



Background Ozone in Surface Air: Origin, Variability, and Policy Implications

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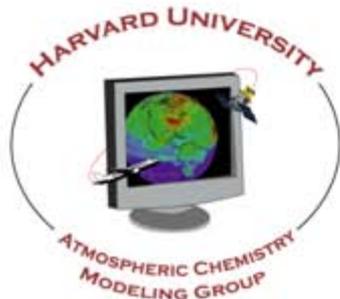
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What is the origin of tropospheric ozone?

Stratospheric O_3

Stratosphere

~12 km

$h\nu$

O_3

Free Troposphere

Hemispheric Pollution

Direct Intercontinental Transport

Boundary layer

(0-3 km)

NO_2

NO

OH

HO_2

VOC CH_4 , CO

NO_x

VOC

O_3

air pollution (smog)

air pollution (smog)

NO_x

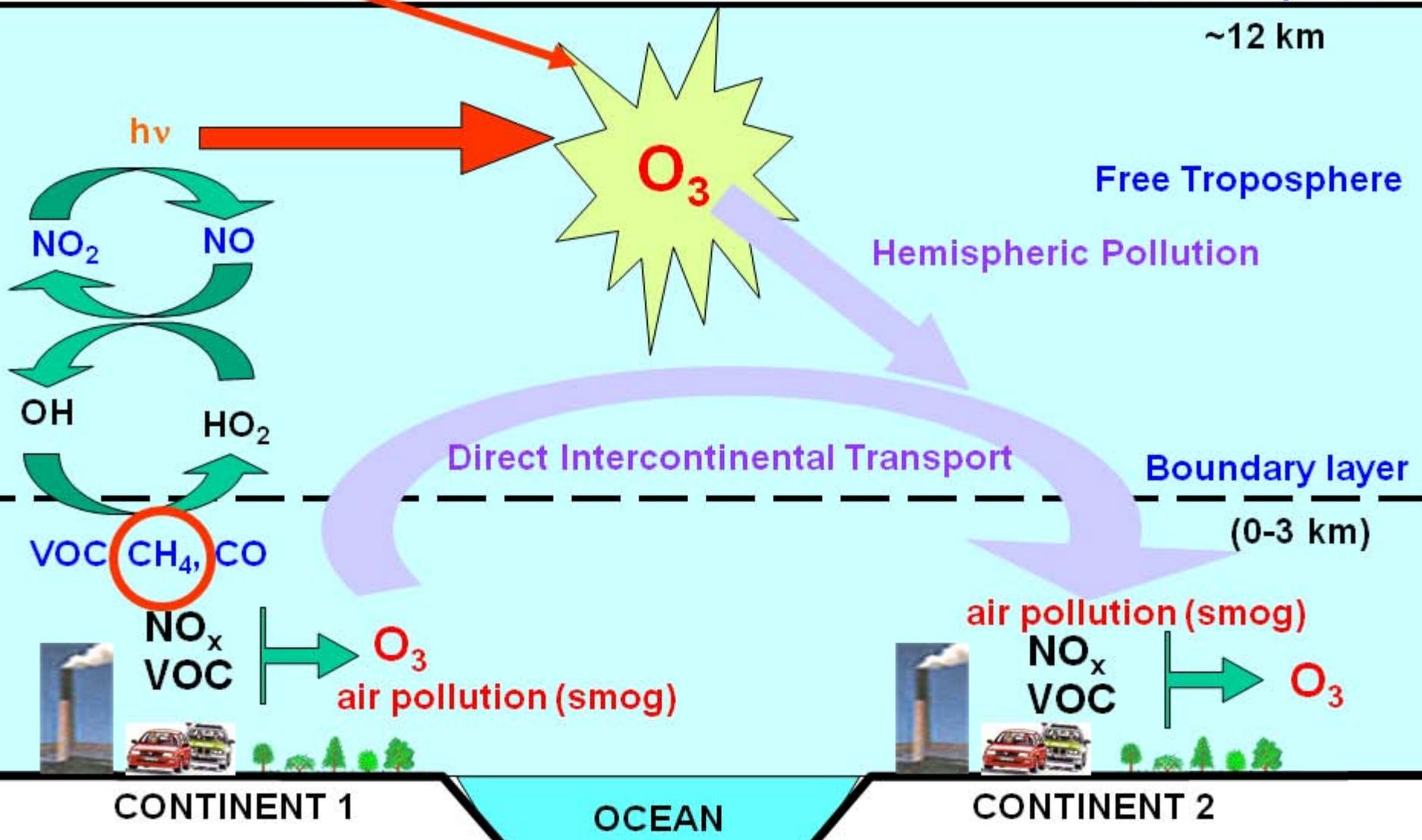
VOC

O_3

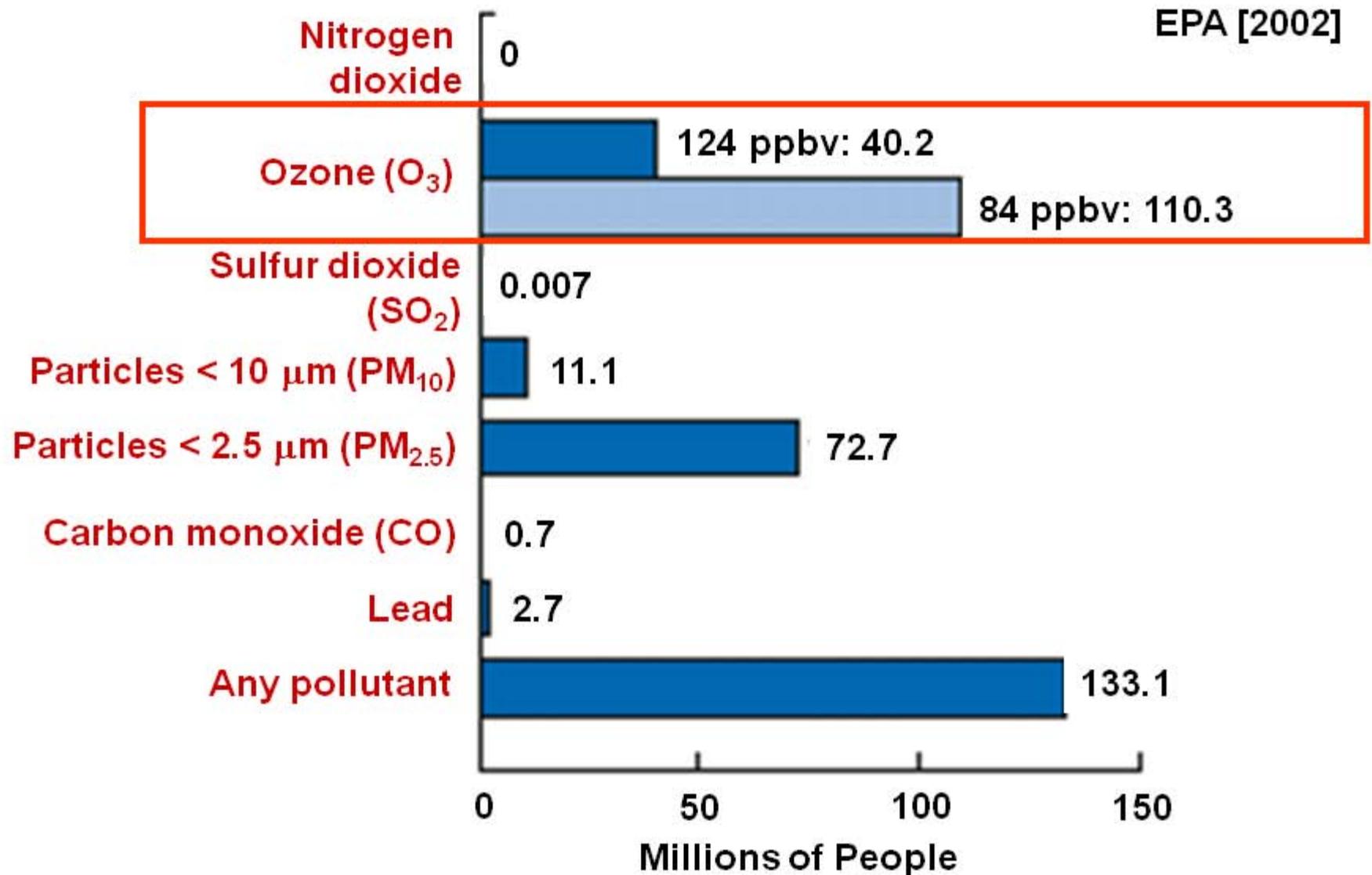
CONTINENT 1

OCEAN

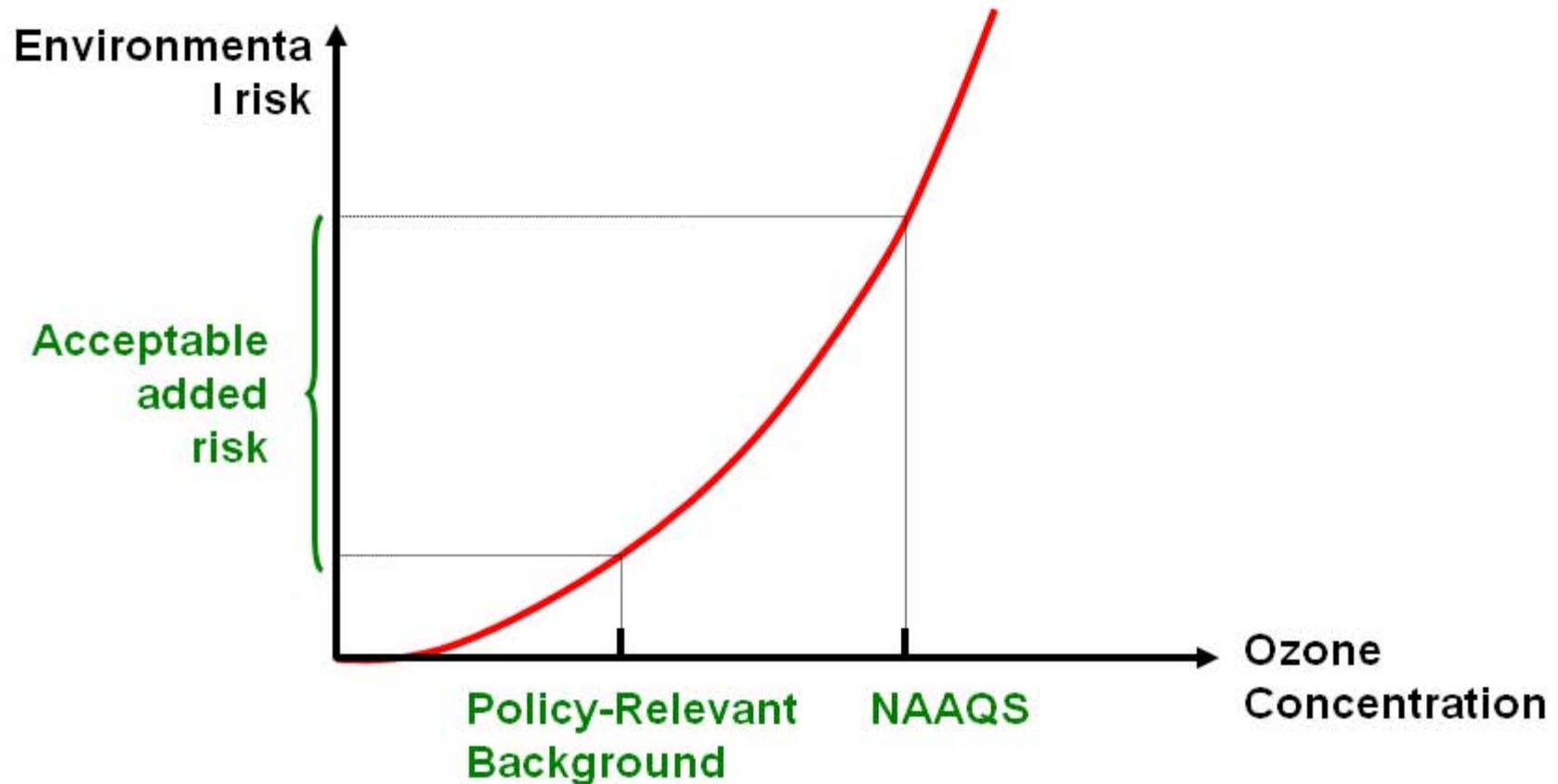
