Impact of Local and Non-local Sources of Pollution on Background US ozone: Role of LEO and GEO Sounders in a Composition Constellation

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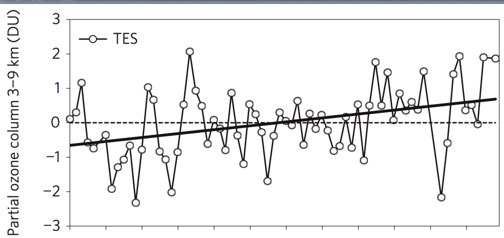
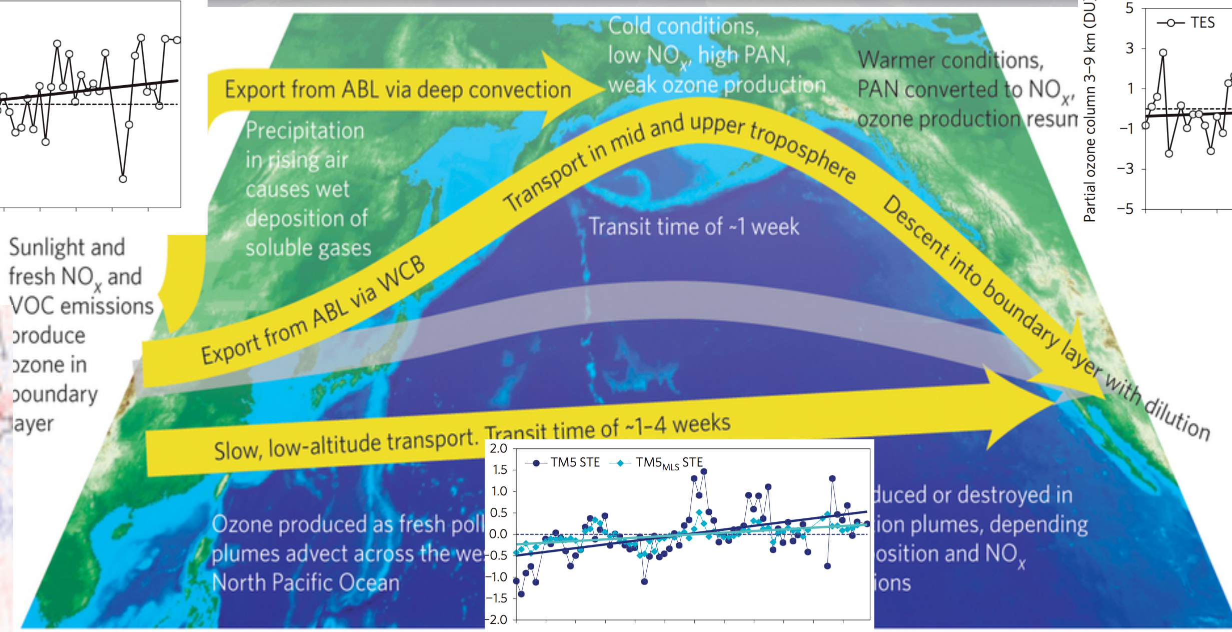
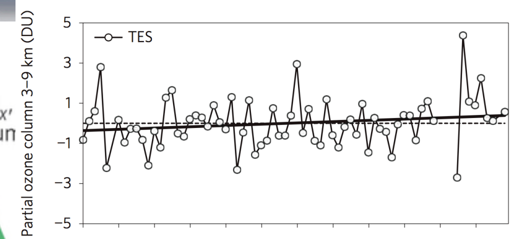
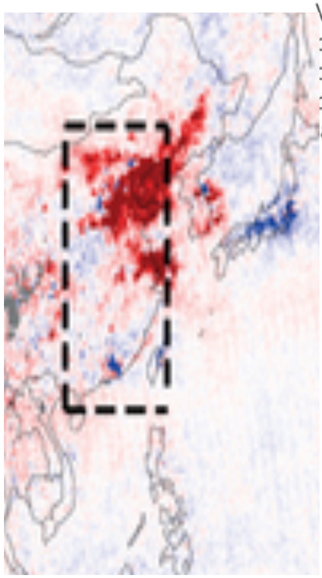
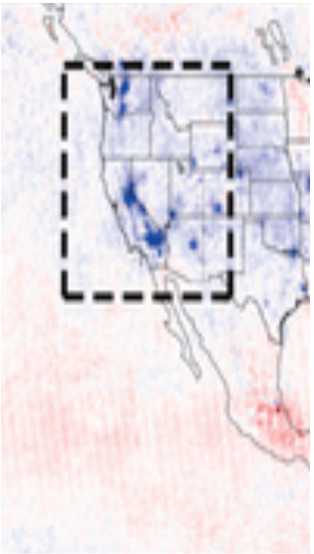
Eastern China emissions offset 43% of the expected reduction in mid-tropospheric ozone over the Western US from 2005-2010 (*Verstraeten et al., Nature Geosci., 2015)*











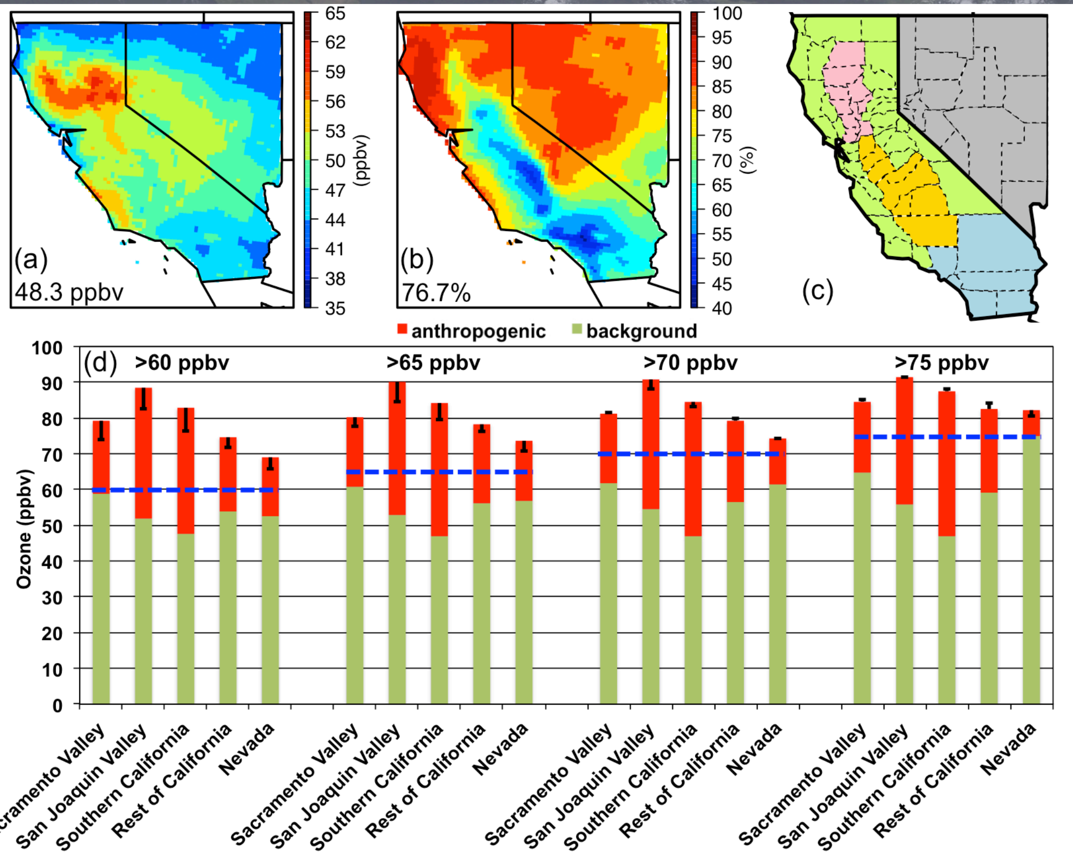
MLS: Temporary increase in downward transport from the stratosphere partly due to 2009-2010 El Nino.
Explains 50% E. China ozone increase
Offsets 57% of W. US ozone decrease

TES: 7% Increase in mid-tropospheric ozone
OMI: 21% increase in NO_x emissions.
Explains 50% of the ozone increase.

