Evaluating CMAQ Simulations of Ammonia Sources and Impacts using Surface, Aircraft, and Satellite Data

C. R. Lonsdale[1], J. D. Hegarty[1], K. E. Cady-Pereira[1], M. J. Alvarado[1], D. K. Henze[2], M. Turner[2], S. Capps[2], J. B. Nowak[3], J. Murphy[4], M. Markovic[4,5], T. VandenBoer[4,6] and R. Ellis[4]

[1]Atmospheric and Environmental Research, Inc.

[2]Univerisity of Colorado – Boulder

[3]Aerodyne Research, Inc.

[4]University of Toronto

[5]NOAA ESRL and CIRES

[6]Memorial University of Newfoundland

NH_3 is an important $PM_{2.5}$ precursor

$NH_3 + HNO_3 \leftrightarrow NH_4NO_3$

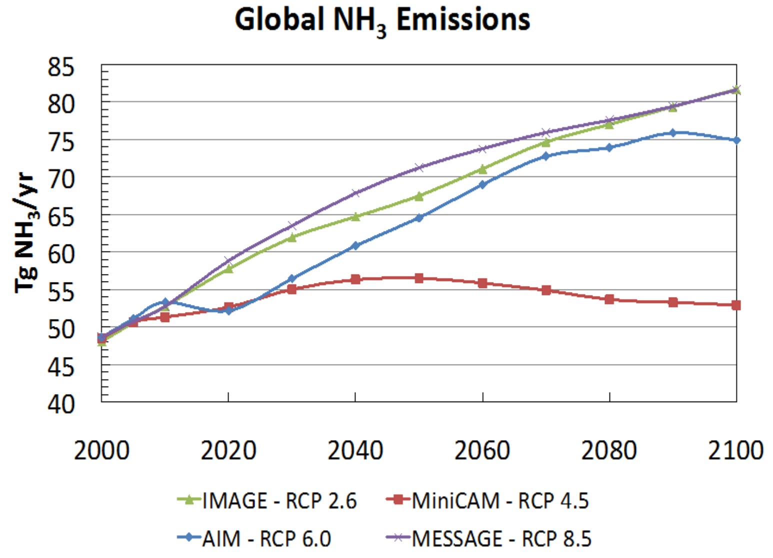
$2\ NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4$

Long-range export

Long-range import

$PM_{2.5}$

SO_2, NO_X decreasing
but NH_3 forecast to increase



Nitrogen Deposition

- **Increase incidence of cardiovascular and respiratory diseases**
- **Increase number of CCN**
- **Harmful algal blooms**
- **Loss of species diversity**

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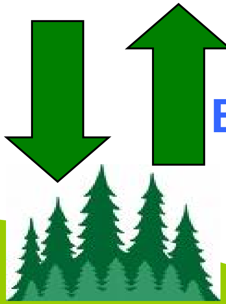
NH_3 sources are not well known

Biomass burning

Automobiles (catalytic converters)
- Large urban centers
  - 50% of NH_3 in LA area (Nowak et al., GRL, 2012)

Industry
- Fertilizer
- Coal Mining
- Power generation

AGRICULTURE
- Animal waste (temperature dependent)
- Fertilizer application



Bi-directional Flux

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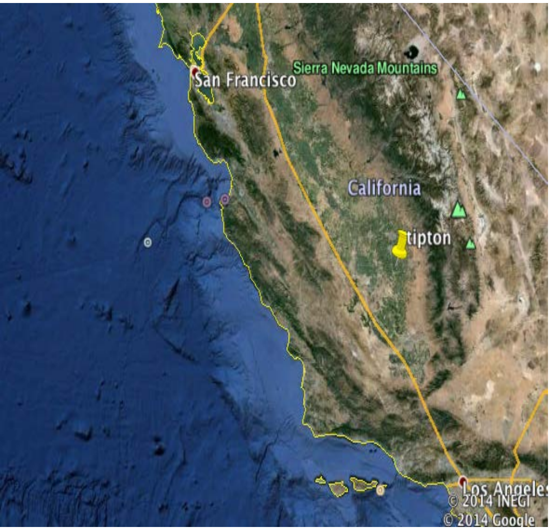
Monitoring NH_3 is difficult

NH_3 is highly reactive

➔ highly variable in space and time

- NH_3 from an Open path Quantum Cascade Laser (QCL) on a moving platform in the San Joaquin Valley during DISCOVER-AQ 2013.



Miller et al., AMT 2013

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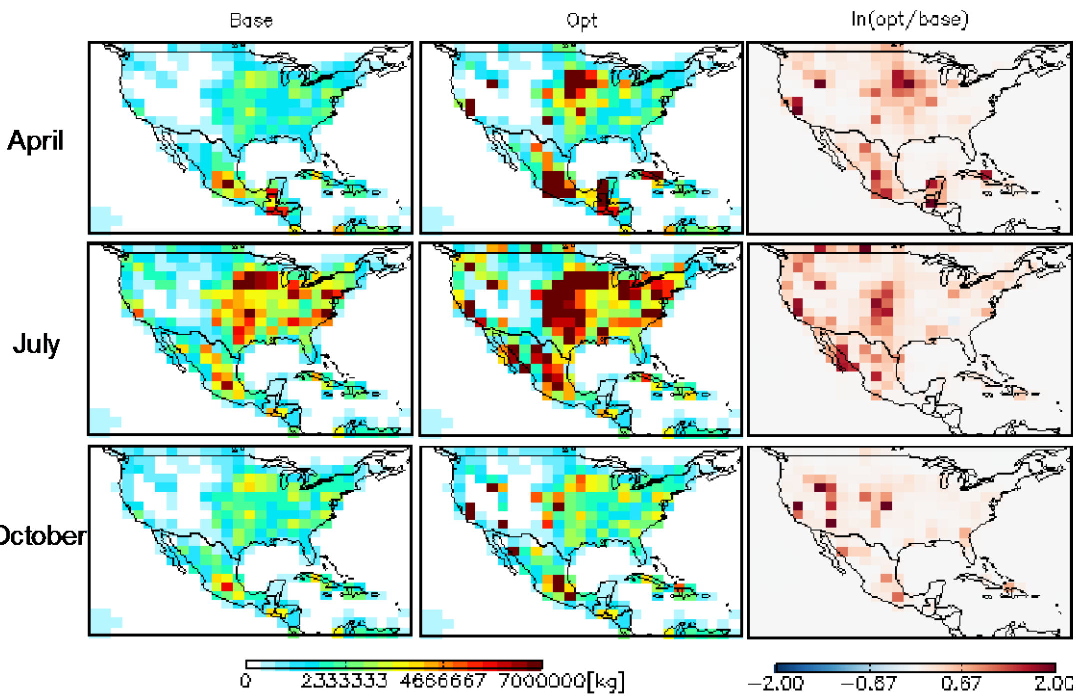
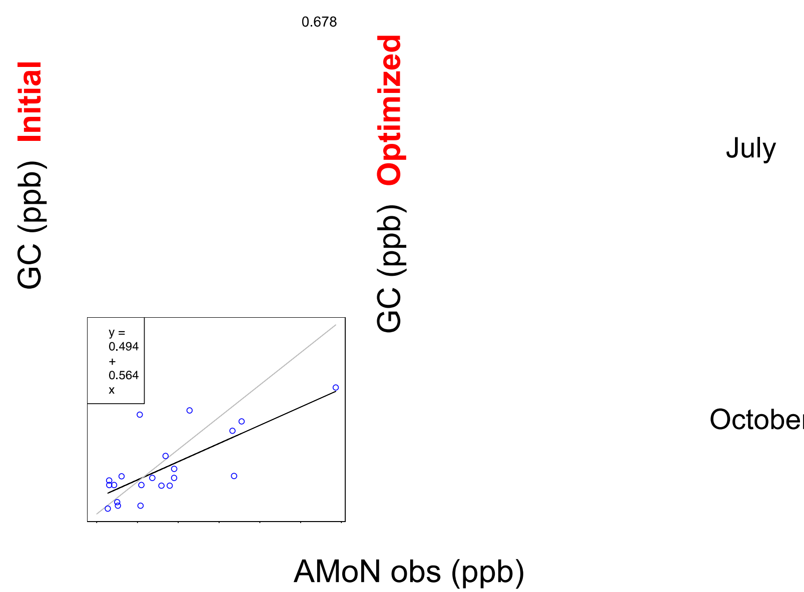
Better emissions with TES NH_3



Largest changes western US and Mexico



- Used GEOS-Chem adjoint with TES NH_3 profiles, averaging kernels and error covariances to optimize model
- Optimized GC shows better agreement with **AMoN** network measurements

Zhu et al., 2013, JGR

Outline

- Using TES NH_3 data, along with surface and aircraft data, to investigate NH_3 emissions during 2010 CalNex Campaign.

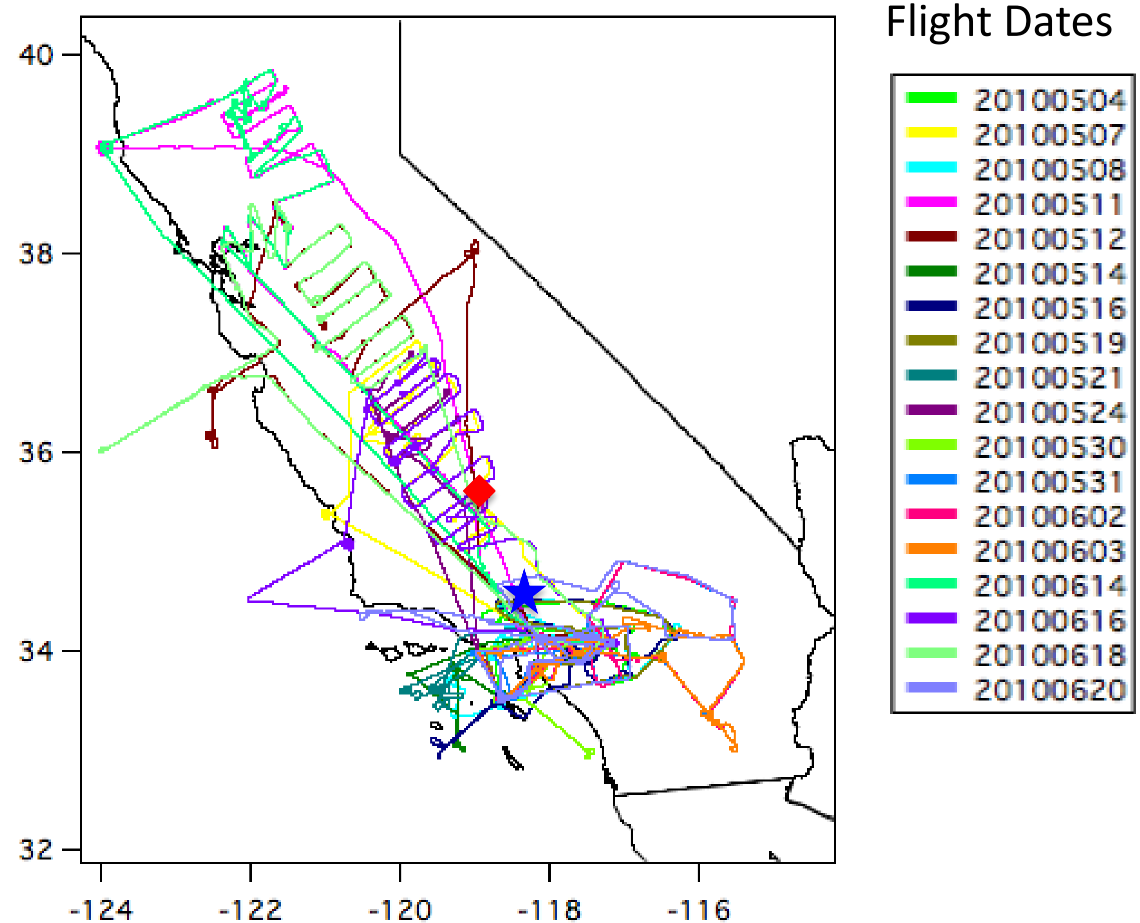
- Using the Cross-Track Infrared Sounder (CrIS) to investigate NH_3 sources in California and Southeast US.

