Ammonia (NH_3) Distributions and Recent Trends by 13-year AIRS Measurements

J. X. Warner[1], Z. Wei[1], L. L. Strow[2], R. R. Dickerson[1], J. B. Nowak[3]
Y. Wang[4], Q. Liang[5, 6]

[1] Dept. of Atmospheric and Oceanic Science, UMCP, College Park, MD, U.S.A.
[2] Dept. of Physics and JCET, UMBC, Baltimore, MD, U.S.A.
[3] Aerodyne Research, INC
[4] Department of Earth and Atmospheric Sciences, University of Houston
[5] NASA Goddard Space Flight Center, Atmospheric Chemistry and Dynamics, Greenbelt, MD, U.S.A.
[6] Universities Space Research Association, GESTAR, Columbia, MD, U.S.A.

*Published in ACP: **The Global Tropospheric Ammonia Distribution as seen in the 13-year AIRS Measurement Record**

*Ready to Submit: **Increases in Atmospheric Ammonia over Major Agricultural Areas from Space Measurements**

* Funded by NASA's The Science of Terra and Aqua Program (NNX11AG39G), and the Atmospheric Composition Program (NNX07AM45G).

Why Ammonia

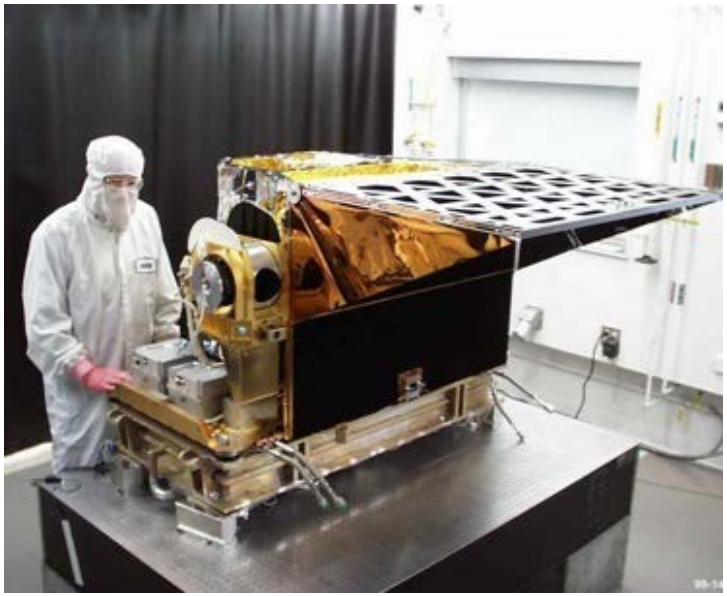
- Ammonia (NH_3) plays an increasingly important role in the global biogeochemical cycle of reactive nitrogen as well as in aerosol formation and climate.

- Measurements with daily and large global coverage are challenging and have been lacking partly because the lifetime of NH_3 is relatively short and partly because it requires high sensitivity for the retrievals that can be only obtained from areas with high thermal contrasts near the surface (Clarisse *et al.*, 2010).

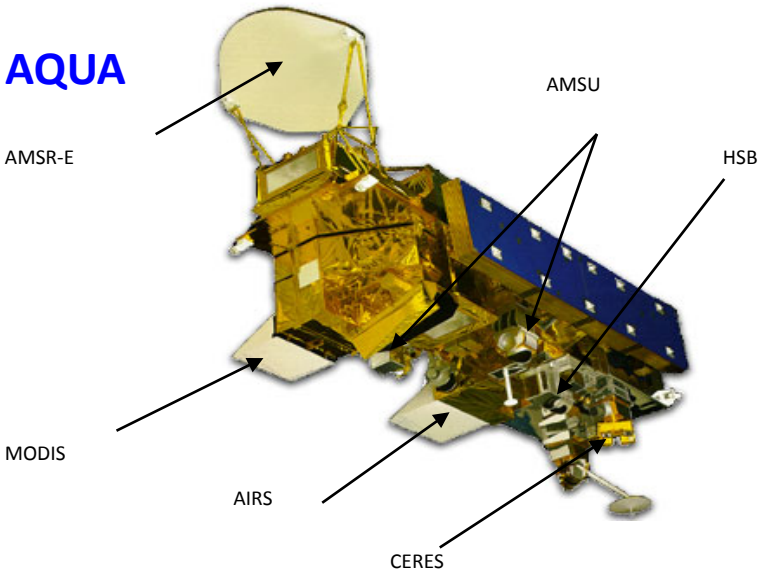
- AIRS afternoon overpasses (1:30pm) are best correlated with the daily emission peak time and during the daily period with the highest thermal contrast. Additionally, AIRS large coverage with wide swaths and cloud-clearing provide daily NH_3 maps. The 13-year data records makes AIRS the best sensor for NH_3 trends and variability studies (to date).

