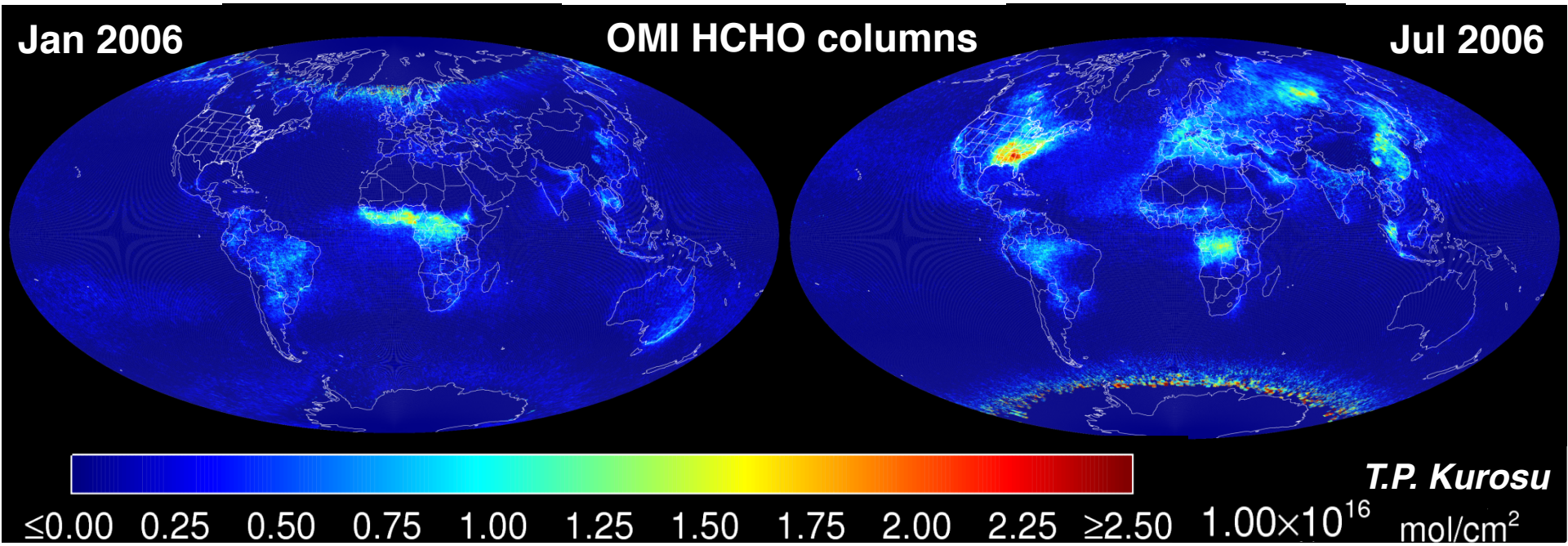
Overview of Southeast U. S. Chemistry: Goals, Types of Flights, DISCOVER-AQ

Daniel J. Jacob (Harvard) and the Southeast US Planning Team

Southeast in summer:
world hotspot for emissions of biogenic volatile organic compounds (BVOCs)



Central questions for Southeast US chemistry

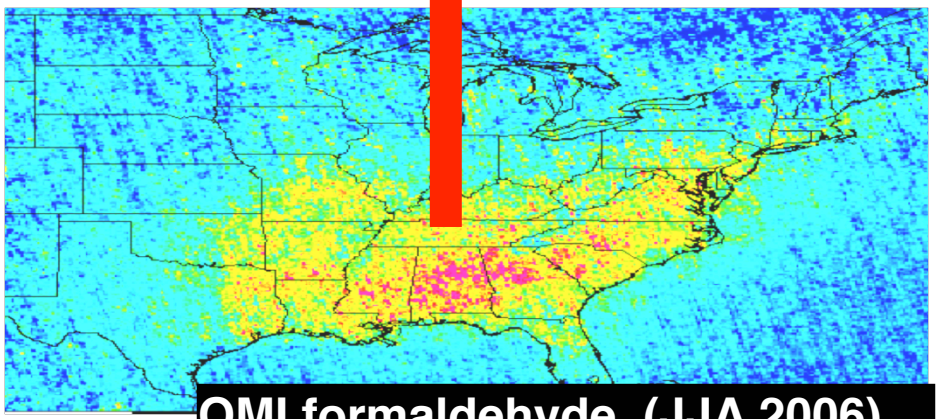
What is the delivery of H_2O and other gases to the lower stratosphere by deep convection, and what are the implications for ozone loss?

tropopause

UT anticyclone

How does aging of convective outflow in the North American UT anticyclone contribute to ozone and aerosol formation?

How efficiently are gases and aerosols scavenged in deep convection?



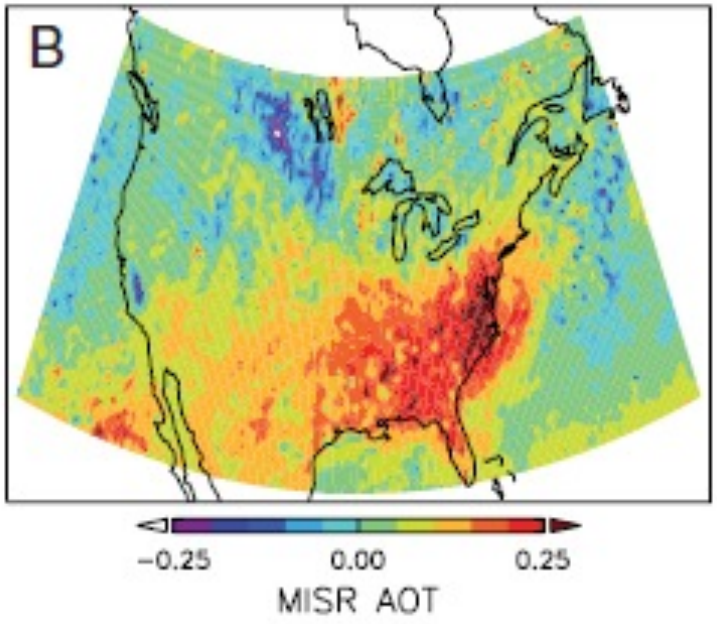
OMI formaldehyde (JJA 2006)

How does human influence affect BVOC chemistry with implications for organic aerosols and ozone?

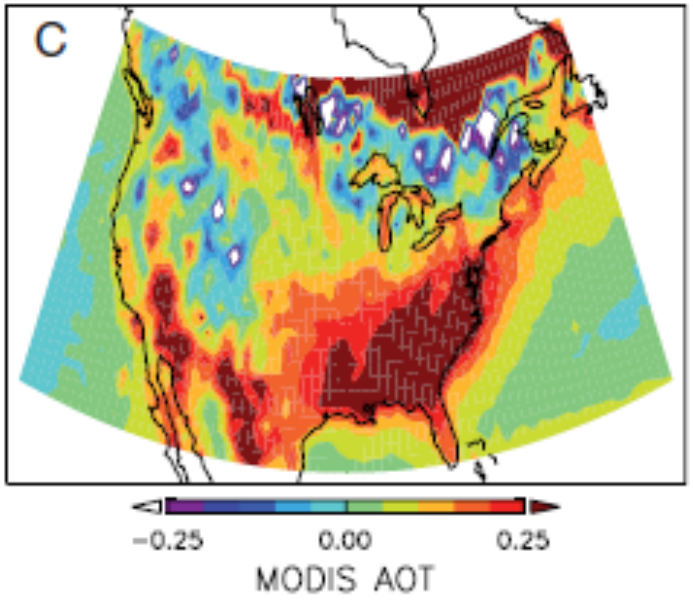
Southeast features a sharp aerosol maximum in summer

Seasonal difference between JJA and DJF aerosol optical thickness (2000-2006)