CalNex 2010 field campaign

- Combined satellite, aircraft and ground-based measurement campaign focused on the California Central Valley and Los Angeles Basin during May – June 2010.

- Provides rich data set for studying NH_3 emissions.

NOAA WP-3D Flight Tracks



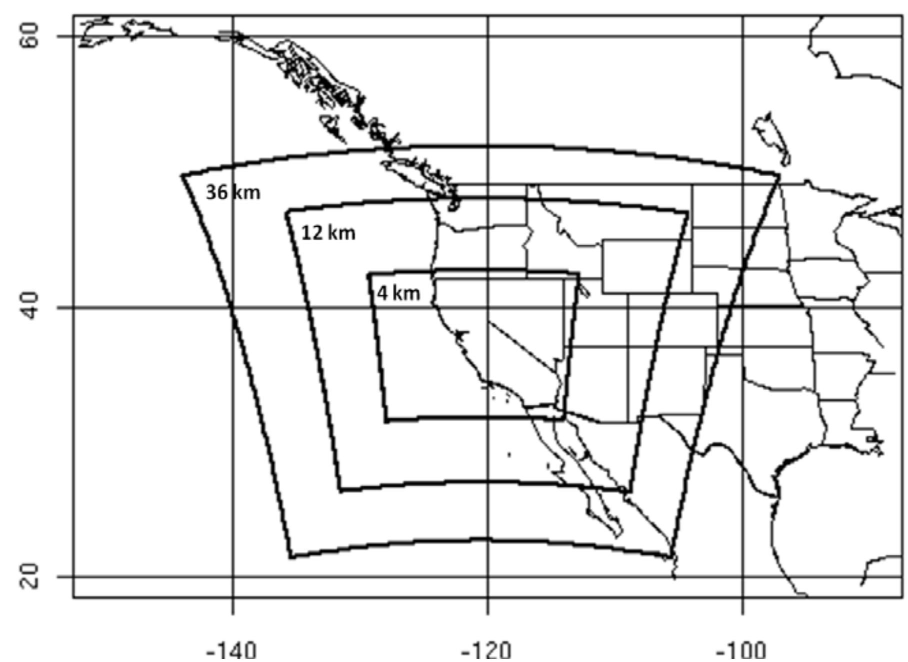
◆ Bakersfield site – mostly agricultural sources

★ Los Angeles site – urban setting: agricultural, industrial and mobile sources

WRF and CMAQ Modeling

- WRF-ARW v3.5 with 3 nest levels of 36, 12 and 4 km
  - 41 levels, 1st layer ~50 m
- CMAQv5.0.1 run on inner 4 km domain only.
  - cb05 photochemistry with updated toluene chemistry
  - ae6_aq - aerosol module 6 with aqueous chemistry
  - No bi-directional NH_3 flux
- CMAQ boundary conditions provided by GEOS-Chem on a $2.0^0 \times 2.5^0$ grid.
- Emissions provided by California Air Resources Board (CARB)

WRF Domains



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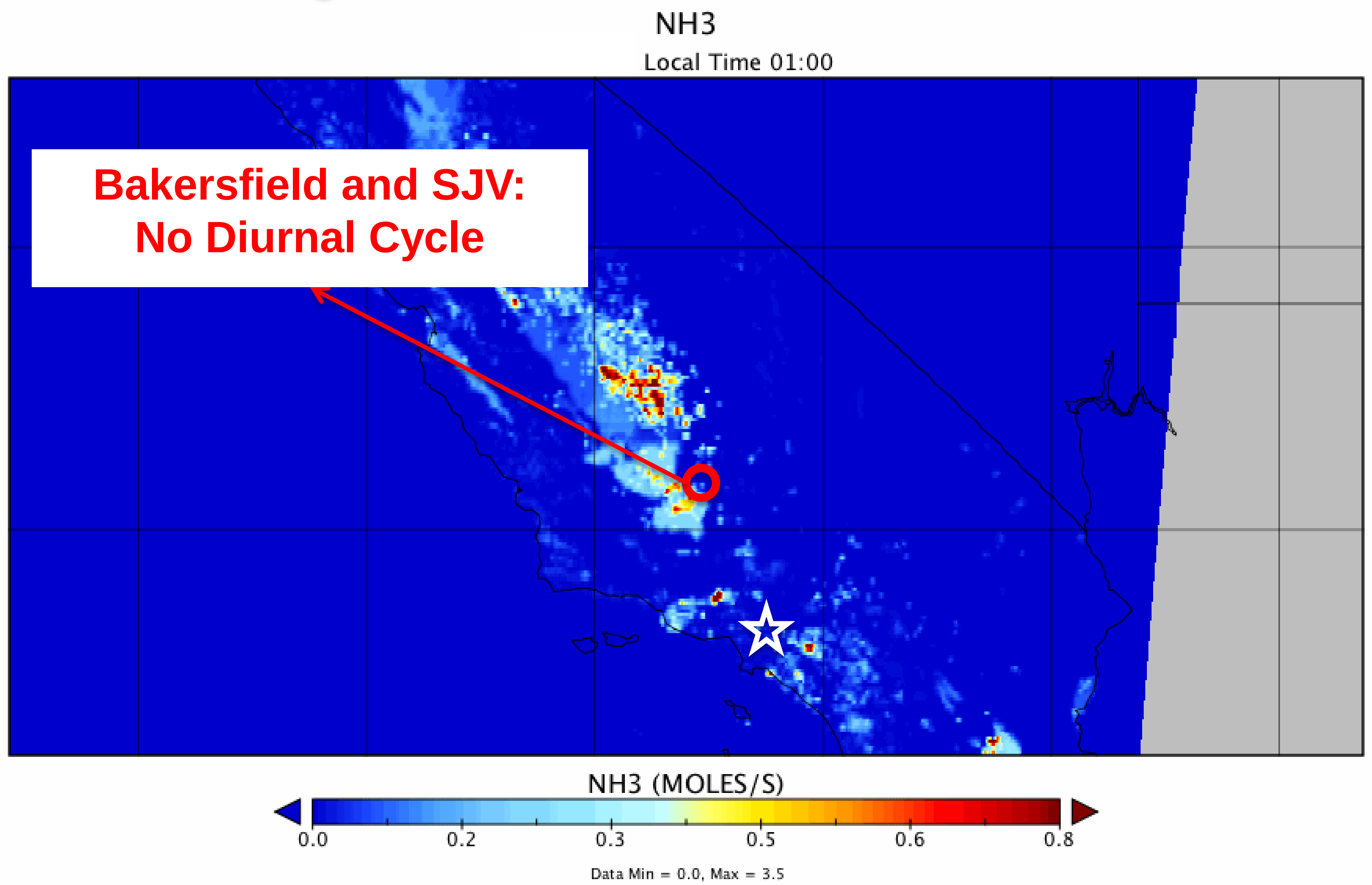
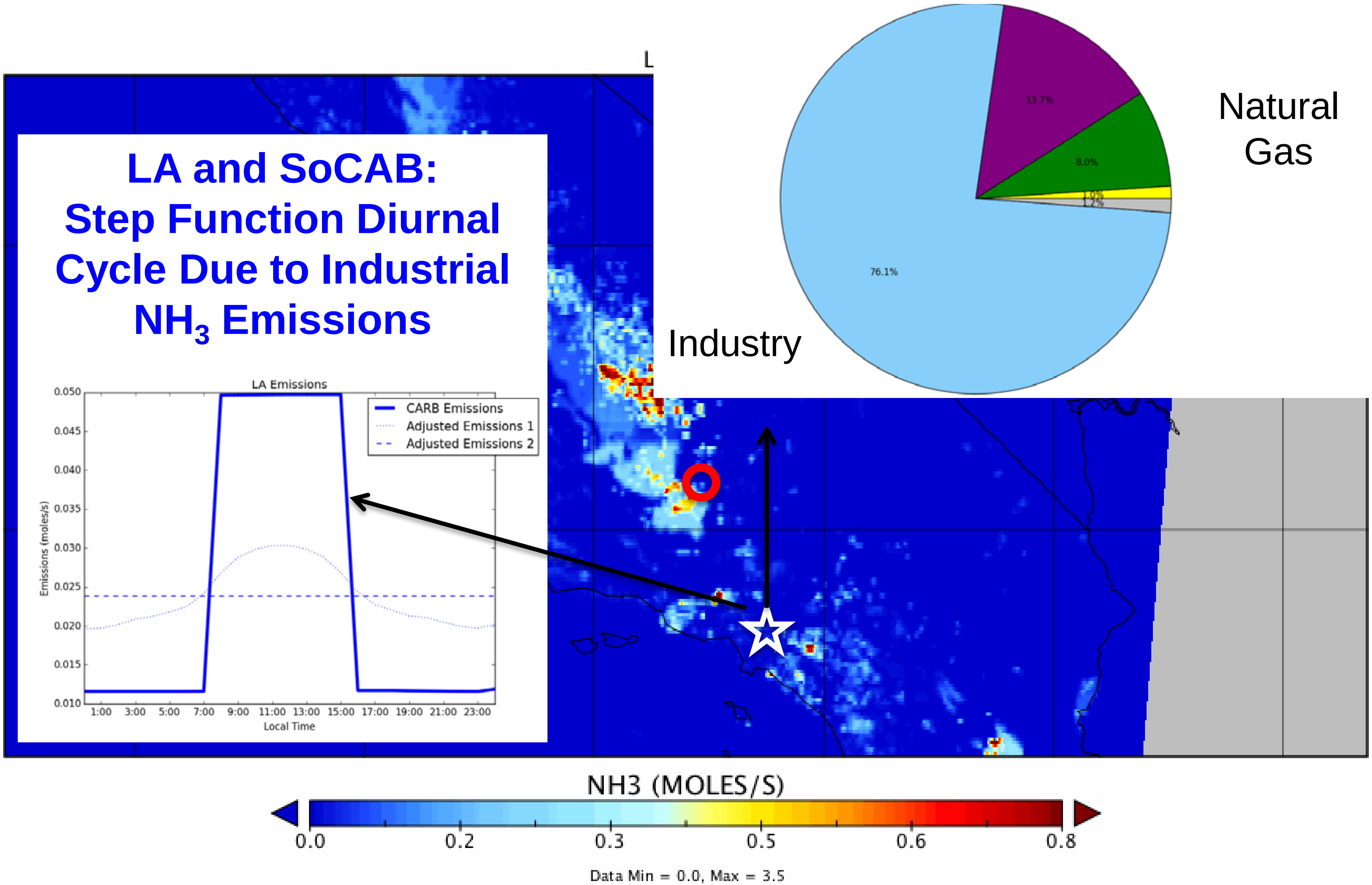
Lonsdale et al., in prep.

