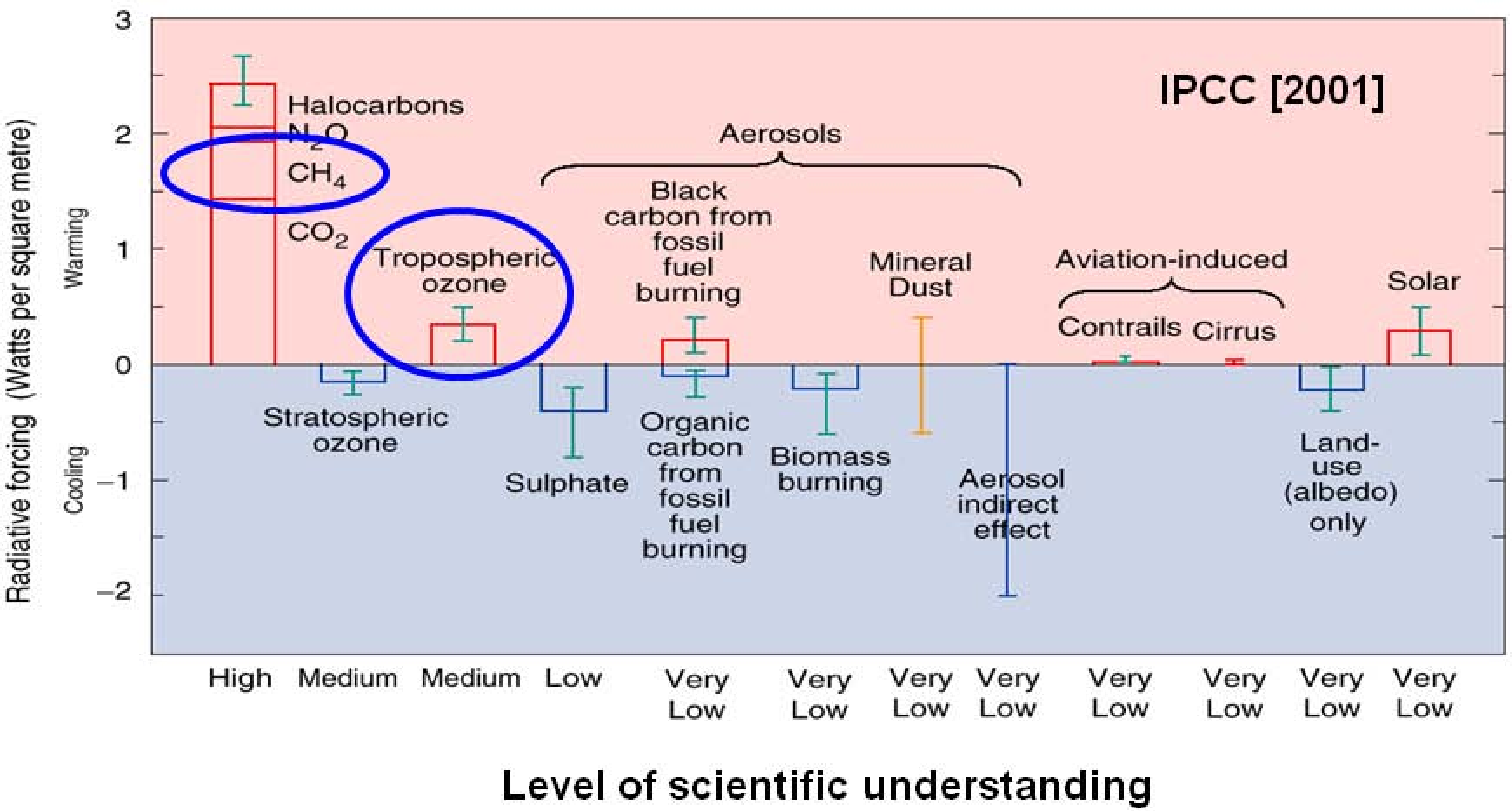


Sensitivity of global tropospheric ozone inventory in GEOS-CHEM to 50% global reductions in anthropogenic emissions

