



# Something wicked this way comes: impact of ozone on air quality and climate.

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# Ozone: The New Standard



## **EPA Strengthens Ozone Standards to Protect Public Health/Science-based standards to reduce sick days, asthma attacks, emergency room visits, greatly outweigh costs**

Release Date: 10/1/2015

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“Based on extensive scientific evidence on effects that ground-level ozone pollution, or smog, has on public health and welfare, the U.S. Environmental Protection Agency (EPA) has strengthened the National Ambient Air Quality Standards (NAAQS) for ground-level ozone to 70 parts per billion (ppb) from 75 ppb to protect public health.”



# Cacophony Chorus



## American Lung Association Responds to EPA Ozone Standard Update, Impact on Public Health

(October 1, 2015) - Washington, DC Today, in response to the updated National Ambient Air Quality Standards for ozone announced by the Obama Administration, Harold P. Wimmer, National President and CEO of the American Lung Association, issued the following statement:

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The EPA's independent scientific advisors reviewed the evidence and concluded that a level of 60 ppb would provide more public health protection than a standard of 70 ppb. Furthermore, leading medical and health organizations and more than 1,000 health and medical professionals continuously voiced strong support for a standard at 60 ppb.



# Cacophony Chorus



NATIONAL ASSOCIATION OF  
Manufacturers

## Ozone Regulations

It's Time For Congress To Act!

“Today, the Obama Administration finalized a rule that is overly burdensome, costly and misguided....,make no mistake: the new ozone standard will inflict pain on companies that build things in America—and destroy job opportunities for American workers. “

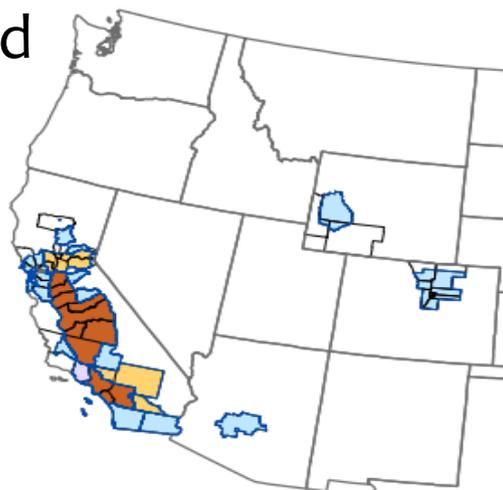
**WHY ARE WE  
CHANGING  
THE OZONE  
STANDARD  
NOW?**



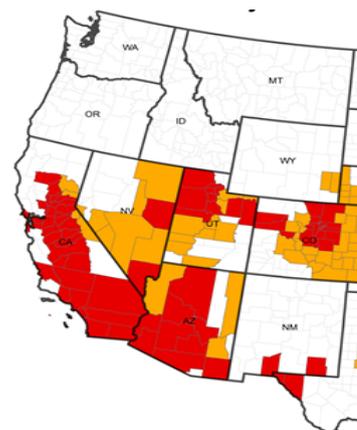
# Who will be effected?

Extended ozone nonattainment areas in the western US after the US ozone standard was lowered in last October

2008 standard  
(75 ppbv)



Updated standard  
(70 ppbv)

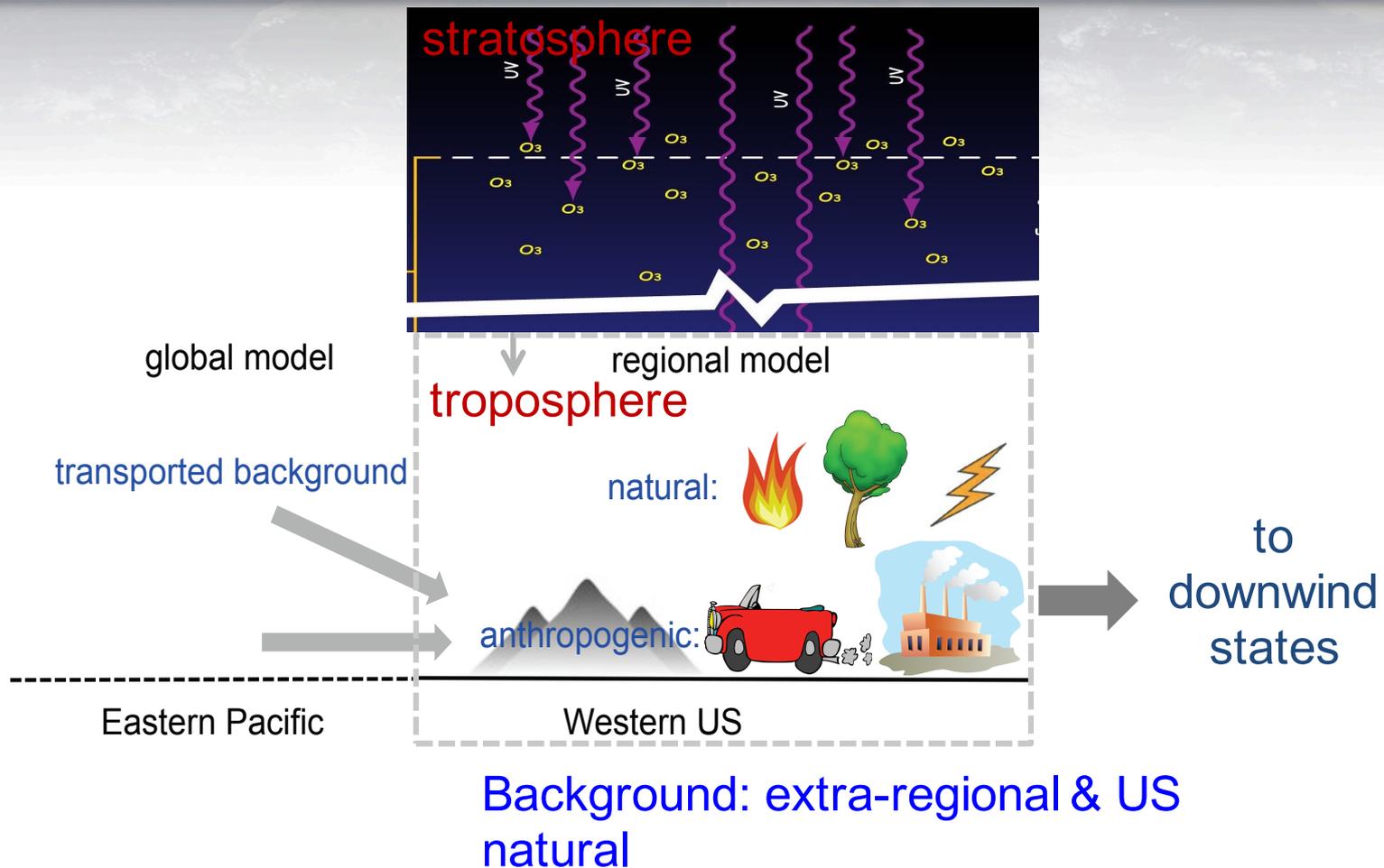


Monitored  
Unmonitored

- A lot of these non-attainment areas are currently not monitored by EPA
- Are they breathing healthy air?



# Every breath you take—and where it came from.



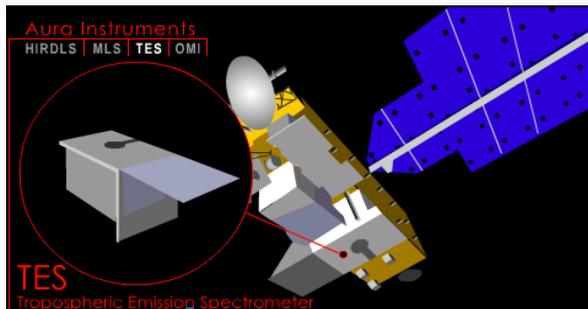
- The air that you breath came in part from somewhere else.
- The Clean Air Act has provisions to address noncompliance due to local versus non-local sources.
- How do you *attribute* the air that you breath?



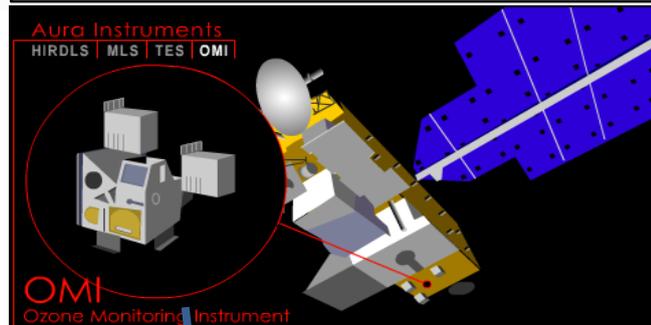
# Multiscale Assimilation System

## Aura Satellite

TES level 2 ozone profiles



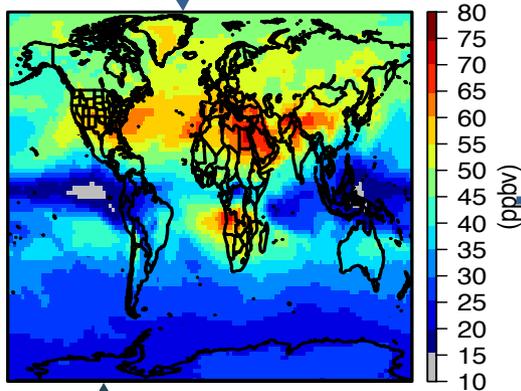
OMI NO<sub>2</sub> tropospheric columns



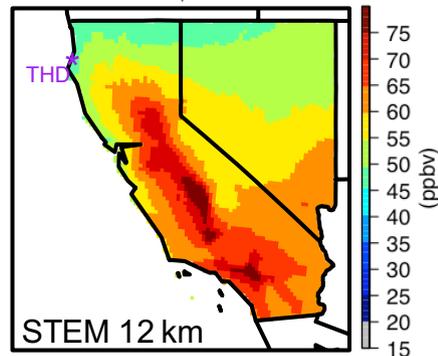
3D-Var  $\mathbf{H}_i(\bullet) = \mathbf{x}_a + \mathbf{A}_i(\bullet - \mathbf{x}_a)$

4D-Var

GEOS-Chem  
adjointv34, 2°x2.5°



top/lateral  
boundary  
conditions



grid-specific  
scaling factors  
on NO<sub>x</sub>  
emissions

co-scale  
VOCs

updated chemical  
fields (total and  
background)

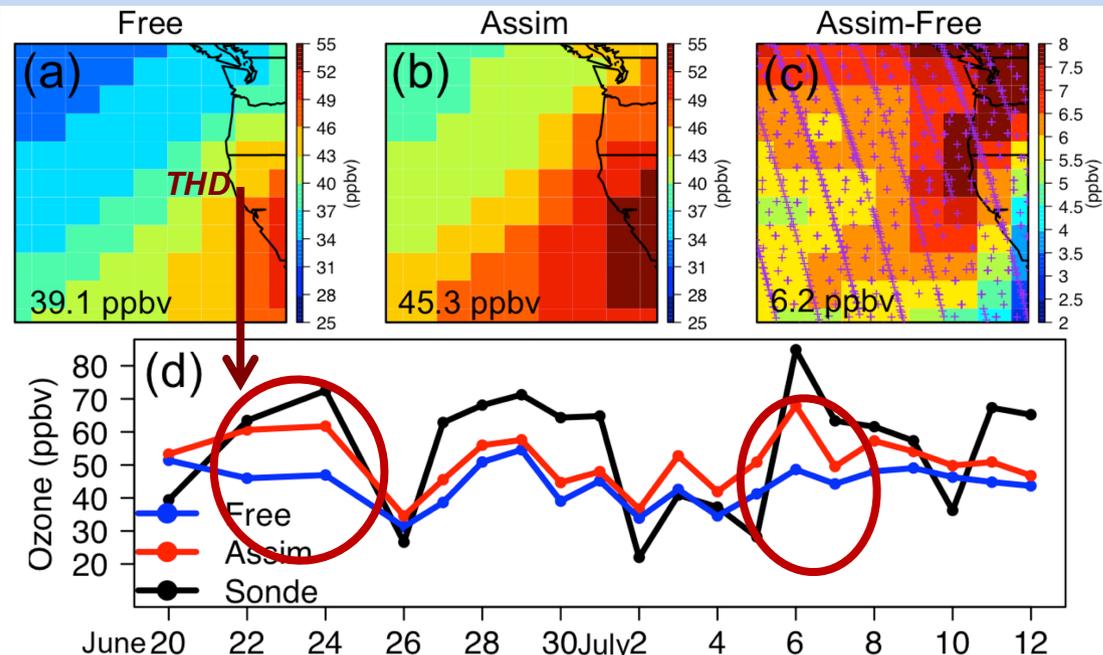
Cross Validation

evaluate with independent in-situ (ozonesonde, surface, aircraft) observations



# Ozone at the gate.

Ozone in boundary condition model (GEOS-Chem) at 700-900 hPa:  
Changes due to assimilating TES ozone

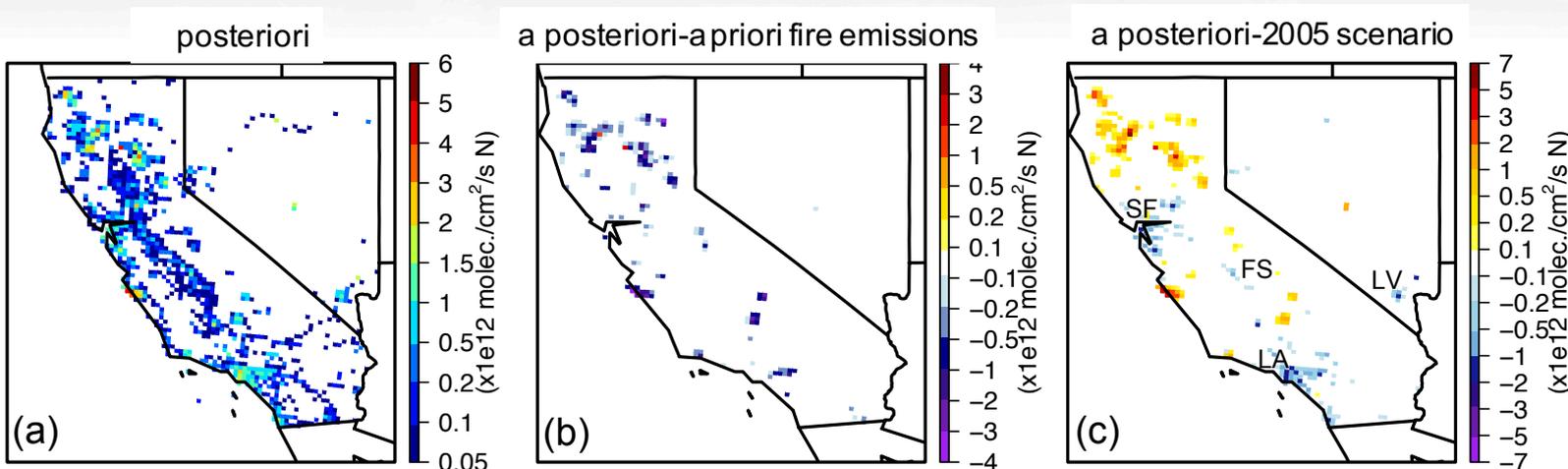


- Assimilating TES ozone enhanced the GEOS-Chem ozone a priori by ~16% (6.2 ppbv): generally increasing by latitude.
- Assimilation reduced (but did not eliminate) the overall negative biases
- Most significant improvement occurred during the long-range transport events (Jun 22-24; Jul 6)



# Emission partition

Regional-scale assimilation spatially redistributed local NO<sub>x</sub> emissions particularly from urban and wildfires



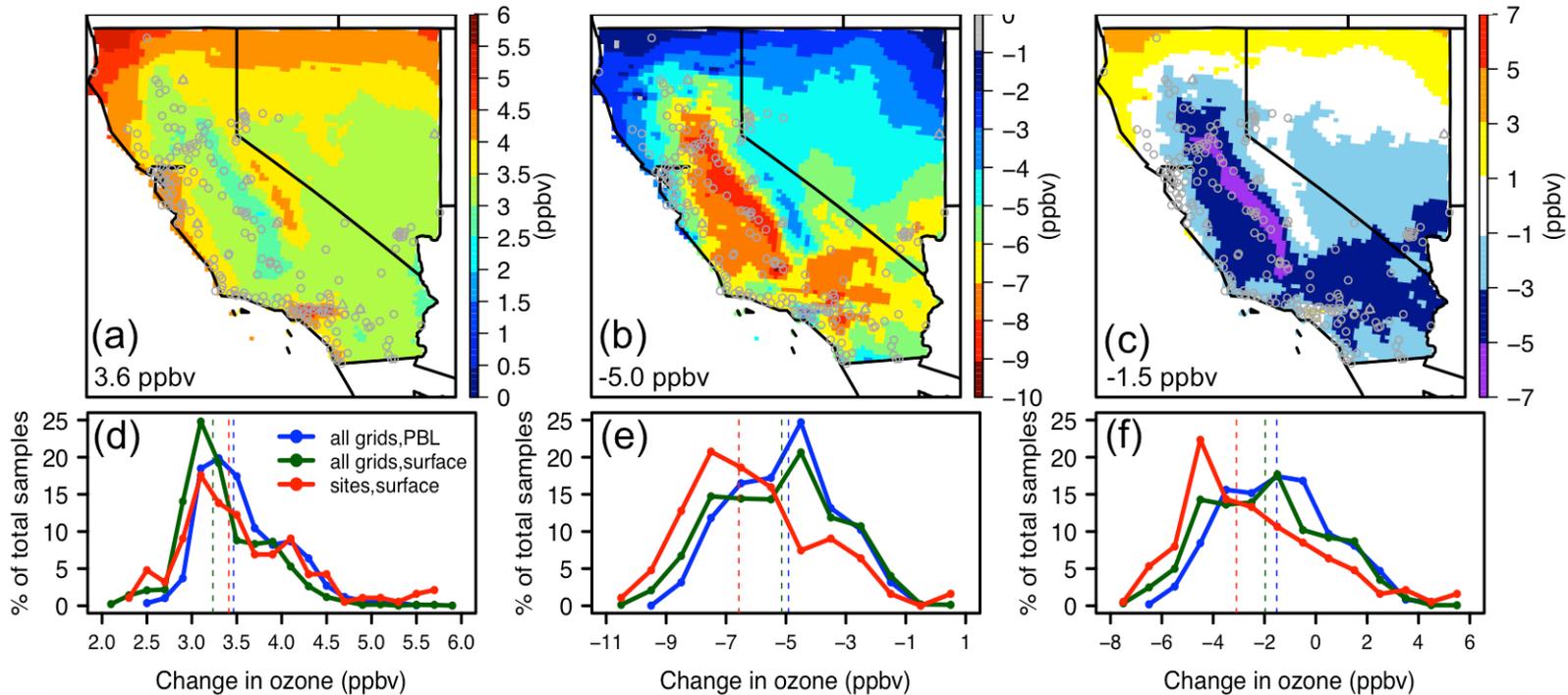
- Anomalously high fire emissions in the Northern California this year
- Original QFED fire emissions overpredicted the NO<sub>x</sub> emissions
- Variable reductions in urban areas due to the emission controls during 2005-2008 and the uncertainties of NEI 2005 relative to its base year of 2005



