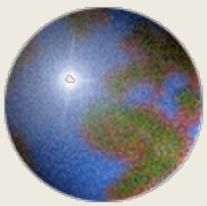

The role of tropospheric ozone in air quality and climate: new insights from satellites, modeling and assimilation.

Kevin W. Bowman

Jet Propulsion Laboratory, California Institute of Technology

Joint Institute for Regional Earth System Science and Engineering, University of California, Los Angeles

Contributors:

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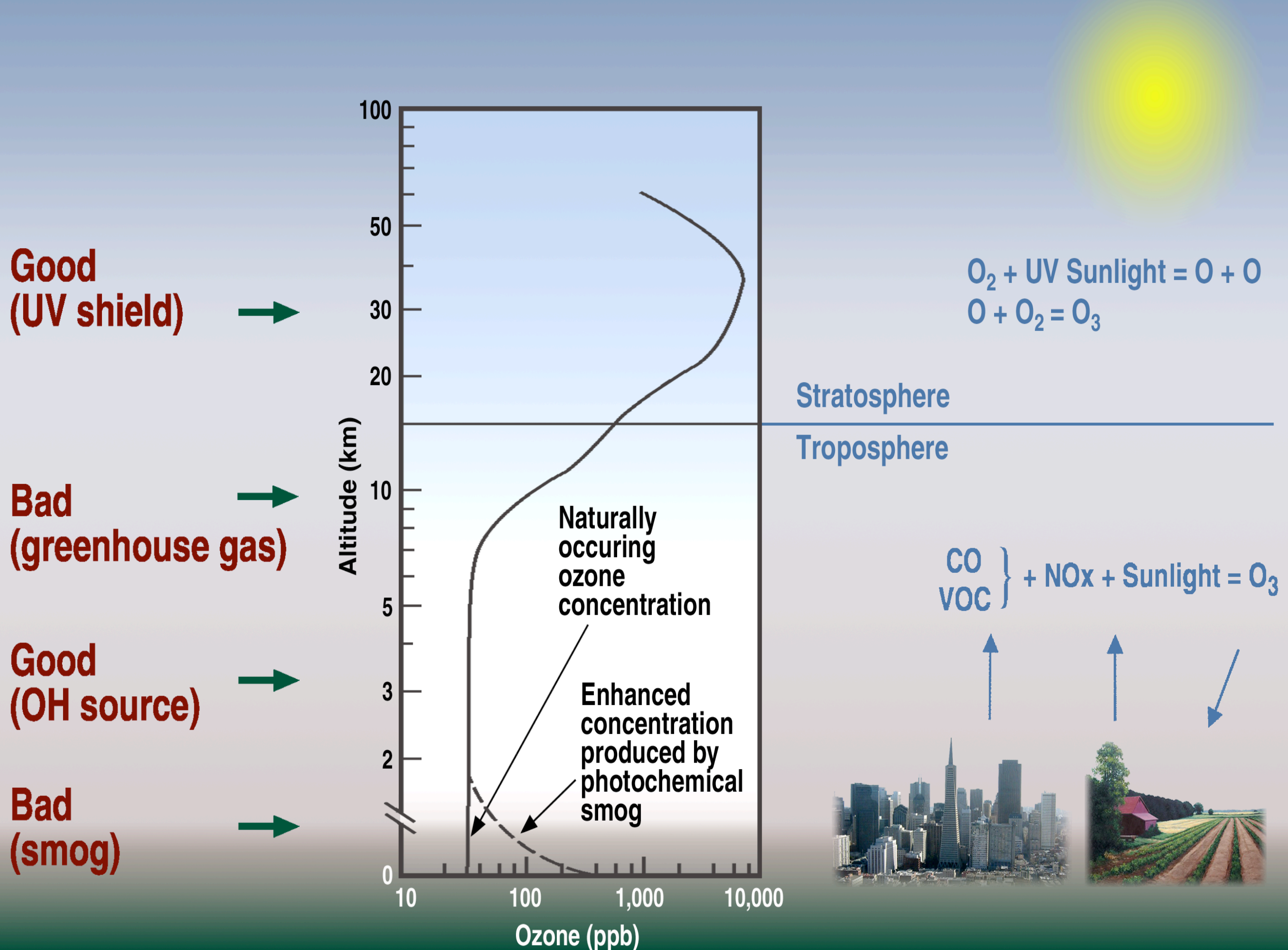
[3]Department of Computer Science, Virginia Tech

Ozone: at the nexus of air quality and climate

Characterization of the vertical distribution of ozone is critical to understanding its role in atmospheric chemistry and climate



Ozone is crucial to air quality

Marion E. Lent makes her way to work as smog dims City Hall in this 1953 photo. (Los Angeles Times), courtesy P. Wennberg-Caltech

Low-level ozone exposure found to be lethal over time

An 18-year study shows an increased annual risk of death from respiratory illnesses, depending on the pollution level. It goes beyond studies that linked brief ozone spikes to short-term effects.

By Thomas H. Maugh II
March 12, 2009

Ozone pollution is a killer, increasing the yearly risk of death from respiratory diseases by 40% to 50% in heavily polluted cities like Los Angeles and Riverside and by about 25% throughout the rest of the country, researchers reported today.

Environmental scientists already knew that increases in ozone during periods of heavy pollution caused short-term effects, such as asthma attacks, increased hospitalizations and deaths from heart attacks.

But the 18-year study of nearly half a million people, reported today in the New England Journal of Medicine, is the first to show that long-term, low-level exposure to the pollutant can also be lethal.

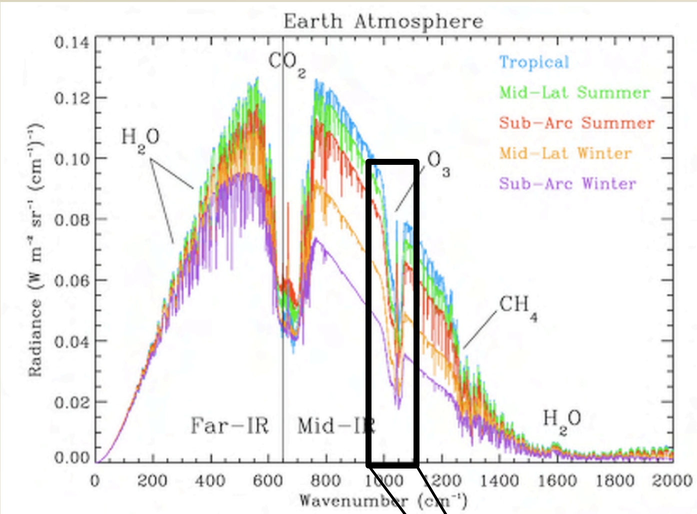
Current standards for ozone pollution cover only eight-hour averages of the colorless gas, but even with that relatively relaxed rule, 345 counties with a total population of more than 100 million people are out of compliance.

The Environmental Protection Agency "has already said that it will revisit the current ozone standards in the country," said Dan Greenbaum, president of the Boston-based Health Effects Institute, one of the study's sponsors.

"Undoubtedly, when it happens these results are going to be a very important part of that review," said Greenbaum, who was not involved in the study.

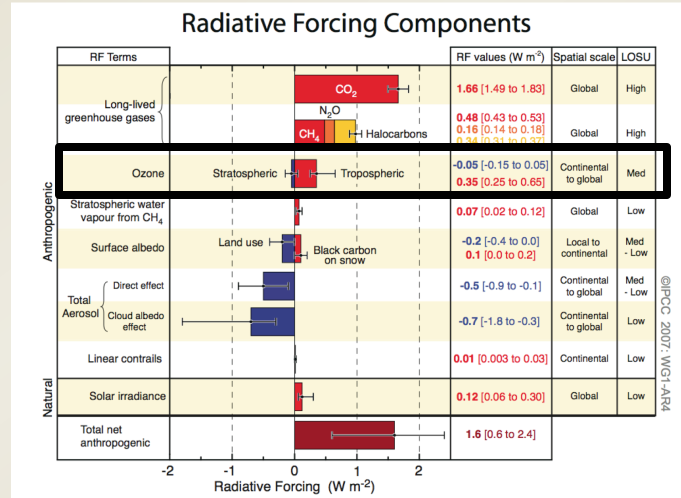
http://www.latimes.com/features/health/la-sci-ozone12-2009mar12,0,6992890.story

Radiative forcing from ozone

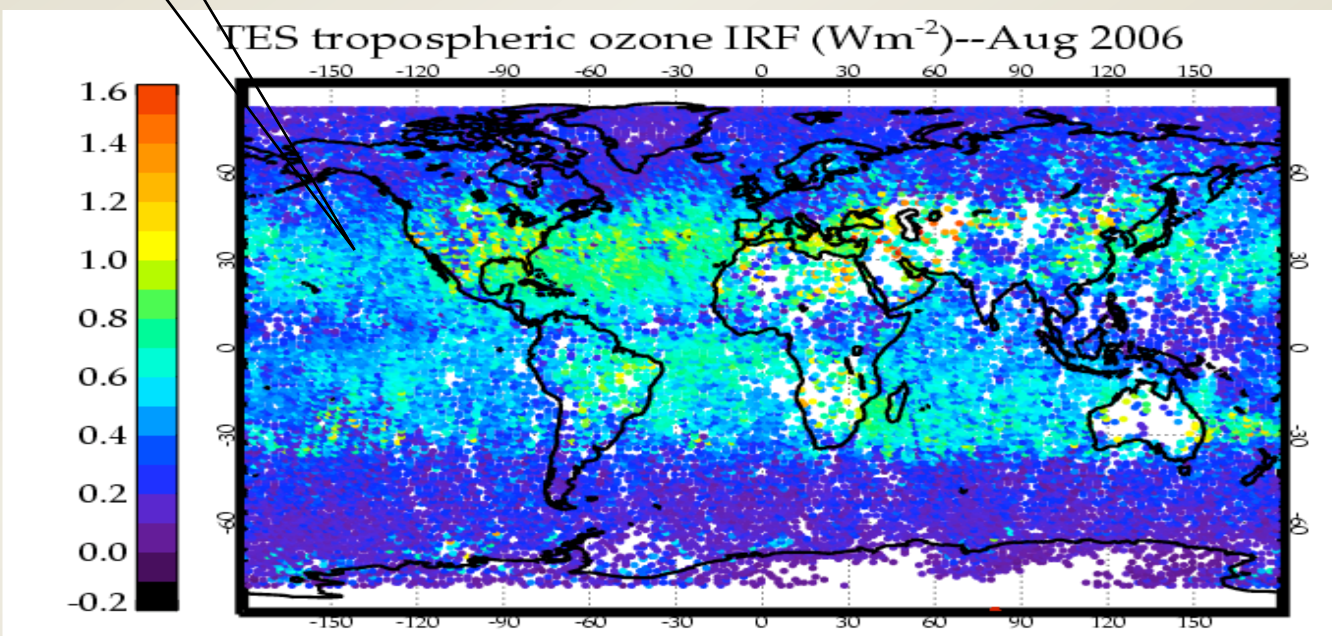


- Radiative forcing from ozone is .35 [0.25-.65] W/m^2

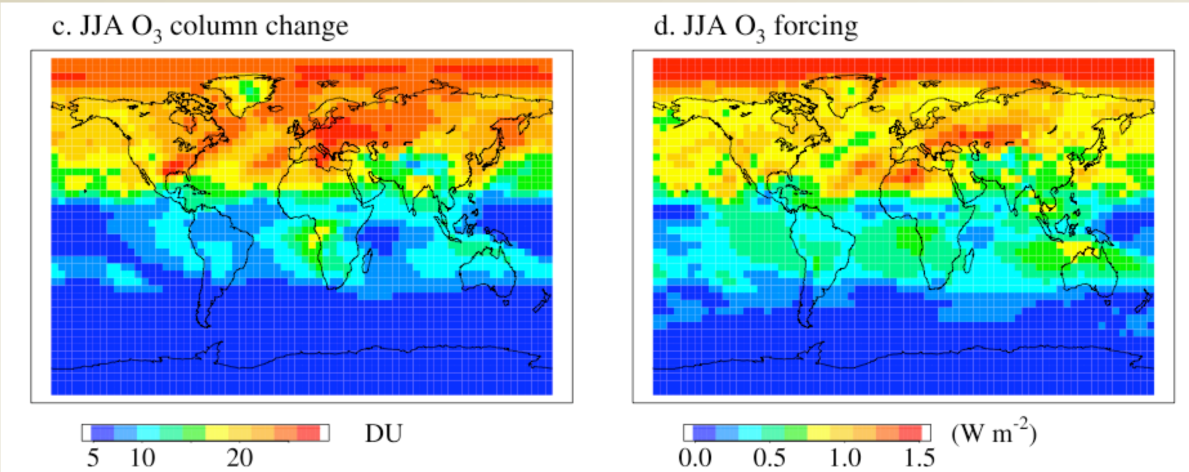
Uncertainties in
- pre-industrial background ozone
- Spatio-temporal ozone distribution