CONTINENT 2



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



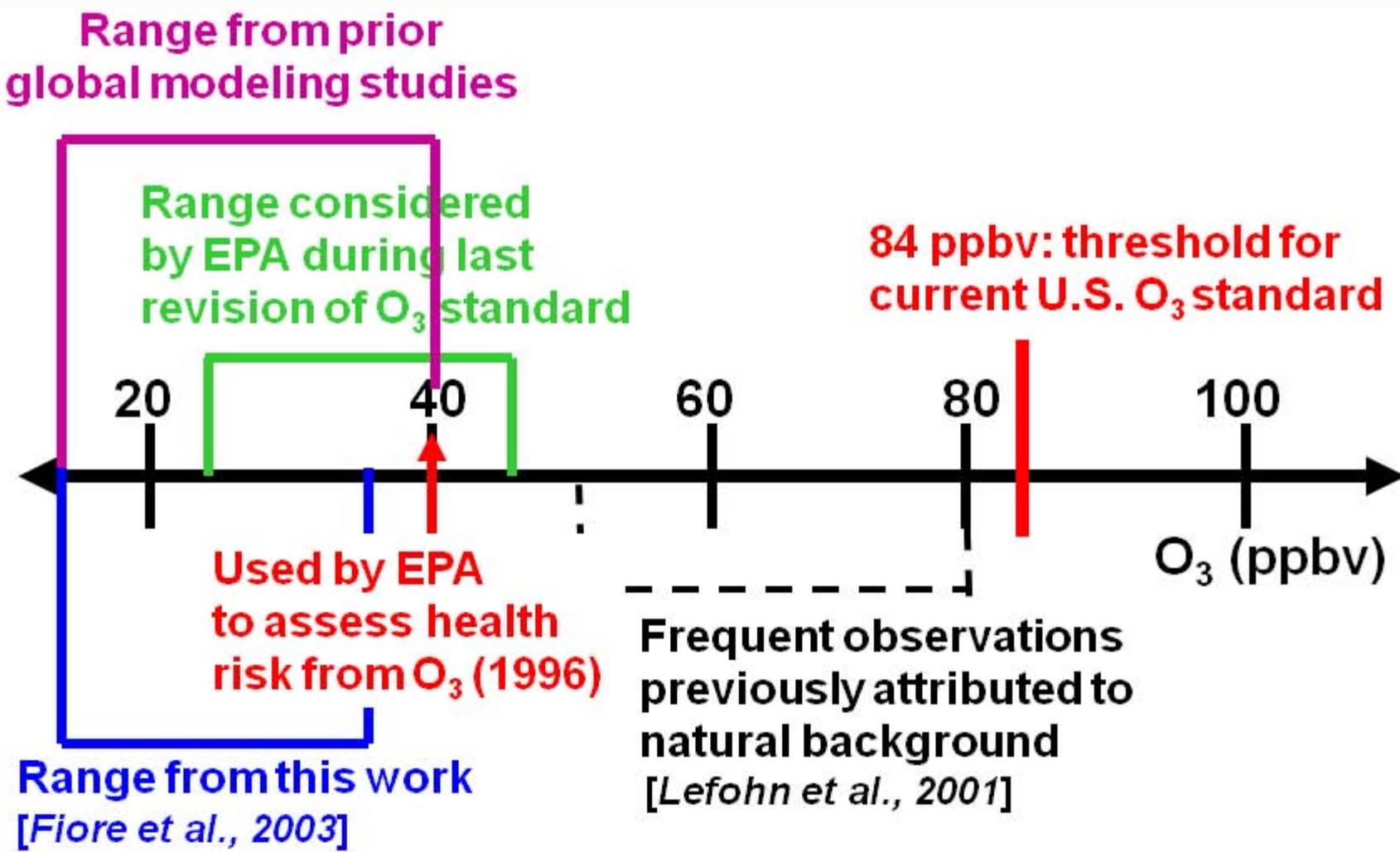
The Risk Increment Above the Background is Considered when setting the NAAQS for Ozone



→ Need a quantitative estimate for background ozone

→ EPA chose a constant value (40 ppbv) in previous review of O₃ standard

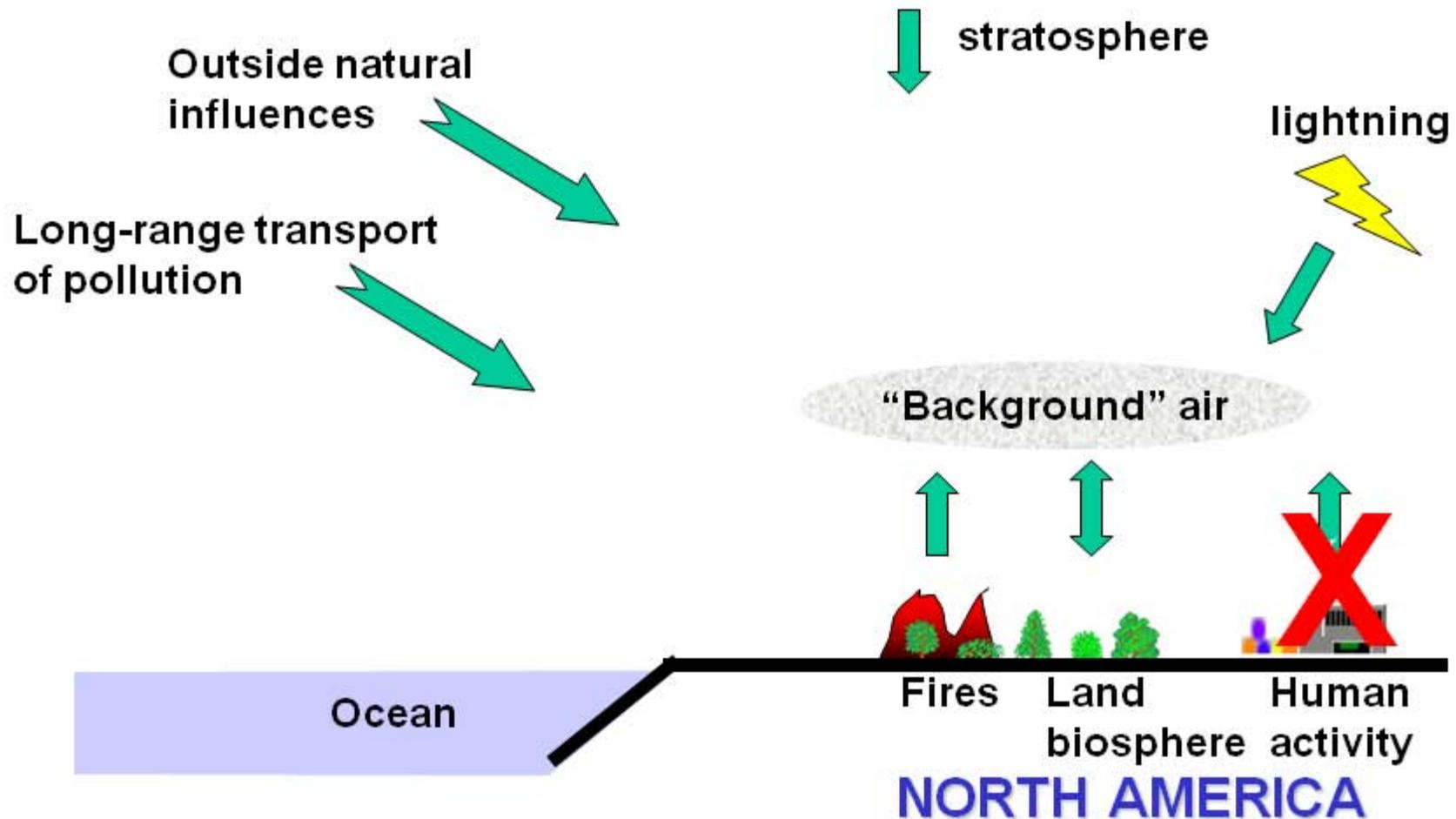
Range of "background O₃" estimates in U.S. surface air



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: Insights from two chemical transport models



GEOS-CHEM [*Bey et al., 2001*]

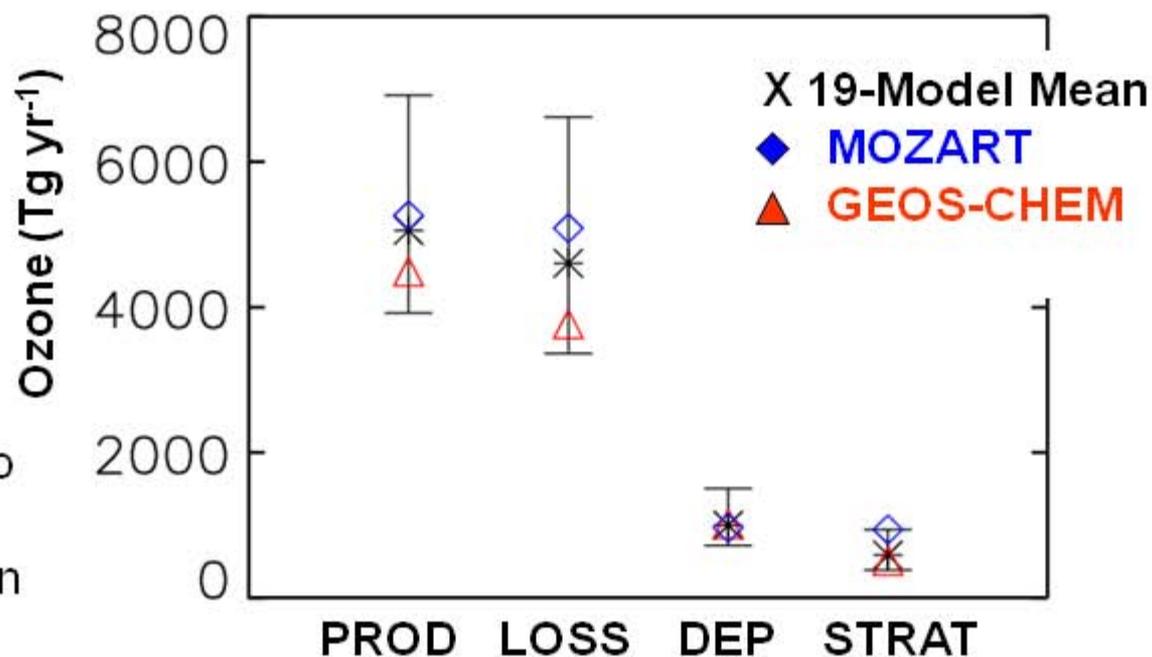
- GEOS-3 GMAO met
- 2°x2.5°; 30 σ -levels
- Synoz upper boundary



