

Ammonia (NH₃) Distributions and Recent Trends by 13-year AIRS Measurements

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- In Print at ACP: **The Global Tropospheric Ammonia Distribution as seen in the 13-year AIRS Measurement Record**
- In Prep: **Recent Trends in the Global Tropospheric Ammonia in the 13-year AIRS Measurement Record**
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Why Ammonia

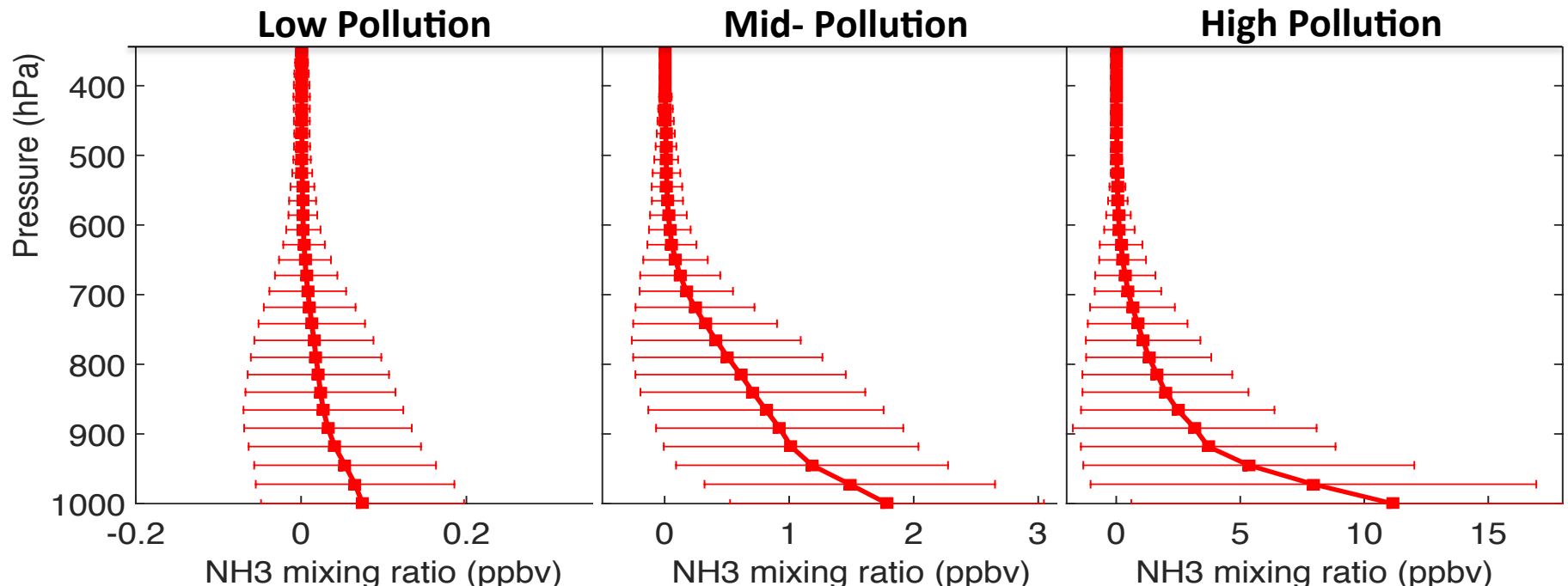
- Ammonia (NH_3) plays an increasingly important role in the global biogeochemical cycle of reactive nitrogen as well as in aerosol formation and climate.

Why AIRS

- Measurements with daily and large global coverage are challenging and have been lacking before the recent satellites of IASI, TES, and AIRS, 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).

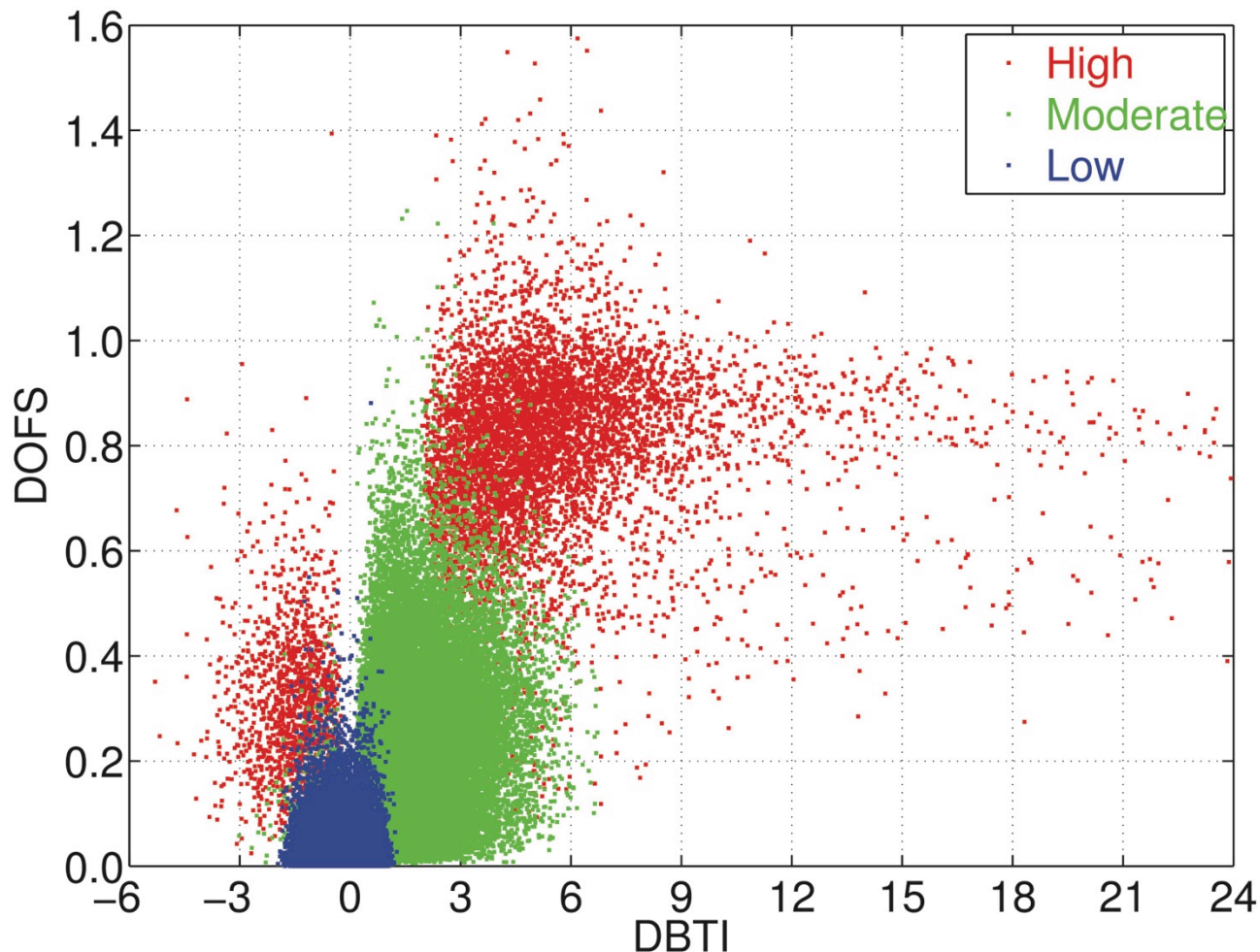
AIRS NH₃ Algorithm - I

- AIRS NH₃ retrievals use Optimal Estimation (OE) technique (Rodgers, 2000);
- CCRs and SARTA are used as in AIRS algorithm for other species;
- Globally one set *a priori* profiles;
- The *a priori* levels are computed from GEOS-Chem;



