Sensitivity of Tropospheric Ozone to Emissions

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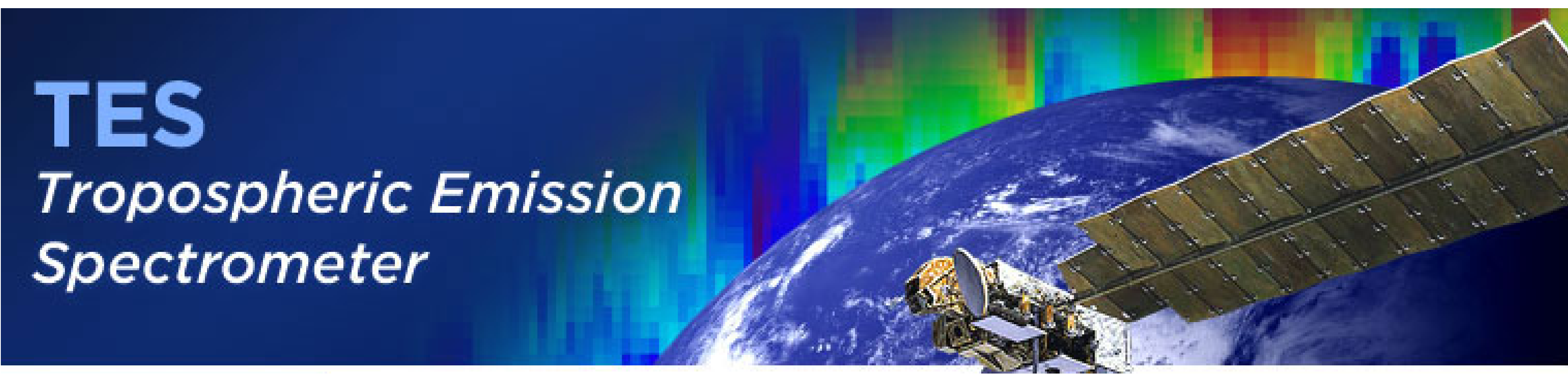
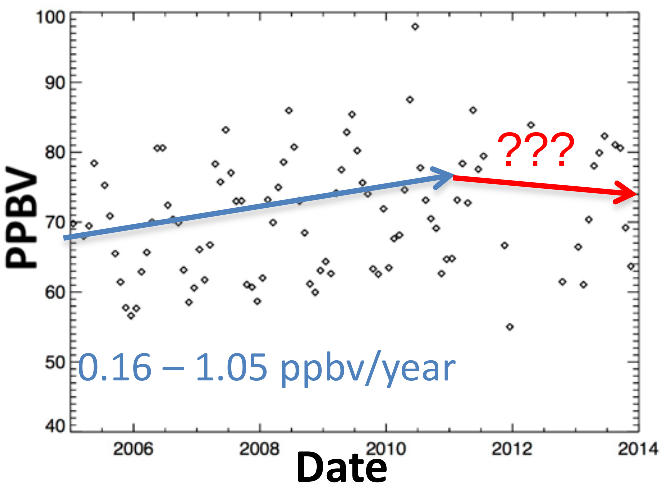
3) KNMI



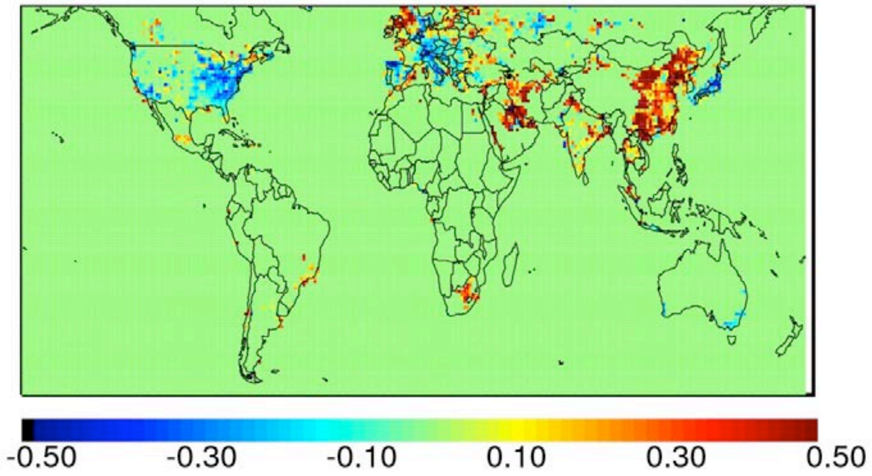
How is Tropospheric Ozone Responding to Rapidly Changing Ozone Pre-Cursor Emissions?

Lamsal et al. GRL 2010, H. Worden ACP 2012; Worden et al. GRL 2013, Verstraeten et al. Nat Geo Accepted

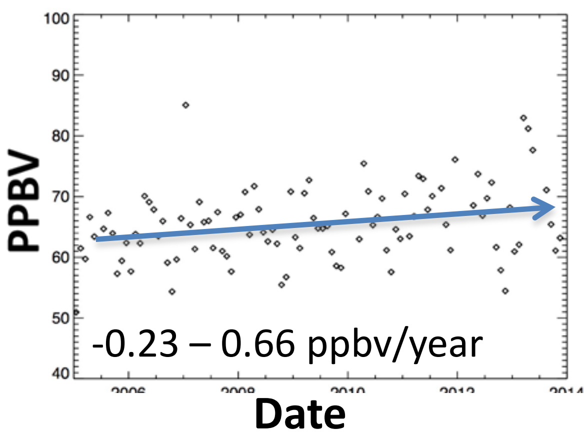
TES Free-Troposphere Ozone (Asia)



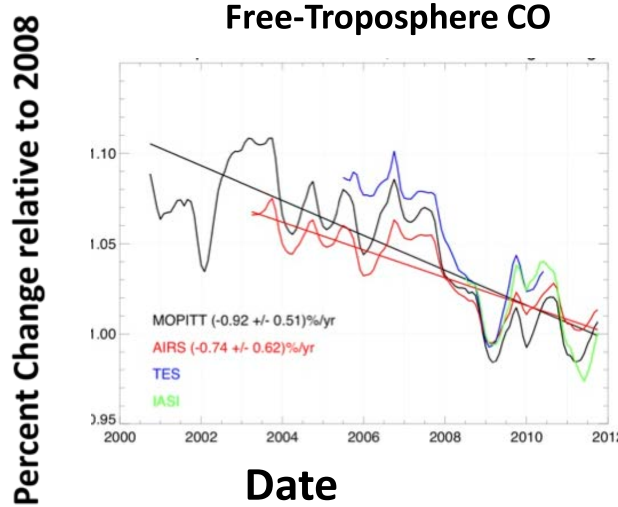
Fractional Change in OMI NO_2 since 2005



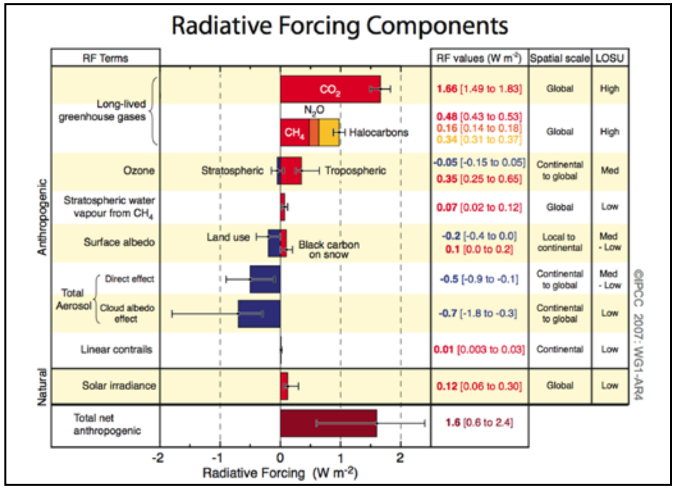
TES Free-Troposphere Ozone (N. America)



Free-Troposphere CO



Tropospheric Ozone is the Third Most Important Greenhouse Gas and an Air Pollutant