MISR



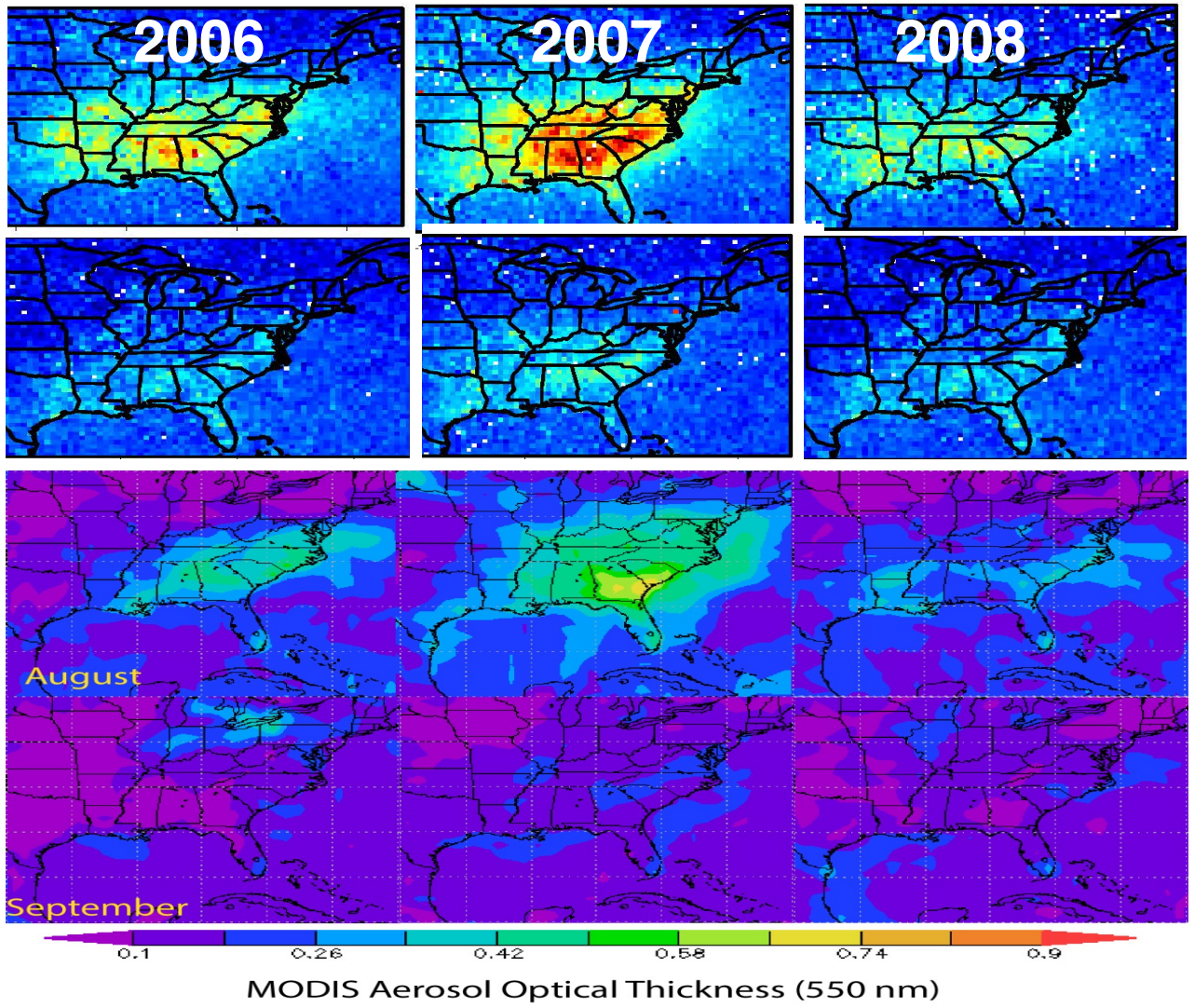
MODIS



Evidence of a large source of oragnic aerosol from BVOCs;
how is this source modulated by anthropogenic pollution (NO_x, aerosol)?

Goldstein et al. [2009]

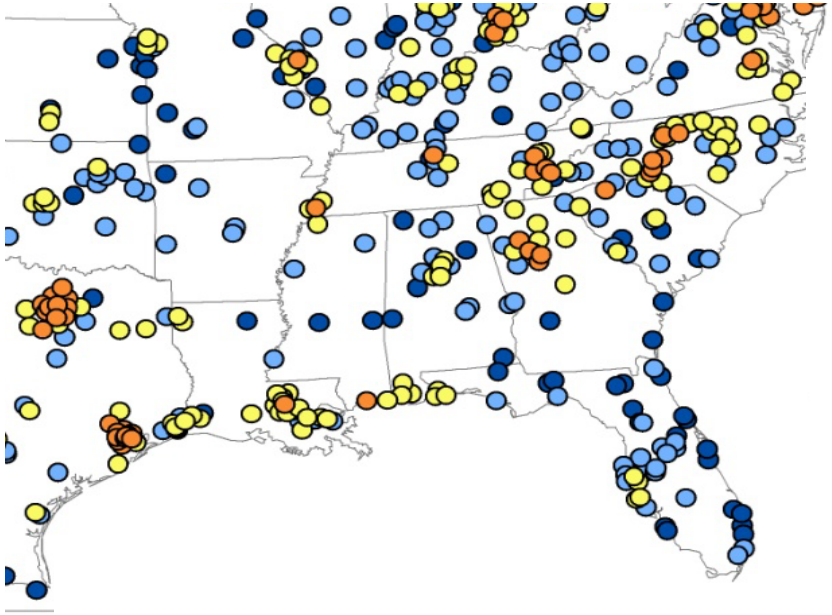
Southeast aerosol has sharp Aug-Sep transition correlated with BVOC emissions



- Seasonal transition in BVOC emissions is mainly driven by temperature
- How do ozone and aerosol formation respond to this transition?

Lei Zhu (Harvard), Jeff Reid (NRL)

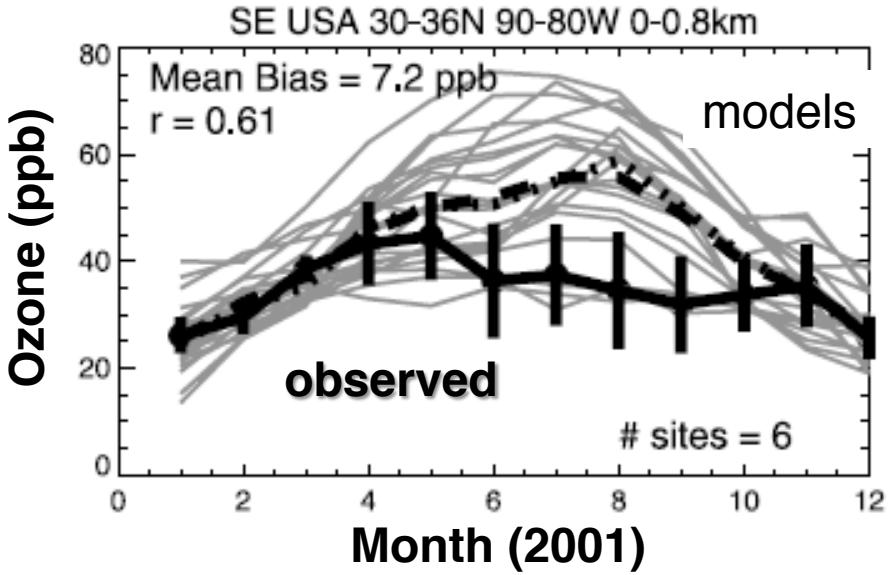
The Southeast also has a major ozone pollution problem…



EPA [2012]

…and models show very large biases in the region

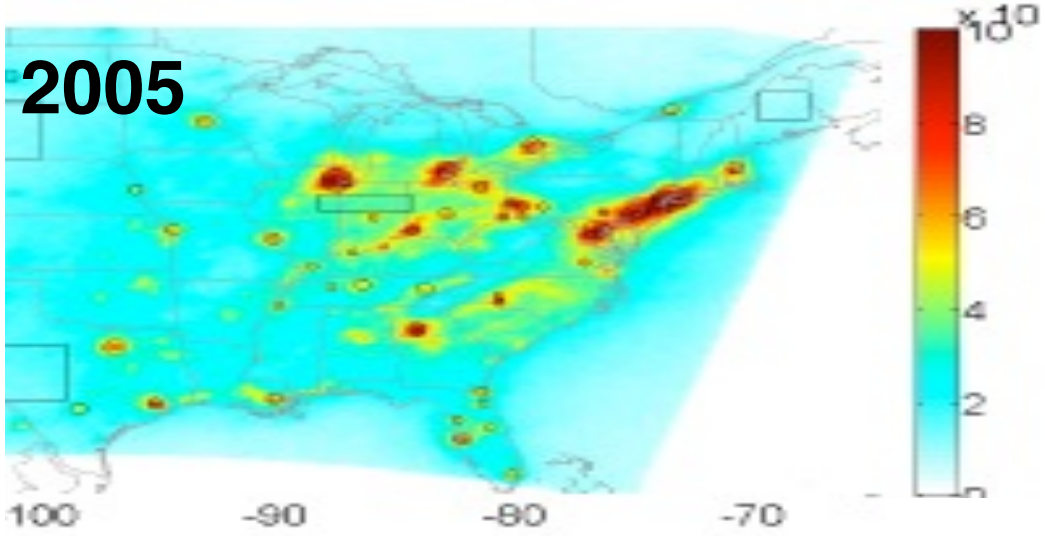
HTAP model intercomparison, 24-h mean ozone [Fiore et al., 2009]

