Fresh constraints on the global reactive nitrogen budget through new TES NH_3 and PAN observations

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NOAA CrIS Workshop
18-19 Sep. 2014
College Park, MD

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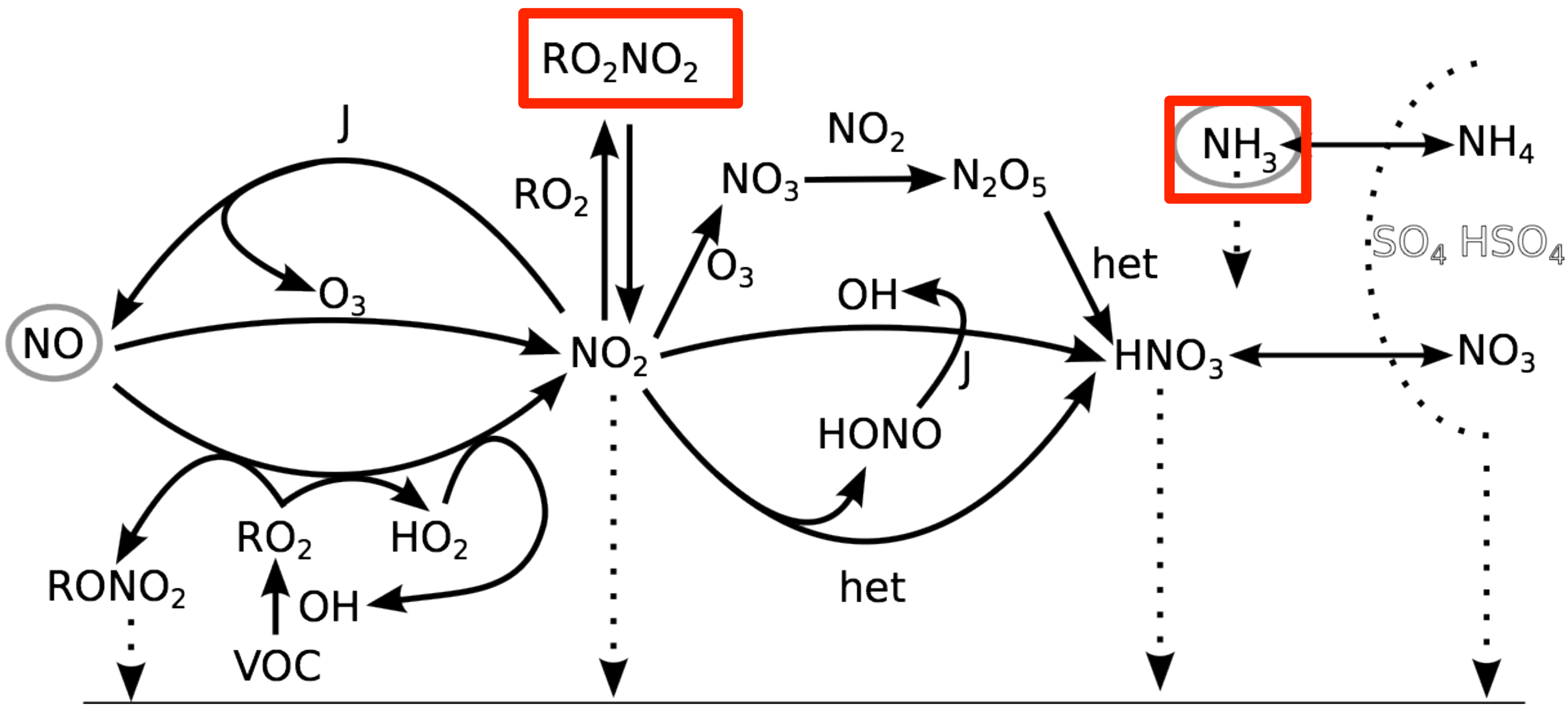
Atmospheric and Climate Application, Inc., CANADA;
M. Luo, NASA JPL, CA.

Funding from NASA and EPA

E.V. Fischer CSU, CO;
V.H. Payne, NASA JPL, CA.

Funding from NASA

NH_3 and PAN play critical roles in atmospheric chemistry.



NH_3

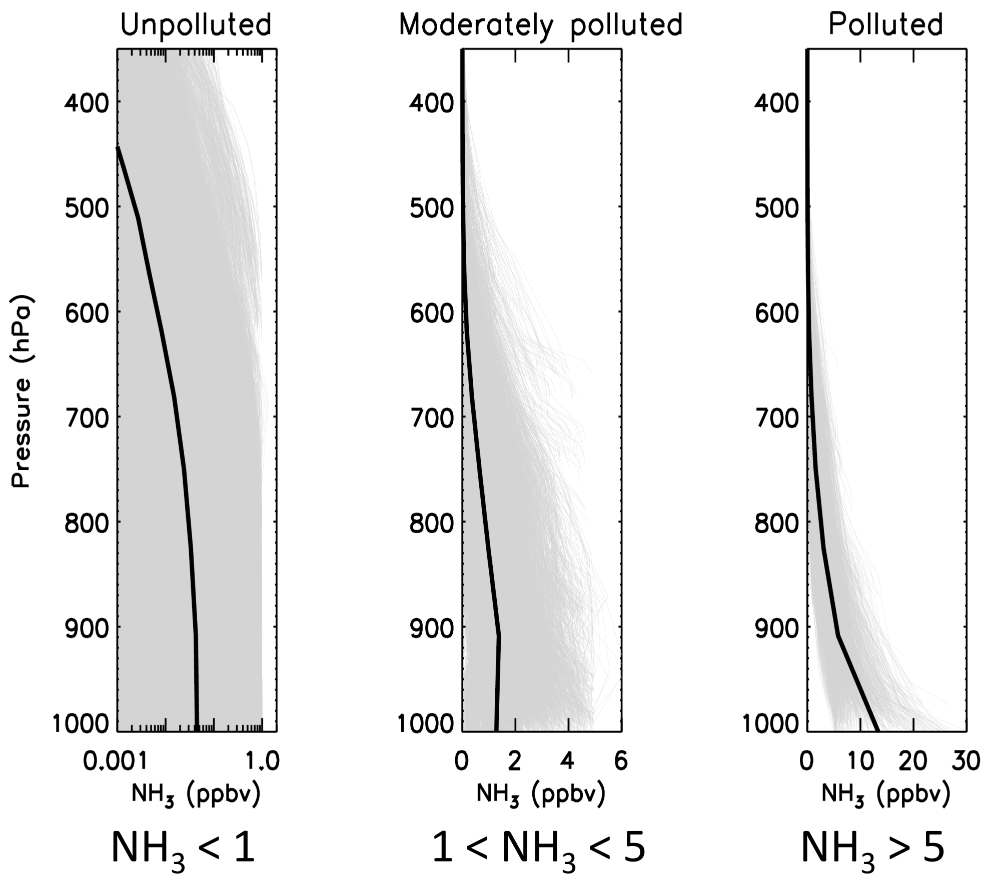
What can TES observations tell us about process-based emissions of ammonia?

PAN

What can TES PAN observations tell us about the sources and transport of PAN and O_3 with attention to anthropogenic intercontinental transport, biomass burning, and lightning?

GEOS-Chem was used to derive 3 a priori profiles of TES NH_3.