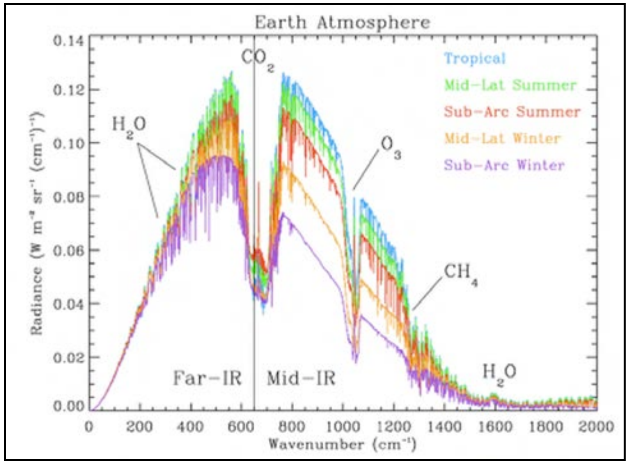
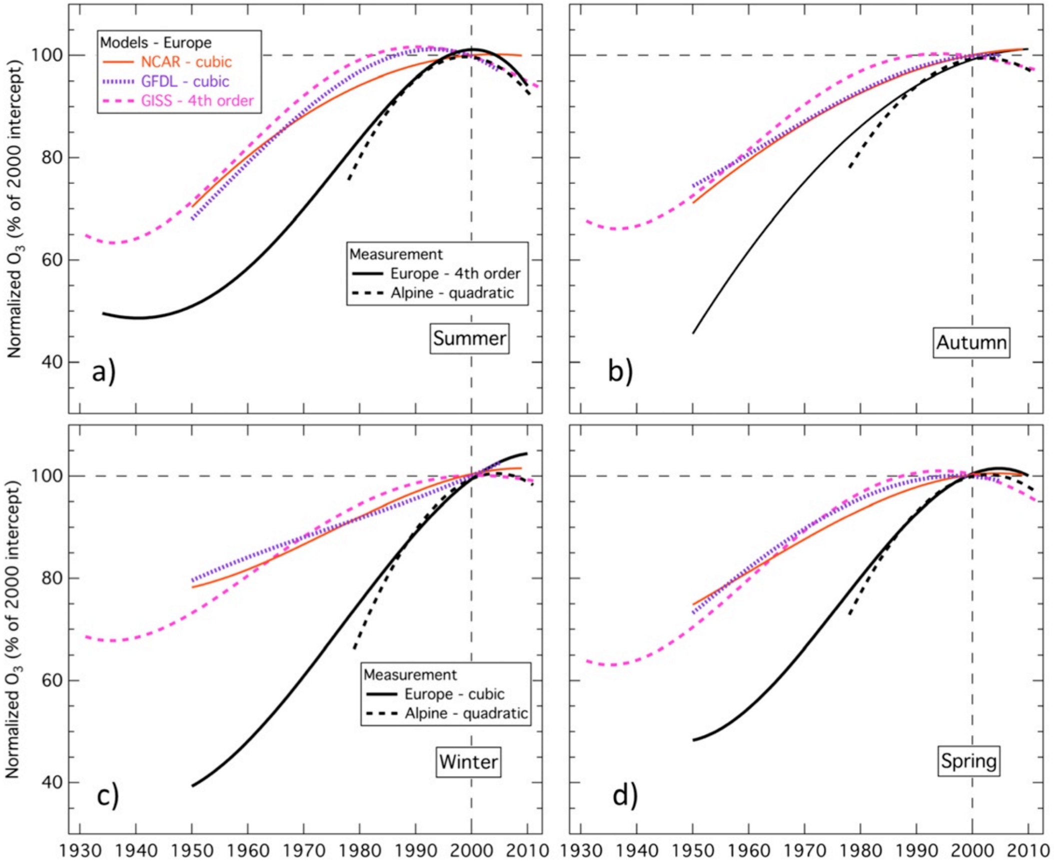


Fig. courtesy M. Mlynzcack (LaRC)

Ozone affects air-quality and plant health

Reduced CO_2 uptake from damaged plants also strongly affects climate

Sensitivity of Free-Tropospheric Ozone Over Europe to Global Emissions

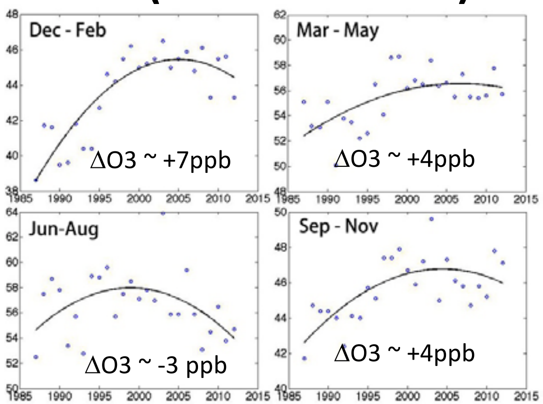


Change in ozone between ~1940 to the present is much larger in data than model

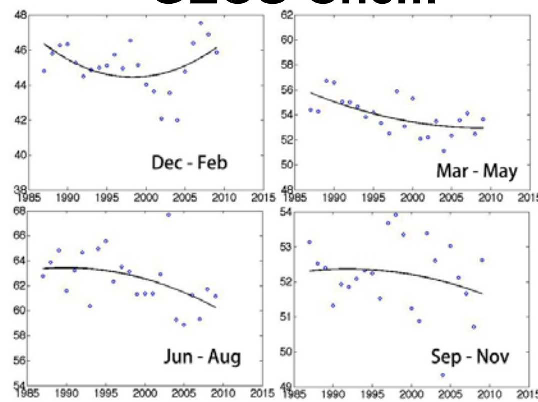
Parrish et al., JGR 2014

Sensitivity of Free-Tropospheric Ozone Over Europe to Global Emissions

Data (1985 – Present)



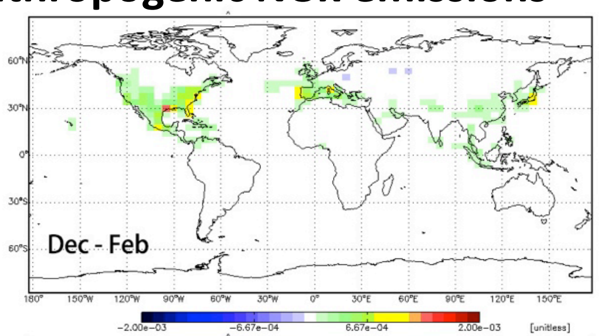
GEOS-Chem

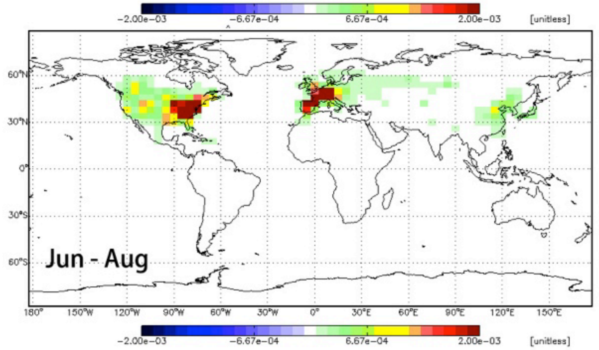


Data show ~ -3 ppb decrease for summer and ~+4 ppb increase for all other seasons (1985 to present)

Model and data ~agree in summer and completely dis-agree for all other seasons

Sensitivity of ozone over Europe to Anthropogenic NOx emissions





- **Sensitivity of free-tropospheric ozone to emissions is relatively small in winter**
- **Natural production and/or strat/trop exchange too high in winter?**
- **Sensitivity to Asian emissions too low?**

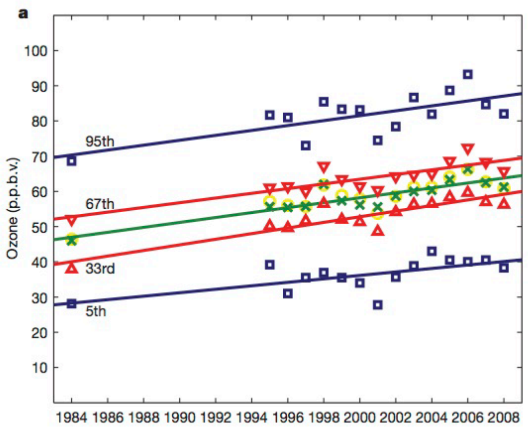
Sensitivity primarily to European and North American Emissions in Summer
→ Decrease since 1997 partly explained by decrease in emissions

How Does Tropospheric Ozone Respond to Asian Emissions?

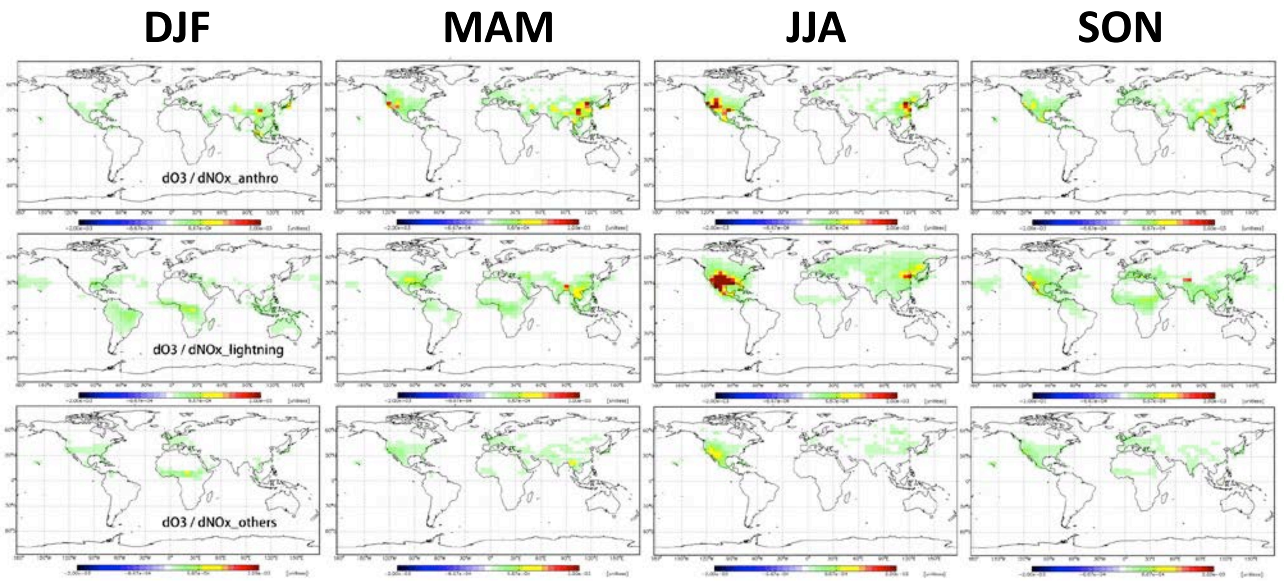
Ozone-Sondes over the Western USA show increasing spring-time free-tropospheric ozone
Cooper et al., Nature, 2010 infers that this increasing ozone is linked to increasing Asian emissions

Sensitivity of Ozone to Asian emissions

Free Tropospheric Ozone Over Western USA from Ozone-Sondes



~0.63 +/- 0.34 ppb / yr

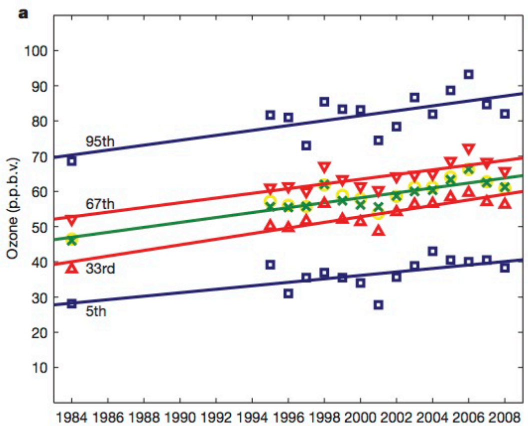


Sensitivity of ozone to Asian emissions (relative to other ozone pre-cursor sources) is largest in the spring

How Does Tropospheric Ozone Respond to Asian Emissions?