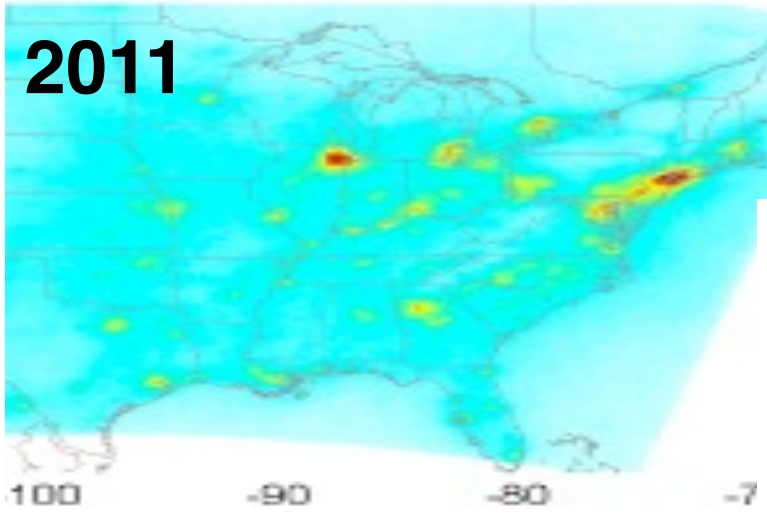


- **Uncertain VOC chemistry?**
- **Overestimate of ozone in Gulf of Mexico inflow?**

NO_x emissions have been decreasing over past decade

OMI tropospheric NO_2, Apr-Sep:
30% decrease over 2005-2011





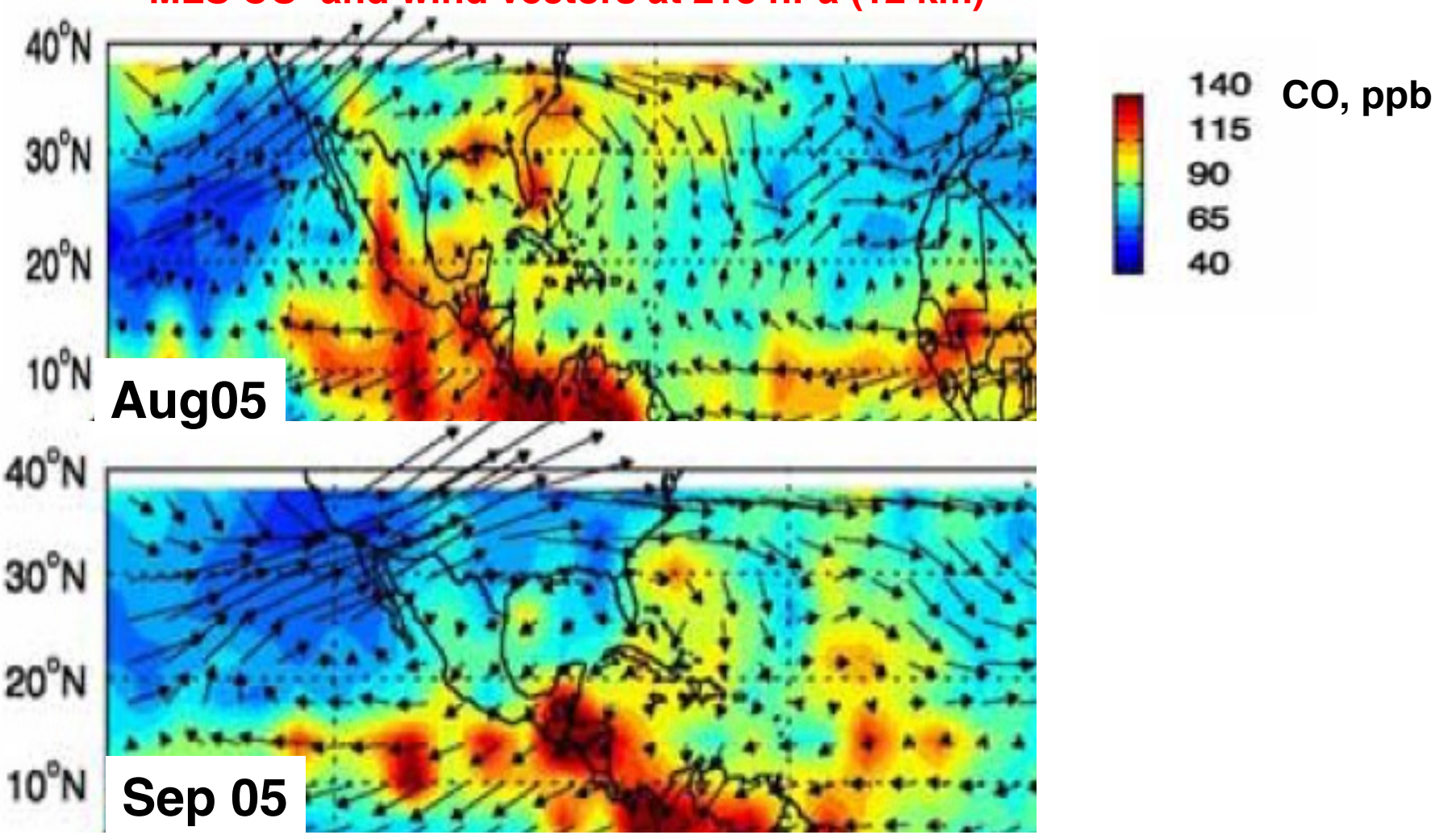
We now have the opportunity to sample both high-NO_x and low-NO_x regimes over the Southeast

- **How is ozone response to decreasing NO_x modulated by BVOC chemistry?**
- **What is the effect of decreasing NO_x on organic aerosol yields?**

Russell et al. [2012]

Deep convection over Southeast and trapping in UT anticyclone

MLS CO and wind vectors at 215 hPa (12 km)

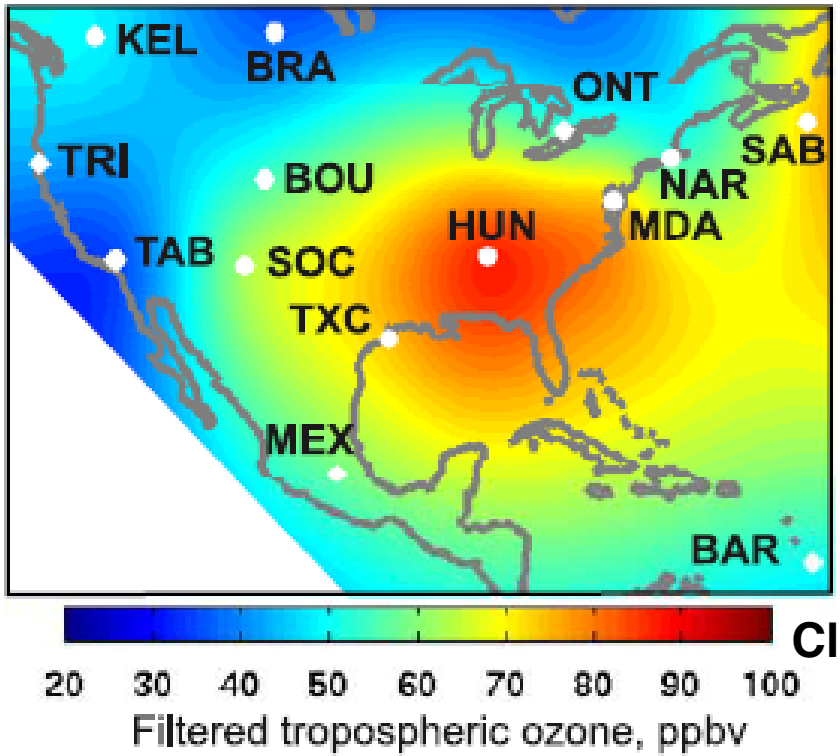


Frequent deep convection and effective trapping over Southeast US in August; sharp transition to less convection and no trapping in September