# Impact of sampling

Impact of the multi-scale assimilation on STEM ozone:  
We repartitioned ozone source contributions from local and non-local sources

*Period-mean near-surface daytime (<2 km a.g.l., 8am-7pm) ozone in STEM, after-before assimilation*

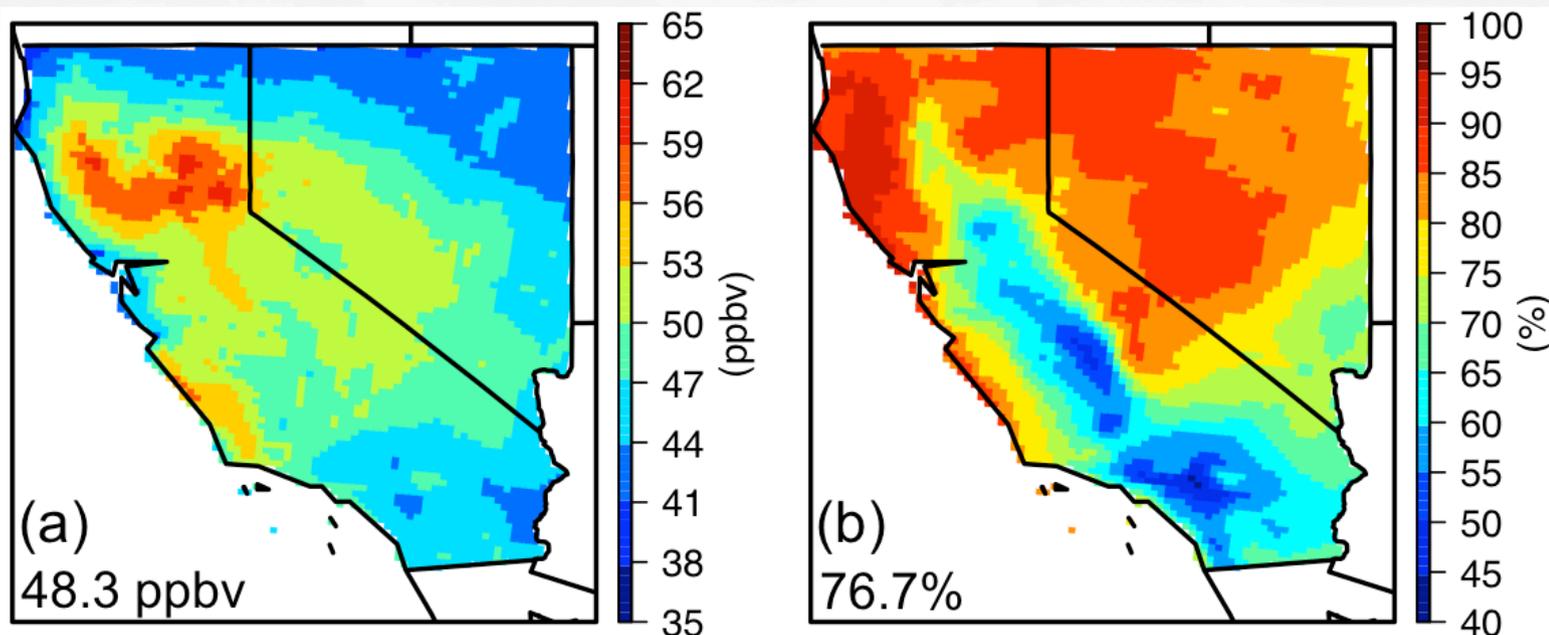


- High terrain regions: more sensitive to extra-regional sources
- Central Valley and SoCal: more sensitive to local emissions
- Monitoring sites unable to capture some strong changes due to the assimilation



# Repartition of local and non-local sources

## Monthly-mean surface background ozone (MDA8)

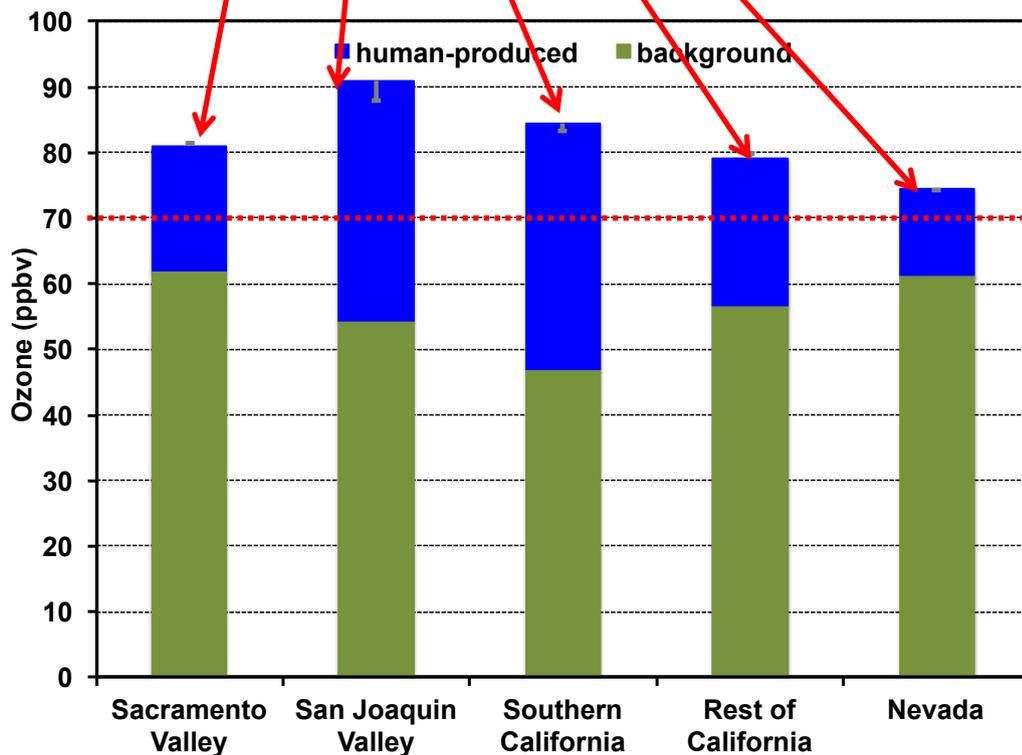
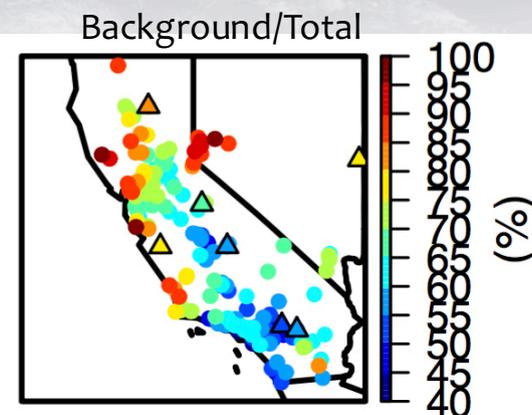
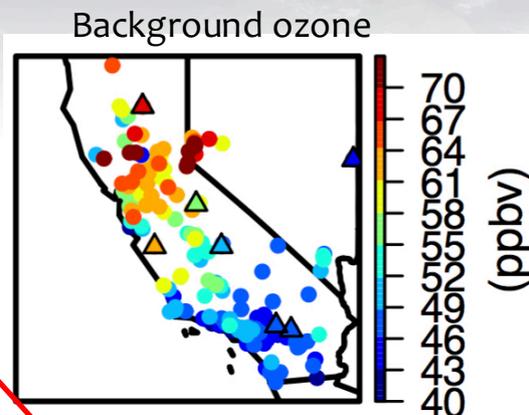


- Assimilation repartitioned background (as well as total) ozone:
- Compared to a priori: 50.7 ppbv
  - +3.3 ppbv from the boundary conditions
  - -5.7 ppbv from the local emissions



# Background ozone contribution

Background ozone contributing to the exceedances (>70 ppbv) at surface sites



Computed also for other levels of exceedances

- Highest background ozone in Sacramento Valley, <10 ppbv below the primary standard on the observed days of exceedances.
- Lowest background ozone in Southern California, and on observed days of exceedances, larger additional anthropogenic contributions (up to 20 ppbv) would be possible without exceeding the threshold.

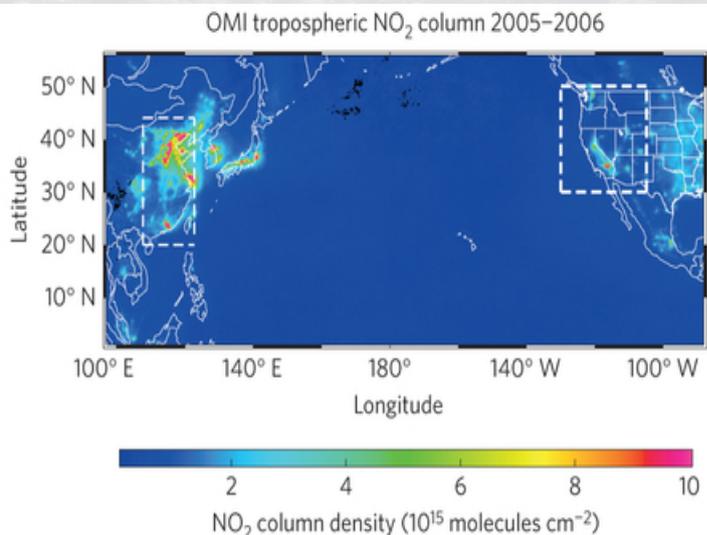


# Is Background Ozone Changing?

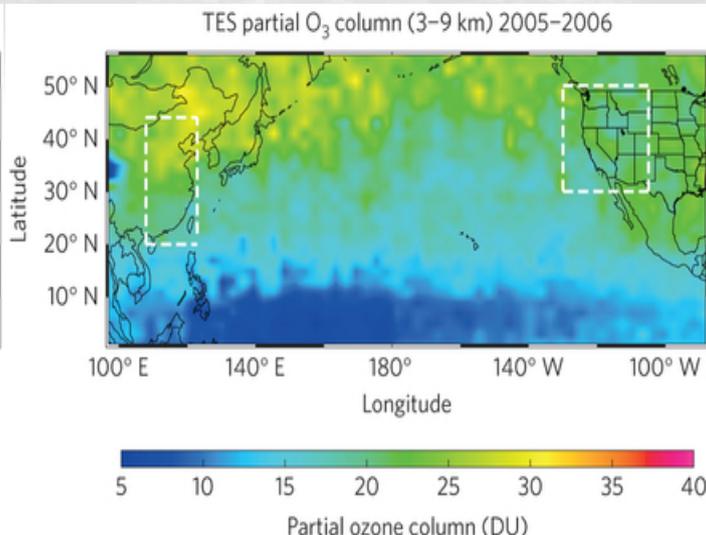


# Aura sees big changes in just 6 years

**NO<sub>2</sub> Column from the Ozone Monitoring Instrument (OMI)**

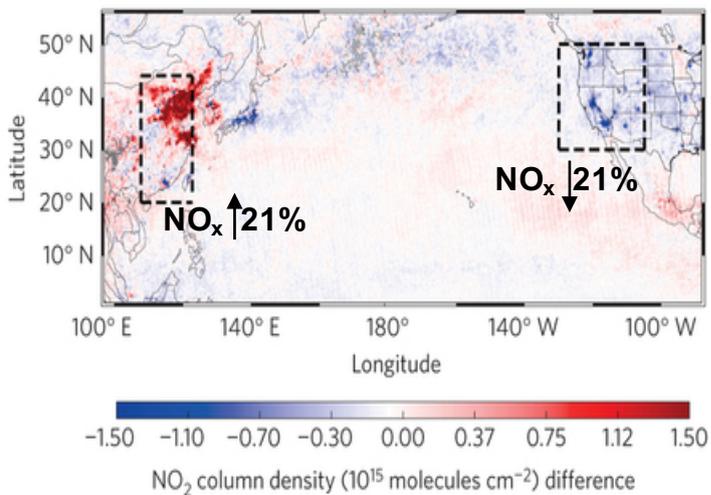


**Mid-tropospheric ozone from the Tropospheric Emission Spectrometer (TES)**

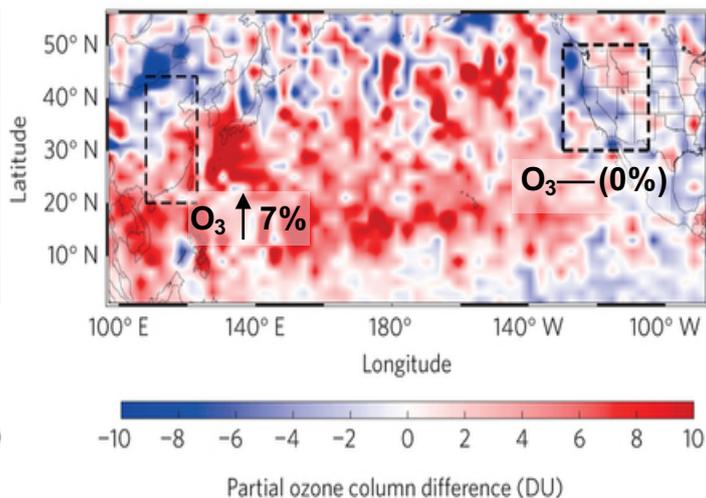


2005–2006 NO<sub>2</sub> and Ozone

2010–2005  $\Delta$  in OMI NO<sub>2</sub> column



2010–2005  $\Delta$  in TES partial O<sub>3</sub> column



Change in NO<sub>2</sub> and Ozone from 2005 to 2010

Nitrogen oxides (NO<sub>x</sub>) are critical ozone-forming pollutants

Mid-tropospheric ozone contributes to regional greenhouse gas warming



# Attribution: Emissions and transport

We used a model (constrained by the Aura observations) to attribute observed ozone changes to:

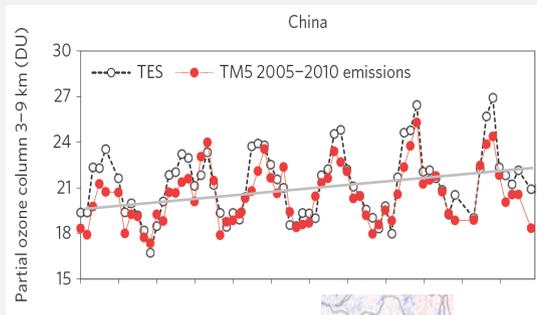
Regional emissions

Long-range transport

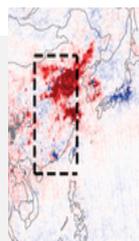
Downward transport from the stratosphere

## Microwave Limb Sounder (MLS): Temporary increase in downward transport from the stratosphere

TES: 7% Increase in mid-tropospheric ozone



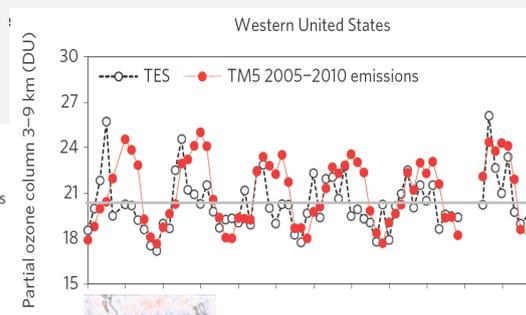
OMI: 21% increase in NO<sub>x</sub> emissions. Explains 50% of the ozone increase.



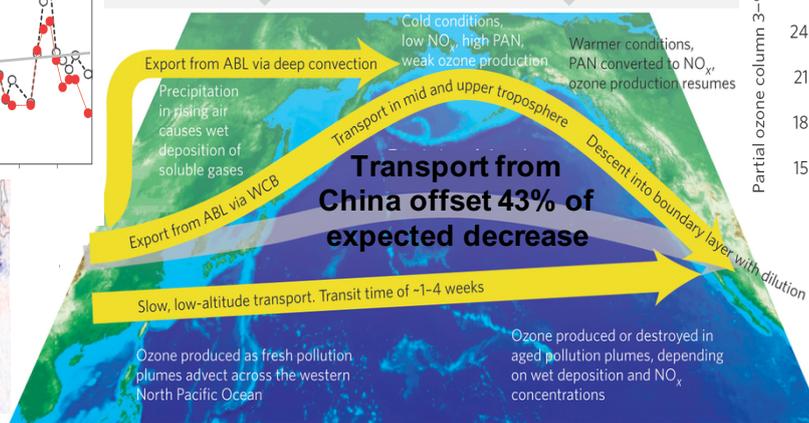
Explains 50% of the ozone increase

Offset 57% of expected ozone decrease

TES: No change in mid-tropospheric ozone



OMI: 21% decrease in NO<sub>x</sub> emissions. Should have given a 2% decrease in ozone

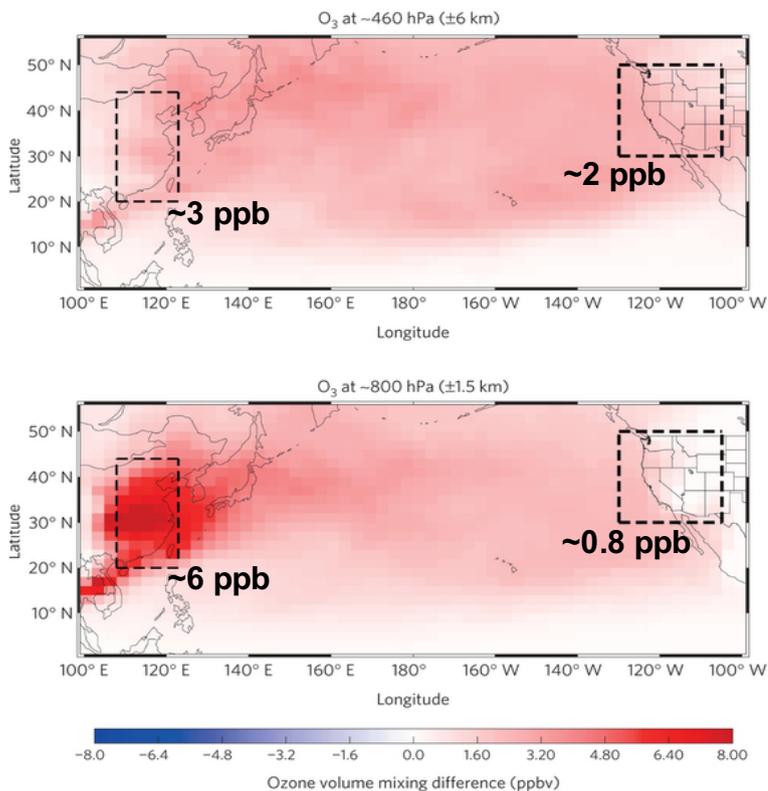




# Export of Chinese Pollution to the Western US

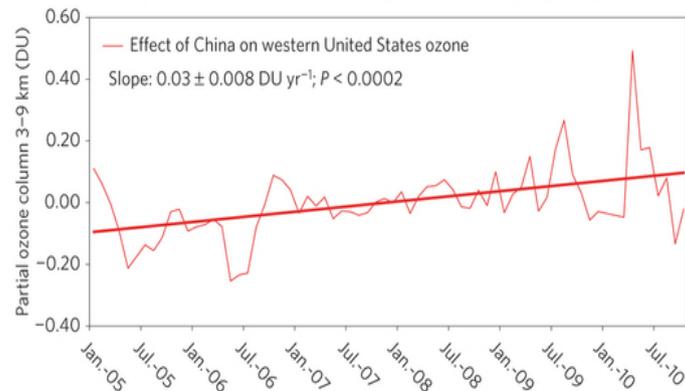
In contrast to the oscillating stratospheric contribution, there was a secular increase in Western US ozone attributable to long-range transport from Eastern China.