CARB NH_3 Emissions



CARB NH_3 Emissions



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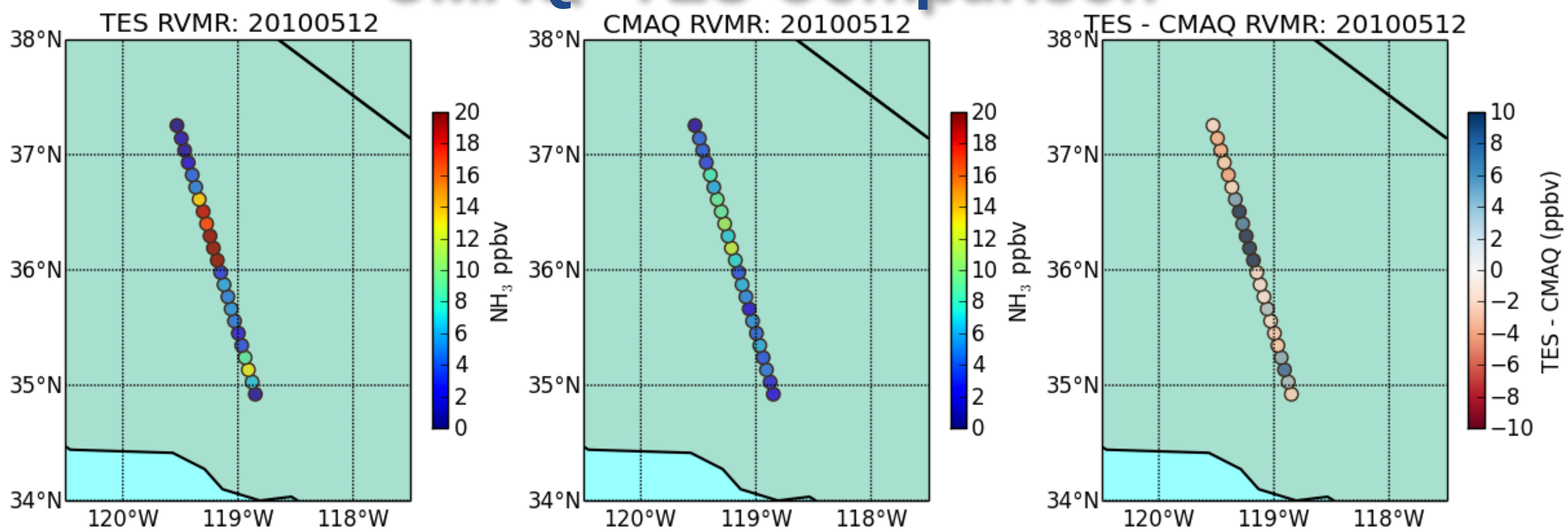
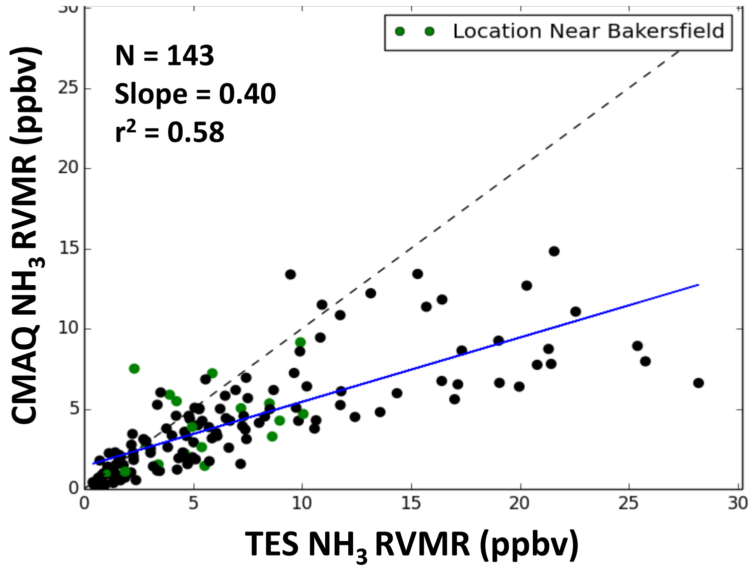
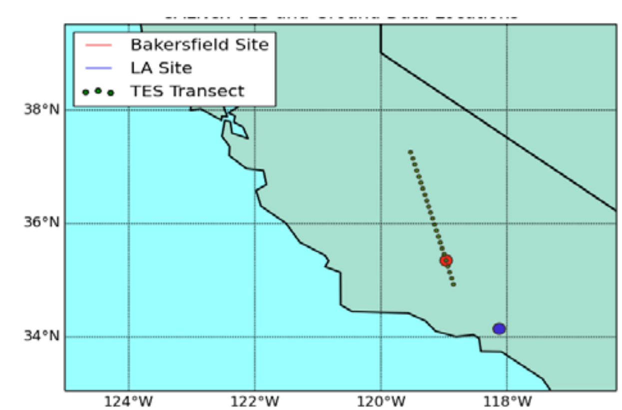
CMAQ -TES Comparison







- 6 TES transect days during CALNEX campaign at ~1:30 pm local time
- CMAQ and TES generally agree on the locations of the high and low NH_3.
- CMAQ seems to be biased low compared to TES for the highest NH_3 RVMRs.

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Surface Observations of NH_3 Diurnal Cycles

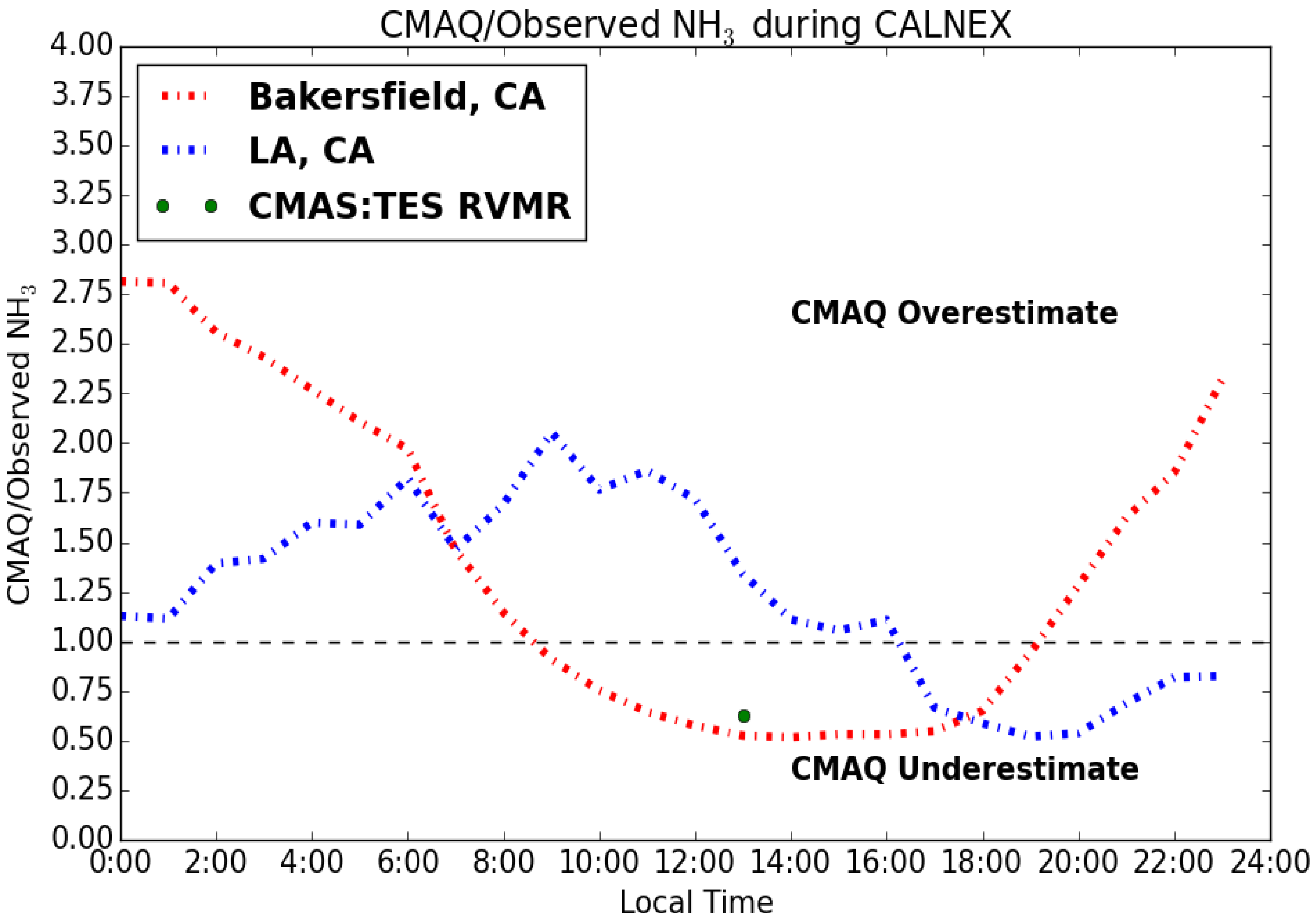
Bakersfield:

- CMAQ low during day, matching TES
- But too high at night

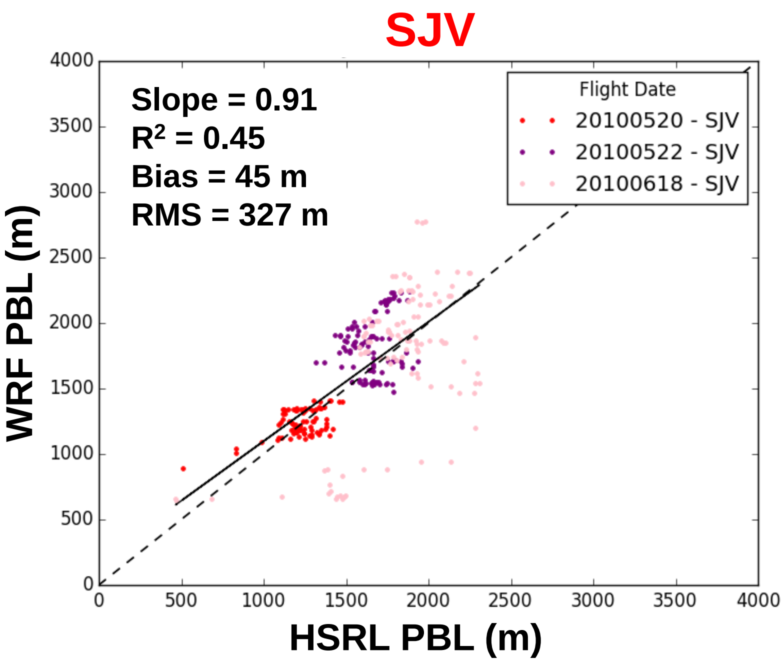
Los Angeles

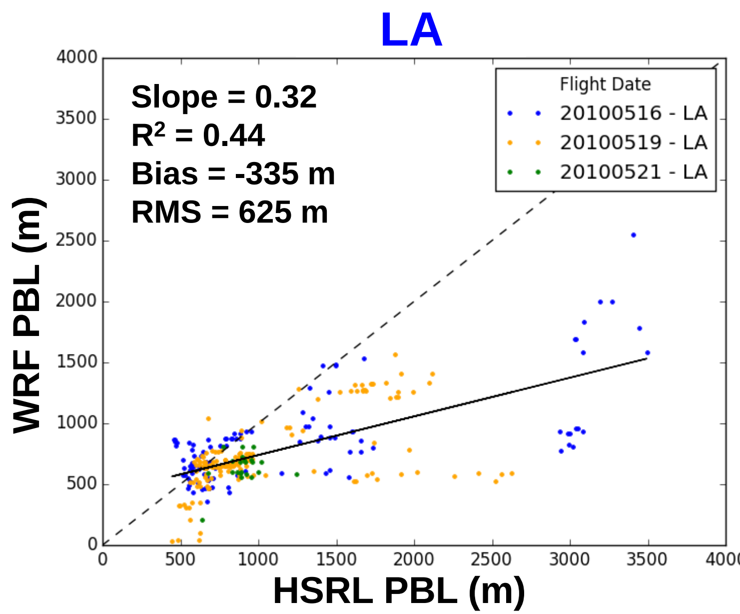
- Opposite pattern

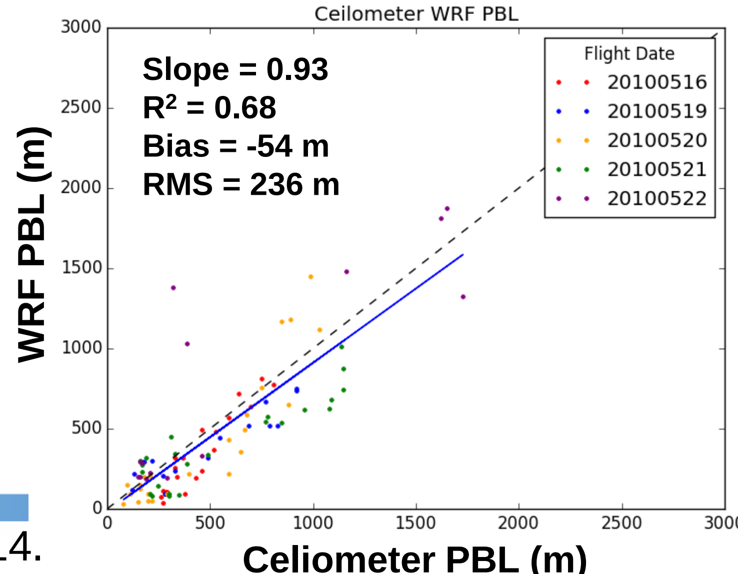
Results aren't sensitive to BCs, gas-aerosol partitioning, or diurnal changes in transport directions.



Are PBL height errors responsible?







SJV: PBL errors negligible.

LA: Celiometer suggests PBL errors negligible, but HSRL shows negative bias consistent with daytime NH_3 overestimate.

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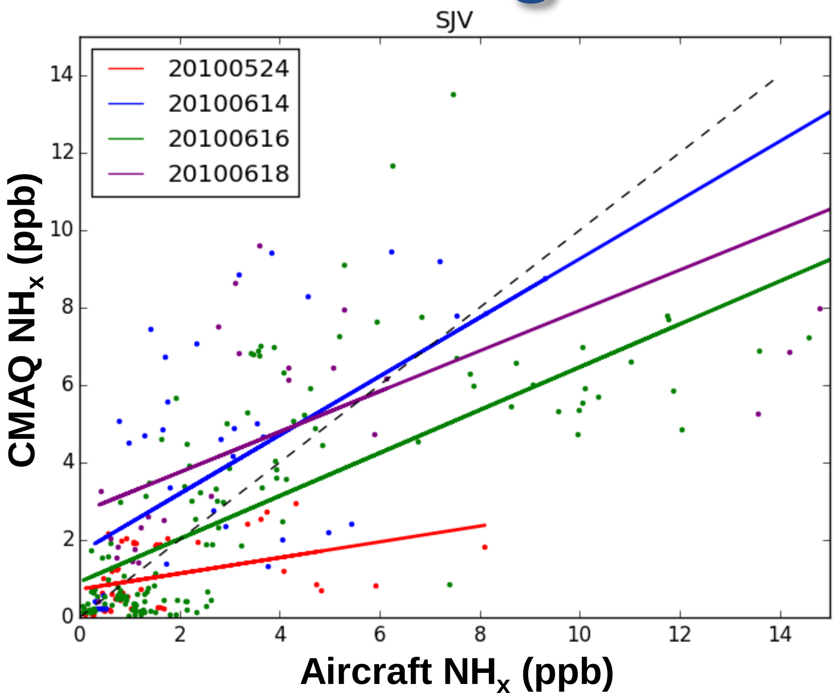
HSRL data from Scarino et al., ACP, 2014.
Celiometer data from Haman et al., JAOT, 2012.

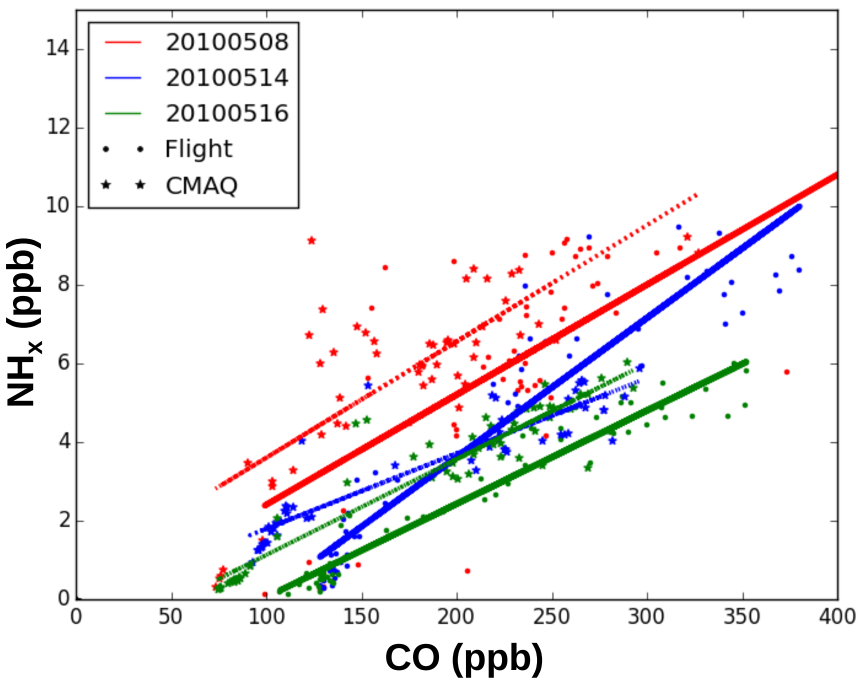
For Bakersfield, satellite, surface, and aircraft data give a consistent picture.



Date	Time (PST)	NH_x	
		Slope	R^2
20100524	16:00-22:00	0.20 ± 0.07	0.17
20100614	11:00-18:00	0.76 ± 0.07	0.73
20100616	11:00-18:00	0.56 ± 0.04	0.55
20100618	11:00-18:00	0.52 ± 0.08	0.64
Average		**0.51 ± 0.13**	

- Afternoon flights also show CMAQ underestimating NH_3 by a factor of 2, consistent with surface and satellite data.
- No such underestimate seen for CO.
- Since HSRL data suggests WRF PBL is good in SJV, **most likely explanation is an error in the diurnal cycle and/or daily magnitude** of SJV NH_3 emissions.