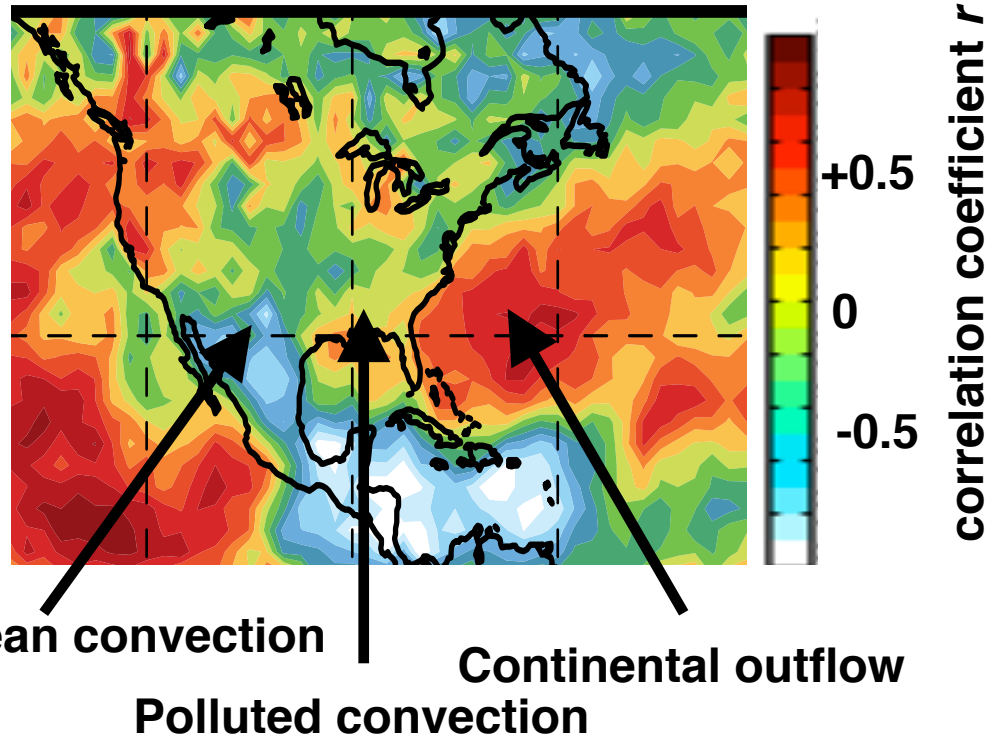
Liu et al. [2013]

UT ozone maximum over Southeast in summer

Tropospheric ozone at 10-11 km from IONS ozonesondes, Aug 2006



Ozone-CO correlations at 700-350 hPa from OMI+AIRS, JJA 2008

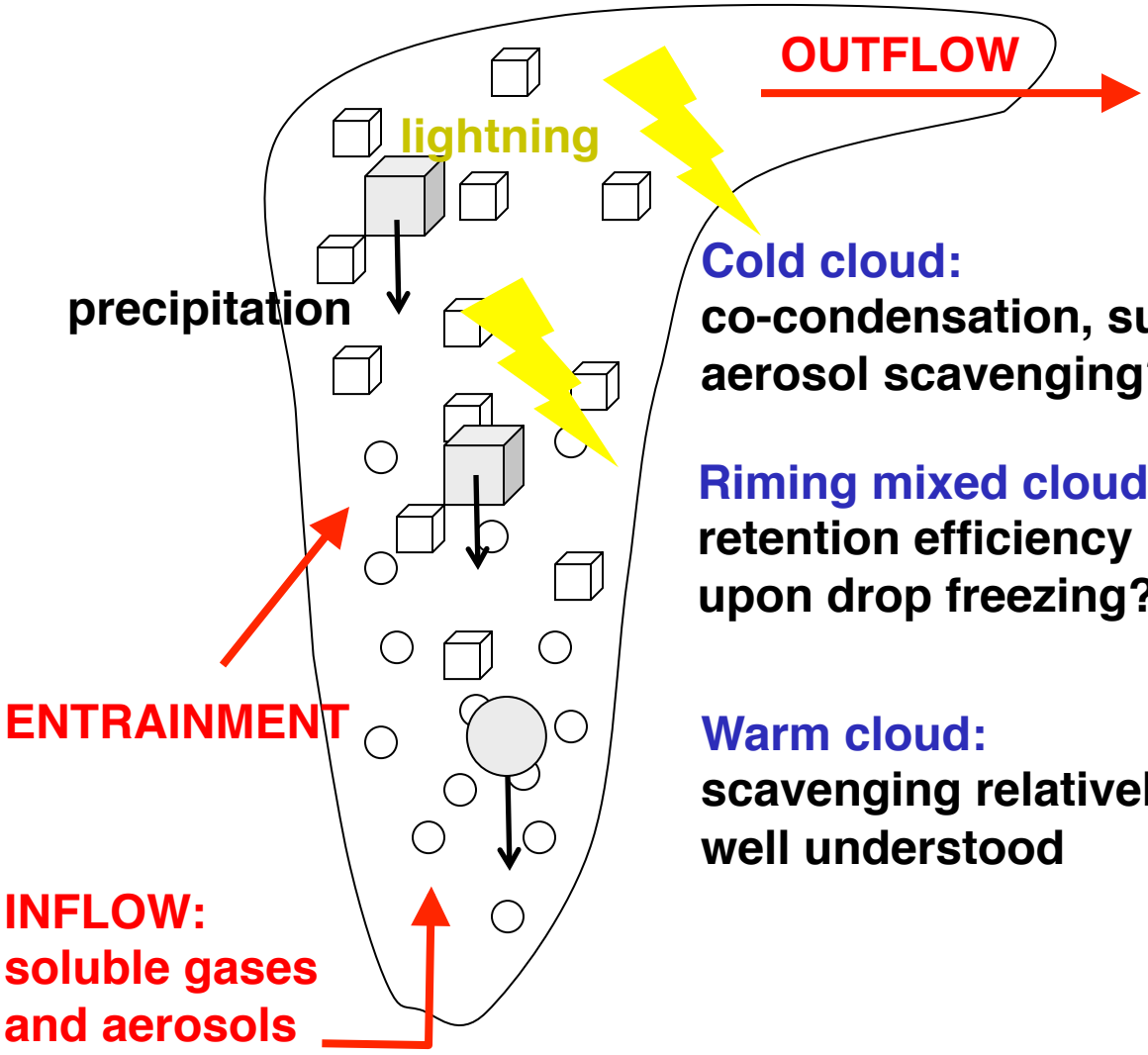


Can we relate the UT ozone maximum to deep convective injection of precursors?

Cooper et al. [2008]; Kim et al. [2013]

Variable gas/aerosol scavenging efficiencies in deep convection

What was learned in DC3 and how can we build on that?

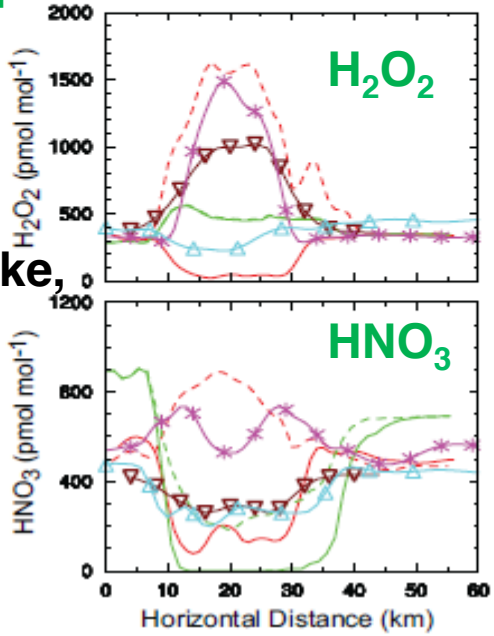


Cold cloud:
co-condensation, surface uptake, aerosol scavenging?

Riming mixed cloud:
retention efficiency upon drop freezing?