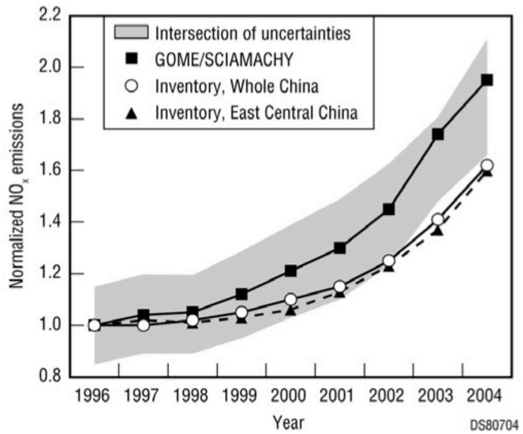
Ozone-Sondes over the Western USA show increasing spring-time free-tropospheric ozone
Cooper et al., Nature, 2010 infers that this increasing ozone is linked to increasing Asian emissions

Free Tropospheric Ozone Over Western USA from Ozone-Sondes



~5PPB in Ozone over Western USA Between 1994 and 2004
Cooper et al. Nature 2010

Change in NO_x over Eastern China



~70% change in NO_2 between 1996 and 2004
Zhang et al., JGR 2007

Change in NO_2 from 1996 - 2006

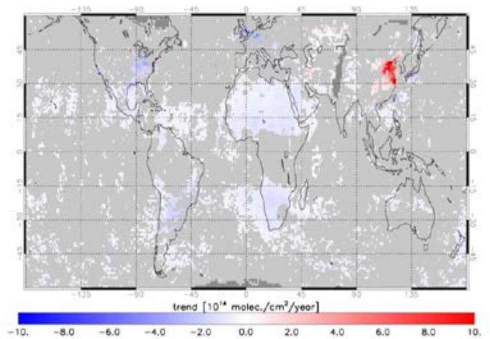


Figure 2. Linear trend per year for tropospheric NO_2 in the period 1996 till 2005 derived from the satellite observation of GOME and SCIAMACHY. For the light grey areas no significant trend has been found in the time series. For the dark grey areas not enough observations were available to construct a time series of tropospheric NO_2.

Likely decrease in NO_2 over USA since 1996 supports explanation that increasing ozone concentrations over USA are from rising Asian Emissions
Van Der A, JGR, 2008

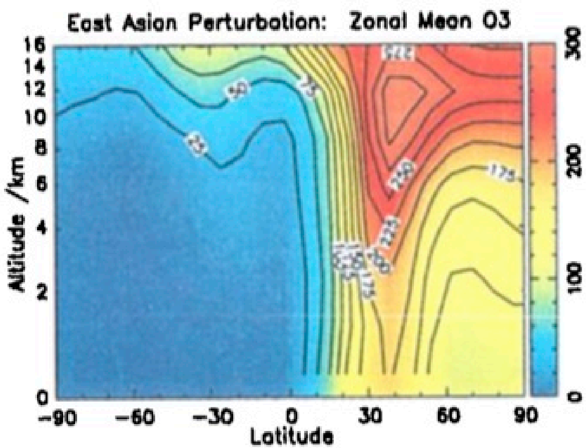
Assuming attribution of increase in ozone is correct and ozone production is limited by NOx then
~10% increase in Asian emissions → 0.7 ppb over USA

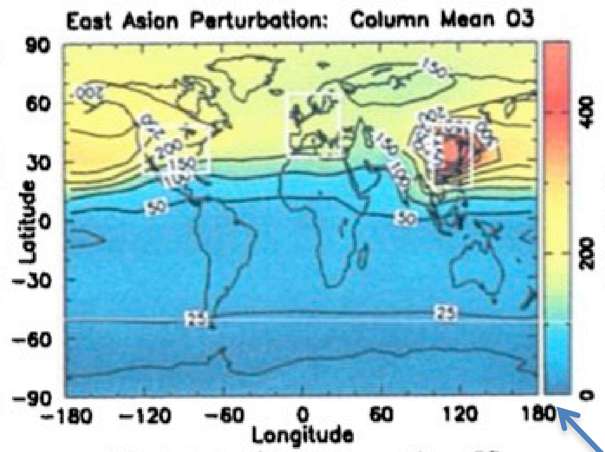
How Does Tropospheric Ozone Respond to Asian Emissions?

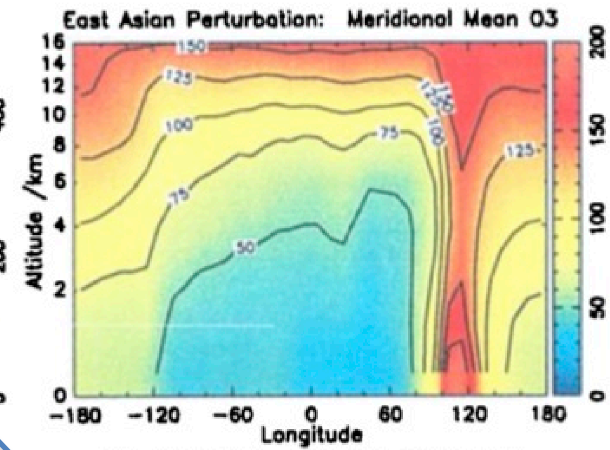
Model and Data Again Show Conflicting Results!

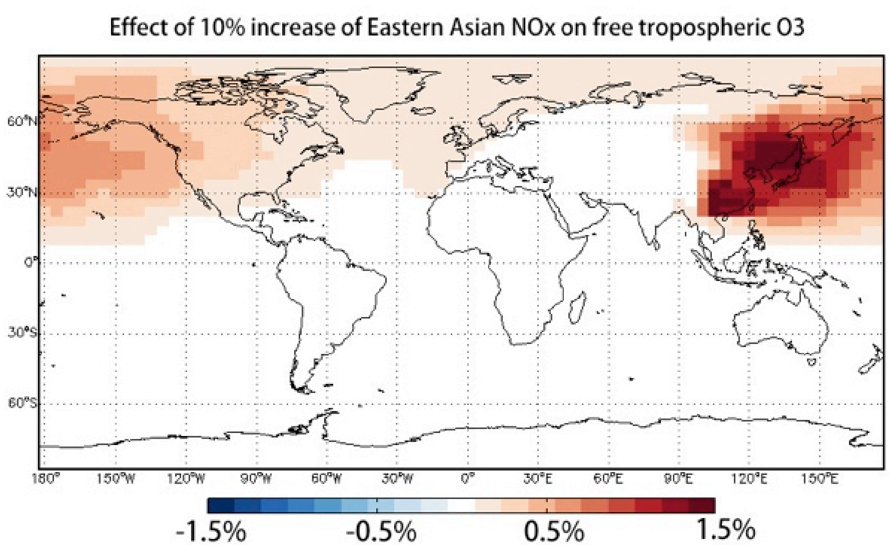
Data (from previous slide) suggest 10% increase in NO_x → 0.7 ppb increase over USA

Models show that 10% increase in NO_x → 0.2 ppb increase over USA









10% increase in East Asian NOx

→ ~0.2 PPB over USA

→ ~0.4 PPB over Ea. Asia

Using UCI model

Wild and Akimoto, JGR, 2001; Fiore et al. 2009

10% increase in East Asian NOx

→ 0.3% or ~0.2 PPB over USA

→ 1.0% or ~0.6 PPB over E. Asia

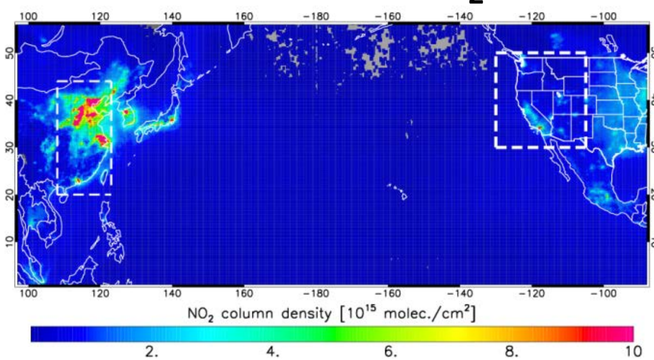
from GEOS-Chem

Jiang et al., in prep

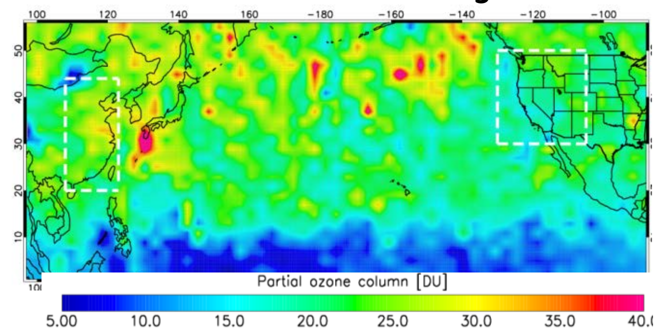
Response of Free-Tropospheric Ozone to Asian and N. American Emissions as Observed by Satellite

