Western US ozone and its source attribution: Multi-scale modeling analyses constrained by aircraft and satellite observations

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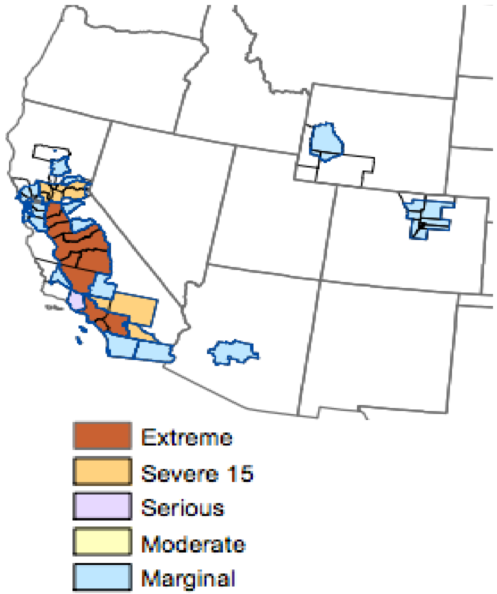
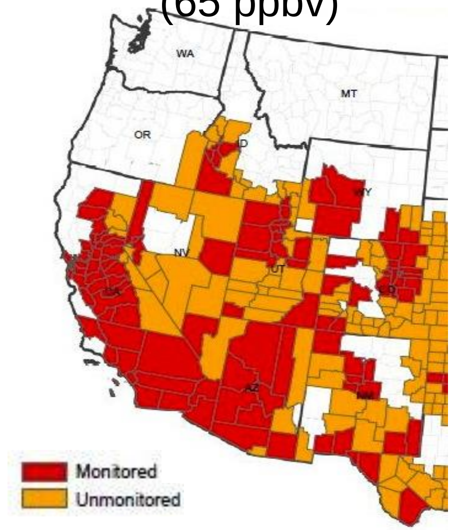
Ozone variability and exceedances in the western US is impacted by local, regional and extra-regional sources

Ozone non-attainment areas

2008 standard (75 ppbv)

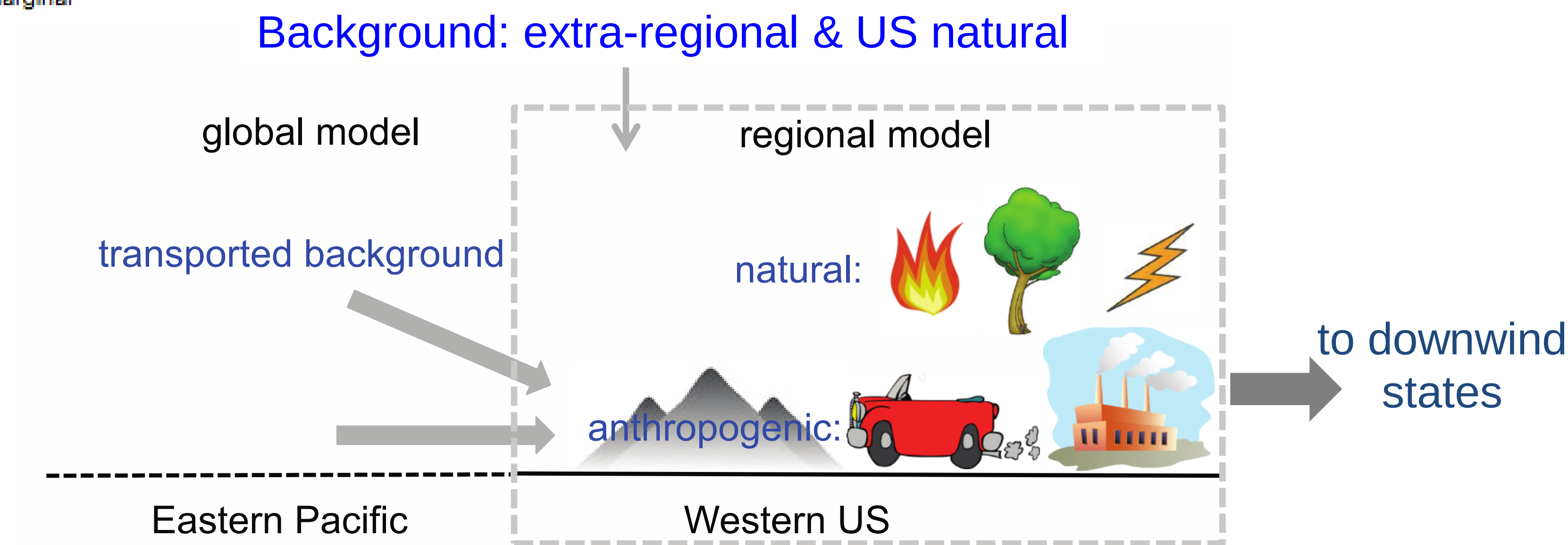


proposed standard (65 ppbv)



