Ammonia (NH₃) Distributions and Recent Trends by 14-year AIRS Measurements

J. X. Warner¹, Z. Wei¹, R. R. Dickerson¹, L. L. Strow², J. B. Nowak³ Y. Wang⁴, Q. Liang^{5, 6}

¹Dept. 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.

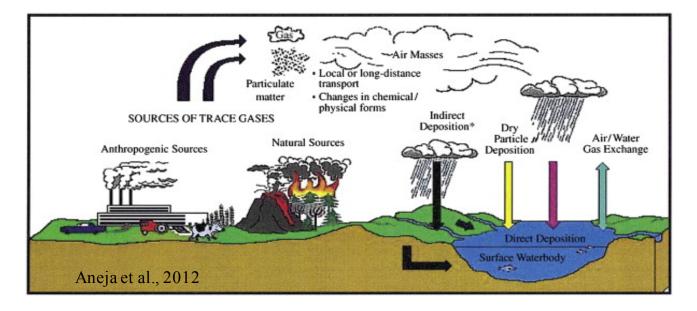
*Published in ACP: The Global Tropospheric Ammonia Distribution as seen in the 13-year AIRS Measurement Record

* Published in GRL: Increased Atmospheric Ammonia over the World's Major Agricultural Areas Detected from Space

* Funded by NASA's The Science of Terra and Aqua Program (NNX11AG39G), the Atmospheric Composition Program (NNX07AM45G), and SCIS (NNX16AQ67G).

Why Ammonia (NH₃)

- affect air quality, ecosystem, and climate, primarily from anthropogenic sources



Sources:

- Fertilizer use for crop production;
- Anima feeding operations;
- Emission increase with:
 - Nitrogen storage and pH of soils;
 - Surface temperature exponentially;
 - Soil moisture, etc.
- Biomass burning and volcanoes.

Sinks:

- Dry and wet deposition (soil acidification and eutrophication);
- Convert to particulate ammonium by reacting with sulfuric and nitric acids, arising from SO_2 and NO_x .

Ammonia – precursor gas of $PM_{2.5}$

- Precursor gases for PM_{2.5} (by secondary inorganic aerosols - a large portion of PM_{2.5}):
 ✓ Sulfer dioxide (SO₂);
 - ✓ Nitrogen oxides (NO_x);
 - ✓ Ammonic (NH_3);
- Only NH₃ is not regulated;
- Ammonia is the limiting species in $PM_{2.5}$ formation, regulating SO_2 and NO_x alone cannot determine the fate of $PM_{2.5}$;
- Meteorological conditions (wind, rain, temperatures, etc.) affects the concentration of ammonia gases.



Why Satellite Remote Sensing

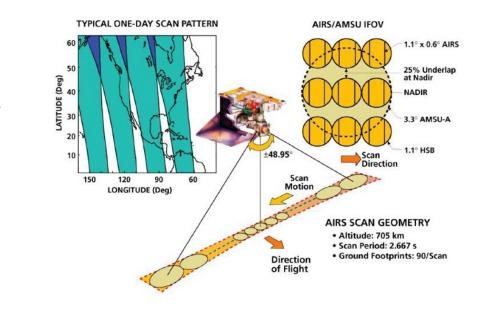
- Ammonia in situ measurements are rare, global coverage is impossible.
- Satellite measurements with daily and large global coverage are challenging and have been lacking partly because *the lifetime of NH*₃ *is relatively short* and partly because it requires high sensitivity for the retrievals that can be only obtained from areas with high thermal contrasts near the surface (Clarisse et al., 2010).
- AIRS (Atmospheric InfraRed Sounder) has the advantages:
 - afternoon overpasses (1:30pm) are best correlated with the daily emission peak time and during the daily period with the highest thermal contrast.
 - AIRS large coverage with wide swaths and cloud-clearing provide daily NH₃ maps.
 - The **15-year data records** makes AIRS the best sensor for NH_3 trends and variability studies (to date).