TES: No change in mid-tropospheric ozone
OMI: 21% decrease in NO_x emissions.
Should have given a 2% decrease in ozone

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

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.

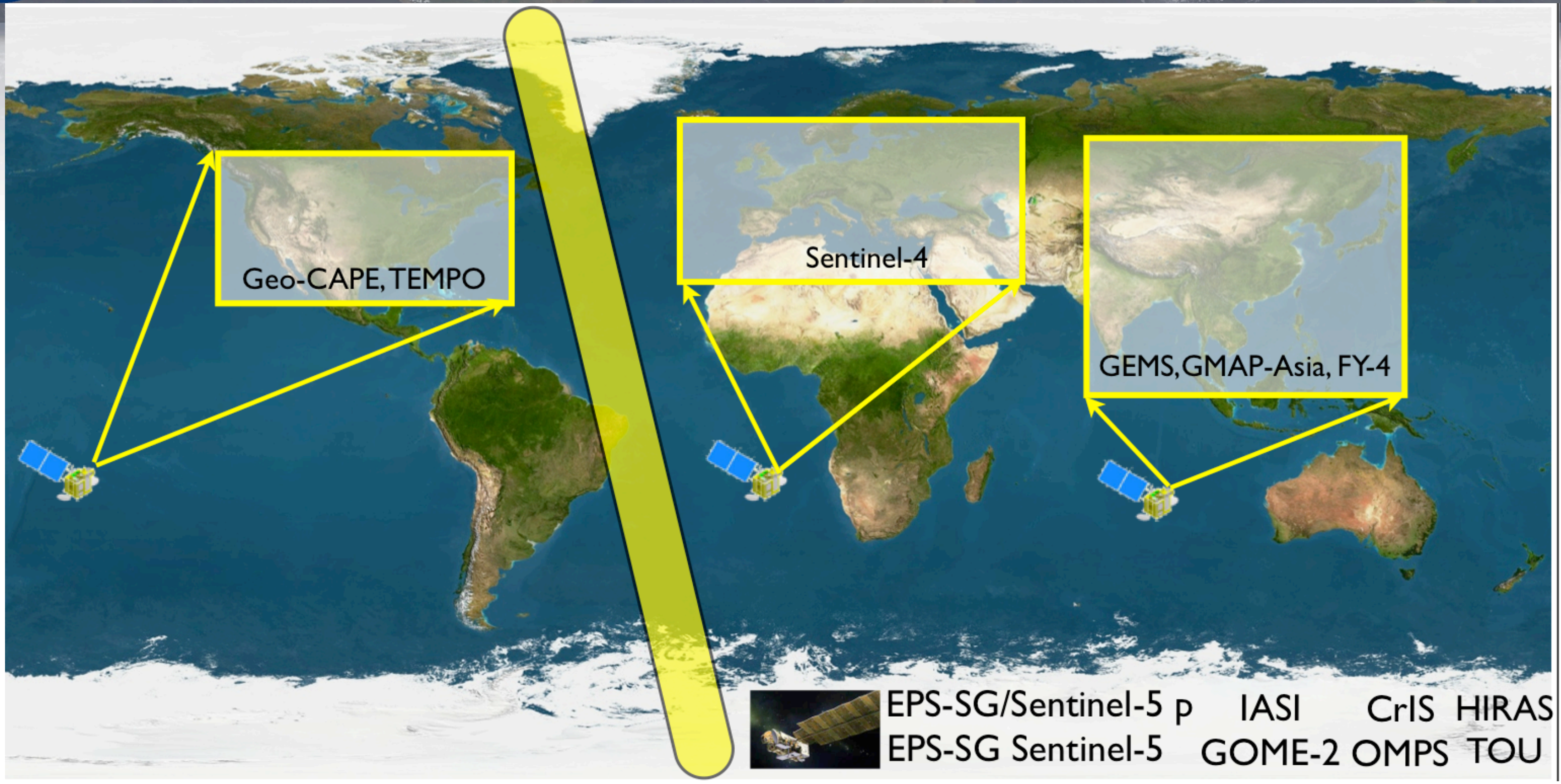


Investigation:
Huang *et al.,* JGR (2015) improved ozone source attribution by integrating Tropospheric Emission Spectrometer (TES) ozone and Ozone Monitoring Instrument (OMI) nitrogen dioxide into a state-of-the-art multi-scale assimilation system. Ozone attribution was estimated at surface monitoring 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 emissions to meet standards.

Toward an Composition-Climate Constellation



Bowman, *Atm. Env.* 2013

- A new generation of geostationary and low-earth orbiting (LEO) sounders will form a new composition-climate constellation.
  - Geostationary sounders including GEO-CAPE, TEMPO, Sentinel-4, GEMS will provide an unprecedented number of composition observations at high spatial resolution.
  - LEO sounders provide the global picture and thread the geostationary observations together.
- How does the constellation improve knowledge of global air quality?

Controls of mean ozone

Define the mean ozone over the Environmental Protection Agency Region 9 (California, Nevada, Arizona) for May, 2006

$$J(E(\mathbf{x};t)+O_3(\mathbf{p};t_0))=\overline{O_3(EPA09)}$$

The change in mean ozone in EPA09 is the product of the change in global emissions and concentrations with the sensitivity of EPA09 to those parameters

$$\delta J=\left(\nabla_E J\right)\delta\mathbf{E}+\left(\nabla_{O_3} J\right)\delta\mathbf{O_3}$$

The sensitivity of mean ozone (δJ) can be efficiently computed with the adjoint of GEOS-Chem.
J is defined separately for mean surface and mean column ozone over EPA.

Regional Distribution of the EPA09 Ozone Sensitivity

$\sum_{Region} \delta J/\delta \ln E \,/\, \sum_{Globe} \delta J/\delta \ln E$

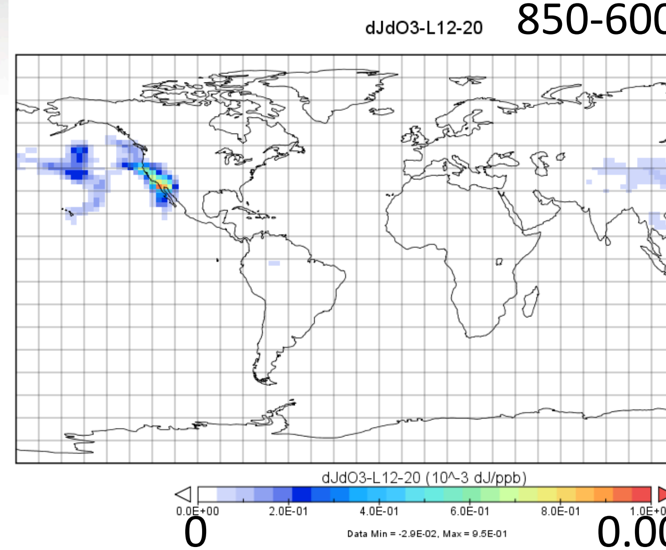
- Reductions in OMI-constrained NOx emission estimates from 2005-2009 decreased mean surface ozone by 0.9% relative to all NOx emission contributions
- Increases in Chinese NOx emissions in 2005-2010 largely offset local EPA09 emission contributions to mean surface ozone.
- China emission changes contributed more to mean column ozone than EPA09 emissions in 2005-2009.
- SE Asia emission changes contributed more to mean column ozone than extra-EPA09 regional emissions 2005-2009.