MOZART-2 [*Horowitz et al., 2003*]

- NCEP meteorology
- T62 (1.9°) ; 28 σ -levels
- Strat O₃ relaxed to climatology

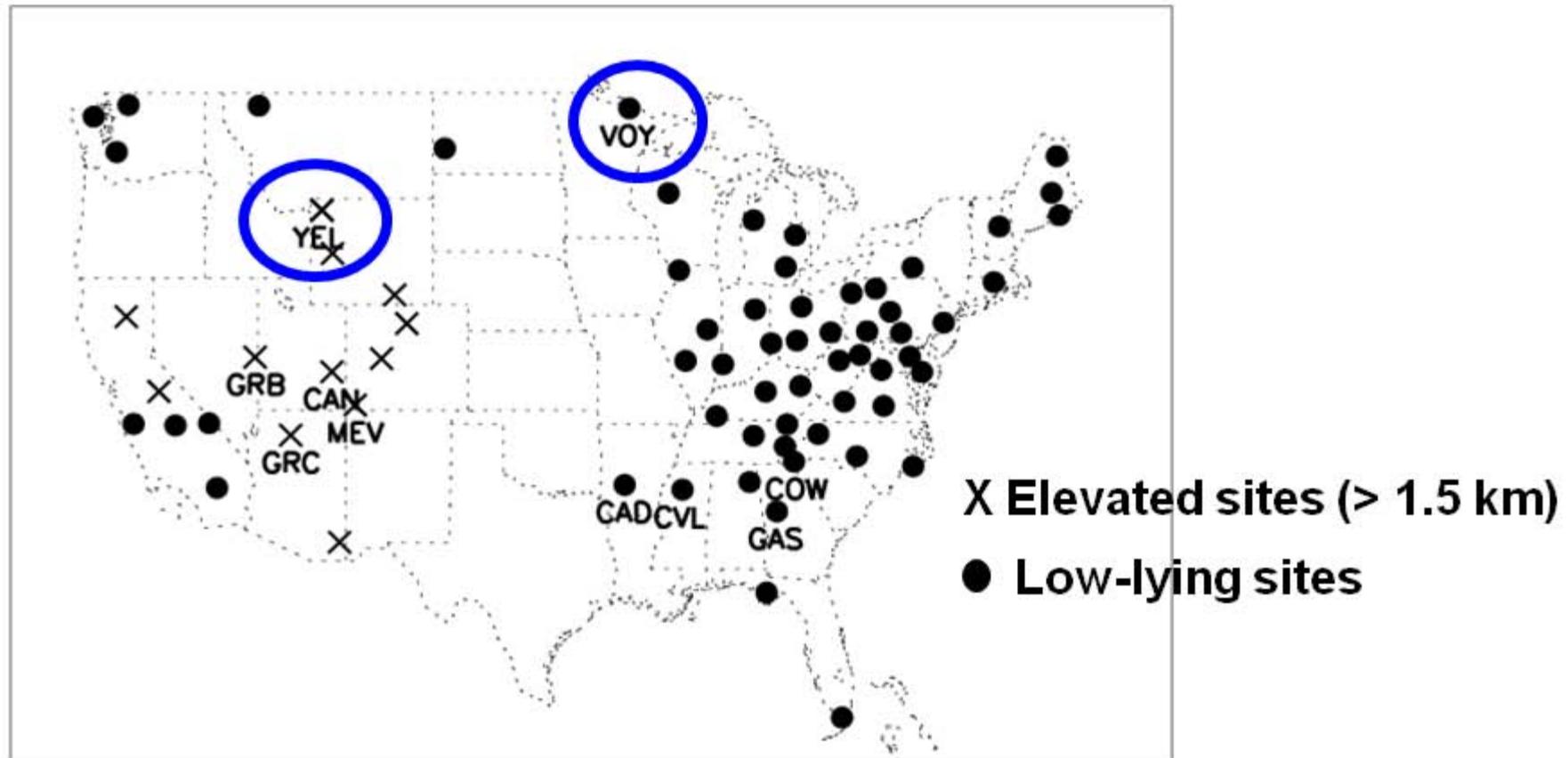
Ozone Budgets in IPCC-AR4 Tropospheric Chemistry Models



CHASER_CTM
CHASER_GCM
FRSGC
GEOS-CHEM
GFDL
GMCCM
GMDAO
gmigis
LLNL-IMPACT
LMDzINCA
LMDzINCAc
NCAR
STOCHEM_HadAM3
TM4
TM5
UM_CAM
MOZECH
STOCHEM_HadGEM
ULAQ

AR4 budgets c/o
Jérôme Drevet
David Stevenson
Frank Dentener

Approach: 2001 CASTNet Observations (EPA, NPS)



1. quantify PRB O₃ and its various sources
2. diagnose origin of springtime high-O₃ events at remote U.S. sites, previously attributed to natural, stratospheric influence

Case Study #1: Voyageurs National Park, Minnesota (May-June 2001)

Lefohn et al. [2001] suggest a stratospheric source as the likely origin of high-O₃ events frequently observed in June

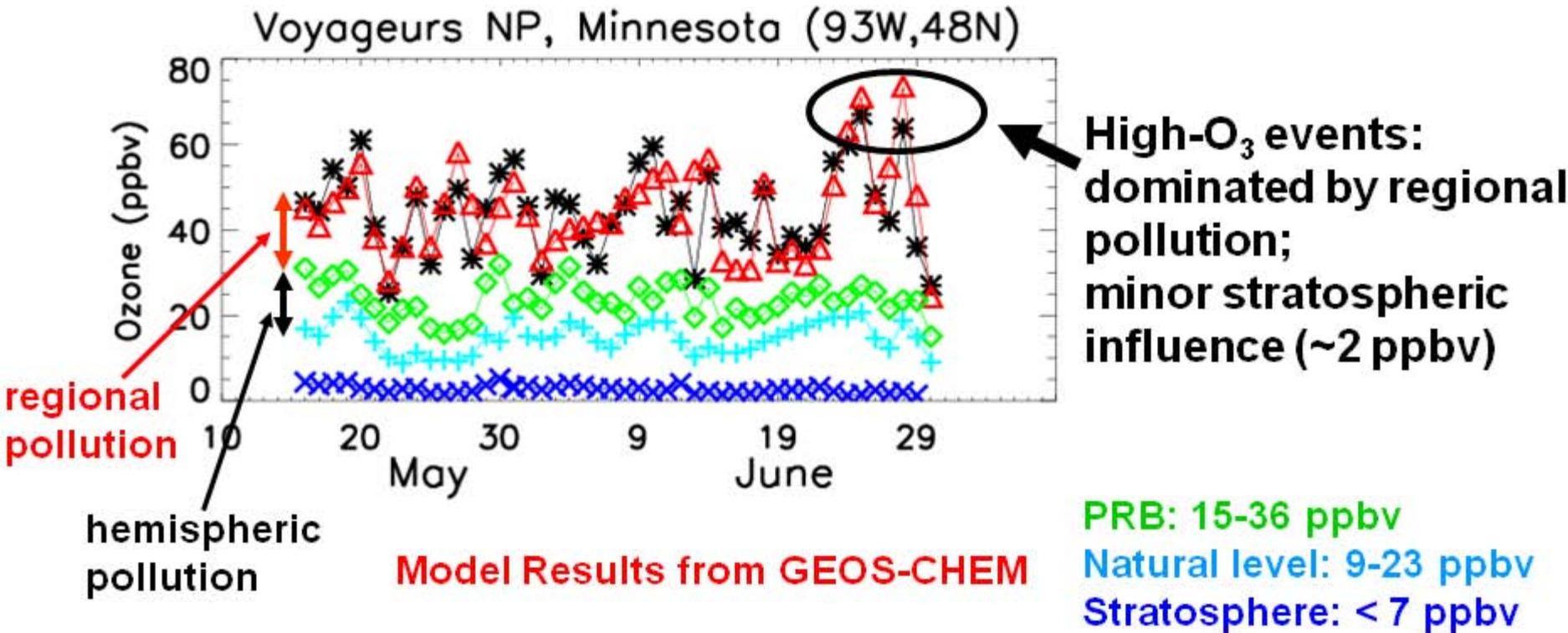
* CASTNet observations

▲ Model } Δ = Regional pollution

◆ Background } Δ = Hemispheric pollution

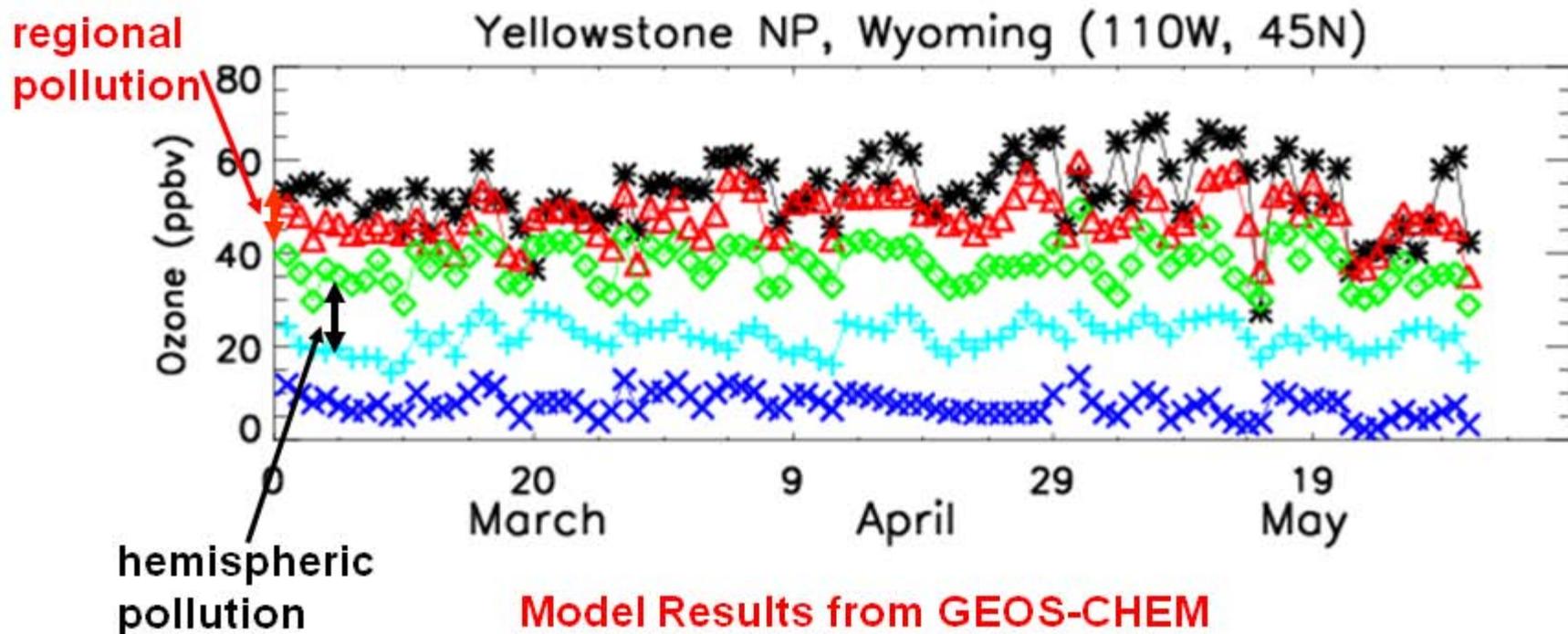
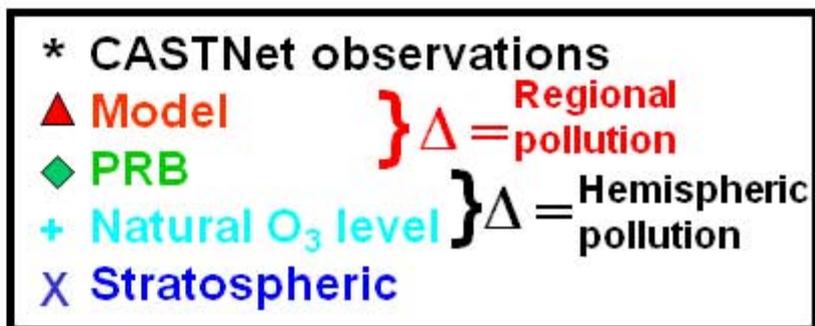
+ Natural O₃ level