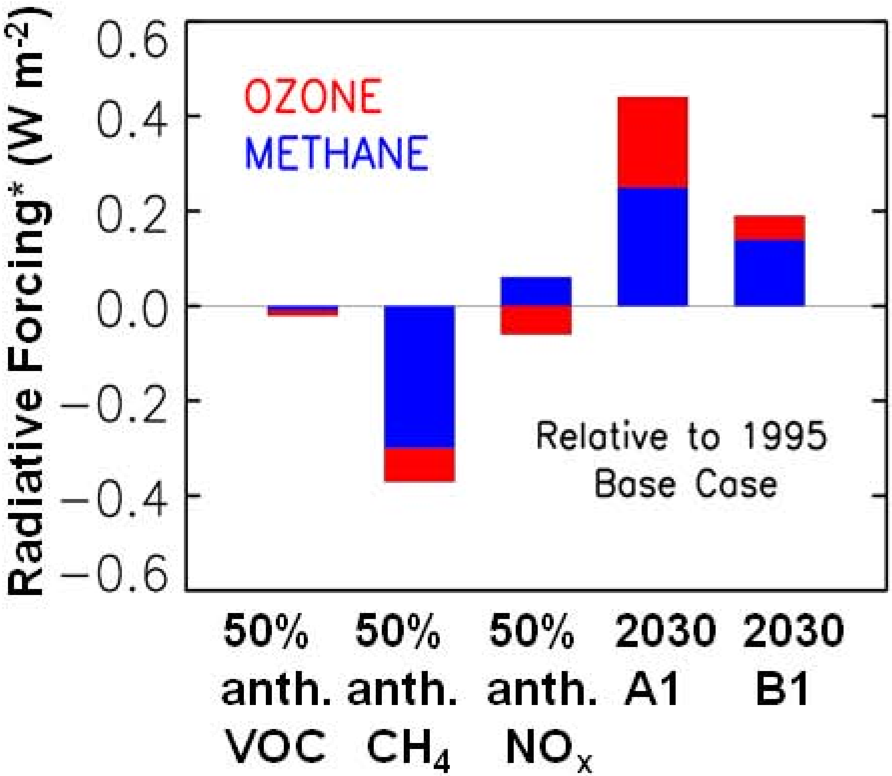
→ Anthropogenic emissions of NO_x and methane have largest influence on tropospheric ozone.... climate? air pollution?

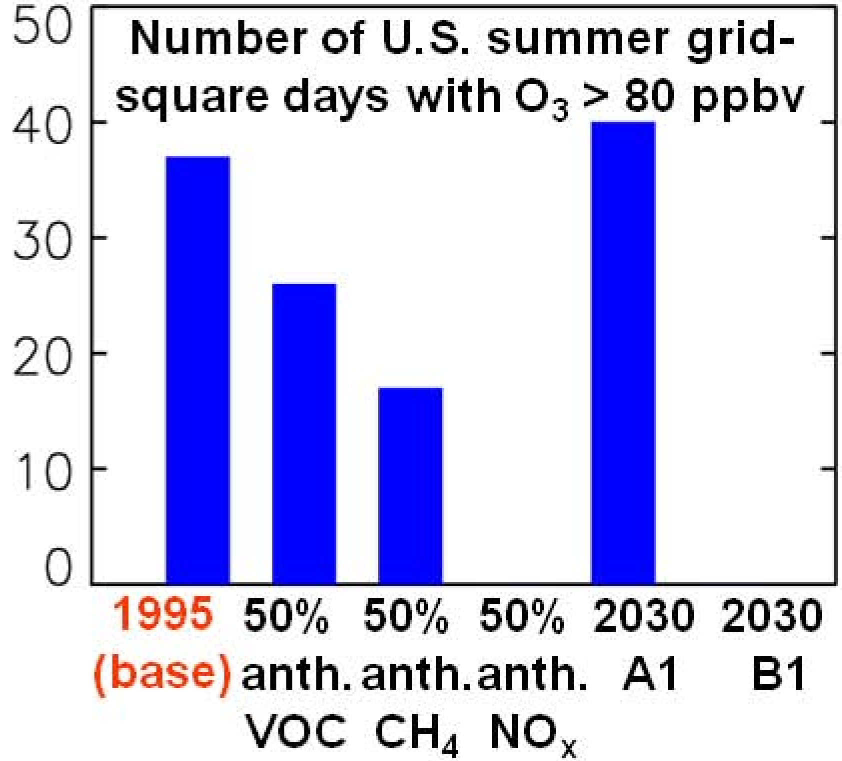
Fiore et al., GRL, 2002

Radiative Forcing of Climate, 1750-Present: Important Contributions from Methane and Ozone



Double dividend of Methane Controls: Decreased greenhouse warming and improved air quality





IPCC scenario	Anthrop. NO_x emissions (2030 vs. present) Global	U.S.	Methane emissions (2030 vs. present)
A1	+80%	-20%	+30%
B1	-5%	-50%	+12%