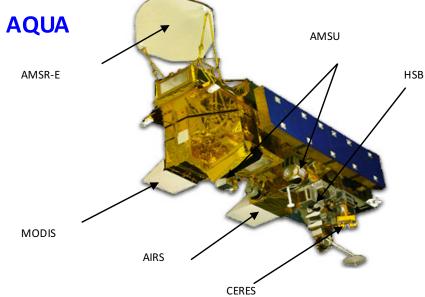
Atmospheric InfraRed Sounder (AIRS)

Launched May 2002; afternoon (1:30pm) overpass; daily global coverage

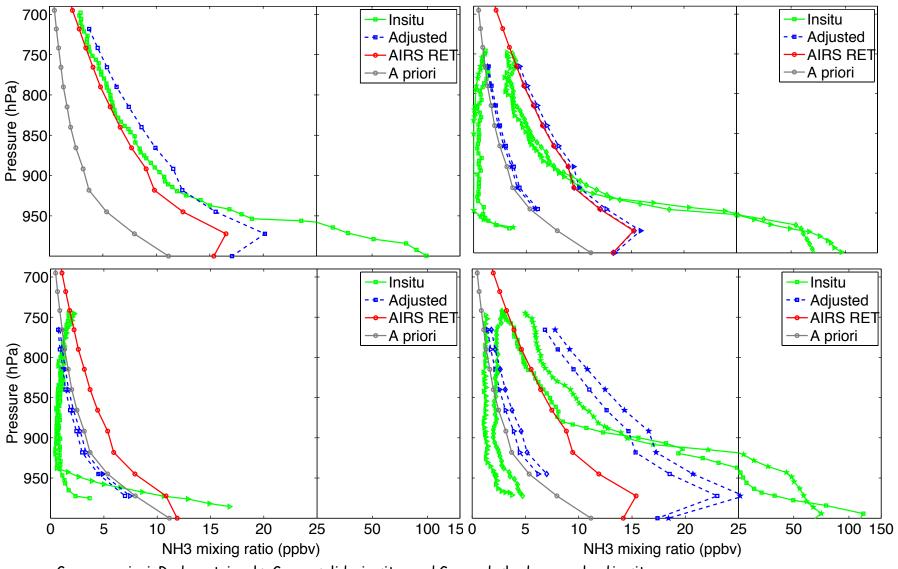


- A grating spectrometer originally designed to improve weather forecast and now also used for climate and air quality studies.
- Spectral resolution at 1/1200 (~ 0.5 cm⁻¹)
- Covers 650-2665 in three bands with a total of 2378 channels
- Spatial resolution 13.5 km² (with retrievals at ~45 km²)
- Wide swaths and cloud clearing provide daily global coverage
- Very high Signal-to-Noise accuracies of 1K over 1 kmlayer.





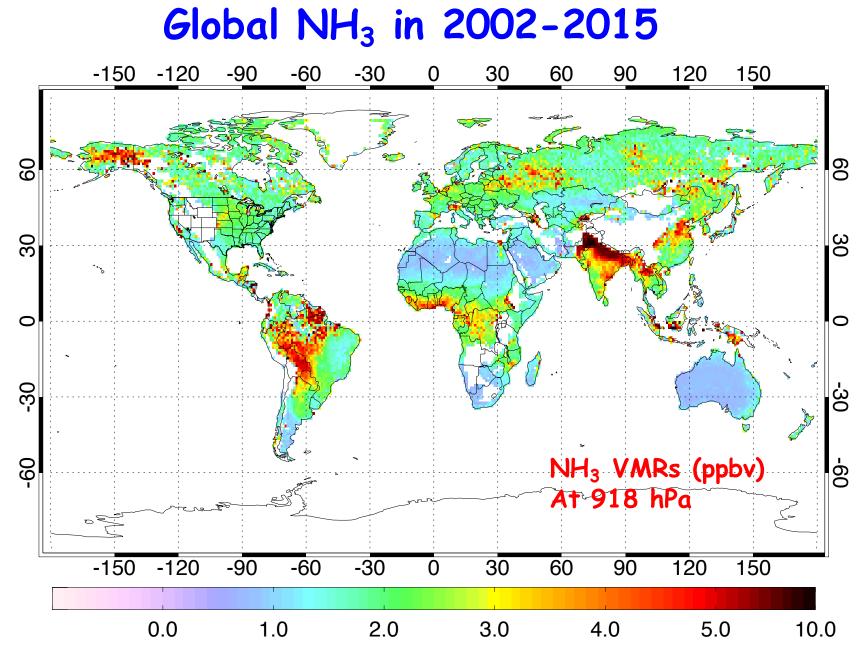
Validation vs CRDS/Picarro in DISCOVER-AQ CA CRDS/Picarro Spiral Profiles Only - 01/16 to 02/06, 2013 (data courtesy of Co-author J. Nowak)



• Gray - a priori; Red - retrievals; Green solid - in situ; and Green dashed - convolved in situ.