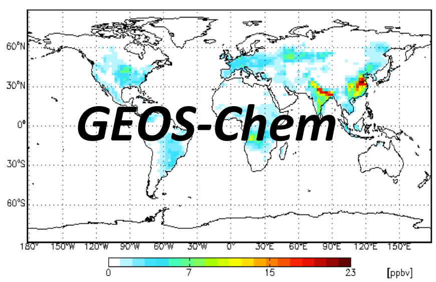
- Sensitivity peak: 700-900 hPa.
- Bias: ~ +0.5 ppb (at 825 hPa).
- Detection limit: ~ 1 ppb.
  (Shephard et al., 2011)
- TES NH_3 spatial and seasonal trends verified by surface obs.
  (Pinder et al., 2011)

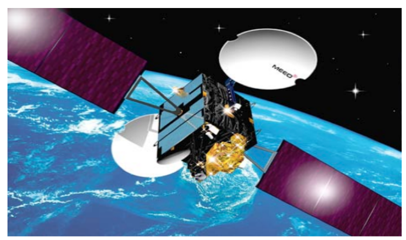


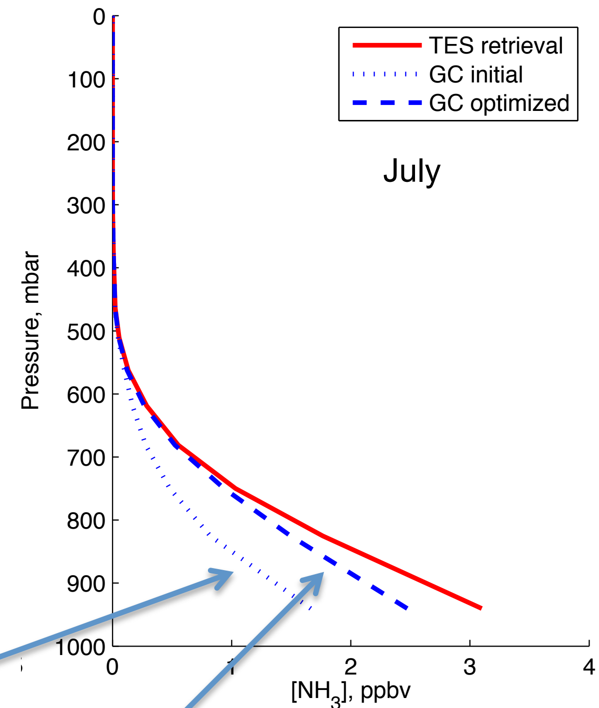
Shephard et al., 2011

Optimized NH_3 profiles more closely resemble TES retrieval.

- 4D-Var inversion (GEOS-Chem adjoint) to adjust NH_3 emissions.
  (North America, 2006 – 2009)



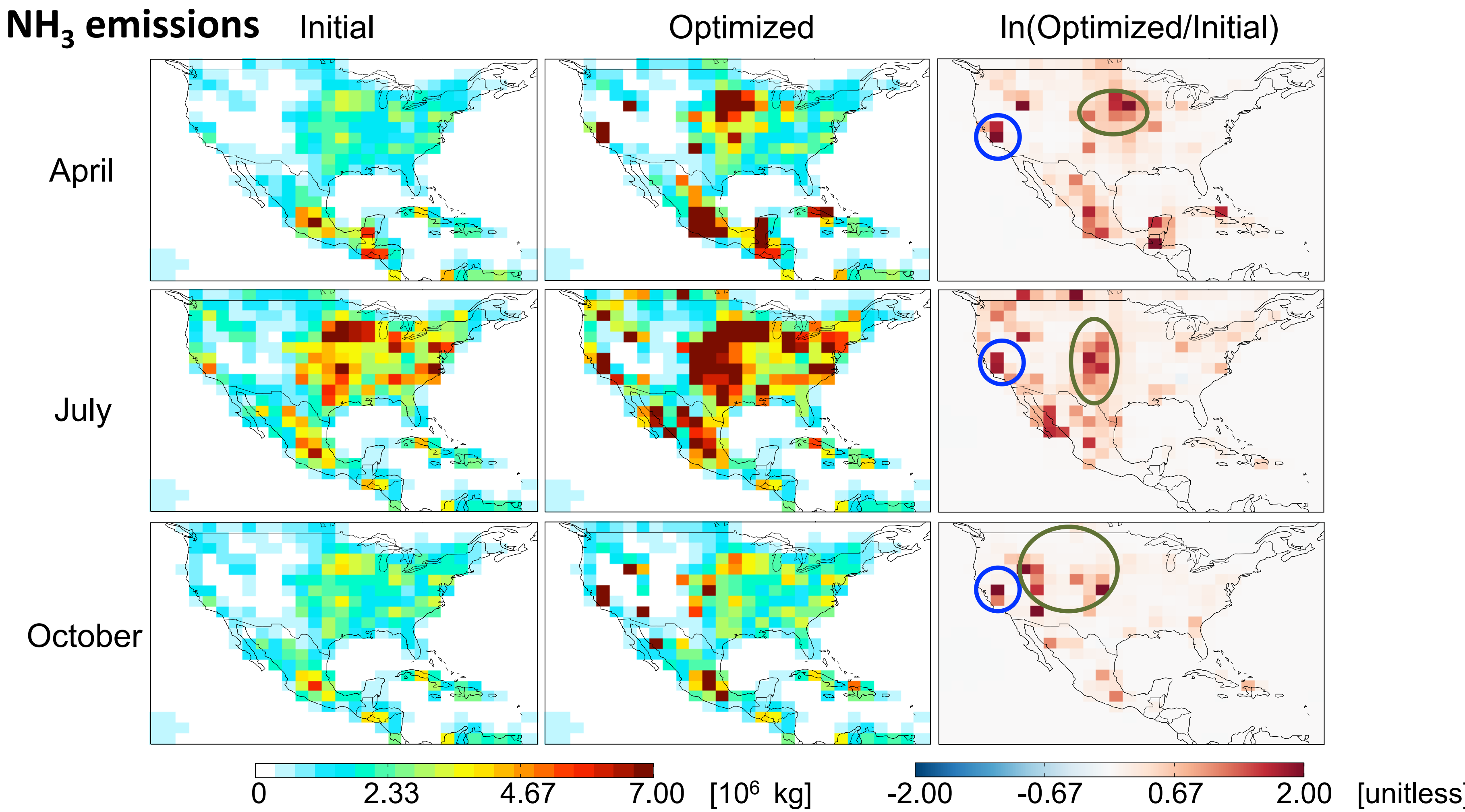




- Prior NH_3 profiles lower than TES retrieval.
- Optimized model still underestimates TES retrievals.
- Reductions of the cost function: 66% for April, 42% for July, 57% for October.

Zhu et al., 2013

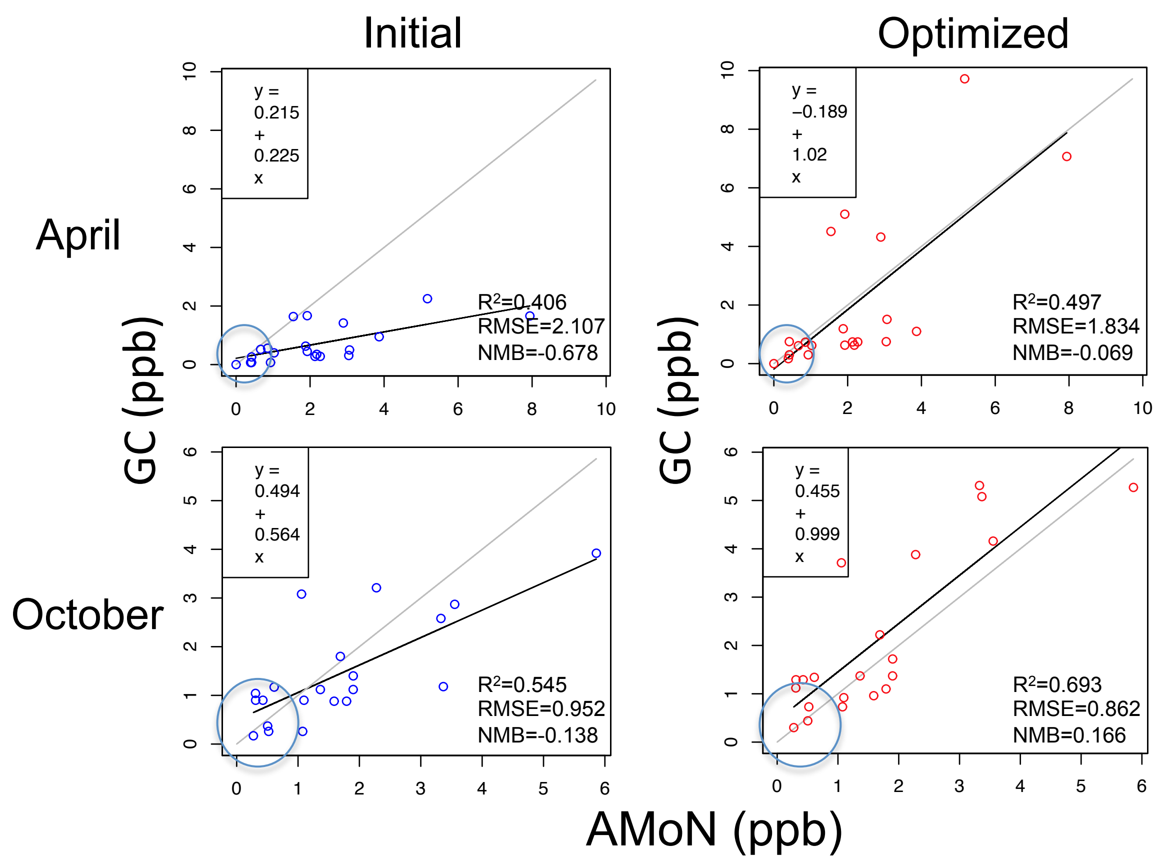
TES improves the model in the central and western US consistent with other studies.