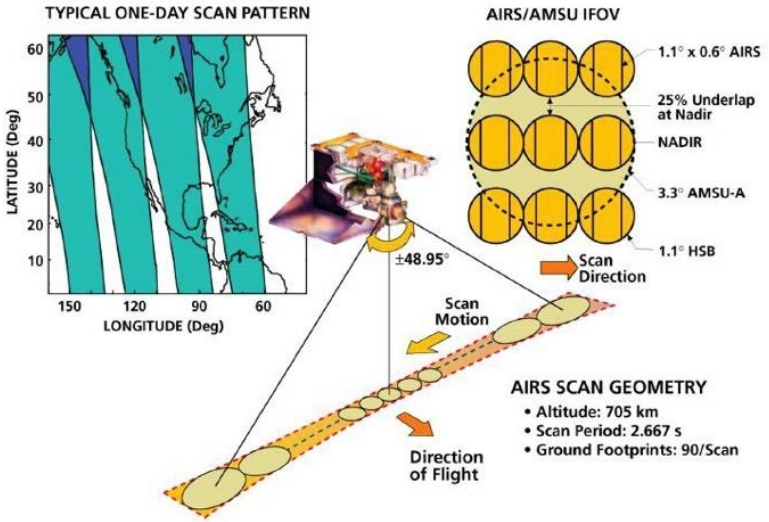
Atmospheric Infrared Sounder

Launched May 2002



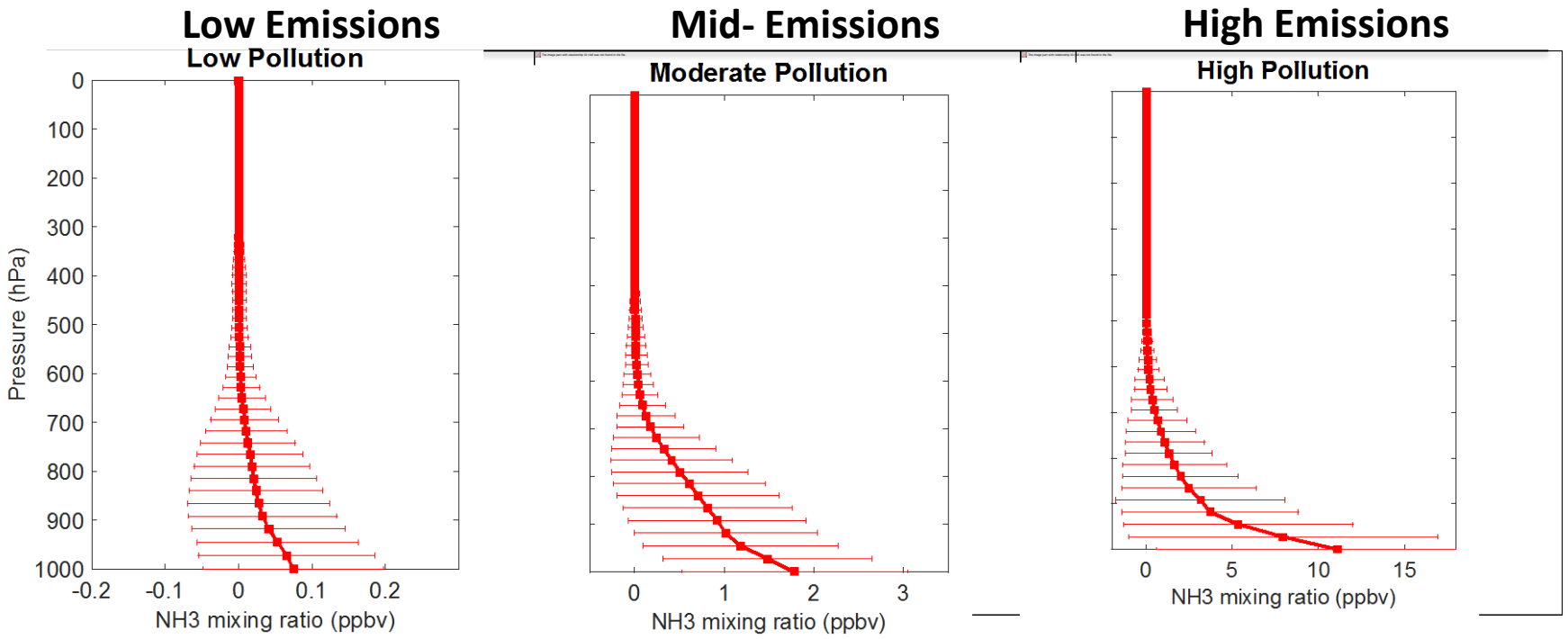
- A grating spectrometer originally designed to improve weather forecast and now also used for climate and air quality studies.
- Spectral resolution at □/1200 (~ 0.5 cm^{-1})
- Covers 650-2665 in three bands with a total of 2378 channels
- Spatial resolution 13.5 km^2 (with retrievals at ~45 km^2)
- Wide swaths and cloud clearing provide daily global coverage
- Very high Signal-to-Noise accuracies of 1K over 1 km-layer.





AIRS NH_3 Algorithm

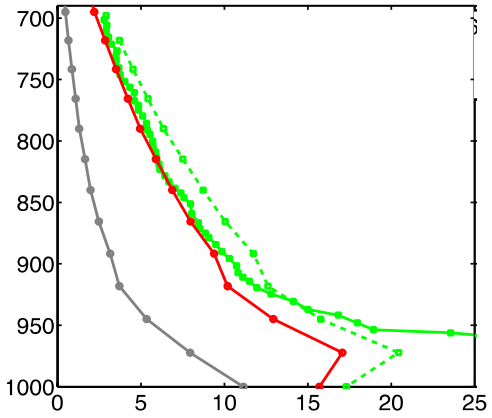
- AIRS NH3 retrievals use Optimal Estimation (OE) technique (Rodgers, 2000);
- CCRs and SARTA are used as in AIRS algorithm for other species;
- The *a priori* levels are computed from GEOS-chem;
- Globally one set *a priori*.
- Select *a priori* levels based on brightness temperature differences weighted by noise (dbti);



Validation vs CRDS/Picarro in DISCOVER-AQ CA

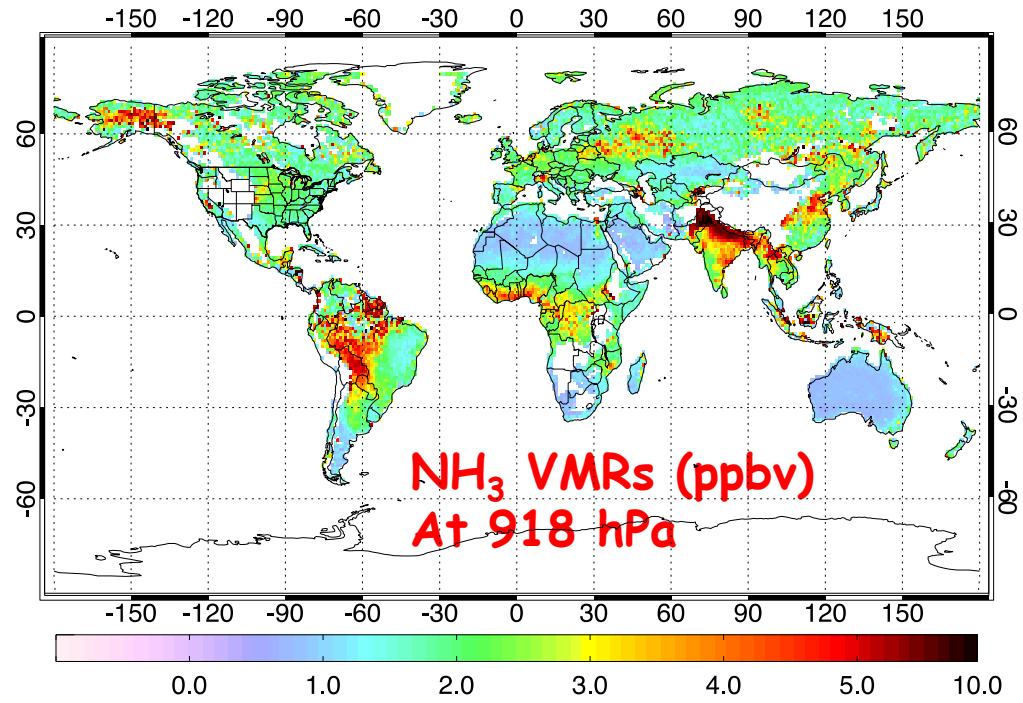
Spiral Profiles Only - 01/16 to 02/06, 2013