Warm cloud:
scavenging relatively well understood

Model intercomparison deep convective outflow

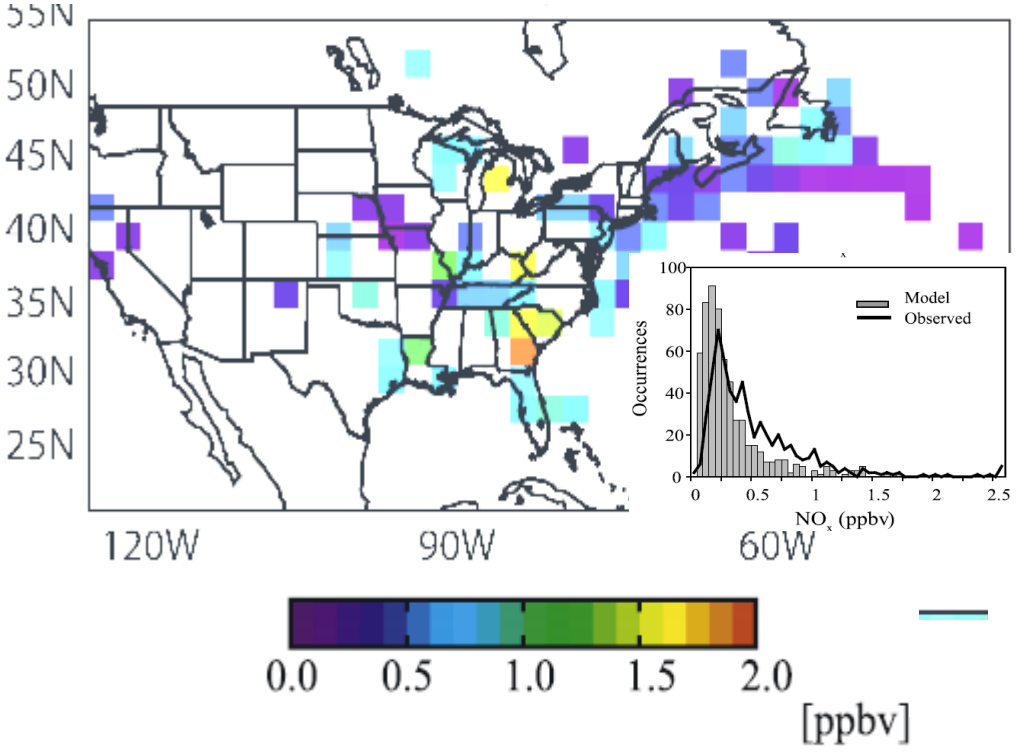


Barth et al. [2007]

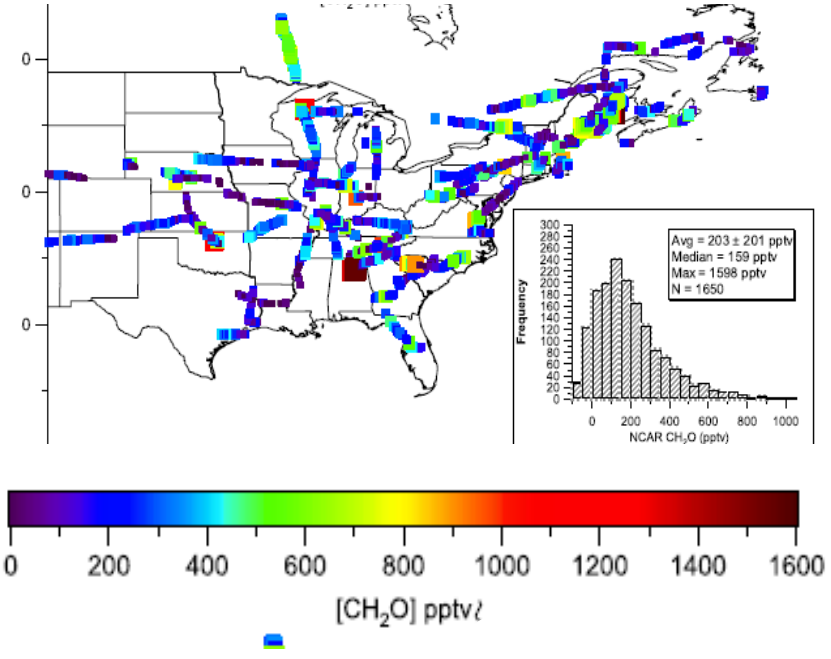
Deep convective injection of photochemical precursors over Southeast

INTEX-A aircraft observations (Jul-Aug 2004)

NO_x (> 8 km)



Formaldehyde (>6 km)

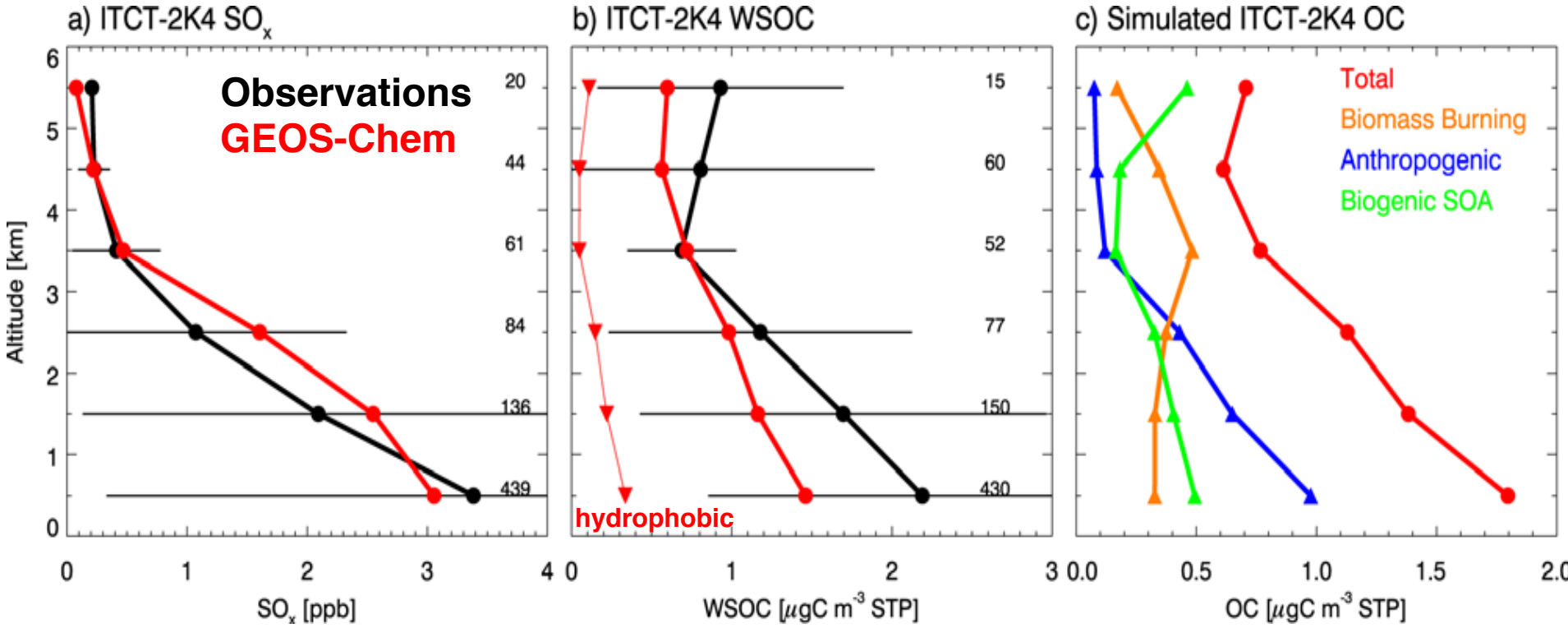


- Models underestimate NO_x in UT (lightning, aging?), overestimates formaldehyde (scavenging?)
- Quantitative link to UT ozone maximum still needs to be made

Hudman et al. [2007], Fried et al. [2008]

Source of secondary organic aerosol from convected precursors?