Sensitivity of EPA ozone column to ozone

Sensitivity of mean column ozone over EPA09 for May 2006 with respect to ozone initial conditions integrated over different pressures

$$\left(\nabla_{O_3(p_i \rightarrow p_j)} J\right)$$

850-600 hPa

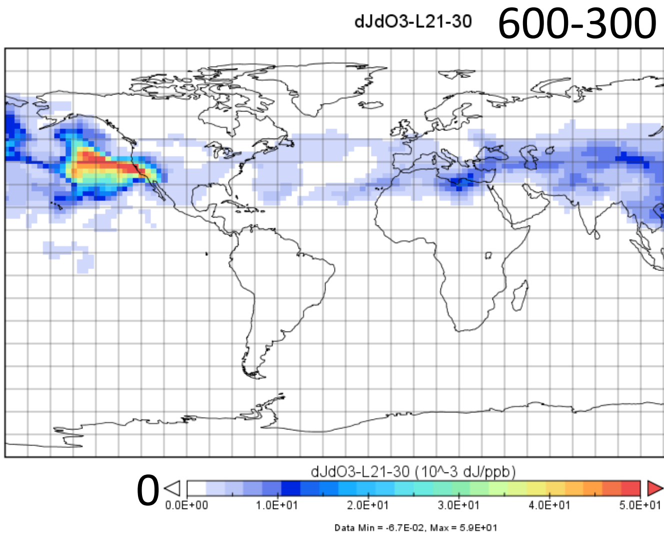


0 — 0.001 ppb/ppb

Changes in the mid-troposphere (600-300) of 1ppb lead to a change in the mean EPA09 column of up to 0.05 ppb.

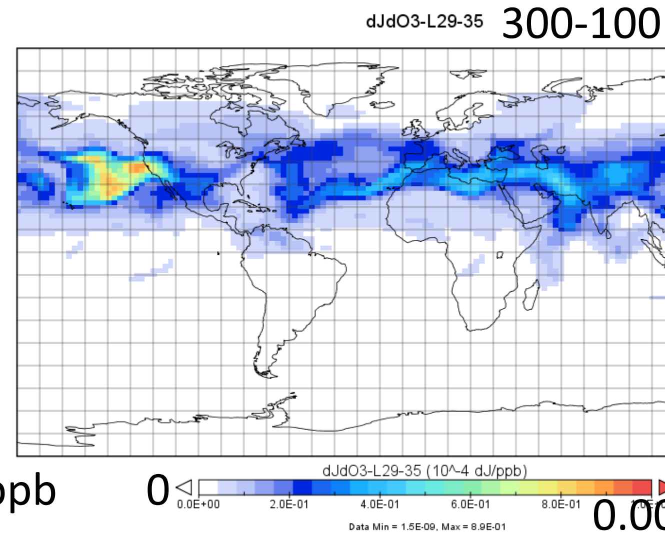
A change of 1ppb across the entire mid-troposphere, i.e., summing sensitivities globally, would lead to a 0.24 ppb. The mid-troposphere dominants the variability of the EPA09 column

600-300 hPa



0 — 0.05 ppb/ppb

300-100 hPa



0 — 0.0001 ppb/ppb

Sensitivity of EPA surface ozone to global ozone

Sensitivity of mean surface ozone with respect to ozone initial conditions integrated over different pressures

$$\left(\nabla_{O_3(p_i \rightarrow p_j)} J_{surf}\right)$$

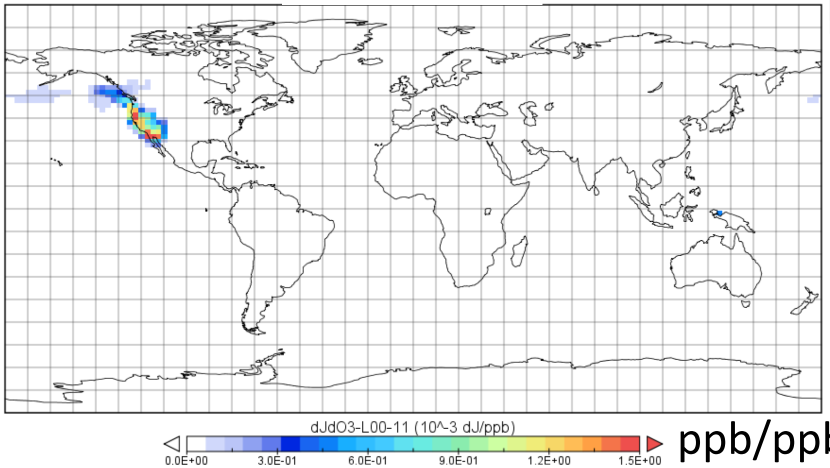
Changes in the near-surface (1000-850) ozone of 1ppb lead to a change in the mean EPA09 surface ozone of up to 0.0015 ppb. Lower tropospheric (850-600) is similar with up to 0.001 ppb.

For example, if Los Angeles ozone changed by 1 ppb, the monthly mean EPA09 ozone would change by ~0.0015 ppb.

$$0.0015\,ppb = \left(\nabla_{O_3(p_i \rightarrow p_j)} J_{surf}\right)\delta O_3^{LA} = \left(\nabla_{O_3(p_i \rightarrow p_j)} J_{surf}\right)[0,\cdots,1ppb,\cdots,0]^T$$

Most of the sensitivity of mean EPA09 ozone is local and near-local ozone. The total sensitivity is 0.083 ppb/ppb (surf-850) and 0.1 ppb/ppb (850-600).