X Stratospheric



Case Study #2: Yellowstone National Park, Wyoming (March-May 2001)

Frequent high-O₃ events previously attributed to natural, stratospheric source
[Lefohn et al., 2001]



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

Both models show that PRB ozone is higher at high-altitude site

* CASTNet observations ▲ GEOS-CHEM Model ▲ MOZART Model
 ◆ GEOS-CHEM PRB ◆ MOZART PRB

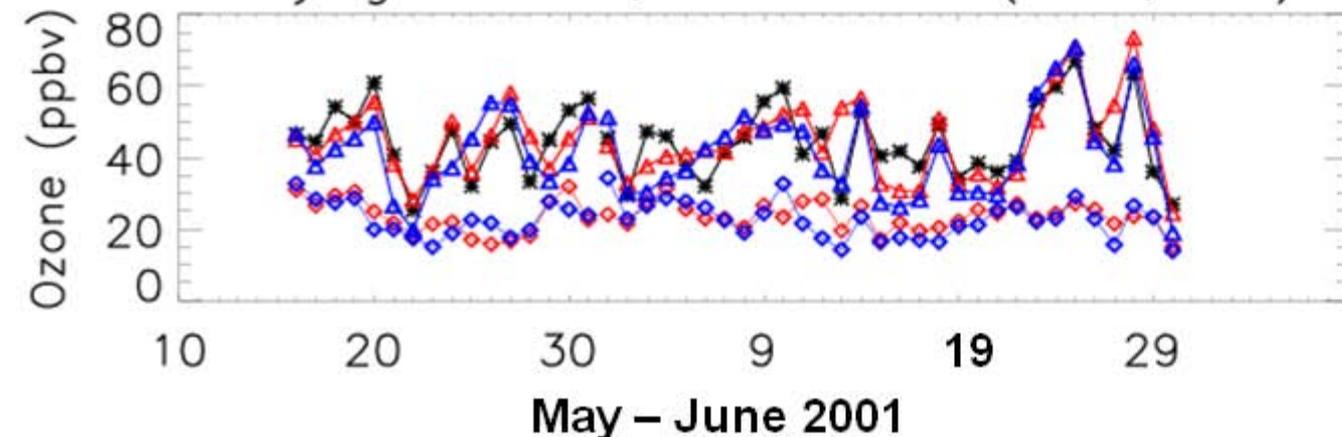
Voyageurs NP, Minnesota (93W,48N)

Elevation: 430 m

PRB Range; mean

15-36; 25

14-34; 23



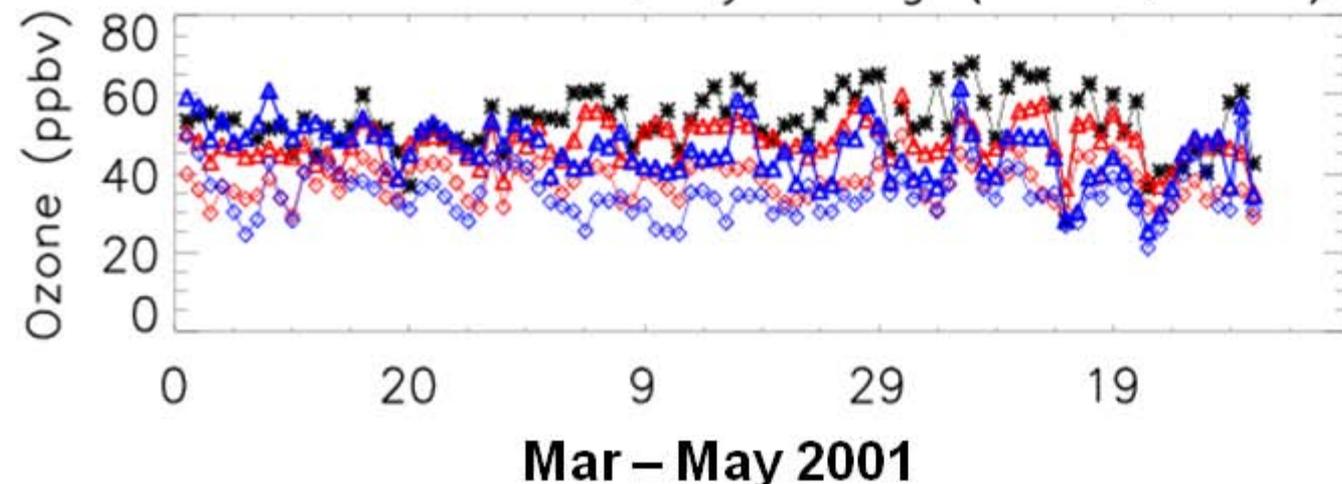
Yellowstone NP, Wyoming (110W, 45N)