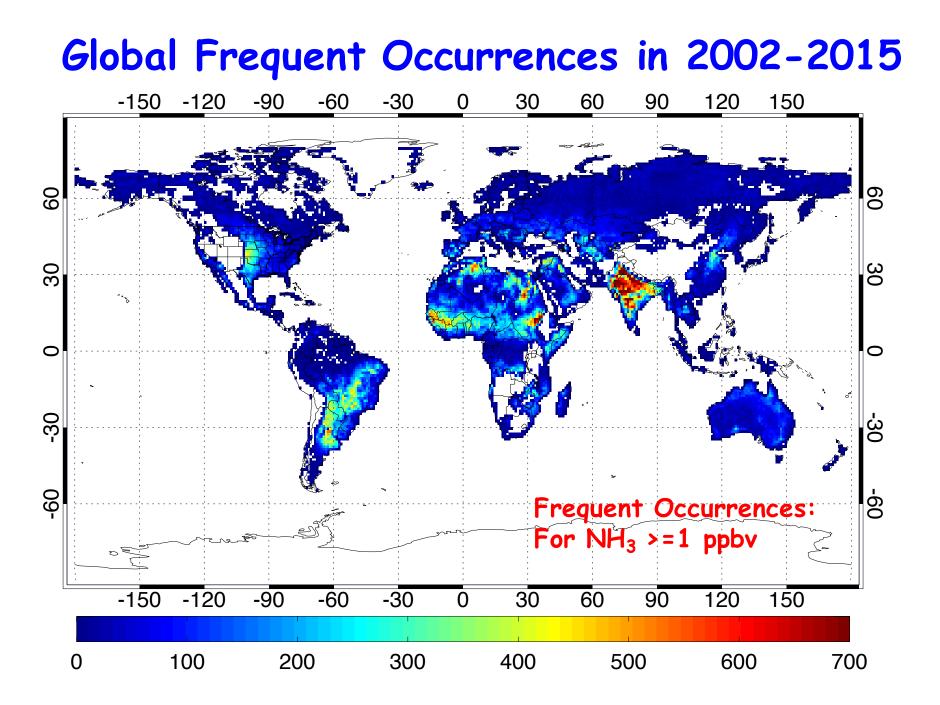
• AIRS L2 pixel sizes are ~45 km², can coincide with multiple in situ profiles.

• AIRS NH₃ measurements are most sensitive at 850-950 hPa layer.

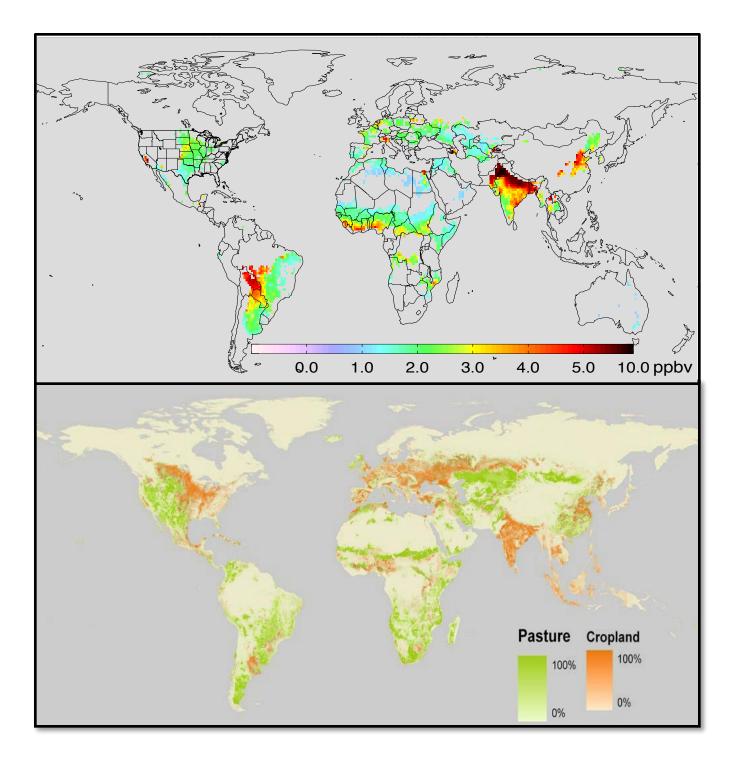


• High concentrations are mainly due to human activities and fires;

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).