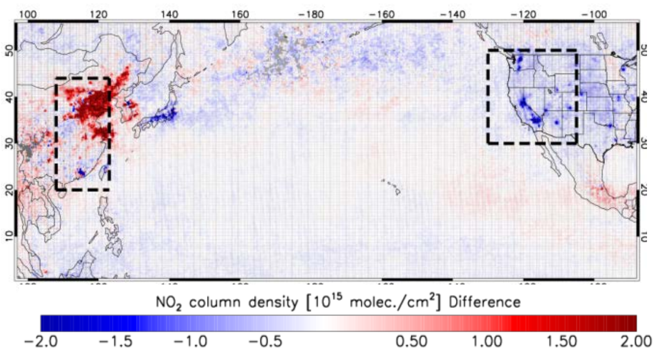
Verstraeten et al., Nature Geo (Accepted)

2009-2010 NO_2

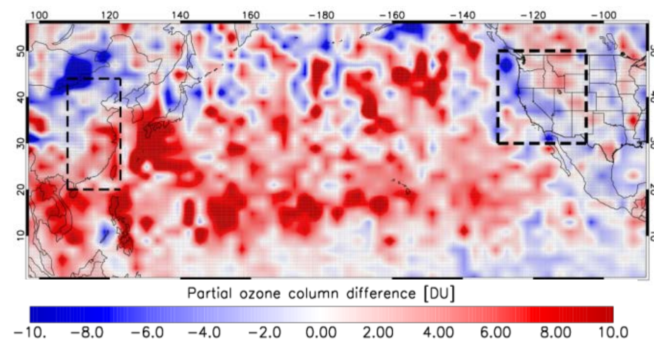


2009-2010 O_3





2010-2005 NO_2



2010-2005 O_3

Does increasing NOx emissions in Asia explain increasing ozone over Asia?

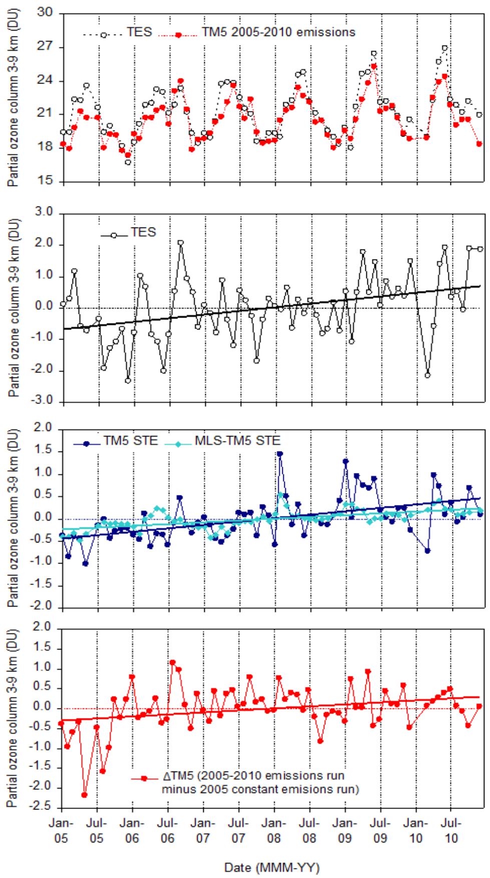
How is tropospheric ozone over N. America responding to increasing Asian emissions and decreasing N. American emissions?

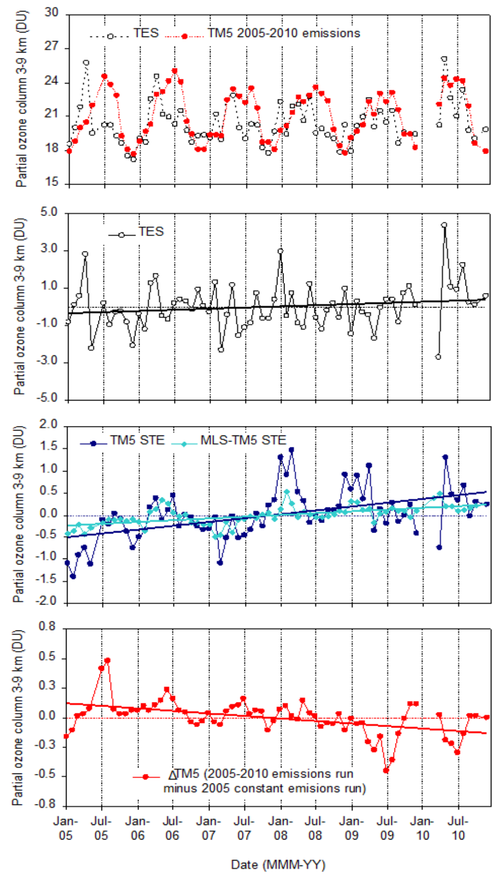
Response of Free-Tropospheric Ozone to Asian and N. American Emissions as Observed by Satellite

Verstraeten et al., Nature Geo (accepted)

Asian Ozone and Budget

W. USA Ozone and Budget





Asian:

Total Ozone Trend
~1.1%/yr Obs
~1.3%/yr Model
Or ~5.5 ppb/6 yrs

Strat/Trop
~0.8% / yr model
~0.5% / yr data

Emissions
~0.6% / yr

W. USA:

Total Ozone Trend
~0.6%/yr Obs
~0.7%/yr Model

Strat/Trop
~0.8% / yr model
~0.5% / yr data

Emissions
~-0.1% / yr

- TM5 model estimates bulk of increase over Asia is due to STE but with statistically significant (P~0.005) contribution from emissions (~10% increase in emissions → ~0.9 ppb ozone over Asia)
- Data from Aura MLS indicate strat/trop exchange only important in 2009/2010 due to QBO
- Asian ozone offsets approximately 47% of N. American emissions reductions.

Summary and Future Directions

Trends in ozone over parts of the globe can partly be explained by change in emissions (e.g. Summertime Northern Europe, Springtime N. America, Asia)

BUT **the response of ozone to emissions is likely larger (and possibly much larger) than expected by models (sensitivity to natural ozone and its pre-cursors too high, sensitivity to Asian emissions too low)**

→ **Radiative Forcing due to ozone is likely larger than expected**

→ **Ability to predict future trajectory of ozone and its chemistry and climate impacts is likely biased**

→ **Controlling ozone pre-cursor emissions will have a much bigger impact on global pollution and climate than expected**

Why Is Modeled and Actual Sensitivity of Ozone to Emissions Different?

- **H1: Larger than Expected Uncertainties in Emissions**

(e.g. Zhang et al., JGR 2007)

- **H2: Stratosphere to Troposphere Exchange**

(e.g., Neu et al., Nature Geo 2014; Verstraeten et al., submitted)

- **H3: Lightning**

(e.g. Miyazaki et al., ACP 2014)

- **H4: Fires**

(e.g. H. Worden et al., ACP 2013)

- **H5: Circulation**

(e.g. Lin et al., Nature Geo 2014)

- **H6: Insufficient Ozone Production in Free Troposphere**

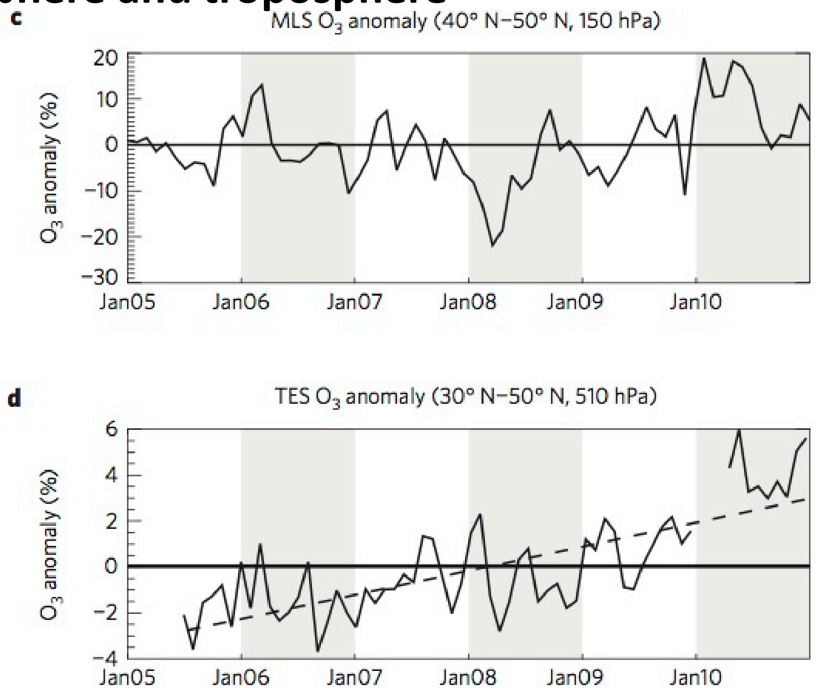
(e.g. Payne at al. AMT 2014; GRL 2015)

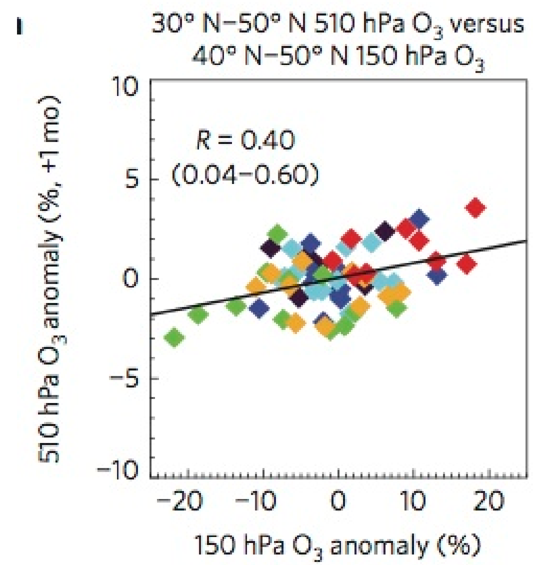
Continued measurements of ozone and its pre-cursors made globally are critical to testing hypothesis that could explain this critical science question.