- Changes in CA consistent with Walker et al. [2012] and Nowak et al. [2012]
- Central US underestimates noted from IIASI [Clarisse et al., 2009; Heald et al., 2012]

Zhu et al., 2013

TES assimilation improves the comparison with AMoN in April & October.

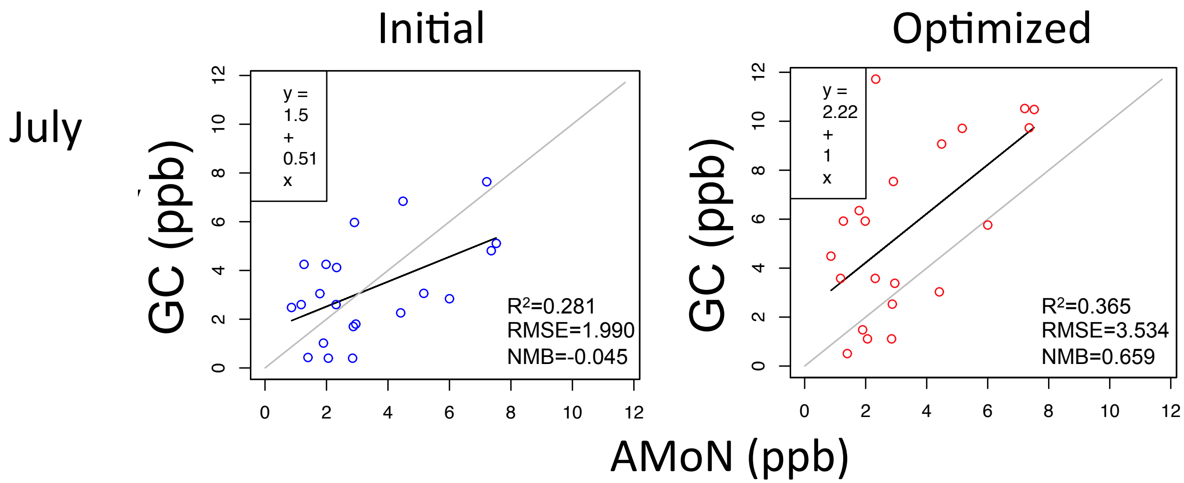


AMoN: 21 sites with 2-week long observation, Middle & Eastern US

- Model values below 1 ppb did not change significantly due to TES detection limit.

Zhu et al., 2013

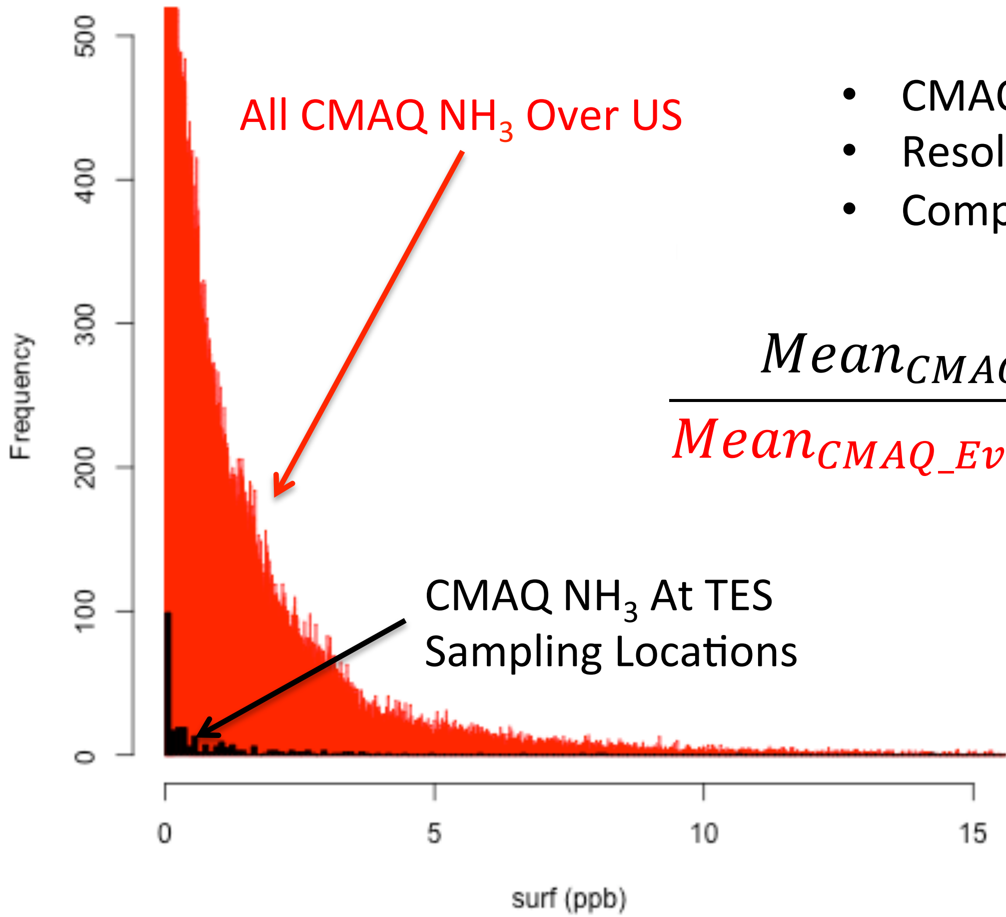
TES assimilation overestimates AMoN in July.



- Possible reasons for July bias:
  - Sampling bias due to TES level-of-detectability (i.e., lack of low values) or spatial sampling.
  - TES overpass time (1:30 AM & PM) points to missing model diurnal variability; new diurnal variability scheme improves comparison with SEARCH obs.
  - Bi-directional exchange was neglected in GC *(Zhu et al., in prep)*.

Zhu et al., 2013

TES sampling strategy leads to a +30% bias in surface concentration.