Courtesy, Adetutu Aghedo, JPL



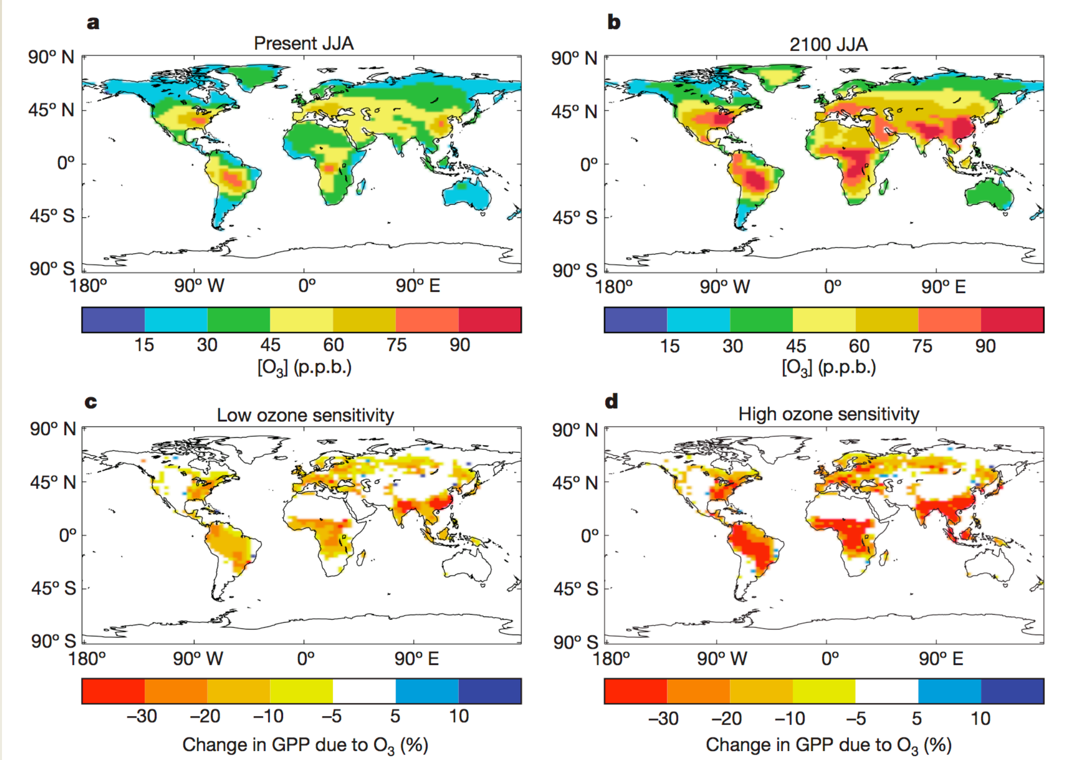
Role of ozone in chemistry-climate coupling



Mickley et al,(2004), *JGR*

Direct effect:
Instantaneous radiative forcing from tropospheric ozone since preindustrial times of .49 W/m^2

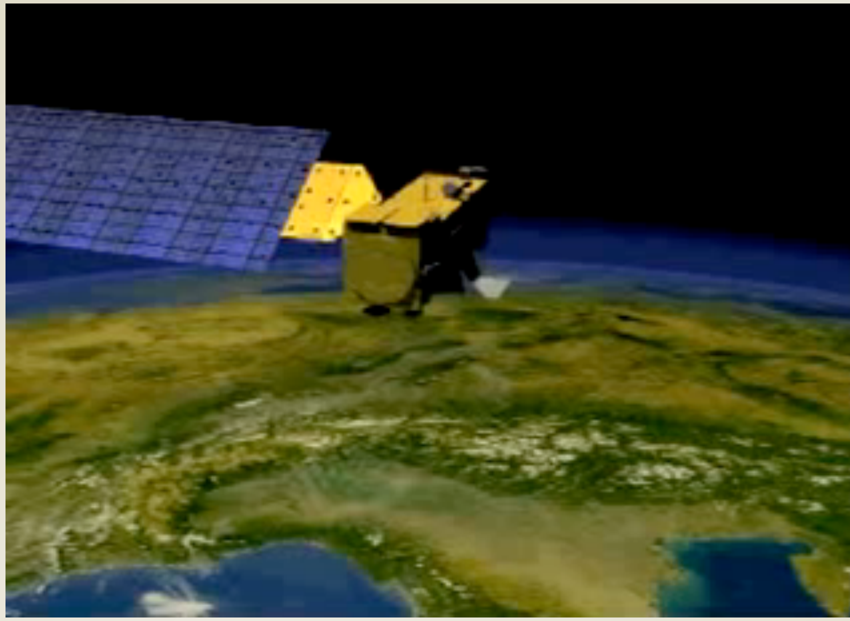
Change of .28^o C increase in global annual mean surface temperature



Indirect effects:
Suppression of carbon uptake by ozone damage to plants could lead to additional 0.62 to 1.09 W/m^2 CO_2 radiative forcing

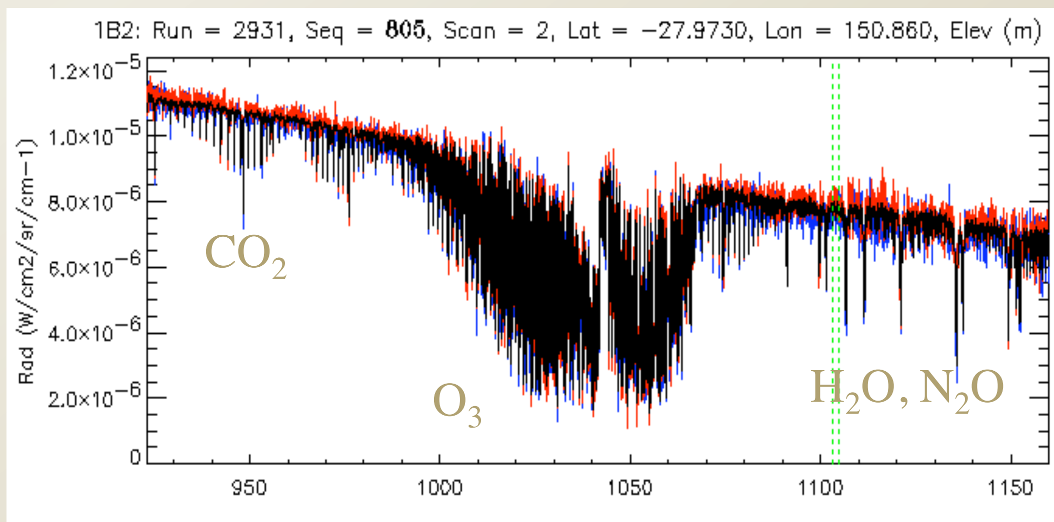
Sitch et al,(2007), *Nature*

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 cm^{-1} (nadir) 0.015 cm^{-1} (hi-res)
Spectral Coverage	650 to 3050 cm^{-1} (3.2 to 15.4 microns)
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

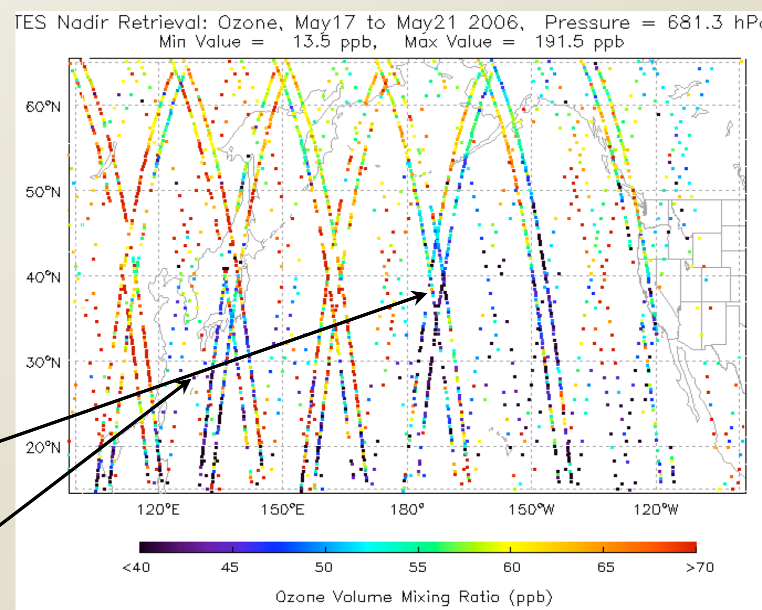


Where does TES observe?

Aura is in a “sun-synchronous” polar orbit with a ~1:43 pm local solar time in the ascending node

Two observational modes:

1. Global survey
   1. Sep 2004-Apr 2005
      - 72 obs/orbit
      - 16 orbits/day
      - 2 nadir/3 limb/obs
      - ~5º separation between obs
   2. May 2005 onwards
      - 73 obs/orbit
      - 3 nadir scans/obs
      - ~1.6º separation between obs
2. Special Observations
   1. Step & Stare
      - Nadir scans covering ~5700 km
      - ~46 km separation between obs.
   2. Transect
      - Nadir scans covering ~480 km
      - ~13 km separation between obs

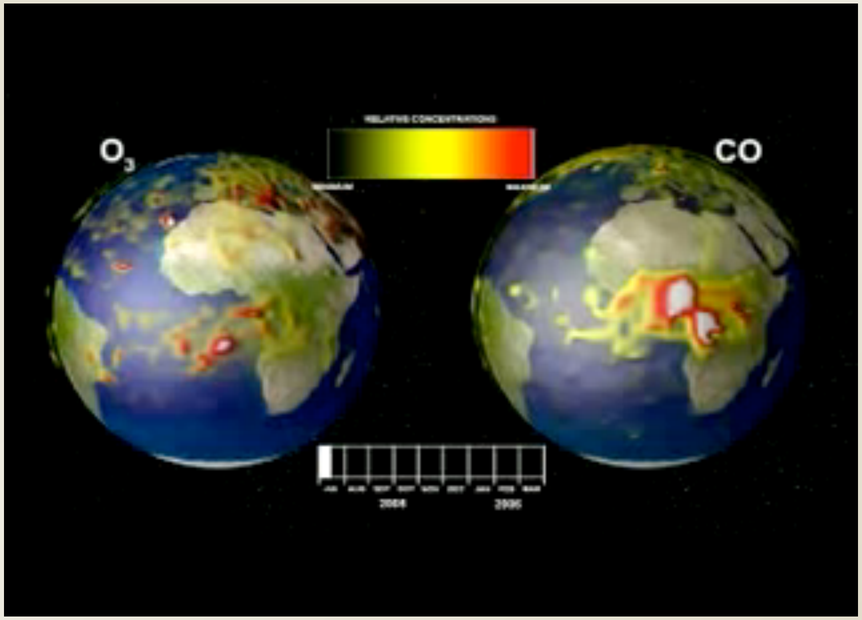


Step/Stare footprints:
~0.4 deg = 45 km apart

Global Survey footprints:
~1.6 deg = 180 km apart

What can TES observe?