Increased Stratosphere to Troposphere Exchange?

From *Neu et al., Nature Geoscience*, 2014

At the hemispheric scale: No obvious increase in exchange of ozone between the stratosphere and troposphere

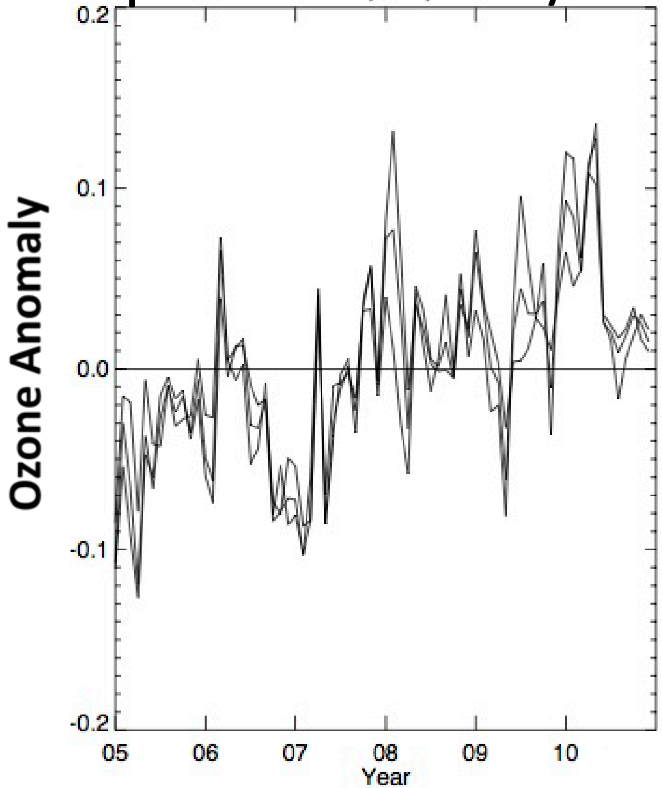




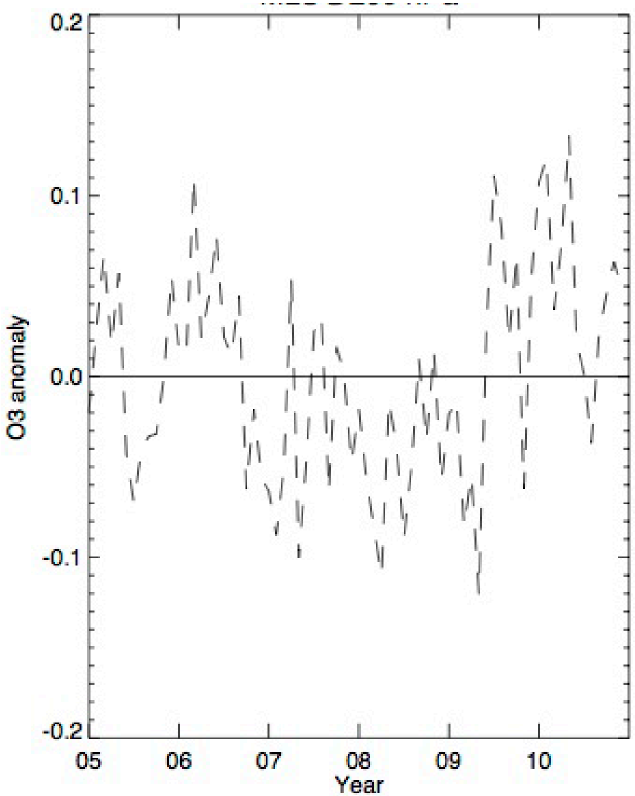
- **Stratospheric mid-latitude inter-annual variability is correlated with tropospheric variability (with 1 month shift).**
- **There is no observed trend in the lower mid-latitude stratospheric ozone or in the stratospheric circulation for this time period**

How Does TM5 Ozone Compare with Aura MLS Near the Mid-Latitude Tropopause?

TM5 Model at 200 hPa over Asia Sampled on Aura MLS Lat/Lon

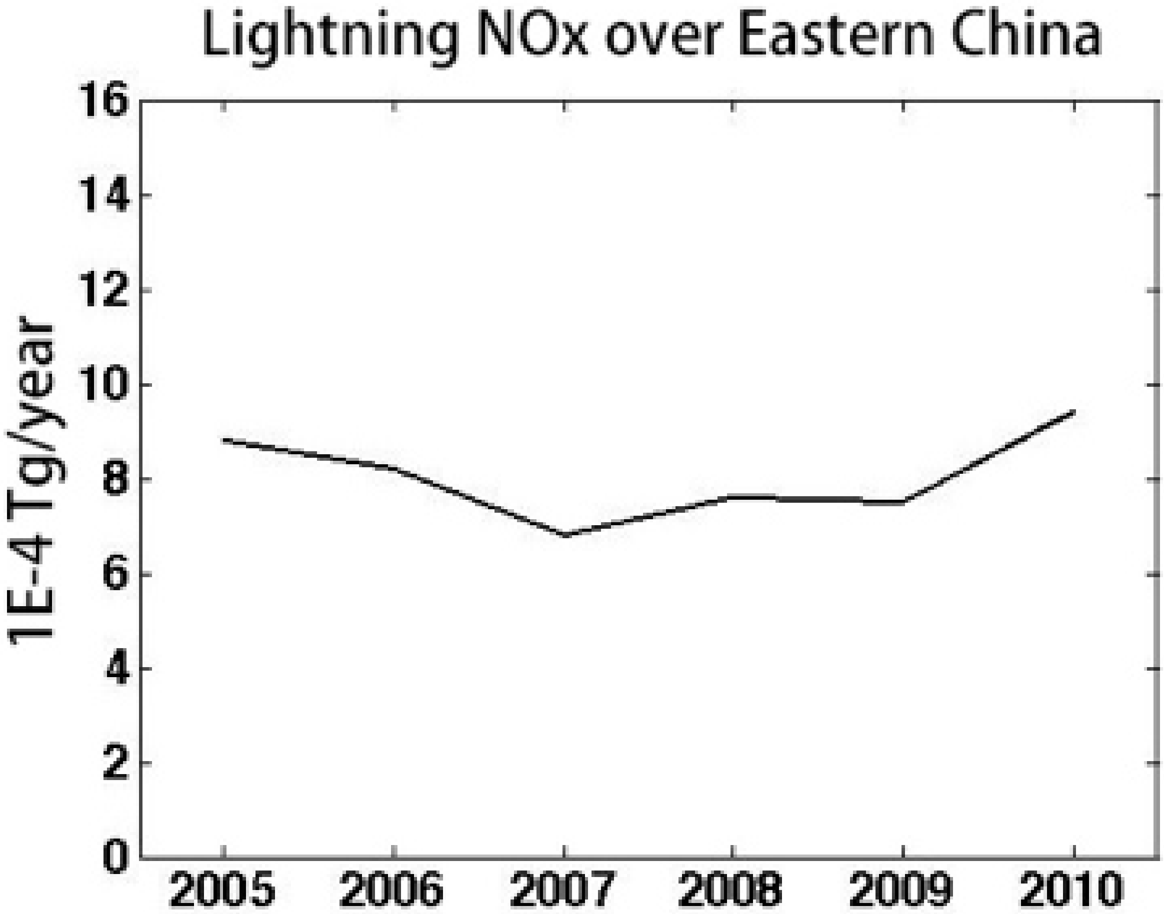


Aura MLS at 200 hPa over Asia



- **At the hemispheric scale, the trend in TM5 is not observed in Aura MLS data → TM5 may have an unrealistic trend in STE at hemispheric scales**
- **We need to evaluate TM5 STE at regional scales**

What about a Regional Increase In Lightning Emissions?



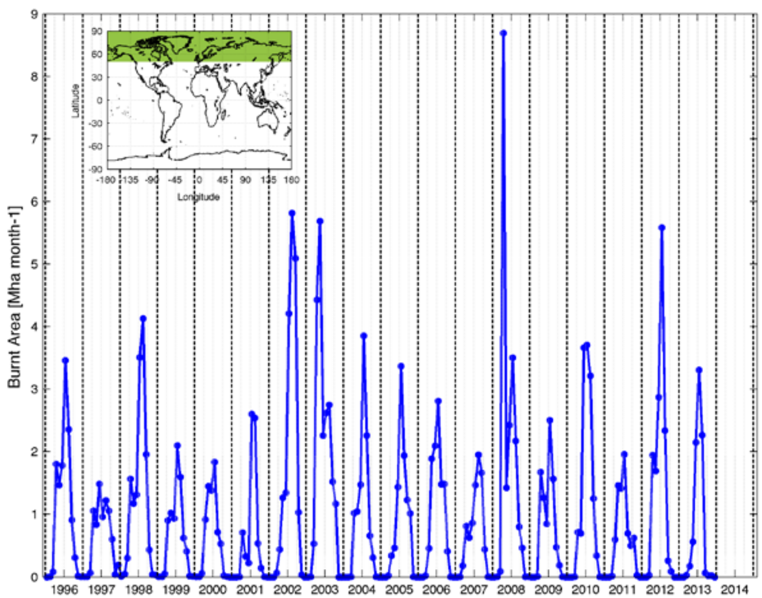
- Lightning NOx in GEOS-Chem is driven by meteorology.
- The meteorological conditions that affect lightning NOx have not changed suggesting that Lightning NOx is not increasing during this time period.