CH_4 links air quality & climate via background O_3

Fiore et al., GRL, 2002

CONCLUSIONS ... and their implications for public policy

1. Background O_3 is typically less than 40 ppbv; even lower under polluted conditions

   → health risk from O_3 underestimated in 1996 EPA risk assessments

2. Pollution from North America contributes to high-O_3 events at remote U.S. sites in spring

   → these events do not represent U.S. background conditions and should not be used to challenge legitimacy of O_3 NAAQS

3. Hemispheric pollution enhances U.S. background

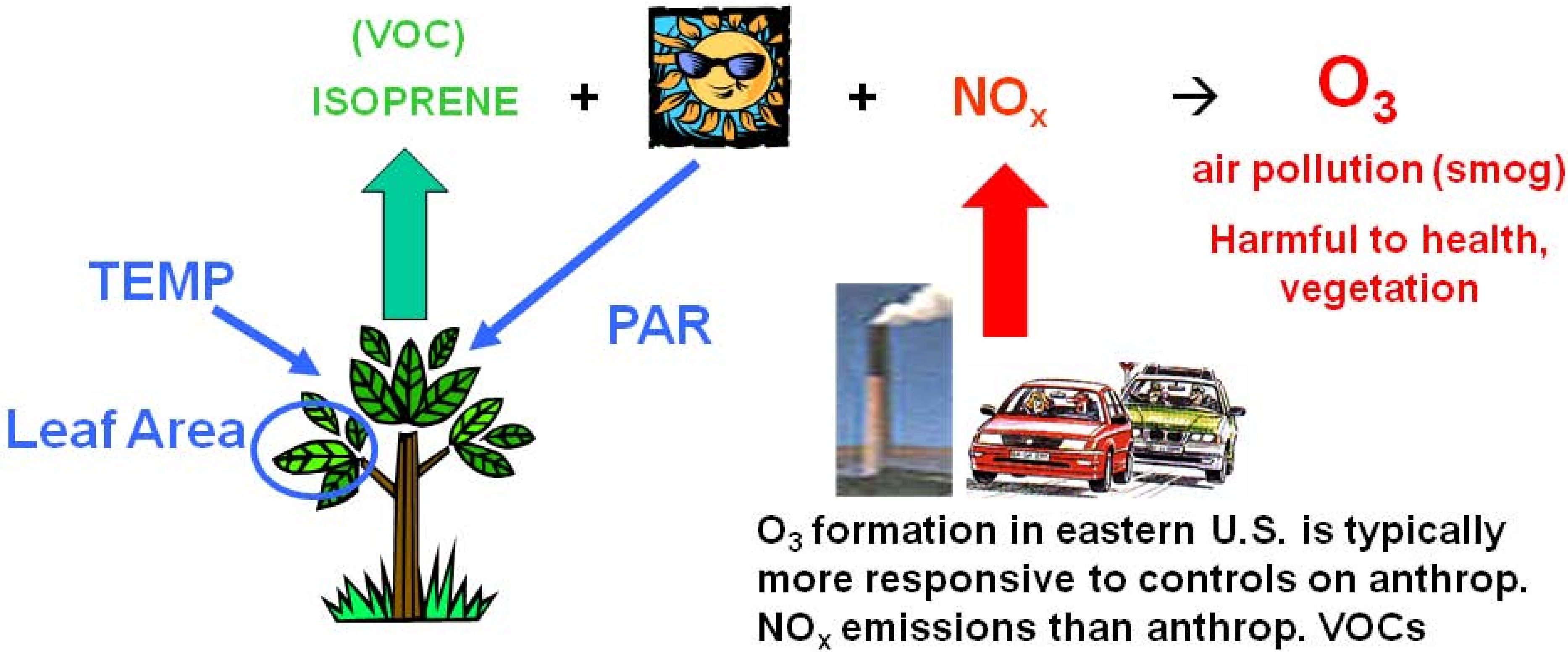
   → international agreements to reduce hemispheric background should improve air quality & facilitate compliance w/ more stringent standards

   → reducing CH_4 decreases both background O_3 and greenhouse forcing; enables simultaneous pursuit of air quality & climate goals

   → global CH_4 controls are viable [J. West seminar next Friday!] and complement local-to-regional NO_x & NMVOC controls

What is the role of uncertainties/changes in "natural" emissions?
First step: BVOC emissions