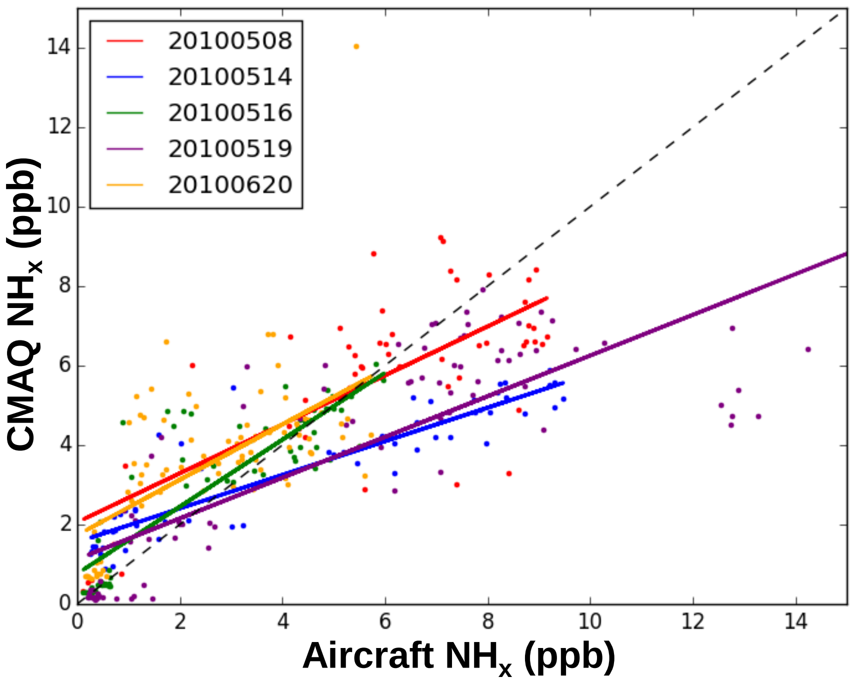
LA is more complicated. CMAQ gives reasonable NH_x/CO slopes relative to aircraft data…



Date	NH_x:CO Slope	
	FLIGHT	CMAQ
20100508	0.028 ± 0.005	0.029 ± 0.004
20100514	0.035 ± 0.002	0.019 ± 0.001
20100516	0.024 ± 0.001	0.024 ± 0.001
20100519	0.036 ± 0.002	0.032 ± 0.001
20100620	0.029 ± 0.002	0.020 ± 0.003
Average	**0.030 ± 0.006**	**0.025 ± 0.005**

- Only using data in LA urban core. Does not include data from dairy farms downwind of LA.
- Model NH_x/CO slopes consistent with afternoon aircraft data and analysis of Nowak et al., GRL, 2012.

…but aircraft data suggest CMAQ underestimates NH_x in afternoon in LA, opposite of surface data.



Date	Time (PST)	NH_x	
		Slope	R^2
20100508	11:00-18:00	0.61 ± 0.09	0.51
20100514	10:00-16:00	0.42 ± 0.03	0.82
20100516	11:00-18:00	0.84 ± 0.07	0.73
20100519	11:00-16:00	0.51 ± 0.04	0.71
20100620	11:00-17:00	0.70 ± 0.10	0.37
Average		**0.62 ± 0.16**	

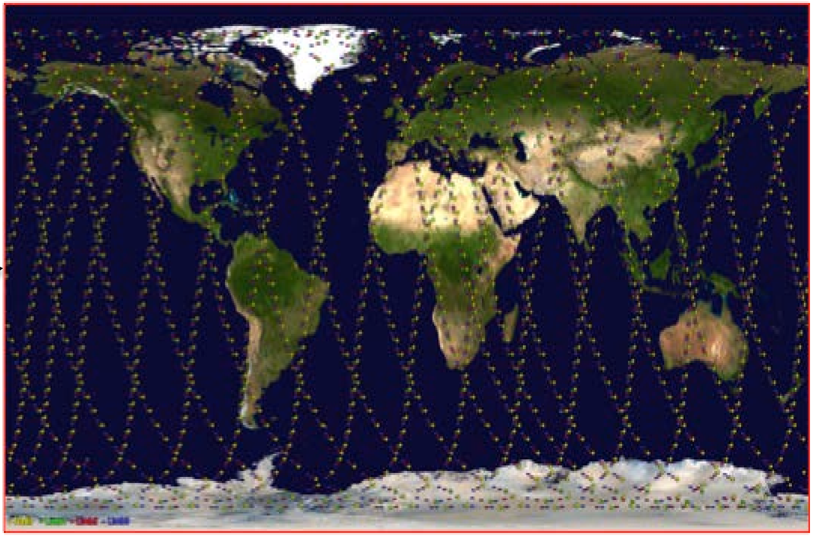
Outline

- Using TES NH_3 data, along with surface and aircraft data, to investigate NH_3 emissions during 2010 CalNex Campaign.

- Using the Cross-Track Infrared Sounder (CrIS) to investigate NH_3 sources in California and Southeast US.

Why switch to CrIS?

- TES is past its design lifetime, taking little new data, and has low spatial coverage
- **CrIS could monitor global NH_3 with high spatial coverage for many more years (>2022)**



	TES	CrIS
Satellite	AURA	NPP
Available Data	July 2004-present	October 2011-present
Resolution	0.06 cm^{-1}	0.625 cm^{-1}
Footprint	5x8 km rectangle	14 km diameter circle
Repeat cycle	Once every 16 days	Daily
Equatorial crossing	1:30 am and 1:30 pm	1:30 am and 1:30 pm
Noise in NH_3 window	0.09 – 0.12 K	0.03 – 0.06 K

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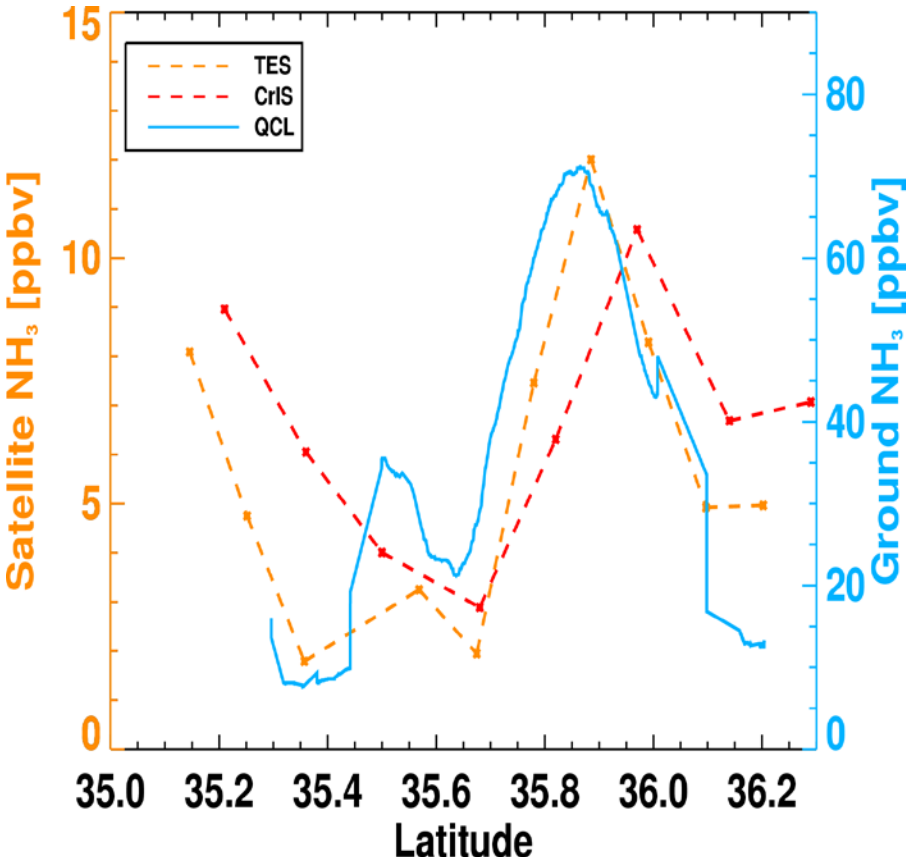
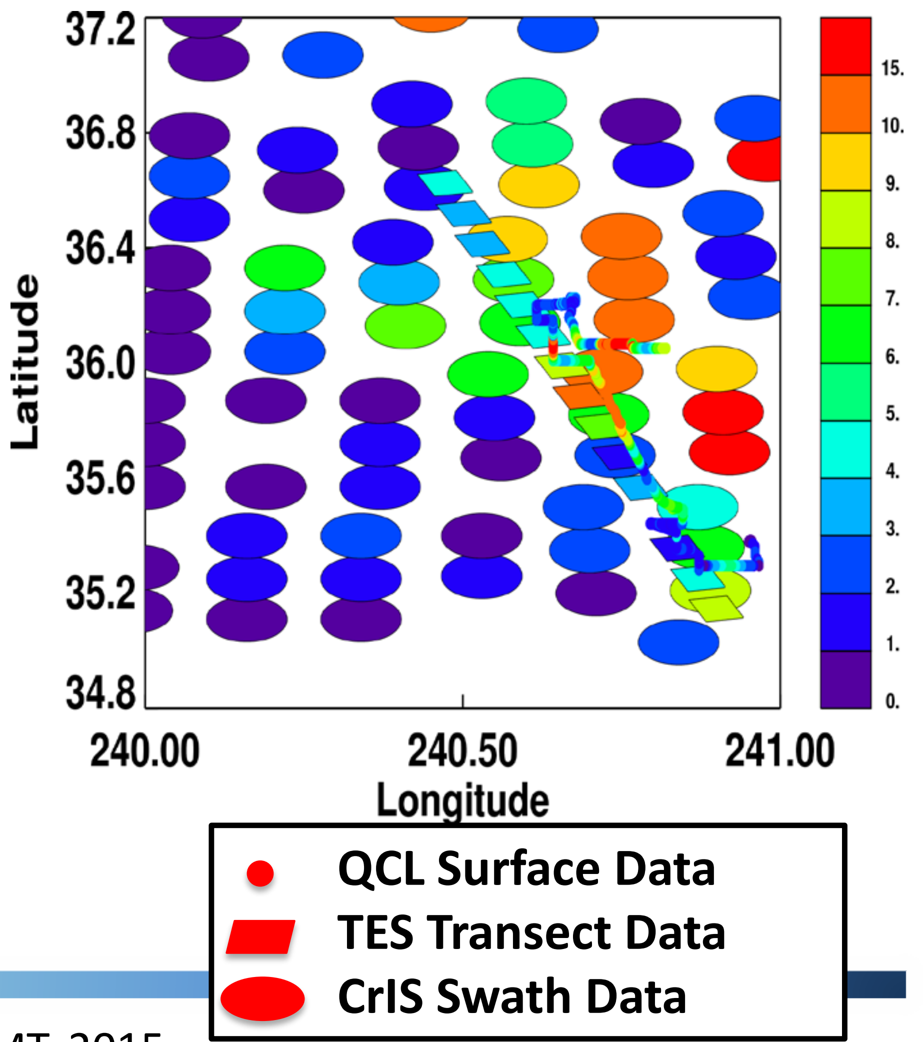
TES and CrIS versus surface NH_3

- QCL directly under TES transect in the San Joaquin Valley on January 28, 2013





Shephard and Cady-Pereira, AMT, 2015.