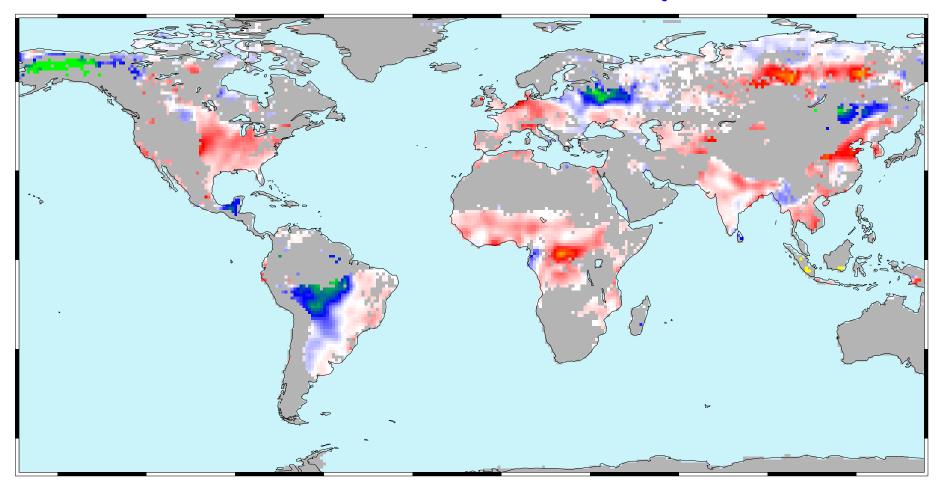
Use occurrences of higher emissions (lower) to distinguish between the two major sources: agricultural (high VMRs & high frequencies); BB emissions (high VMRs & low frequencies).

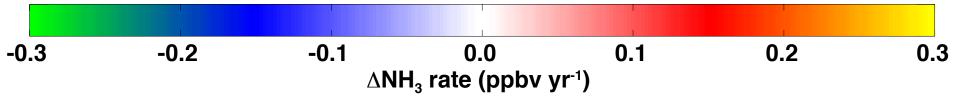


<u>Top panel</u>: The NH_3 VMRs from the persistent sources, i.e., \geq 1.4 ppbv for more than 40 days;

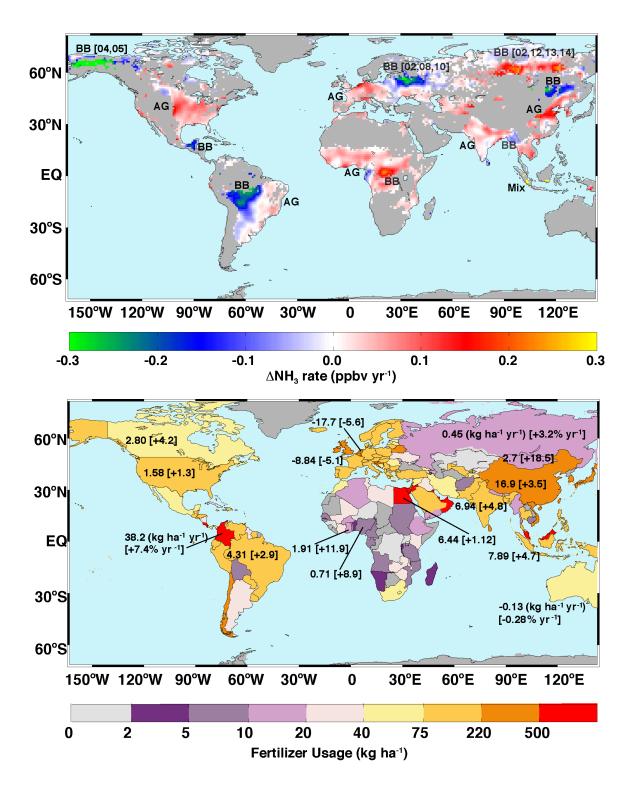
Bottom panel: Pasture and Cropland Map (http://OurWorldInData.org)