AIRS NH₃ Algorithm - II

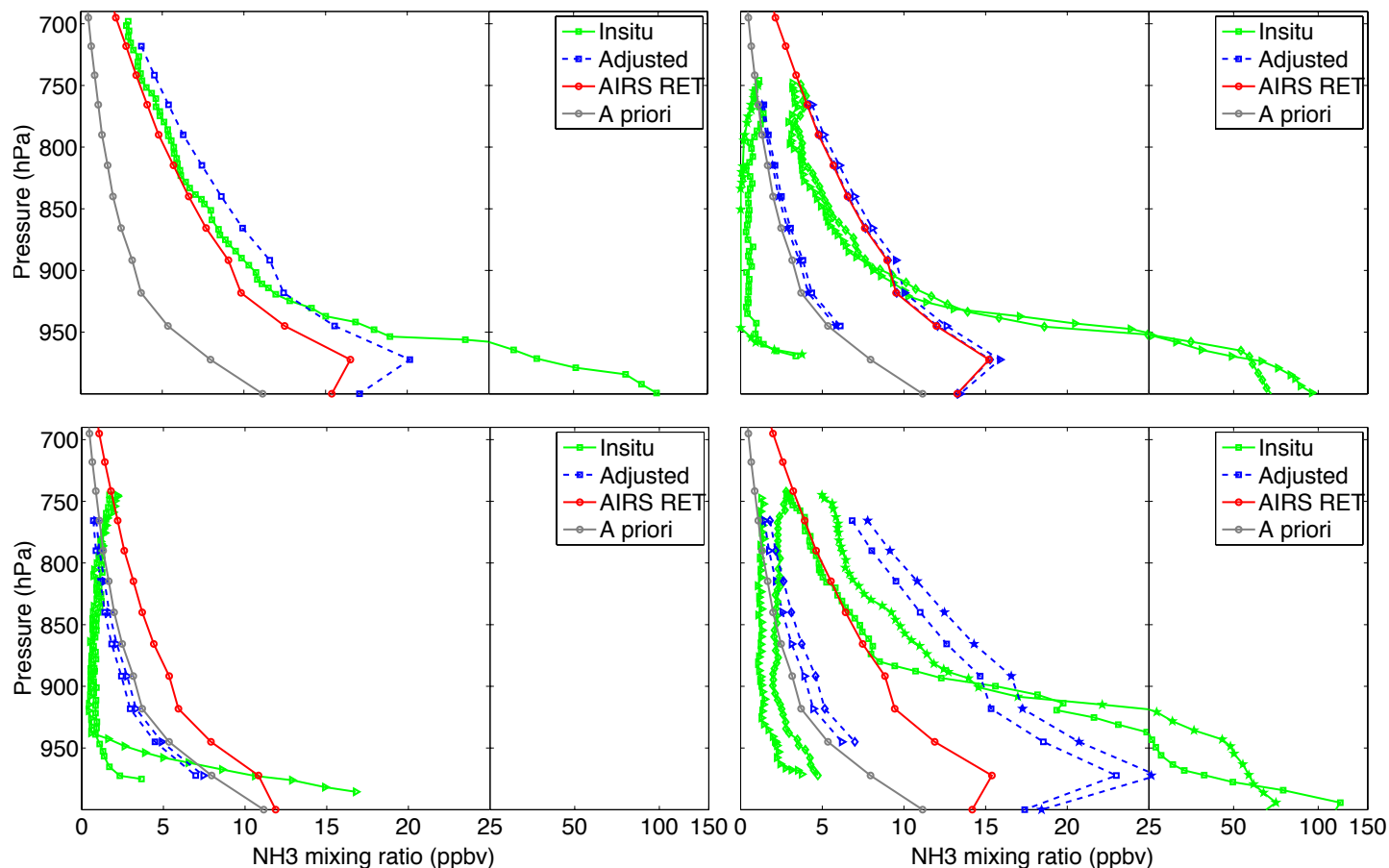
- Select *a priori* pollution scenarios based on brightness temperature differences weighted by noise (DBTI);
- Higher polluted scenarios are correlated with high retrieval DOFS.



Validation vs CRDS/Picarro in DISCOVER-AQ CA

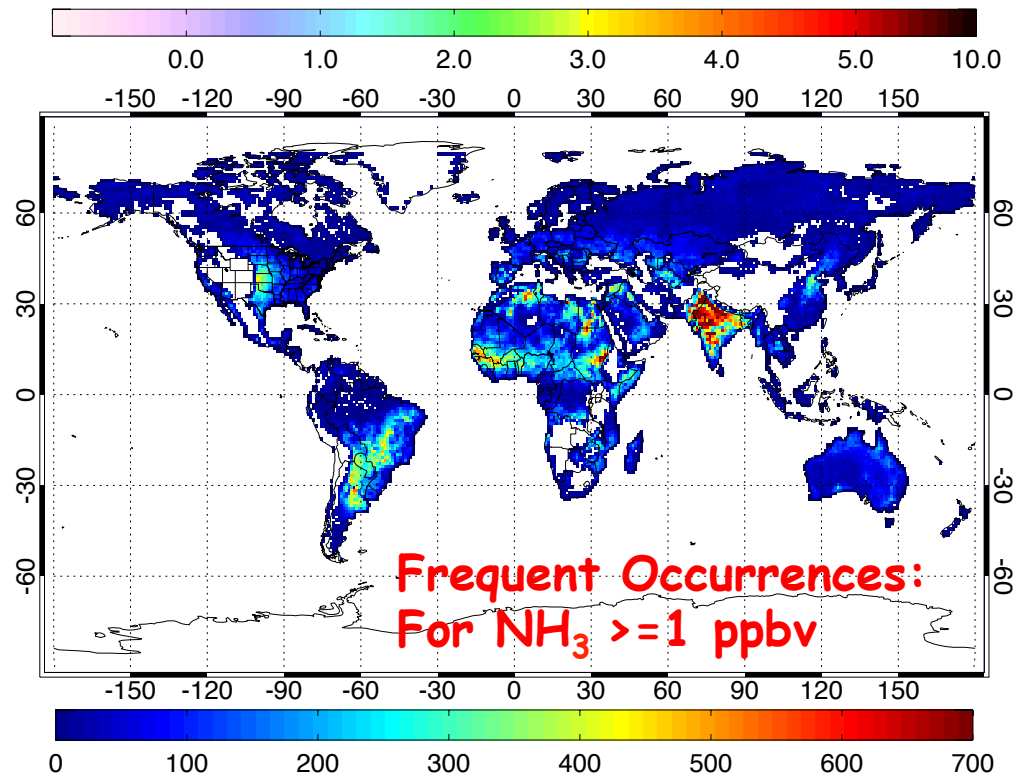
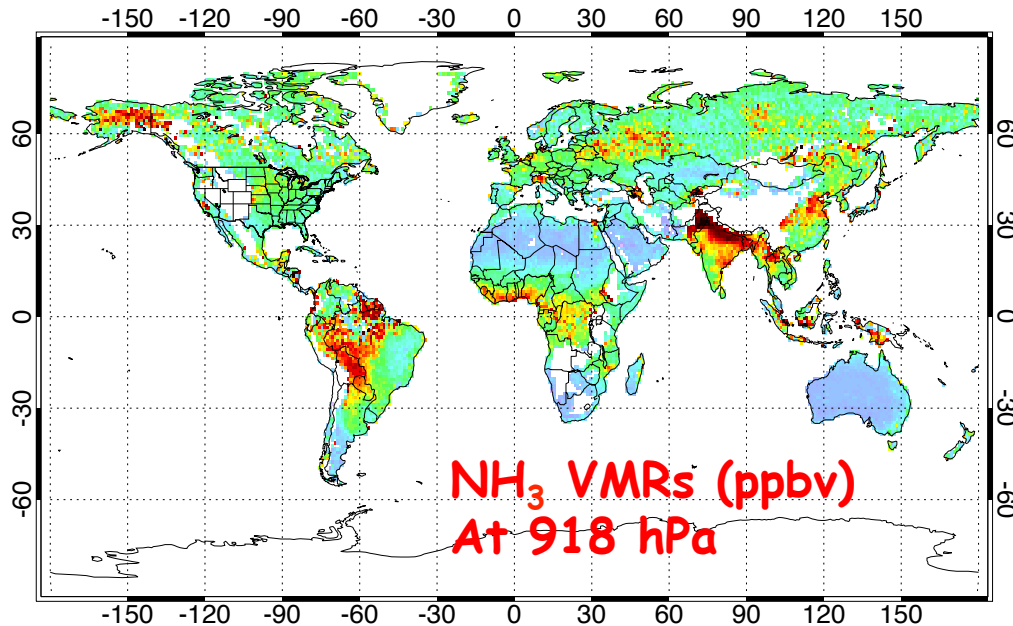
Spiral Profiles Only - 01/16 to 02/06, 2013