## Ozone associated with Chinese Emissions for April-May 2010



## Contribution of Chinese Emissions to Western US Mid-tropospheric Ozone

Monthly deseasonalized tropospheric  $O_3$  in the western US originating from China



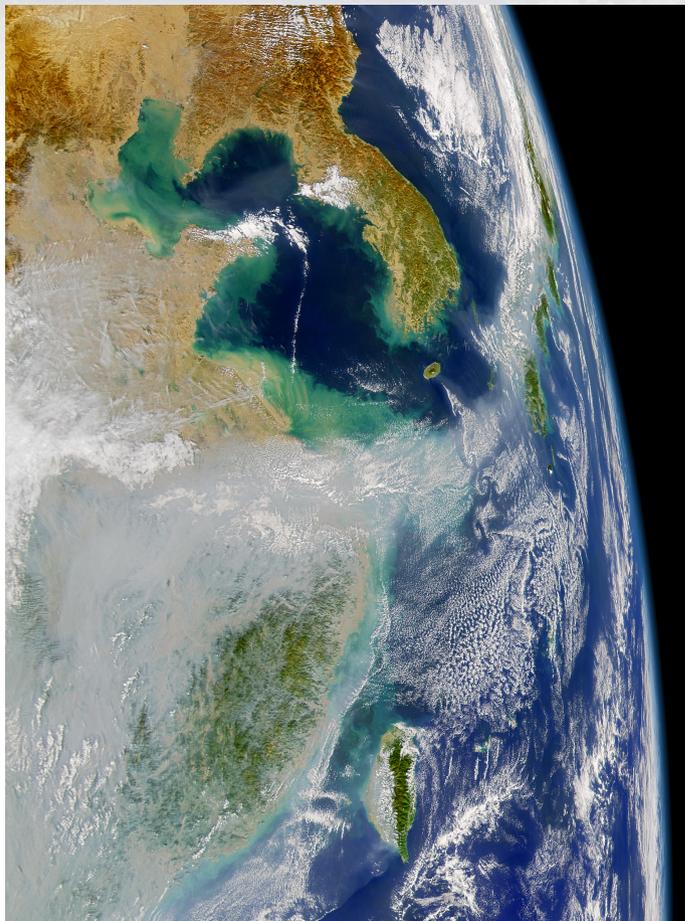
The model indicates that changes in surface ozone over Eastern China have been  $\sim 2x$  larger than mid-tropospheric ozone changes.

The impact of long-range transport from Eastern China on Western US background surface ozone is  $< 1/2$  what we see in the mid-troposphere.

Other studies have shown that individual transport events can increase Western US surface ozone by  $> 8$  ppbv.



# Summary



- Combining data from OMI, TES, and MLS, we find that tropospheric ozone over Eastern China increased by 7% between 2005 and 2010, which can be equally attributed to an increase in  $\text{NO}_x$  emissions and a temporary increase in stratospheric input.
- Tropospheric ozone over the Western US did not decrease despite a 21% reduction in  $\text{NO}_x$  emissions. The expected reduction was offset by a combination of long-range transport from China and the increased stratospheric input.
- While the impact of Eastern China's emissions on ozone at and above the surface of the Western US have thus far been small, our study indicates that external sources of pollution need to be considered in developing new air quality and regional climate change mitigation policies.

Verstraeten et al, Nature Geosciences (2015)



# What is an air pollutant?

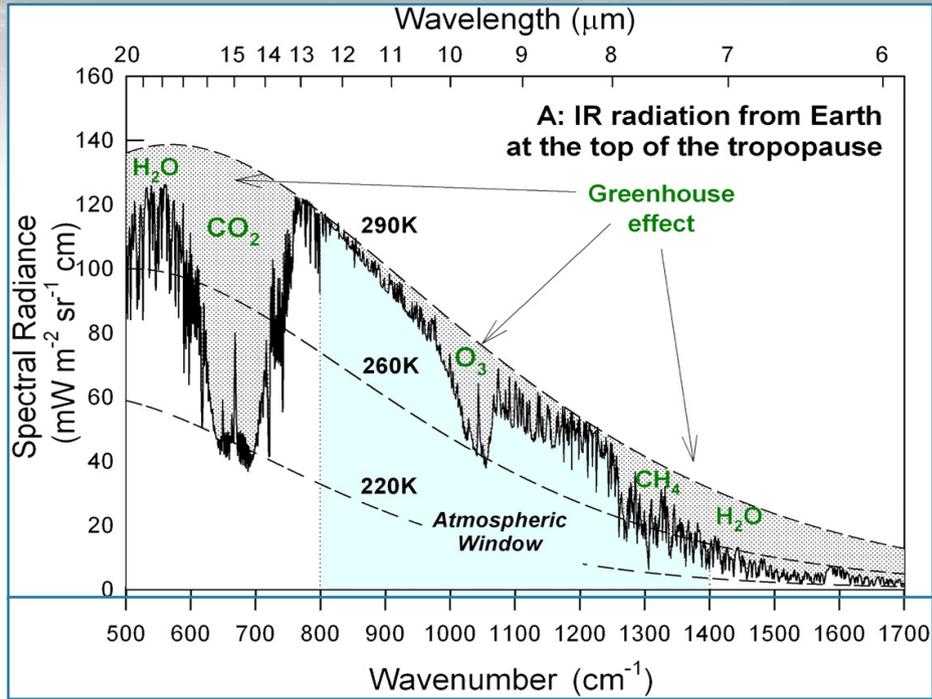
“Because greenhouse gases fit well within the Clean Air Act’s capacious definition of “air pollutant,” we hold that EPA has the statutory authority to regulate the emission of such gases from new motor vehicles.”

MASSACHUSETTS *v.* EPA  
Opinion of the Court, April 2<sup>nd</sup>, 2007  
Justice Stevens

- Air pollutants now include Greenhouse Gases (GHG)
- Air quality constituents, e.g., ozone and black carbon, are also GHGs
- Air quality can effect both human and environmental health, with implications for carbon cycle feedbacks

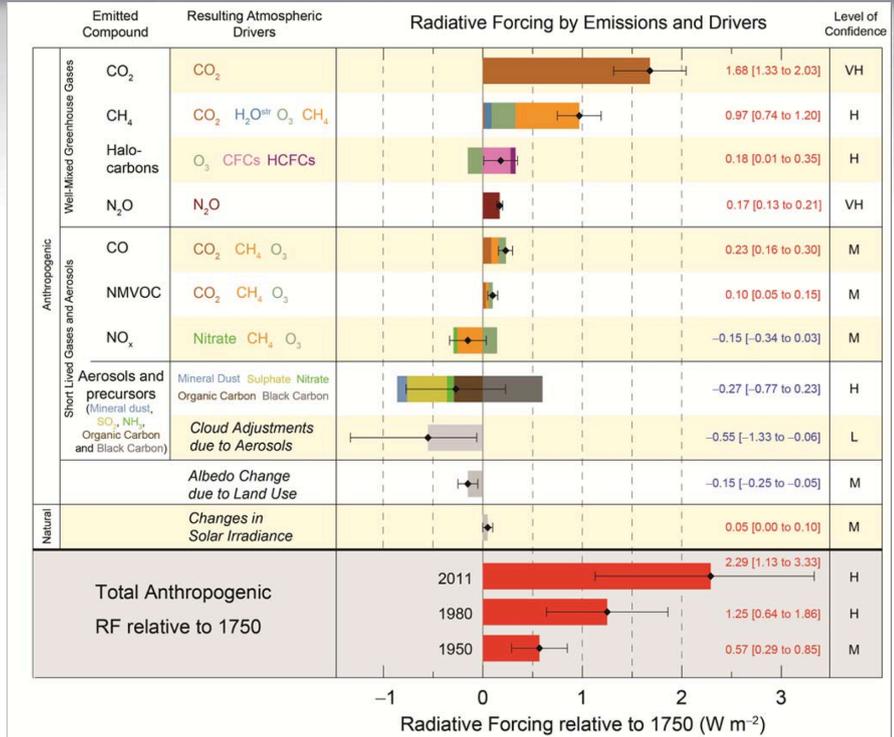


# Radiative forcing from atmospheric composition



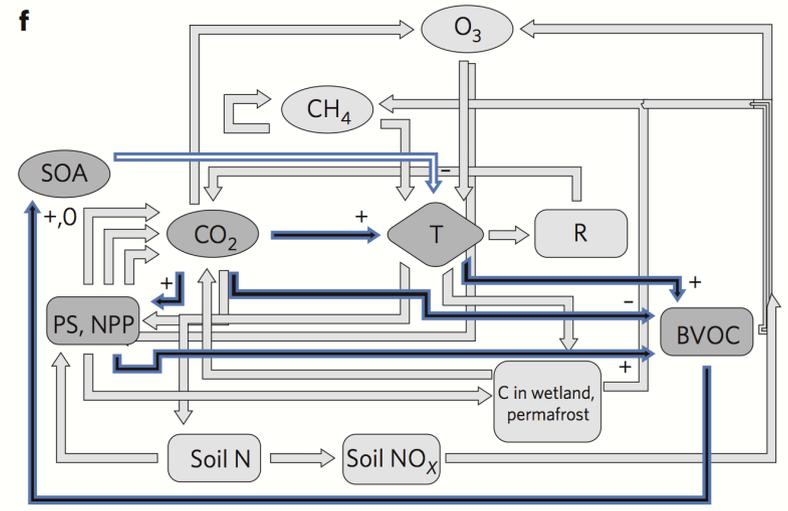
Wallington T J et al. PNAS 2010;107:E178-E179

IPCC AR5



Carbon dioxide, methane, and ozone are the three most important greenhouse gases resulting from anthropogenic activities.

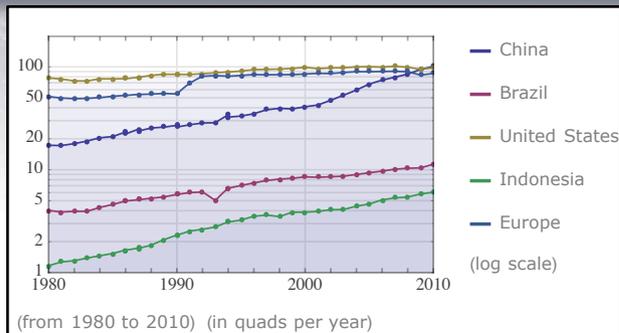
These gases are coupled through common sources and coupled within the Earth System.



Arneeth et al, 2010, Nat. Geo. Sci.



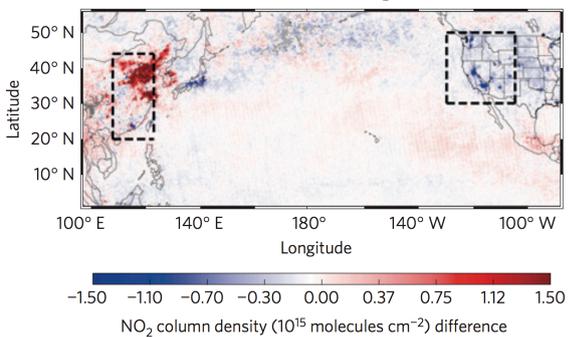
# Earth Observing System epoch



The EOS epoch has borne witness to a dramatic global redistribution of energy.

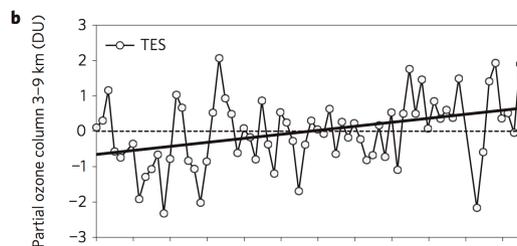
## NO<sub>2</sub>

2010–2005  $\Delta$  in OMI NO<sub>2</sub> column



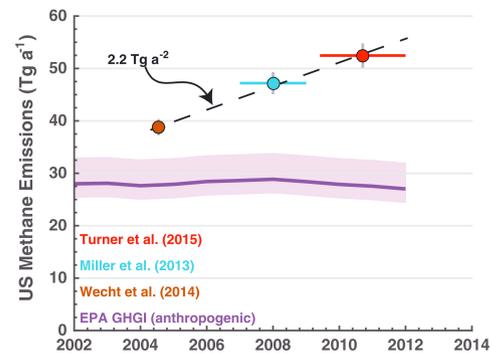
Verstraeten et al, 2015

## Ozone



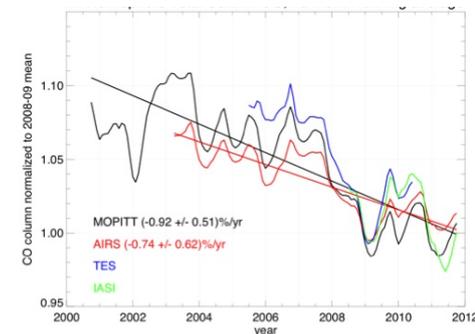
Verstaeten et al, 2015

## Methane



Turner et al, 2016

## Carbon Monoxide



Worden et al, 2013

The past decade has altered the sources of atmospheric composition with profound implications for climate and human health.

The trajectory of atmospheric composition sources/sinks and its impact on the Earth System is both highly uncertain and critically important for climate and environmental health mitigation.



# Chemistry-climate coupling: ozone

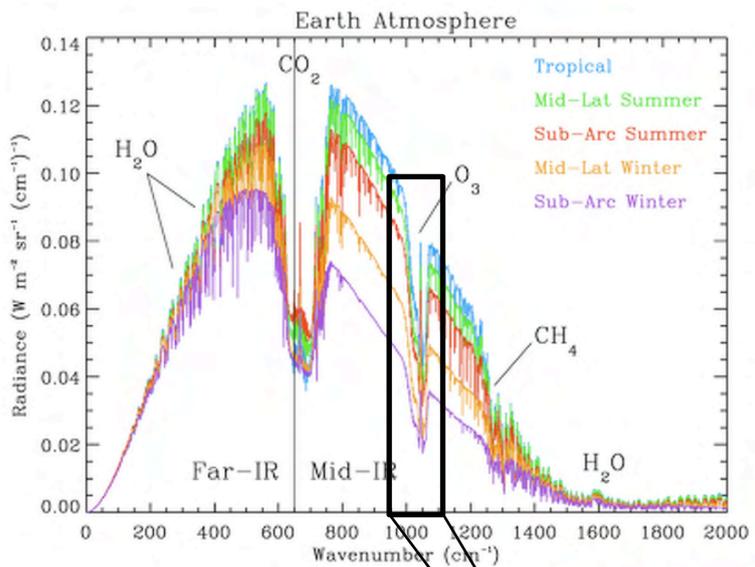
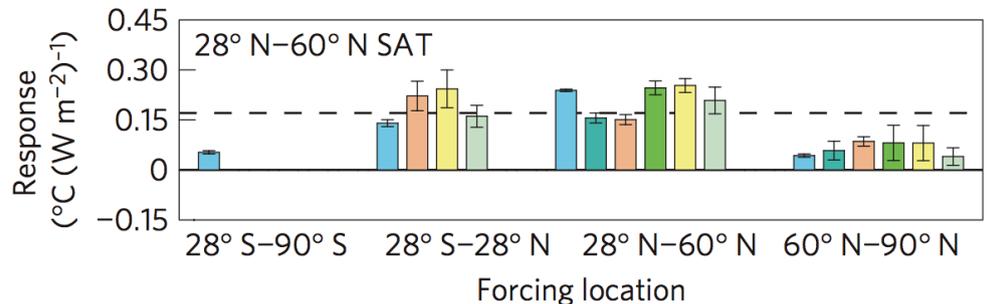
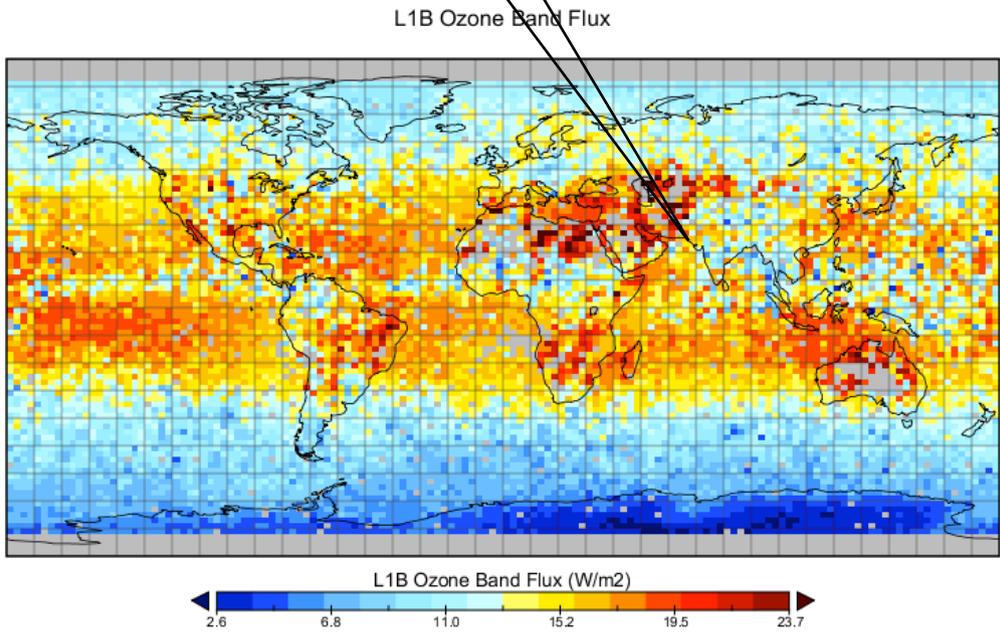
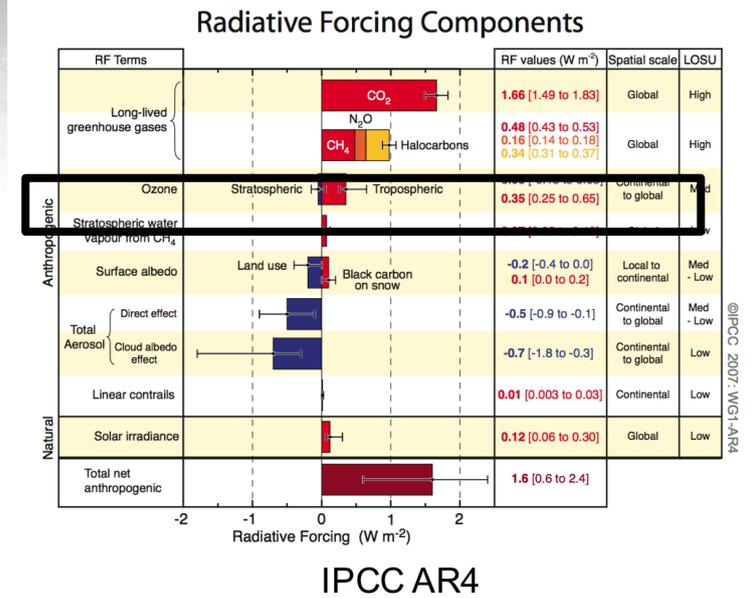