NH₃ Trends - Last 14 years



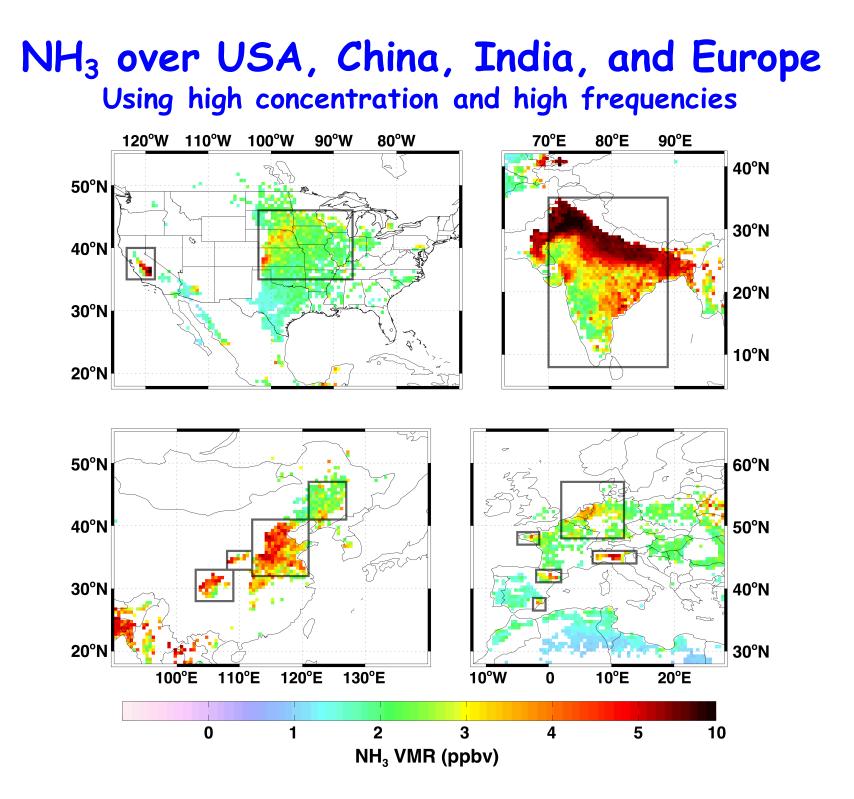


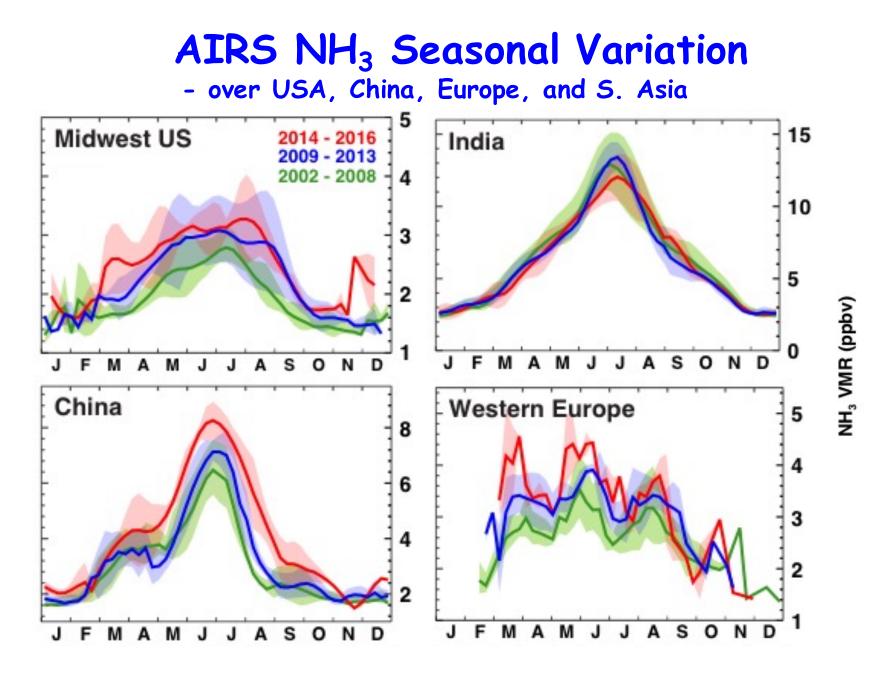
- Global trends in atmospheric NH_3 (i.e., VMRs at 918hPa for each 1x1 grid)
- Red-yellow colors represent increases due to agriculture emission increases and reduced scavenging by acid aerosols
- Blue-green colors represent decreases due to possibly reduced agricultural burning and fewer wild fires