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

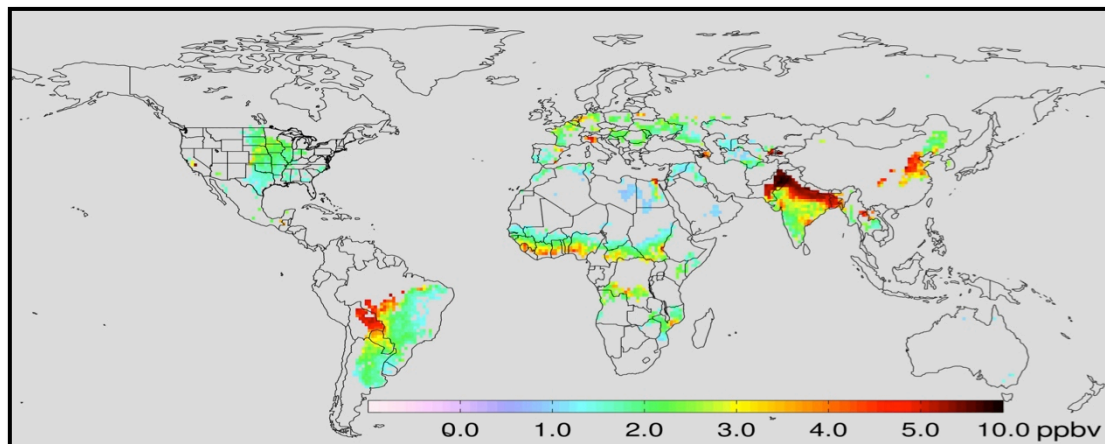


- Gray - a priori; Red - retrievals; Green solid - in situ; and Blue dashed - convolved in situ.
- AIRS L2 pixel sizes are $\sim 45 \text{ km}^2$, can coincide with multiple in situ profiles.
- AIRS NH₃ measurements are most sensitive at 850-950 hPa layer.
- Profile locations and quality assurance criteria are in Warner et al., 2016

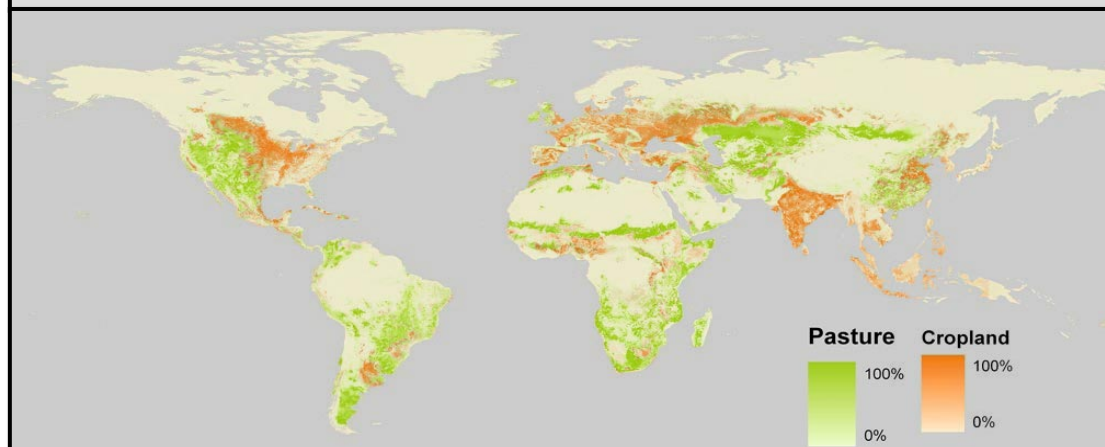
Global NH_3 in 2002-2015



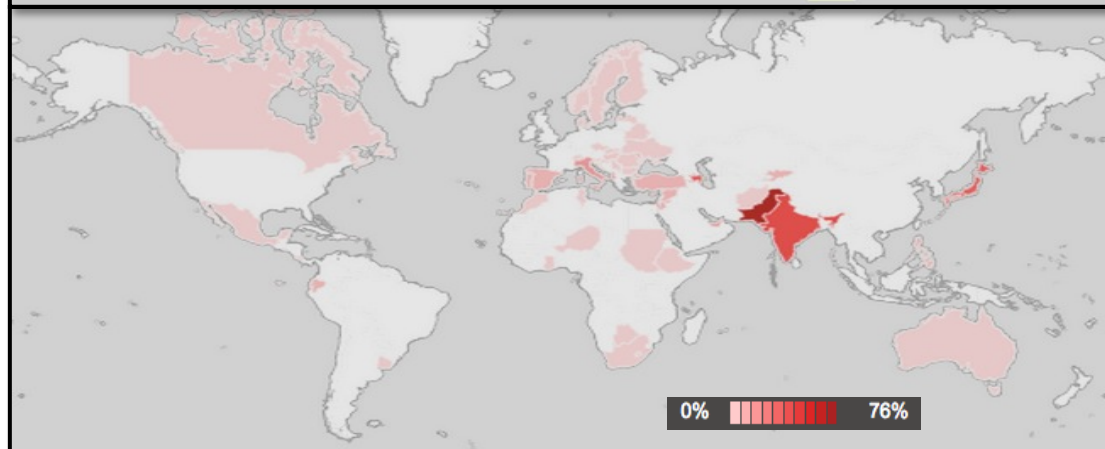
- AIRS NH_3 at 918 hPa for daytime and land only averaged over Sept. 2002 to Aug. 2015;
- Use Q0; DOFS ≥ 0.1 ;
- High concentrations are mainly due to human activities and fires;
- Use occurrences of higher emissions (lower) to distinguish between the two major sources: agricultural (high VMRs & high 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 NH₃ VMRs from the persistent sources filtered with the collocated occurrences of elevated concentrations (≥ 1.4 ppbv) using a threshold of 40 days;