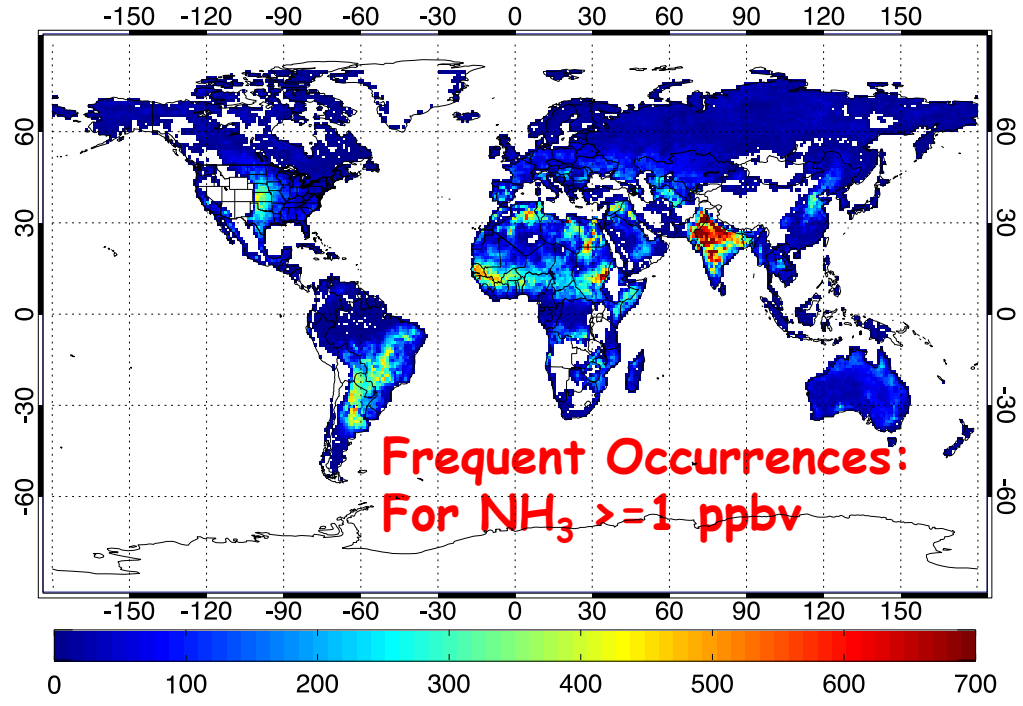
CRDS/Picarro data courtesy of Co-author J. Nowak



m, –0.469h

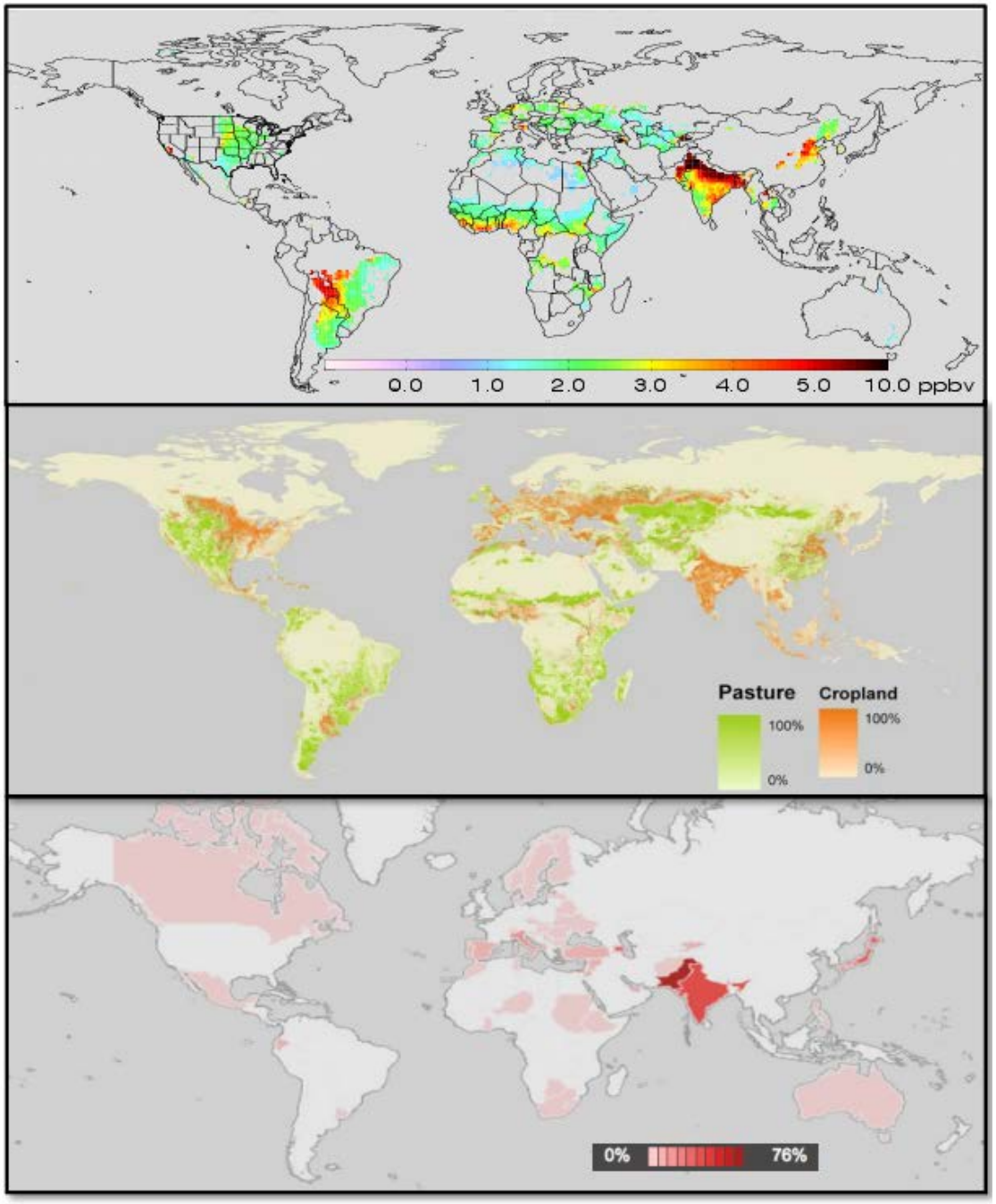
- Gray – a priori; Red – retrievals; Green solid – in situ; and Green dashed – convolved in situ.
- AIRS L2 pixel sizes are ~45 km^2, can coincide with multiple in situ profiles.
- AIRS NH_3 measurements are most sensitive at 850-950 hPa layer.