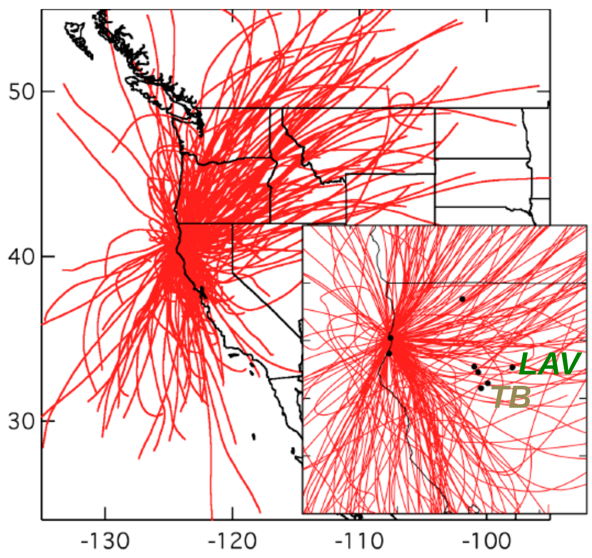
API, 2014

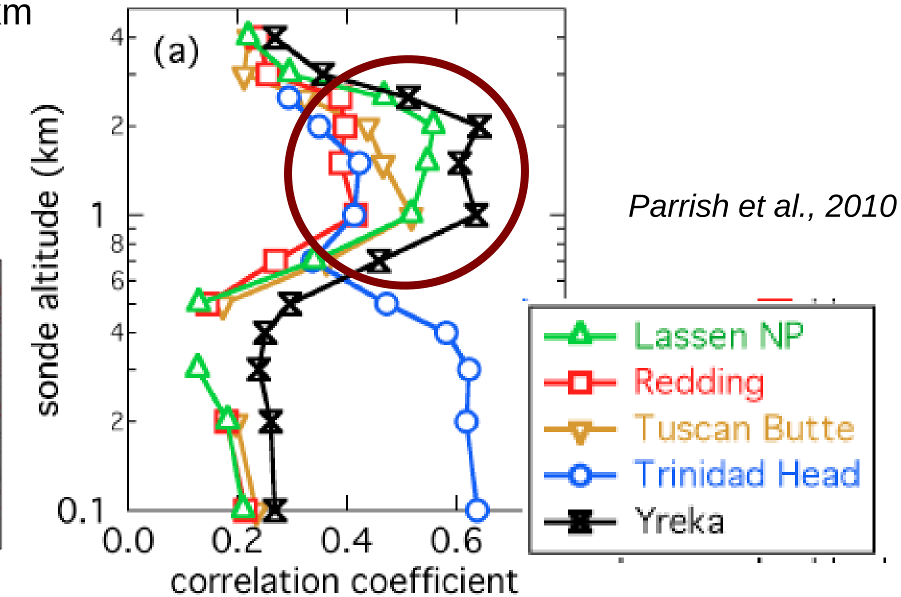
- If national ozone standard is tightened in the future: extended nonattainment areas in California and its downwind states
- Surface observations do not cover all areas that may violate the ozone standards
- Increasing importance of better understanding the total ozone and its source attribution for all areas



California surface ozone increases proportionally to the transported background

HYSPLIT from Trinidad Head ~2.5 km





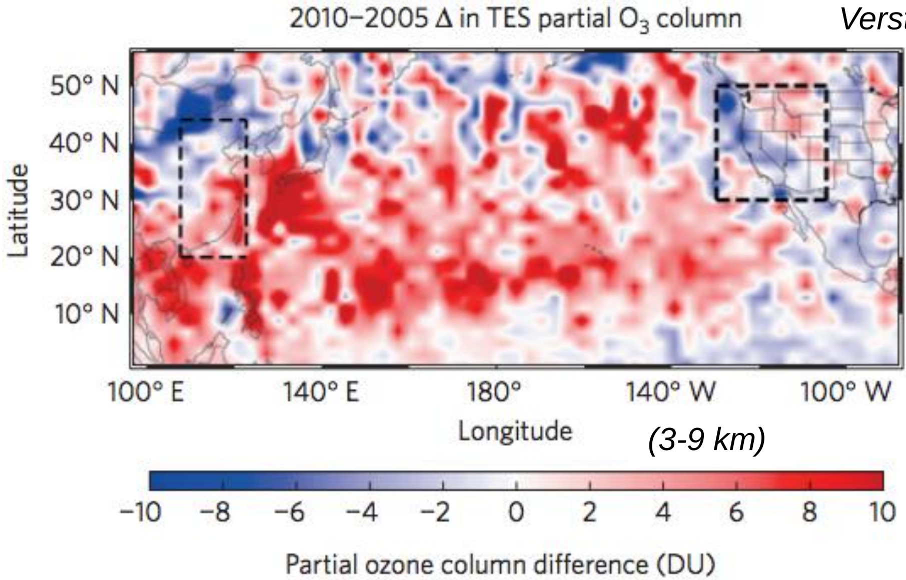
Parrish et al., 2010

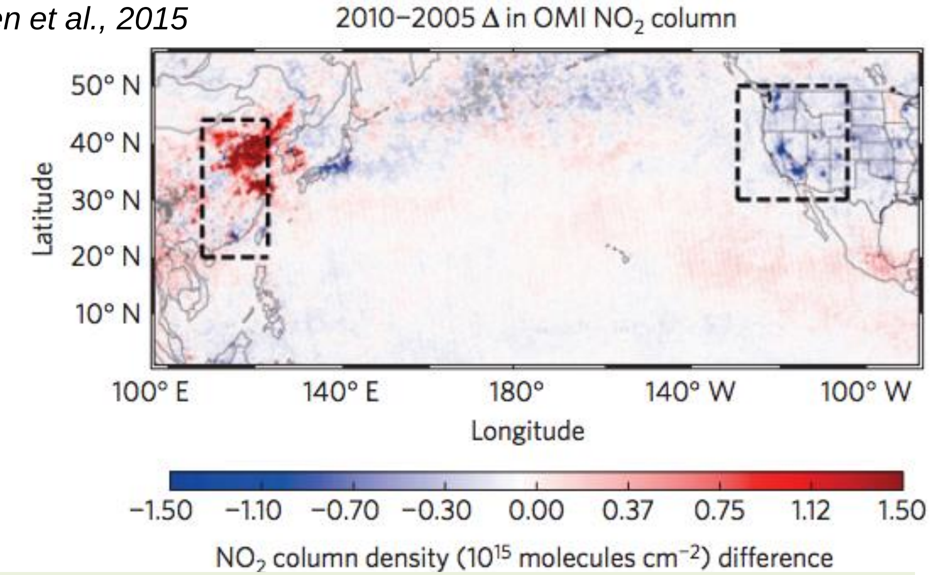
<u>Coastal-inland transport impacts total ozone and its partitioning:</u>

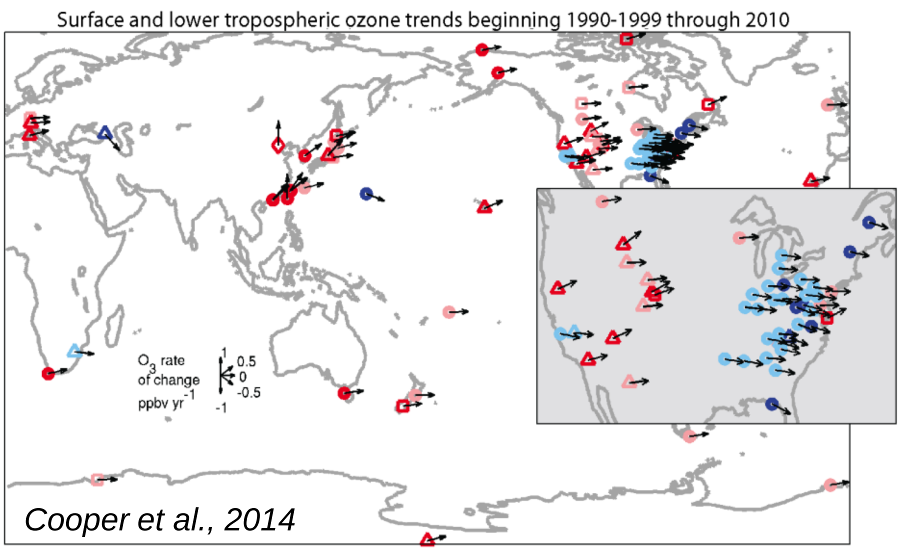
- Ozone sondes at <u>1-2.2 km</u> above Trinidad Head correlated with surface MDA8 at inland sites with different time lags, based on long-term observations:
  - Tuscan Butte (TB): 22 h, r=0.53
  - Lassen NP (LAV): 23 h, r=0.6
- Therefore, regional-scale ozone modeling over these regions can benefit from accurate boundary conditions (typically from global models)

Models well representing ozone and its source contribution in the western US is important but challenging