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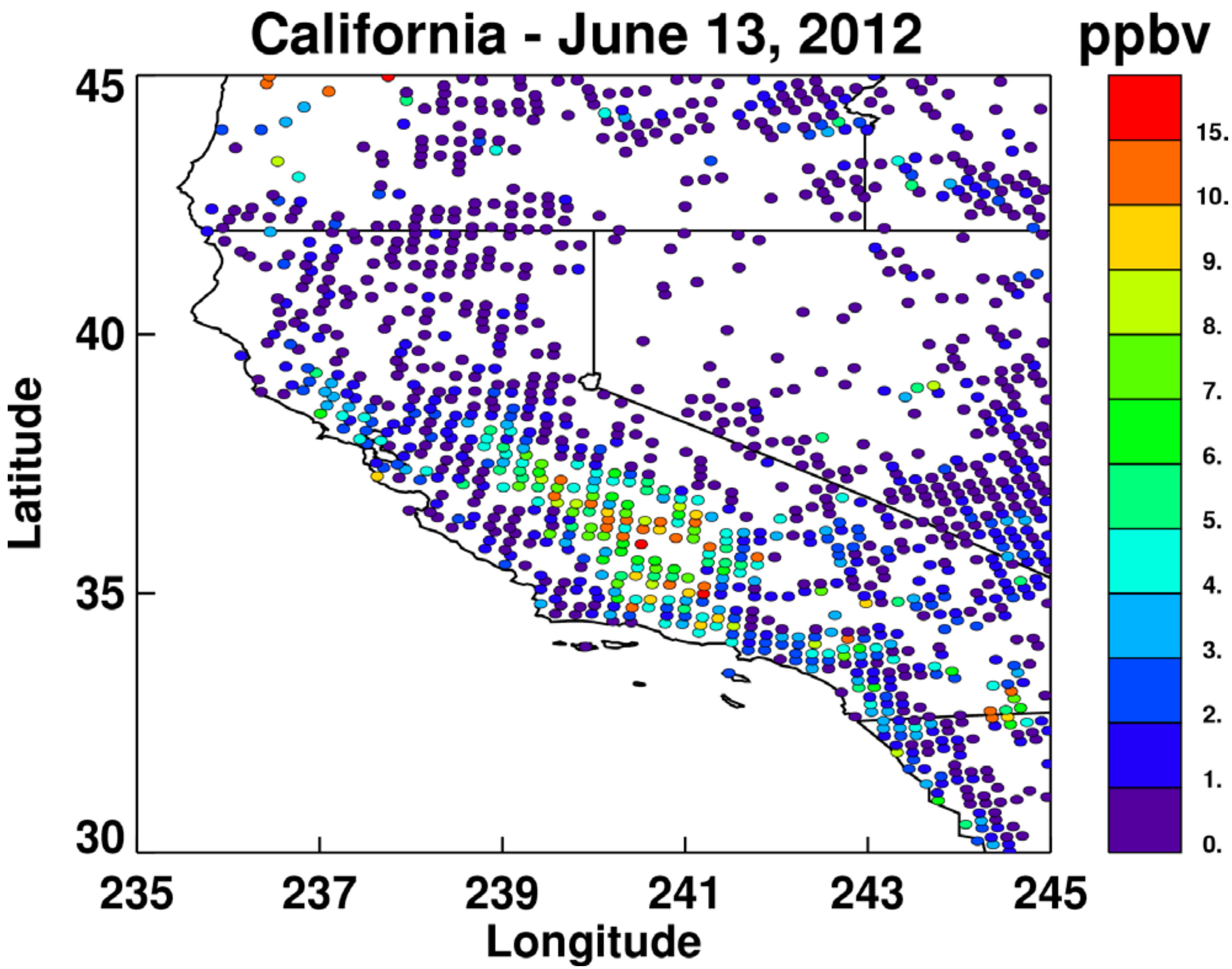
Application of CrIS NH_3 to California



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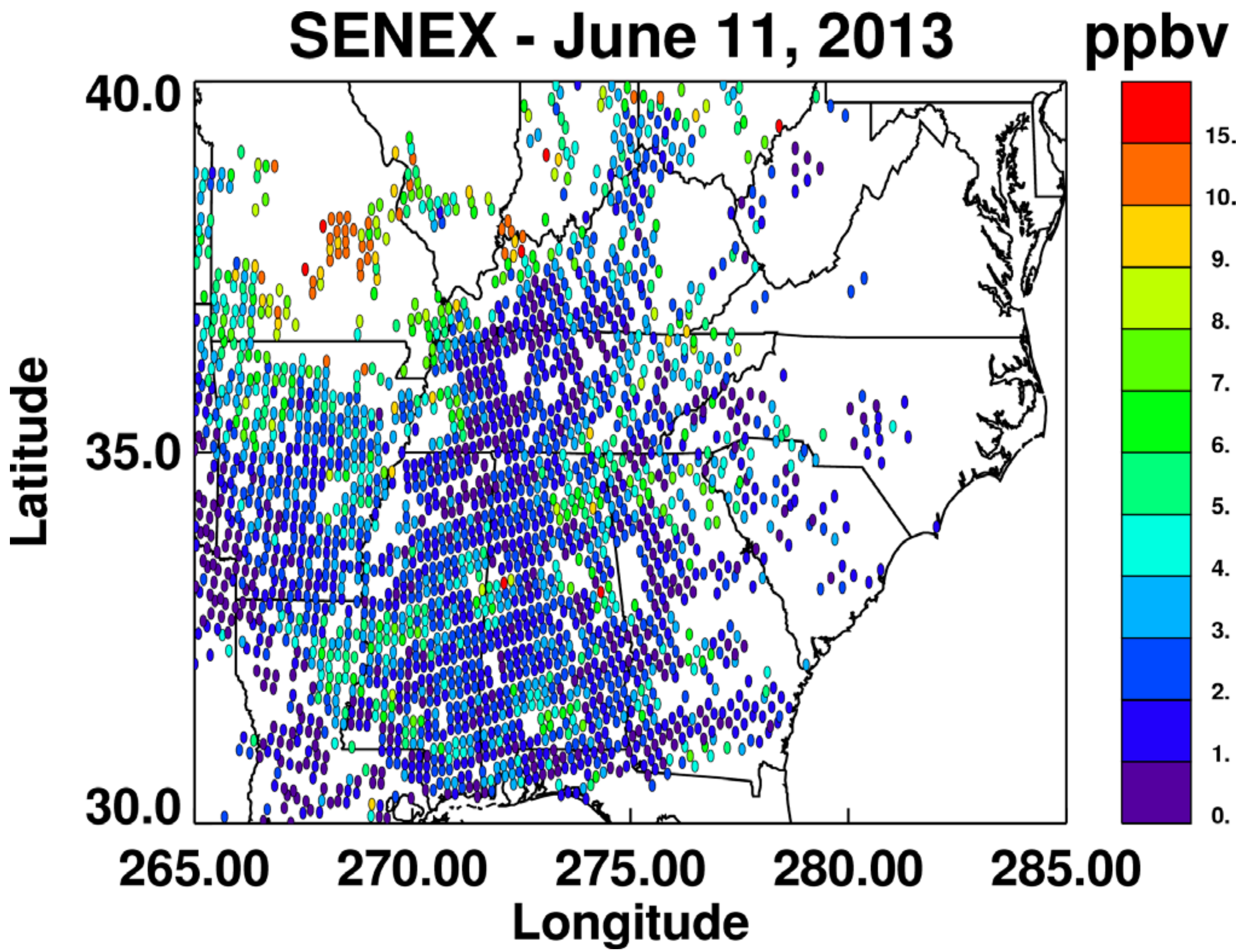
Application of CrIS NH_3 to SENEX



Summary

- Satellite, surface, and aircraft data all suggest diurnally constant NH_3 emissions in CARB CalNex inventory for California Central Valley are likely incorrect.
- For LA, surface observations suggest CARB estimates of industrial NH_3 emissions are either too high or are more constant through the day, but aircraft observations give conflicting information.
- The CrIS satellite instrument can detect NH_3 as well as TES, but has much greater spatial coverage, providing much more data for model evaluation.

Future Work

- Use CMAQ Adjoint, along with CrIS, CalNex, and SENEX data, to constrain NH_3 emissions in California and Eastern US.

Acknowledgements

- Juliet Zhu, Colorado State University
- John Walker and Robert Pinder, EPA
- Armin Wisthaler, University of Innsbruck
- Kang Sun and Mark Zondlo, Princeton
- Amy Jo Scarino, NASA Langley
- Jeremy Avise and Leo Ramirez, CARB
- Ming Luo, Jet Propulsion Laboratory, California Institute of Technology
- NOAA CPO AC4 Program Grant Numbers NA13OAR4310060 and NA14OAR4310129
- TES Science Team
- NASA Suomi-NPP Science Team

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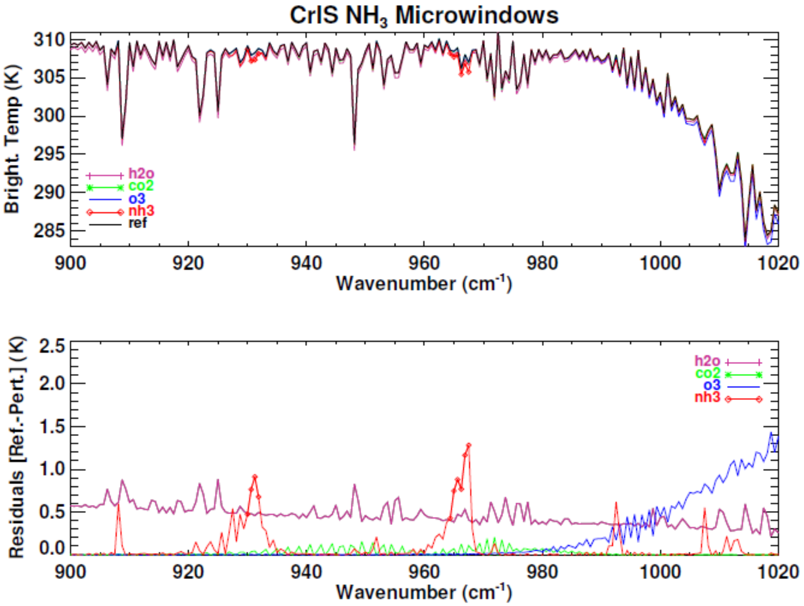
BACKUP SLIDES

CrIS microwindows and constraints

- Lower spectral resolution of CrIS required different microwindows.
- *A priori* and constraints from TES (Shephard et al., 2011)
  - Polluted, Moderately polluted, and Unpolluted profiles
- *A priori* selected based on signal to noise ratio (SNR) and thermal contrast