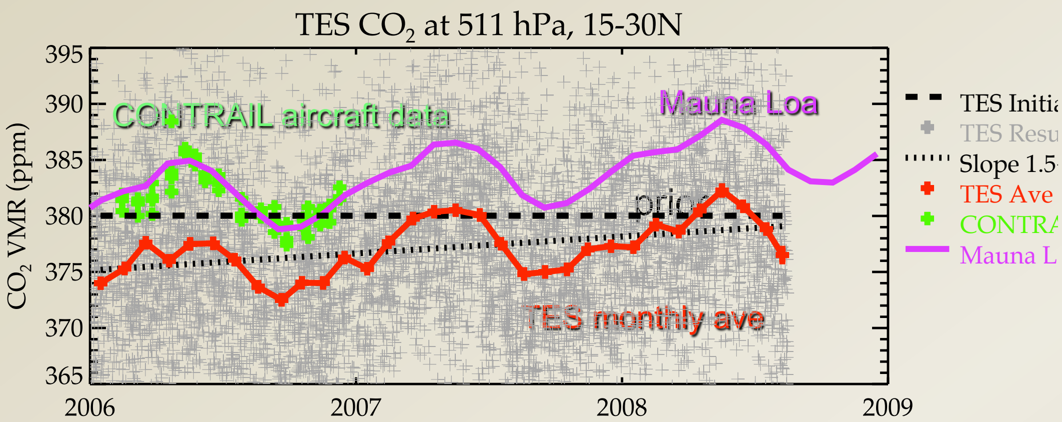
TES observations
Are sensitive to
- Ozone
- Carbon monoxide
- Carbon dioxide
- Temperature
- Water vapor
- HDO
- Emissivity
- Effective cloud parameters
  - Cloud optical depth
  - Cloud height
- Nitric acid
- Sulfur dioxide
- …and more

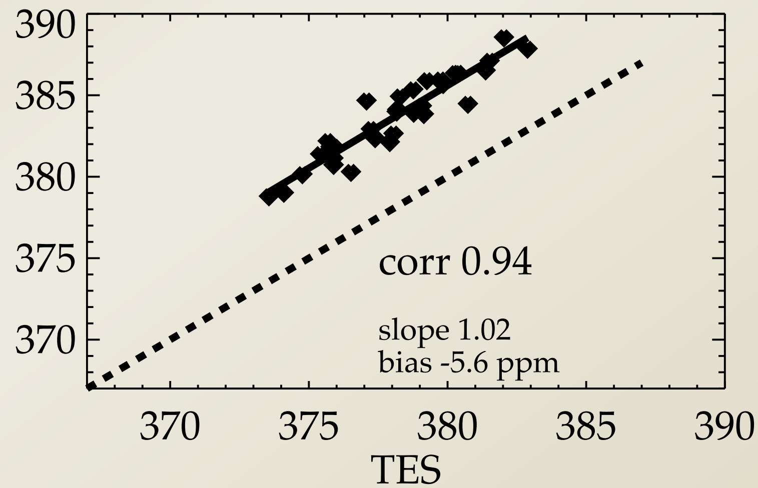


Fundamentally, TES observes *any* atmospheric or surface quantity that emits thermal radiation to space

However, information about these quantities is a function of their spectral sensitivities and natural variability

Mid-tropospheric CO_2





Monthly averages of ~200 targets

Monthly mean error is 0.9 ppm with 5.6 ppm bias

Bias close to estimated spectroscopic error of ~4 ppm

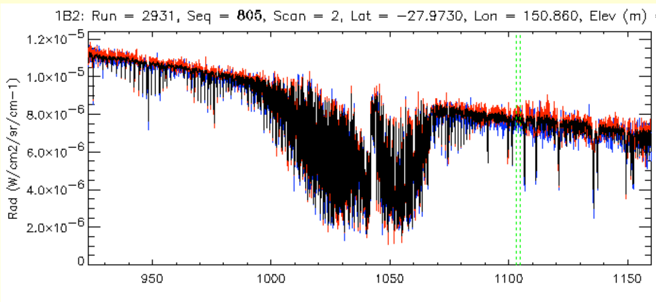
Greatest sensitivity in middle Troposphere

Validated for low cloud, ocean, 40S to 40N targets

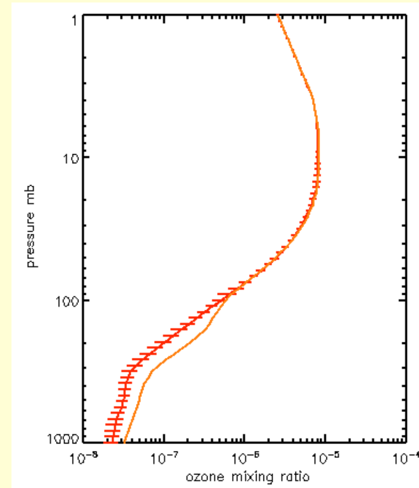
Courtesy S. Kulawik JPL

TES observation operator:

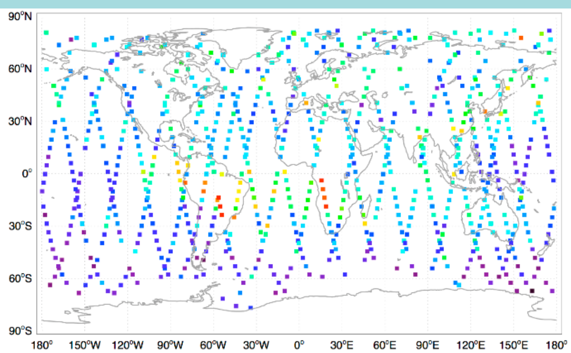
connecting measurements to assimilation



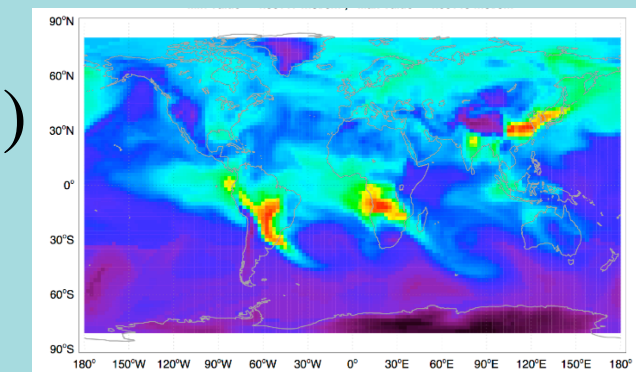
$$\left\|\mathbf{y}-\mathbf{F}(\mathbf{x}_a)\right\|^2_{\mathbf{S}_n^{-1}}+\left\|\mathbf{x}-\mathbf{x}_a\right\|^2_{\mathbf{S}_a^{-1}}$$



$$\hat{\mathbf{x}}=\mathbf{x}_a+\mathbf{A}(\mathbf{x}-\mathbf{x}_a)+\mathbf{G}\mathbf{n}$$



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



$$\sum_i\left\|\hat{\mathbf{x}}_i-\mathbf{H}_i(\mathbf{x})\right\|^2_{(\mathbf{G}_i\mathbf{S}_n^i\mathbf{G}_i^T)^{-1}}+\left\|\mathbf{x}_0-\mathbf{x}_B\right\|^2_{\mathbf{B}^{-1}}$$

What is the impact of background ozone on air quality?

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 113, D18307, doi:10.1029/2007JD009341, 2008

Click Here for Full Article

Estimating the summertime tropospheric ozone distribution over North America through assimilation of observations from the Tropospheric Emission Spectrometer

M. Parrington,[1] D. B. A. Jones,[1] K. W. Bowman,[2] L. W. Horowitz,[3] A. M. Thompson,[4] D. W. Tarasick,[5] and J. C. Witte[6,7]

GEOPHYSICAL RESEARCH LETTERS, VOL. 36, L04802, doi:10.1029/2008GL036935, 2009

Click Here for Full Article

Impact of the assimilation of ozone from the Tropospheric Emission Spectrometer on surface ozone across North America

M. Parrington,[1] D. B. A. Jones,[1] K. W. Bowman,[2] A. M. Thompson,[3] D. W. Tarasick,[4] J. Merrill,[5] S. J. Oltmans,[6] T. Leblanc,[7] J. C. Witte,[8] and D. B. Millet[9]

Chemical Data Assimilation Methodology

Sequential Sub-optimal Kalman filter

$$\hat{\mathbf{x}}_k^a = \mathbf{x}_k^f + \mathbf{K}_k[\mathbf{y}^{\mathrm{obs}} - \mathbf{H}_k\mathbf{x}_k^f]$$

Kalman Gain Matrix: $$\mathbf{K}_k = (\mathbf{H}_k\mathbf{P}_k^f\mathbf{H}_k^{\mathrm{T}} + \mathbf{R}_k)^{-1}\mathbf{P}_k^f\mathbf{H}_k^{\mathrm{T}}$$

Analysis Error Cov. Matrix: $$\mathbf{P}_k^a = (\mathbf{I} - \mathbf{K}_k\mathbf{H}_k)\mathbf{P}_k^f$$

- Observation operator (**H**) accounts for TES averaging kernels and a priori profiles
- Analysis error variance transported as a passive tracer

Models and Data Streams

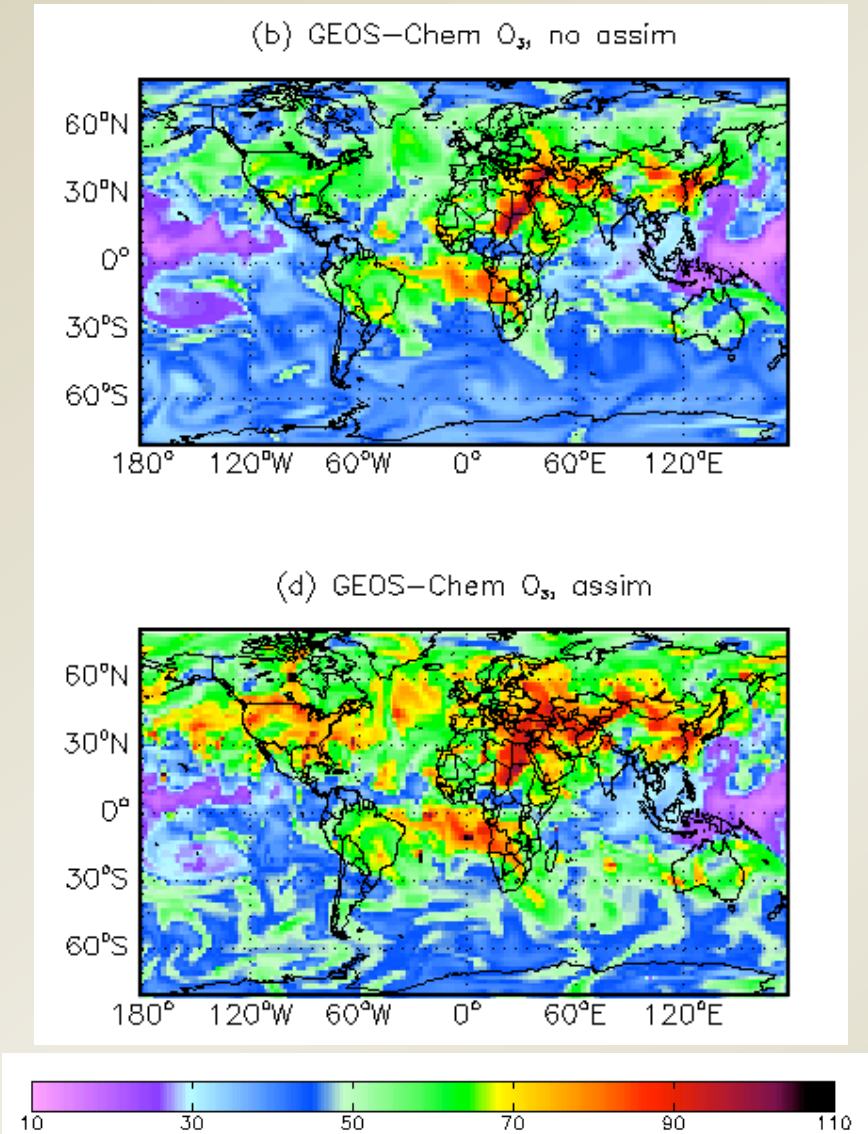
- **GEOS-Chem with full nonlinear tropospheric chemistry**
- **O_3 and CO profile retrievals from TES for July 1 through August 31 2006**
- **6-hour analysis cycle**
- **Assumed forecast error of 20% for CO and 50% for O_3**
- **Neglected horizontal correlations in forecast and observation error covariance matrices**
- **Results presented for 15 August 2006**