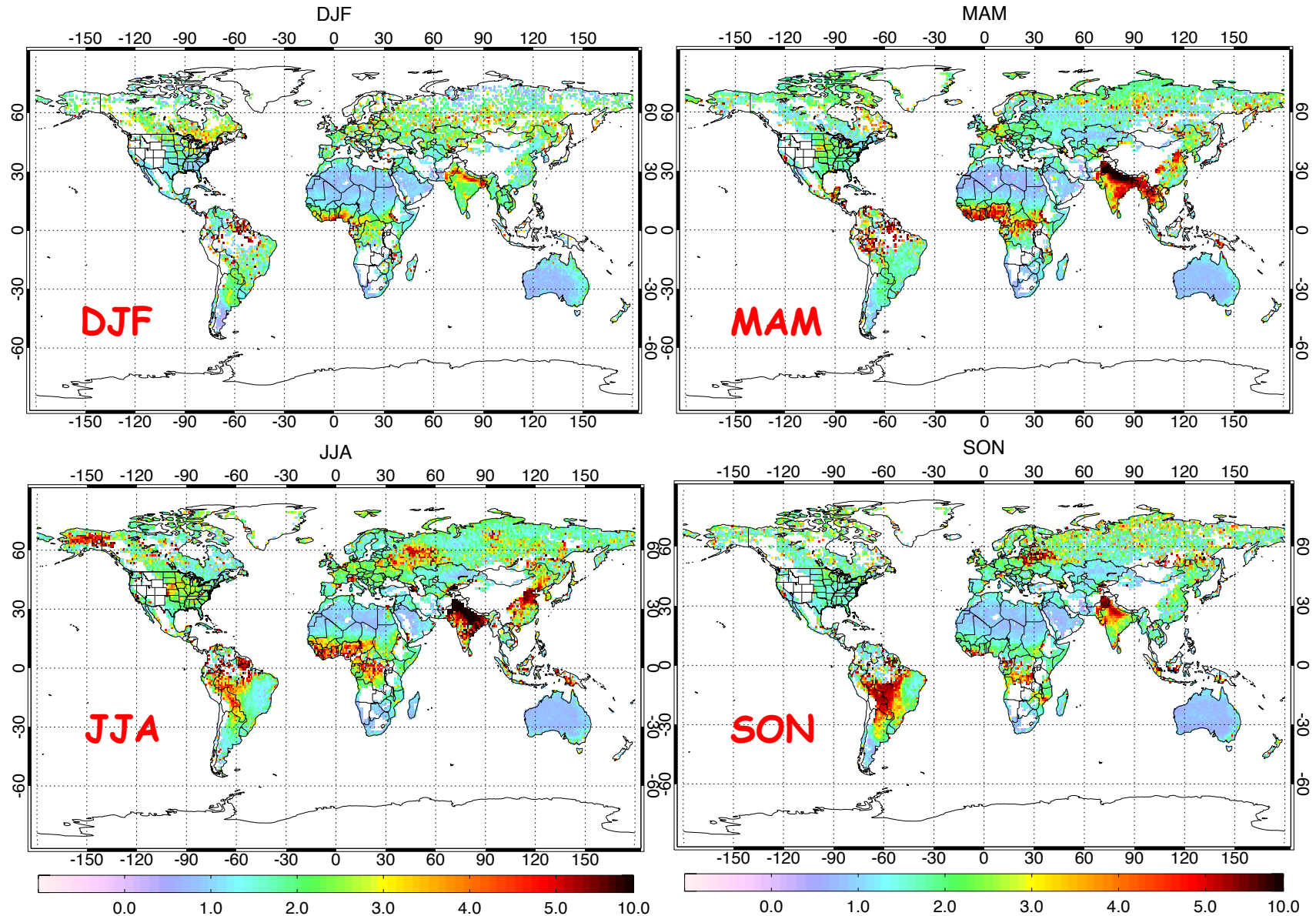


Middle panel: Pasture and Cropland Map



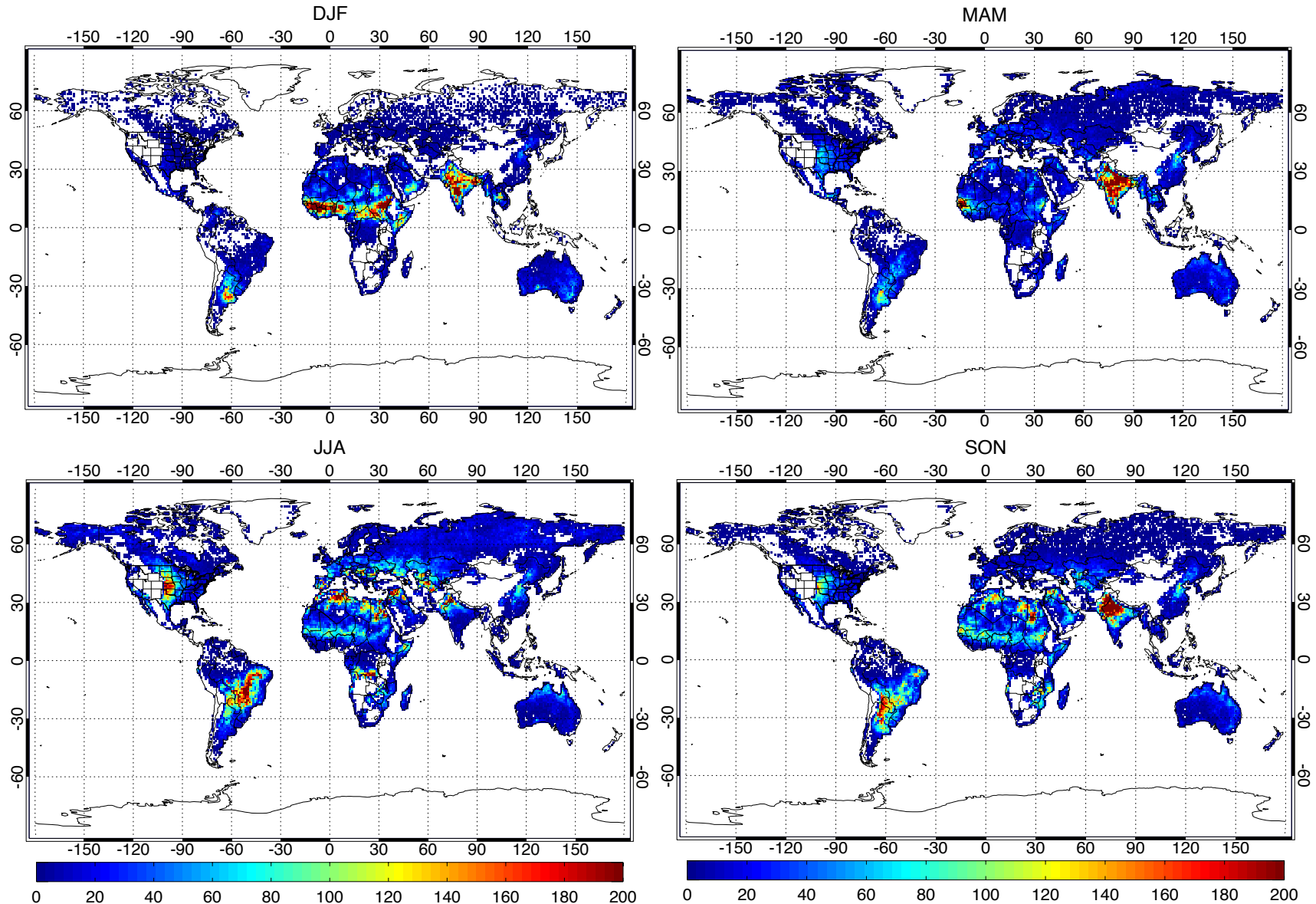
Bottom panel: irrigated agricultural land areas.