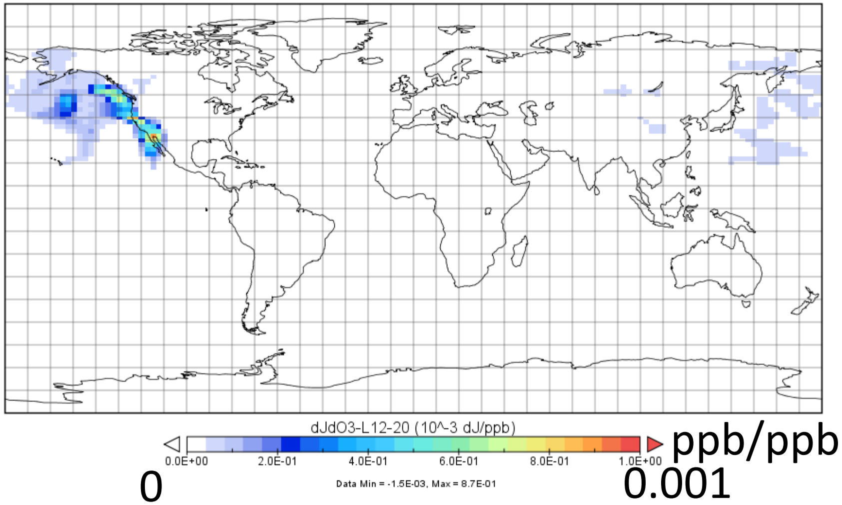
surf-850 hPa



ppb/ppb

0 0.0015

850-600 hPa



ppb/ppb

0 0.001

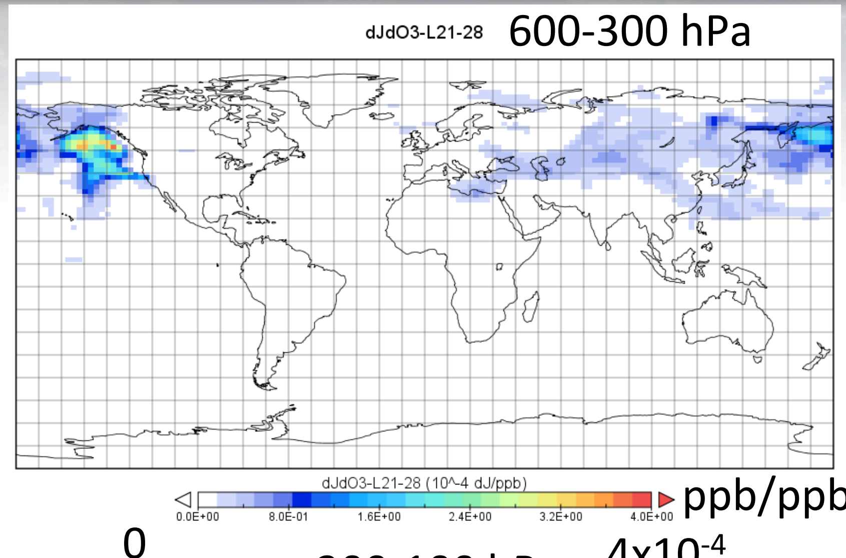
Sensitivity of EPA surface ozone to global ozone

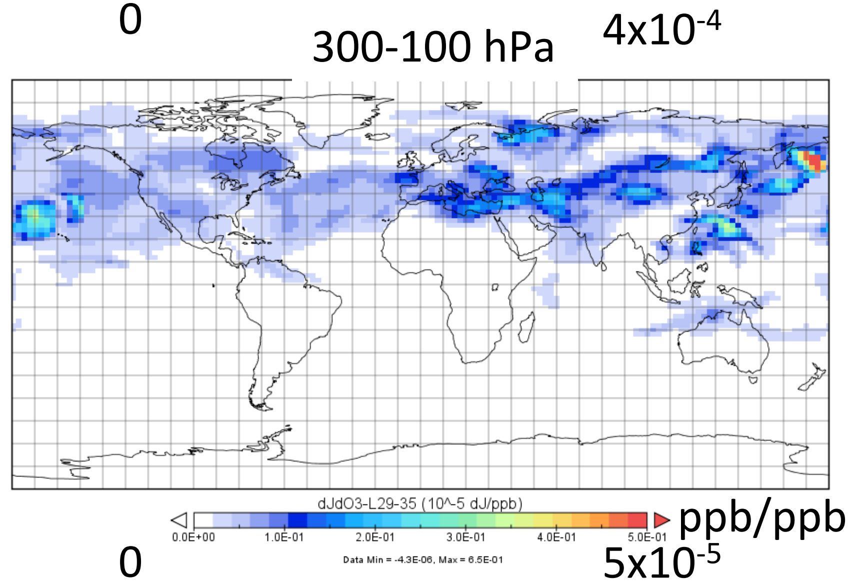
Surface EPA ozone has a different sensitivity to the spatial origin of ozone in the mid-troposphere than the column.

The total sensitivity to surface ozone to 600-300 hPa ozone is 0.097 ppb/ppb, which is comparable to the surface sensitivity, while 300-100 hPa is 0.003 ppb/ppb

These sensitivities define the pollution pathways between non-local sources of ozone and the surface.

These pathways can be shared by both stratospheric intrusions and long-range pollution transport.

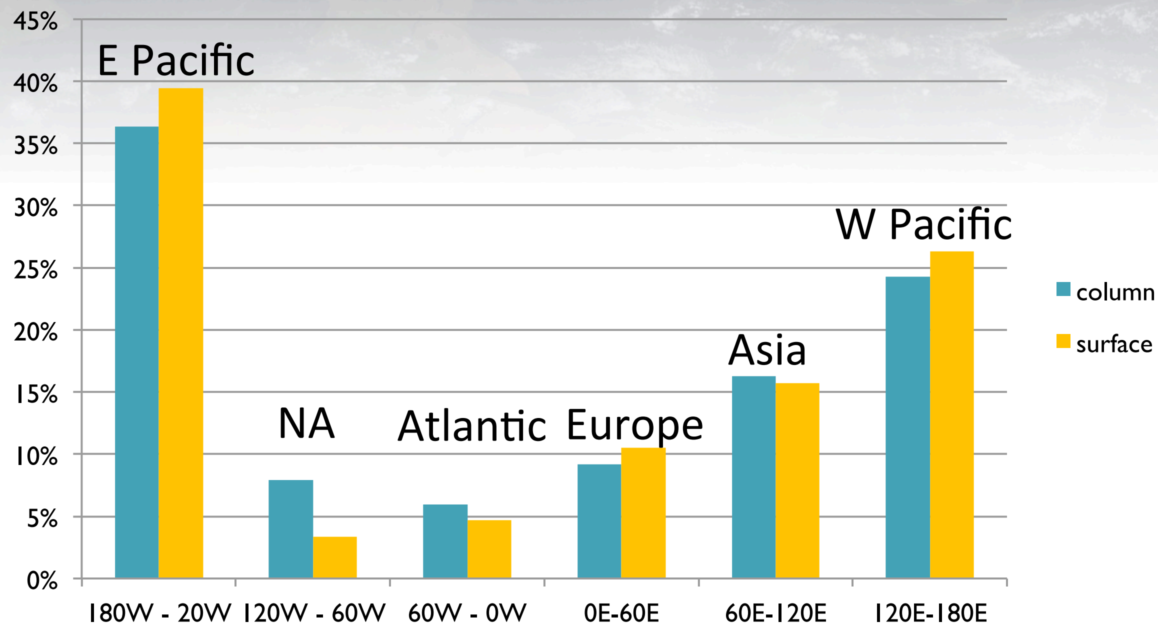




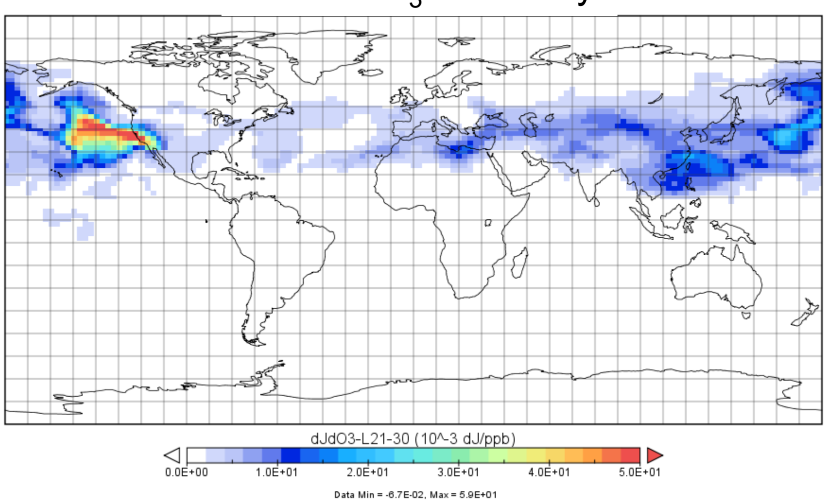
Zonal mid-tropospheric contribution to EPA09 ozone

The greatest sensitivity of EPA09 mean surface ozone is from the E.Pacific, which accounts for almost 40% of the total sensitivity.