Global NH_3 in 2002-2015

- AIRS NH_3 at 918 hPa for daytime and land only averaged over Sept. 2002 to Aug. 2015;
- Use Q0; DOFS ≥ 0.1;
- High concentrations are mainly due to human activities and fires;
- Use occurrences of higher emissions (lower) to distinguish between the two major sources: agricultural (high VMRs & high frequencies); BB emissions (high VMRs & low frequencies);
- Sources are seen in valleys (e.g., San Joaquin Valley, California in the U.S., the Po Valley, Italy, Fergana Valley, Uzbekistan, and the Sichuan Basin in China); Agricultural especially in irrigated lands (e.g., Azerbaijan, Nile Delta and near Nile River in Egypt, the Mid-West U.S., in the Netherlands, in Mozambique and Ethiopia, Africa, and especially the Indo-Gangetic Plain of South Asia).

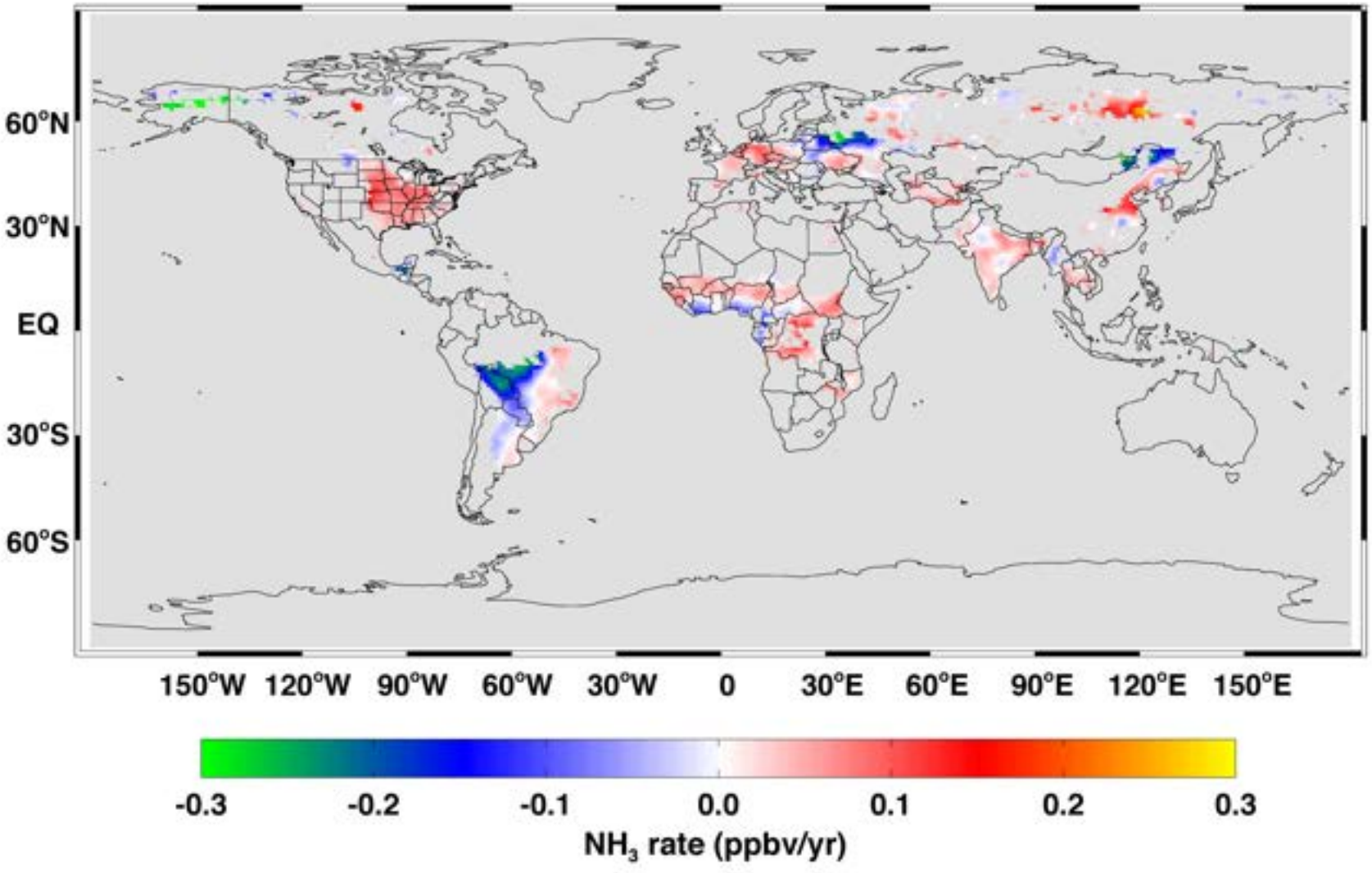


Top panel: The NH_3 VMRs from the persistent sources filtered with the collocated occurrences of elevated emissions (≥ 1.4 ppbv) using a threshold of 40 days;

Middle panel: Pasture and Cropland Map

Bottom panel: irrigated agricultural land areas.