Fig. courtesy M. Mlynzcack (LaRC)

• Inter-model radiative forcing from ozone: .35 [0.25-.65]  $W/m^2$  (IPCC AR4)

Radiative Forcing uncertainties:

- Pre-industrial background ozone
- Spatial distribution
- Vertical distribution



Shindell and Faluvegi, *Nature Geosciences*, 2008

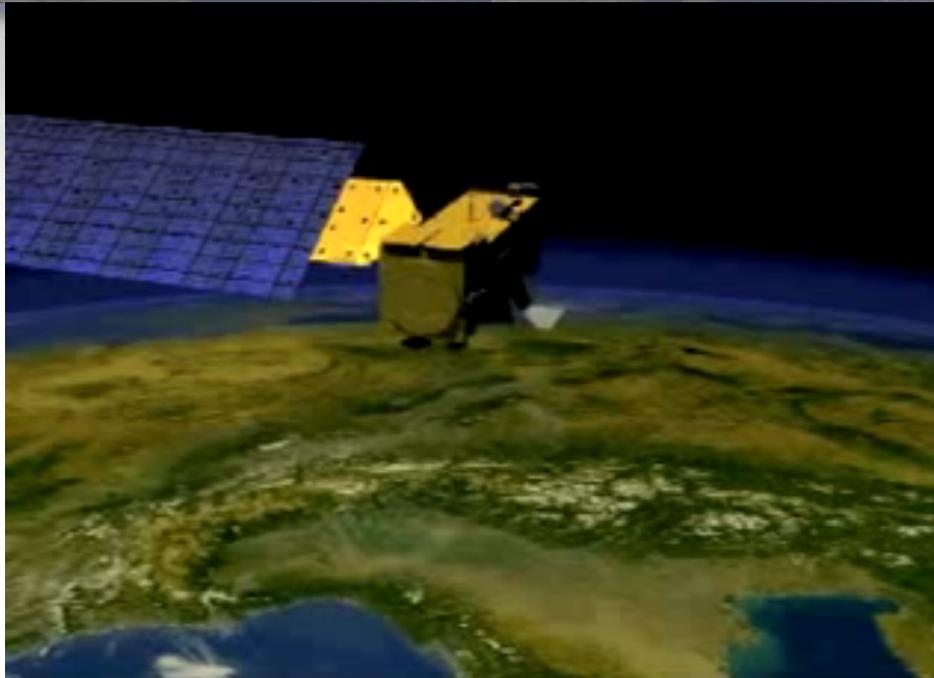


The regional temperature response is a function of the forcing location

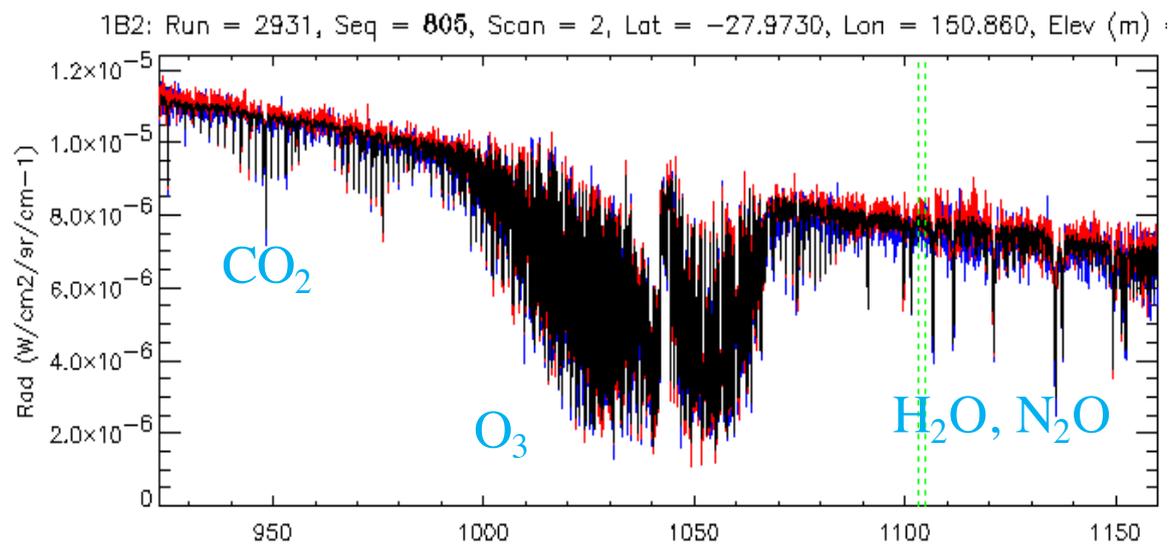


# Tropospheric Emission Spectrometer

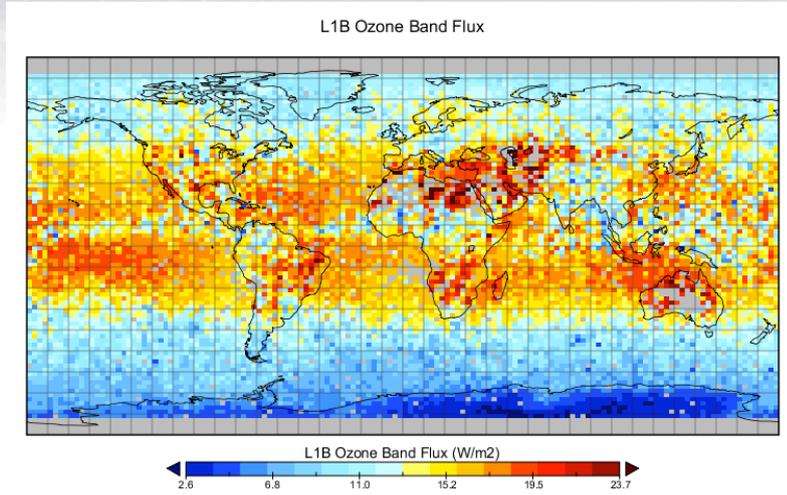
TES, launched aboard the Aura spacecraft in 2004, is a Fourier Transform Spectrometer measures infrared spectral radiances from 3.2 to 15.4 microns.



Spectral Resolution (unapodized)	0.06 $\text{cm}^{-1}$ (nadir) 0.015 $\text{cm}^{-1}$ (hi-res)
Spectral Coverage	650 to 3050 $\text{cm}^{-1}$ (3.2 to 15.4 $\mu\text{m}$ )
Global survey coverage	72 observations/orbit 16 orbits/day
Spatial Resolution	0.5 x 5 km (nadir) 2.3 x 23 km (limb)
Nadir NEDT @290K (Noise Equivalent Delta Temperature)	2B1: 1.08 K 1B2: 0.36 K 2A1: 0.36 K 1A1: 2.07 K

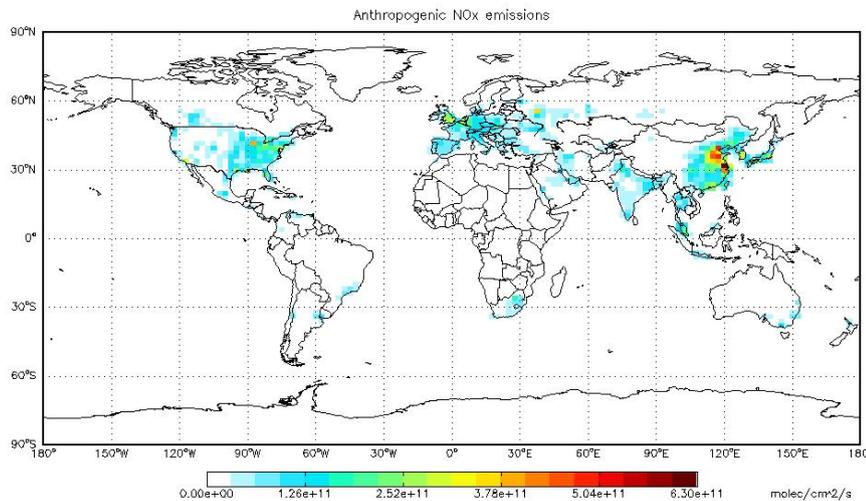


Mean



$$\mathcal{J} = \frac{1}{N} \sum_{i \in \text{TES}} F_i$$

↑  $\delta J(\text{NO}_x)$



$$\lambda = \nabla_{\mathbf{E}} \mathcal{J}$$



# Connecting TOA to emissions

Sensitivity of TOA at one location with respect to precursor emissions  $\mathbf{E}=[E_1, E_2, \dots, E_N]$

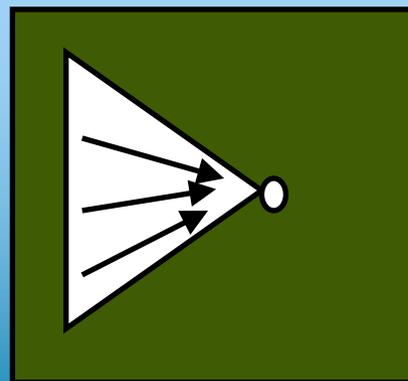
Use chain-rule to link TES Instantaneous Radiative Kernels to emissions through GEOS-Chem adjoint

$$\lambda^i = \frac{\partial F_i}{\partial \mathbf{E}}$$

$$\lambda^i = \left( \frac{\partial \mathbf{c}_i}{\partial \mathbf{E}} \right)^T \frac{\partial F_i}{\partial \mathbf{c}_i}$$

Model adjoint      TES IRK

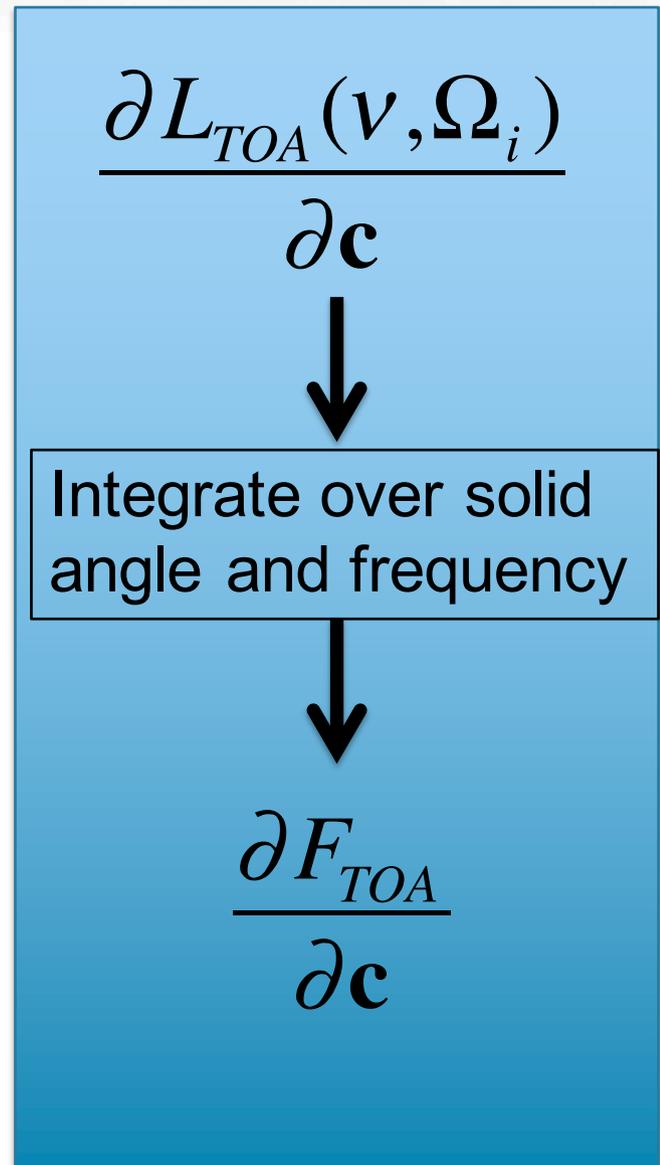
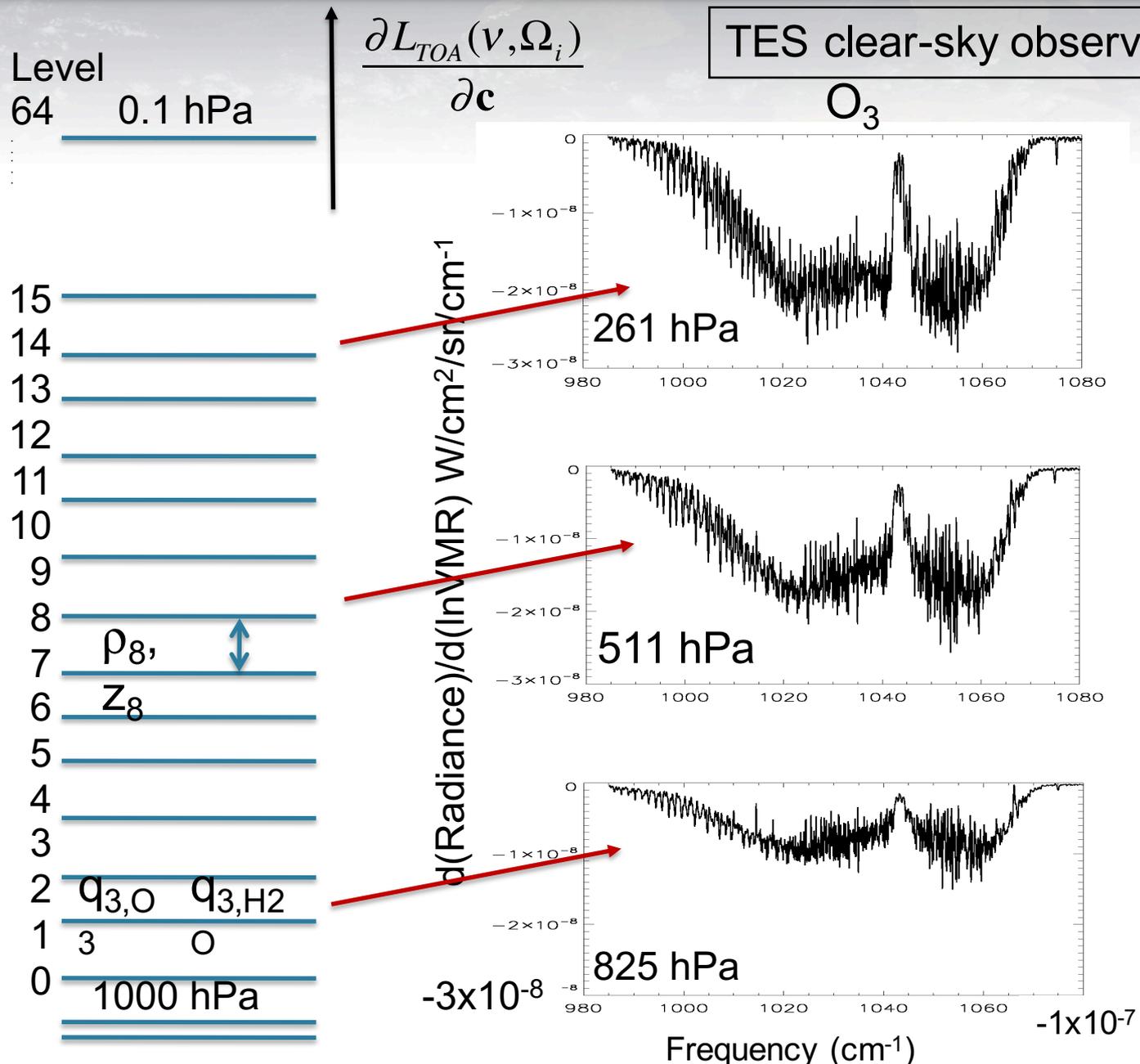
$$\frac{\partial \mathbf{c}^i}{\partial \mathbf{E}} = \frac{\partial \mathbf{M}_{i-1}}{\partial \mathbf{c}_{i-1}} \dots \frac{\partial \mathbf{M}_0}{\partial \mathbf{E}}$$