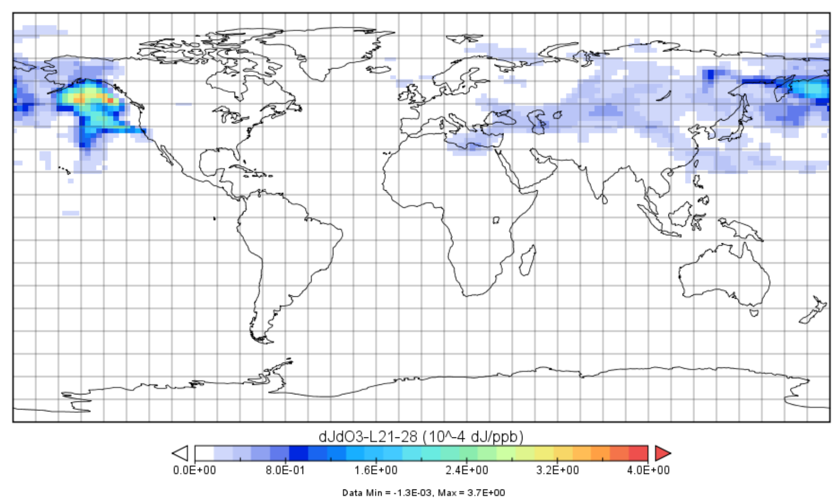
The Pacific accounts for ~65% of the total sensitivity, much of which will not be measured by the geostationary part of the constellation.



Column O_3 sensitivity



Surface O_3 sensitivity



Sensitivity of EPA surface ozone to global ozone sampled by LEO sounders

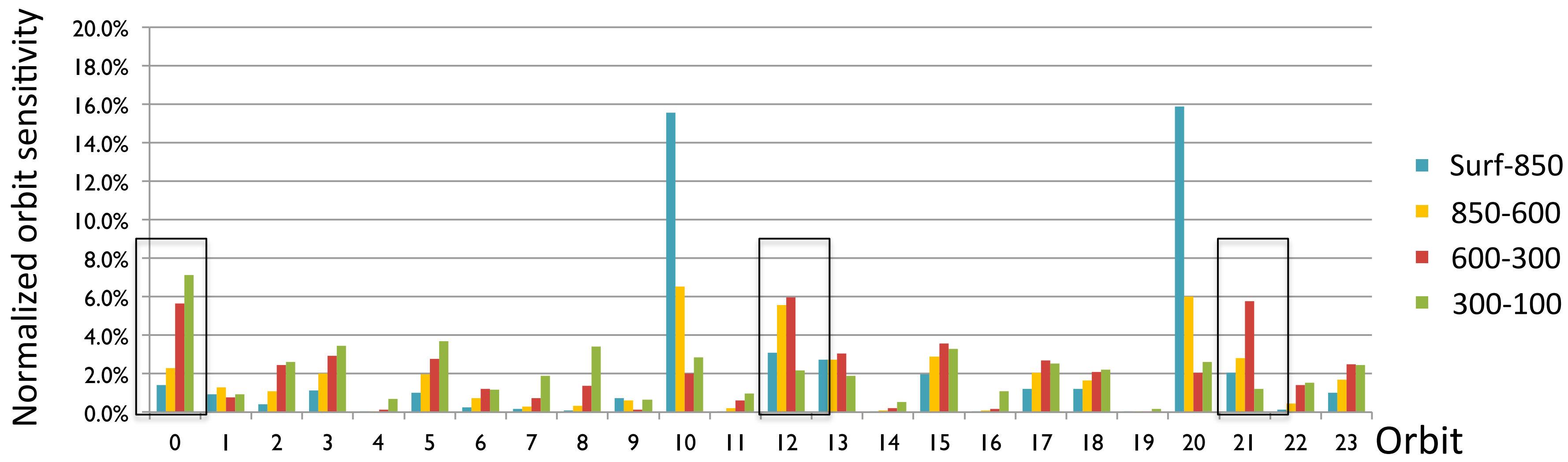
LEO sounders like AIRS or CrIS provide daily coverage but sample different parts of the world at different times.

The sensitivity of EPA09 surface ozone to global ozone sampled along individual sounder orbits can be calculated (neglecting transport over the initial day).

$$\left(\nabla_{O_3(p_i \rightarrow p_j, orbit_k)} J\right)$$

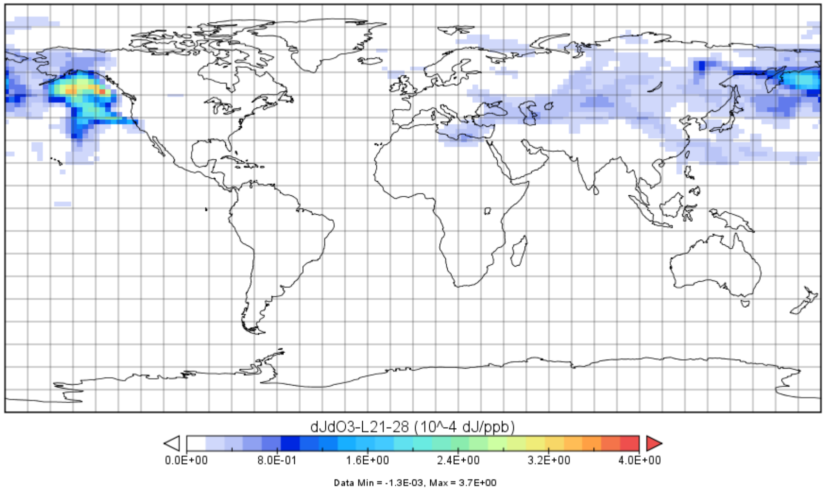
$$\frac{\sum_{i \in altitude} \left(\nabla_{O_3(p_i \rightarrow p_j, orbit_k)} J\right)}{\sum_{l \in globe} \left(\nabla_{O_3(l)} J\right)} \times 100\%$$