Elevation: 2470 m

PRB Range; mean

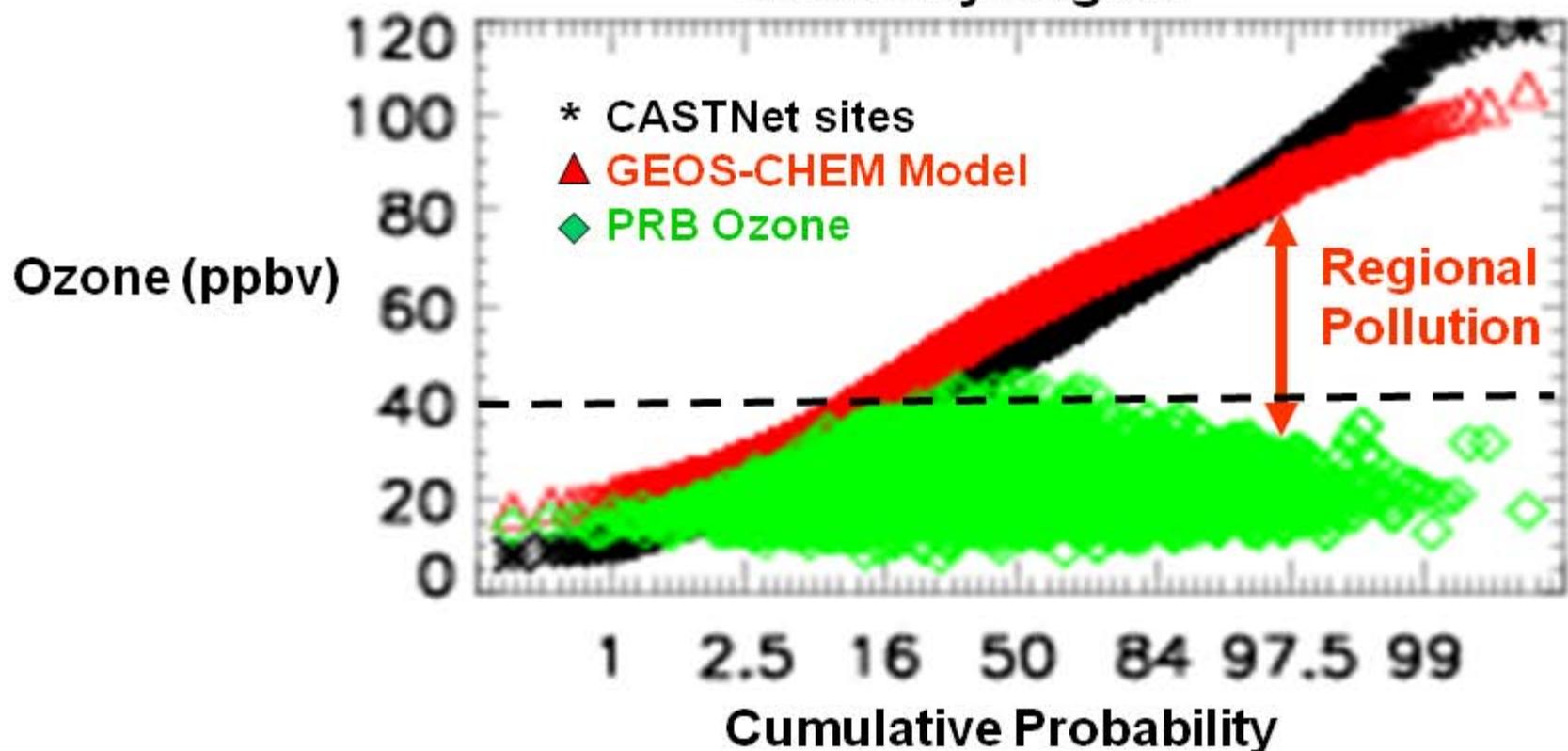
29-50; 38

21-56; 35



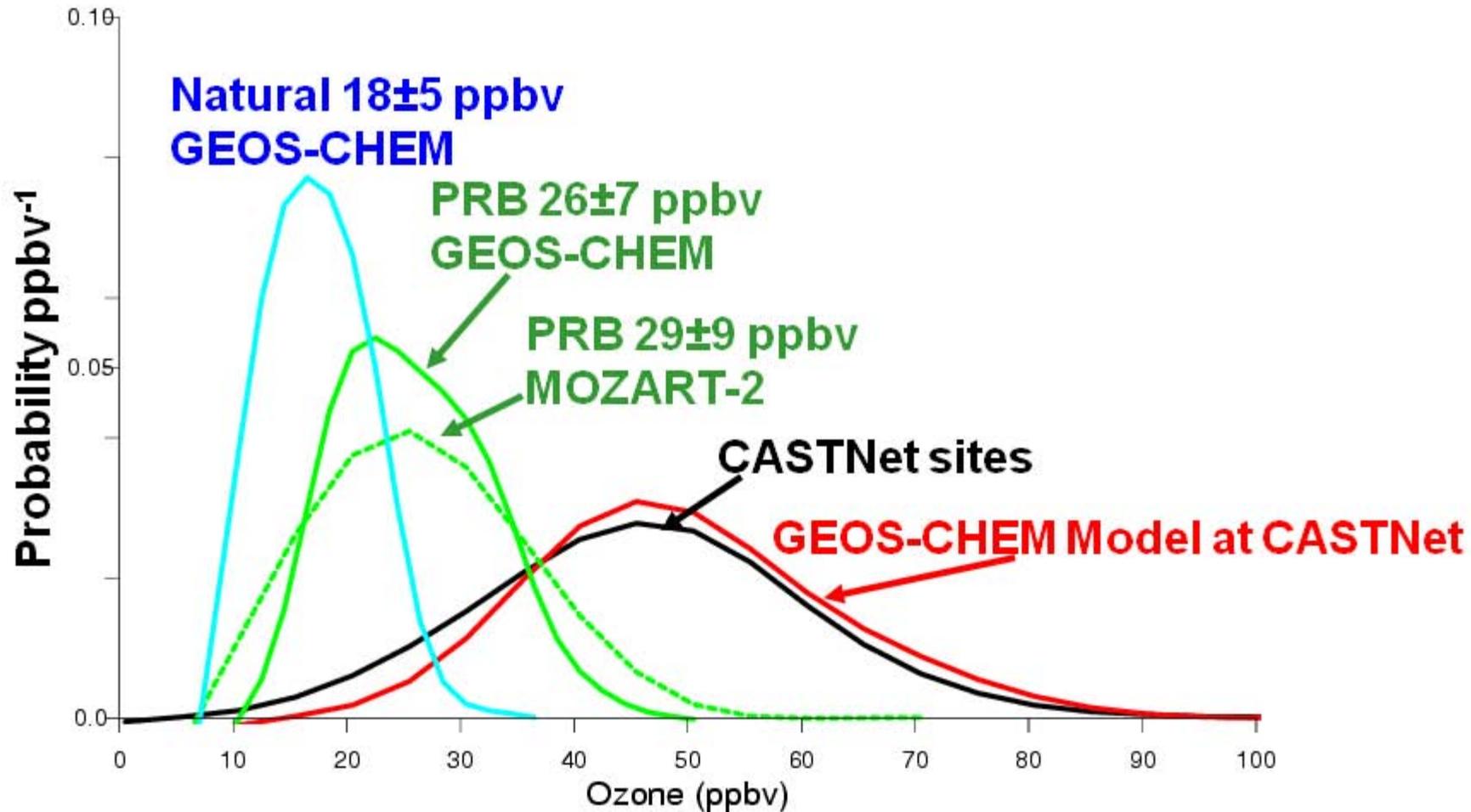
PRB ozone is lower under polluted conditions:
typically below 25 ppbv

Daily mean afternoon O₃ at 58 low-elevation U.S. CASTNet sites
June-July-August



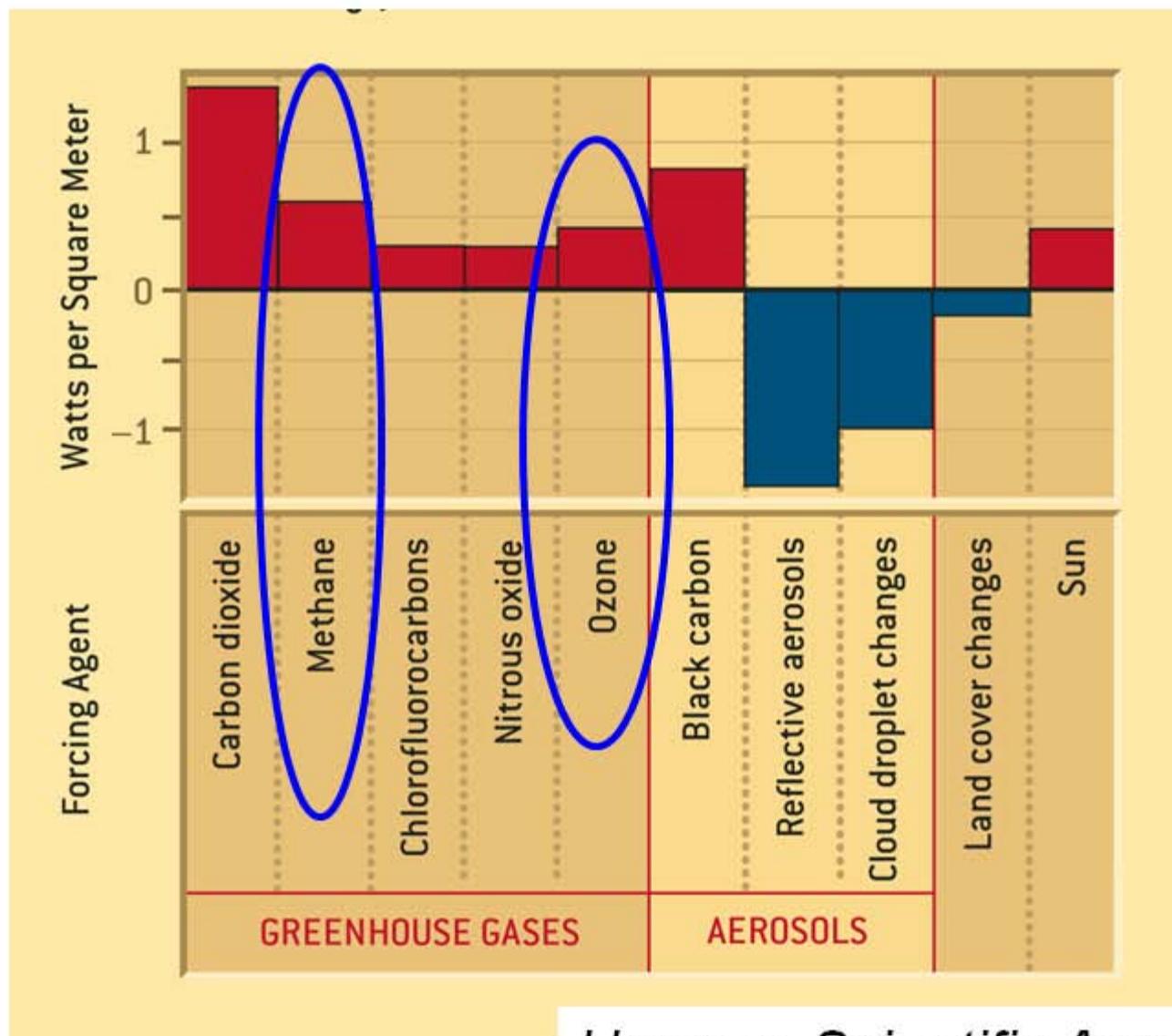
→ Assuming constant 40 ppbv background underestimates health risks on most polluted days

Compiling daily afternoon (1-5 p.m. mean) surface ozone from all
CASTNet sites for March-October 2001:
PRB ozone is typically 20-35 ppbv



- Hemispheric Pollution enhances U.S. PRB by 8 ± 4 ppbv
- Prior work suggests 6 ppbv from anthrop. CH_4 on average

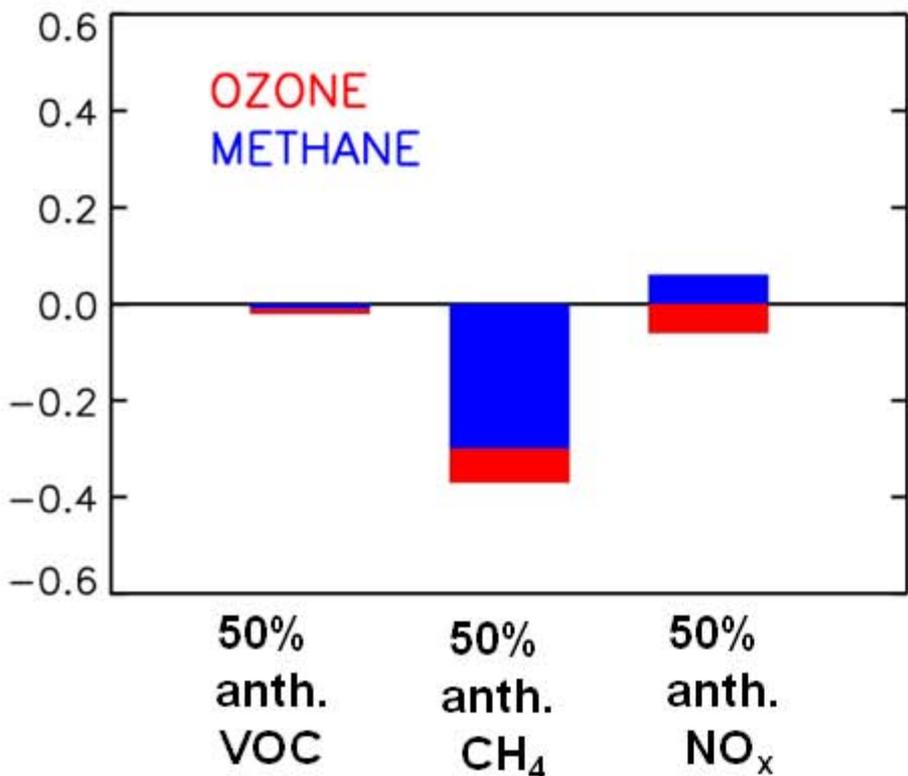
Radiative Forcing of Climate from Preindustrial to Present: Important Contributions from Methane and Ozone



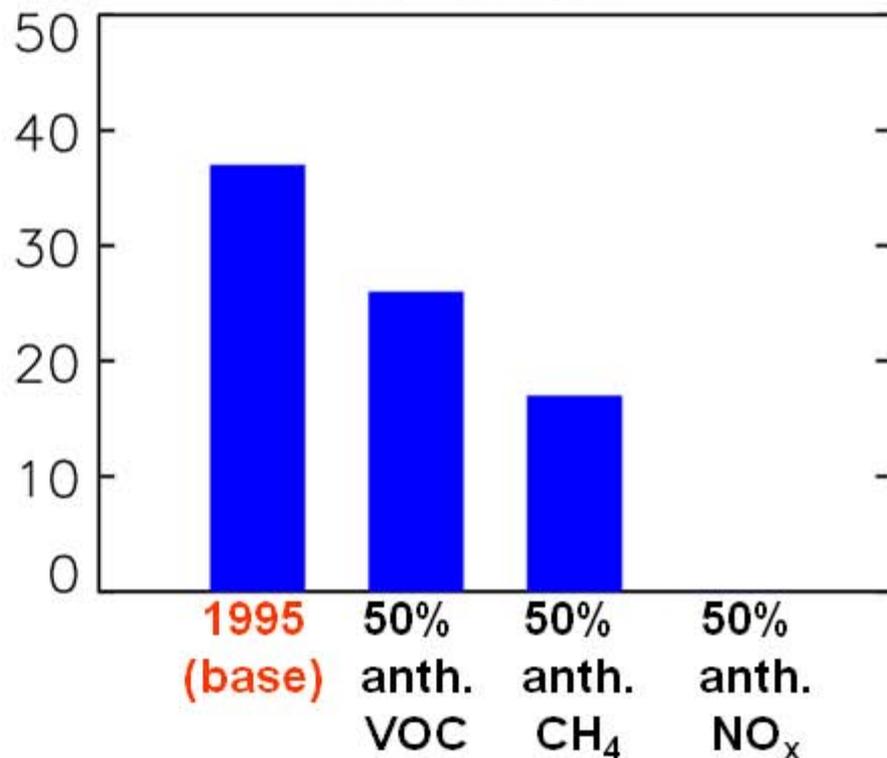
Hansen, Scientific American, 2004

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

CLIMATE: Radiative Forcing (W m^{-2})