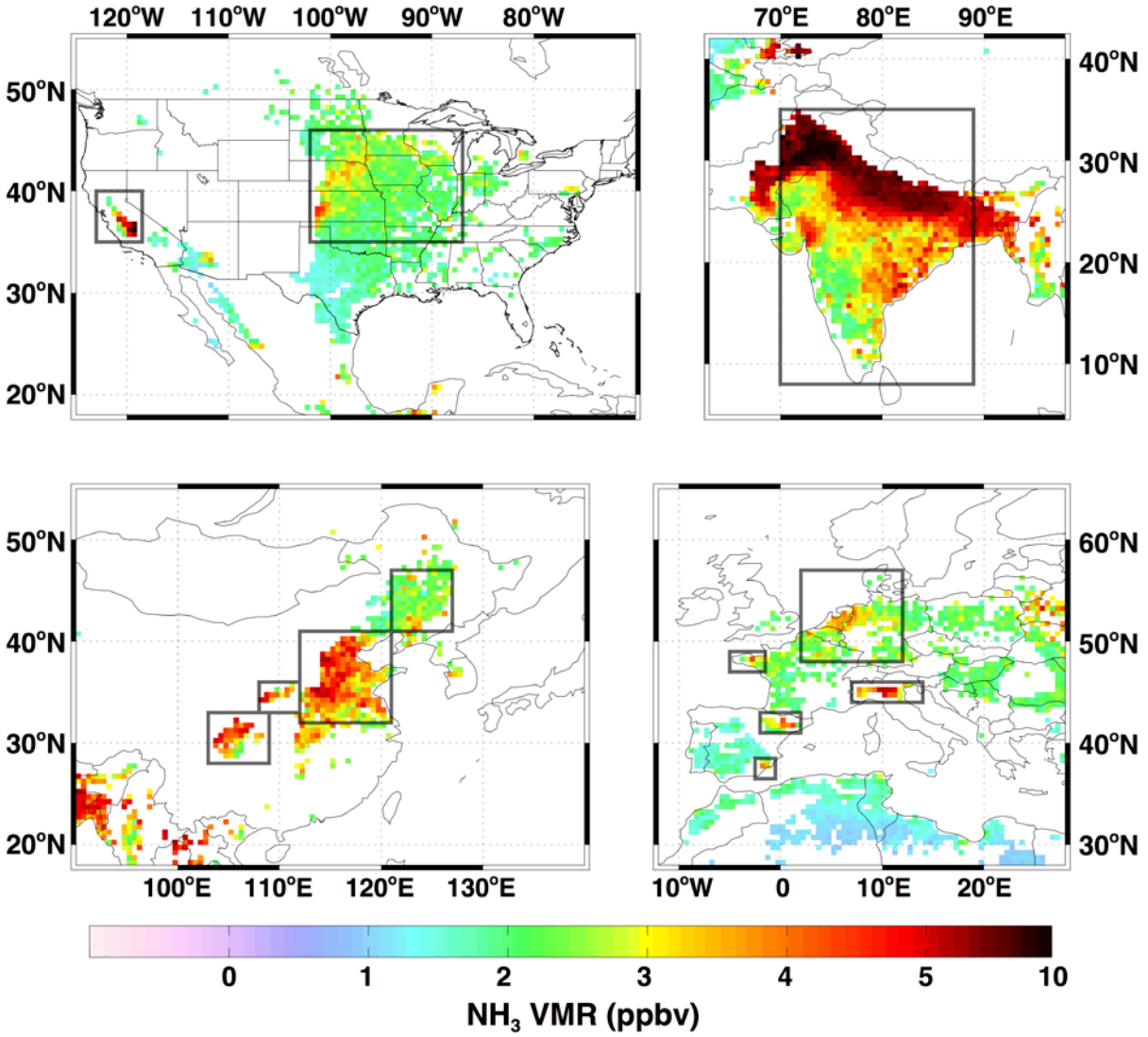
NH_3 Trends - Last 13 years



- Concentrations of anthropogenic emissions increased and BB decreased
- Trends due to BB are not conclusive due to the short record.