(e.g., Martin et al. 2006 and refs therein)

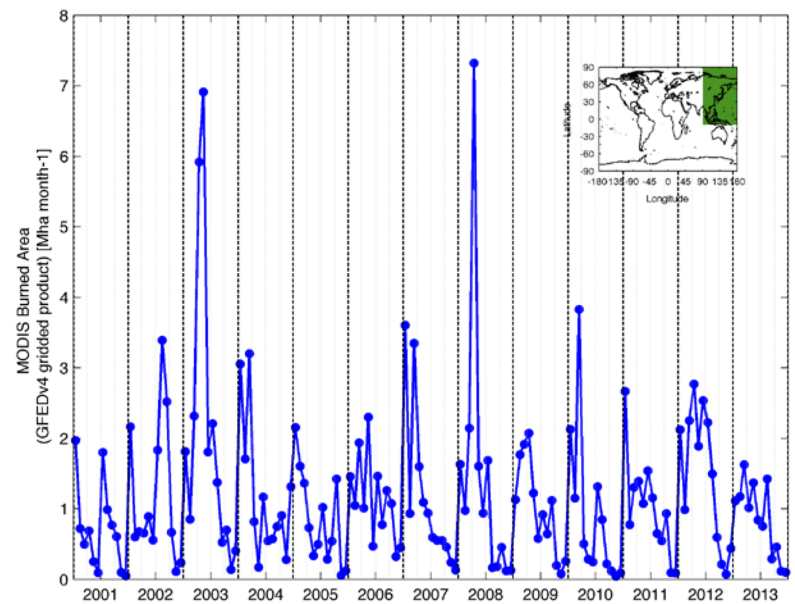
Changes in Fires?

TES
Tropospheric Emission Spectrometer

MODIS Burnt Area for Latitudes > 55 N



MODIS Burnt Area for East Asia

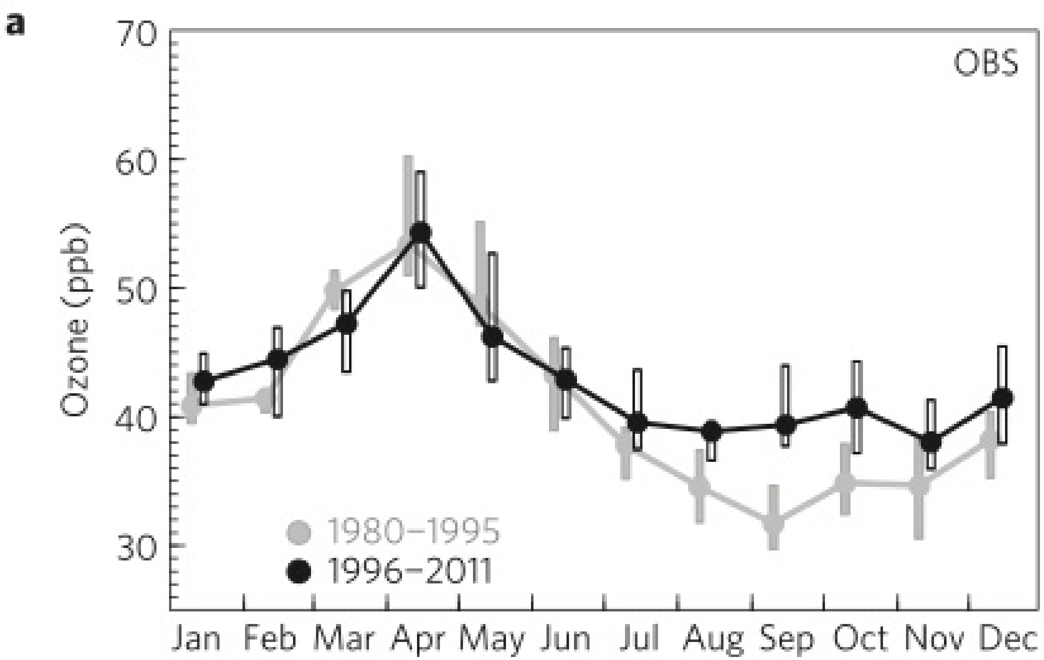


No obvious trend in fire emissions

Changes in Circulation?

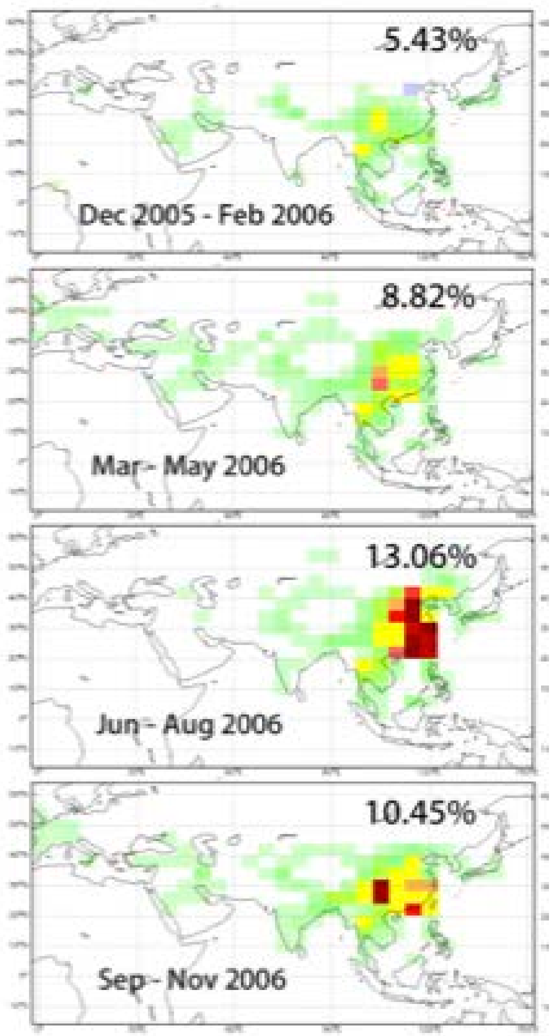
Increase in Summer/Fall Ozone observed at Mauna Loa Observatory is attributed to change in circulation bringing more ozone and ozone-precursors to Hawaii from Asia

Lin et al., Nature Geo, 2014



Changes in Circulation?

Sensitivity of Free-Tropospheric Ozone to Global NOx Emissions



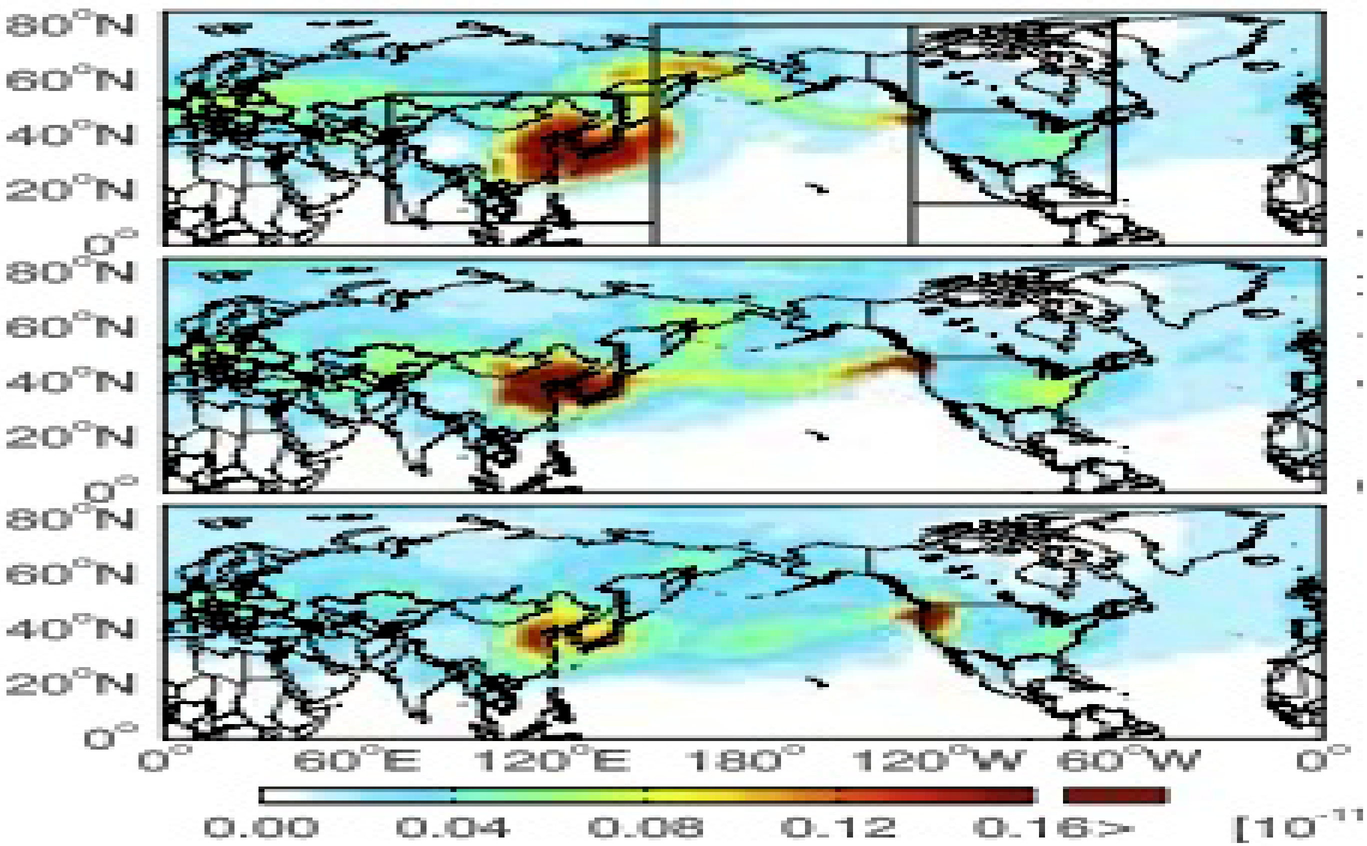
Percentages indicate relative contribution of Asian emissions to ozone over East Asia → largest increase is in the summer due to increased convective lofting.