- CMAQ Model
- Resolution: 12km x 12km
- Comparable to TES footprint 5km x 8 km

$$\frac{Mean_{CMAQ\_TES}}{Mean_{CMAQ\_Everywhere}} = \frac{1.27\ ppbv}{0.98\ ppbv} = 1.3$$

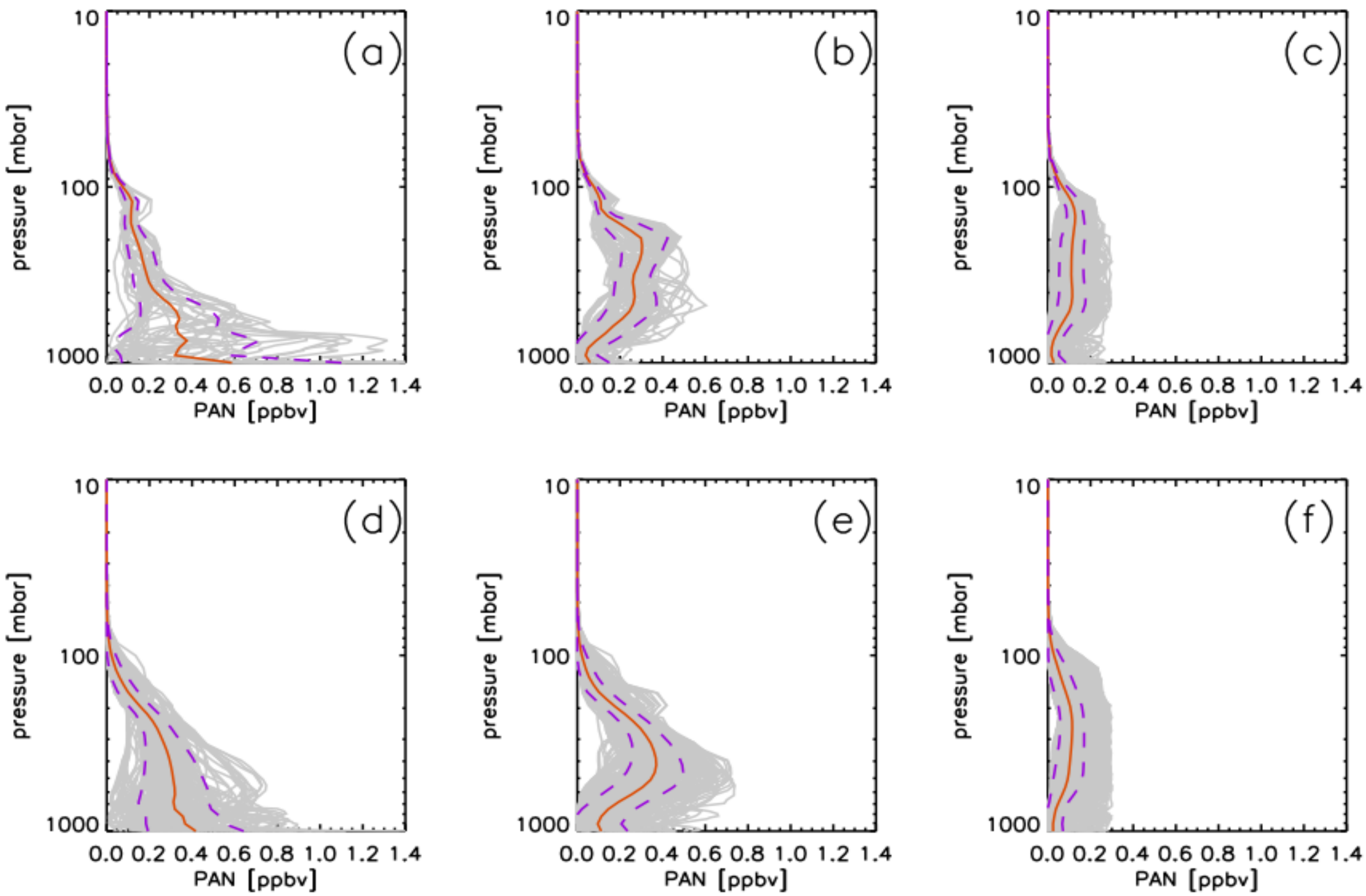
Future solution:
Include more retrievals with peak value of profile below 1 ppbv.

Zhu et al., 2013

A new GEOS-Chem PAN simulation was used to develop 6 a priori profiles.