This sensitivity is normalized by the total sensitivity

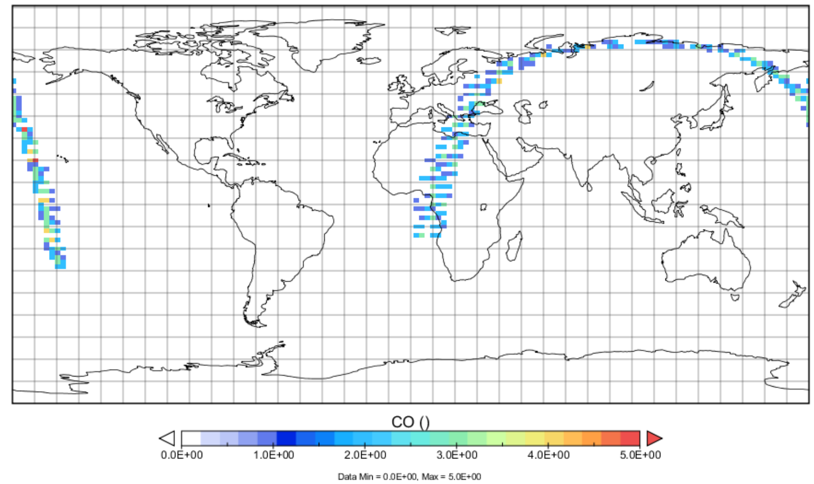


Simulated AIRS sampling

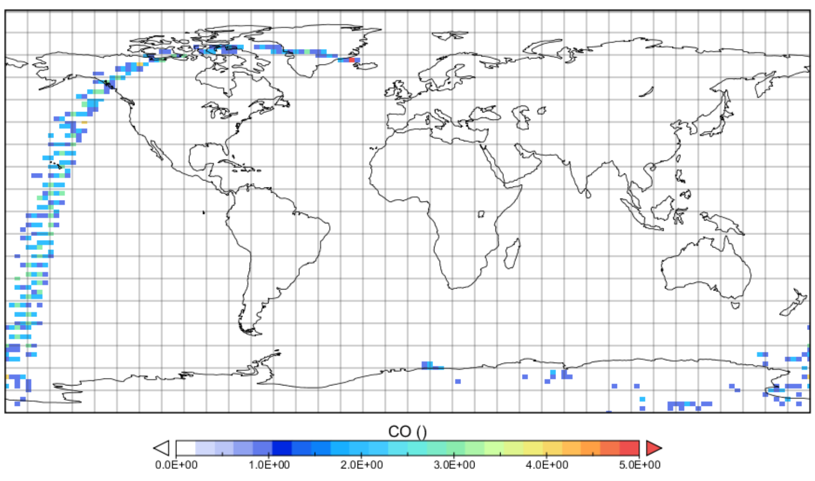
Sensitivity (600 hPa – 300 hPa)



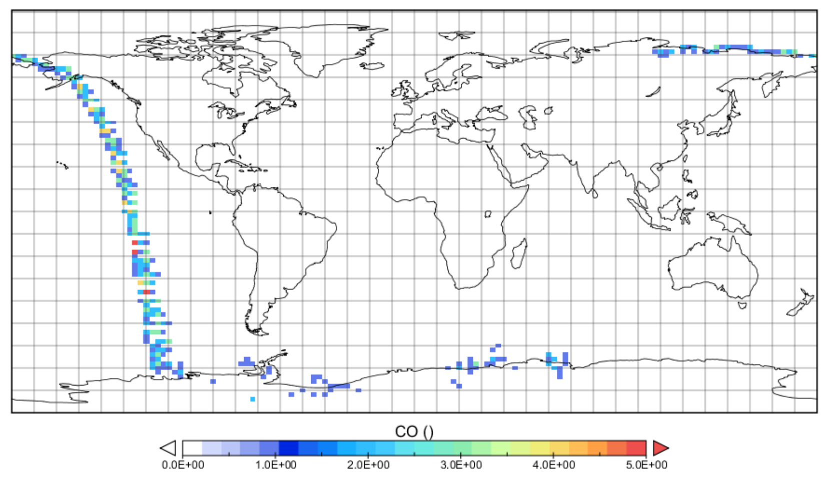
Samples during 00:00 – 01:00



Samples at 12:00 – 13:00



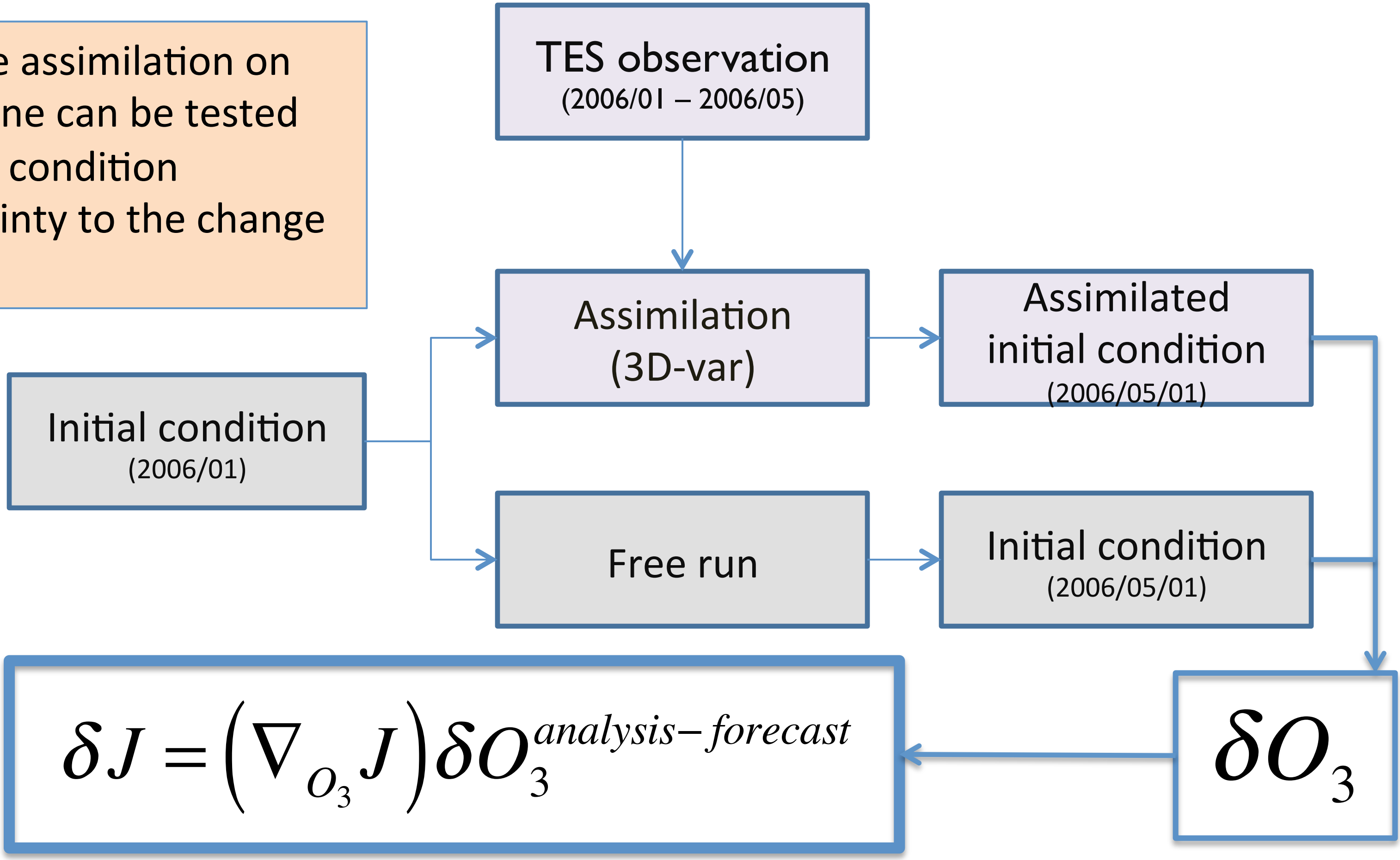
Samples during 21:00 – 22:00



Impact of initial condition uncertainty in surface and column ozone

The attribution of background ozone requires a good knowledge of ozone boundary conditions, especially the altitude at which ozone enters.