Verstraeten et al., 2015

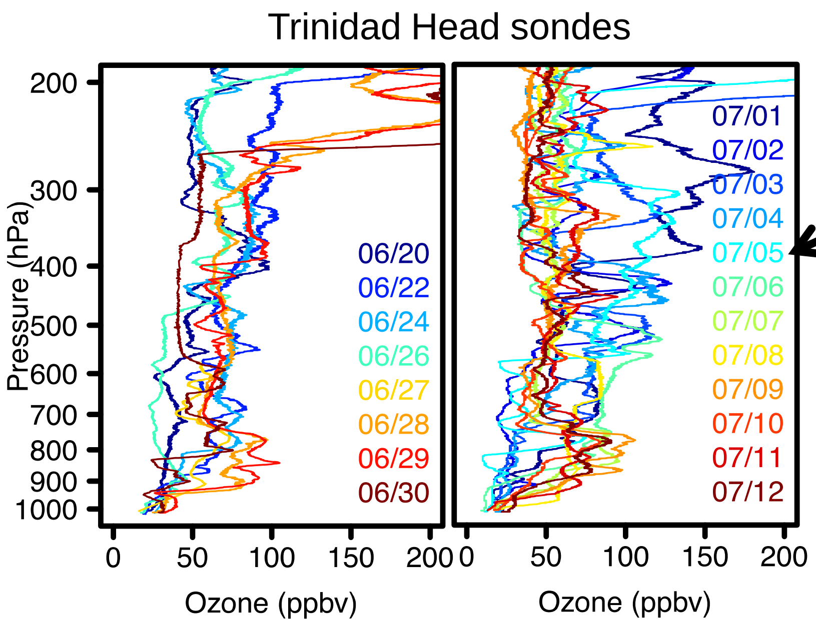


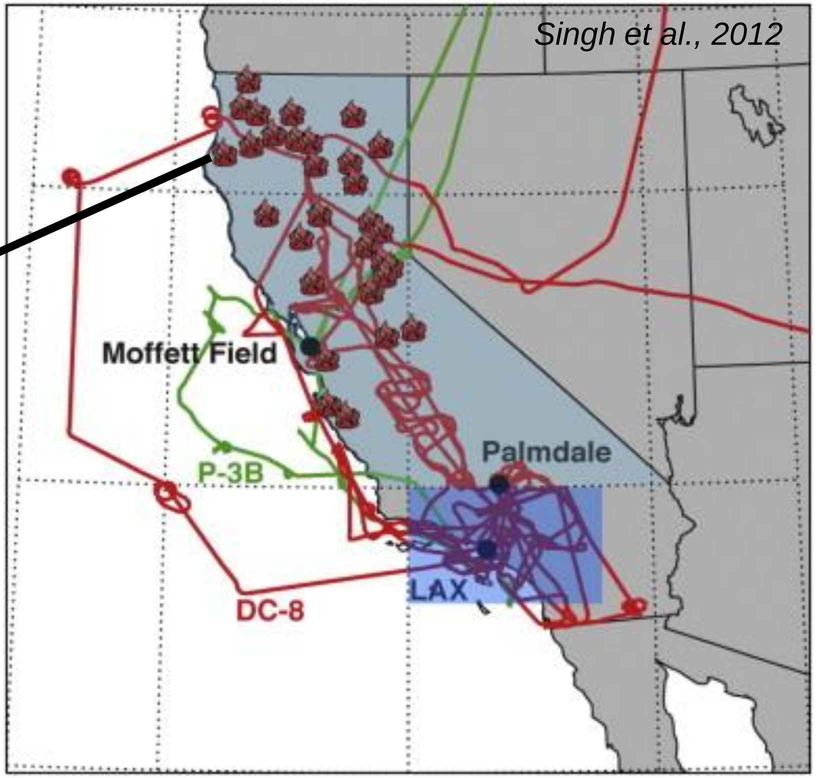


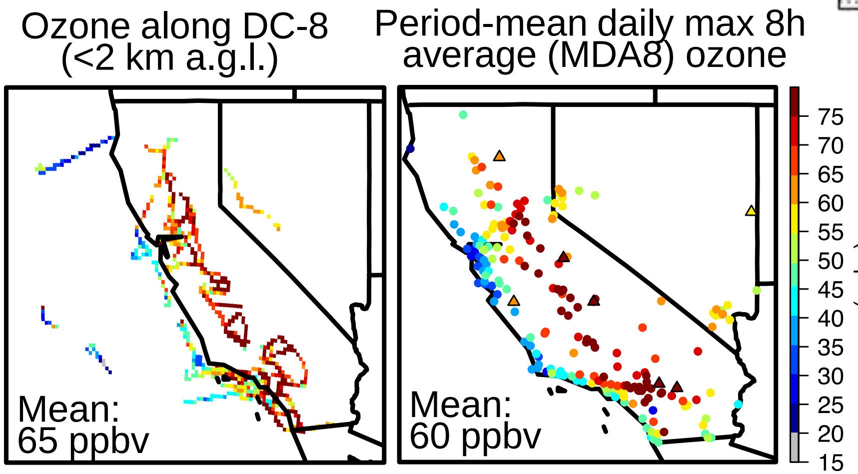


- Satellite and surface in-situ observations revealed that US and non-US air pollution changed through time
- Well representing those changes by models and their inputs (e.g., emissions) is important to AQ forecast and policy decision-making but still challenging
- How much can we benefit from aircraft and satellite observations?

We study California/Nevada June 15-July 14 2008 (during NASA's ARCTAS-CARB campaign): rich observations available