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

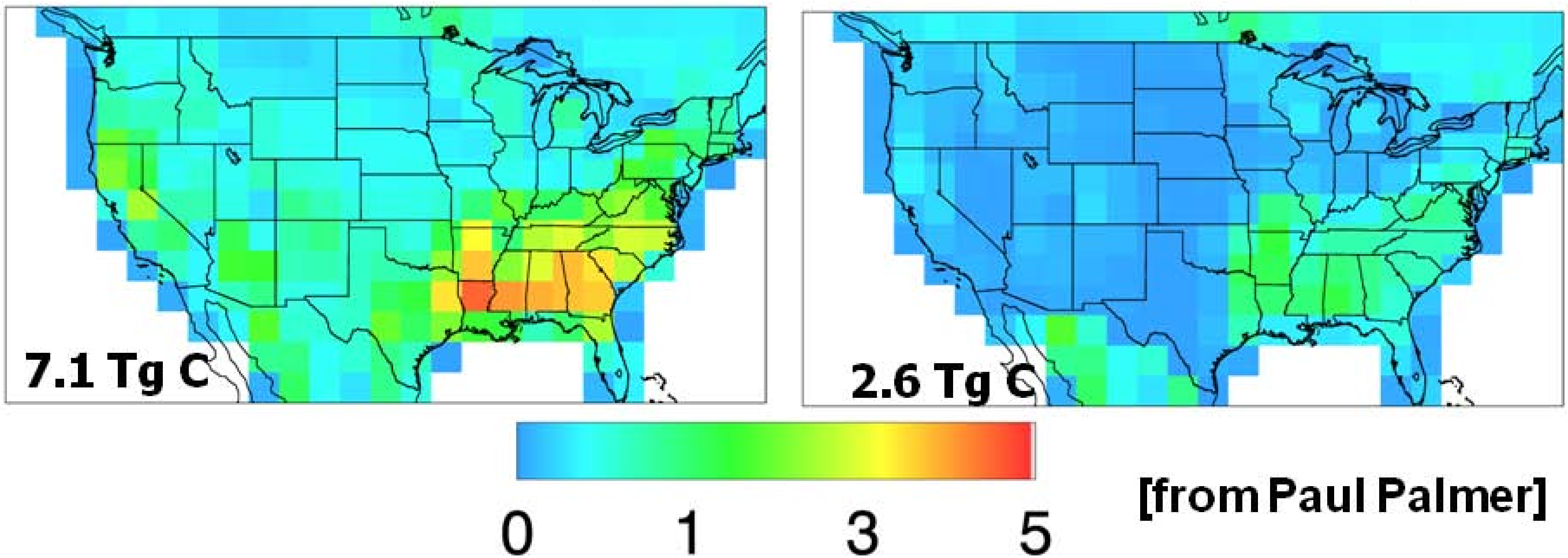
-- Land-use

Isoprene Emission Inventories uncertain by at least a factor of 2

Isoprene emissions – July 1996

GEIA: Global Isoprene Emission Inventory

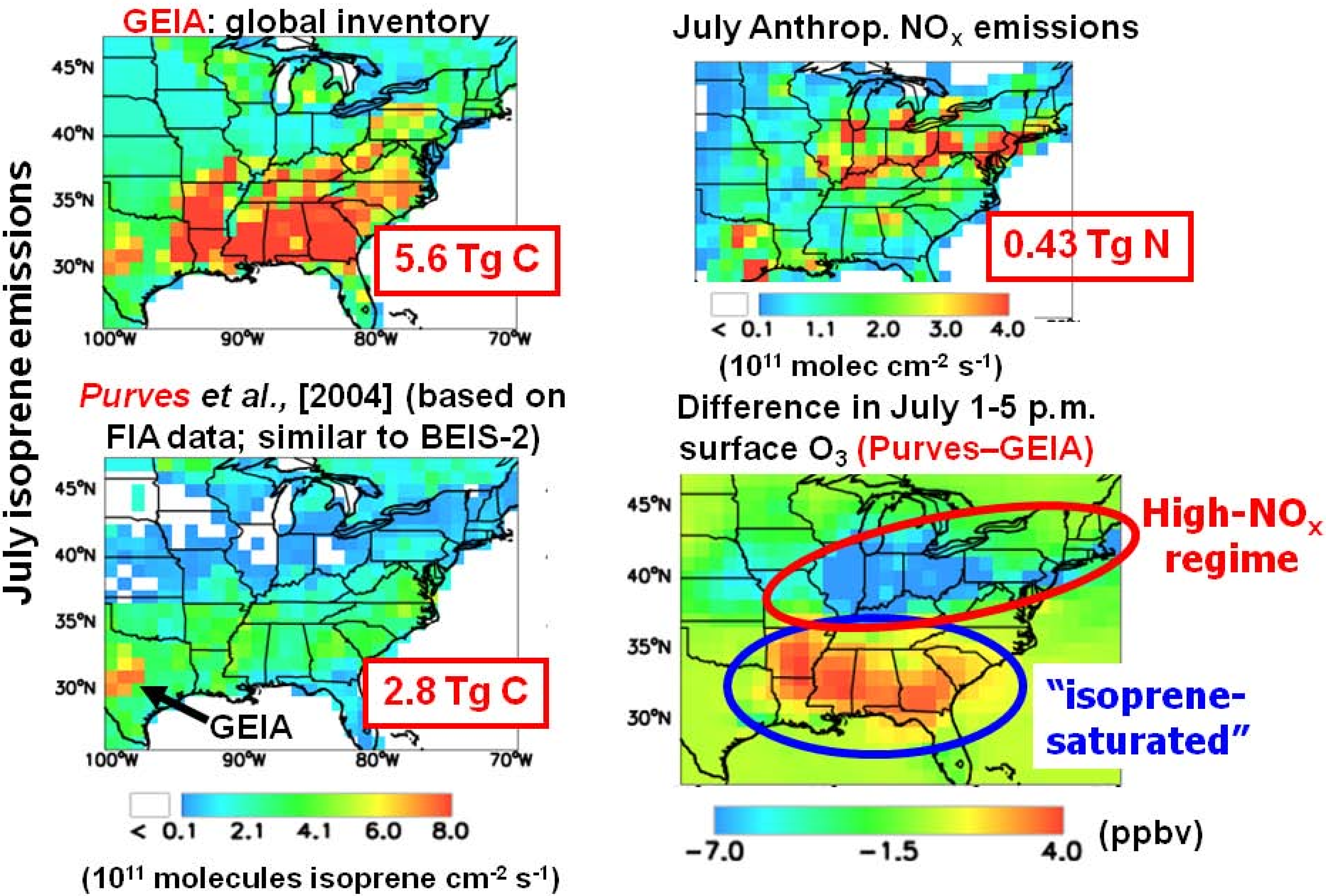
BEIS2: Regional Isoprene Emission Inventory



$[10^{12}$ atom C cm^{-2} $s^{-1}]$

[from Paul Palmer]

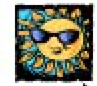