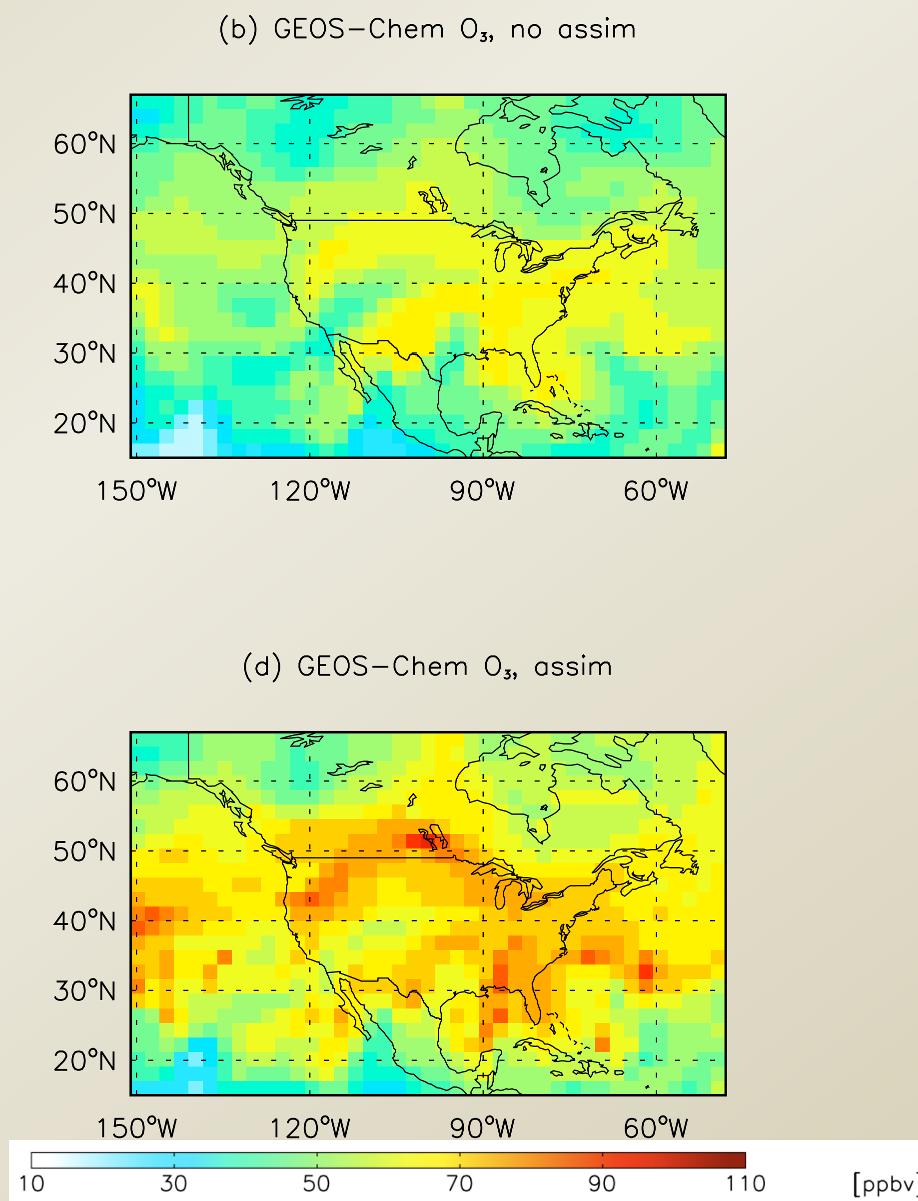
GEOS-Chem

- **Chemical transport model**
- **2.0° latitude x 2.5° longitude or 4° latitude x 5° longitude, 55 vertical levels (top level approx. 0.01 hPa)**
- **Model transport driven by GEOS-4 GMAO analyses**
- **Linoz parameterization of stratospheric ozone vertical distribution**

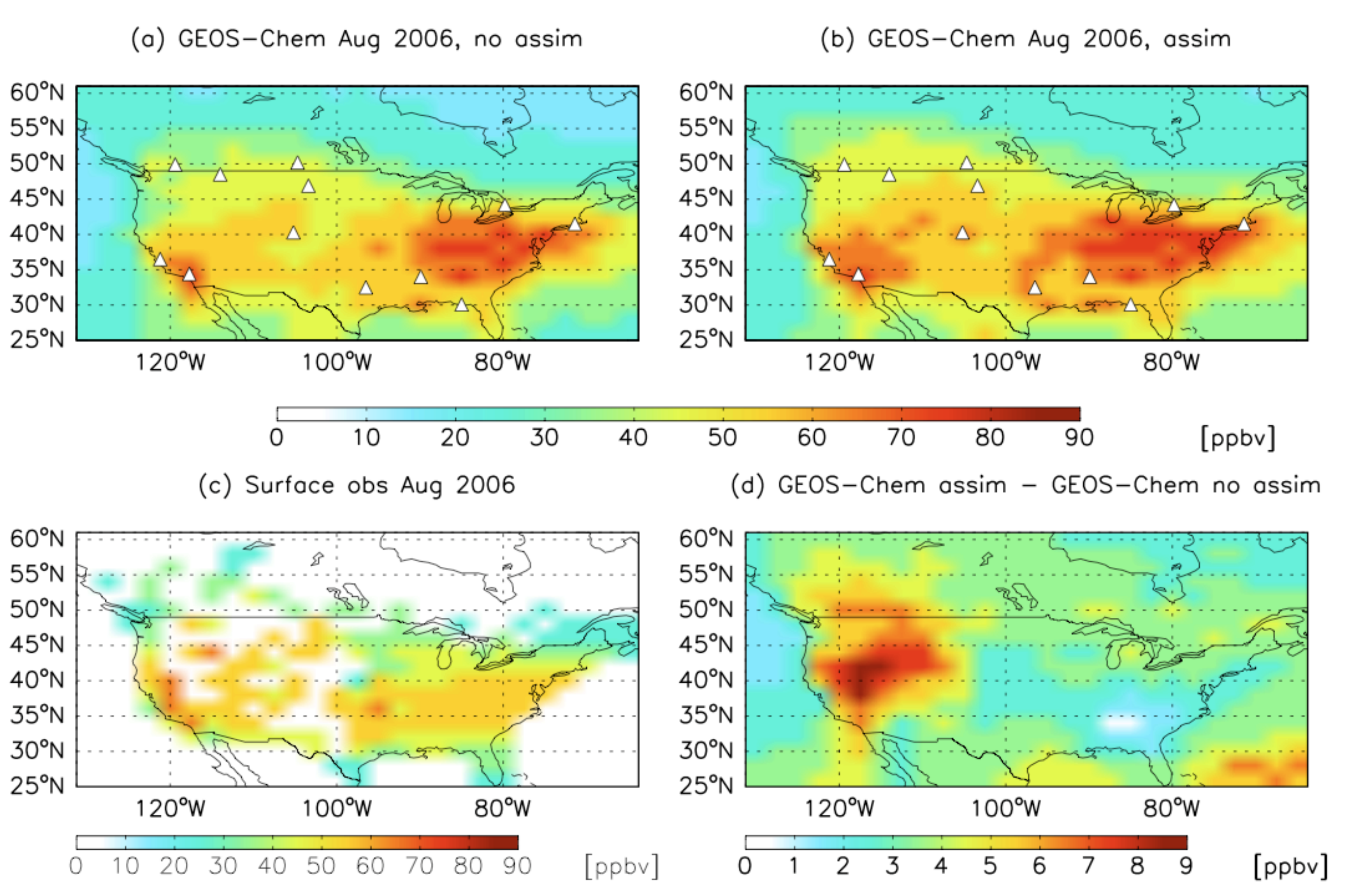
Impact of ozone assimilation at 5km

Aug 15, 2006





Impact of assimilation on surface ozone



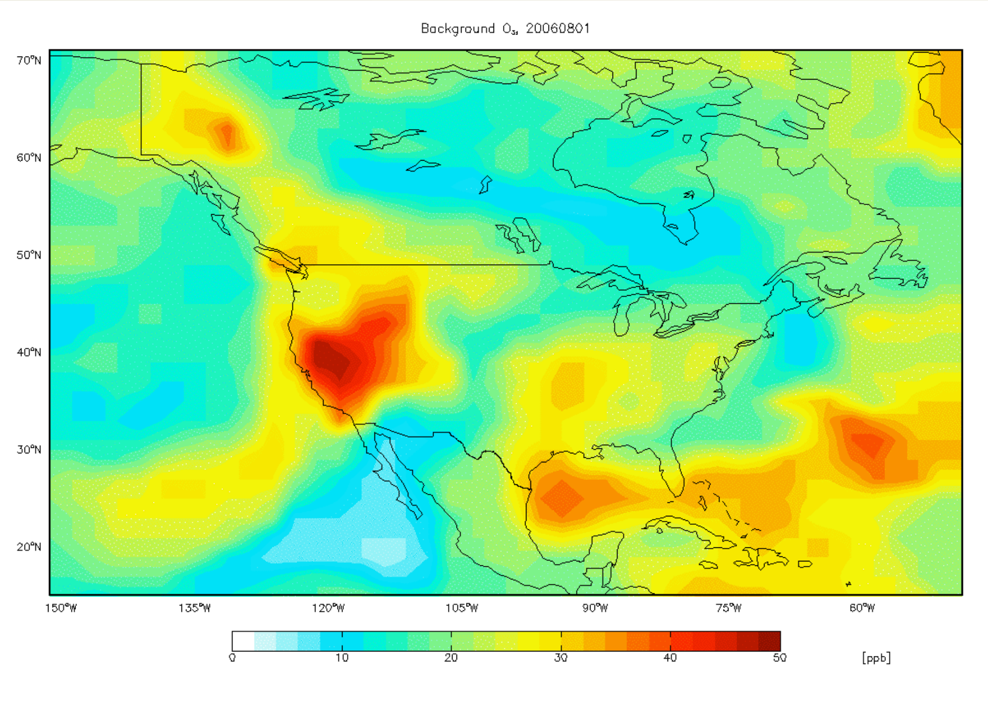
Contribution of background ozone to boundary layer ozone

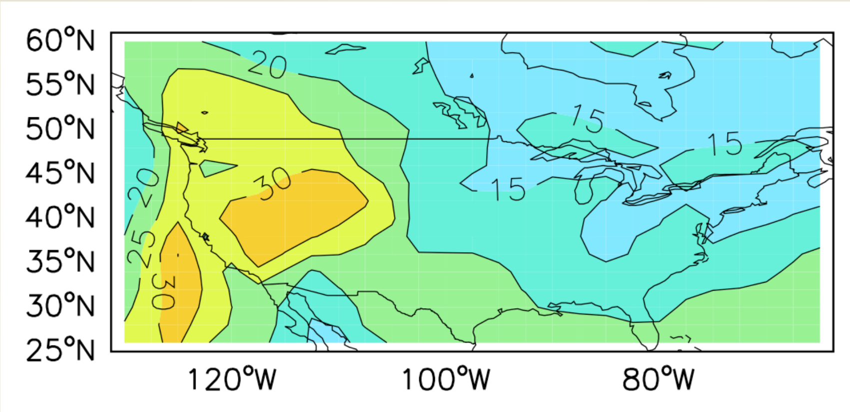
Background ozone is defined as ozone generated outside North American boundary laye

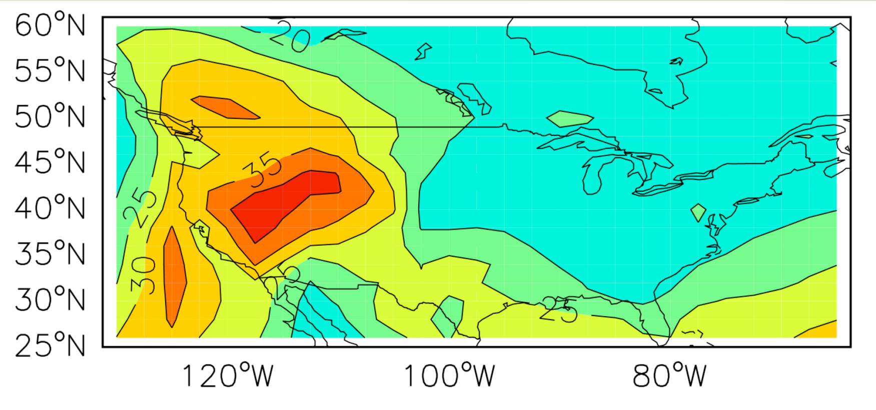
Calculated using a tagged O3 simulation where the O3 chemistry is linearized with 24-hr averaged production rates and loss frequencies for odd oxygen

Animation shows daily mean background ozone.