Tropic

Outside tropics



Peak surface

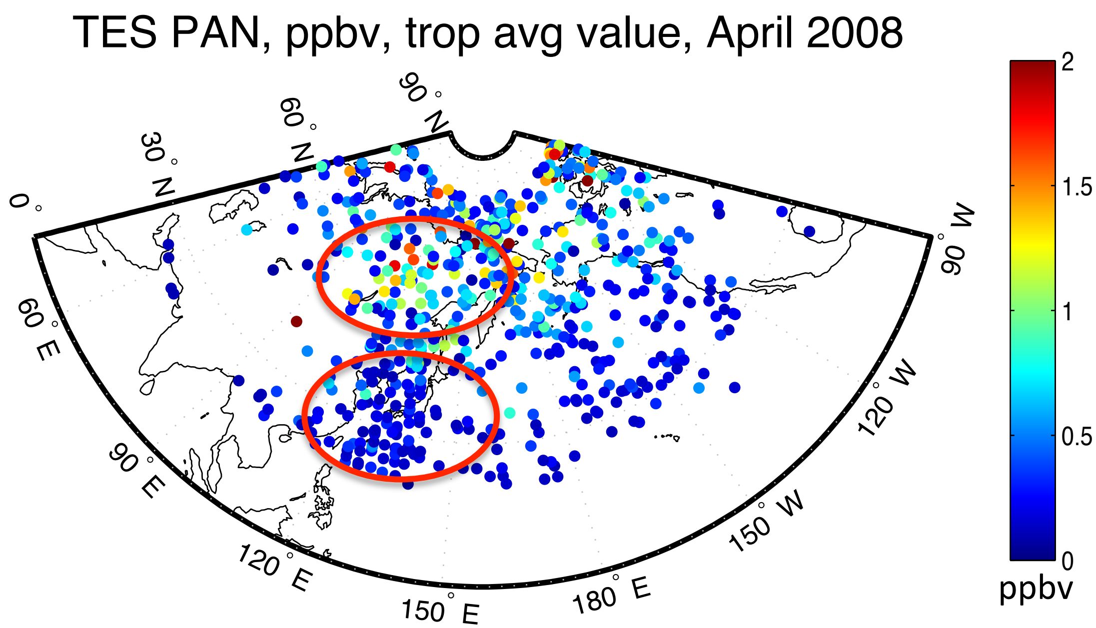
Peak aloft

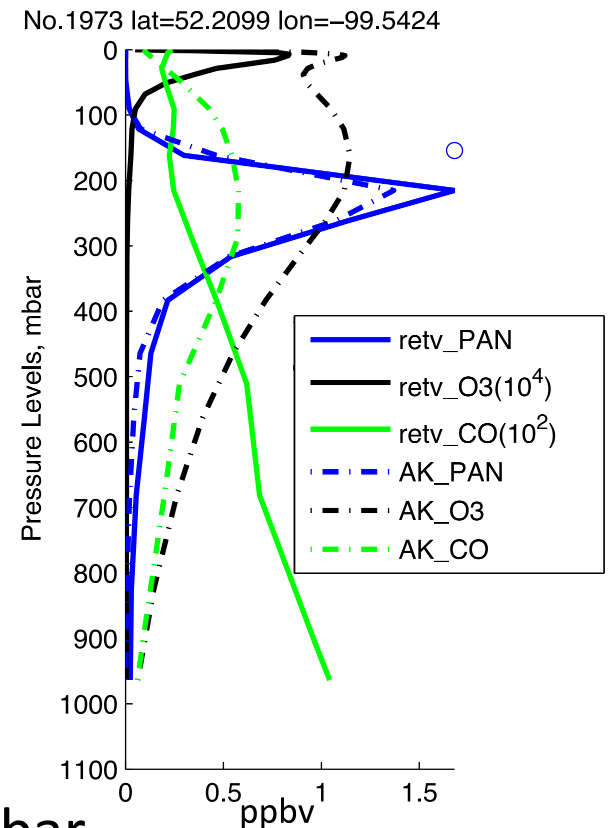
Peak value < 0.3 ppbv

Detection limit: 0.2 ppbv

Payne et al. 2014, ACPD

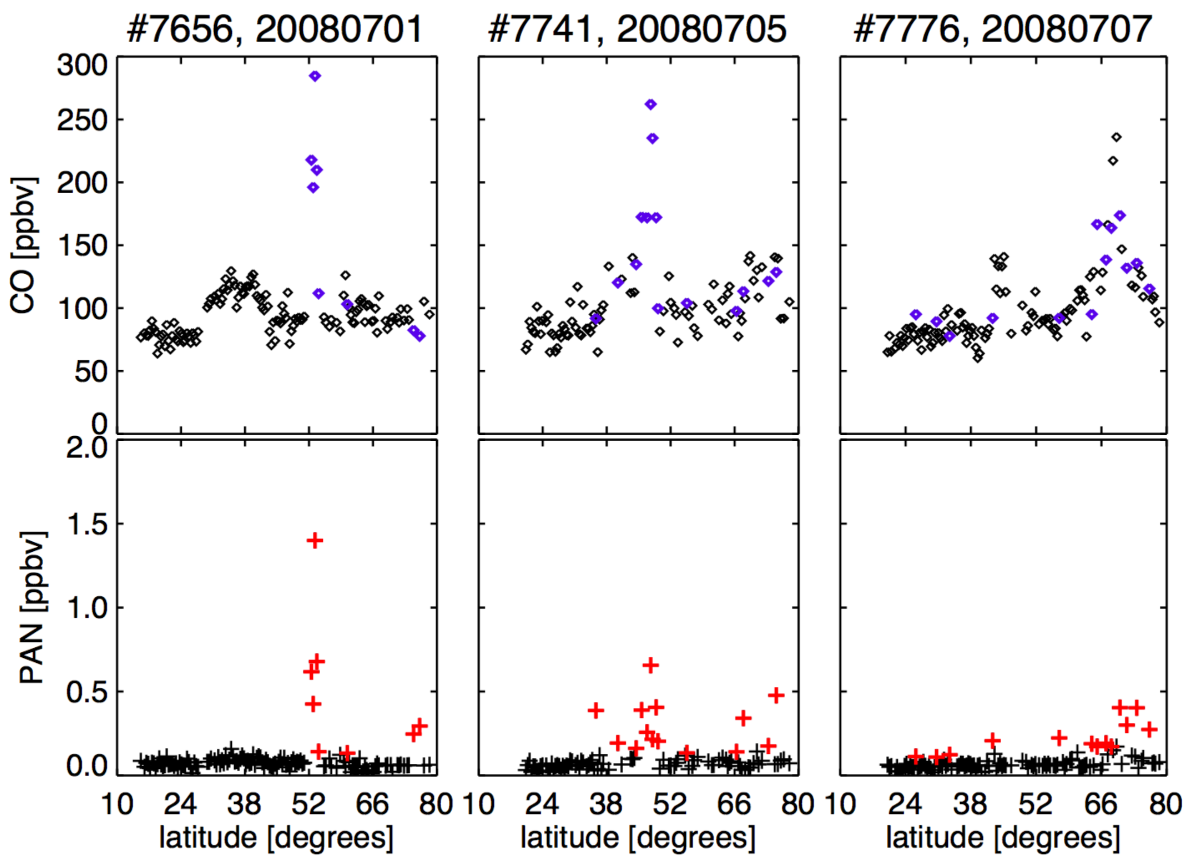
East Asia is a major PAN export region. We are focusing here first.





- 45°E – 90°W, 15°N – 80°N
- trop avg : average of levels between 800 mbar and tropopause

Elevated TES PAN is associated with elevated CO in fire plumes.

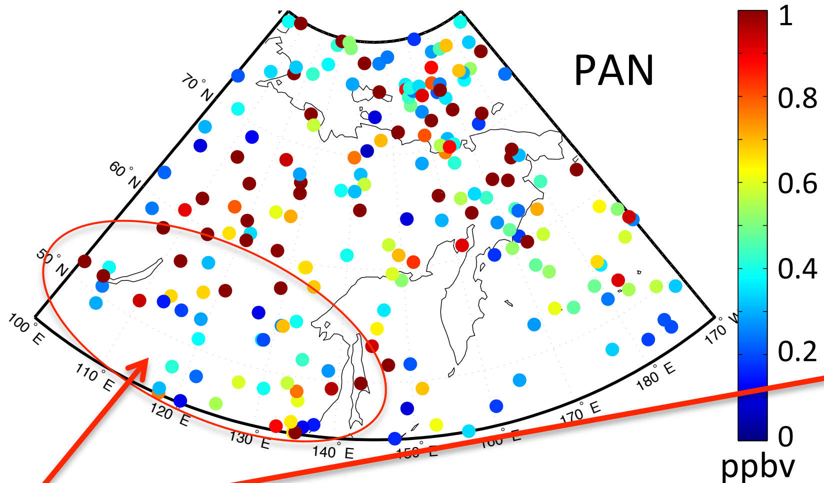


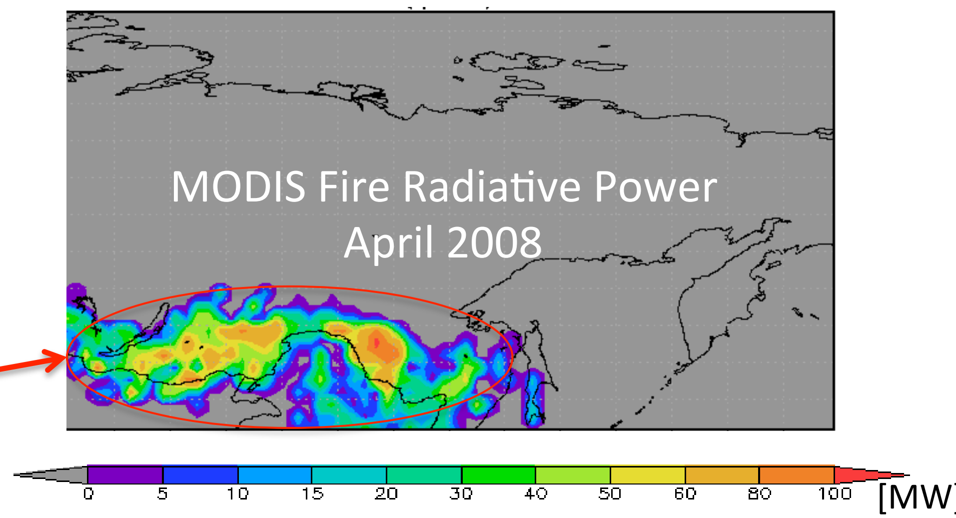
- TES observations from July 2008 ARCTAS campaign
- Colored points: DOFS > 0.6
- Previously identified by Alvarado et al. 2010

Payne et al. 2014, ACPD

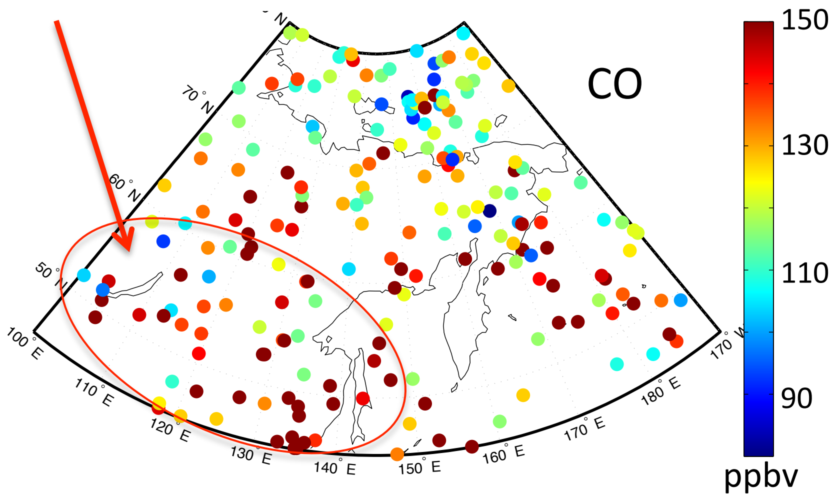
Spring 2008 was an extreme year for biomass burning in East Asia.