Choice of isoprene inventory critical for predicting base-case O_3
(2001 meteorology; 1x1 nested GEOS-CHEM [Wang et al., 2004; Li et al., 2004])



Complicated Chemistry: Isoprene may also decrease surface O_3 in low-NO_x, high isoprene settings

ISOPRENE +  + NO_x → O_3

ISOPRENE

(very fast) OH → RO_2 —NO→ NO_2  ⇒ O_3 ↑ High-NO_x

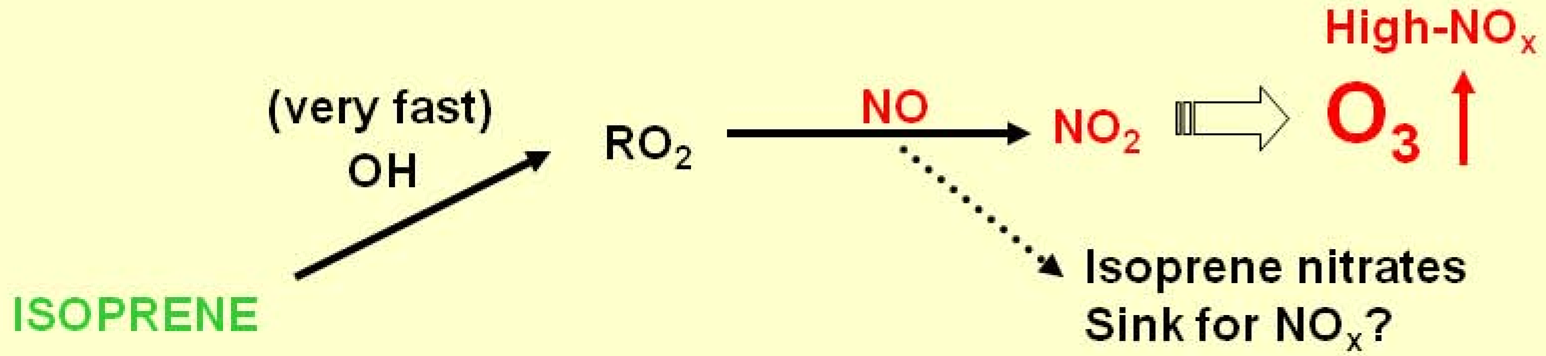
⋯→ Isoprene nitrates
Sink for NO_x?