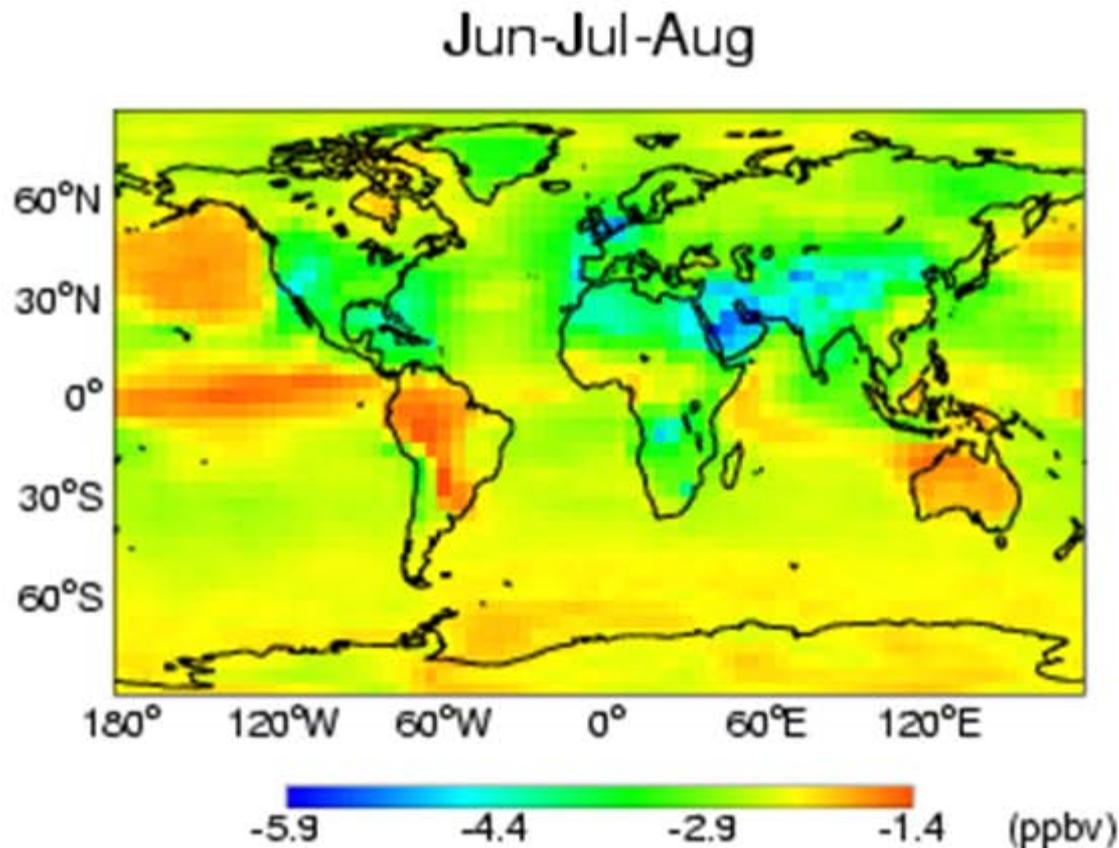


AIR QUALITY: Number of U.S. summer grid-square days with $\text{O}_3 > 80$ ppbv



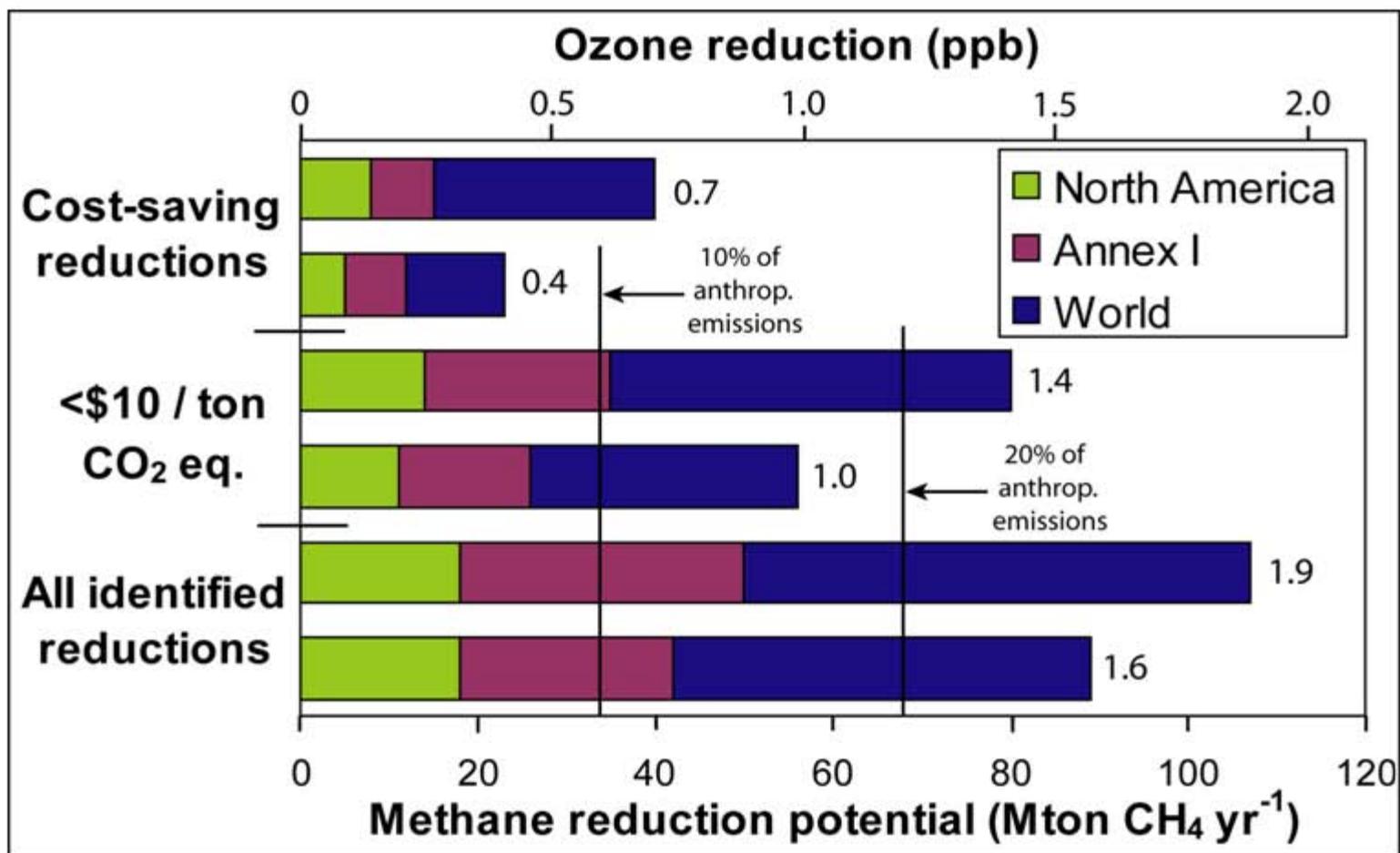
→ Methane links air quality and climate via background ozone

Response of Global Surface Ozone to 50% decrease in global methane emissions (actually changing uniform concentration from 1700 to 1000 ppbv)



- Ozone decreases by 1-6 ppb
- ~3 ppb over land in US summer
- ** ~60% of reduction in 10 yr; ~80% in 20 yr.

How Much Methane Can Be Reduced?

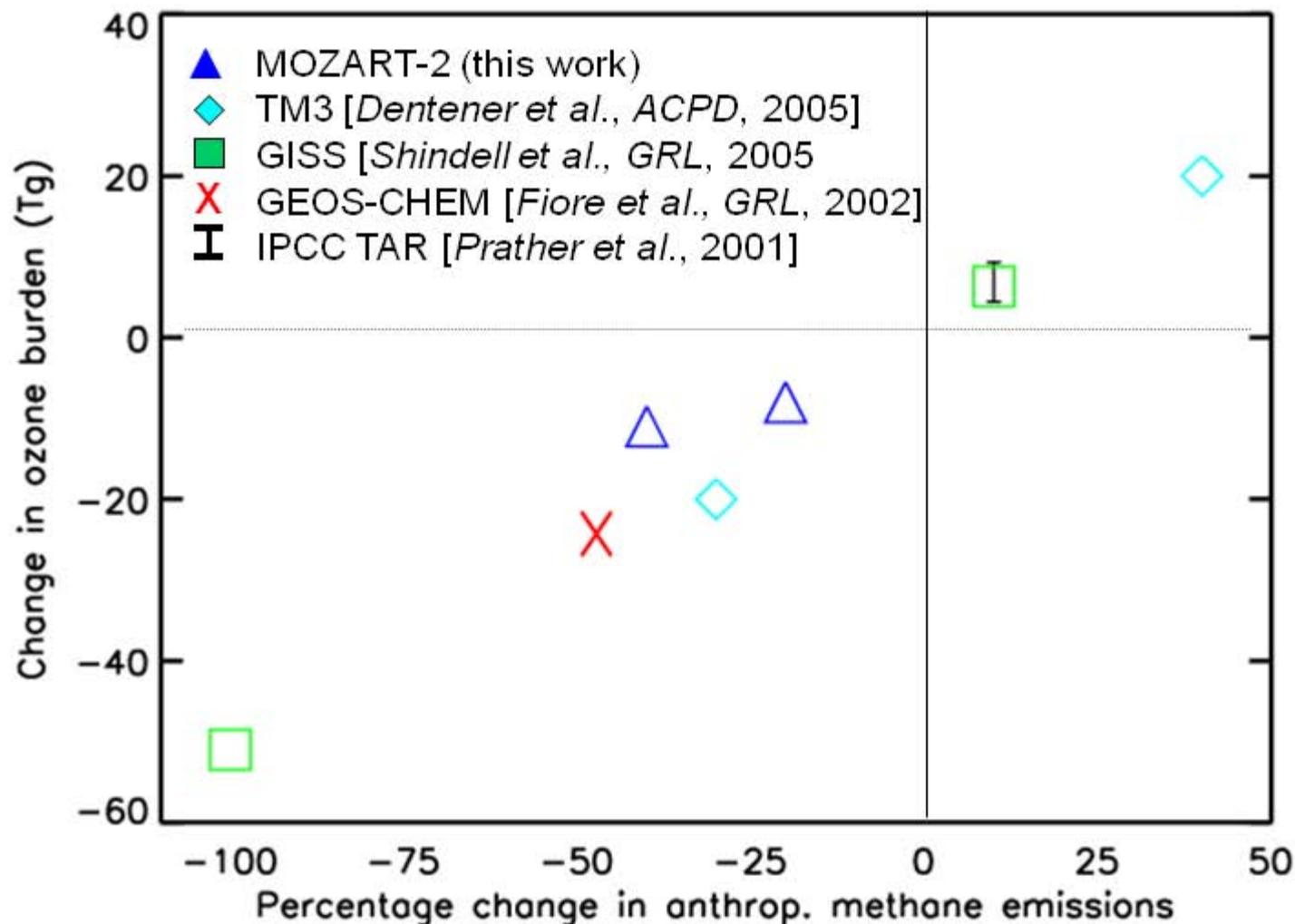


Top bar: IEA (2003), for 5 industrial sectors.