Adjoint accounts for both transport and chemical transformation



# TES Instantaneous Radiative Kernels (IRK)



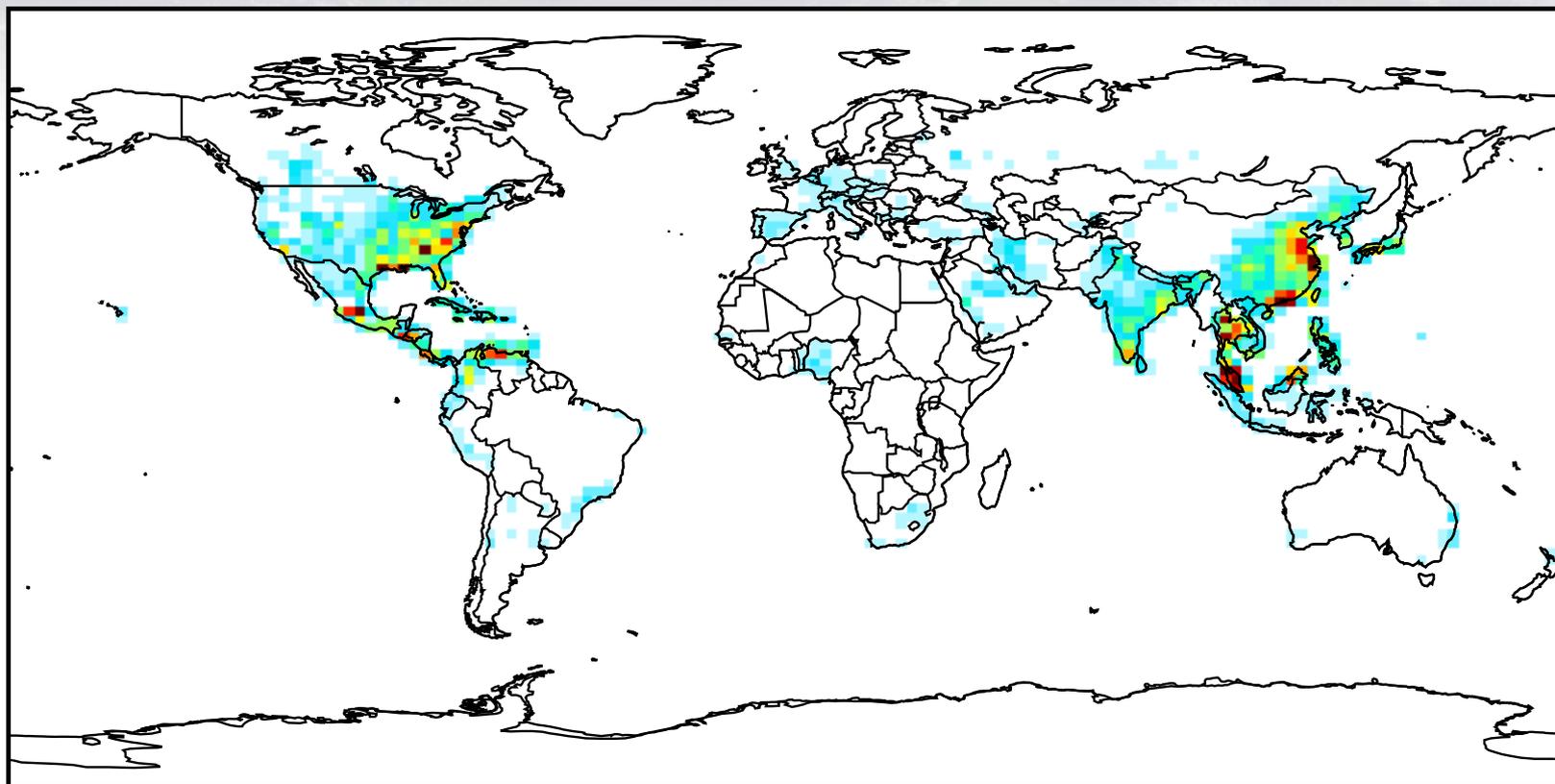
Worden et al, 2008, 2011, Doniki et al, 2015



# Attribution of ozone RF to NO<sub>x</sub> emissions: location matters

(a) NO<sub>x</sub>

August, 2006



Bowman and Henze, GRL, 2012

0 0.04 0.08 0.12 mW m<sup>-2</sup>

- Beijing and Guandong drive O<sub>3</sub> RF in part due to large emissions.
- Anticyclones over Atlanta trap O<sub>3</sub> in the upper troposphere.
- Indonesia and Malaysia are very efficient because of high temperature and convection.

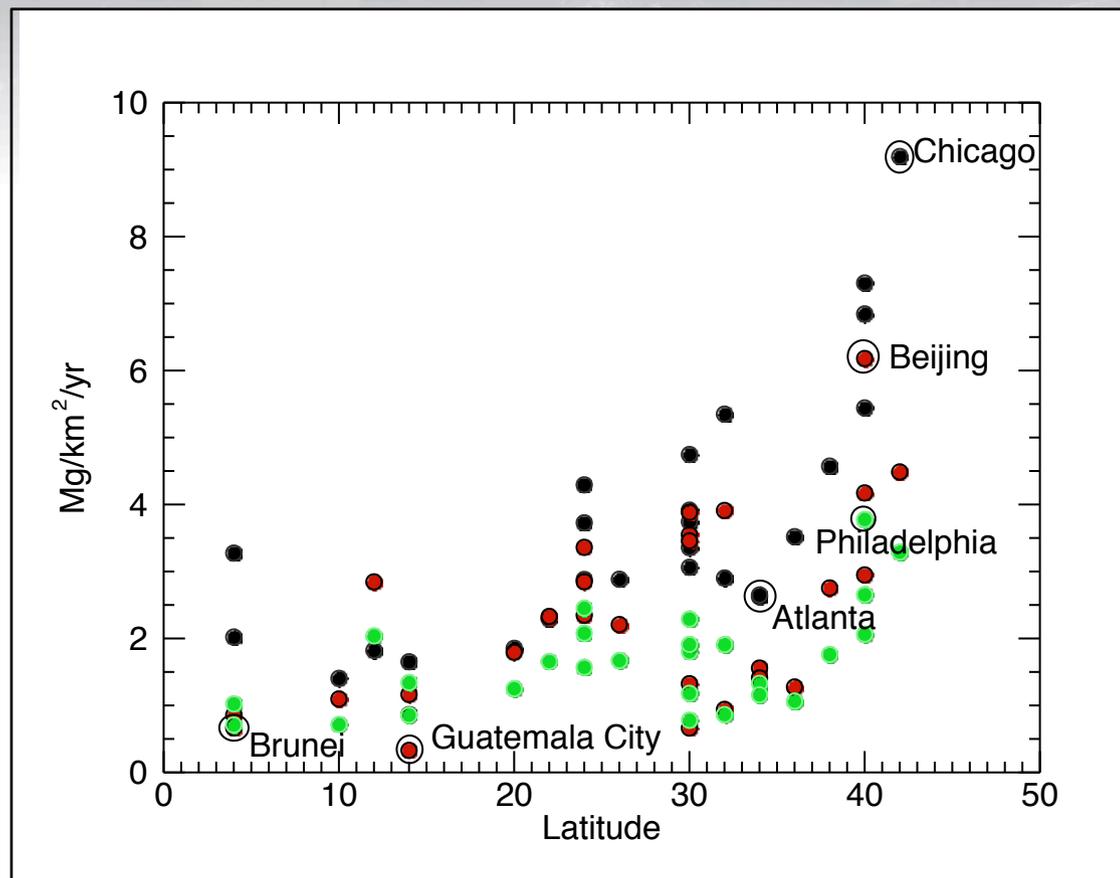


# Change in absolute emissions

NO<sub>x</sub> emissions from Atlanta are over a factor of 3 smaller than Chicago but with comparable ozone DRF.

CO emissions from Guatemala City are a factor of 20 smaller than Beijing.

Brunei NMHC emissions are a factor of 5 smaller than Philadelphia



Bowman and Henze, GRL, 2012

Regional air quality/climate co-benefits can be quantified in terms of ozone direct radiative forcing

CO emission reductions decrease methane lifetime—a more potent greenhouse gas—while NO<sub>x</sub> emissions reductions increase lifetime. Quantification of methane feedbacks necessary for a full air quality-climate co-benefit analysis.

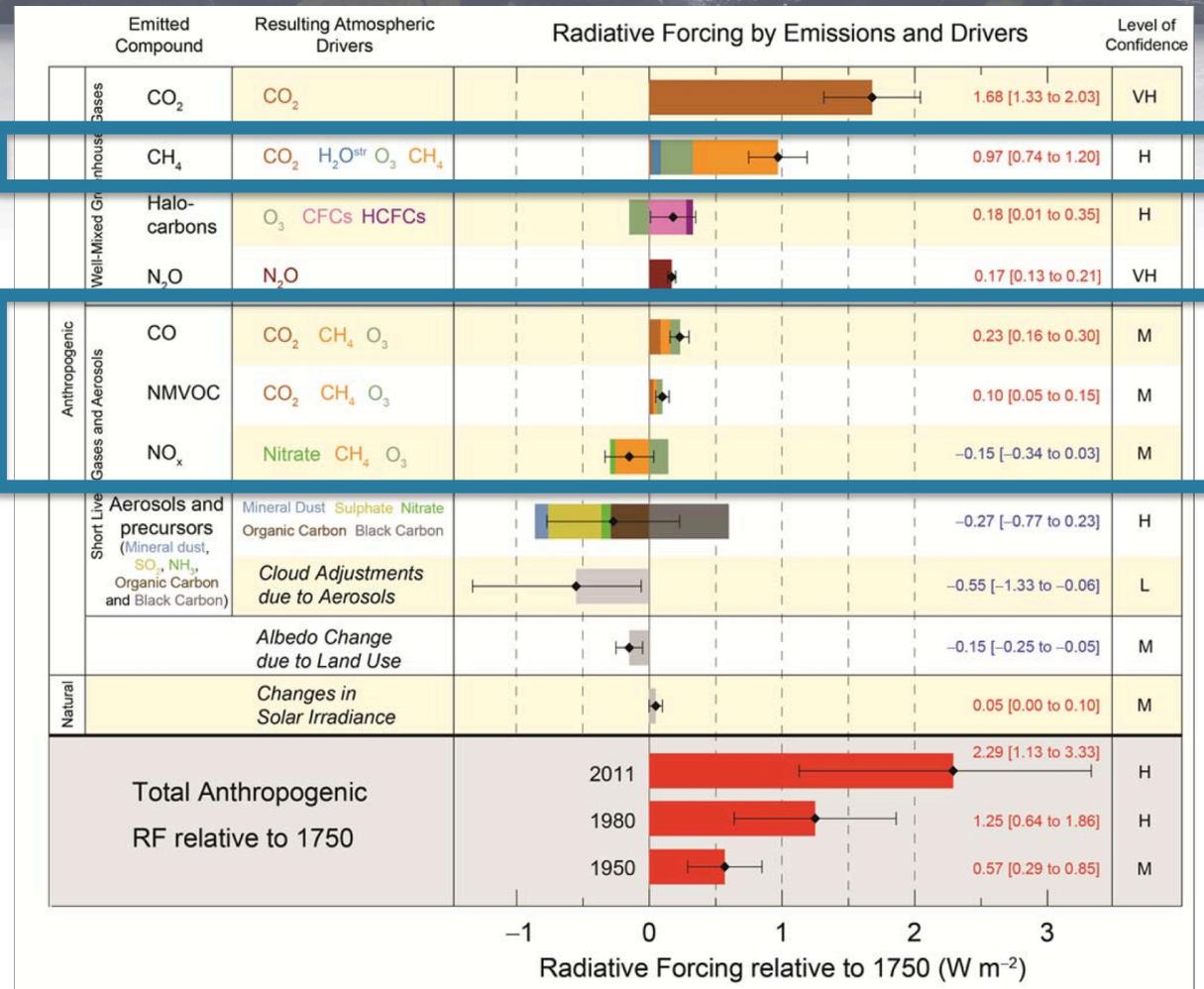


# Methane RF

Historic Methane RF ~1 W/m<sup>2</sup>

Net CH<sub>4</sub> is the balance of emission and chemical prod/loss

$$CH_{4_{net}}(x) = CH_{4_{emiss}}(x) - CH_{4_{chem}}(x)$$



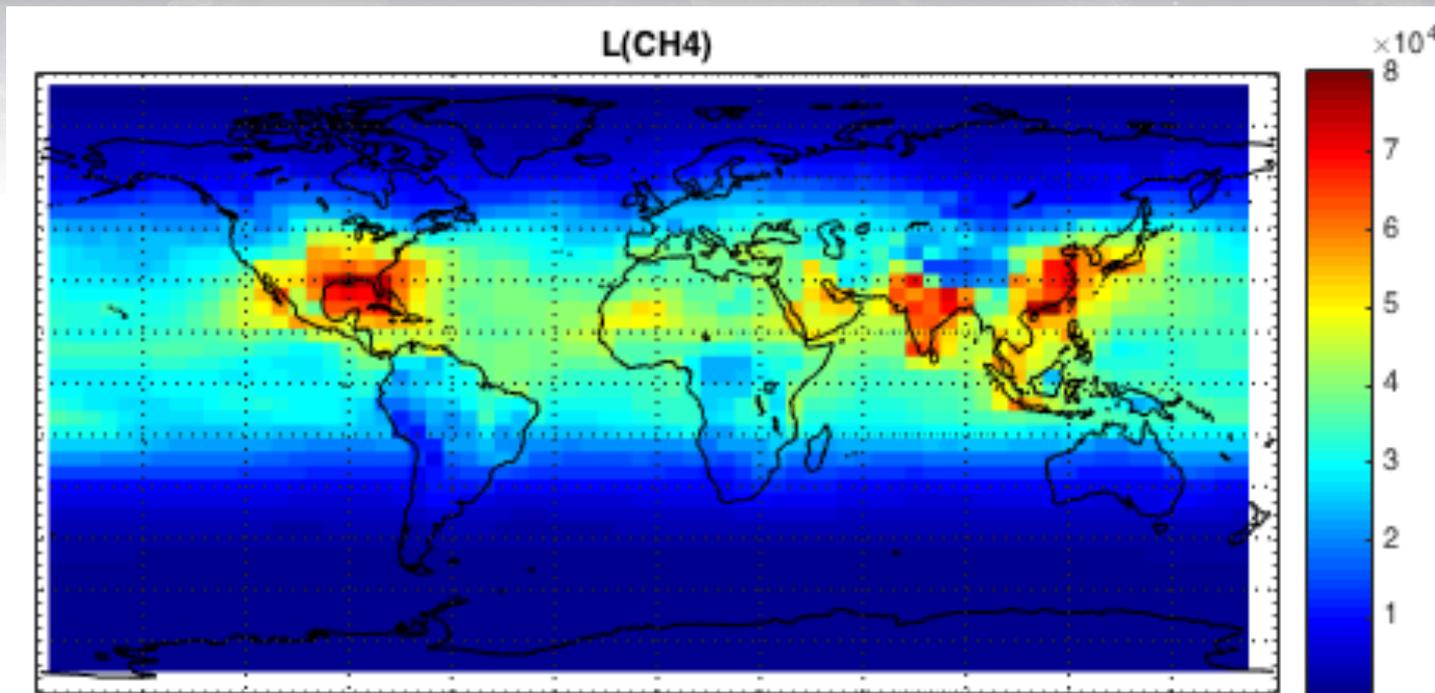
Methane emissions: ~640 (342 Anth+300 natural)

Methane chemistry: ~638 Tg

Ozone precursor emissions affect methane radiative forcing by changing its lifetime through control of OH.



# Losing methane



Total chemical loss of methane is a function of the global distribution of OH

$$L(CH_4) = \sum_{i \in D} \kappa_i [OH]_i [CH_4]_i$$

Sensitivity of methane loss to precursor emissions can be calculated with the adjoint

$$\nabla_{\mathbf{E}} L(CH_4)$$

From methane lifetime change, radiative forcing can be calculated from simplified RT

$$L(CH_4) \rightarrow \Delta CH_4 \rightarrow \Delta RF$$

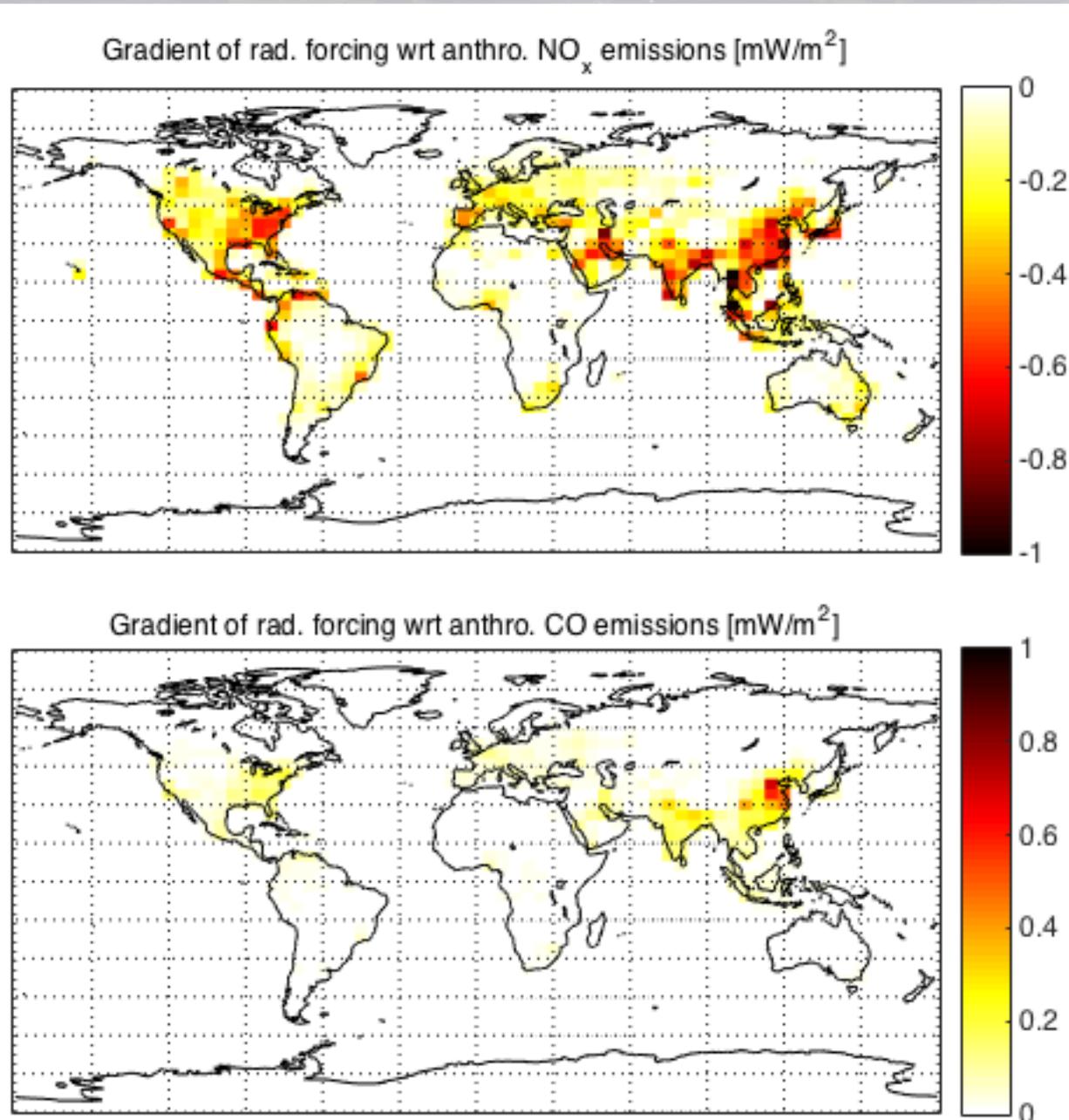


# Spatial attribution of methane RF

NO<sub>x</sub> emissions are a *negative* climate forcer through methane by increasing OH but a *positive* climate forcer through increases in ozone.