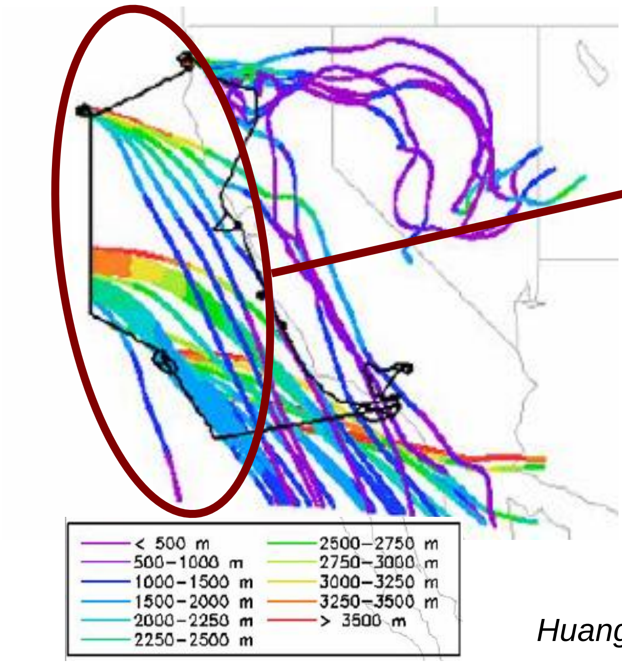


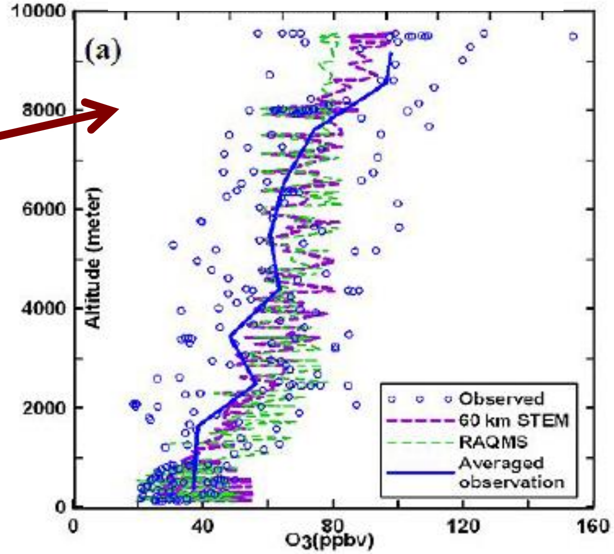


"extra-regional" pollutants mixed with local pollution from various emission sources (e.g., wildfires, urban anthropogenic)

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

Impact of boundary conditions on surface ozone during a long-range transport event starting from June 22





Huang et al., 2010

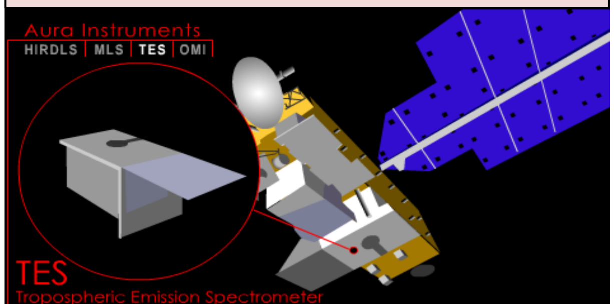
- Switching boundary conditions from positively-biased RAQMS to the aircraft-based reduced the positive biases in STEM-modeled surface ozone at N. California surface sites by ~10 ppbv on the next day

Longer-period analysis and improvement on other ozone contributors needed

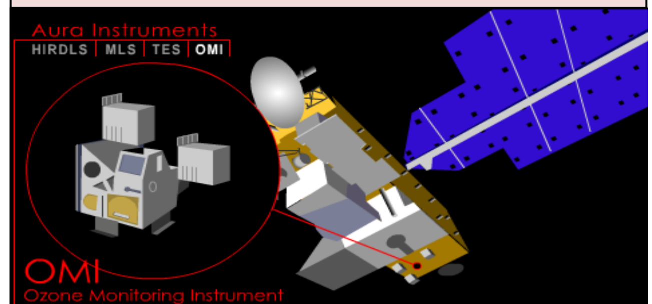
Developing a multi-scale satellite chemical data assimilation system to improve modeled ozone contributions from non-local and local sources

Using multi-model multi-scale chemical data assimilation to improve the estimated impacts on ozone from trans-boundary pollutants and US ozone precursors' emissions from different sources (e.g., urban, wildfires)