TES, trop avg value, East Russia



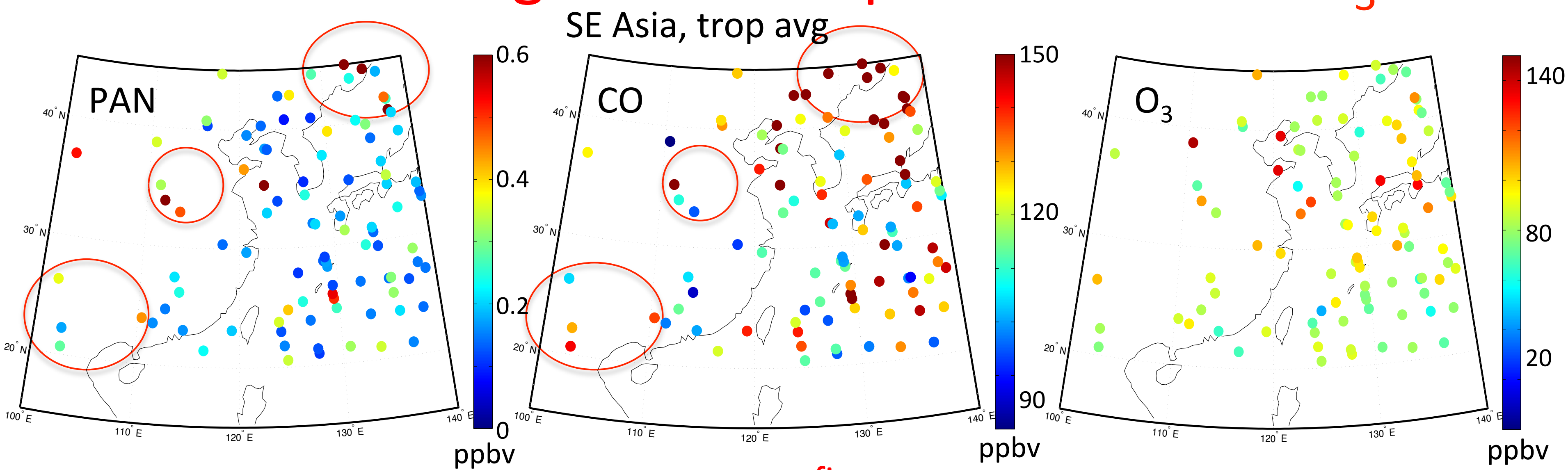


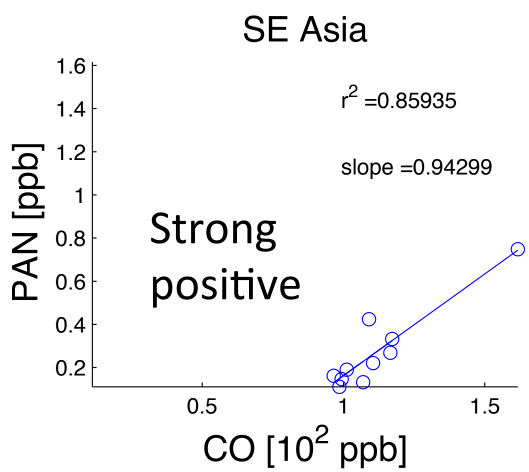
fires

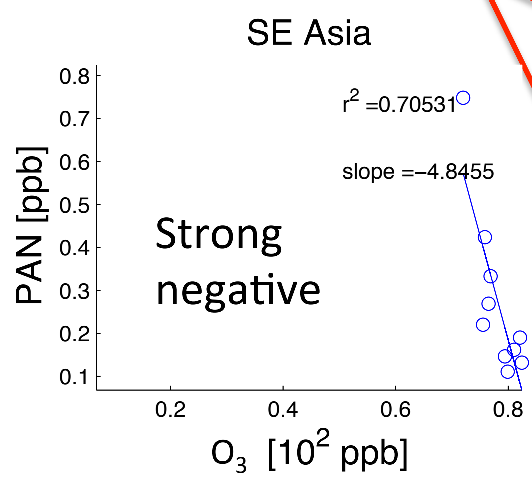


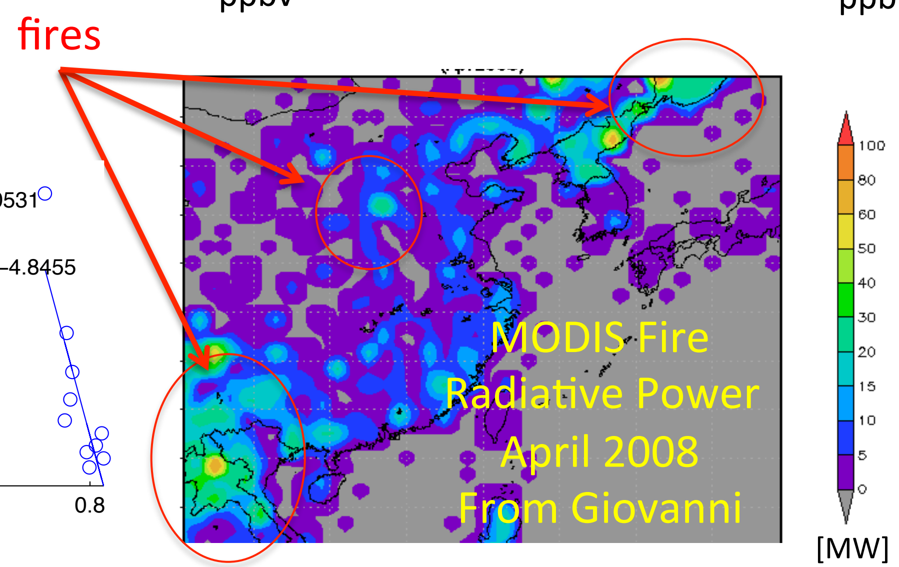
TES PAN and CO reflect this.

In April 2008, fires are a main source of extreme PAN and strong relationships with CO and O_3.