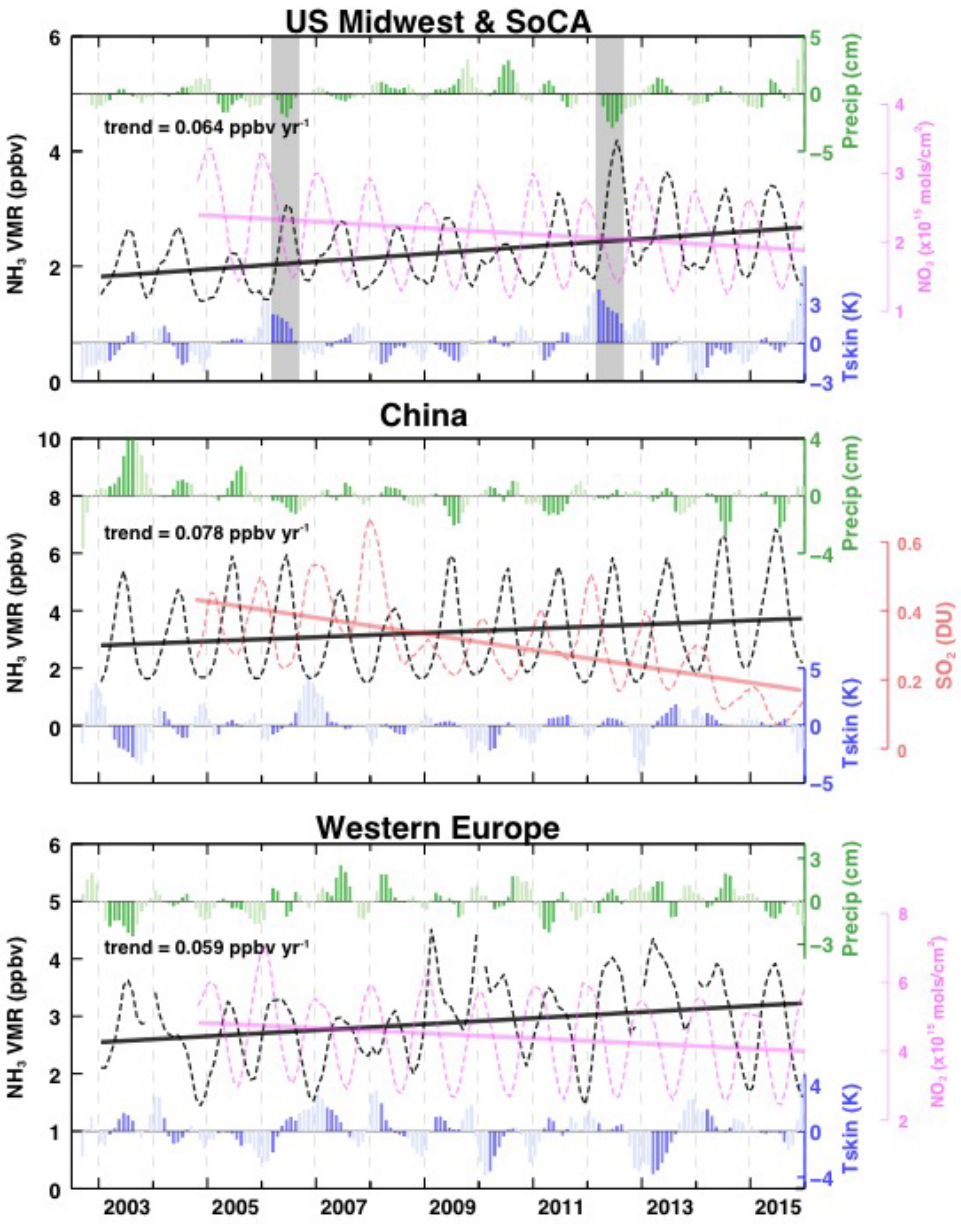
NH_3 over USA, China, India, and Europe

Using high concentration and high frequencies



Black boxes are regions used for follow up trend studies.

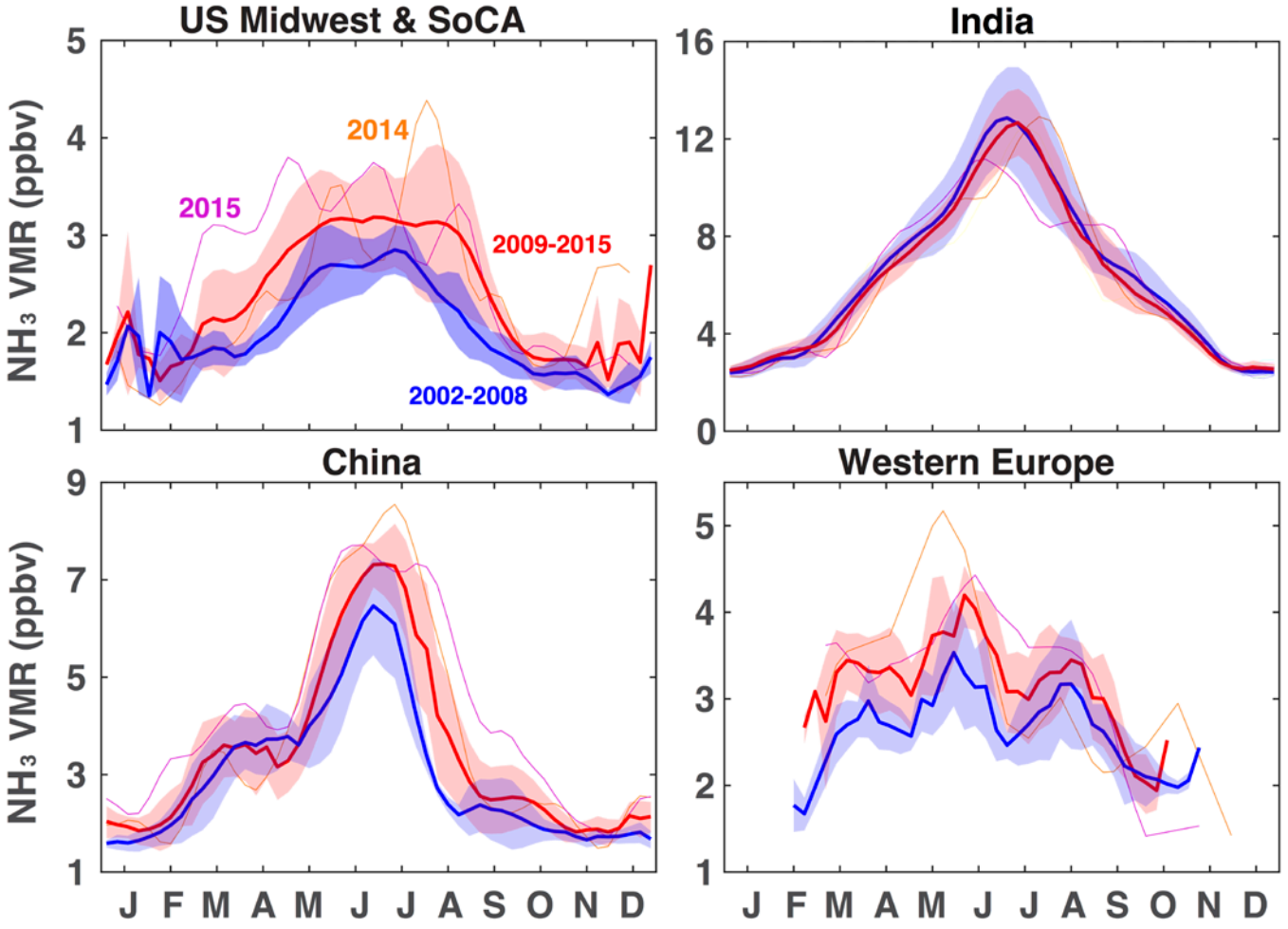
AIRS NH_3 vs OMI NO_2 for US (top), SO_2 for China (middle) and NO_2 for Western Europe (lower)



- All 3 regions show increasing NH_3 trends in the last 13 years, in black.
- Decreased SO_2 from OMI largely explains the reason of NH_3 increases in Midwest U.S. (not shown), China, and Europe (not shown).
- OMI NO_2 decreasing explains winter NH_3 increasing over the US and Europe.
- Meteorological conditions also affect NH_3 concentrations (high surface temperatures and low precipitation), see top panel shaded areas.