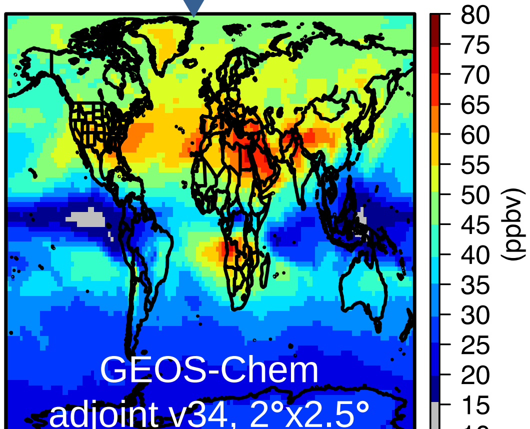
TES L2 ozone profiles



OMI NO_2 tropospheric columns

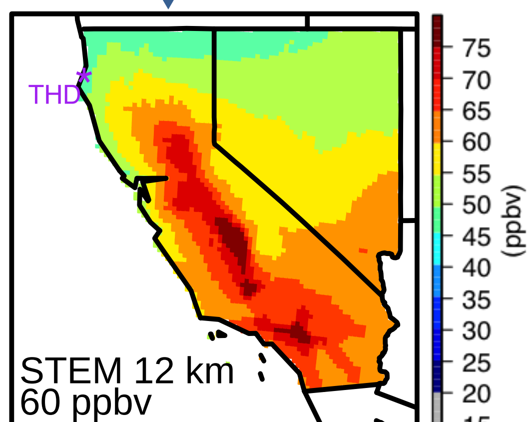


3D-Var



top/lateral boundary conditions →

4D-Var



grid-specific scaling factors on NOx emissions

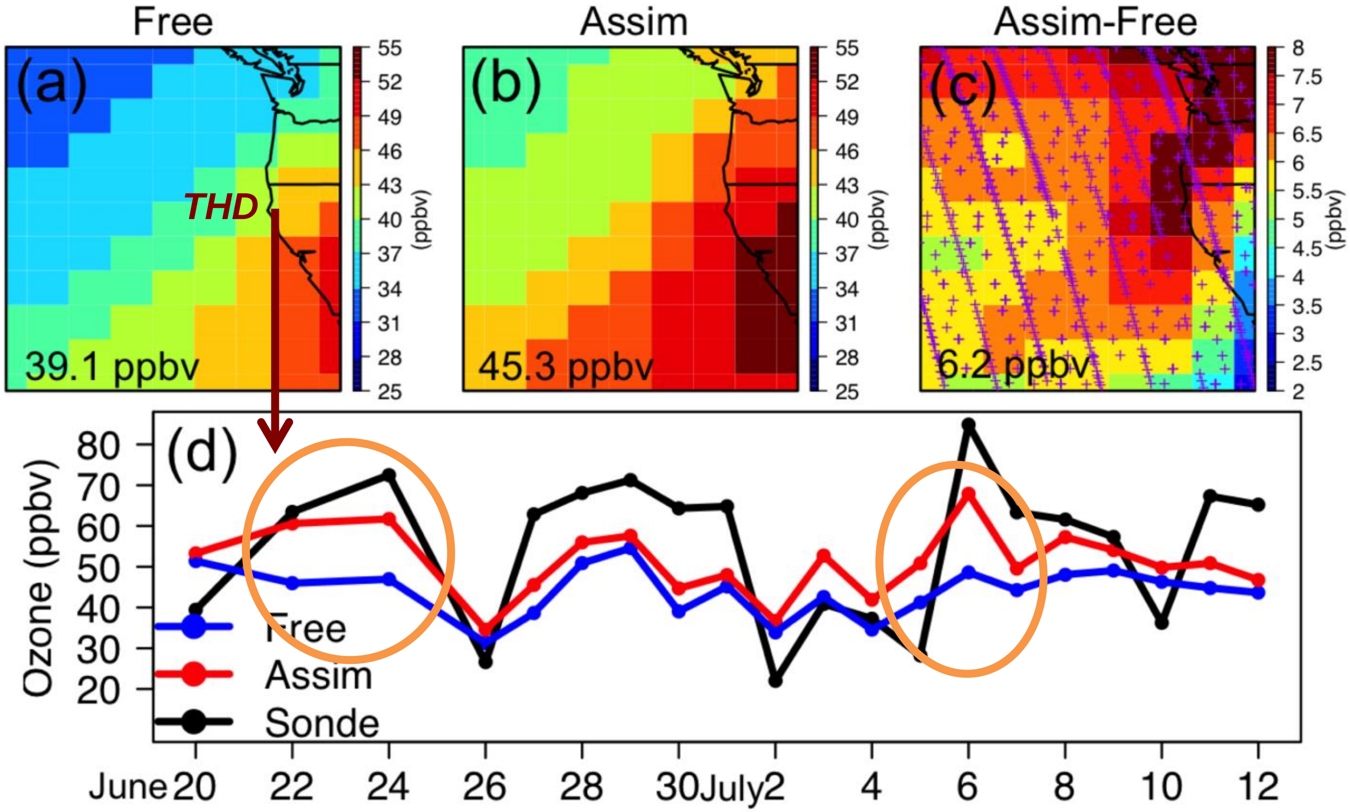
co-scale VOCs

updated chemical fields (total and background)

Cross Validation

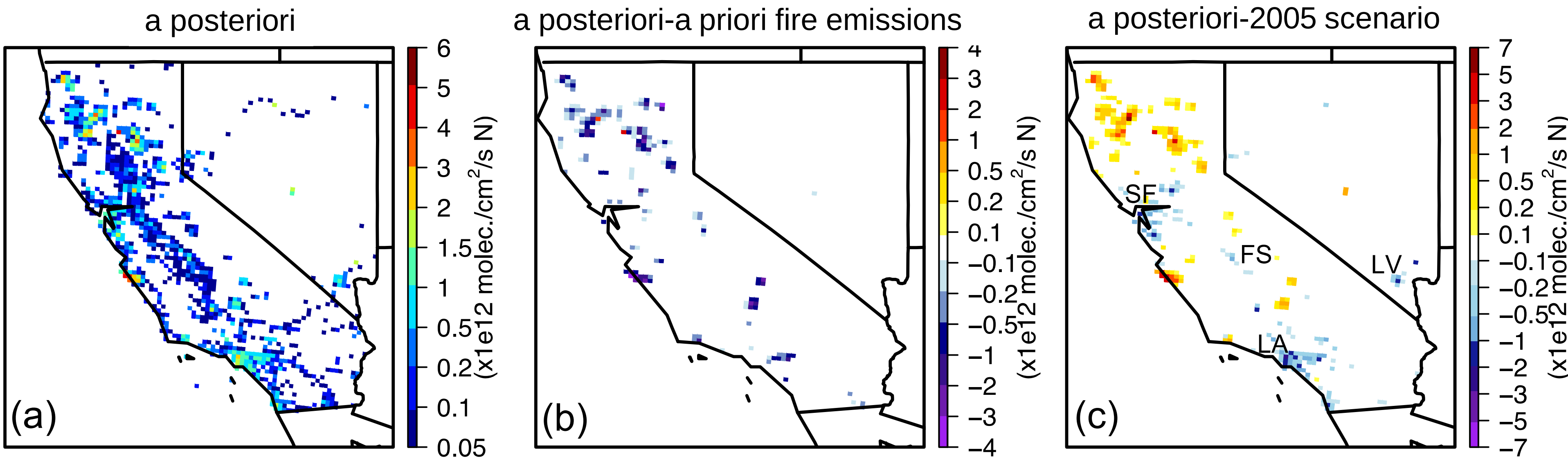
evaluate with independent in-situ (ozonesonde, surface, aircraft) observations

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



- Assimilating TES ozone enhanced 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)

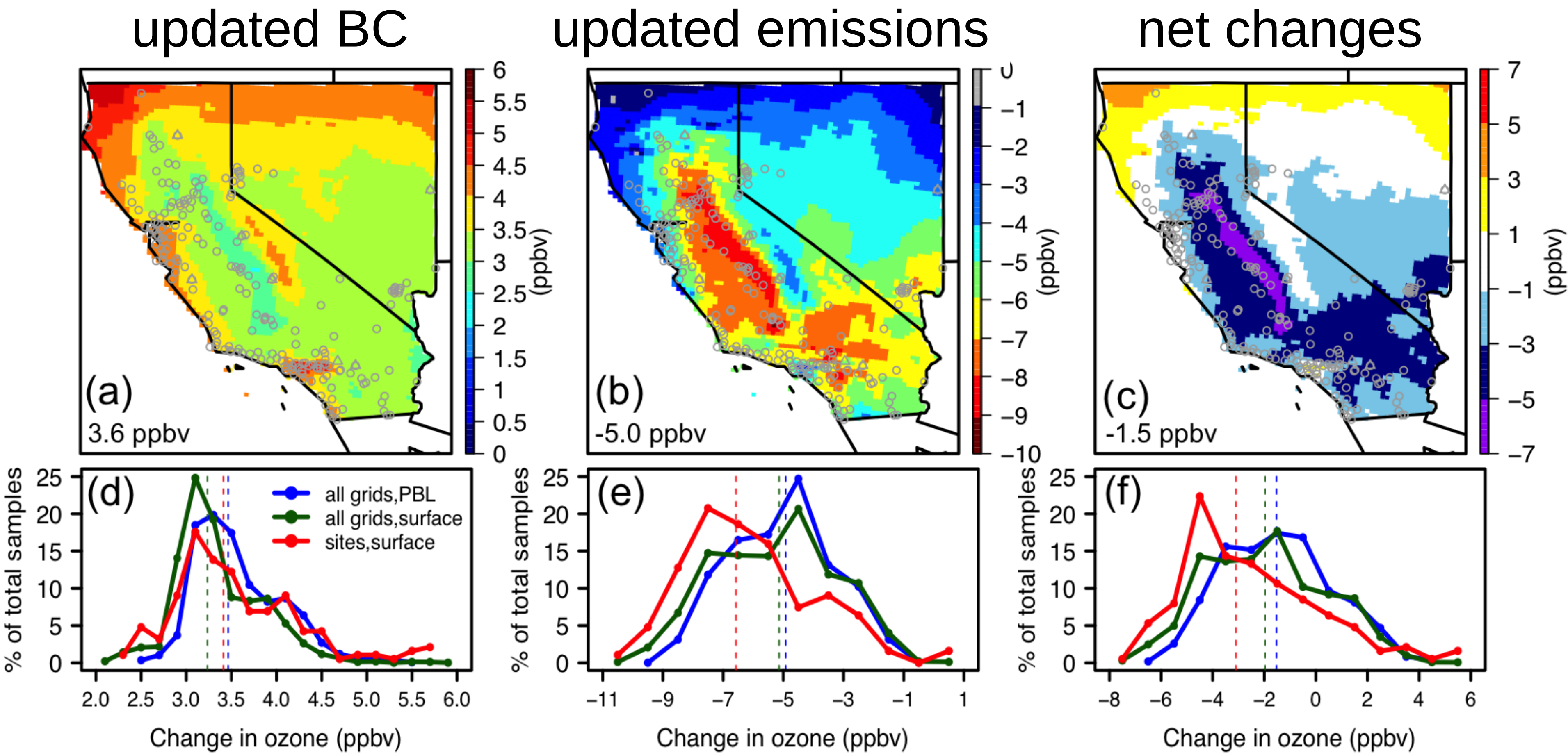
Regional-scale assimilation spatially redistributed local NOx emissions particularly from urban and wildfires