As with ozone RF, tropical, convective regions are a more efficient forcer.

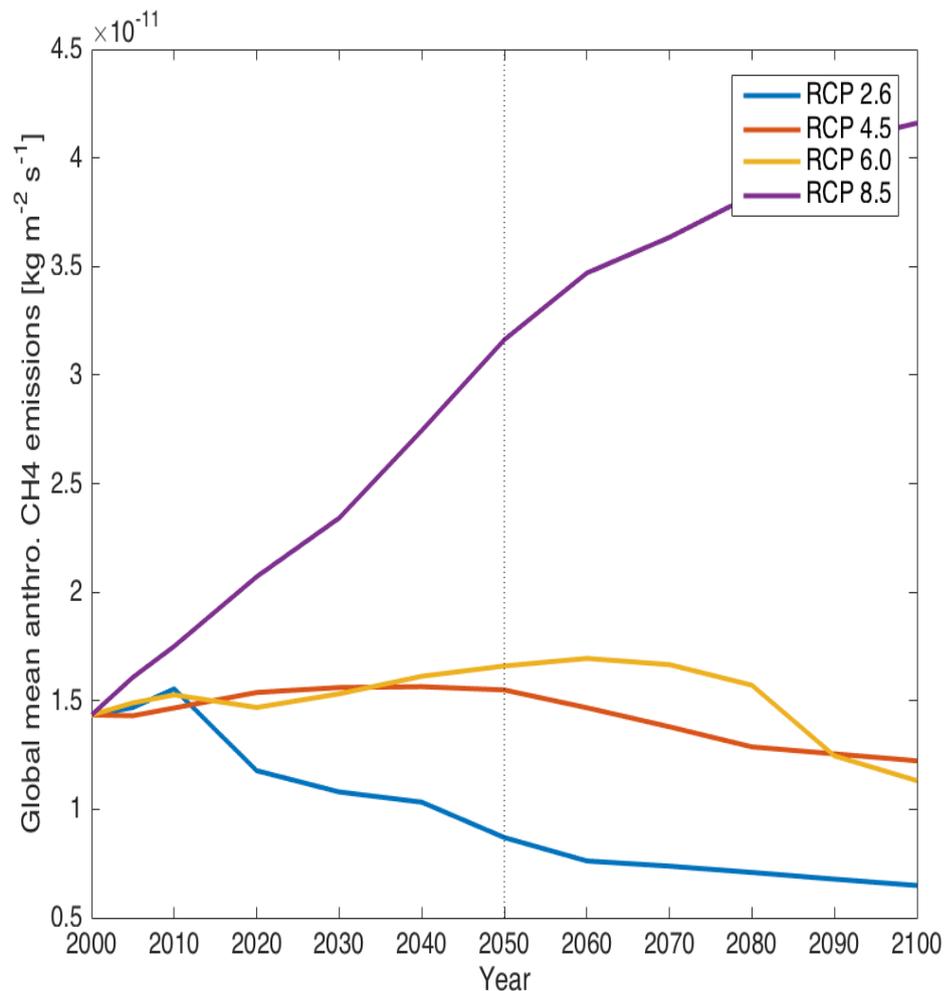
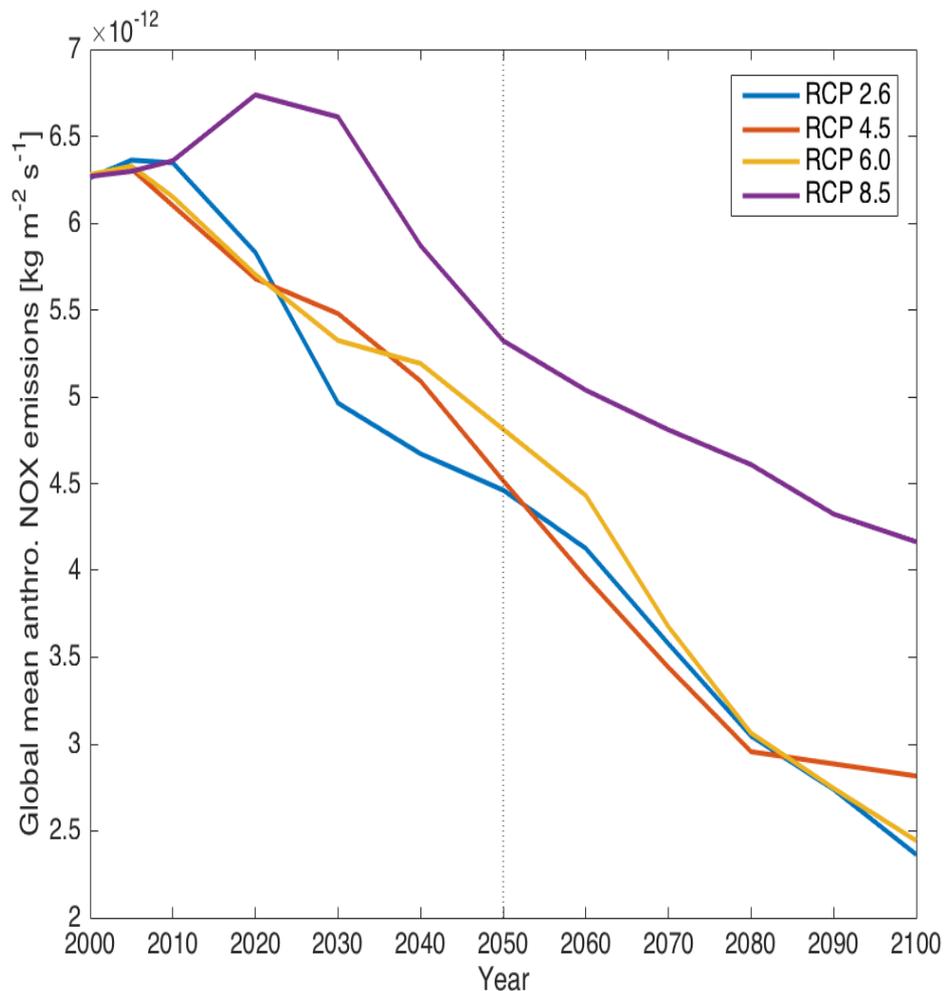
CO emissions are a *positive* climate forcer through methane by reducing OH and increasing ozone.



Walker and Bowman, *in prep.*



# Representative Concentration Pathways



RCP 6.0 includes monotonic NO<sub>x</sub> reductions and but non-monotonic CH<sub>4</sub> increase



# Radiative Forcing in 2050 (RCP6)

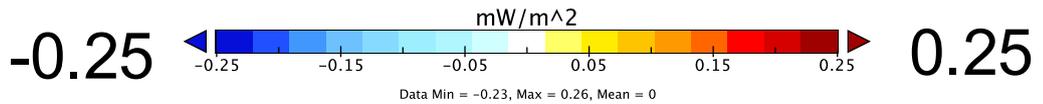
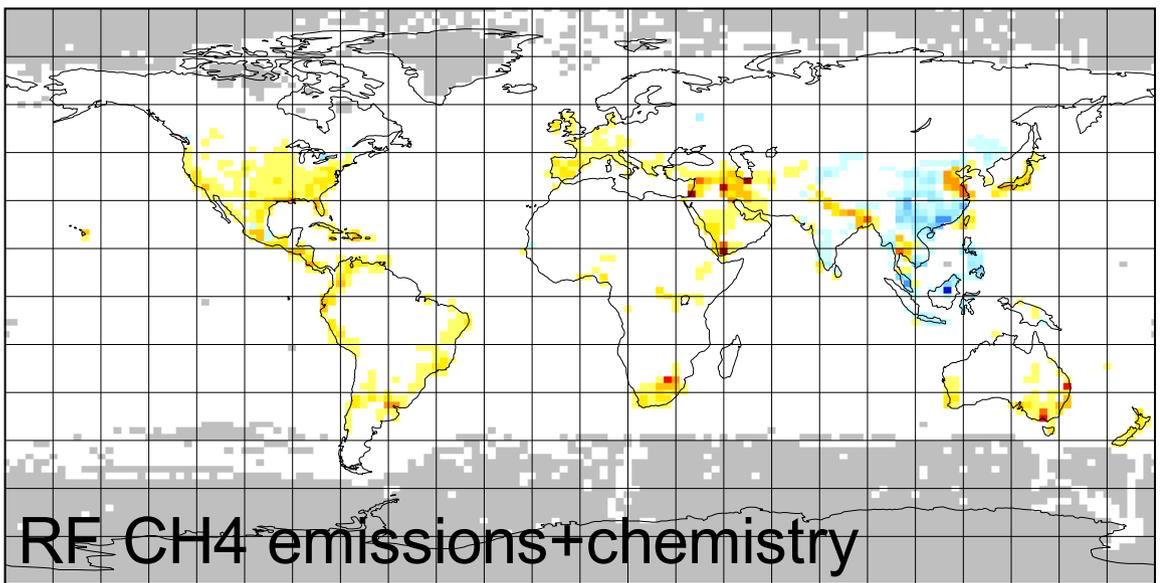
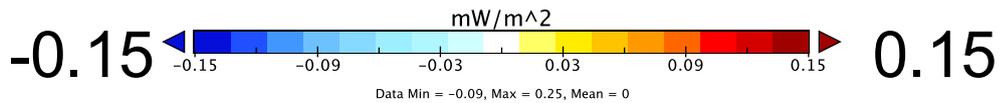
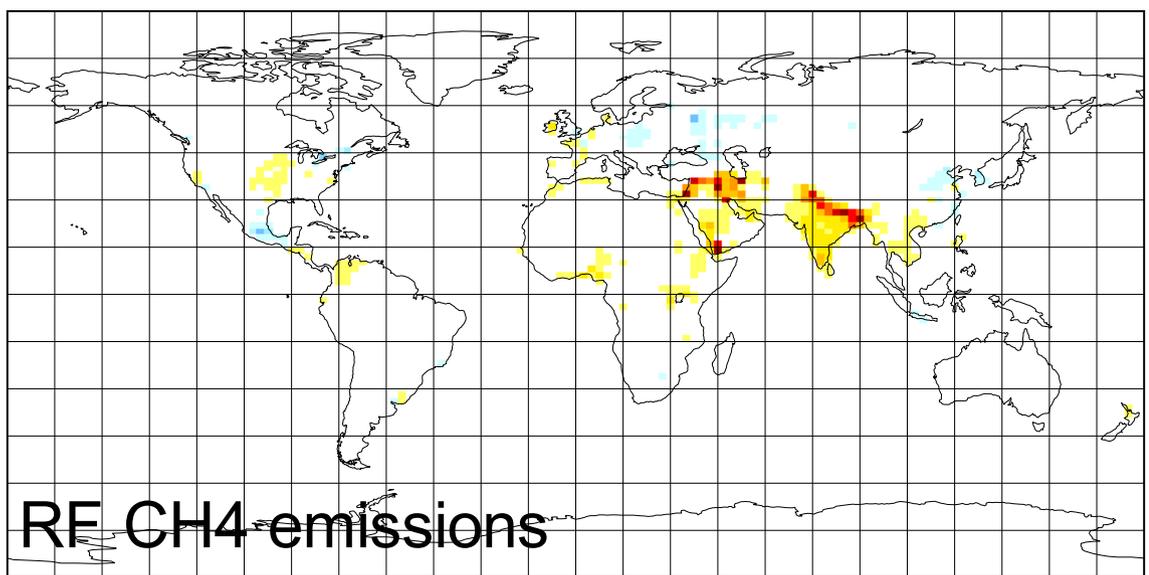
CH4 emissions RF is driven primarily in the Middle East And in Northern India (Gangetic plain)

Total CH4 RF is balanced between chemical reductions in central/south China and increases in US/Europe.

Chemical-driven increases are a consequence of improved air quality standards

Chemical-driven decreases are a consequence of deteriorating air quality.

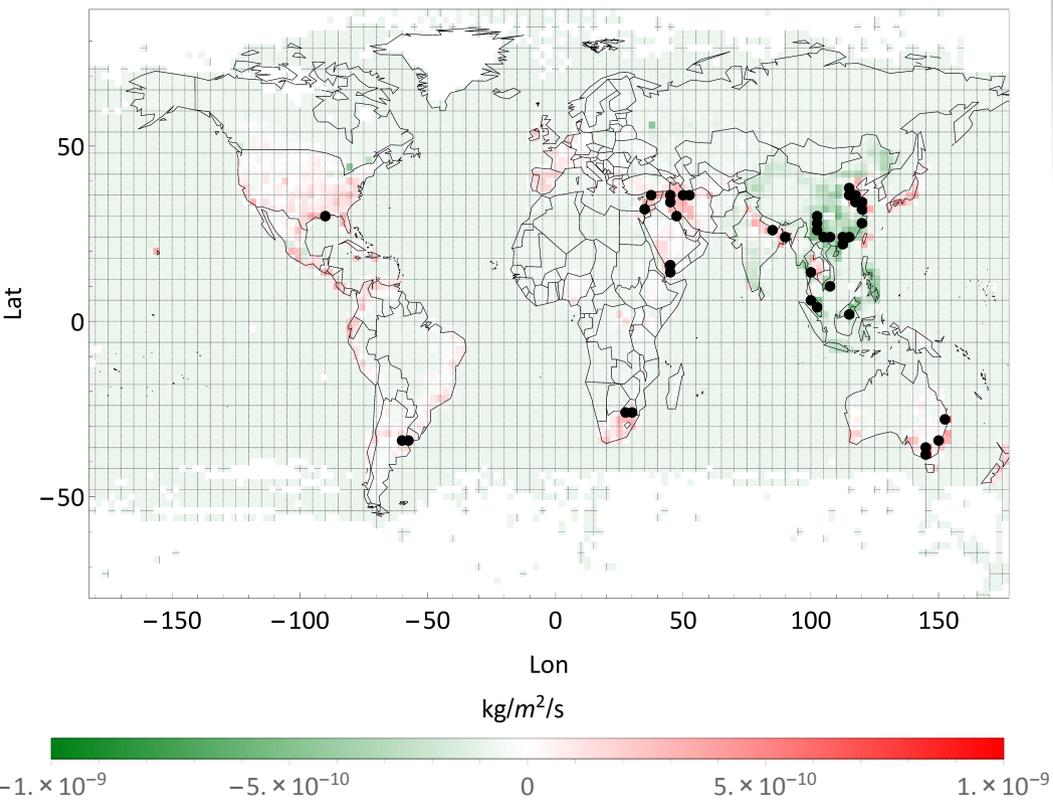
Air quality-climate *disbenefits*



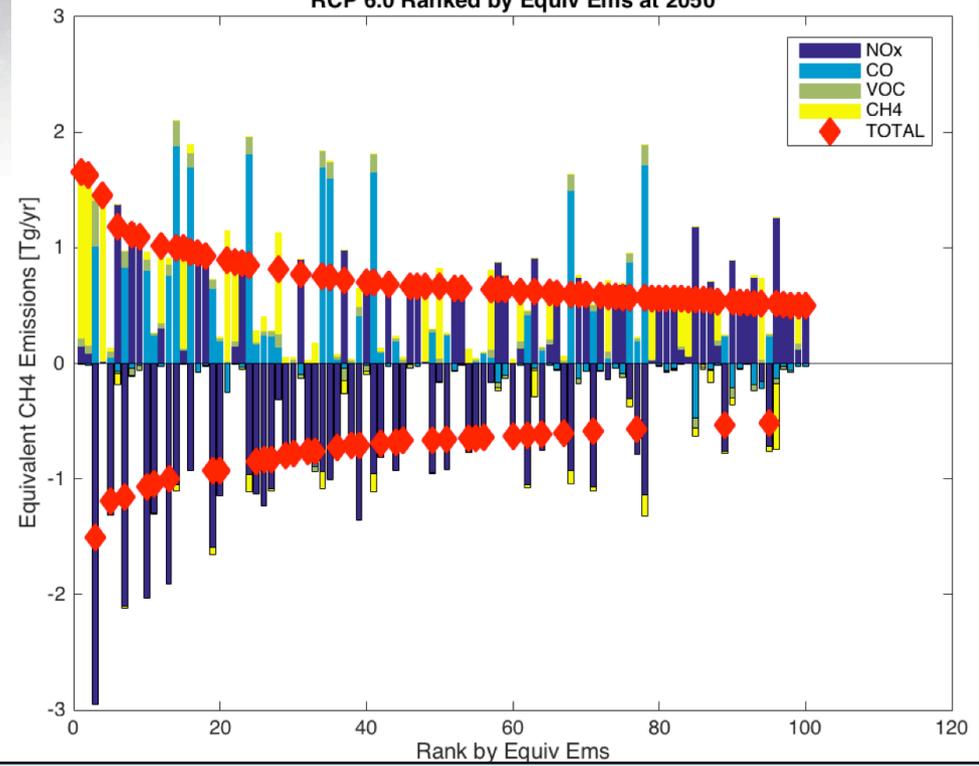


# Ranking total CH4 RF

Total Eq. CH4 emissions—RCP2050



RCP 6.0 Ranked by Equiv Ems at 2050



RF CH4 from NOx, CO, and VOC can be converted to an equivalent emission for RF CH4.

Top 100 equivalent emissions accounts for ~30% of total global impact.