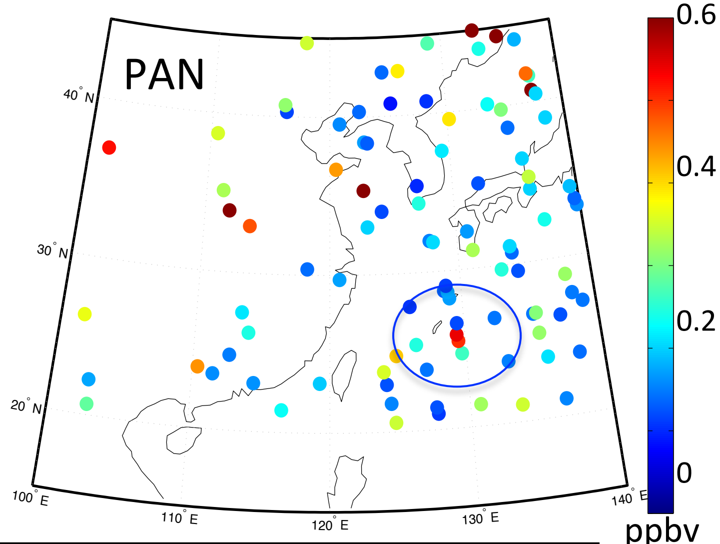


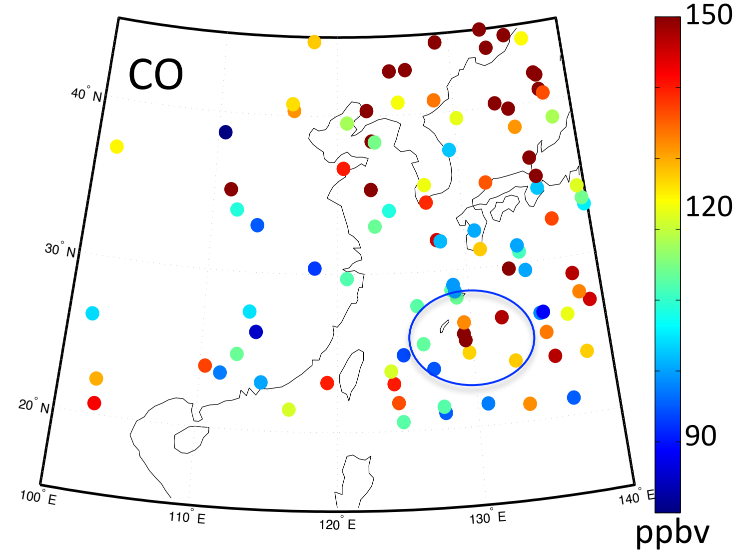


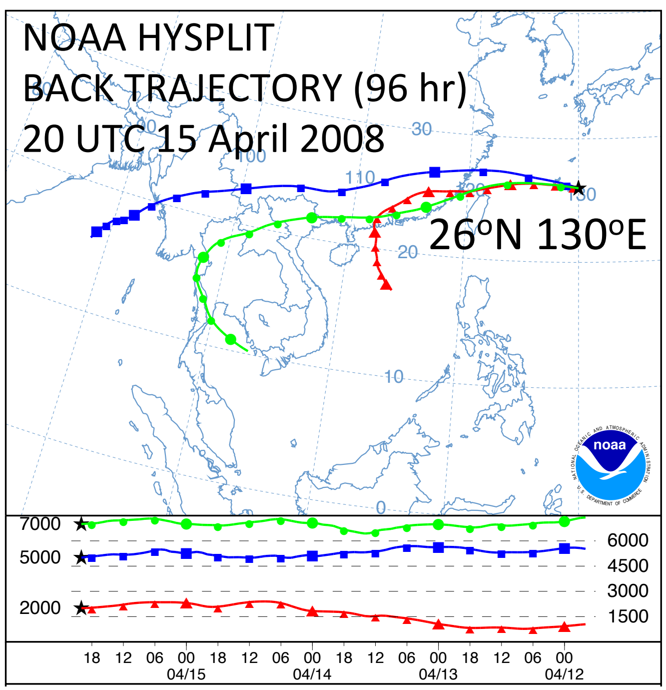


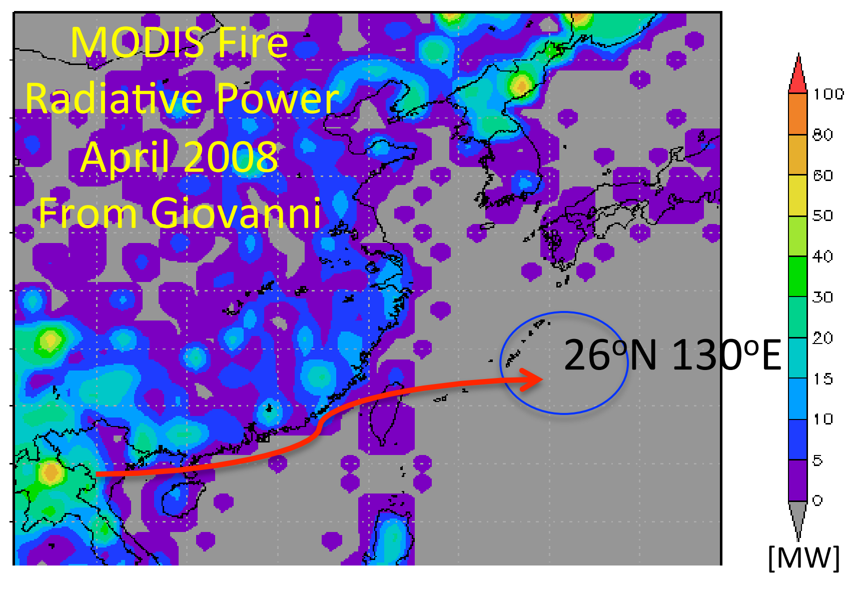
Binned by every 10 points

There is evidence of fire PAN and CO being exported over the Pacific in April 2008.









Conclusions

- TES NH_3 values are higher than initial model but potentially biased.
- TES assimilation enriches our understanding of NH_3 emissions in the West and Midwest US .
- TES assimilation better captures the range and variability of surface NH_3 in April and October, but it is biased high in July.

Preliminary Conclusions

- Fires are a main source of extreme PAN in April 2008 and we see strong relationships with CO and O_3 in East Asia.
- Elevated TES PAN is associated with elevated CO in fire plumes.
- There is evidence of fire PAN and CO exported over Pacific.

Thanks !

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NH_3 has large impacts on human health and the environment.

$PM_{2.5}$ causes **bronchitis**, **asthma**, **premature mortality**…

Smog

Decreases visibility**.**

Eutrophication

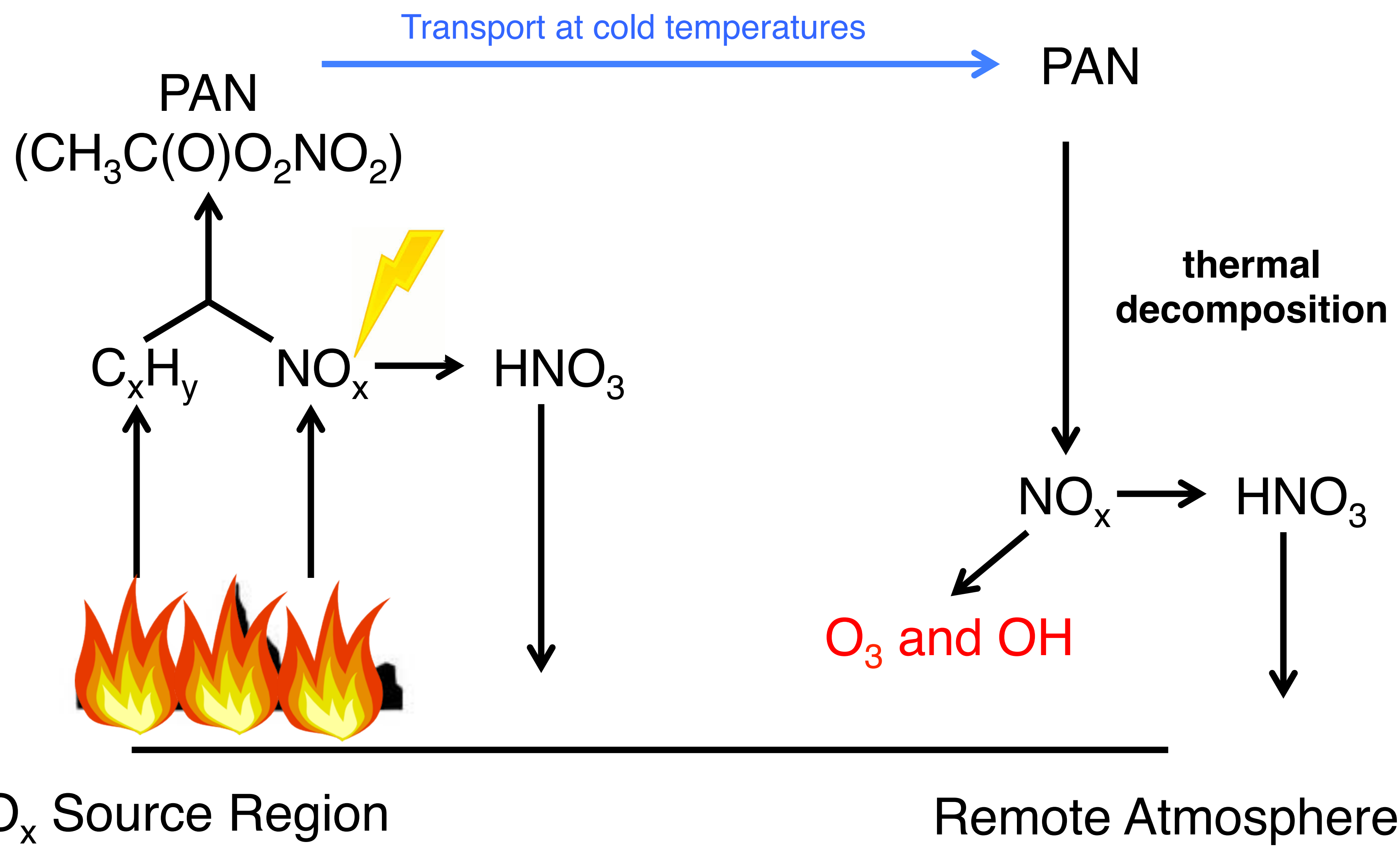
Alga blooms; Hypoxia;
Cloudy, colored water.