- Anomalously high fire emissions in the Northern California this year
- Original QFED fire emissions overpredicted the NOx emissions, CO emissions less worse→ perhaps the emission factor (from Andreae and Merlet 2001) issue
- Variable reductions in urban areas due to the emission controls during 2005-2008 and the uncertainties of NEI 2005 relative to the base year of 2005

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

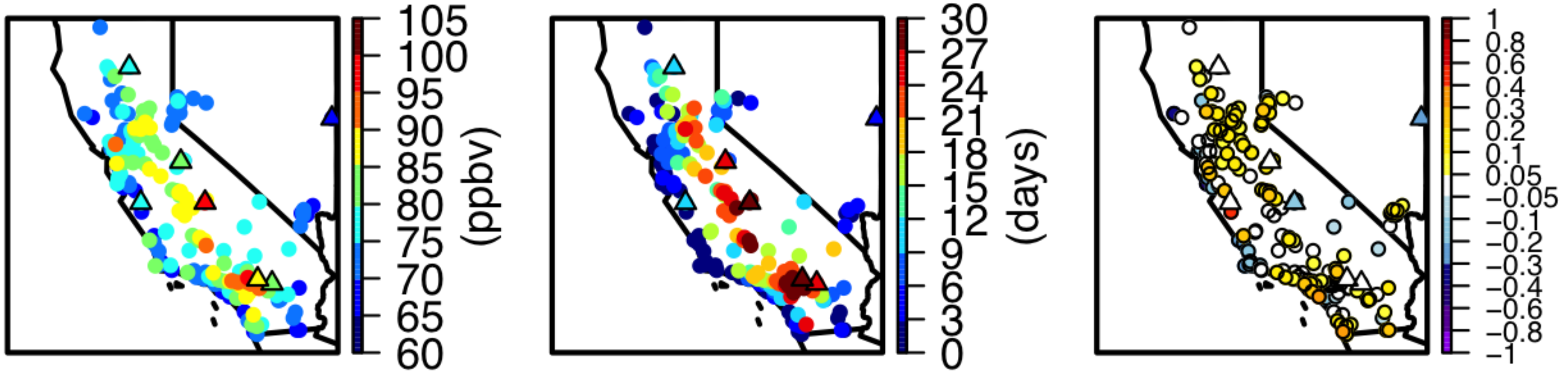


- High terrain regions: more sensitive to extra-regional sources
- Central Valley and SoCal: more sensitive to local emissions
- Monitoring sites unable to capture some strong changes due to the assimilation

Impact of the multi-scale assimilation on STEM ozone: Evaluation using "Fractional Bias (unitless)": 2x(model-obs)/(model+obs)

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

days/sites exceeding >65 ppbv

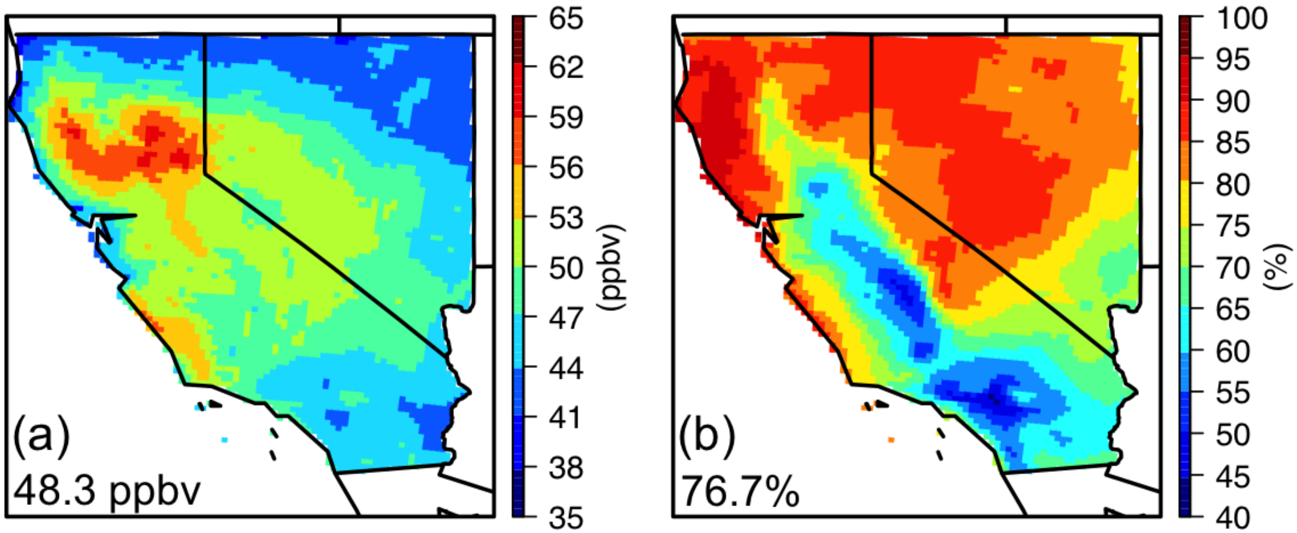


- Improvement occurred after constraining boundary conditions and emissions with satellite observations, despite the remaining positive biases

- Different magnitudes of improvement:

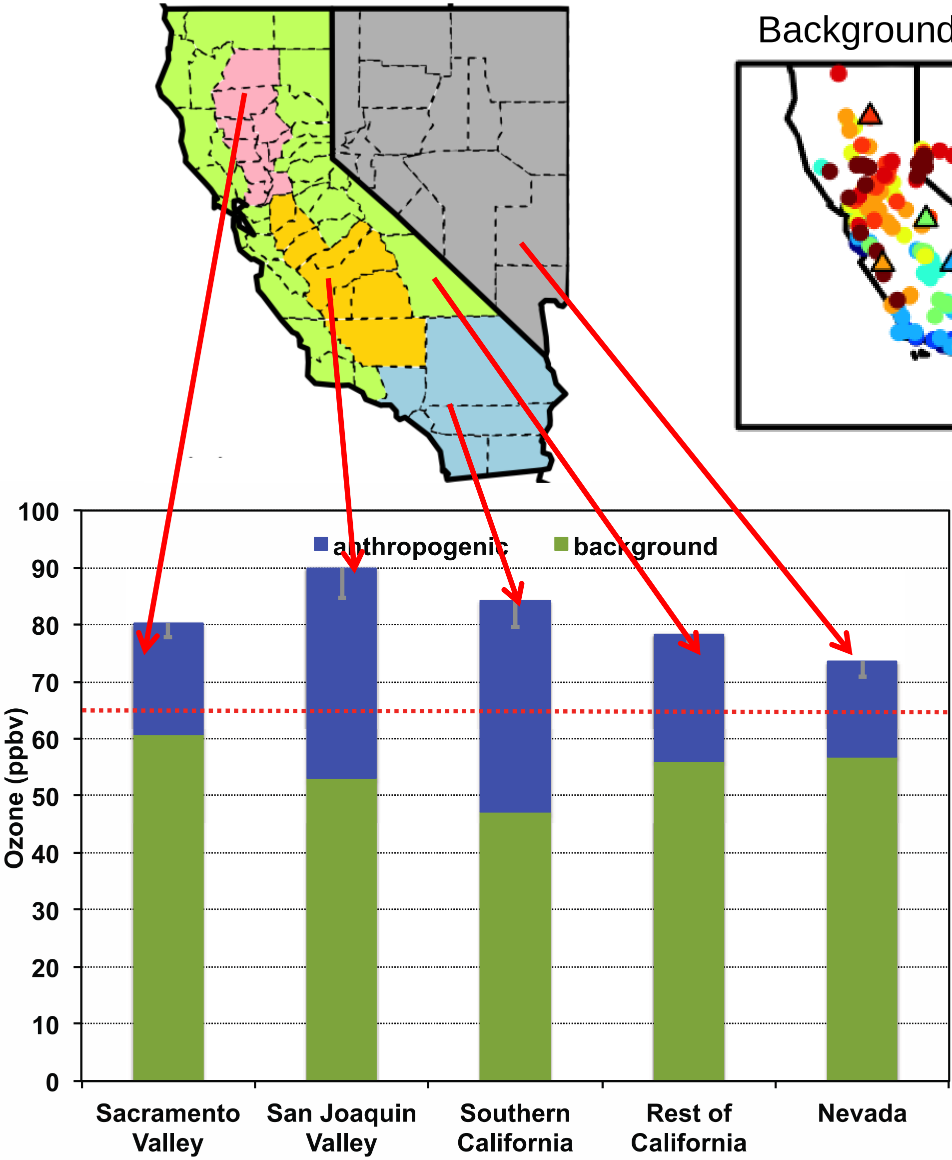
- 0.15 → 0.10 (33%) for days/sites exceeding 65 ppbv
- 0.11 → 0.09 (18%) for days/sites exceeding 75 ppbv
- 0.22 → 0.26 (15%) for all days/sites
- 0.05 → 0.04 (20%) along aircraft <2 km a.g.l.

Monthly-mean surface background ozone (MDA8)

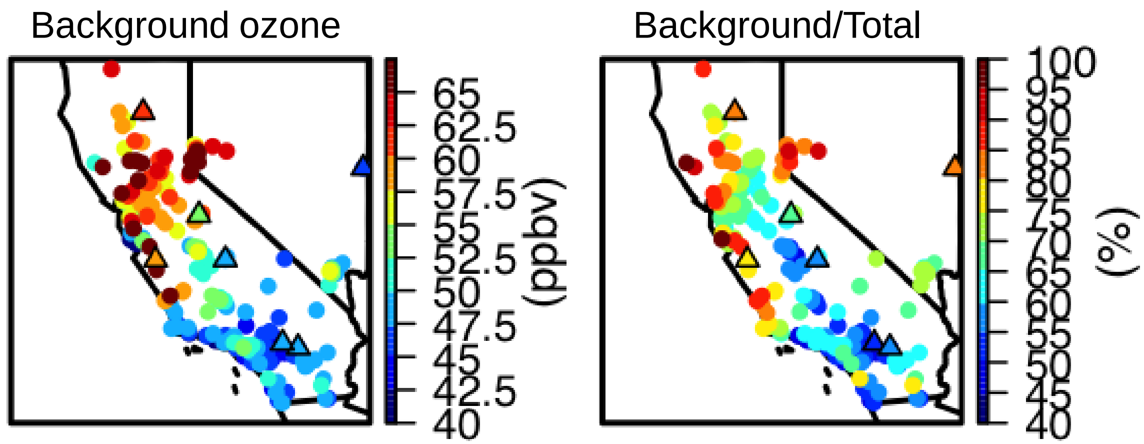


- Assimilation repartitioned background (as well as total) ozone: Compared to a priori: 50.7 ppbv (+3.3 ppbv from boundary conditions; -5.7 ppbv from local emissions)

Background ozone contributing to exceedances (>65 ppbv) at surface sites



Computed also for other levels of exceedances



- Highest background ozone in Sacramento Valley, <10 ppbv below the current and several proposed primary standards 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 thresholds.

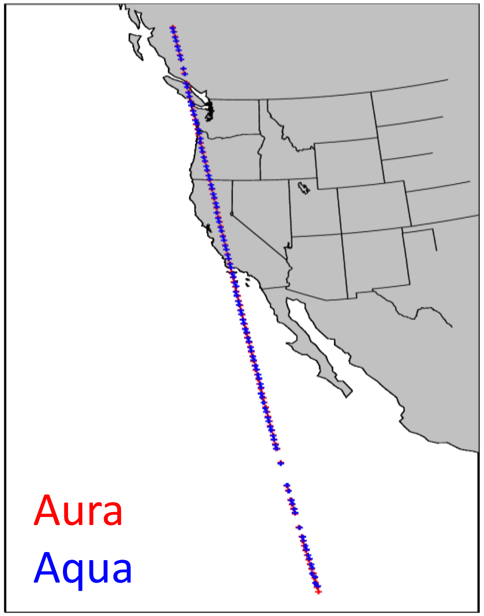
Take-home messages

- Huang et al. (2010): boundary conditions derived from aircraft vertical profiles of pollutants in the eastern Pacific can improve total and transported background ozone in downwind areas–useful during events