O_3 (slower) → O_3 ↓ Low-NO_x, high-isoprene

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+hν -> OH (11d)

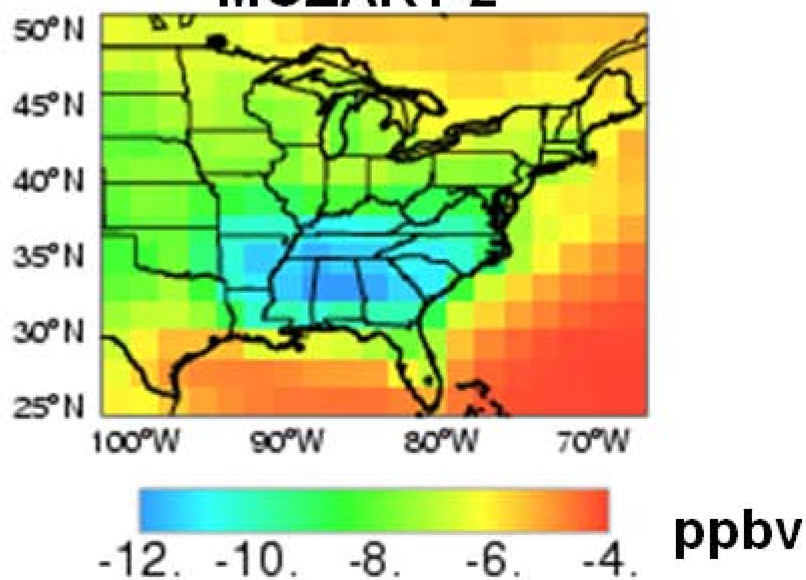
What is the O_3 sensitivity to the uncertain fate of organic isoprene nitrates?



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

→ 6-12 ppbv impact!