Water-soluble organic aerosol (WSOC) on NOAA plane during ICARTT

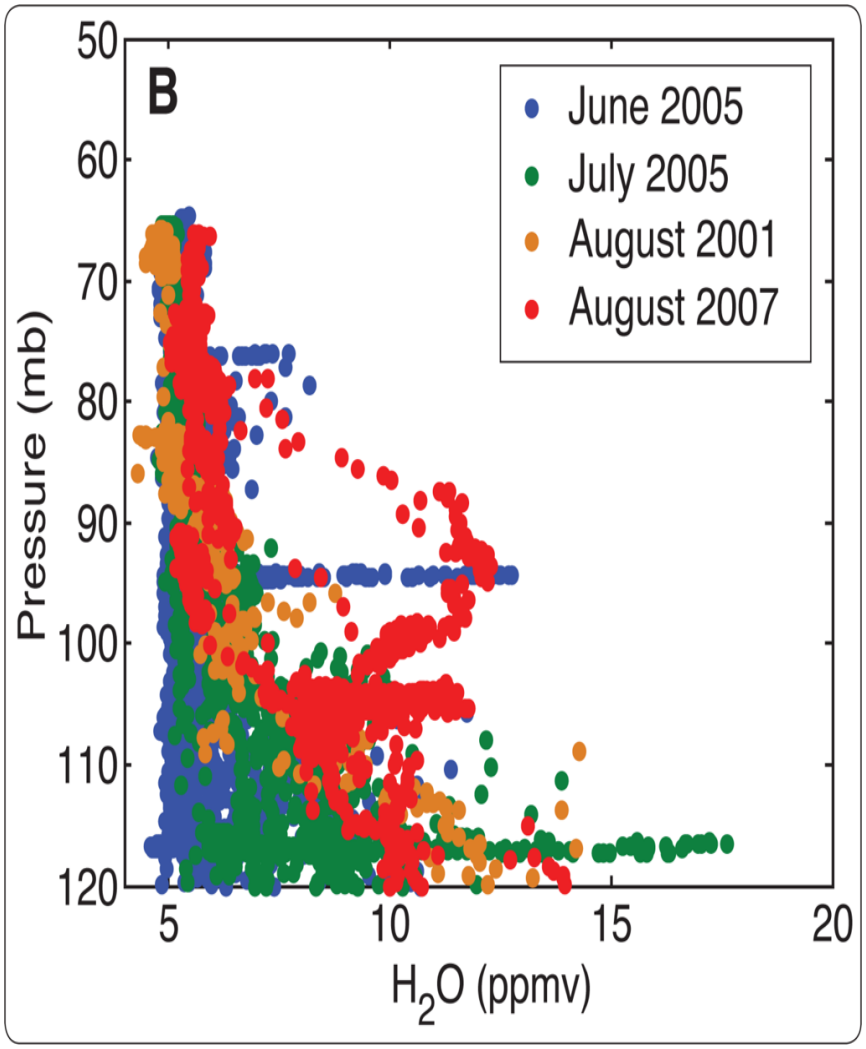


Vertical gradient much weaker for WSOC than for sulfate: implies either less effective scavenging or sources aloft

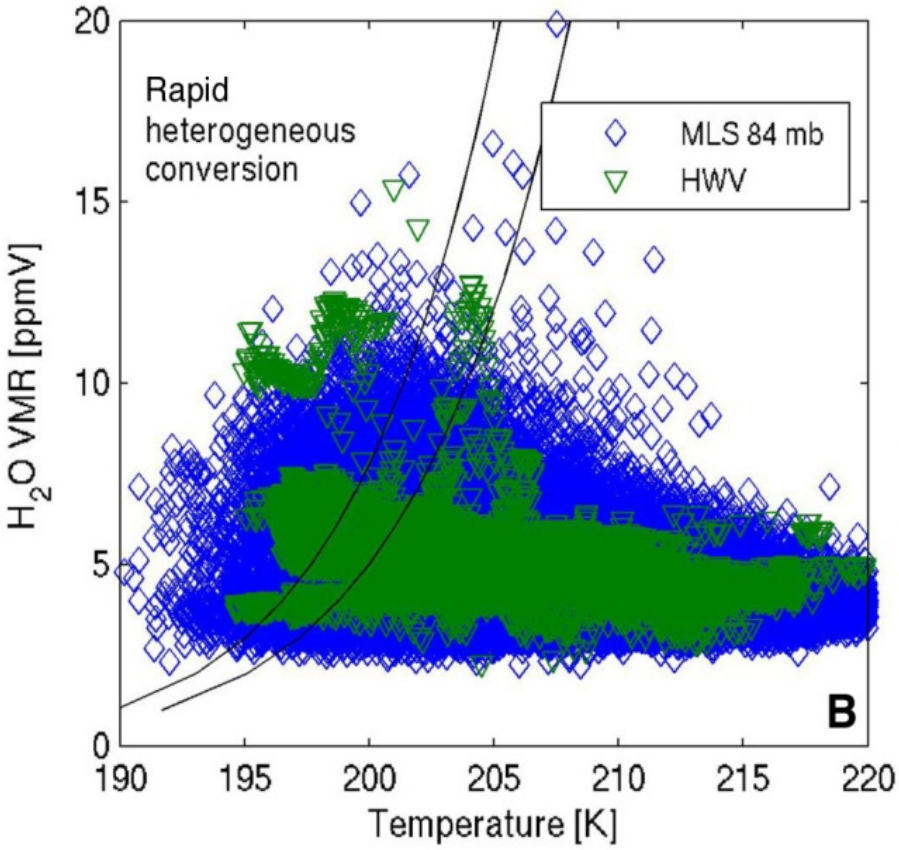
Heald et al. [2006]

H_2O injection into lower stratosphere over US in summer

ER-2 vertical profiles, S-Central US



MLS H_2O vs. *T* at 84 hPa



Frequent conditions for PSC-like chemistry

How does injection of other deep convective gases (NO_x, formaldehyde…) affect the chemistry for ozone loss?

Anderson et al. [2012]

A platter of scientific questions for Southeast US chemistry

- **Boundary layer chemistry**
  - **How well can satellites quantitatively constrain VOC and NO_x emissions?**
  - **How do anthropogenic NO_x and aerosol modulate organic aerosol formation from BVOCs?**
  - **How does BVOC chemistry modulate the response of ozone to NO_x controls?**

- **Deep convective transport and scavenging**
  - **What are the scavenging efficiencies of aerosols and gases in deep convection?**
  - **How do these relate to cloud microphysics?**
  - **Do NO_x and VOC oxidation products modulate the chemical effect of water vapor injected into the lower stratosphere?**

- **Aging of deep convective outflow**
  - **Can the UT ozone maximum over the Southeast US be explained on the basis of chemical production from convected precursors?**
  - **Do we understand the NO_x chemistry in UT convective outflow, aged air?**
  - **Is there a significant organic aerosol source in the free troposphere from convection of precursors?**

Implications for flight plans

1. Boundary layer sampling

- Long boundary layer transects along chemical gradients under clear skies (fair-weather Cu OK), hot/cool cases, daytime
- One transect from Gulf of Mexico to inland under inflow conditions
- Porpoise vertical profiles to observe shallow convection, cloud processing
- Deep vertical profiles to link with remotely sensed data: MetOp (09:30 LT), Aura (13:30 LT), AERONET, Huntsville (ozone lidar)
- Coordinate with fire objective

2. Sampling of mature deep convective systems

- Learn from DC3 experience
- Sample inflow, outflow, near-field chemical/aerosol evolution
- Sample deepest systems with ER-2
- Coordinate with cloud objectives; requirements should be compatible

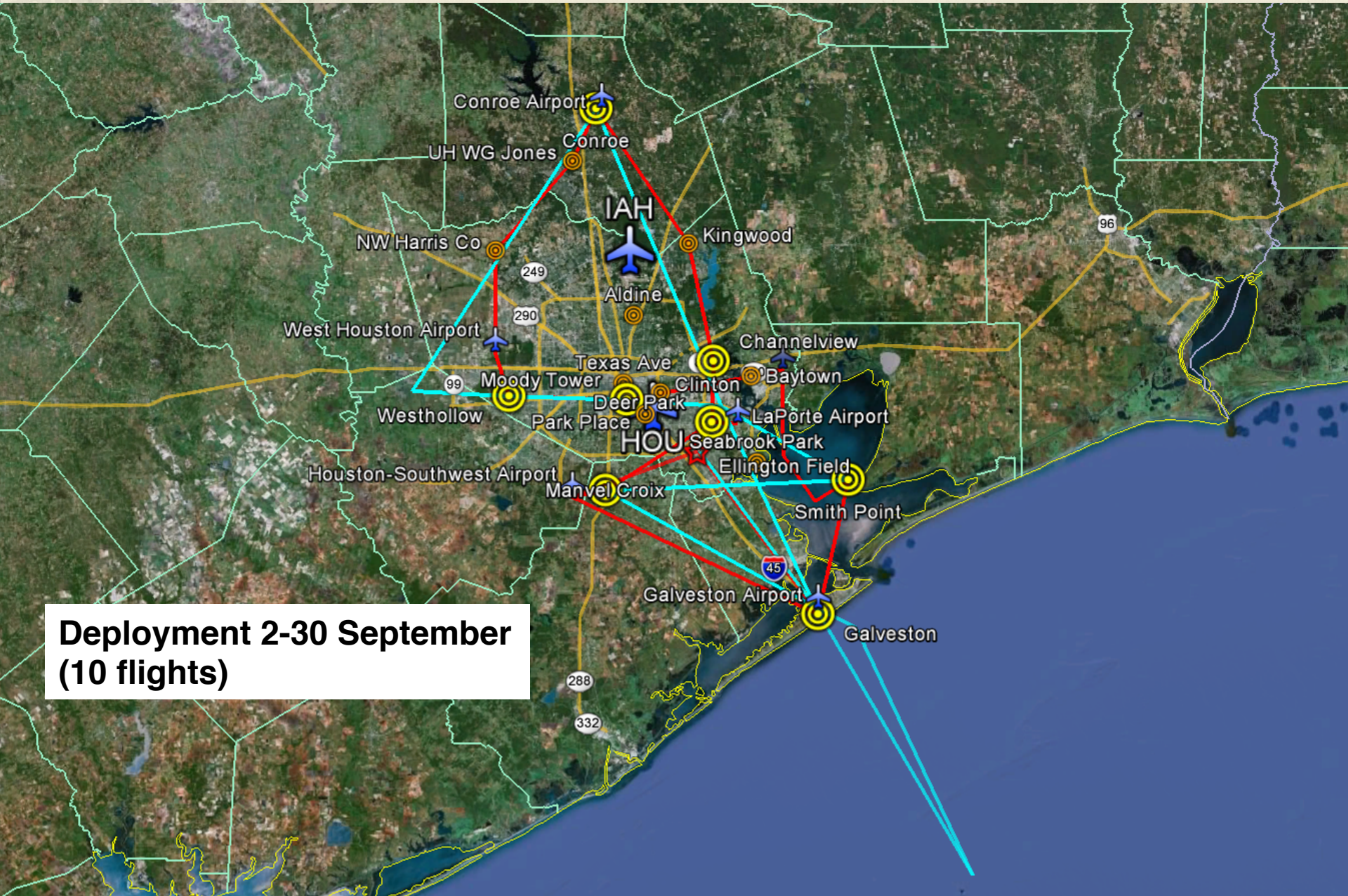
3. Chemical/aerosol evolution in aged deep convective outflow