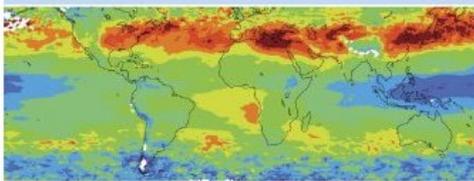
## Largest impacts are in

- Middle East
  - Methane emissions
- Southeast Asia
  - NOx emissions
  - efficient OH loss
- China
  - High CO emissions
  - High NOx emissions



# Supporting mitigation policies

## GLOBAL SOURCES OF LOCAL POLLUTION

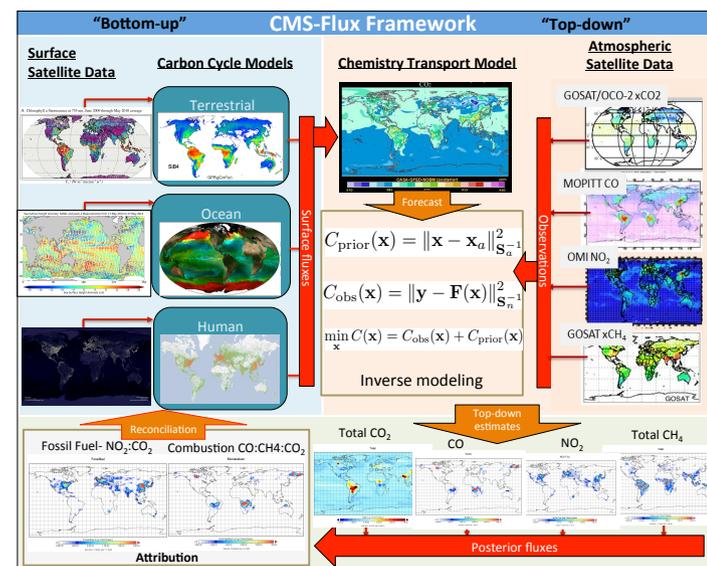
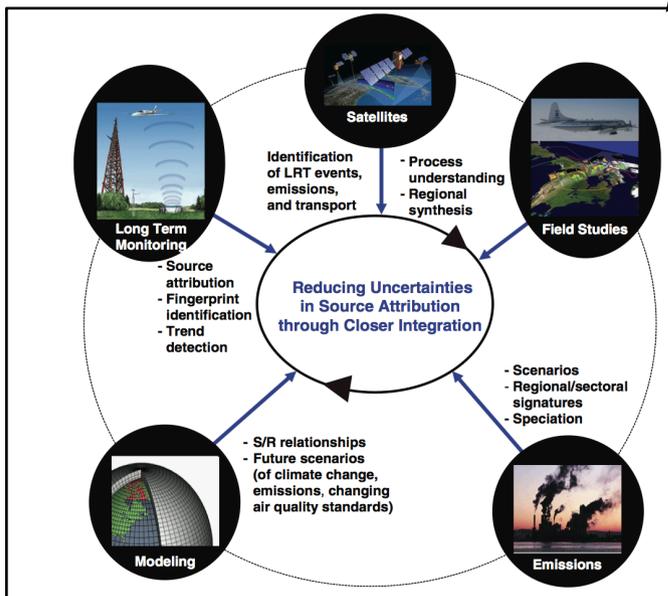
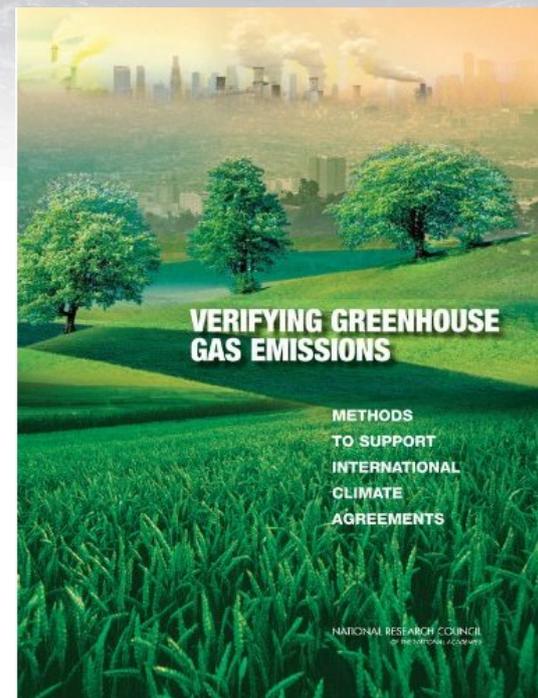


An Assessment of Long-Range Transport of Key Air Pollutants to and from the United States



NATIONAL RESEARCH COUNCIL OF THE NATIONAL ACADEMIES

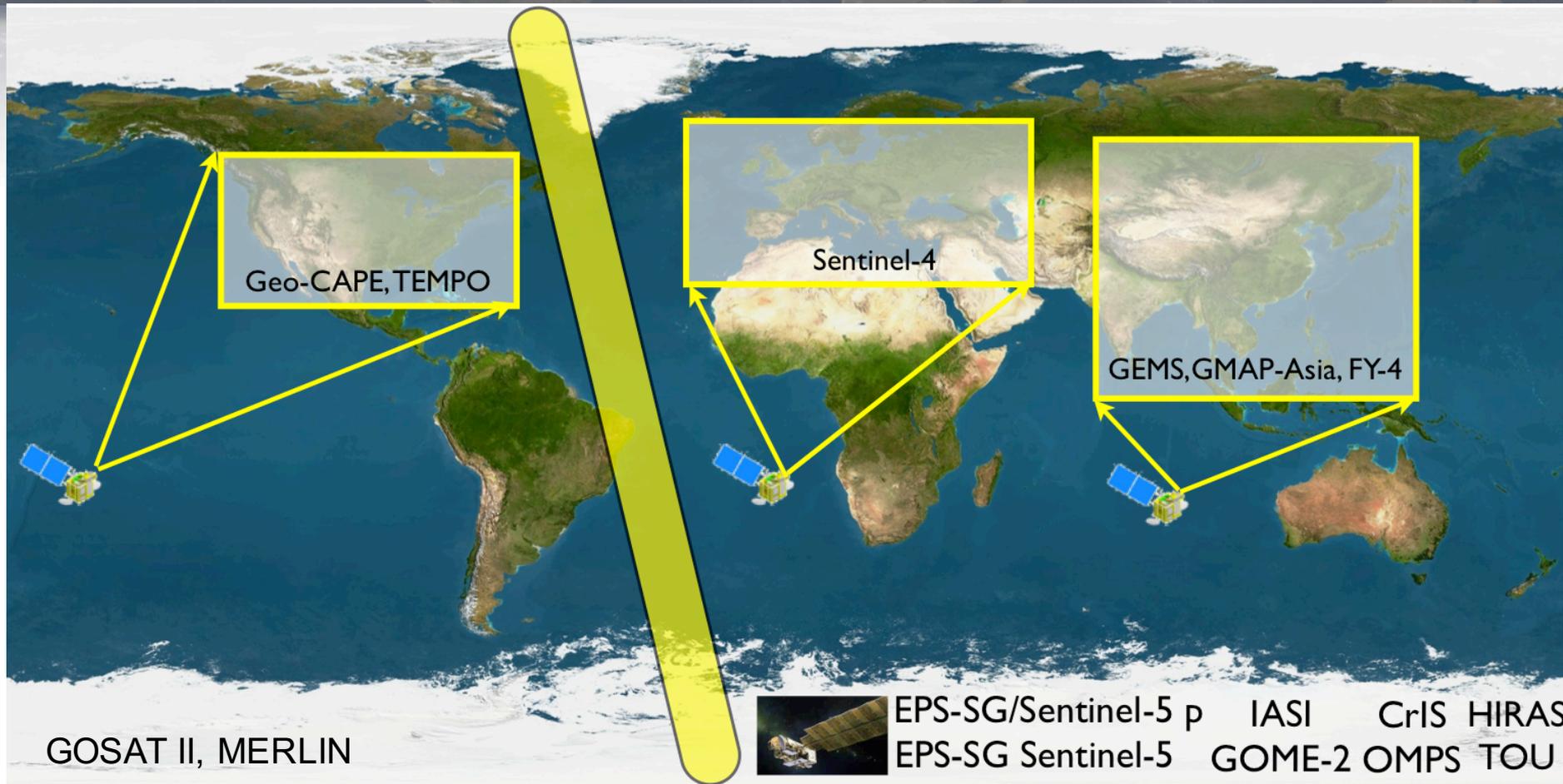
How can we develop science-based mitigation policies and monitor their effectiveness?



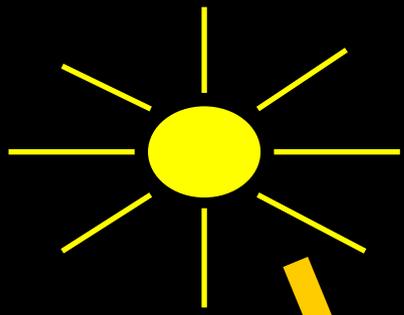


# Toward an Air Quality-Climate Constellation

Bowman et al, Atm.Env. 2013



- LEO:
  - IASI+GOME-2, AIRS+OMI, CrIS+OMPS could provide UV+IR ozone products for more than a decade.
  - Combined UV+IR ozone products from GEO-UVN and GEO-TIR aboard Sentinel 4 (Ingmann *et al*, 2012 *Atm. Env.*)
  - Sentinel 5p (TROPOMI) will provide column CO and CH4.
- GEO
  - TEMPO, Sentinel-4, and GEMS, would provide high spatio-temporal air quality information.



**UV: Provides partial column information**

$$\frac{\Delta S}{\Delta X}$$



**NIR: Provides column information**

$$\frac{\Delta S}{\Delta X}$$



**solar backscatter**

**TIR: Provides FT profile information**

$$\frac{\Delta S}{\Delta X}$$



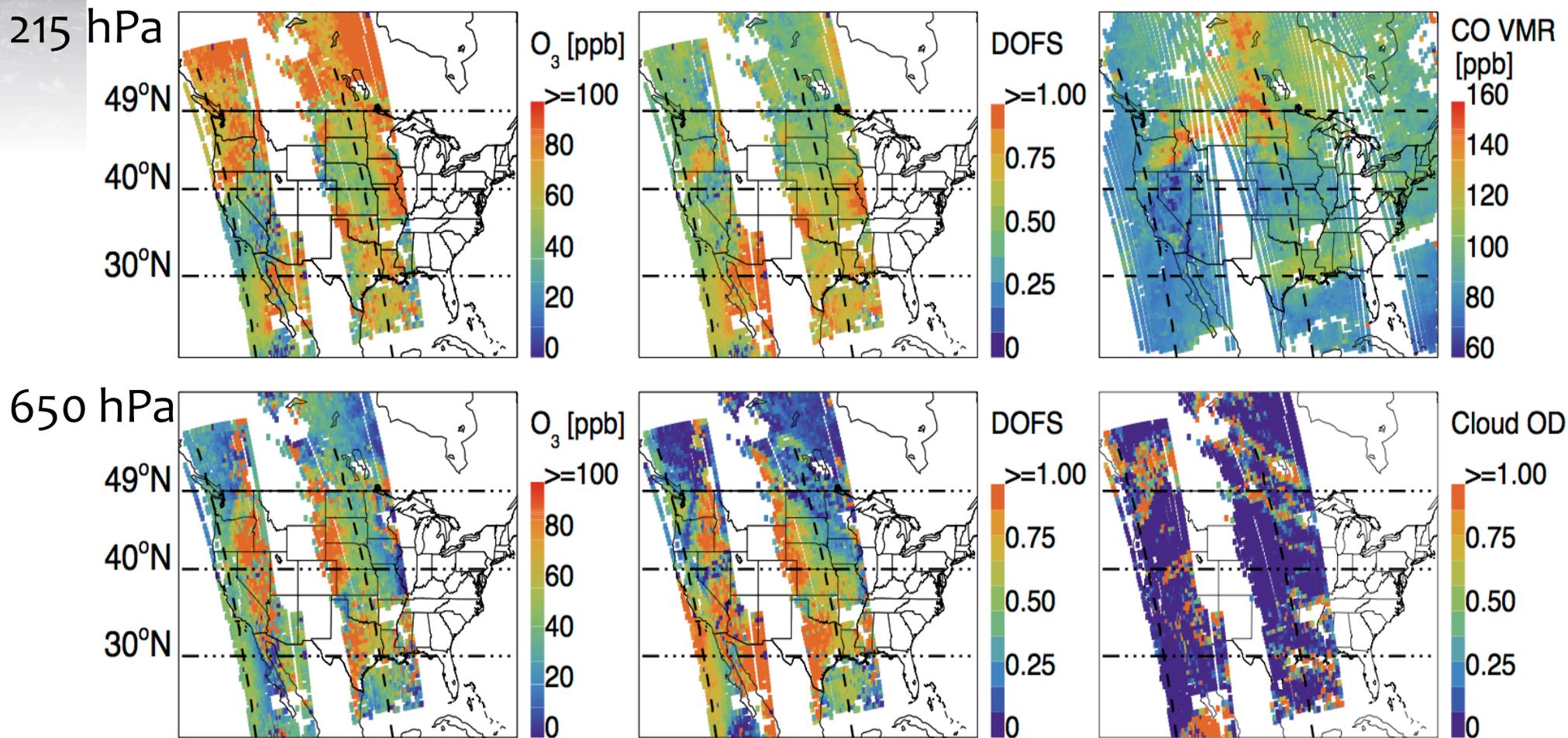
**atmospheric emission**

**surface emission**

# Multi-Spectral Remote Sensing



# AIRS/OMI--Aug 23<sup>rd</sup>, 2006



- The TES optimal estimation retrieval algorithm was applied to the combination of AIRS and OMI radiances to infer O<sub>3</sub> and CO.
- Elevated CO and O<sub>3</sub> in Pacific NW associated with biomass burning.
- DOFS show skill in separating upper and lower troposphere.
- DOFS and Cloud OD show the British Columbia obscured by clouds
- Elevated Texan LT ozone associated with agricultural burning and anthropogenic emissions



# Summary

- What is background ozone?
  - Average background ozone was estimated at 48.3 ppbv or 76.7% of the total ozone in California-Nevada region in summer 2008 but was repartitioned between non-local pollution, which was enhanced by 3.3 ppbv from TES ozone assimilation, and local wildfires, which was reduced by 5.7 ppbv from OMI nitrogen dioxide assimilation.
  - Less than 10 ppbv of local anthropogenic ozone would be possible without violating a 60 ppbv threshold for some regions.
- Is background ozone changing?
  - Tropospheric ozone over Eastern China increased by 7% between 2005 and 2010, which can be equally attributed to an increase in  $\text{NO}_x$  emissions and a temporary increase in stratospheric input.
  - Western US did not decrease despite a 21% reduction in  $\text{NO}_x$  emissions. The expected reduction was offset by a combination of long-range transport from China and the increased stratospheric input.
- How do precursor emissions impact climate?
  - Approximately 8% of the ozone direct radiative forcing from precursor emissions can be attributed to 15 regions, which are predominantly located in China and the United States (US).
  - Under RCP6, the top 100 equivalent emissions accounts for ~30% of total global impact on  $\text{CH}_4$  radiative forcing.
- How can the efficacy of emission changes on global air quality and climate be assessed?
  - A new air quality/climate constellation will provide unprecedented observations
    - New multispectral satellite measurements, e.g., AIRS/OMI, IASI/GOME-2, CrIS/TROPOMI, can provide global information
    - Geostationary sounders, e.g., TEMPO, Sentinel-4, GEMS, will high resolution data.



Backup

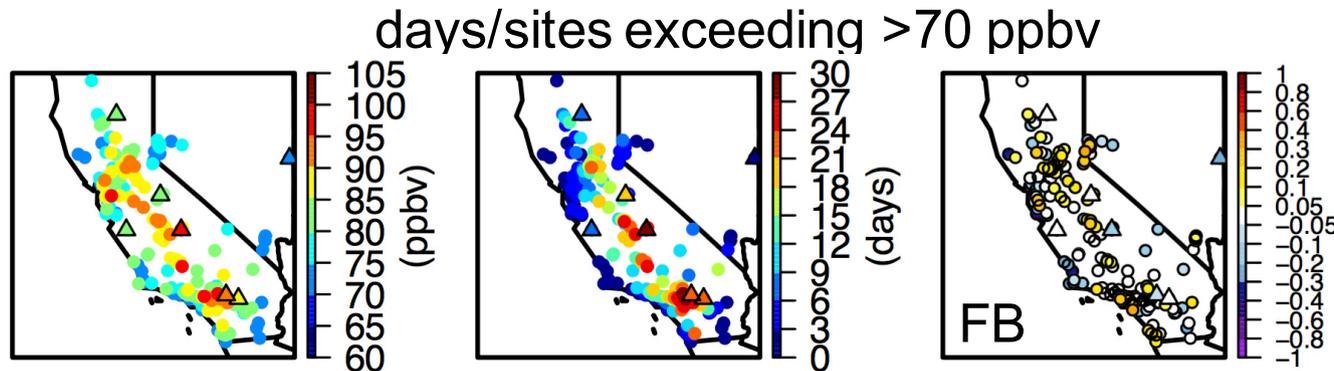
**BACKUP**



# Ozone evaluation

Impact of the multi-scale assimilation on STEM ozone:  
Evaluation using “Fractional Bias (unitless)”:  $2x(\text{model-obs})/(\text{model+obs})$

*Monthly-mean surface (AQS & CASTNET) daily-max 8h average ozone (MDA8)*

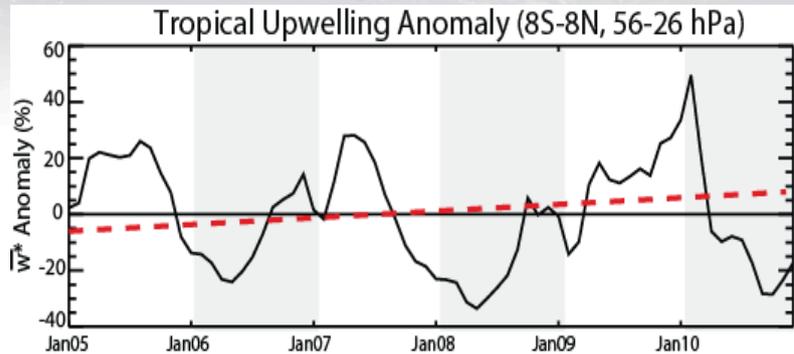


- Improvement occurred after constraining boundary conditions and emissions with satellite observations, despite the remaining positive biases
- Different magnitudes of improvement:
  - 0.13 → 0.10 (23%) for days/sites exceeding 70 ppbv
  - 0.11 → 0.09 (18%) for days/sites exceeding 75 ppbv (previous standard)
  - 0.22 → 0.26 (15%) for all days/sites
  - 0.05 → 0.04 (20%) along aircraft <2 km a.g.l.