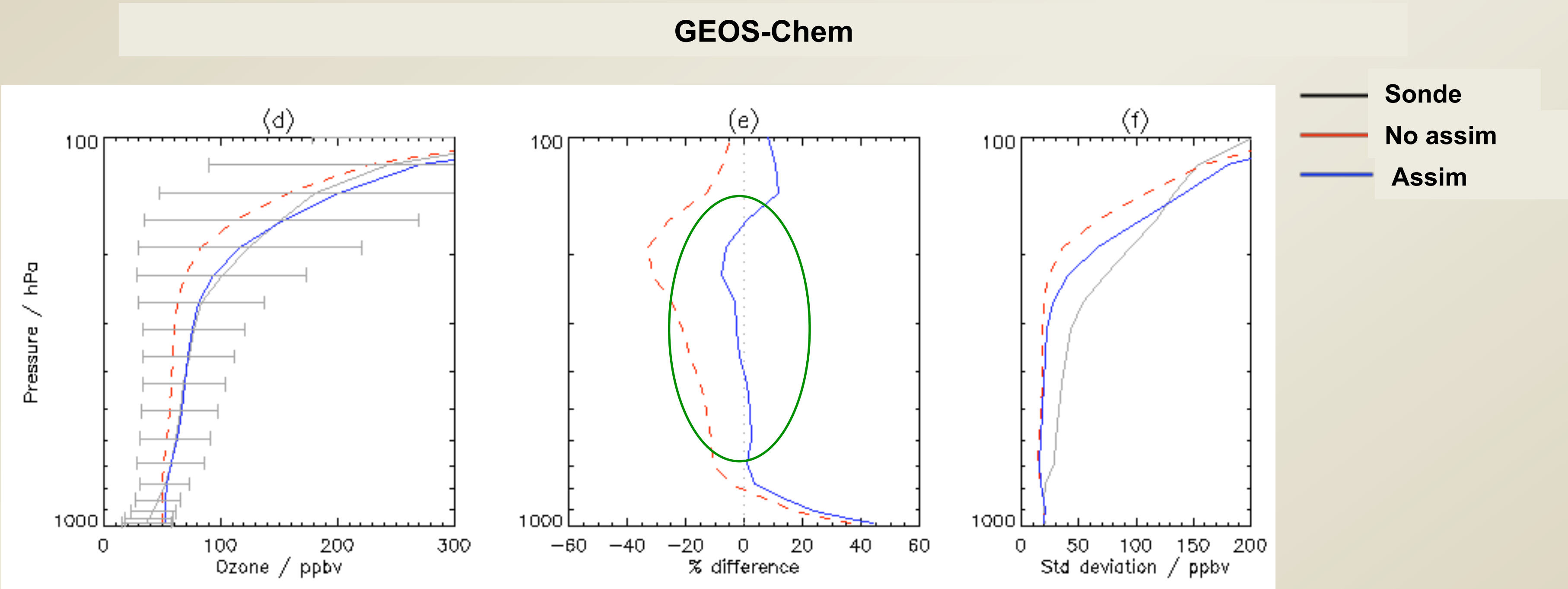
Background O3 values with the assimilation are about 20 – 40 ppb across North American, an increase of 20 – 30% over background O3 in standard model







Comparison to IONS ozone sondes

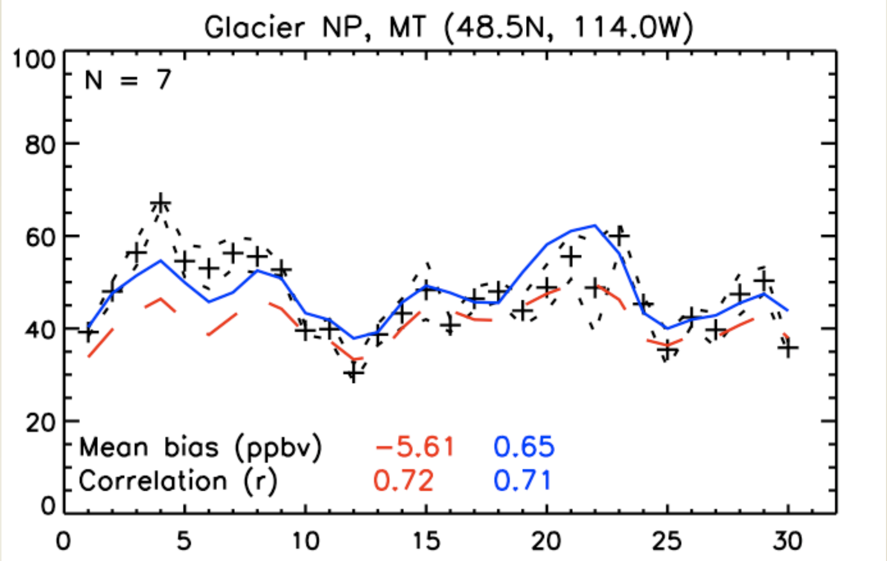


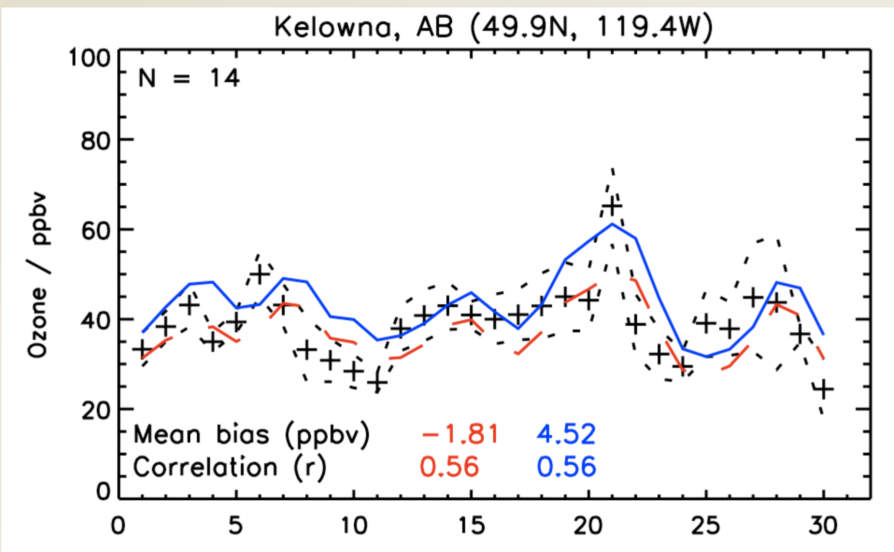
Significant improvement in mid-tropospheric ozone distribution following assimilation.

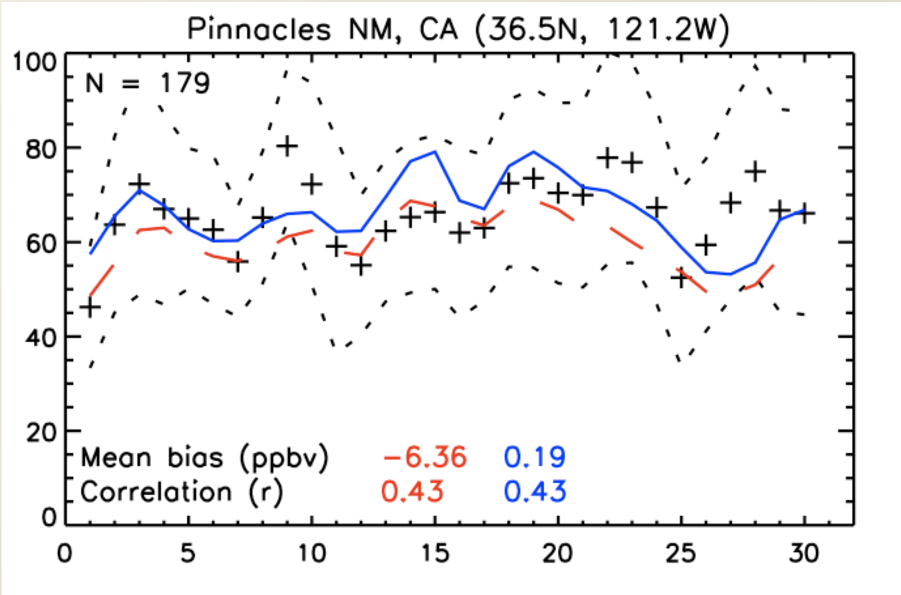
Marginal improvement in mid-tropospheric ozone variance following assimilation.

Comparison to surface sites

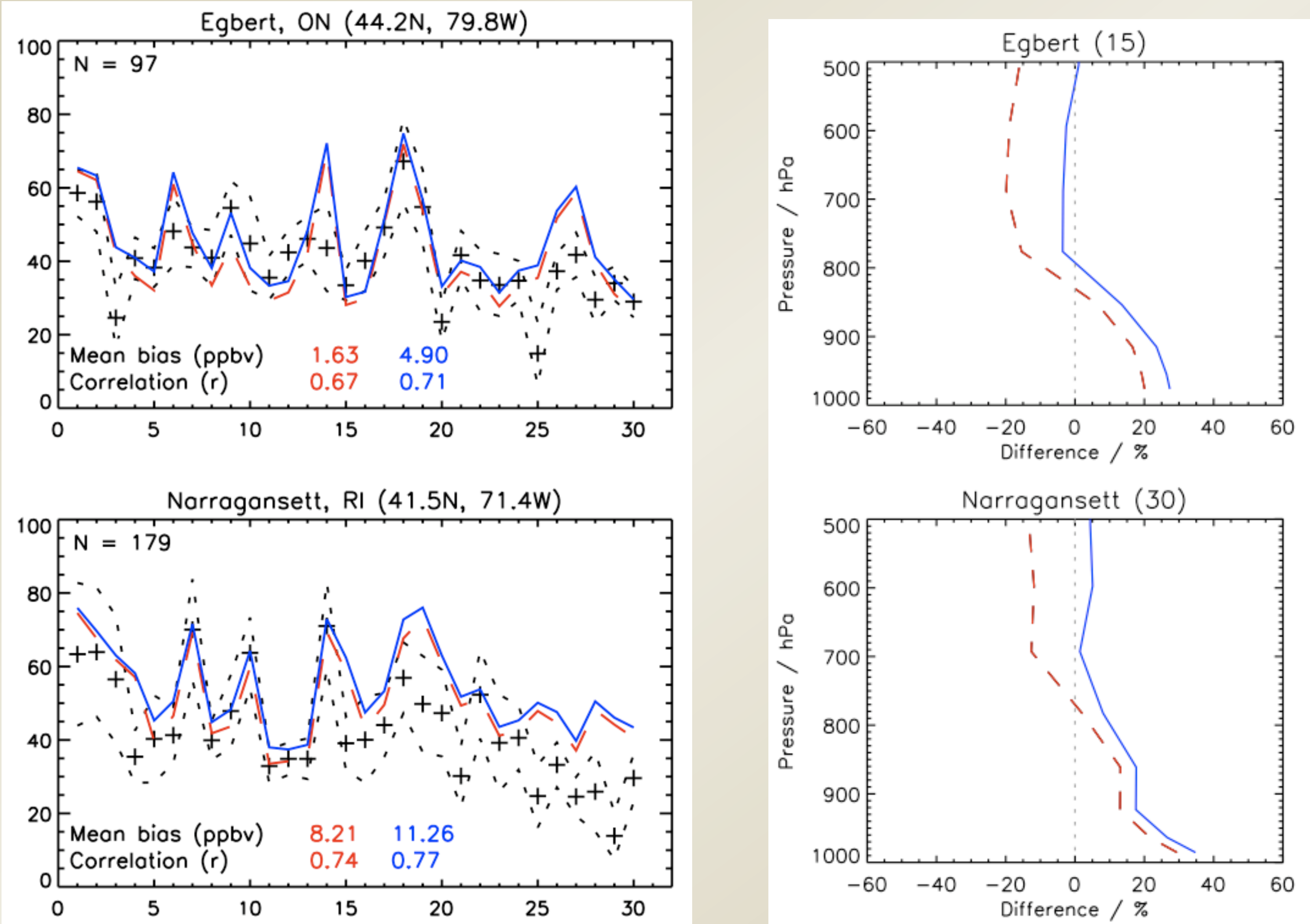
TES assimilation generally reduces bias in the West---up to 9ppb







Comparison to Eastern Sites

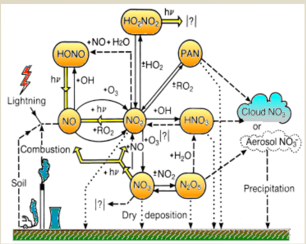


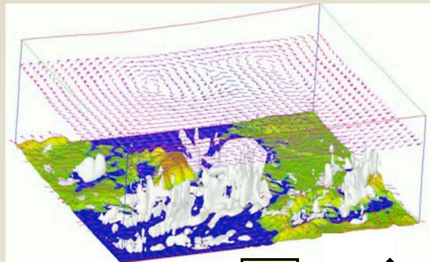
Assimilation paradigm

Transport

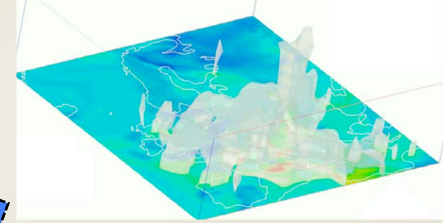
Meteorology

Chemical kinetics

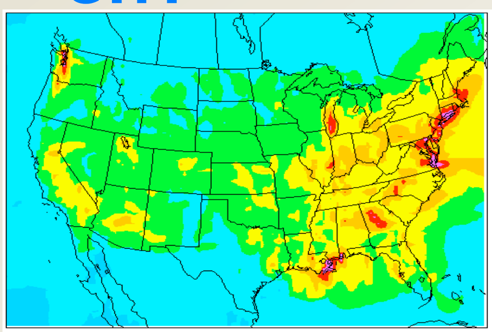




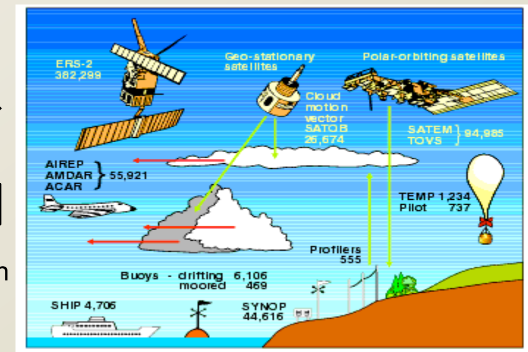
Optimal analysis state



CTM

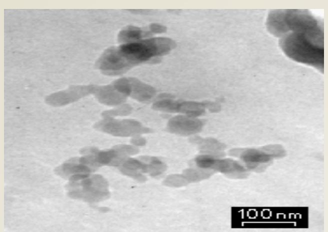


Observations



State/parameter estimation

Aerosols



Emissions

$$\min_{\mathbf{x}_0} C(\mathbf{x}) = \left\{ \sum_i (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x}))^\top (\mathbf{S}_n^i)^{-1} (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x})) + (\mathbf{x}_0 - \mathbf{x}_a)^\top \mathbf{S}_a^{-1} (\mathbf{x}_0 - \mathbf{x}_a) \right\}$$