- Sample convective outflow on successive days.
- Survey UT anticyclone
- Coordinate with N America monsoon objective

- August is generally best! – Avoid delay at front end of mission
- Observing Aug-Sep transition is a great opportunity but only needs a few flights in September

DISCOVER-AQ

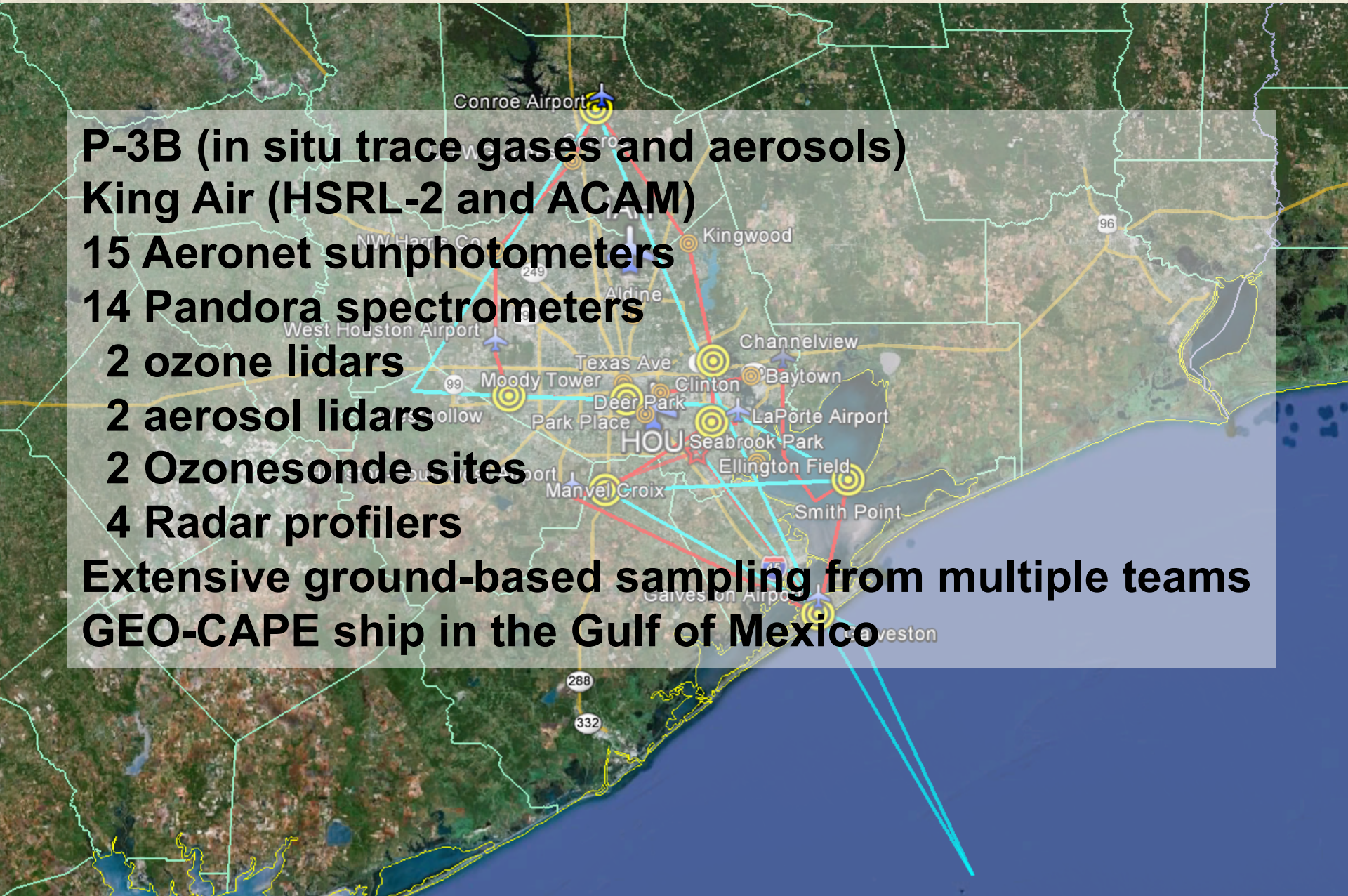
Observation Domain



Deployment 2-30 September (10 flights)

DISCOVER-AQ

Observational Assets

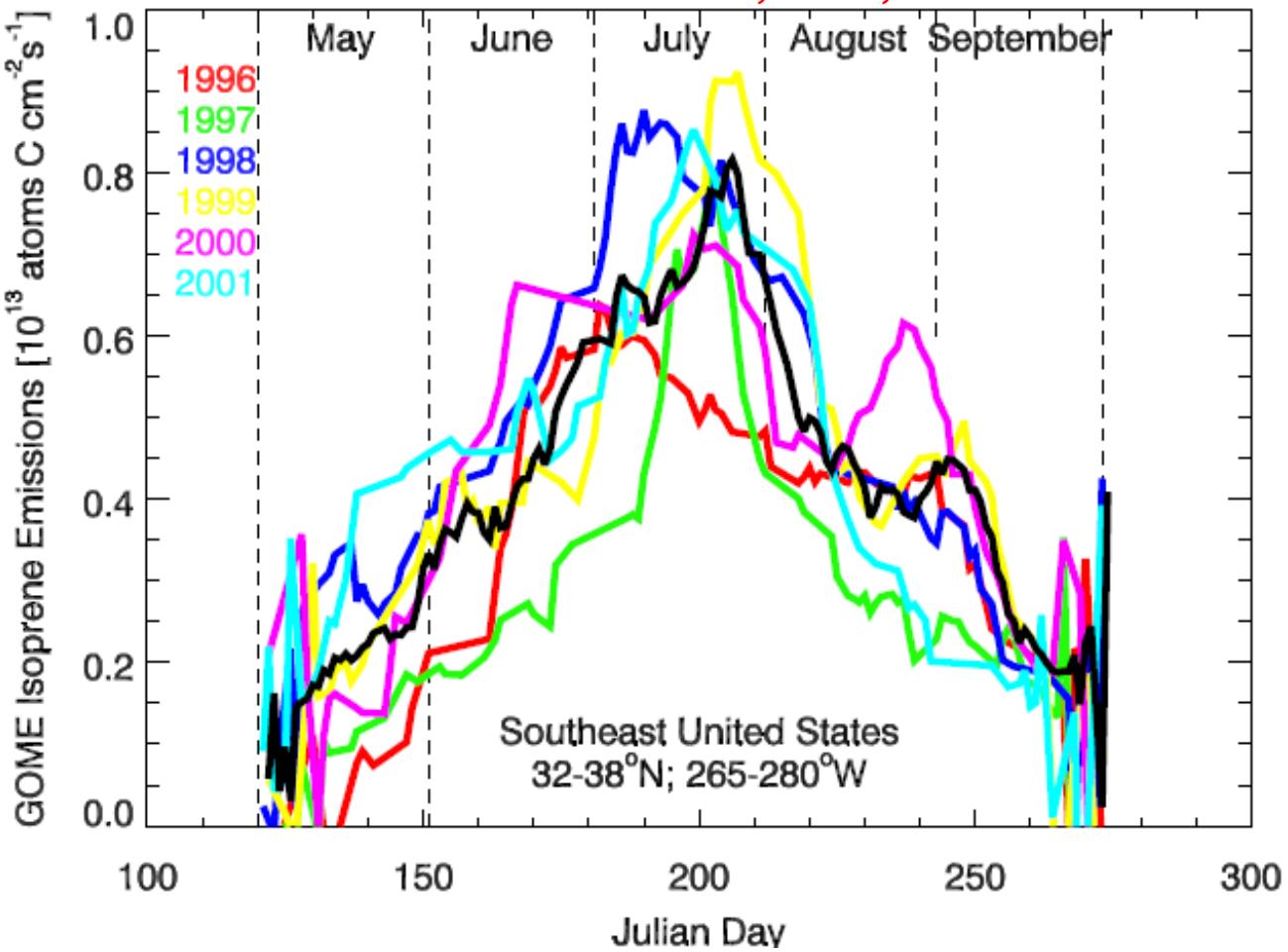


P-3B (in situ trace gases and aerosols)
King Air (HSRL-2 and ACAM)
15 Aeronet sunphotometers
14 Pandora spectrometers
  2 ozone lidars
  2 aerosol lidars
  2 Ozonesonde sites
  4 Radar profilers
Extensive ground-based sampling from multiple teams
GEO-CAPE ship in the Gulf of Mexico