GEOS-Chem adjoint shows that ozone in the free-troposphere over Asia strongly depends on remote sources (e.g., Jiang et al., ACPD 2014)

We cannot yet rule out that a shift in atmospheric circulation is contributing to the increase in ozone over Asia and the USA

Not Enough Ozone or Ozone Pre-Cursor Emissions Injected into Free Troposphere?

Ozone Production is more efficient in the free-troposphere
Ozone life-time is longer in the free troposphere

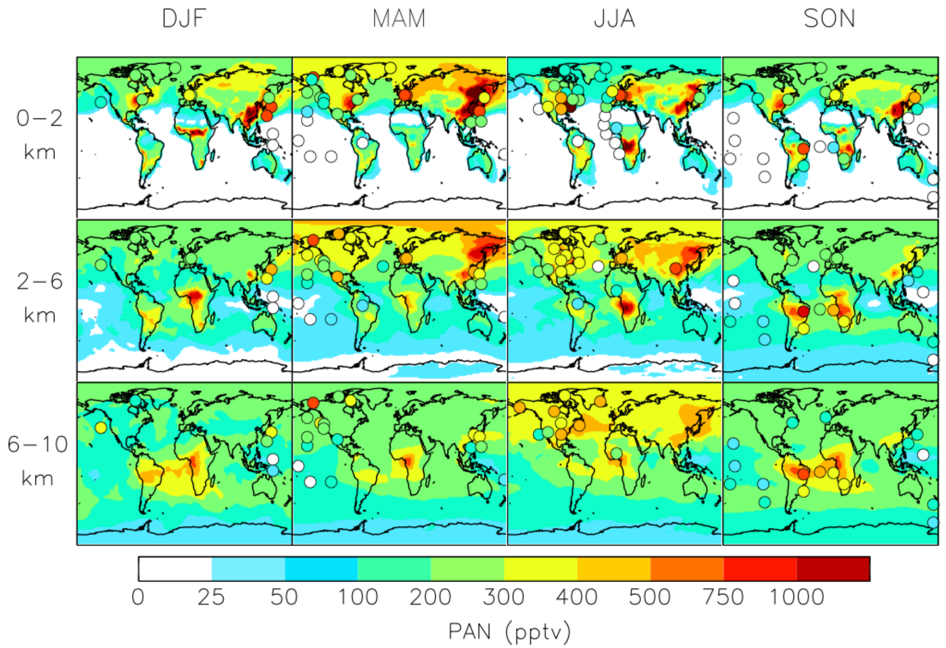


Sensitivity of ozone over MBO to ozone production worldwide

If models are not injecting enough ozone pre-cursors into the atmosphere then the sensitivity of free-tropospheric ozone to surface emissions is too small

Hypothesis: Are Models Not Injecting Enough Ozone Pre-cursors Into The Free Troposphere?

- Lets look at PAN because (1) PAN is a significant ozone pre-cursor and (2)we have new PAN data from the Aura TES satellite instrument
- PAN is a product of NOx and NMVOC's
- PAN has longer life times at lower temperatures found in the free-troposphere →PAN can be transported long distances
- At higher temperatures (lower altitudes) PAN → NO_X → ozone



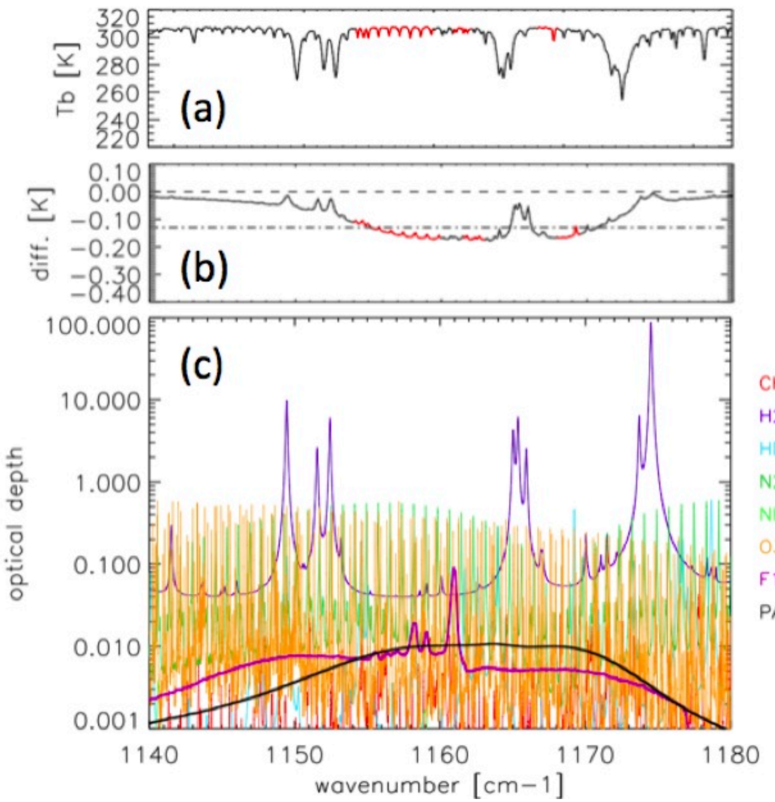
Recent GEOS-Chem with increased PAN pre-cursors and higher injection heights based on aircraft and ground data predicts that PAN peaks in the summer over Asia due to convective lofting of surface emissions and lightning (Fischer et al., ACP, 2014)

Hypothesis: Are Models Not Injecting Enough Ozone Pre-cursors Into The Free Troposphere?

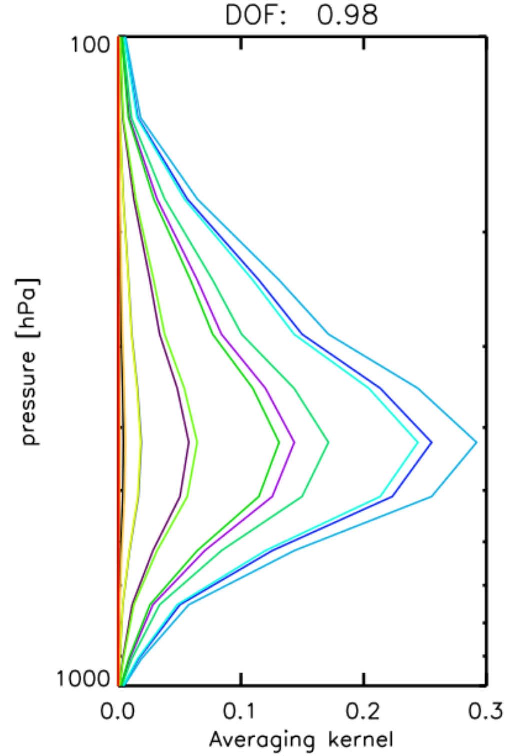
Coheur et al., ACP, 2009; Alvarado et al, Atm, 2011; Payne et al. AMTD 2014

- TES can measure elevated PAN (>200 pptv) in the free troposphere
- Uncertainty ~30% for free-tropospheric average

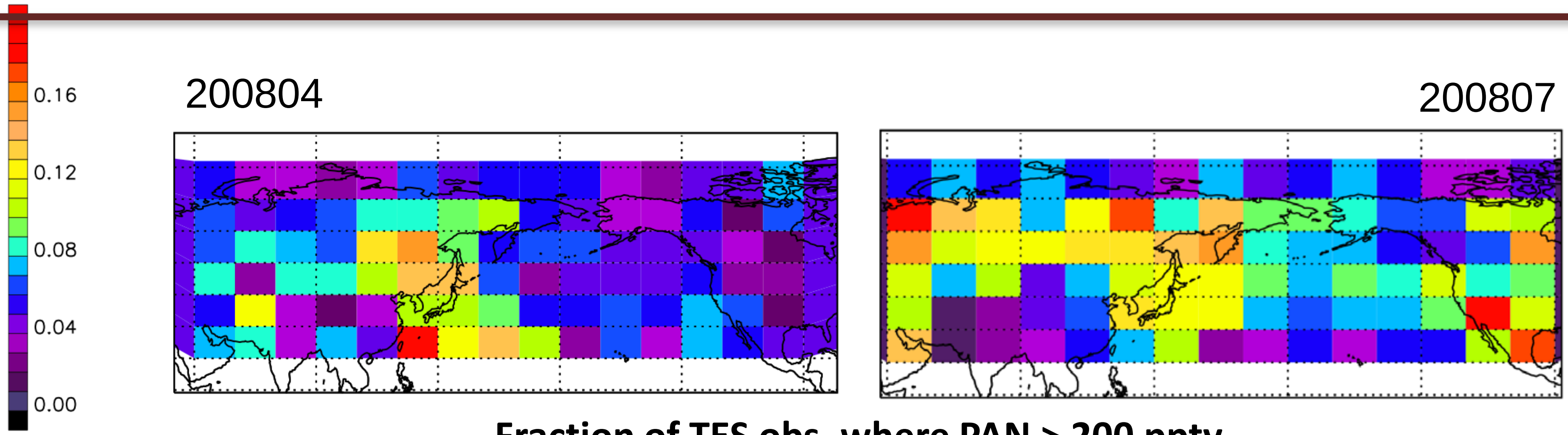
Observed PAN Spectral Signature is Small but Measurable



Peak Sensitivity in Middle / Upper Troposphere



Hypothesis: Are Models Not Injecting Enough Ozone Pre-cursors Into The Free Troposphere?