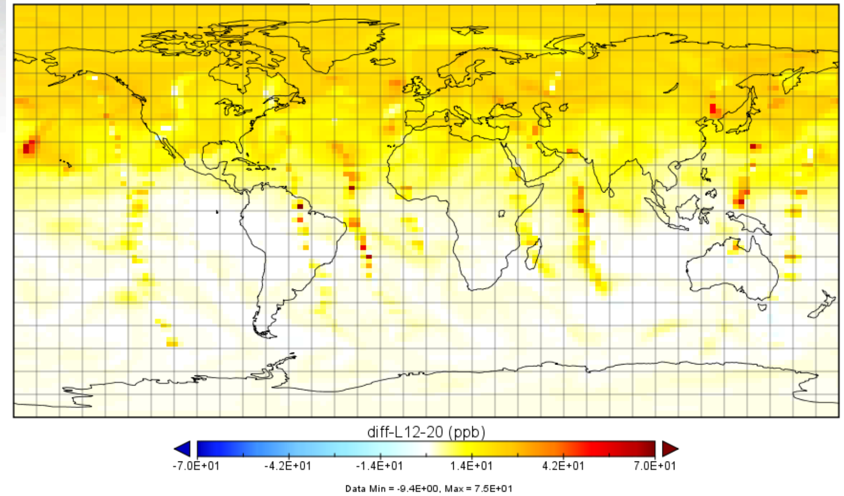
The impact of ozone assimilation on the background ozone can be tested by projecting initial condition differences/uncertainty to the change in background

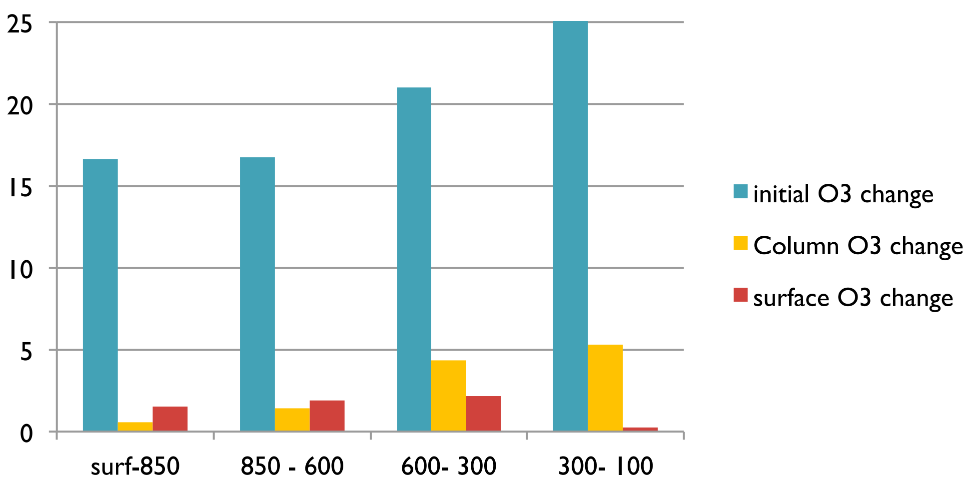


Impact of assimilated ozone on background EPA09 ozone

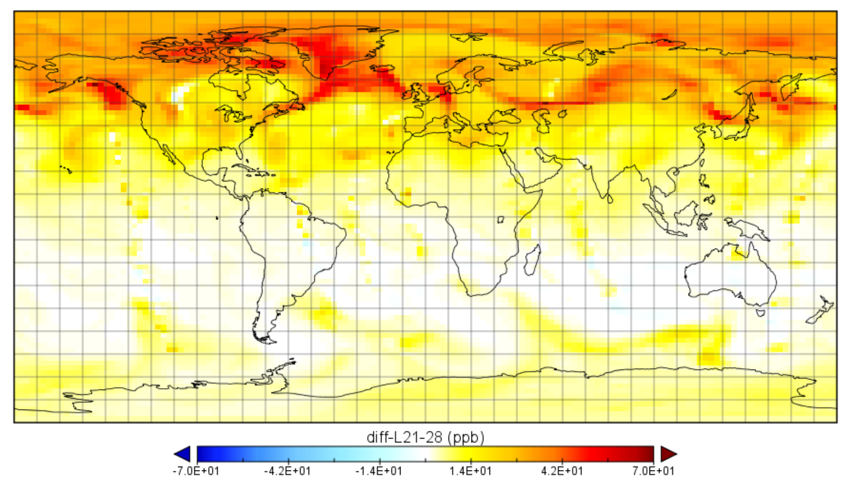
Vertical range	Pressure range (hPa)	O_3 change / cell (ppb)	Column O_3 (ppb)	Surface O_3 (ppb)
L00-11	Surface – 850	16.63	0.59	1.52
L12-20	850-600	16.75	1.42	1.9
L21-28	600-300	20.98	4.34	2.17
L28-35	300-100	137.39	5.31	0.24
Total	Surface -100	N/A	11.67	5.82
J (EPA-09)			57.07	39.25

850-600 hPa





300-100 hPa



Summary/Future Directions

- Changes in TES-observed tropospheric ozone Chinese emissions have offset 43% of the air quality reductions in the Western US (Verstraeten et al, 2015)
- Assimilation of TES and OMI show that localities would have less than 10 ppb ozone "margin" for a 60 ppb ozone standard (Huang et al, 2015).
- NOx adjoint sensitivity analysis shows similar offset in Western US surface ozone and attributes those changes to Chinese emissions at metropolitan scales
  - Potential of GEMS NOx observations in constellation
- Ozone analysis shows a ~0.2 ppb/ppb sensitivity of monthly mean EPA09 surface ozone to tropospheric ozone initial conditions
  - 65% of that sensitivity is in the Pacific
- Pacific ozone sensitivity quantifies pollution pathways
  - Necessity of advanced LEO sounders, e.g., AIRS/OMI, CrIS/OMPS, IASI-NG in constellation
- Ozone assimilation accuracy critical for EPA09 background ozone estimates
- Test sensitivities in summer; incorporate ozone standards
- Implement observation operators
- Relate ozone pathways to emissions
  - Links LEO and GEO sounders in constellation
- Link top-down NOx emission uncertainty to background ozone uncertainty
  - 4D-var and EnKF??
- Continued investigation of ozone assimilation impact on background ozone
  - Use nature run and compare performance with current observations?

Backup

Future arc of emissions

Update with Berkley Earth numbers

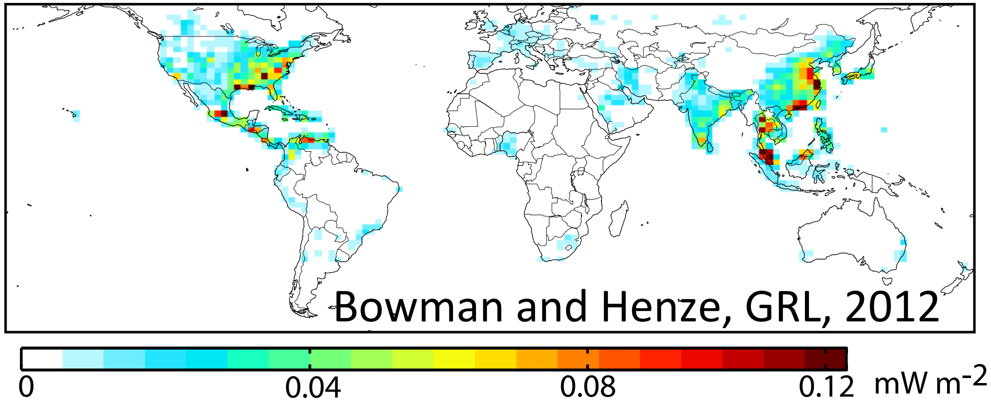
Deteriorating air quality in China has led to ~500,000 premature deaths/yr (Chen et al, Lancet 2014) prompting a "war on air pollution" from government officials.