Lower bar: EPA (2003), for 4 industrial and 1 agricultural sector.

Comparison: Clean Air Interstate Rule (proposed) reduces 0.86 ppb over the eastern US, at \$0.88 billion yr⁻¹, through NO_x control.

Tropospheric ozone response to anthropogenic methane emission changes is fairly linear



Shindell et al., 2005 report that tropospheric ozone responds linearly to 10, 25, 50, 75, 100% decreases in anthropogenic CH₄

Methane Emissions in EDGAR inventory early 1990s; Tg CH₄ yr⁻¹

Anthropogenic

Natural

Energy, landfills, wastewater	95
Ruminants	93
Rice	60
Biomass burning	86
Ocean	10
Biogenic	204
TOTAL	547

→ Are ozone decreases independent of CH₄ source location?

**Cut Global Anthropogenic by 40%: (1) All in Asia
(2) Everywhere in the globe**

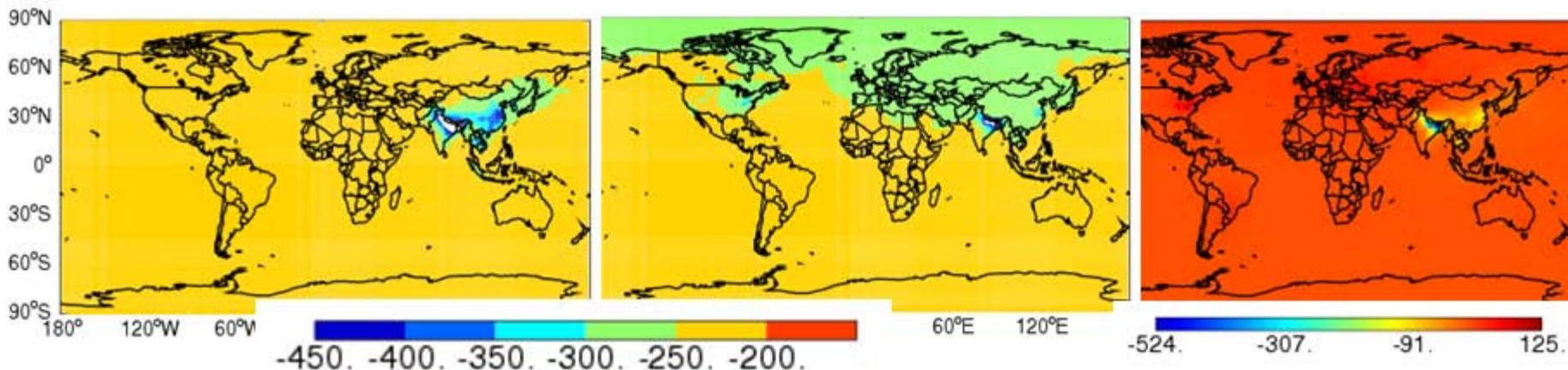
July 2000 surface O₃ change mainly independent of CH₄ source location, slightly larger response in source region
(Transient simulations with EDGAR 1990 emissions, beginning 1990)

Zero anthrop. CH₄ emissions from Asia

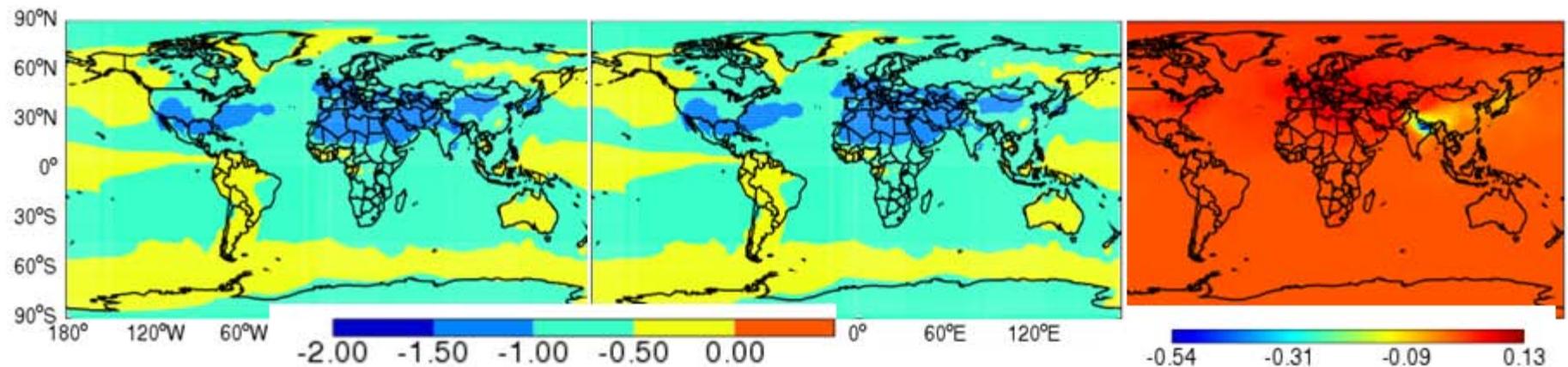
Global 40% decrease in anthrop. CH₄ emis.

Zero Asia – global 40%

METHANE



OZONE



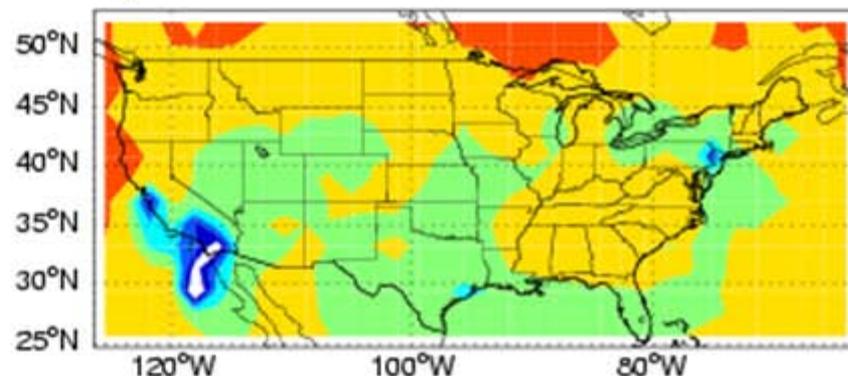
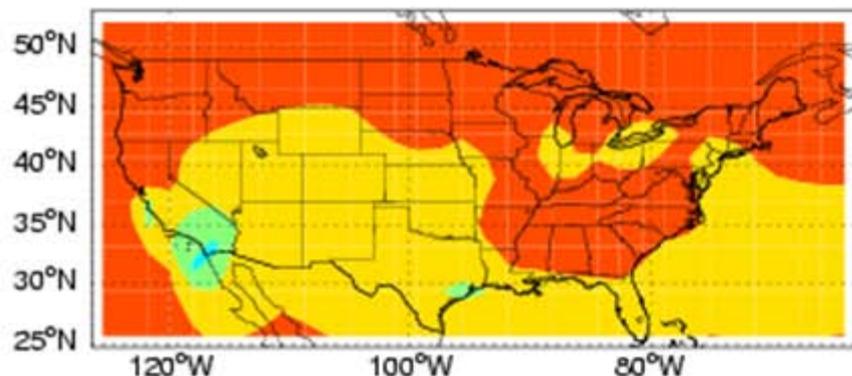
U.S. Surface Afternoon Ozone Response in Summer also independent of methane emission location

MEAN DIFFERENCE

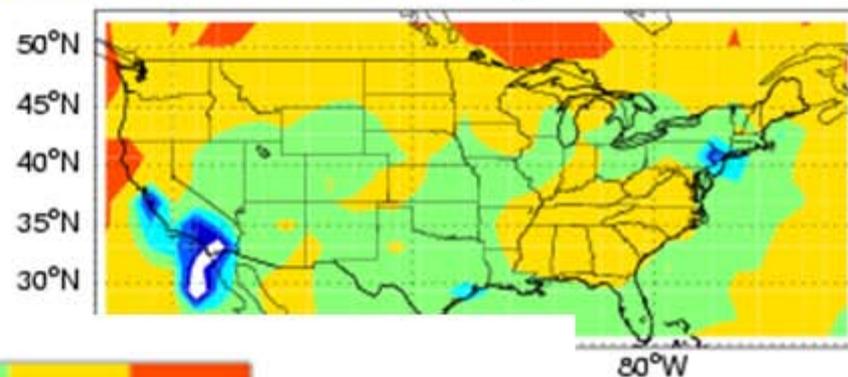
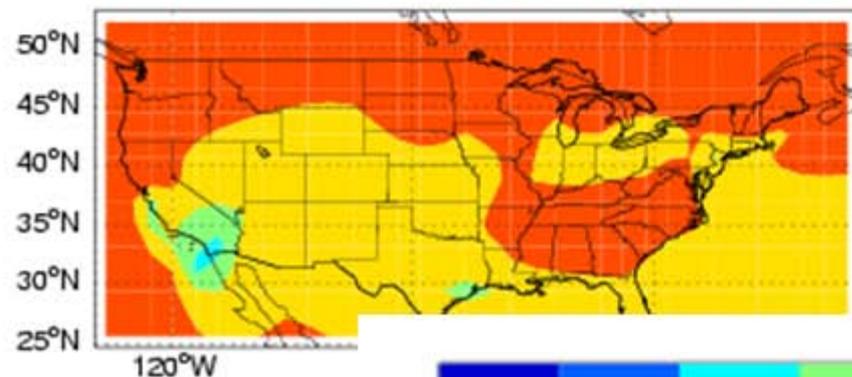
MAX DIFFERENCE

(Max daily mean afternoon JJA)