Global NH_3 Seasonality over 13 years



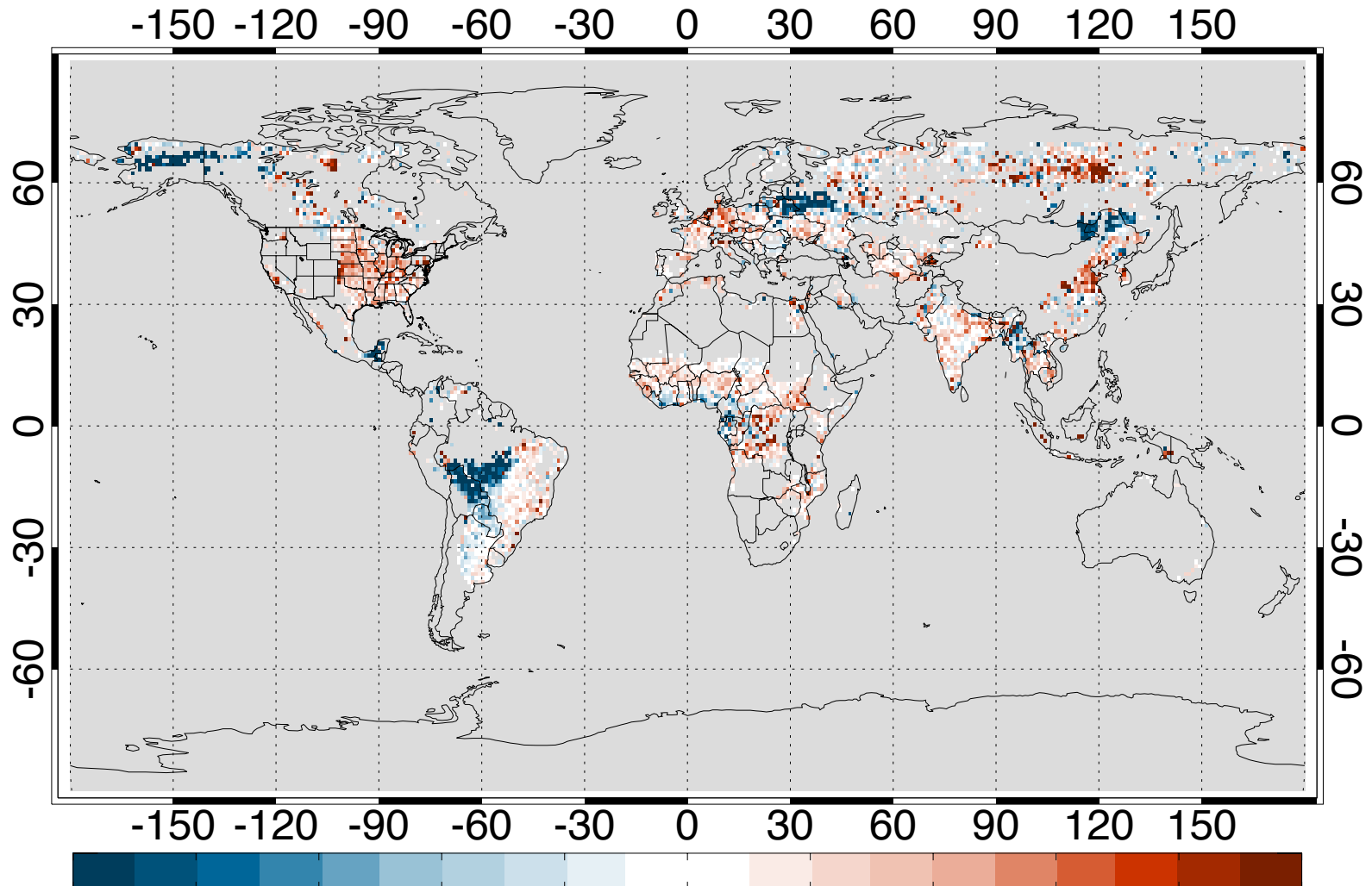
- The strongest non-BB emissions in the NH occur in the spring and summer
- Highest non-BB emissions are over India, China, USA, and Europe.

Occurrences of High NH_3 DOFS > 0.1



- Distinguish occasional BB events from the livestock and agricultural activities.