Soil acidification

Nitrification of NH_4^+ into NO_2^-, releasing H^+.

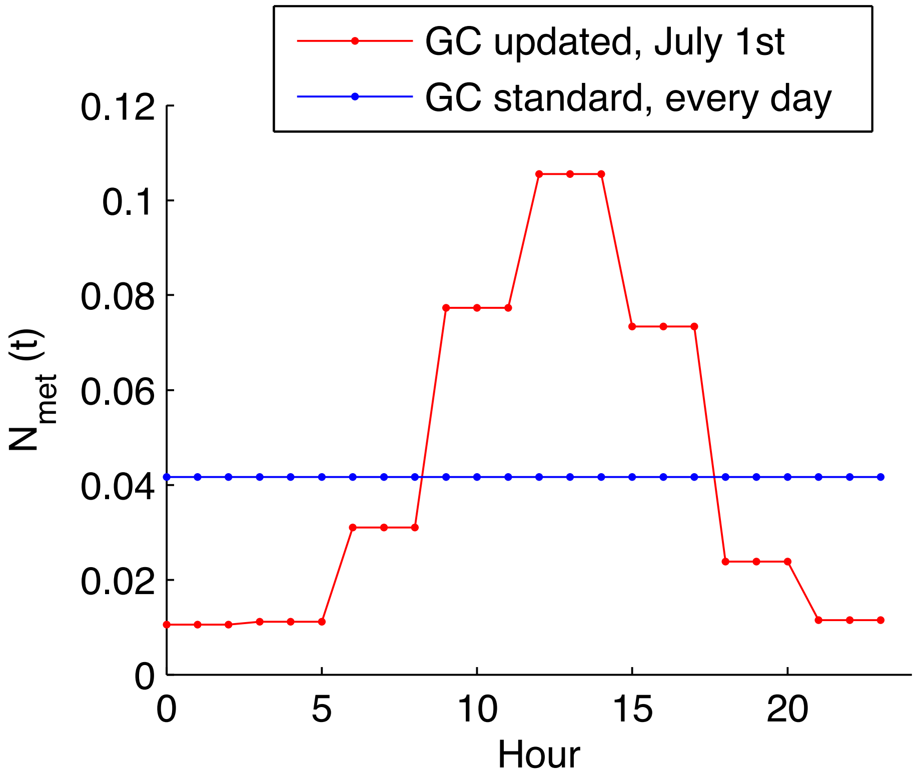
Large uncertainties in NH_3 inventories.

PAN is the route for NO_x to reach the remote troposphere.



Jacob 1998

Implementing diurnal variability for livestock NH_3 emissions reduces bias.



$E_h(t) = E_d * N_{met}(t)$

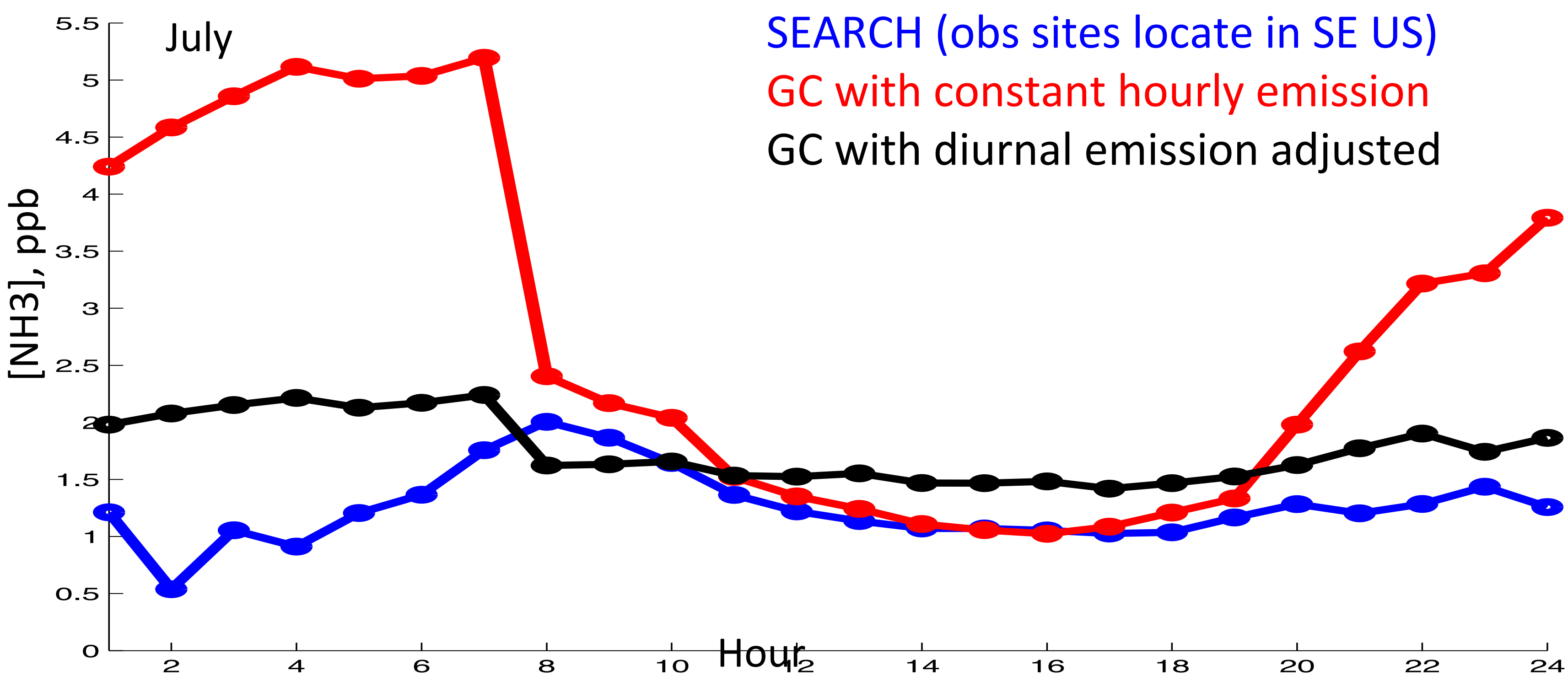
t: hour, 1,2, … , 24;
E_d: daily NH_3 emission;
$E_h(t)$: NH_3 emission at hour t;
$N_{met}(t)$: fraction of NH_3 emission diurnal variation, depends on T, $R_{aerodynamic}$

$$N_{met}(t) = \frac{H(t)/R_a(t)}{\sum_1^n (H(t)/R_a(t))}$$

$$H(t) = \frac{161500}{T} e^{-10380/T}$$ (Nemitz et al., 2000)

Scheme developed based on field studies downwind of livestock facilities in North Carolina (Bash et al., in prep.)

Implementing diurnal variability for livestock NH_3 emissions reduces bias.



- NH_3 decreased at night by several ppb; increased in day up to 1 ppb.
- Monthly average surface NH_3 (and NO_3^-!) decreased.
- NH_3 concentration (at TES overpass time 13:30) can be impacted without changing total emissions.
- Improves TES assimilation results compared to Zhu et al. 2013.