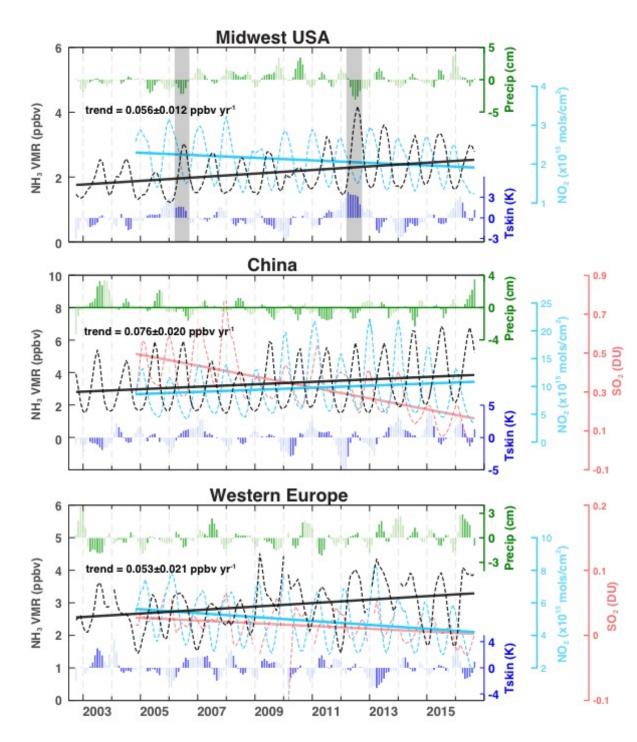
(top) Temporal trends; BBbiomass burning and AGagricultural regions

(bottom) National averaged annual N fertilizer usage in 2002–2013 in kg ha⁻¹ and trends (percentage changes)





The highest NH₃ concentrations in average occur in South Asia, and China. Note scales.
NH₃ in India seasonal variation are broad and no obvious increasing/decreasing trends;
NH₃ for USA, China and Europe have increased, with peaks in both spring and summer;



AIRS NH₃ vs OMI NO₂ for US (top), SO₂ for China (middle) and NO₂ for Western Europe (lower)

All show increasing NH_3 trends (black) 2002-2016.

Decreased SO_2 largely explains the NH_3 increases in Midwest U.S. (not shown), China, and Europe.

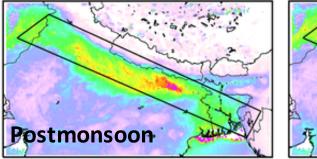
OMI NO_2 decreasing explains winter NH_3 increasing over the US and Europe.

Meteorological conditions also affect NH₃ concentrations (high surface temperatures and low precipitation), see shaded anomalies in the top panel.

ECMWF surface skin temperatures show increases over the US and China can possibly link climate change to the increased NH_3 emissions.