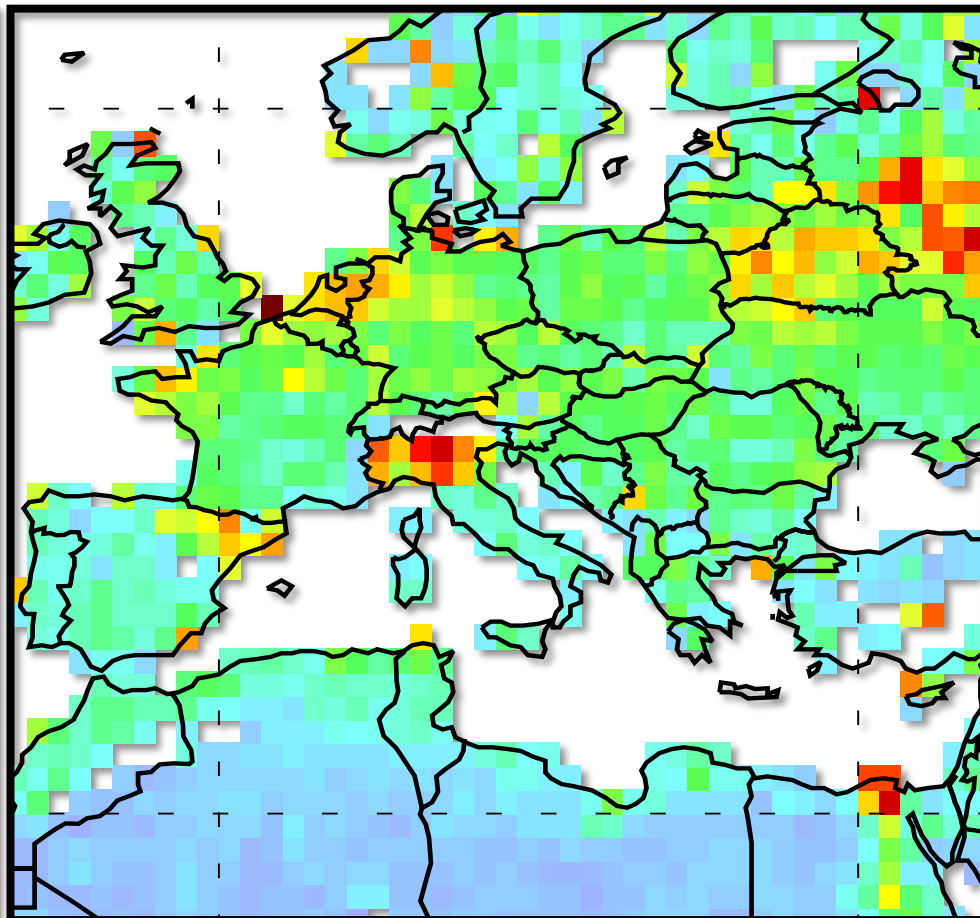
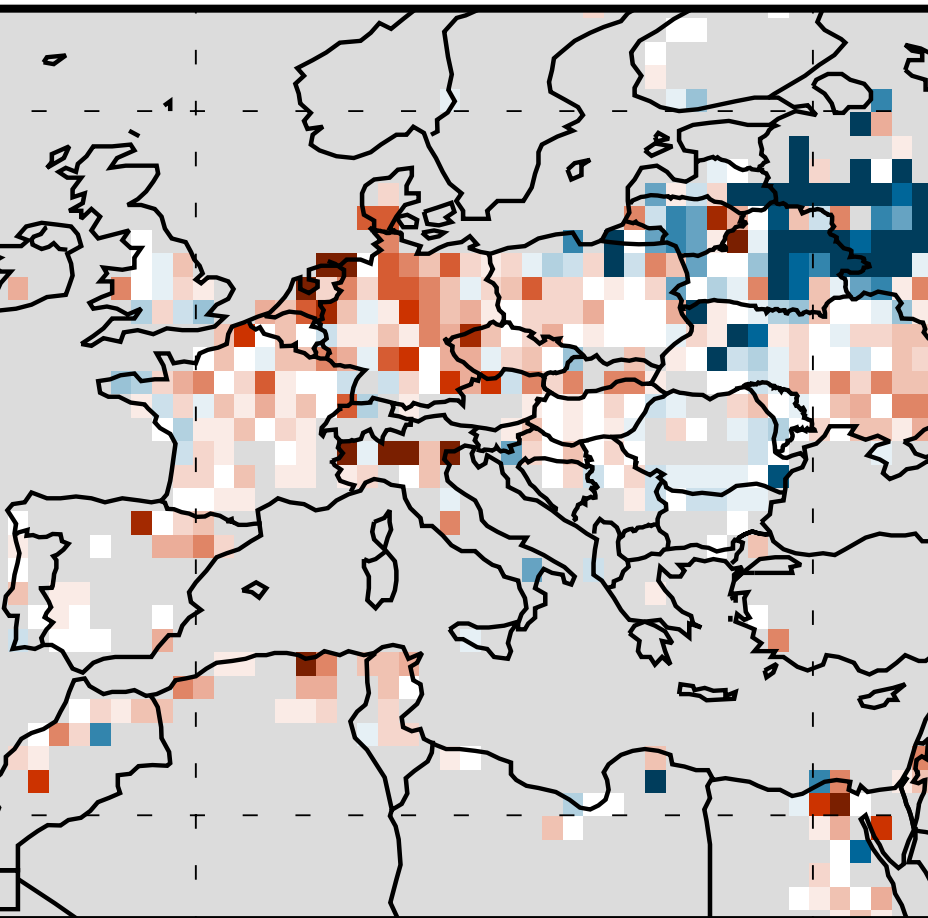
NH₃ Short-term Trends - Last 13 years



-0.20 -0.16 -0.12 -0.08 -0.04 0.00 0.04 0.08 0.12 0.16 0.20

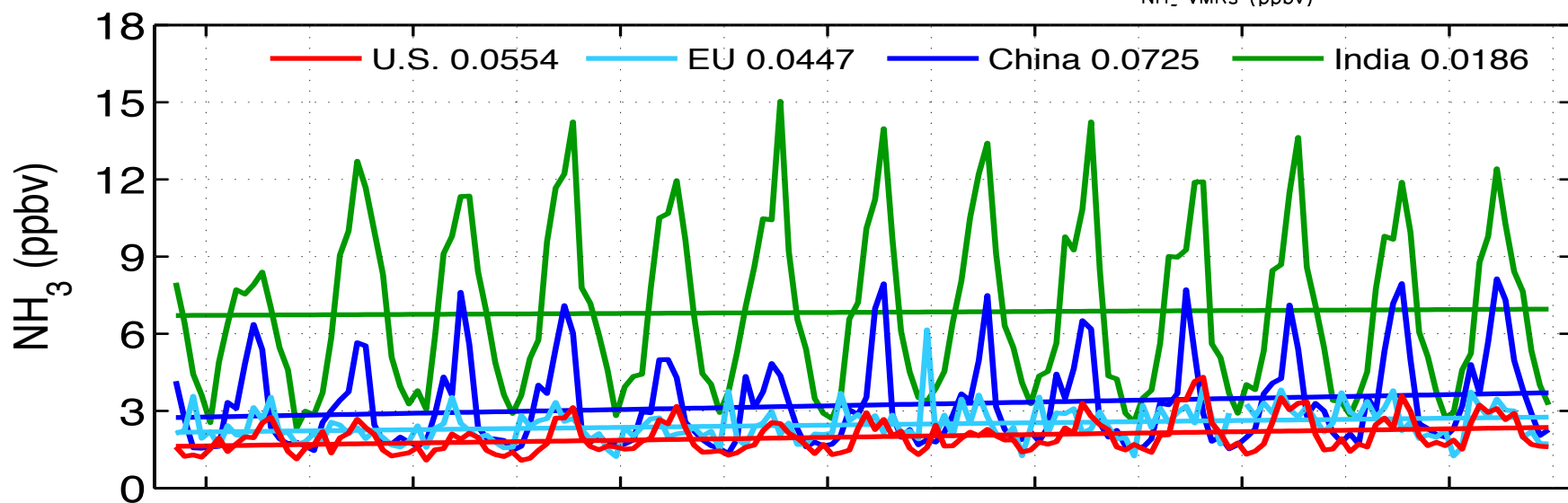
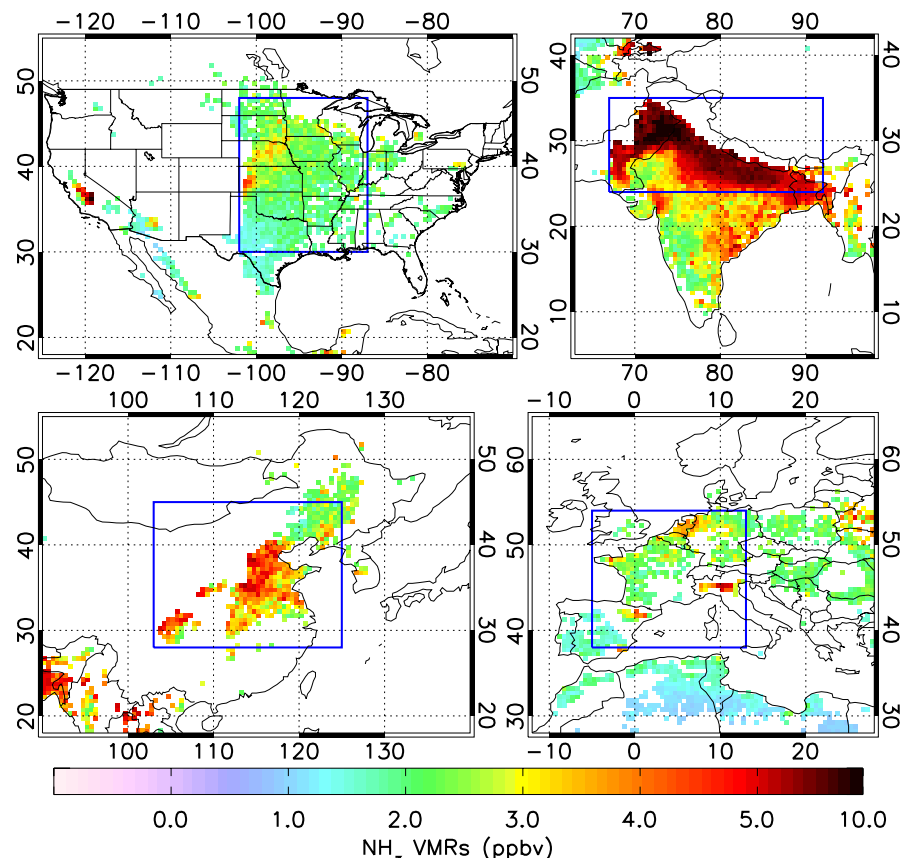
- Slopes of linear fit of NH₃ VMRs for each 1x1 grid.
- Concentrations of anthropogenic emissions increased and BB decreased
- Trends due to BB are not conclusive due to the short record.