# Why is there a “trend” in stratospheric input?

Changes in Stratospheric Circulation From  
MLS H<sub>2</sub>O Measurements



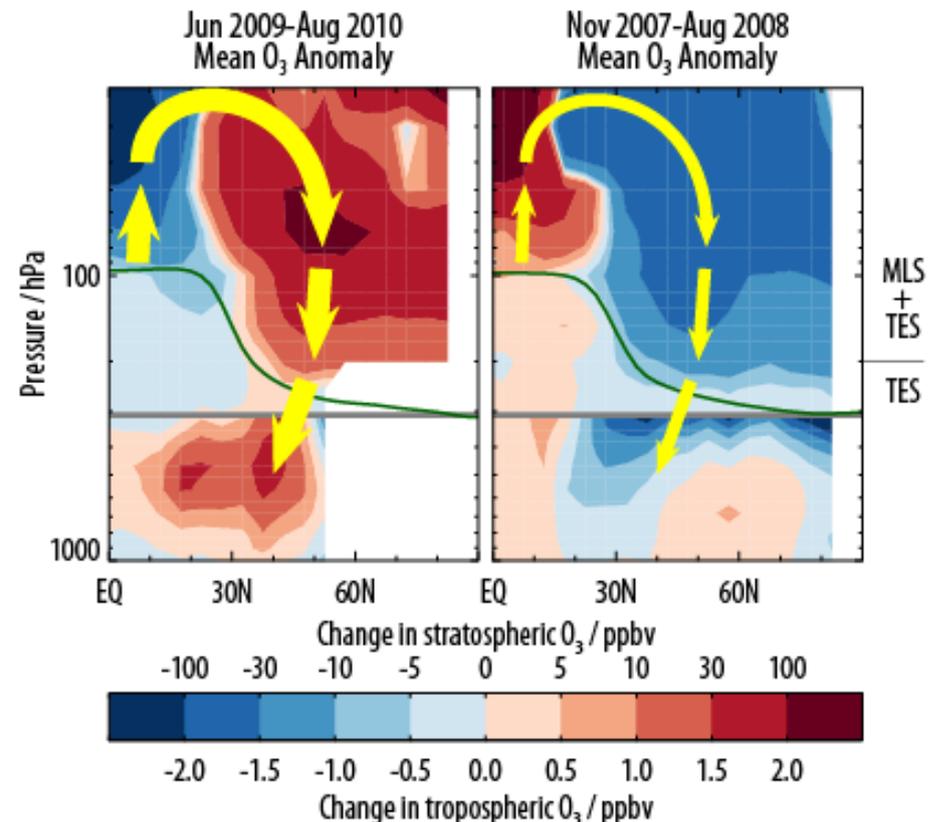
The stratospheric circulation oscillates every ~2 years due to a combination of El Niño / Southern Oscillation and the stratospheric quasi-biennial oscillation

When the circulation is stronger, more stratospheric ozone is carried into the troposphere (Neu et al., 2014).

A large uptick in circulation at the end of the time series gives an apparent trend in the stratospheric contribution to tropospheric ozone.

If a different set of years were used, this “trend” would disappear.

Ozone Changes For Strong and Weak  
Circulation Years

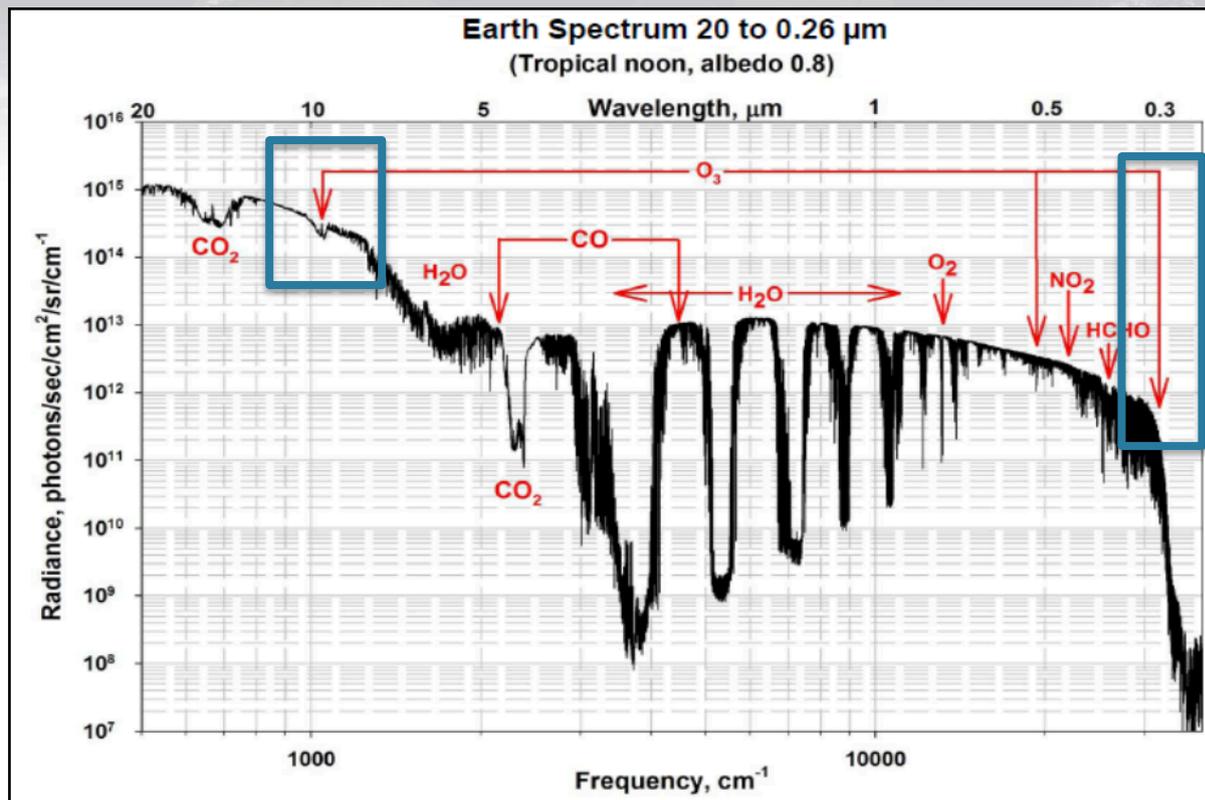




# Two eyes are better than one: IR and UV

## IR

ATMOS  
CLAES  
HALOE  
IMG  
MIPAS  
TES  
AIRS  
ACE-FTS  
IASI  
CrIS



## UV

TOMS  
SAGE  
SBUV  
SOLCE  
OSIRIS  
SCIAMACHY  
OMI  
GOME  
GOME-2

UV and IR measurements provide complimentary sensitivity to ozone. Worden et al, GRL, 2007 and Landgraaf and Hasekamp, JGR, 2007 showed the feasibility of estimating boundary layer ozone. Fu et al, ACP, 2013 and Cuesta et al, ACP, 2013 have demonstrated the potential for TES and IASI.



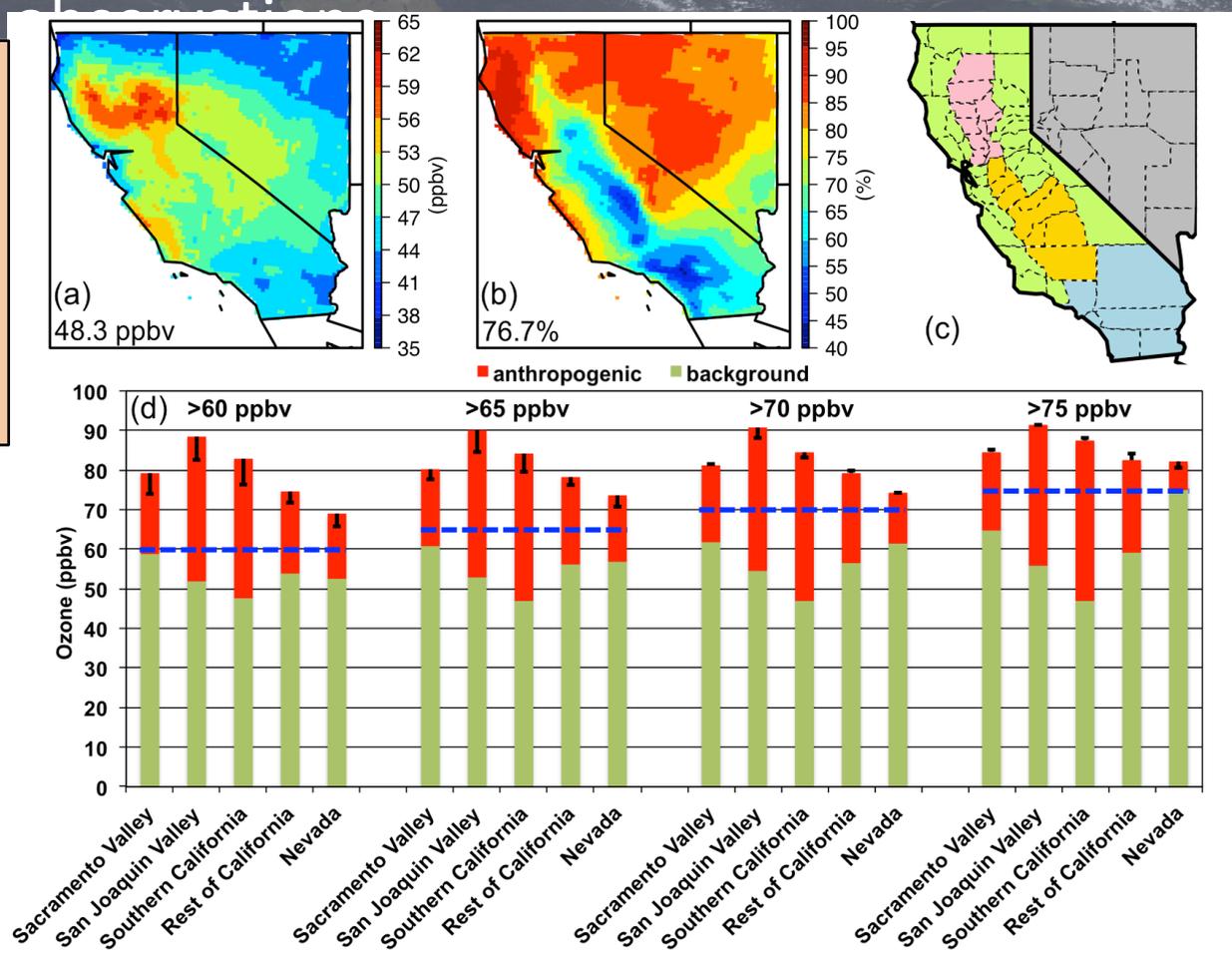
# Background versus local anthropogenic contributions to Western US ozone pollution constrained by Aura TES and OMI

## Science problem:

Proposed reductions in EPA primary ozone standard increases the importance of accurate attribution of background (non-local and local natural) and local human ozone sources.

## Investigation.

Huang *et al.*, JGR (2015) improved ozone source attribution by integrating Tropospheric Emission Spectrometer (TES) ozone and Ozone Monitoring Instrument (OMI) nitrogen dioxide into a state-of-the-art multi-scale assimilation system. Ozone attribution was estimated at surface monitoring sites when total ozone exceeded current and potential thresholds (Fig. c-d).



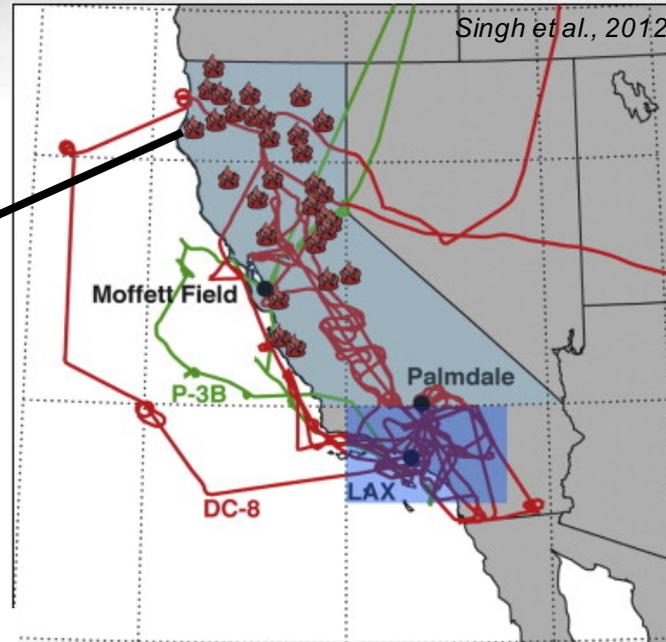
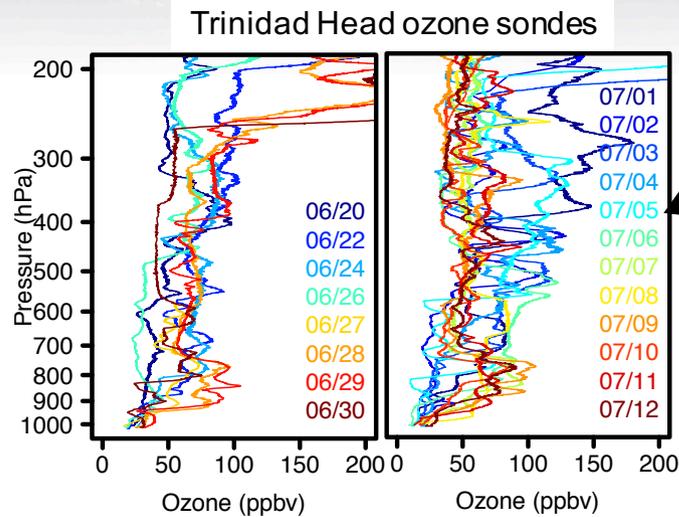
## Key Findings:

- Average background ozone was estimated at 48.3 ppbv or 76.7% of the total ozone in California-Nevada region in summer 2008 (Fig. a-b) but was repartitioned between non-local pollution, which was enhanced by 3.3 ppbv from TES ozone assimilation, and local wildfires, which was reduced by 5.7 ppbv from OMI nitrogen dioxide assimilation.
- Background ozone varied spatially with higher values in many rural regions. Except Southern California, less than 10 ppbv of local anthropogenic ozone would be possible without violating a 60 ppbv threshold.

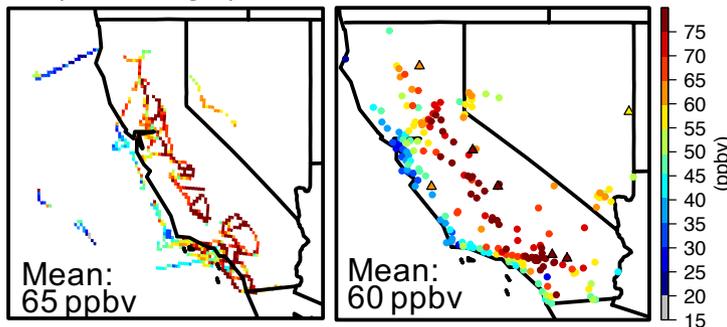
Increases in non-local pollution and local wildfires will require additional reductions in local anthropogenic



# NASA ARCTAS-CARB Campaign June 15-July 14 2008



Ozone along DC-8 (<2 km a.g.l.)

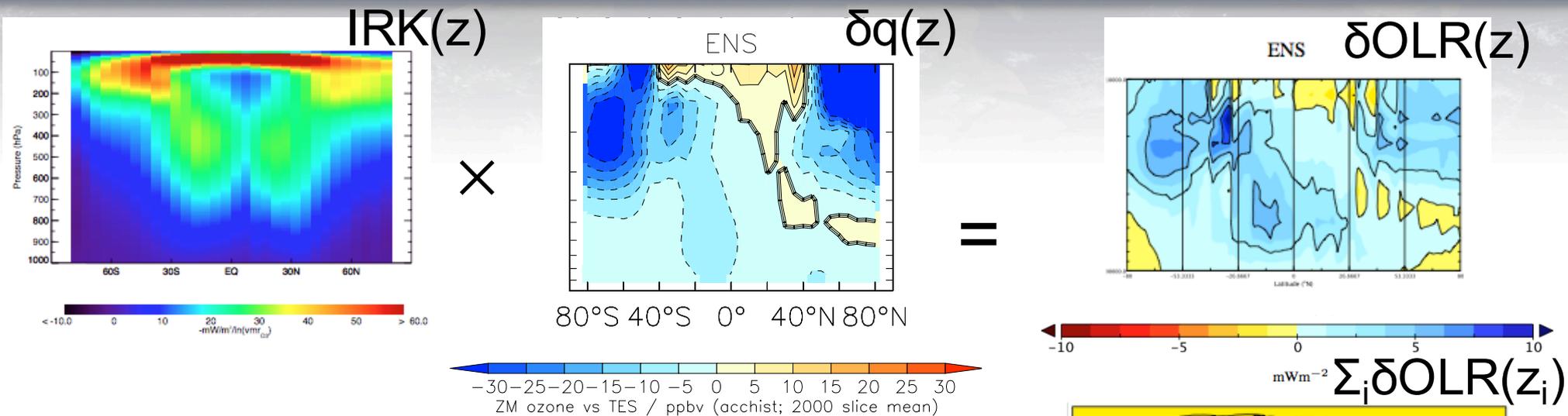


“extra-regional” pollutants mixed with local pollution from various emission sources (e.g., wildfires, urban anthropogenic)

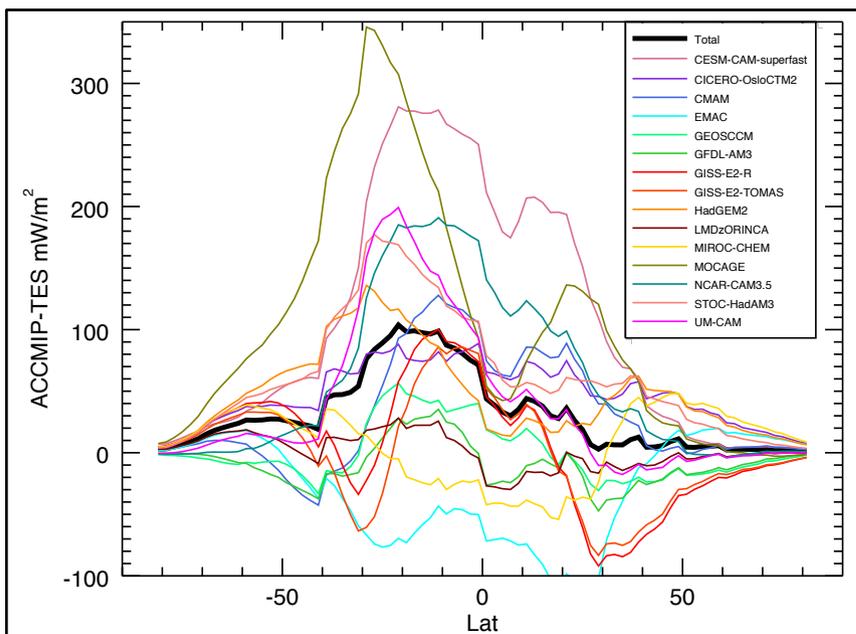
- Strong variability in observed ozone in the free troposphere
- Expanded areas of ozone exceedances near the surface



# OLR bias in chemistry-climate models



The Atmospheric Chemistry-Climate Model Intercomparison Project (**ACCMIP**) estimated historic radiative forcing (RF) and future response using consistent emissions for the IPCC 5<sup>th</sup> assessment. (Lamarque et al, 2013)



Bowman et al, 2013

In the tropics, discrepancies lead to over 300 mWm<sup>-2</sup> for individual models and up to 100 mWm<sup>-2</sup> for the ACCMIP ensemble.