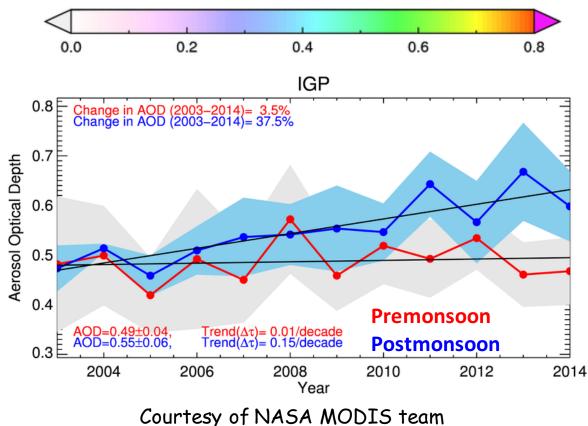
South Asia Aerosols Trends

2003-2007

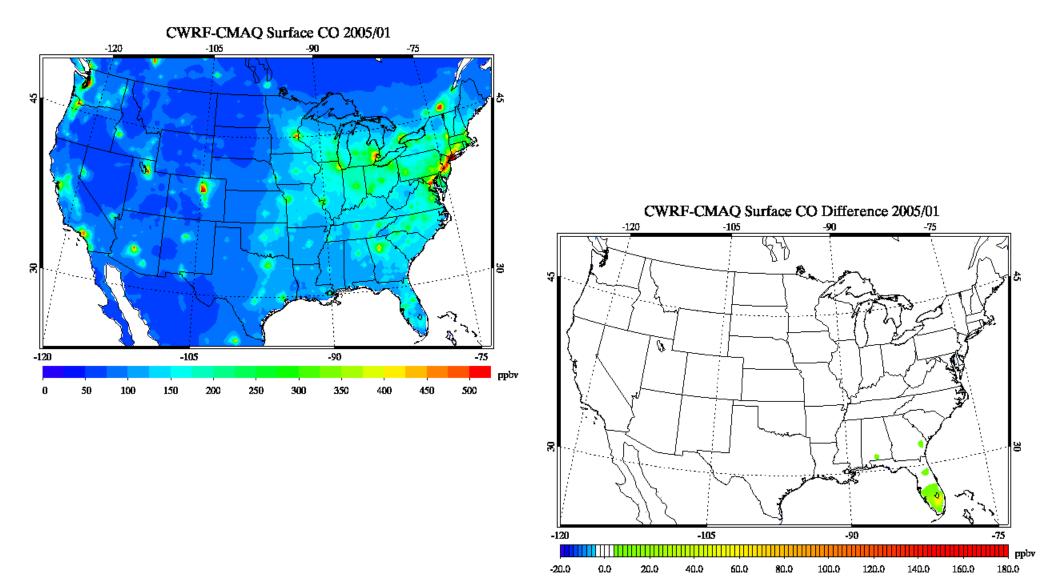


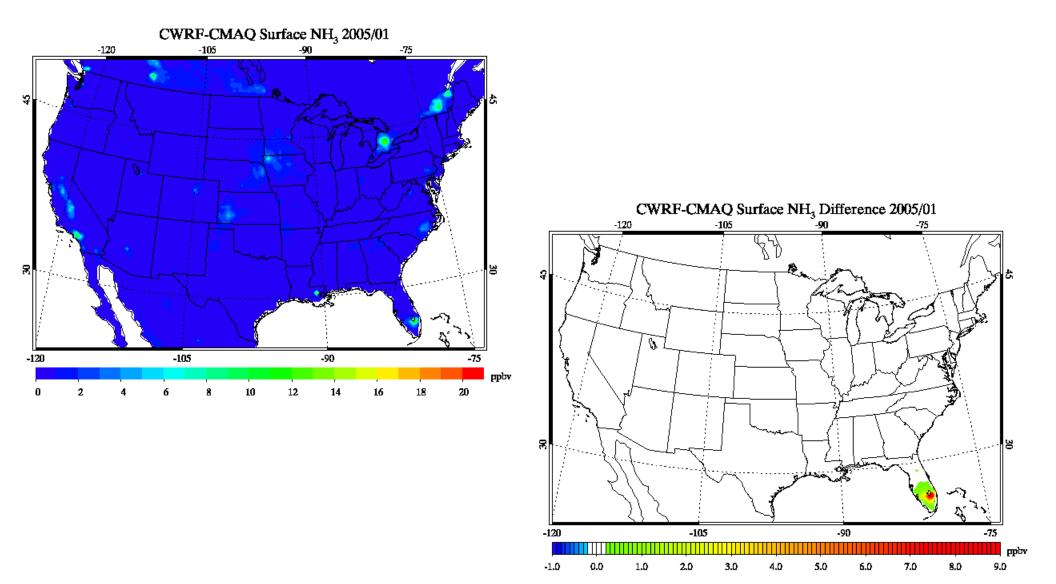
2008-2014

Aerosol Optical Depth at 550 nm



- No obvious NH₃ increased in the AIRS observations over South Asia, while the fertilizer usage has increased;
- Recent increases in SO₂ and NO_x from uncontrolled coal combustion and other sources;
- Greater conversion of gaseous NH₃ into particulate sulfates and nitrates.
- Dust influence in premonsoon and sea salt during monsoon, more fine mode aerosols in postmonsoon.





Summary

- AIRS NH_3 products not only include 14 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 concentrations increase resulted primarily from decreases in concentrations of acidic aerosols (sulfate and nitrate), an unintended consequence of effective controls of NO_x and SO₂ emissions.