- We want to understand the sensitivity of ozone to processes
- We can observationally constrain parameters defined within those processes

Courtesy, Adrian Sandu, VT

Putting it together: adjoint-based source and state estimation

$$\min_{\mathbf{x}_0} C(\mathbf{x}) = \left\{ \sum_i (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x}))^\top (\mathbf{S}_n^i)^{-1} (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x})) + (\mathbf{x}_0 - \mathbf{x}_a)^\top \mathbf{S}_a^{-1} (\mathbf{x}_0 - \mathbf{x}_a) \right\}$$

$$\text{subject to } \mathbf{x}_{i+1} = \mathbf{M}_i(\mathbf{x}_i, \mathbf{p}_i)$$

The gradient for the cost function with respect to the initial value is

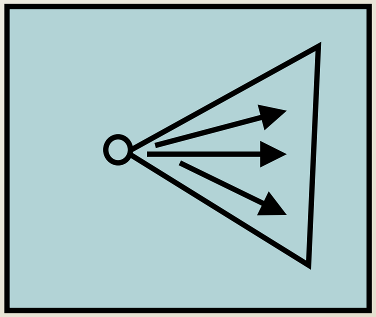
$$\lambda = \nabla_{\mathbf{x}_0} C(\mathbf{x}) = \left(\frac{\partial C(\mathbf{x})}{\partial \mathbf{x}_0} \right)^\top =$$

$$\mathbf{S}_a^{-1}(\mathbf{x_0} - \mathbf{x}_a) + \sum_i \left(\frac{\partial \mathbf{x}^i}{\partial \mathbf{x}^0} \right)^\top \mathbf{F}_i^\top (\mathbf{S}_n^i)^{-1} (\mathbf{y}_i - \mathbf{F}_i \mathbf{x}_i)$$

Sensitvity between states or parameters

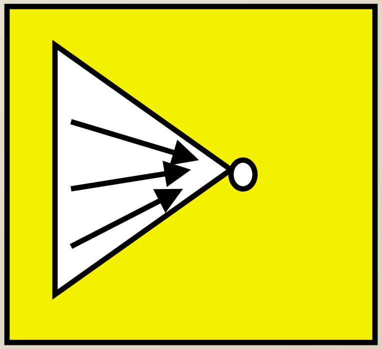
The sensitivity between states can be calculated this way

$$\left(\frac{\partial \mathbf{x}^i}{\partial \mathbf{x}^0}\right)^\top = \left(\frac{\partial \mathbf{M}_{i-1}}{\partial \mathbf{x}_{i-1}} \cdots \frac{\partial \mathbf{M}_0}{\partial \mathbf{x}_0}\right)^\top$$



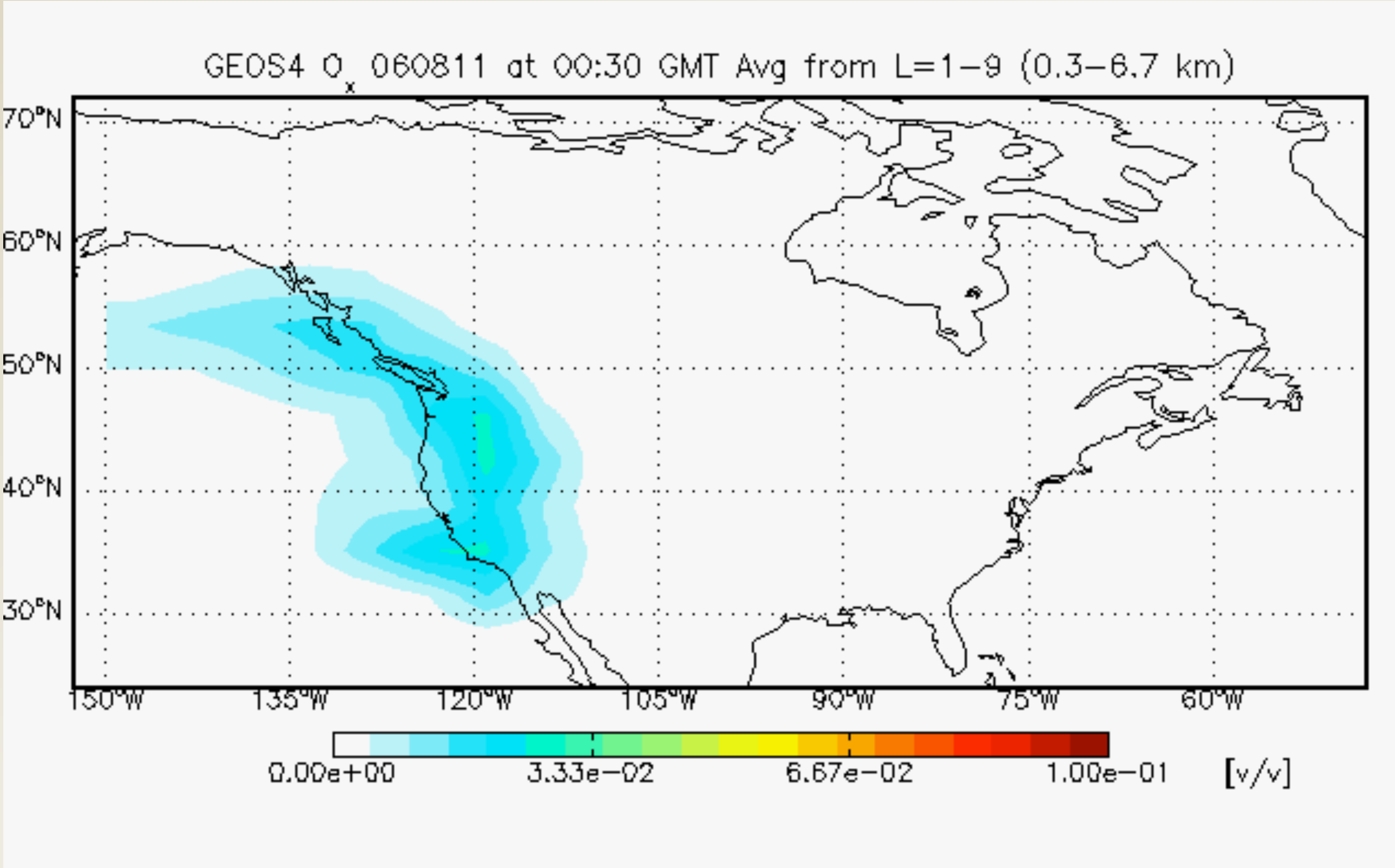
or that way

$$\left(\frac{\partial \mathbf{x}^i}{\partial \mathbf{x}^0}\right)^\top = \left(\frac{\partial \mathbf{M}_0}{\partial \mathbf{x}_0}\right)^\top \cdots \left(\frac{\partial \mathbf{M}_{i-1}}{\partial \mathbf{x}_{i-1}}\right)^\top$$