EPA has proposed lower ground ozone standards (65-70 ppb), prompting concerns about background ozone

"Airpocalypse": Harbin, China

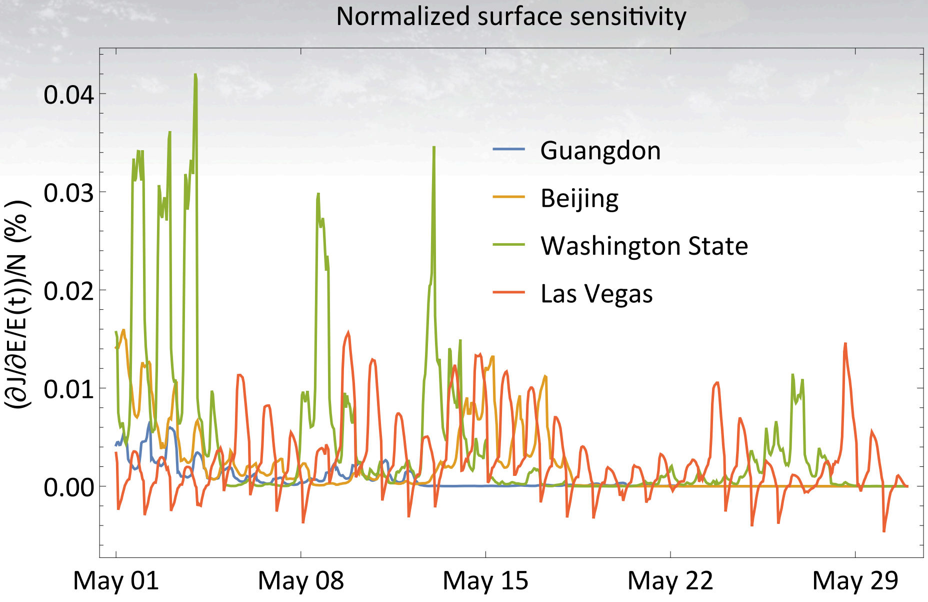
Shifts in emissions towards the equator enhance potency of short-lived climate pollutants (SLCP) and alter mitigation strategies (Schmale et al, Nature Climate Change, 2014)

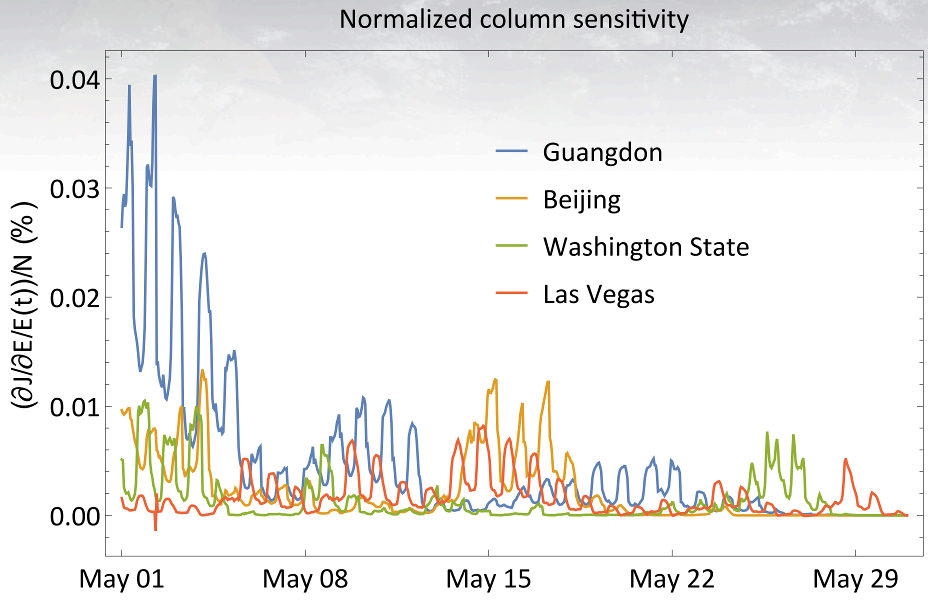
Contribution of NO_2 emissions to Ozone Radiative Forcing



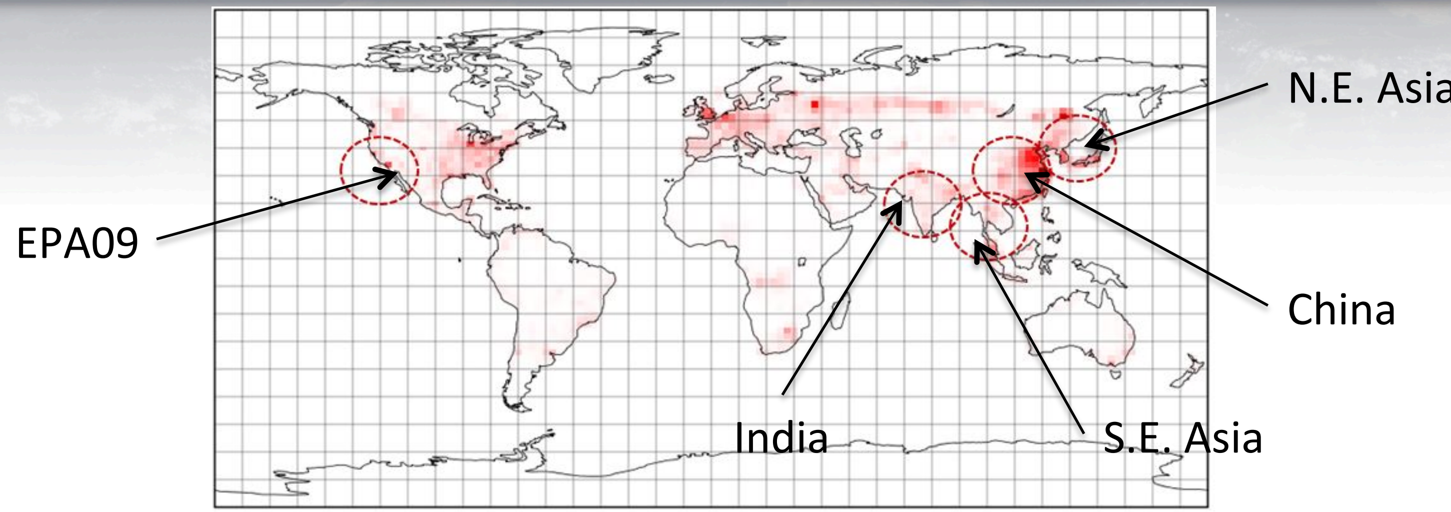
Impact of changing emissions on global air quality and climate requires a comprehensive and sustained observation system.

regional contributions





Regional Distribution of the EPA09 mean ozone sensitivity to NOx emissions



35% of mean surface ozone in EPA09 comes from emissions outside EPA09