AIRS NH_3 Seasonal Variation

- over USA, China, Europe, and India

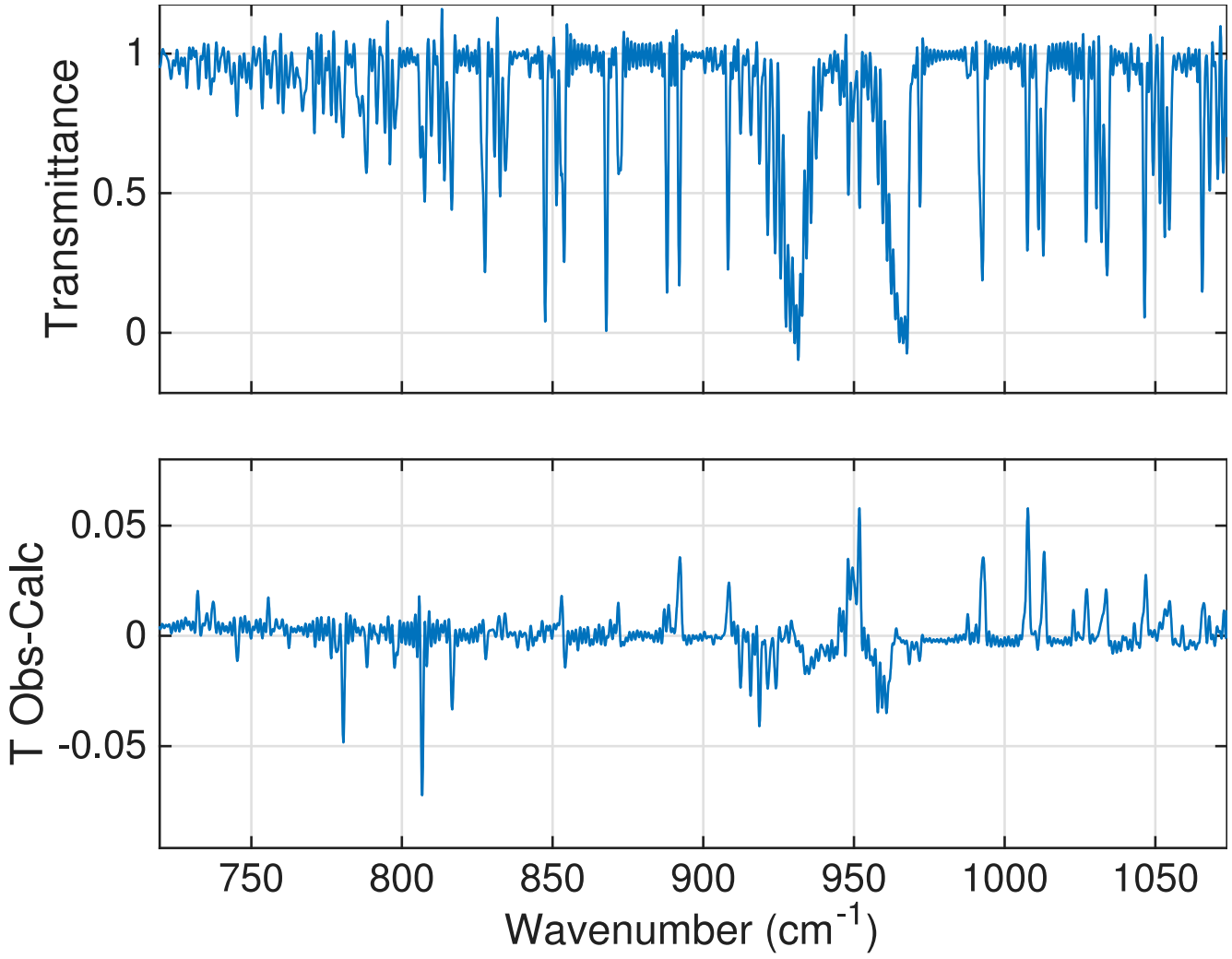


- The highest NH_3 concentrations in average occur in India/Pakistan, and China. Note scales.
- NH_3 in India seasonal variation are broad and no obvious increasing/decreasing trends;
- NH_3 for USA, China and Europe have increased, with peaks in both spring and summer;
- Clear increasing trends over US Midwest, China, and Western Europe.