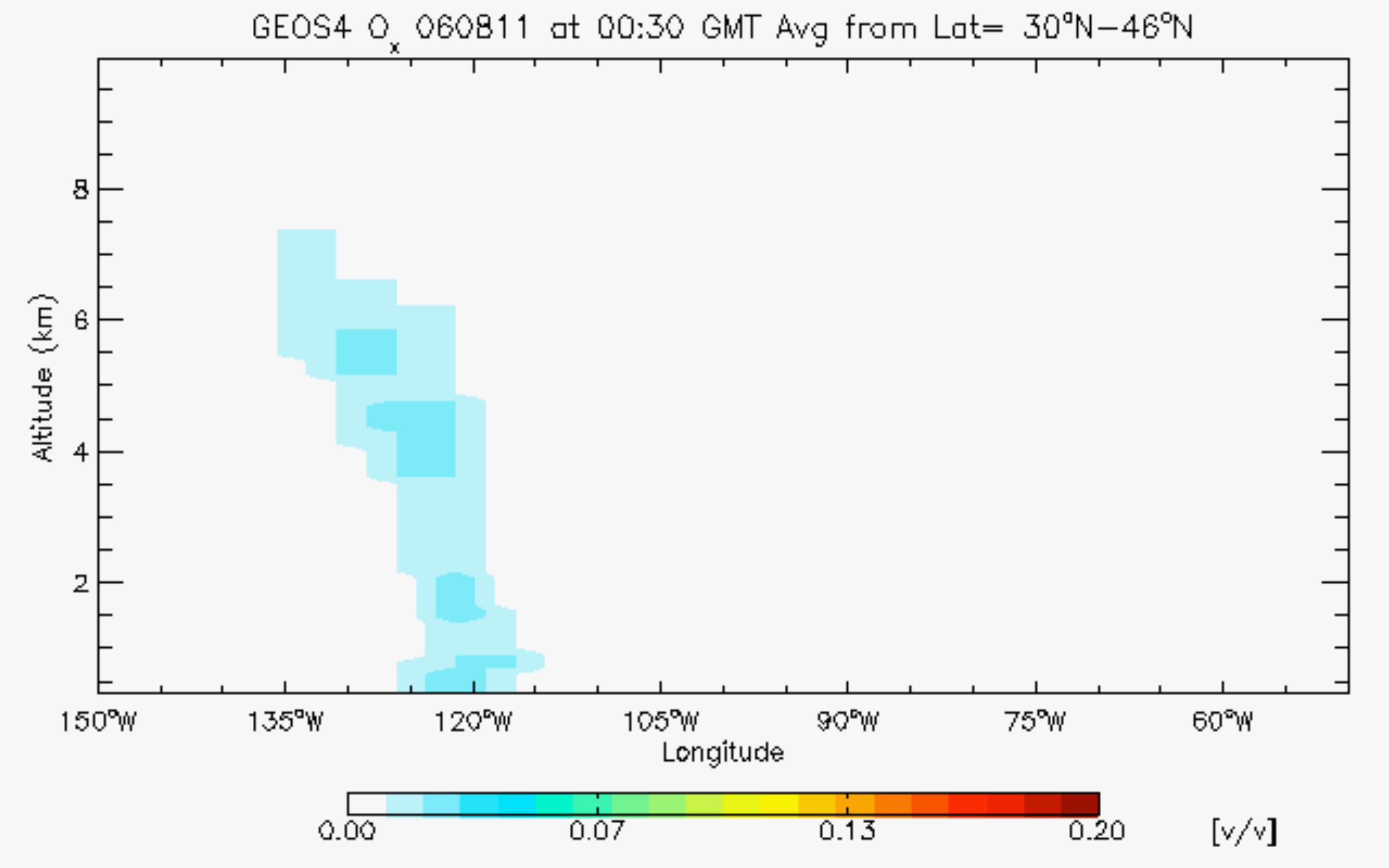


Sensitivity analysis over West Coast

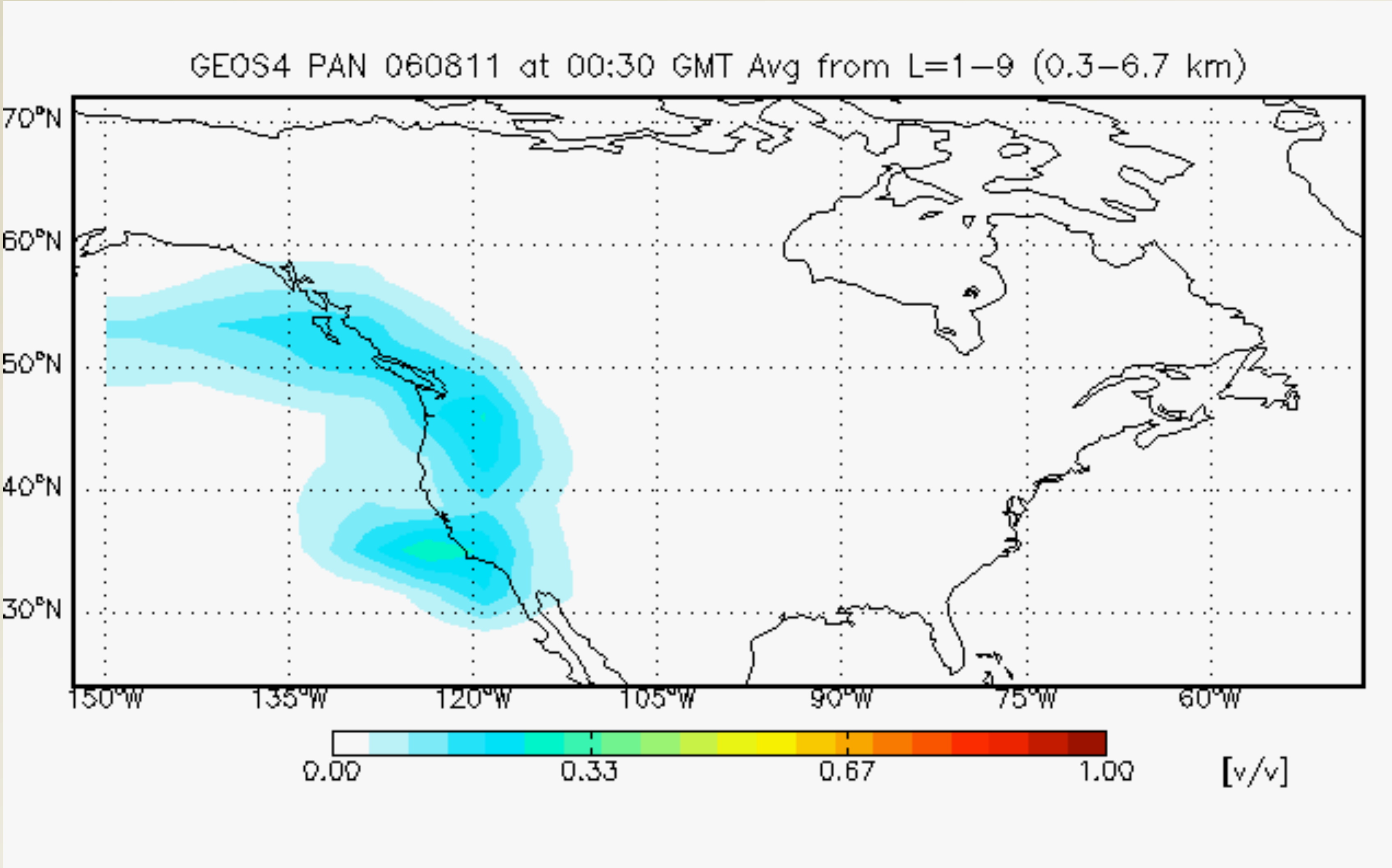
Adjoint sensitivity of PBL ozone wrt to tropospheric ozone on Aug 15, 2006, 1:30pm