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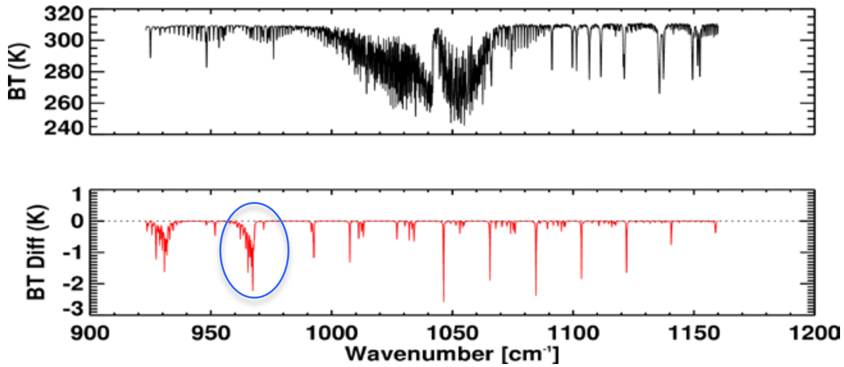
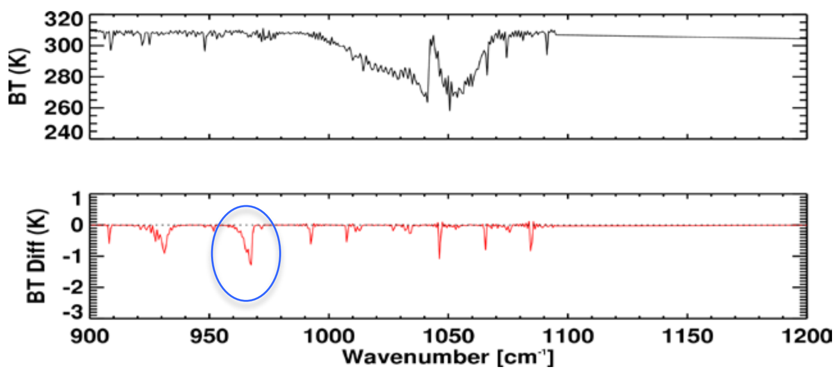
NH_3 signal from TES and CrIS

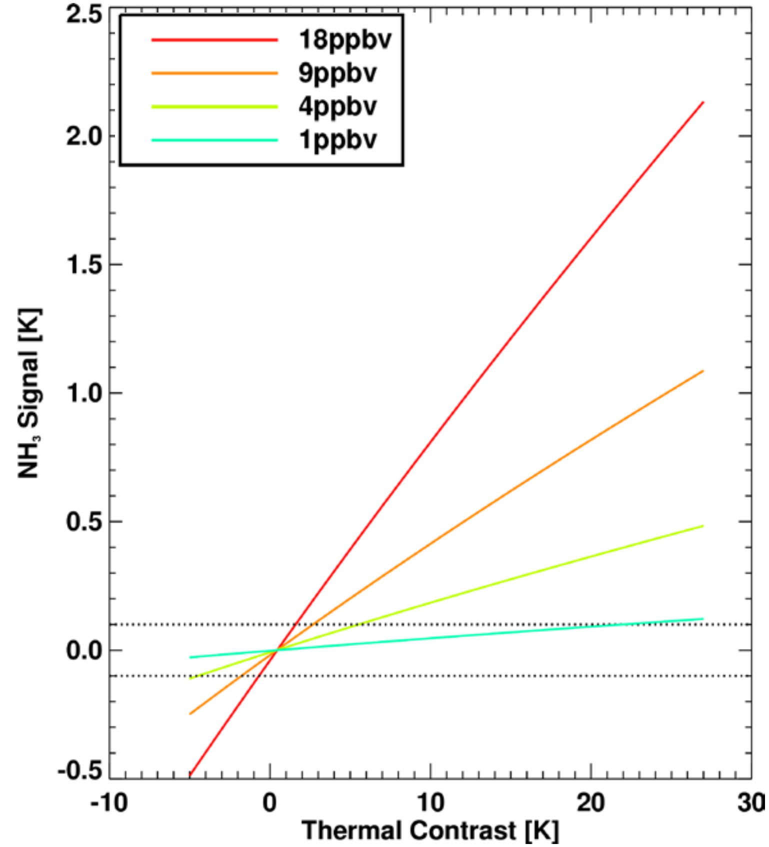
Simulated spectra and NH_3 signal
18 ppbv at surface

TES



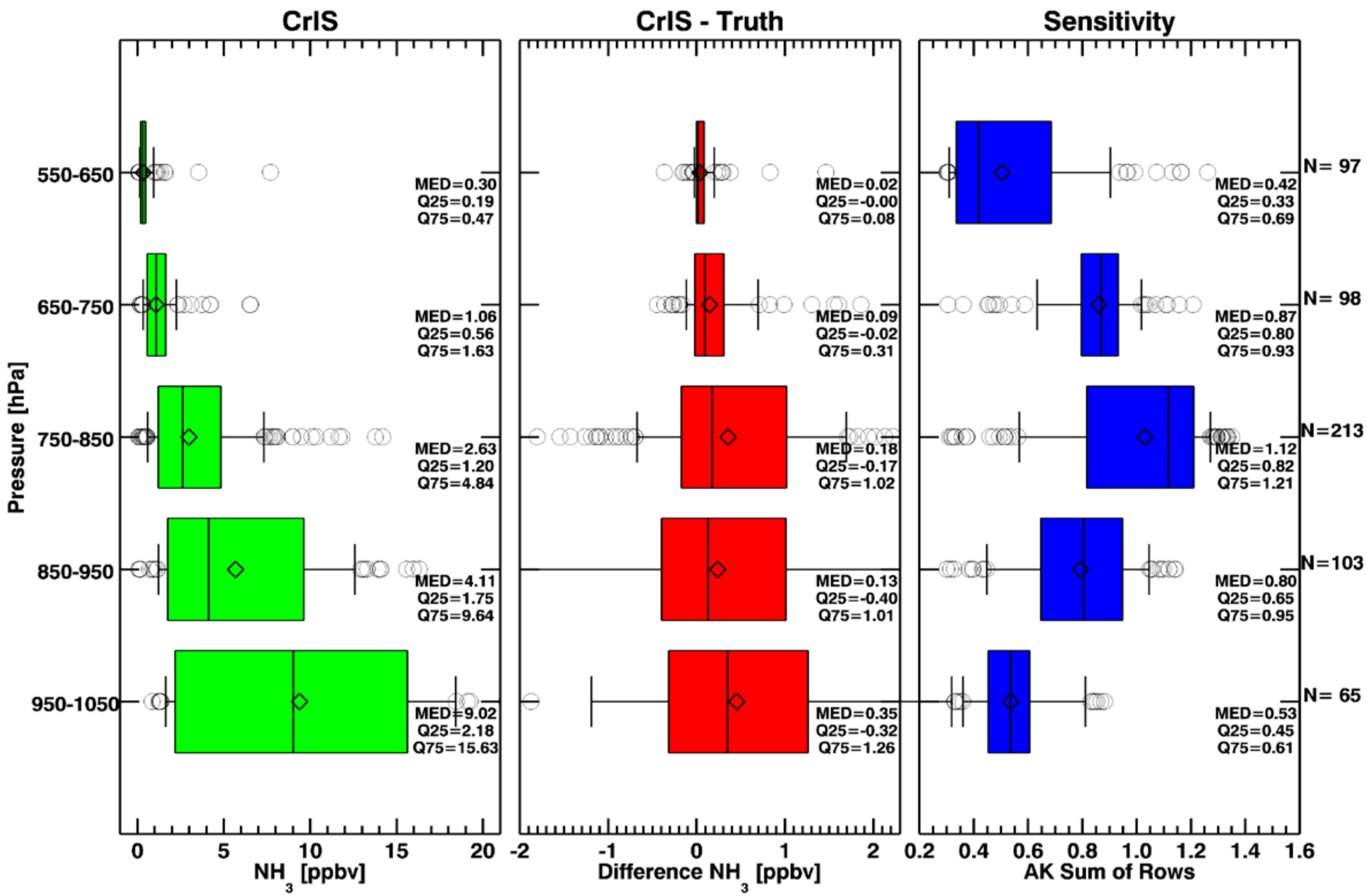
CrIS





- Detectability is ~1 ppbv under ideal conditions
- But thermal contrast also plays a role

CrIS NH_3 Retrieval: Simulated Spectra



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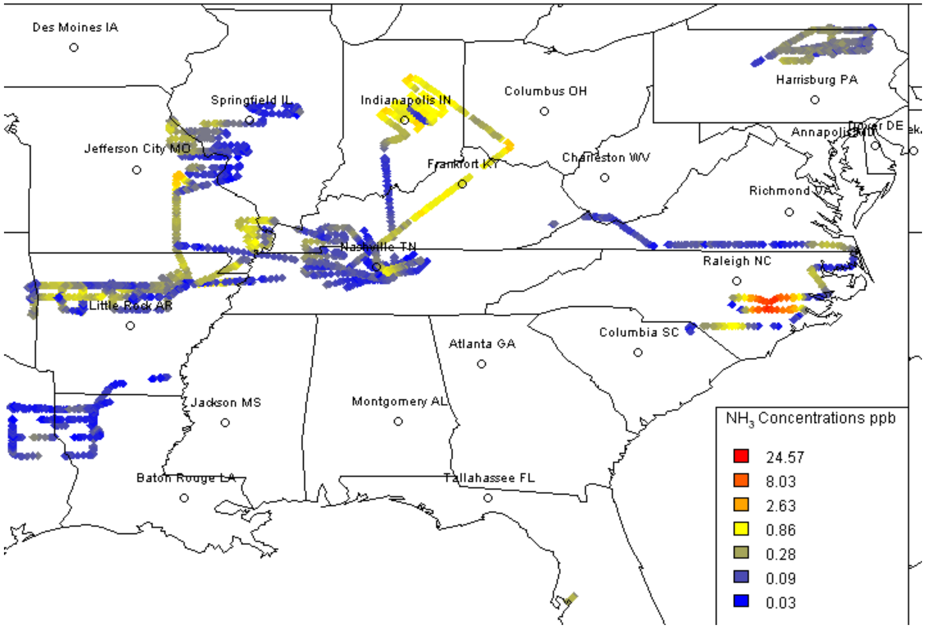
Future Work on CrIS NH_3 Retrieval

- Validate against SENEX, FRAPPE, and other field NH_3 measurements.
- Use CMAQ adjoint to test ability of CrIS to optimize NH_3 emissions.
- Deliver CrIS NH_3 retrieval algorithm to NASA

SENEX Flights with NH_3 Measurements



NH_3 mixing ratios (ppbv) measured by the NOAA WP-3 aircraft during SENEX 2013. (Figure courtesy of Jesse Bash, US EPA NERL.)