NH₃ Short-term Trends - Last 13 years



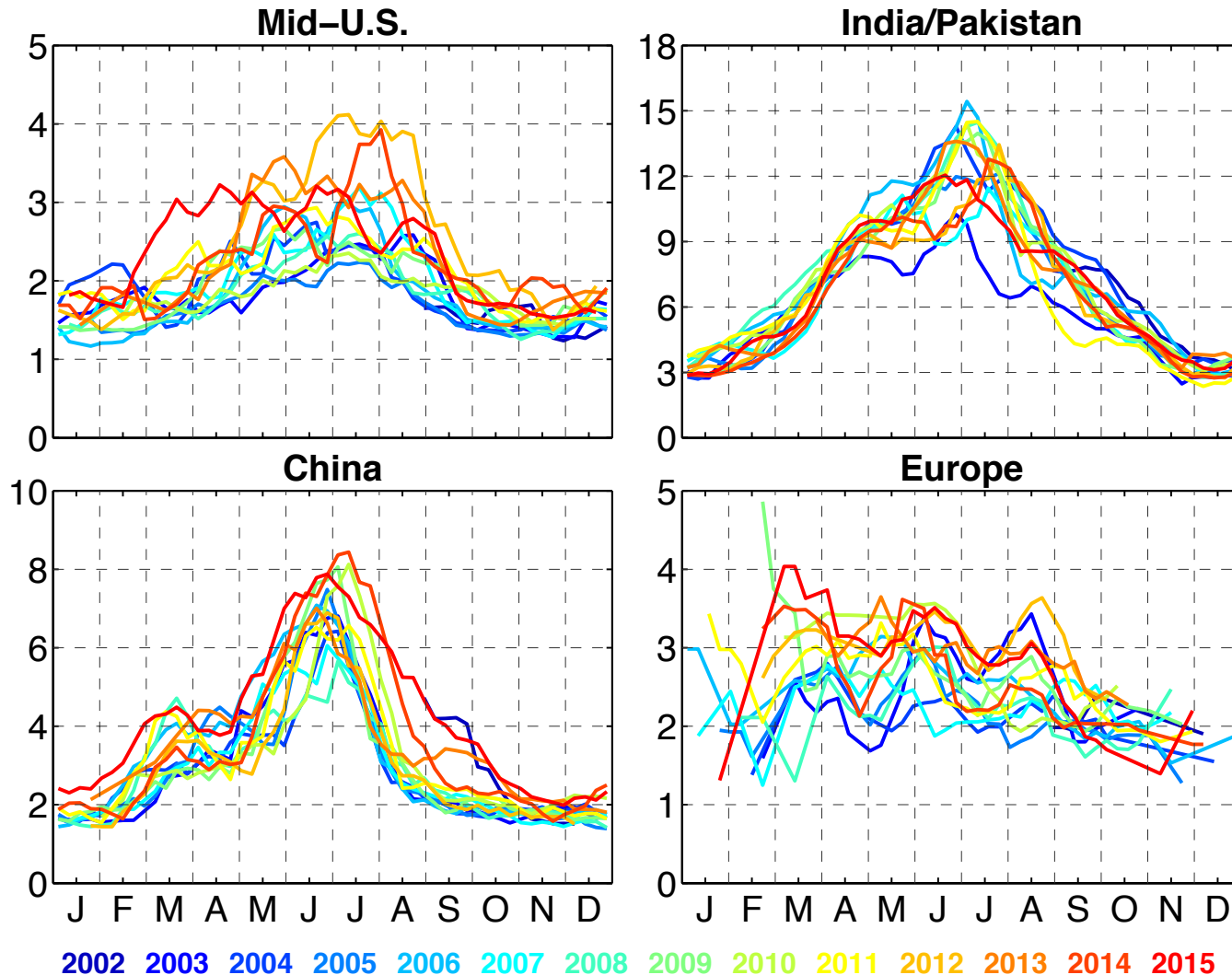
NH₃ VMRs at 918hPa US, China, India, & Europe

- Using high concentration and high frequent occurrences;
- The highest NH₃ concentrations in average occur in India/Pakistan & China.
- All 4 regions show increasing NH₃ trends in the last 13 years.
- Examine against OMI SO₂ & NO₂