Extra slides

Finer seasonal structure and interannual variability of seasonal transition of isoprene emission in Southeast

Derived from GOME HCHO, data, 1996-2001

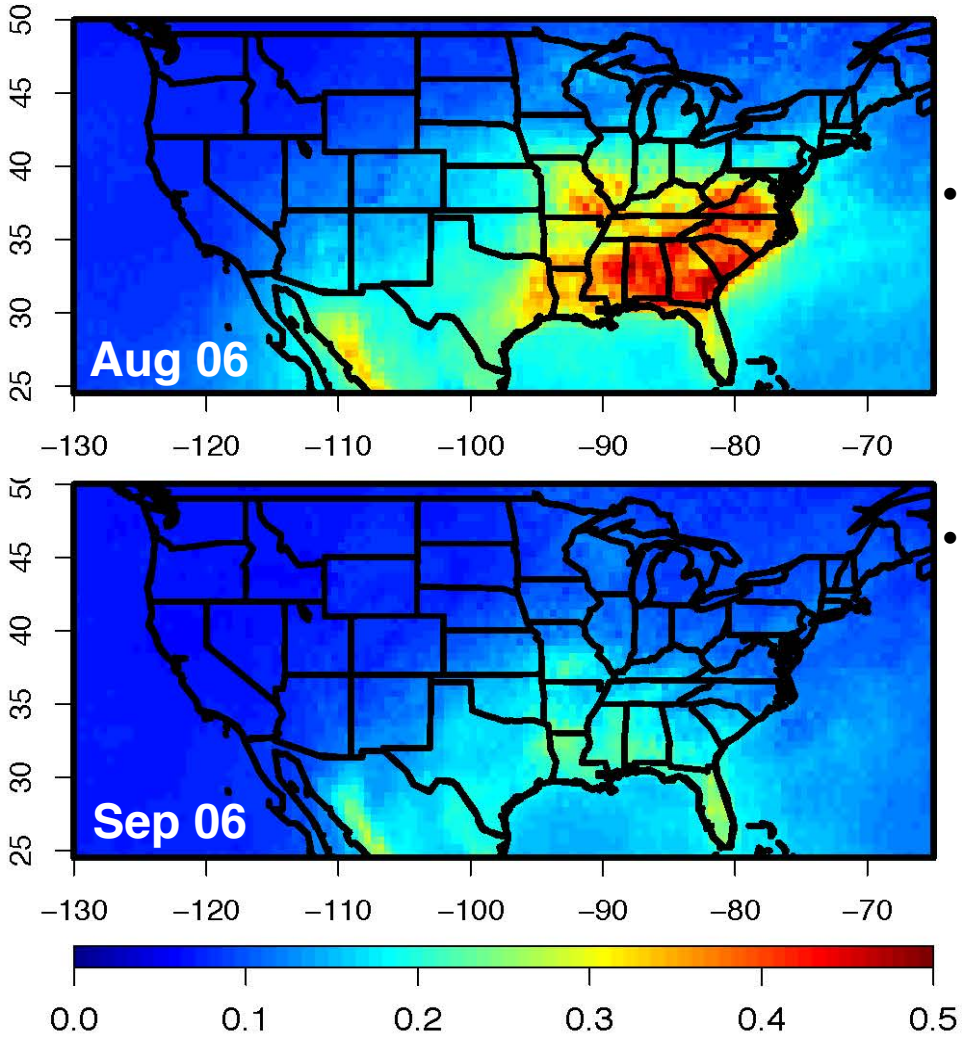


Timing of seasonal transition varies from year to year, depending on temperature

Palmer et al., 2006

Deep convective UT influence over the Southeast US

Mean formaldehyde (ppb) at 314 hPa (GEOS-Chem)



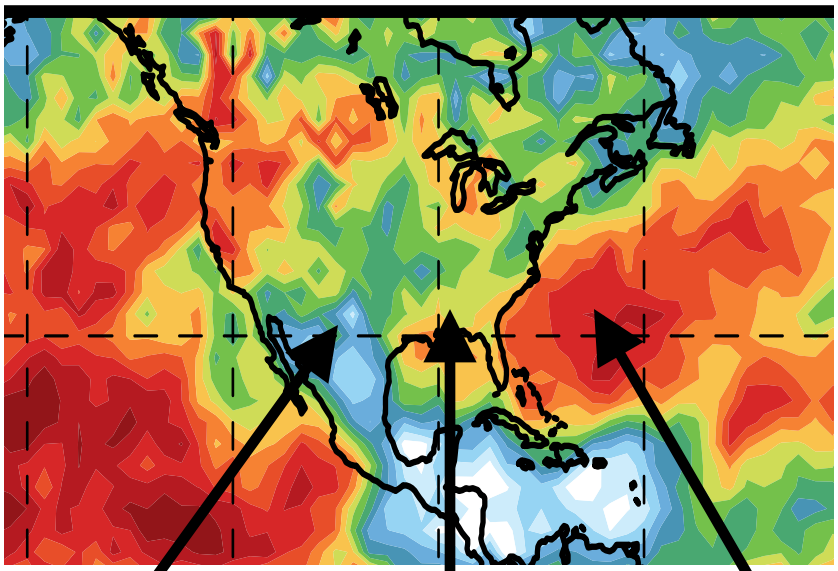
- **Deep convective injection of VOC oxidation products together with anthropogenic and lightning NO_x can drive fast UT photochemistry to produce ozone maximum**
- **Sharp transition in convection between Aug and Sep**

Lei Zhu (Harvard)

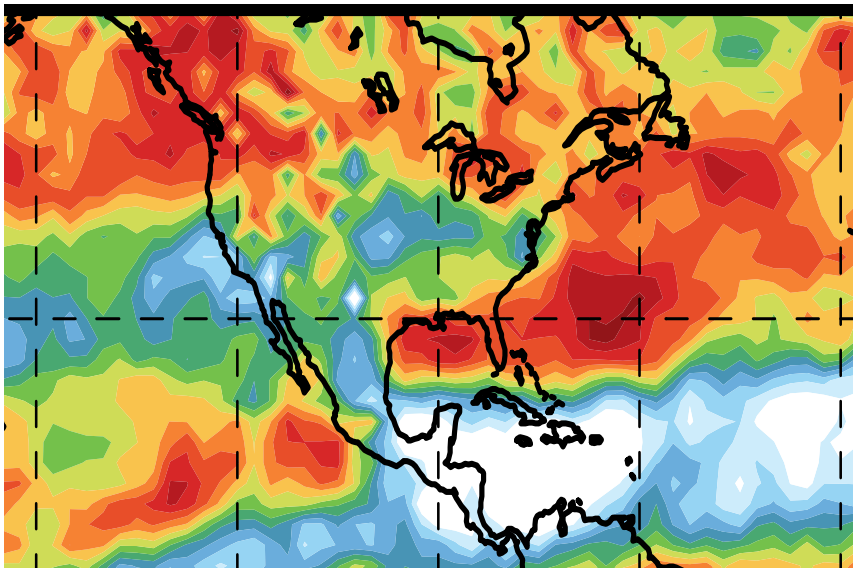
Free tropospheric ozone – CO Correlations from OMI+AIRS

700-350 hPa, JJA 2008

OMI + AIRS



GEOS-Chem



Clean convection

Polluted convection

Continental outflow

Correlations provide constraints on sources of free tropospheric ozone; SEAC^{4}RS will allow to test interpretation

Kim et al., submitted