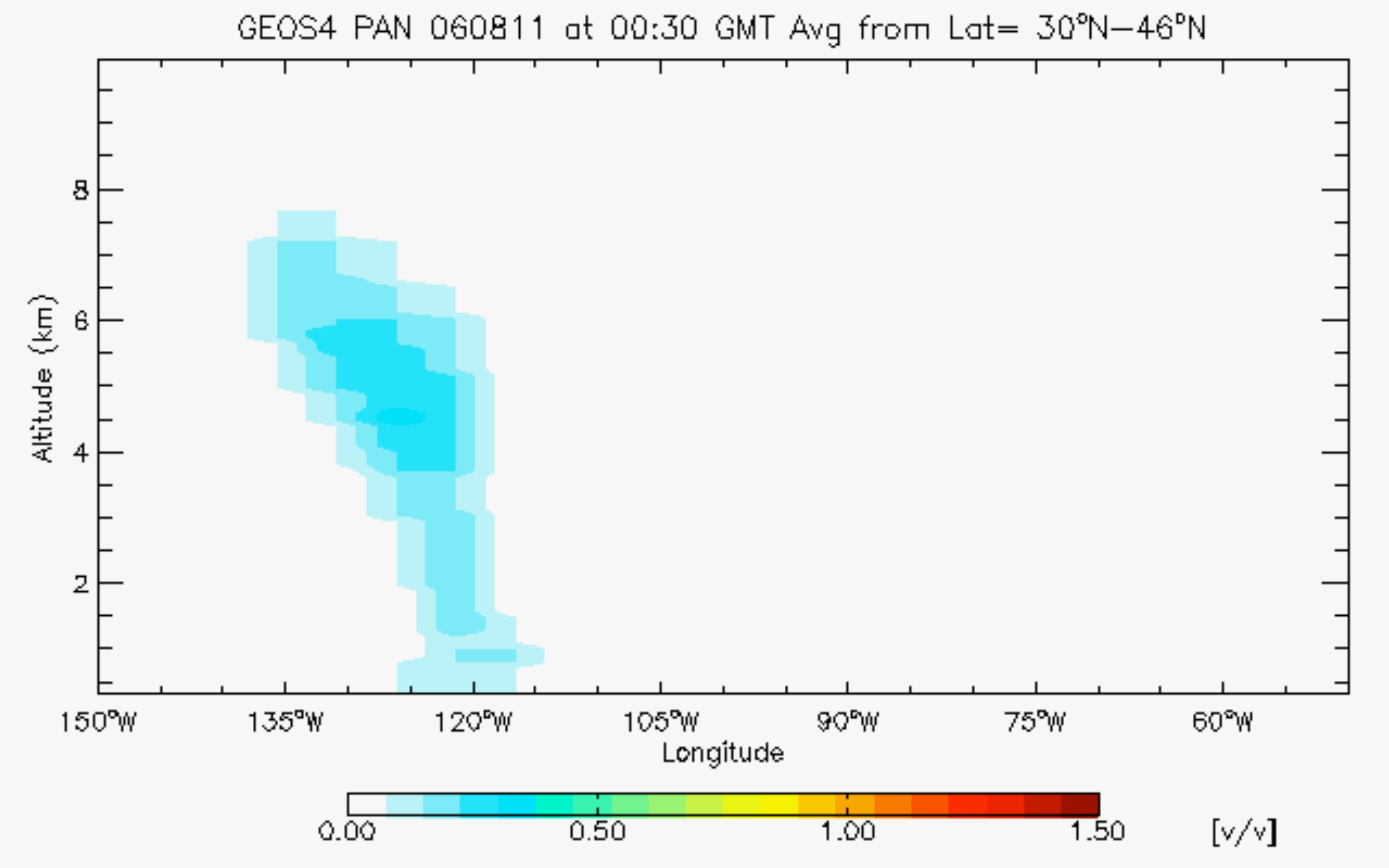
Sensitivity over the West Coast



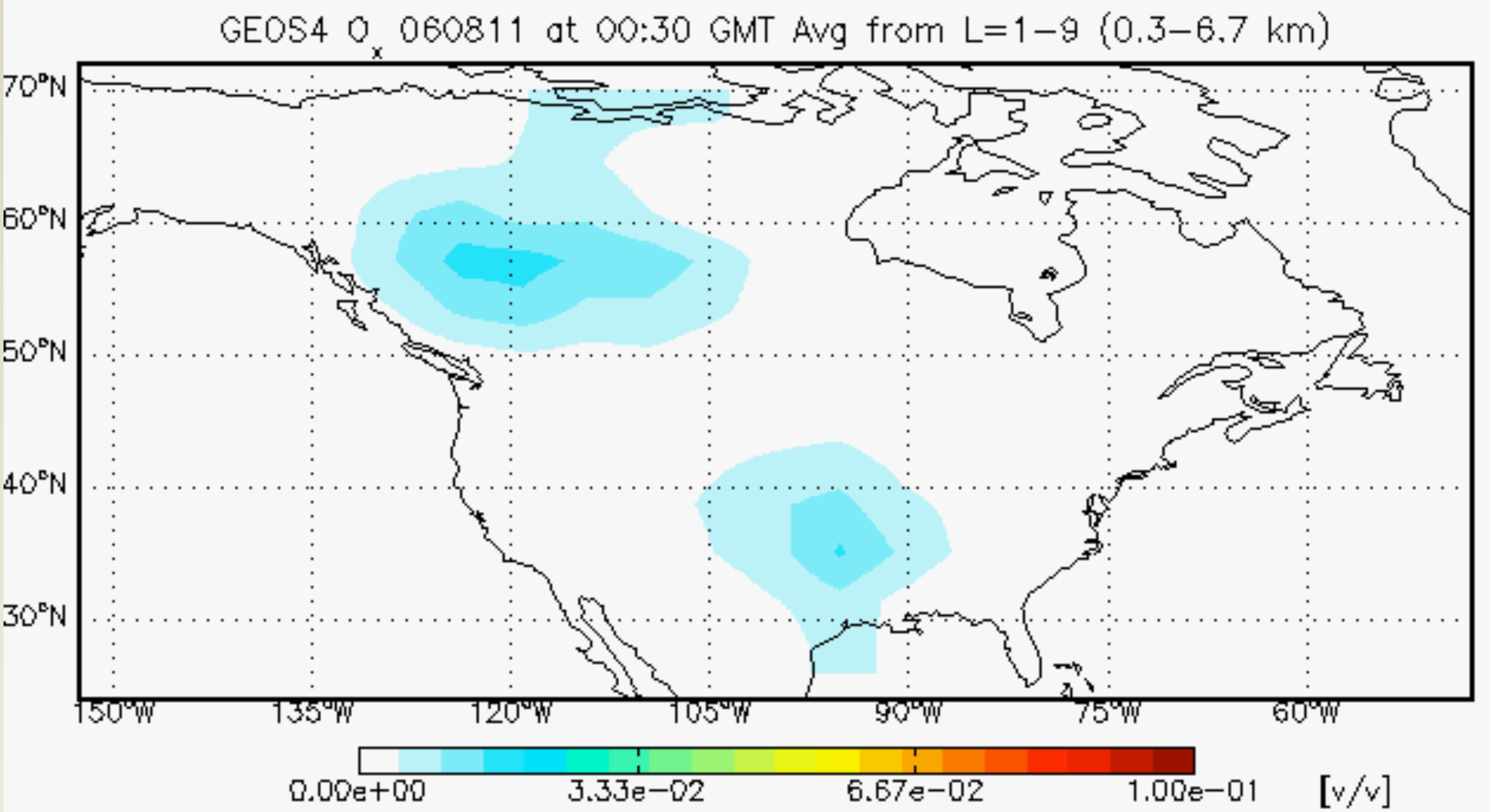
Sensitivity of O3 to PAN



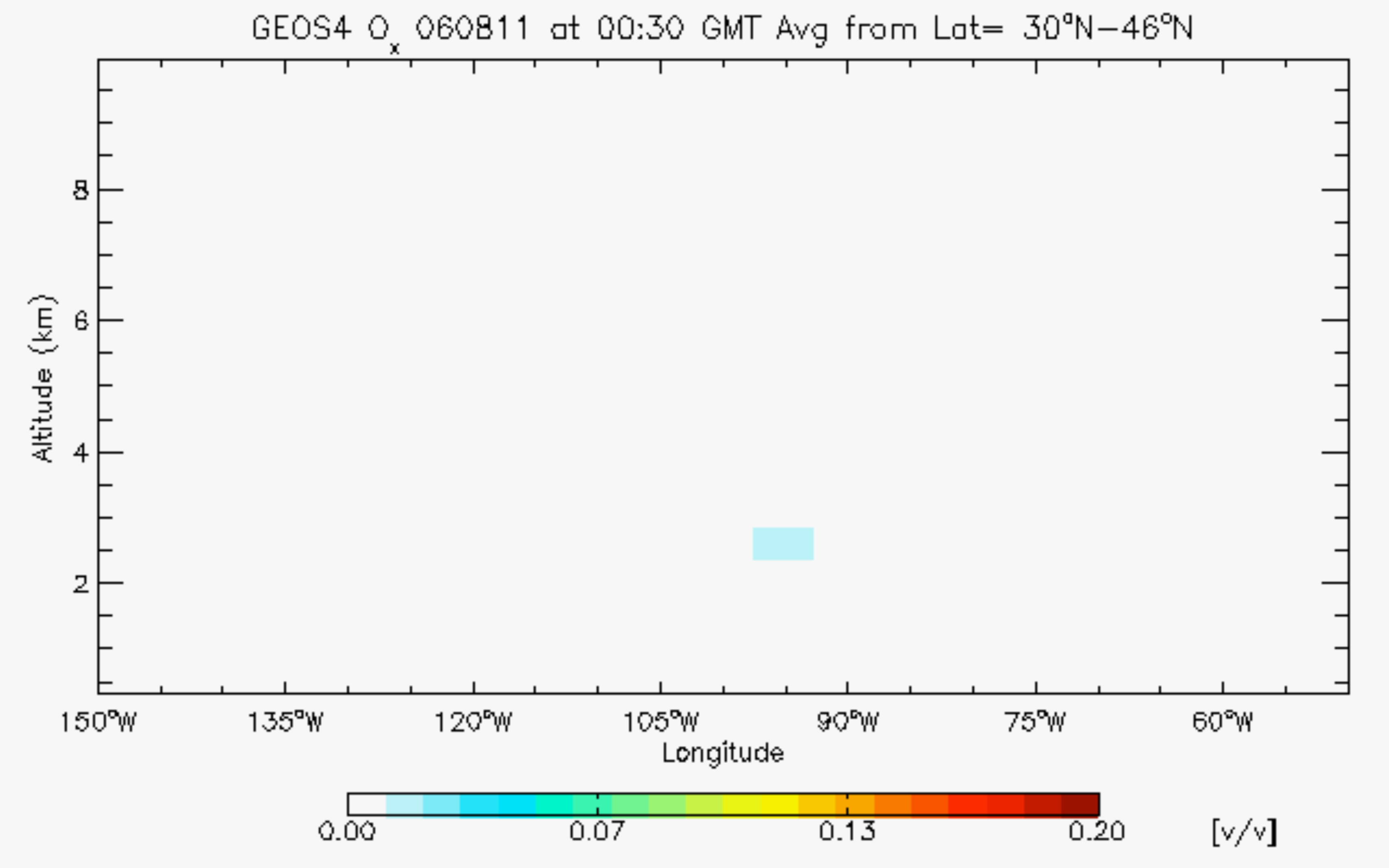
Sensitivity O3 to PAN



Sensitivity over the East Coast



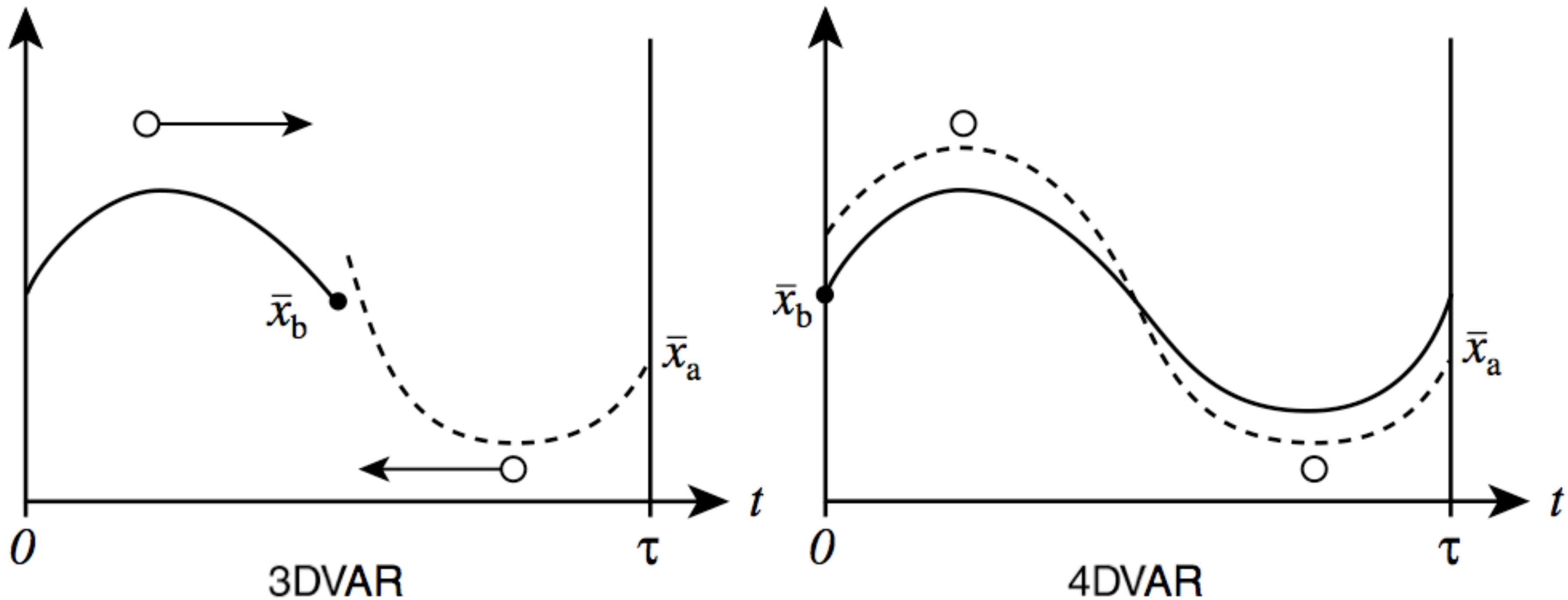
Sensitivity over the East Coast



Towards 3D-var and 4D-var assimilation

$$\min_{\mathbf{x}_0} C(\mathbf{x}) = \left\{ \sum_i (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x}))^\top (\mathbf{S}_n^i)^{-1} (\mathbf{y}_i - \mathbf{F}_i(\mathbf{x})) + (\mathbf{x}_0 - \mathbf{x}_a)^\top \mathbf{S}_a^{-1} (\mathbf{x}_0 - \mathbf{x}_a) \right\}$$

$$\text{subject to } \mathbf{x}_{i+1} = \mathbf{M}_i(\mathbf{x}_i, \mathbf{p}_i)$$



Courtesy E. Holm, ECMWF

Comparison of 3D-var and 4D-var

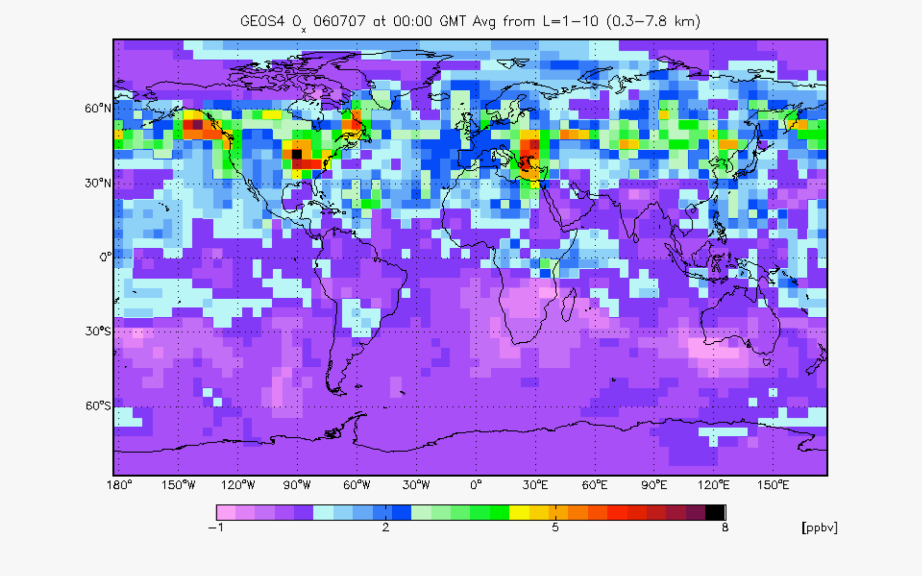
Assimilation for July 7th, 2006
0 GMT

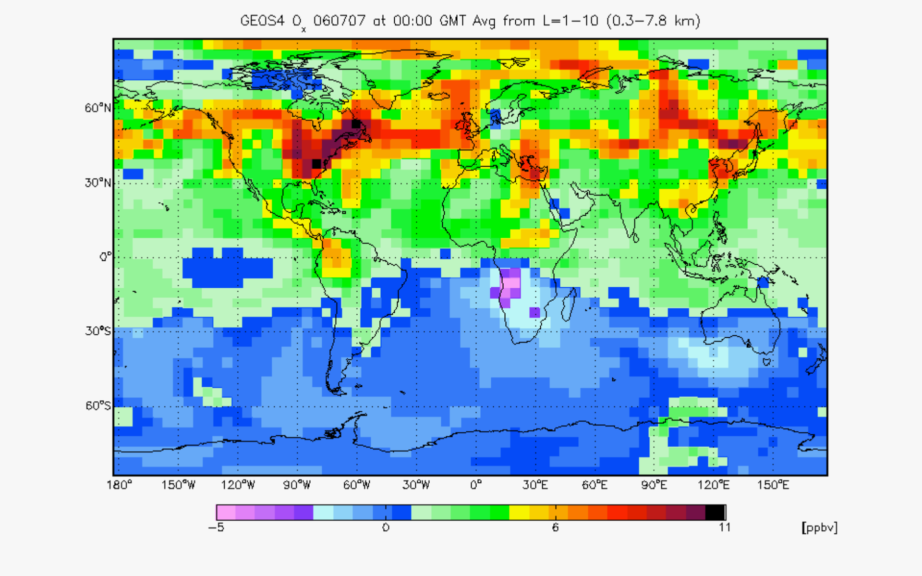
Plots taken from data averaged from 0.3-7.8km

3D-var is based on 4 hour assimilation window

4D-var uses 6 day assimilation window

Diagonal background used for these analyses



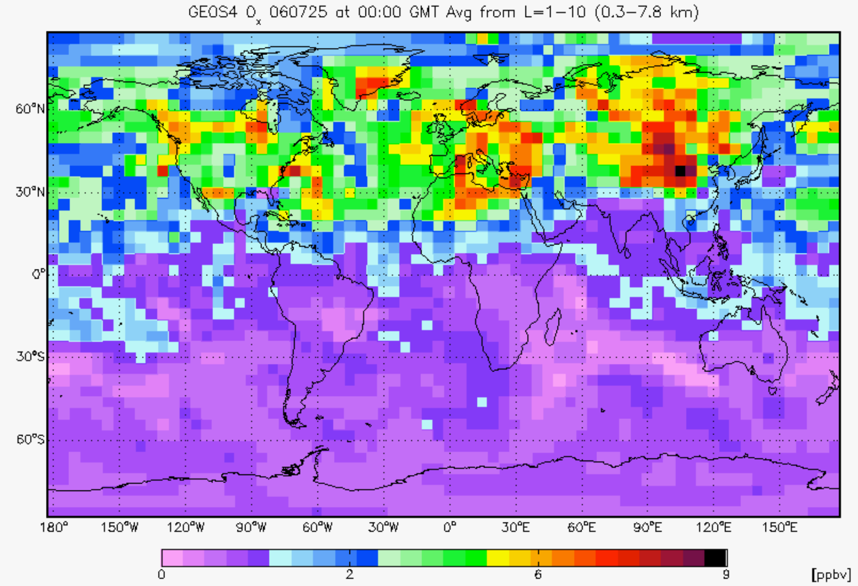


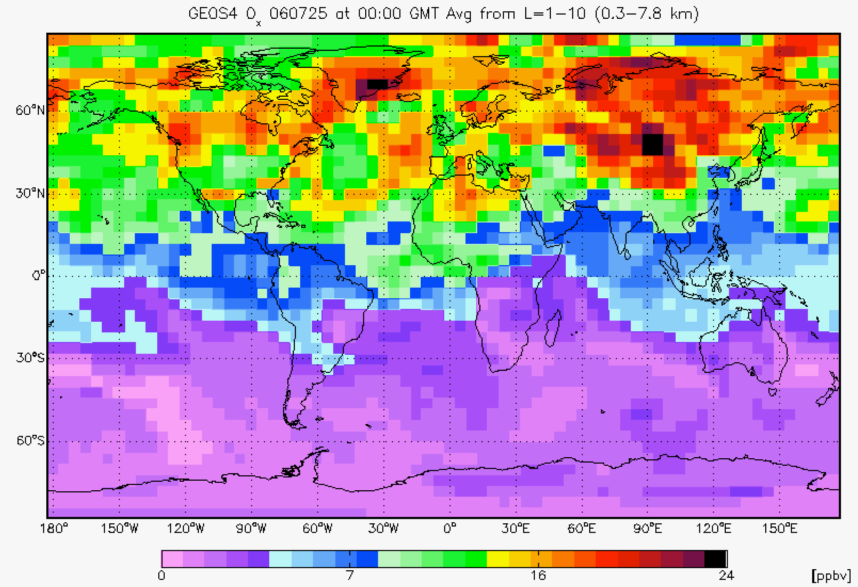
Comparison of 3D-var and 4D-var

July 25th, 2006, 0 GMT

Data averaged from 0.3-7.8km

Up to 15 ppb difference between 3D-var and 4D-var





Conclusions

- The assimilation reduces the negative bias in the modeled free tropospheric ozone
- Surface ozone abundances increased by as much as 9 ppb in western North America and by less than 2 ppb in the southeast
- Total background source of ozone of 20 – 40 ppb.
- Improved agreement with western surface ozone observations
- Decreased disagreement with the eastern sites
  - despite the agreement between the assimilation and ozonesonde measurements in the free troposphere,
  - Suggests errors in the ozone sources or sinks or in boundary layer mixing in the model.
- Adjoint sensitivity analysis is a powerful tool investigate the exchange between boundary layer and free troposphere
- 4D-var assimilation promises to be a significant improvement over sub-optimal Kalman filtering for AQ studies
- Application of TES data on Houston and Dallas AQ during TexAQS 2006 is discussed by Pierce *et al,* JGR, 2009