AIRS NH₃ Seasonal Variations

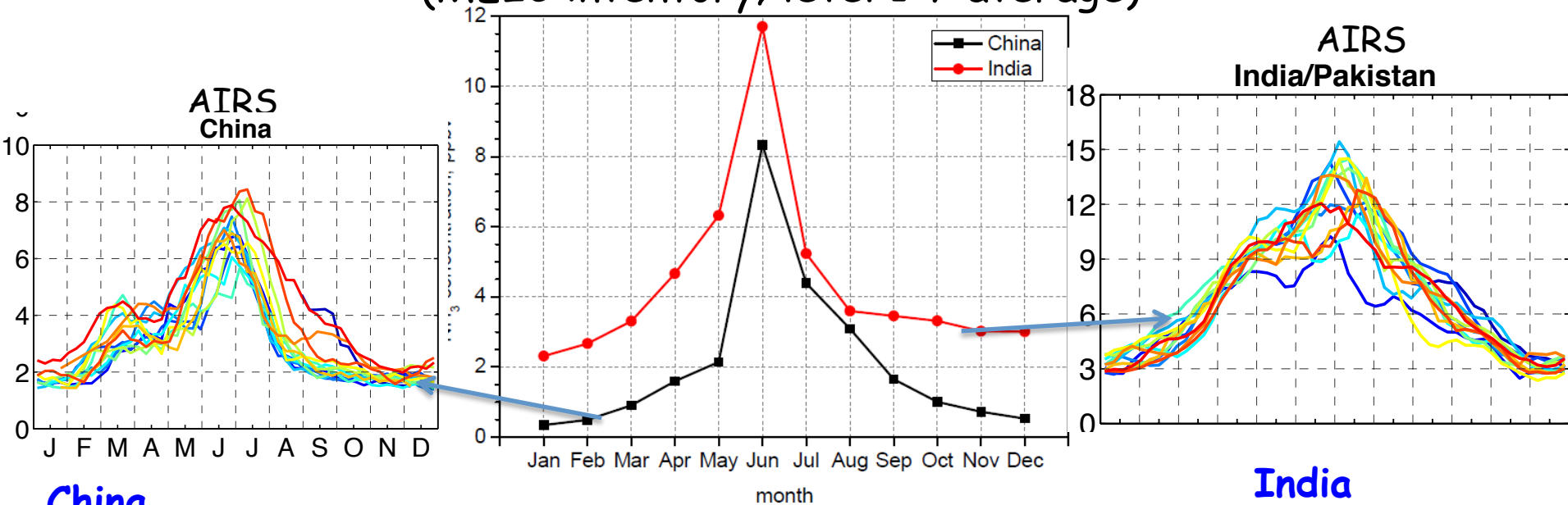
- over USA, China, Europe, and India



- NH₃ in India seasonal variations are broad and no obvious increasing/decreasing trends;
- NH₃ for USA and China are similar, with peaks in both spring and summer;
- NH₃ low seasonal changes for Europe, regions selected are too large.

Comparison with GEOS-Chem simulation

Model NH_3 over China and India (2010)
(MEIC inventory; level 1-7 average)*



China

- Summer peaks are consistent (~8 ppbv)
- Model is 2X lower in winter**
- Model misses the spring peaks

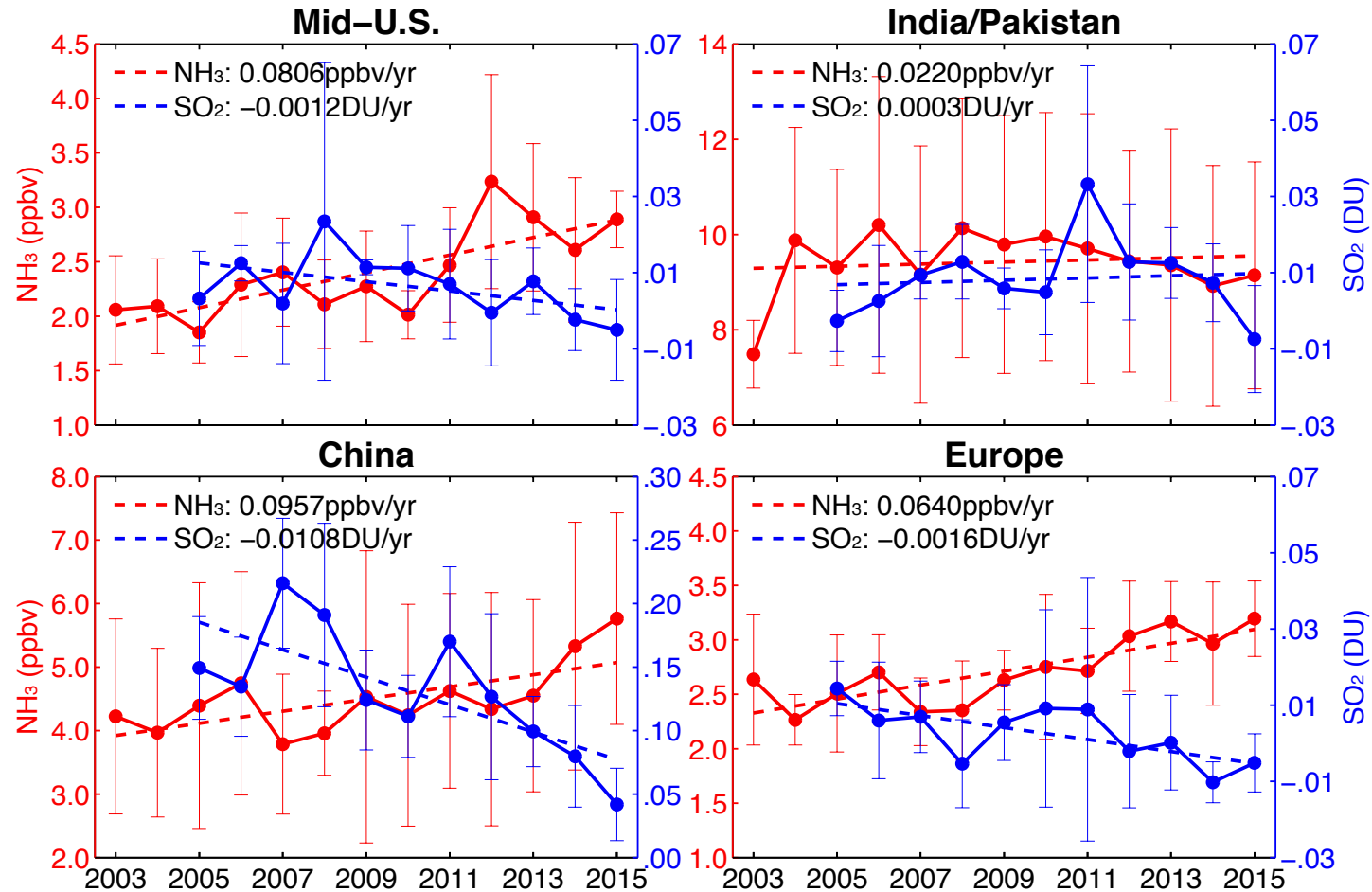
India

- Model lower in both winter and summer by 20~30%
- Model misses the broad spring shoulder

AIRS NH_3 is consistent with model in:
(1) Magnitude of peaking concentrations
(2) India is overall higher than China

* GEOS-Chem v9-02, nested-grid
** Note low sensitivity values are excluded!

AIRS NH_3 (top) vs OMI SO_2 (mid-) and NO_2 (lower) over Mid-US (red), China (blue), India (green) and Europe (cyan)



- The highest NH_3 concentrations in average occur in India/Pakistan, and China.
- All 4 regions show increasing NH_3 trends in the last 13 years.
- Decreased SO_2 from OMI largely explains the reason of NH_3 increases in Midwest U.S., China, and Europe.
- In India, SO_2 slightly increase except for 2015, NH_3 has not varied significantly.

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 valley 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).
- Results are land only, daytime only, and over relatively higher thermal contrast regions. More work is needed to study the more complicated surface types, and regions with lower thermal contrasts.
- More validation over broader emission types and different regions are needed.