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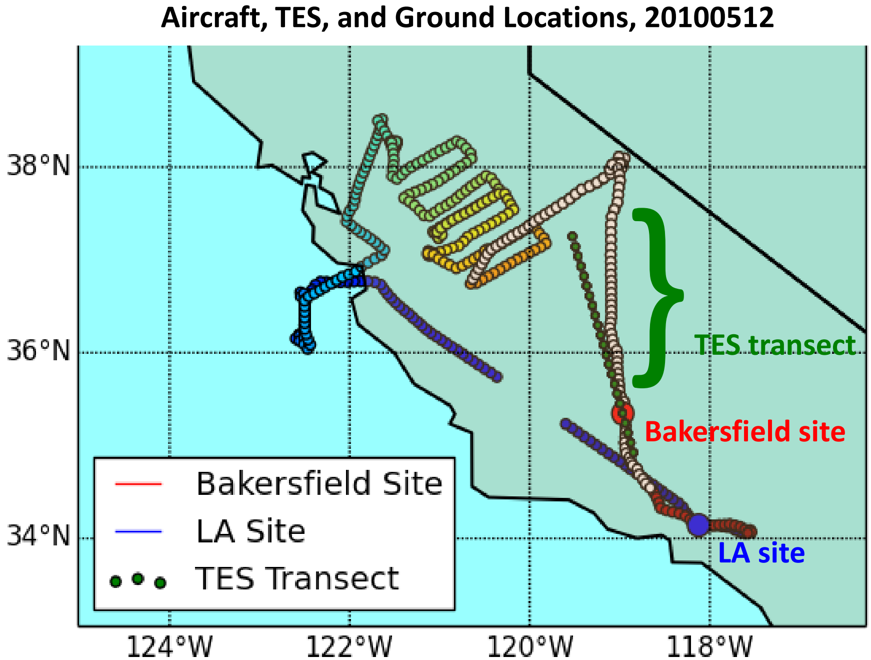
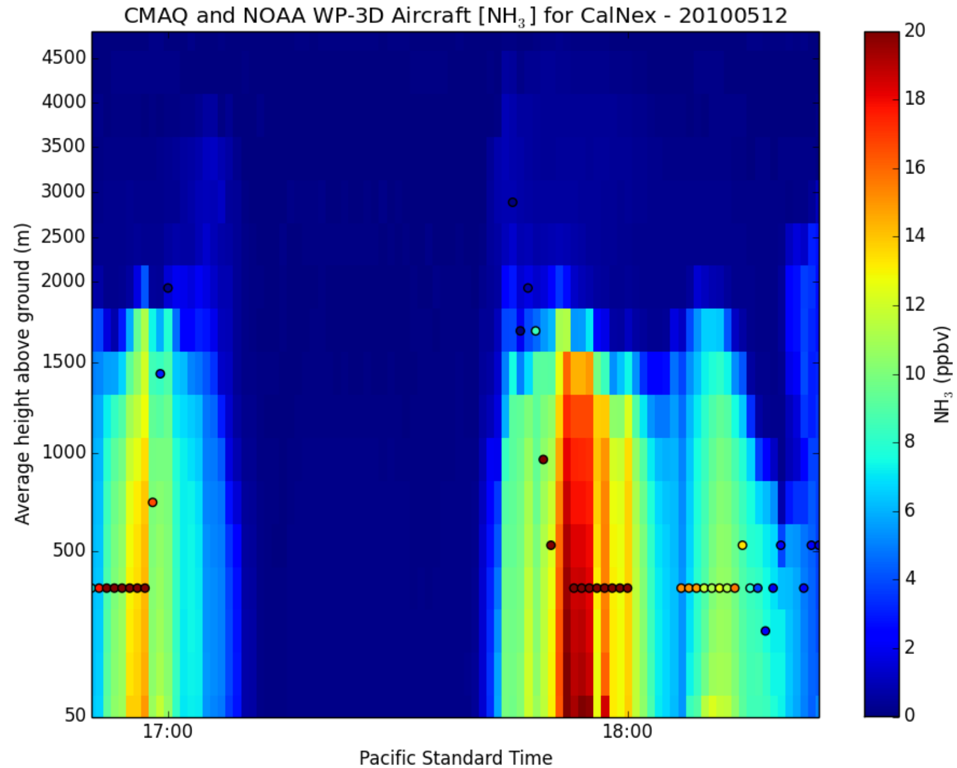
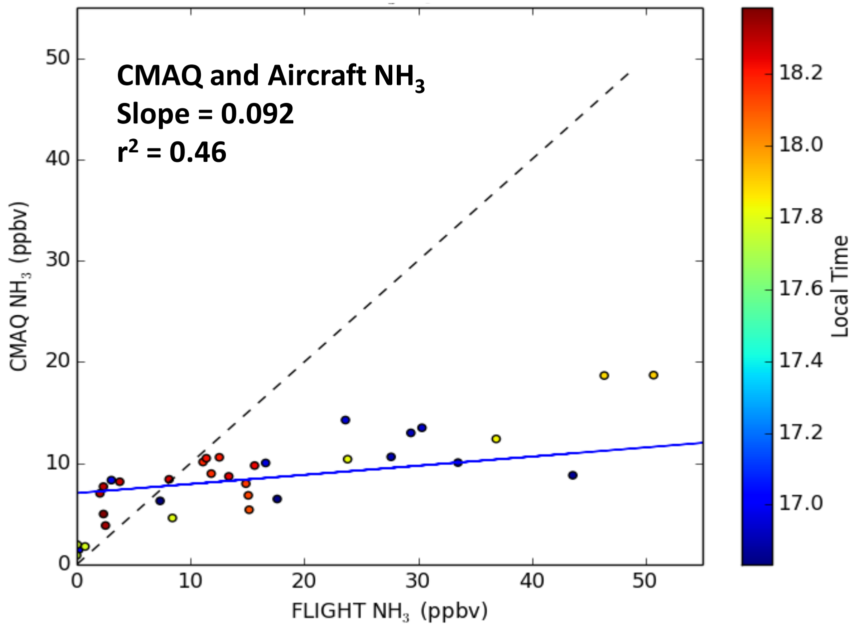
CMAQ – CALNEX Aircraft Comparison

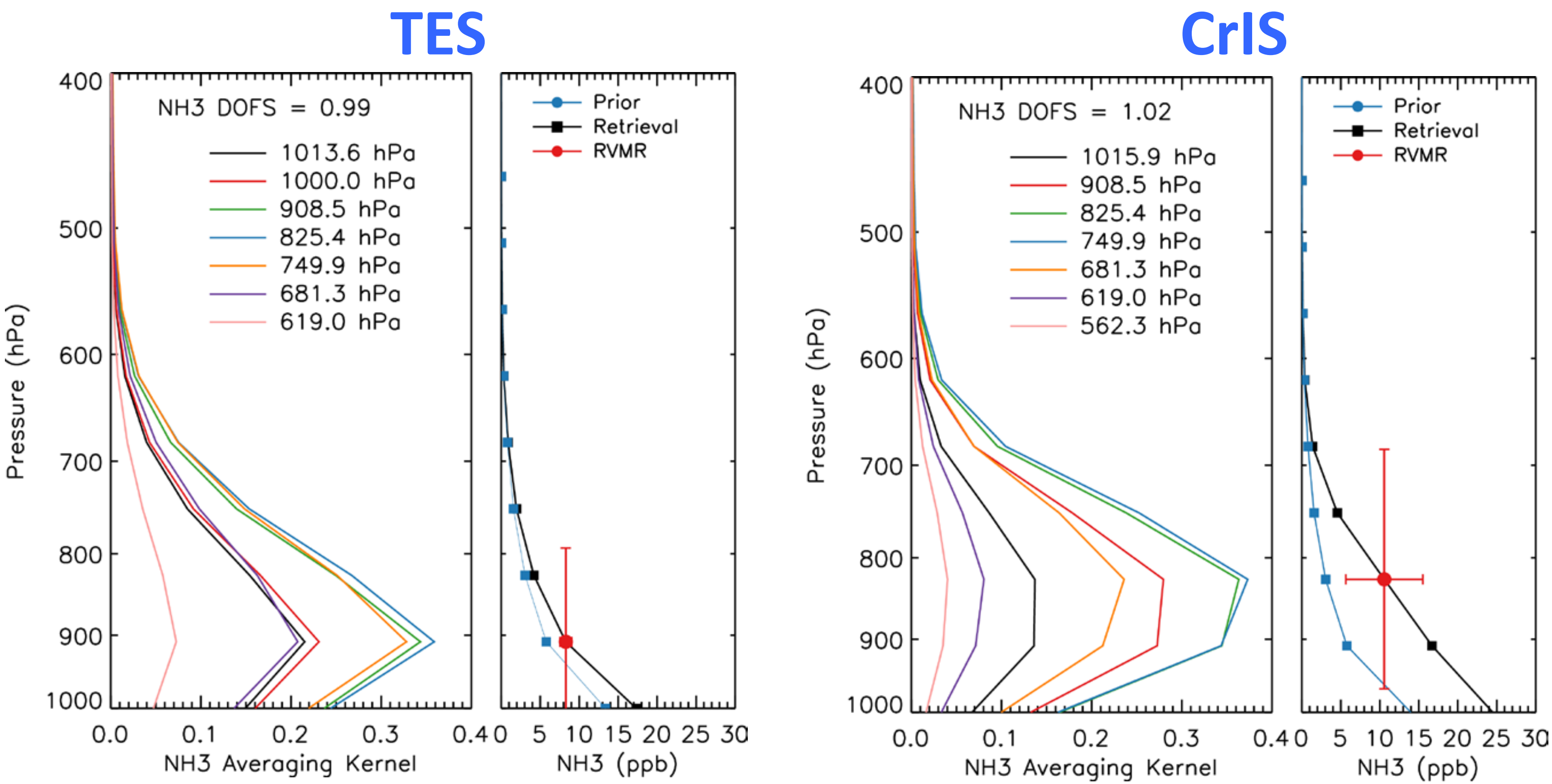






- CMAQ with CARB emission inventory generally captures the locations of the NH_3 plumes observed by aircraft.
- But absolute concentrations too low.

TES and CrIS Sensitivity to NH_3



- Both instruments most sensitive to NH_3 between 950 and 600 mbar
- TES is more sensitive to amounts lower in the atmosphere
- 1 piece of information or less: DOFS<1.0
- Collapse all information to a single point: RVMR
    - Easier to compare with *in situ* measurements, models and other instruments