MOZART-2

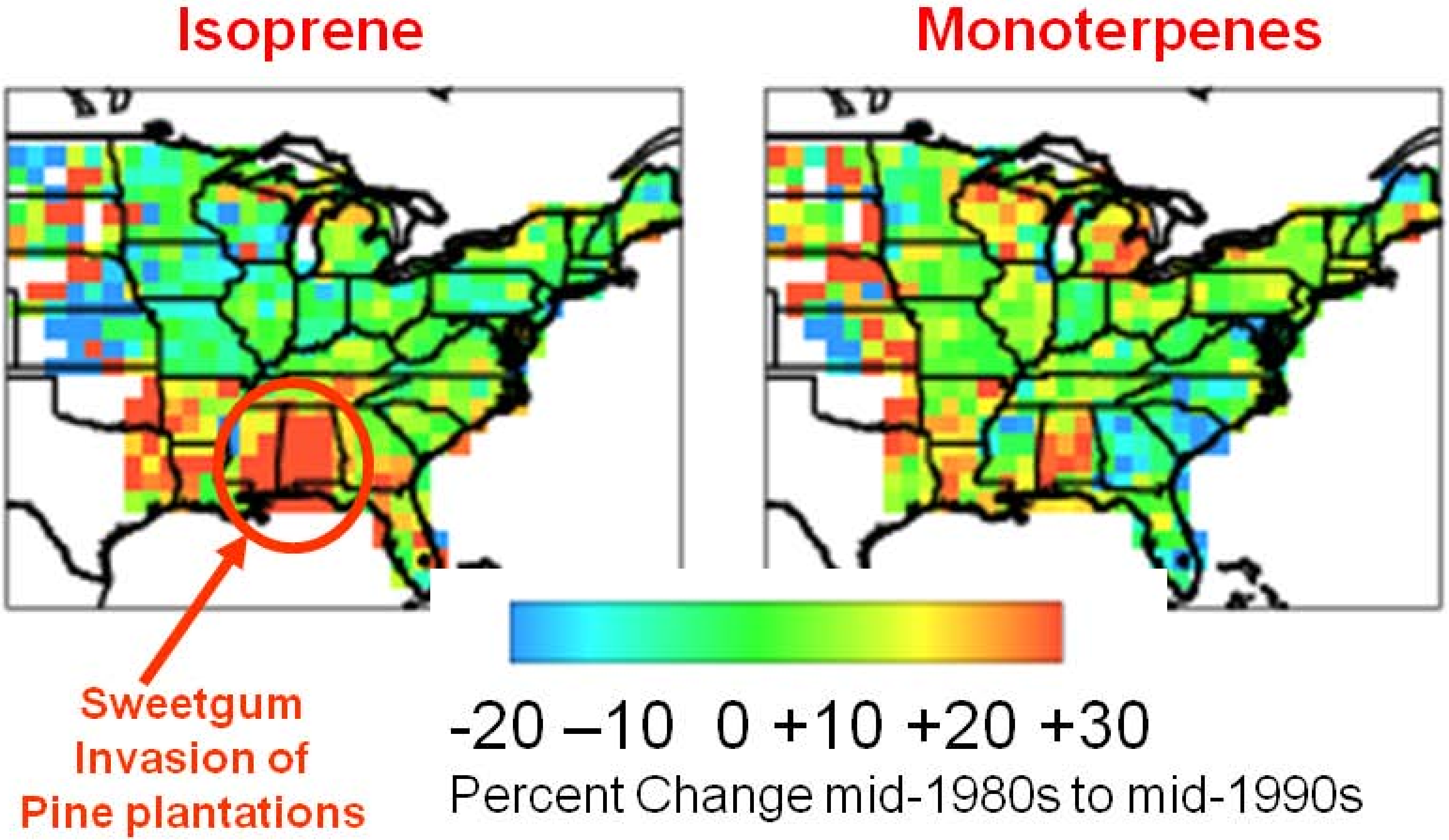


Recent Changes in Biogenic VOC Emissions

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

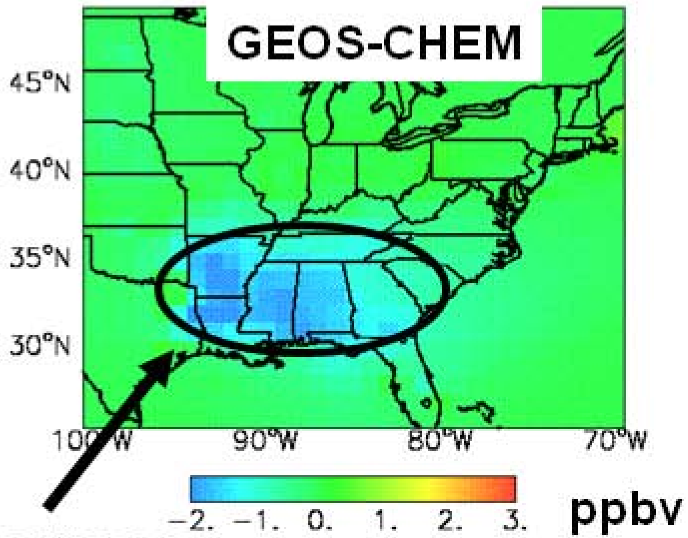
→ Substantial isoprene increases in southeastern USA largely driven by human land-use decisions

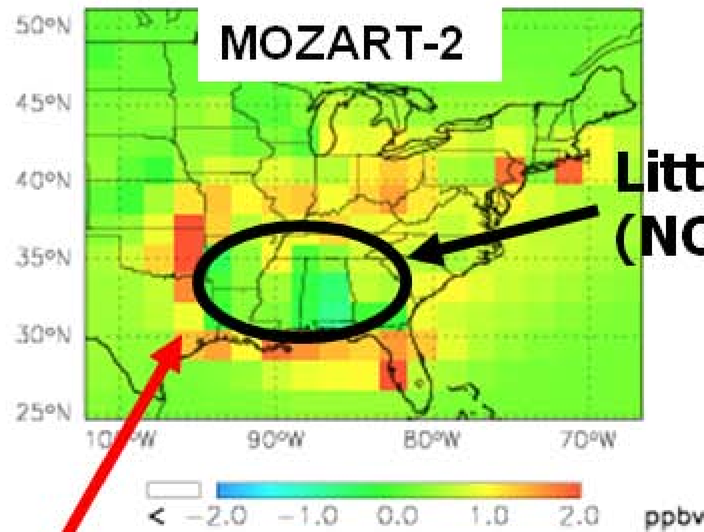
→ Land-use changes not presently considered in CTMs



Do these recent changes in isoprene emissions influence surface O_3? A Two-Model Perspective