NO ASIAN CH₄



GLOBAL 40% DECREASE IN ANTHROP. CH₄



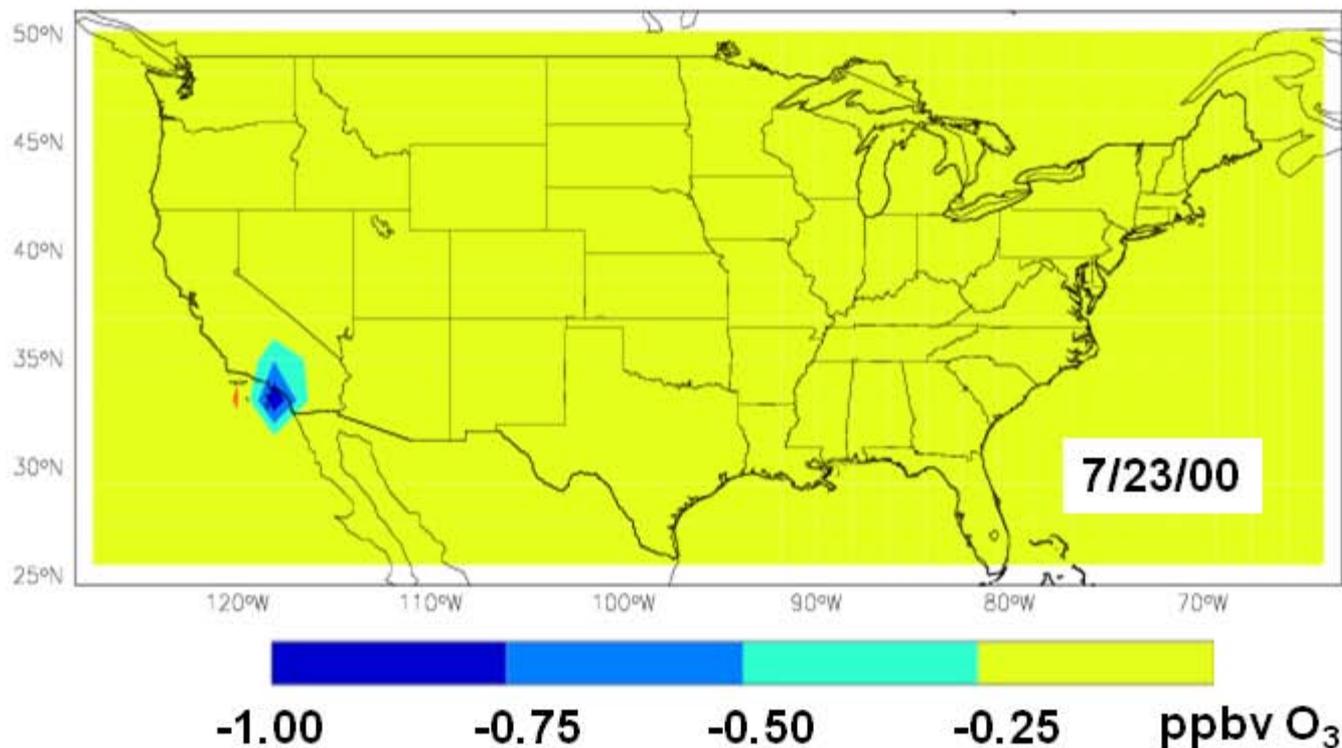
-3.5 -3.0 -2.5 -2.0 -1.5 -1.0

ppbv

→ Stronger Sensitivity in NO_x-saturated regions (Los Angeles)

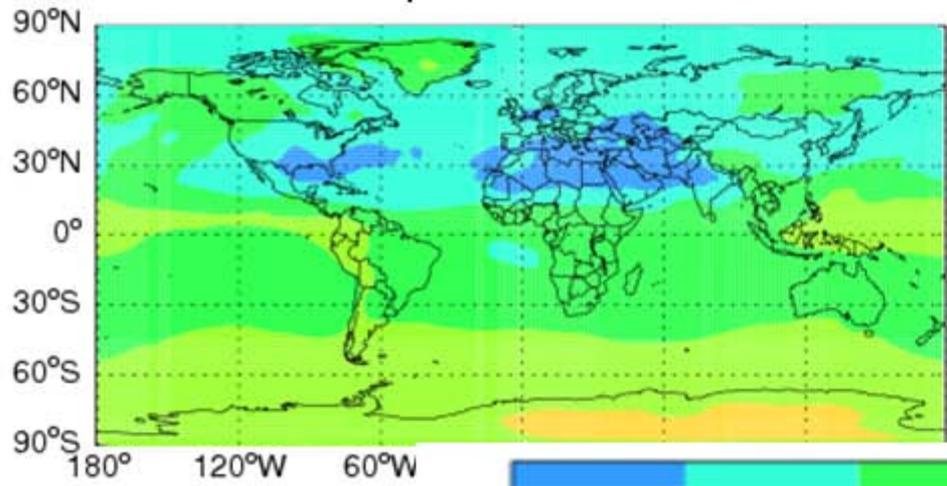
Local CH₄ oxidation contributes up to ~30% of the total decrease in surface O₃ from lowering CH₄ concentrations in a NO_x-saturated region

Change in MOZART-2 surface afternoon (1-5 p.m.) O₃ concentration after 1 day, when a 300 ppbv decrease in CH₄ concentrations is imposed below 800 hPa in Los Angeles

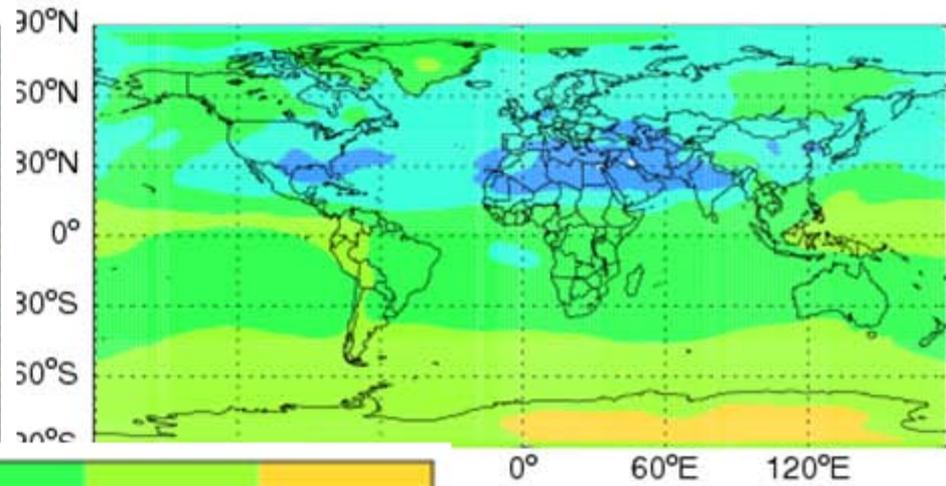


CLIMATE IMPACTS: Change in July 2000 Trop. O₃ Columns (to 200 hPa)

40% decrease in global anthrop. CH₄ emissions

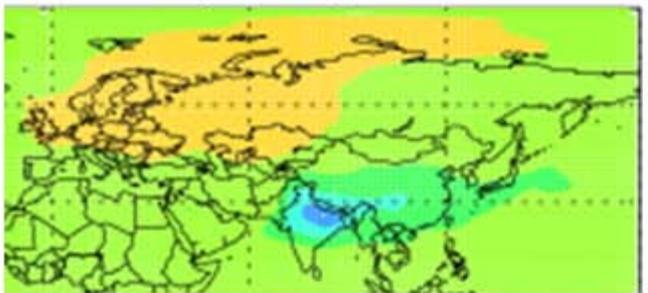


Zero CH₄ emissions from Asia (= 40% decrease in global anthrop.)



-1.00 -0.80 -0.60 -0.40 -0.20 Dobson Units

No Asia – (40% global decrease)

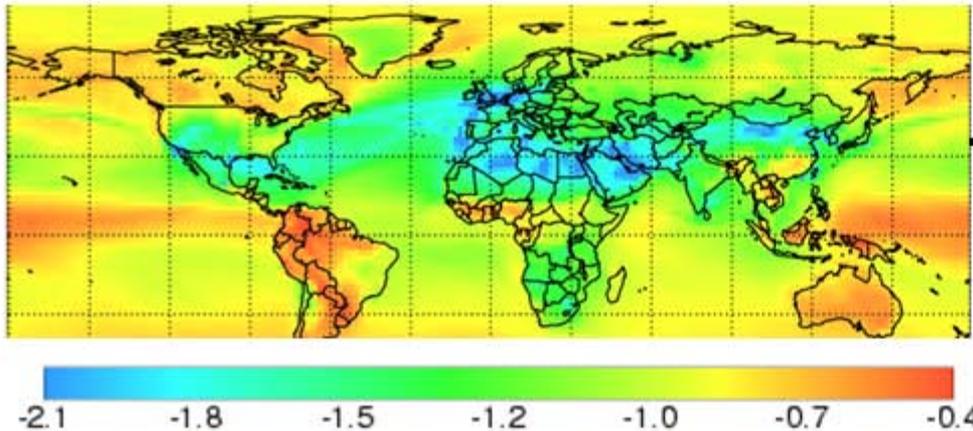


Tropospheric O₃ column response is independent of CH₄ emission location except for small (~10%) local changes

-0.15 -0.10 -0.05 -0.02 0.02 DU

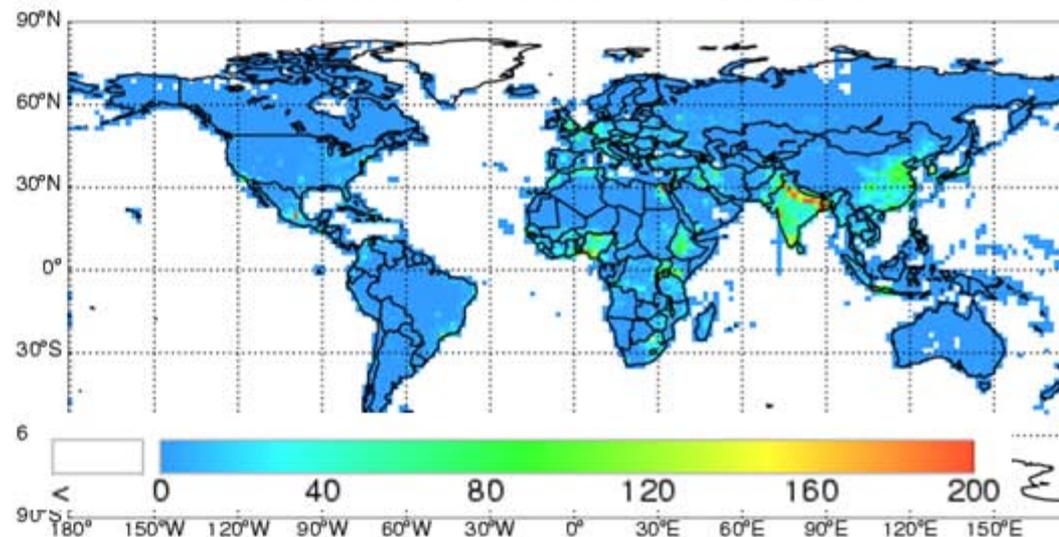
AIR POLLUTION IMPACTS: 2030 Avoided Premature Mortalities (A2 scenario - 65 Tg CH₄ emissions)

Change in 8-hr summer surface ozone from a 20% decrease in global anthrop. methane emissions



Assume 25 ppb threshold,
CH₄ reductions starting in 2000

Avoided Mortalities in 2030



Reducing anthrop. CH₄ emis.
by 20% (starting in 2000):

-- decreases global surface
ozone (~1 ppbv in populated
areas)

-- prevents ~34,000 premature
deaths in 2030

West et al., 2005

Conclusions

- 1. Policy-Relevant Background** in surface air over the United States:
 - **25 ± 5 ppbv** at low-elevation sites in summer
 - varies with season, altitude; anti-correlated with high domestic O_3
 - **8 ± 4 ppbv** from hemispheric pollution ($\sim 5-7$ ppbv from anthr. CH_4)
 - **40 ppbv** previously used by EPA underestimates health risks
 - international negotiations to reduce hemispheric background would facilitate compliance with more stringent standards
- 2. 20% reductions in anthrop. methane emissions possible now:**
 - reduce surface ozone globally by ~ 1 ppbv in populated regions
 - lower global radiative forcing by $\sim 0.13 \text{ W m}^{-2}$
 - avoid 34,000 premature mortalities in 2030
- 3. Climate and air quality benefits from controls on anthropogenic methane emissions are largely independent of source location**
 - small enhancements ($\sim 10\%$) in source region
 - enhanced surface ozone response (up to $\sim 30\%$ total response) to CH_4 changes in NO_x -saturated regions