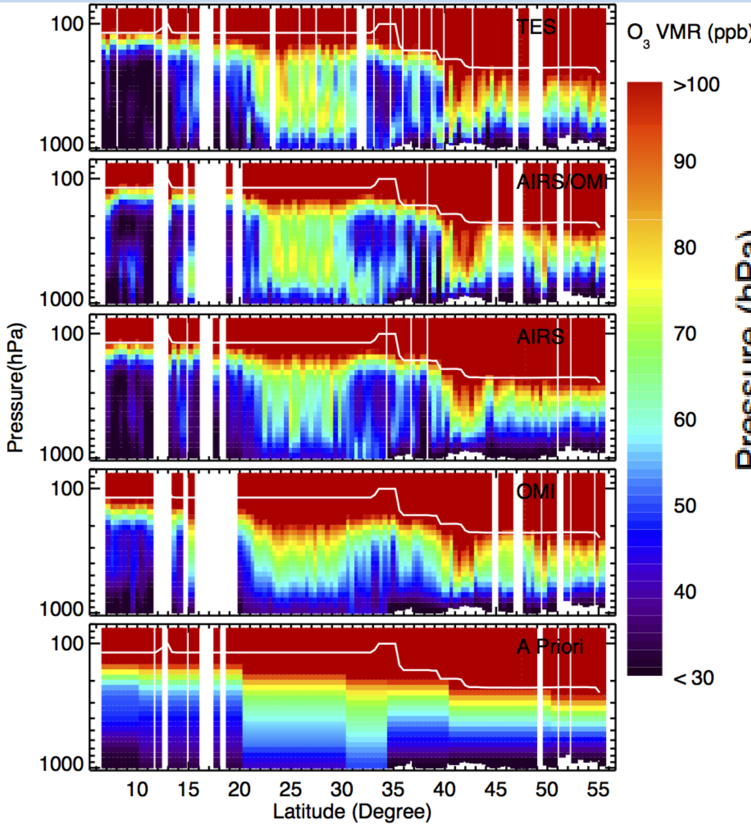
- Huang et al. (2015): boundary conditions from TES-constrained GEOS-Chem, together with the OMI-constrained US emissions improved total and background ozone–adaptable to other regions/times

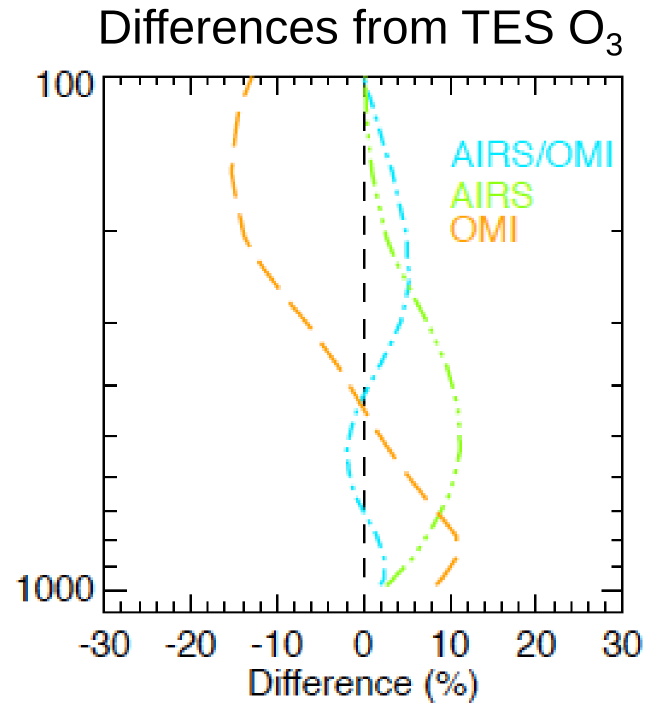
- Future directions: using single and multi-spectral satellite products from other/newer instruments for studying other locations and times

Using multi-spectral ozone product (e.g., OMI/AIRS; CrIS/OMPS) for model evaluation and assimilation

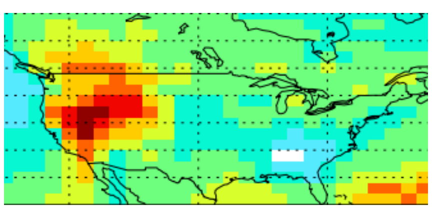


D. Fu et al.

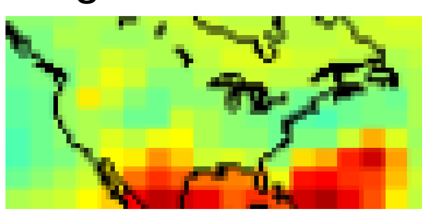




Ozone differences due to assimilating TES (L) and OMI/AIRS (R) ozone into GEOS-Chem, Aug 2006



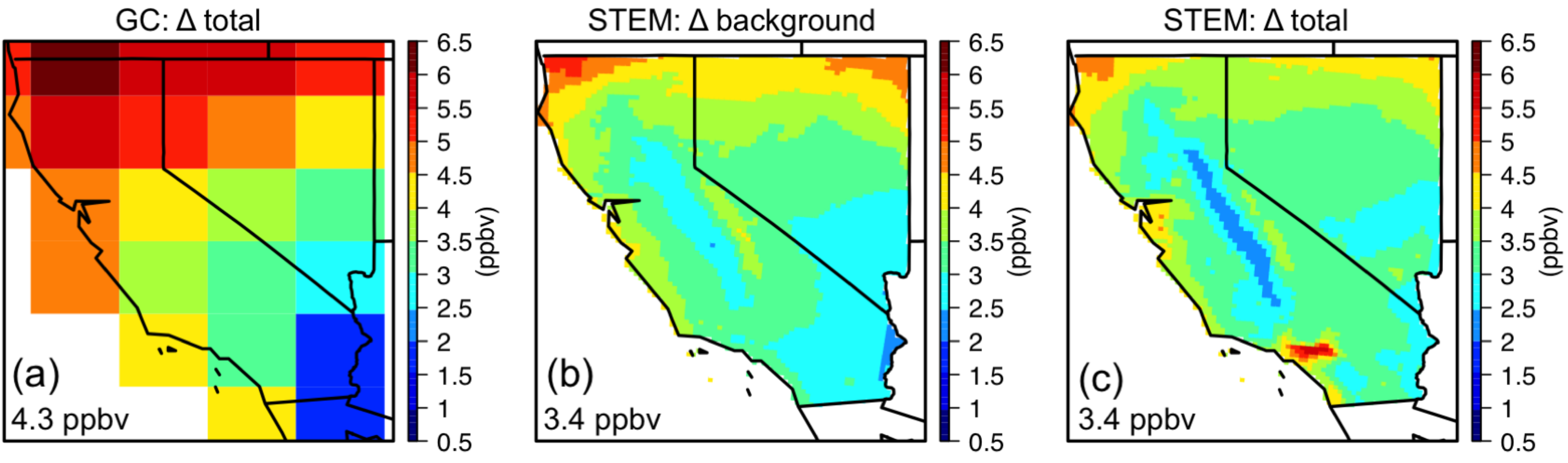
M. Parrington et al.



T. Walker et al.

- from TES ozone to OMI/AIRS, and CrIS/OMPS
- NO_2 from OMPS, TROPOMI, GOME-2
- from California to other states/periods: TexAQS (Aug 2006); DISCOVER-AQ and SEAC4RS (Aug-Sep 2013)

Differences due to assimilation in global and regional models



GEOS-Chem surface ozone enhancements are ~1 ppbv > STEM's surface background and total ozone responses to the updated GEOS-Chem boundary conditions: different model terrain and transport in part due to the resolution

The changes in STEM surface total and background ozone are different in spatial distributions despite their similar domain-wide mean values: reflecting the non-linear chemical coupling between trans-boundary and locally produced ozone.