Change in July 1-5 p.m. surface O_3

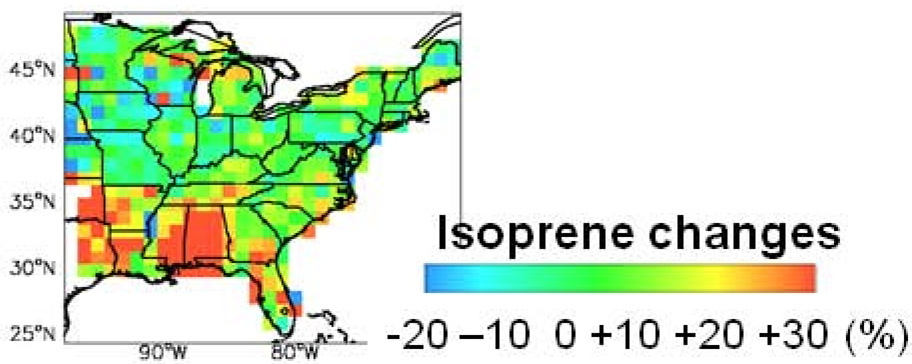




Little change (NO_x-sensitive)

O_3 decreases (same as previous result)

O_3 increases (1-2 ppbv)



Isoprene nitrates are not a NO_x sink in standard MOZART-2;

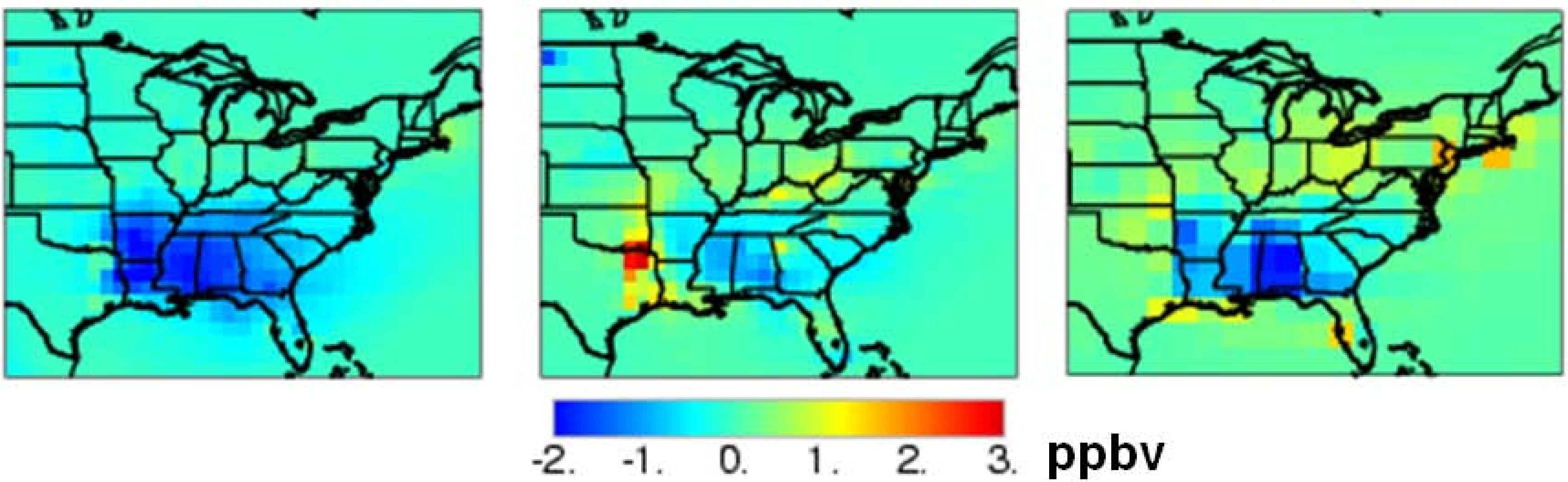
Results are sensitive to this assumption!

Chemical uncertainty: MOZART-2 shows similar results to GEOS-CHEM if isoprene nitrates are a NO_x sink

Change in July 1-5 p.m. surface O_3 (ppbv)
(due to isop emis changes from mid-1980s to mid-1990s)

With 12% yield of isoprene nitrates

GEOS-CHEM: GEIA | **GEOS-CHEM: Purves** | **MOZART-2: GEIA**



➡ **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 Eastern U.S. surface O_3
  2. how O_3 responds to both anthrop. and biogenic emission changes

  → Utility of satellite CH_2O columns?
  → New inventories (MEGAN, BEIS-3) more accurate?
  → Insights from aircraft campaigns?

- Recent isoprene increases may have reduced surface O_3 in the SE

  → Does this regime actually exist?
  → Fate of organic nitrates produced during isoprene oxidation?

Fiore et al., JGR, 2005