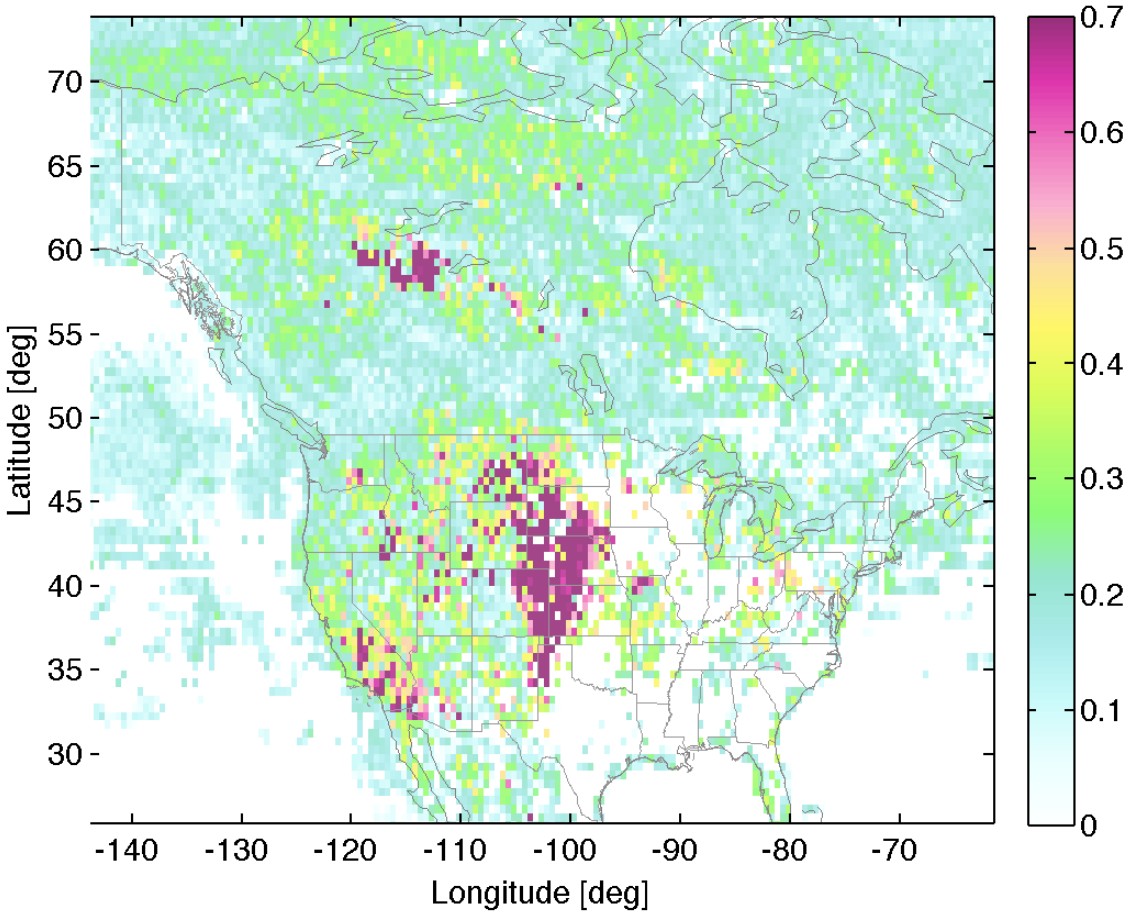
Summary

- AIRS NH_3 products not only include 13 years data record, it also provide daily maps!
- AIRS retrieved vertical profiles show good agreement (~5 – 15%) with in situ profiles from the 2013 DISCOVER-AQ field campaign in central California.
- AIRS daily measurements captures the strong continuous NH_3 emission sources from the anthropogenic (agricultural) source regions, as well as emissions from biomass burning (BB).
- Ammonia trends increase over agriculture regions, where fertilizers are used as routine practice, decrease over BB regions (with insufficient records).
- Ammonia increases resulted primarily from dramatic decreases in concentrations of acidic aerosols (sulfate and nitrate), an unintended consequence of effective controls of NO_x and SO_2 emissions.

CrIS NH3 (TVAC Gas Cell)

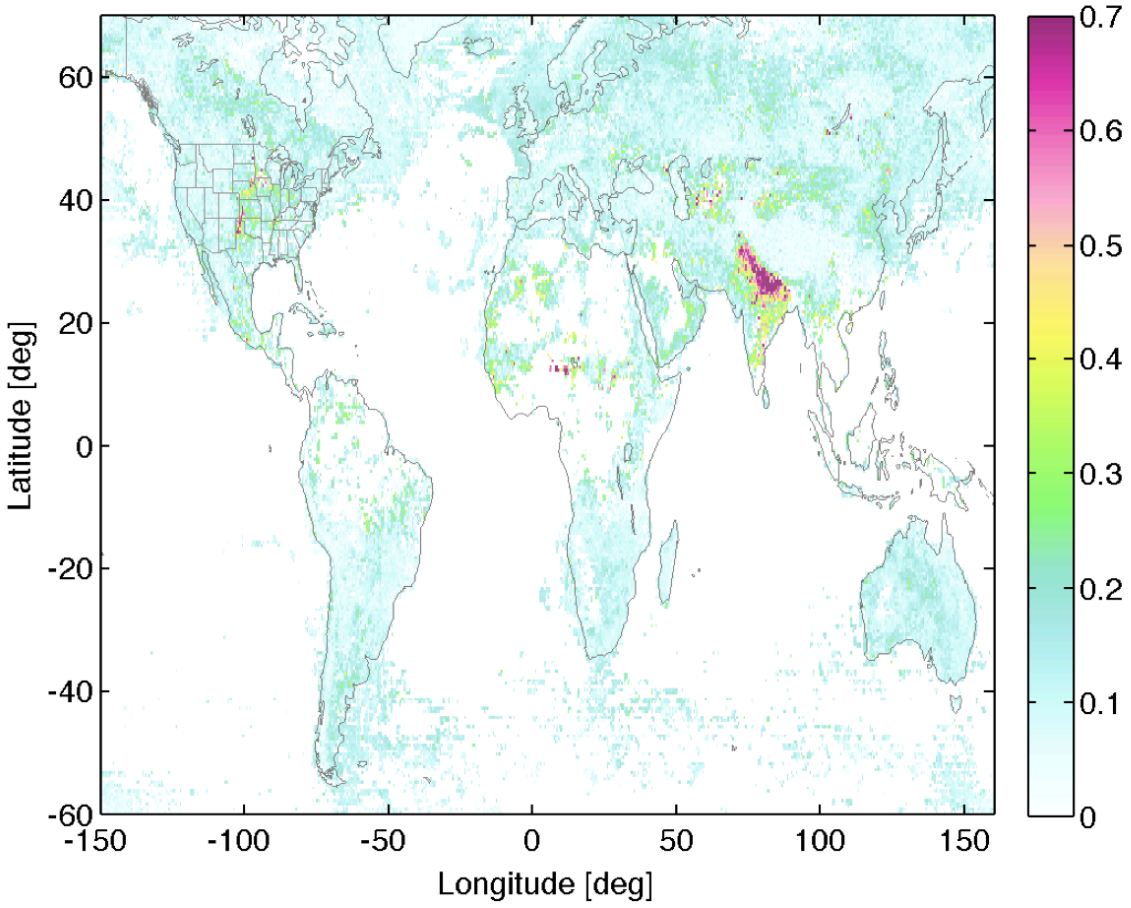


CrIS NH3 B(T) Signals (on/off line)



July 2012

CrIS NH3 B(T) Signals



May 2012