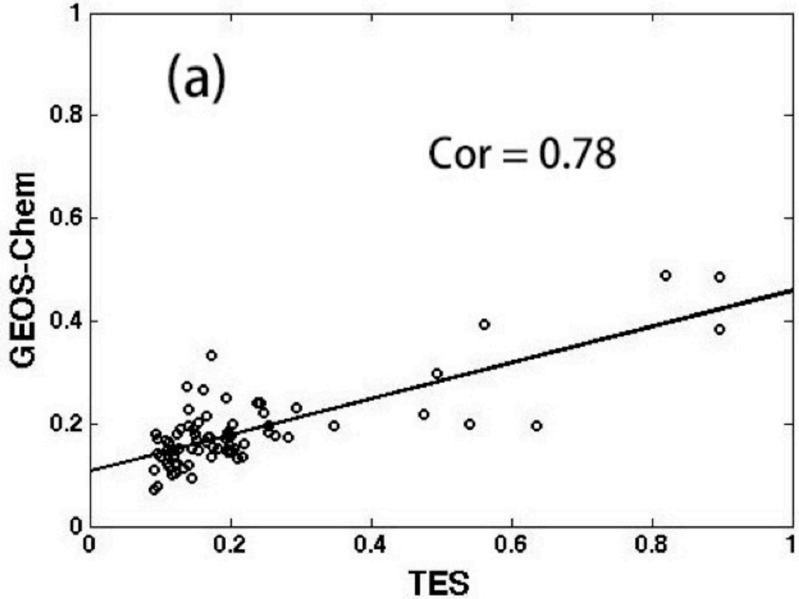
Fraction of TES obs. where PAN > 200 pptv

Seasonality of data and model agree

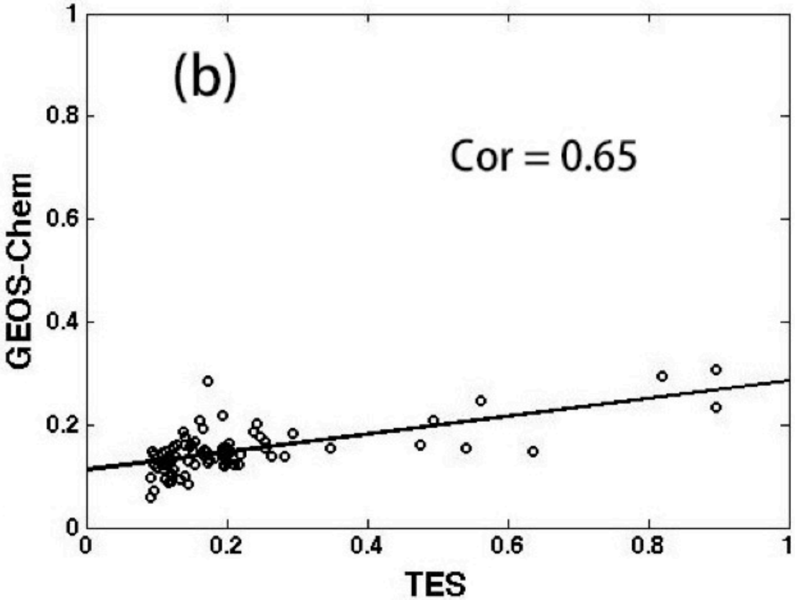
Hypothesis: Are Models Not Injecting Enough Ozone Pre-cursors Into The Free Troposphere?

TES Tropospheric Emission Spectrometer

Updated GEOS-Chem with Increased PAN Pre-cursor Emissions



Previous GEOS-Chem

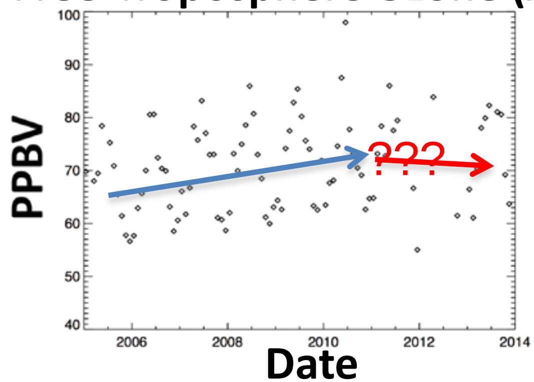


TES data indicate additional sources of PAN pre-cursors are needed → consistent with Fischer et al., ACP, 2014

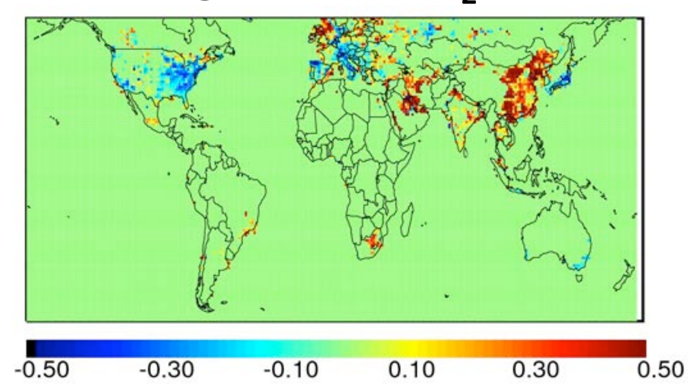
Summary and Conclusions

- Global ozone pre-cursor emissions have changed dramatically, even over the last decade, with strong increases in Asia and decreases in N. America, Europe, and Japan
- Data from last 20 years suggest important role of Asian emissions on N. American ozone
  10%/yr NOx → 0.7 PPB/yr ozone (in free troposphere)
- Chemical transport models show a smaller role for Asian emissions
  10%/yr NOx → 0.2-0.4 PPB/yr ozone
- Possible explanations for difference between data and model:
  - *Changes in lightning and fires: Not likely*
  - *Changes in STE: Model estimates might be too large, additional "regional" studies needed*
  - *Changes in circulation: Cannot rule out this effect*
  - *Not enough surface emissions and ozone lifted into free-troposphere? New TES PAN data support hypothesis but magnitude of effect is still to be determined*

TES Free-Troposphere Ozone (Asia)



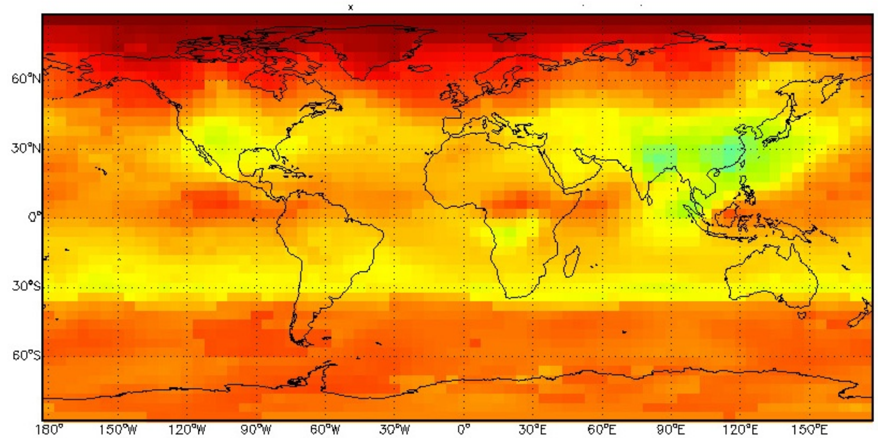
Fractional Change in OMI NO_2 since 2005



Hypothesis: Are Models Not Injecting Enough Ozone Pre-cursors Into The Free Troposphere?

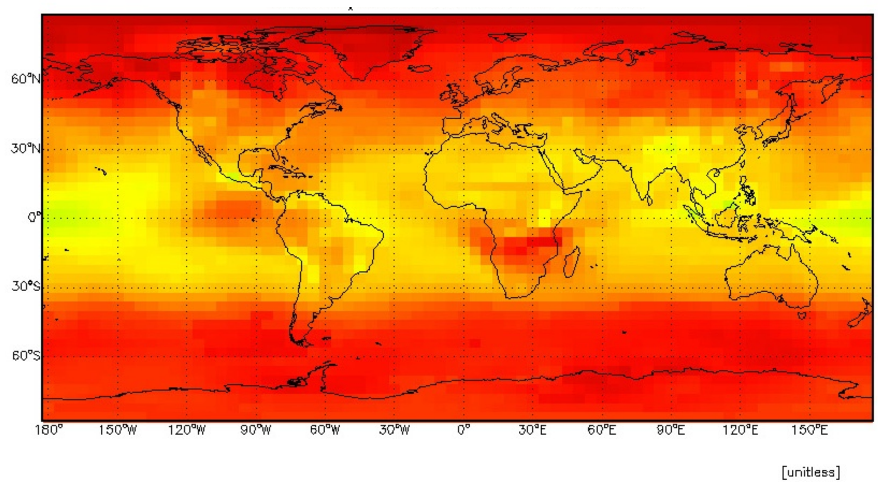
Fractional Increase of New to Older GEOS-Chem (July 2008)

O_3



~0% to 20%

PAN



~10%-50%

Increased PAN in the free-troposphere leads to increased O_3 production