Molecular perspective for global modeling of upper atmospheric NH_3 from freezing clouds

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Nanjing University Information Science and Technology
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First measurement of ammonia in free troposphere

Gaseous ammonia and ammonium ions in the free troposphere

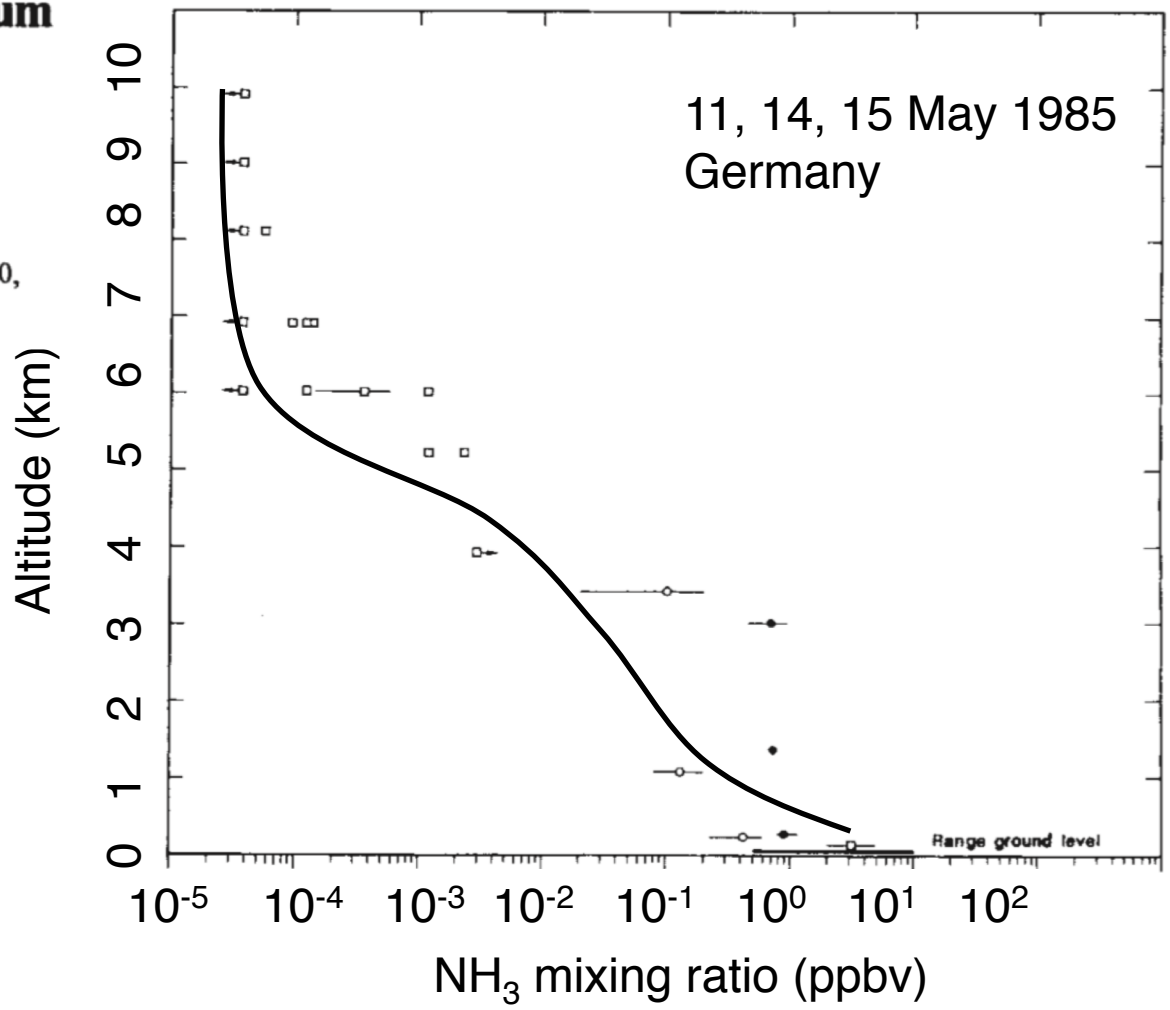
Nature, 1986

H. Ziereis & F. Arnold

Max-Planck-Institut für Kernphysik, Postfach 103980, D-6900 Heidelberg, FRG

Measurement of NH_3 can be affected by interference of NH_4^+—containing particles.

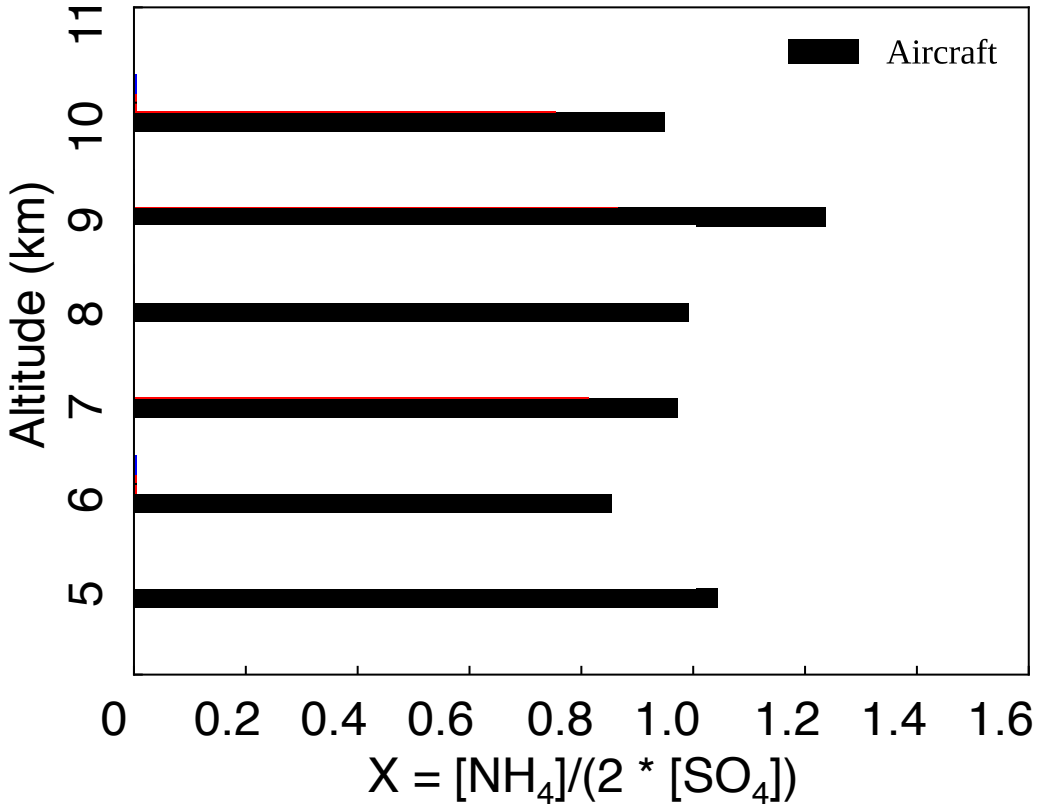
Detection of gaseous NH_4^+ 'cluster ions' in free troposphere can provide sensitive means for measuring gaseous NH_3.



Sulfate particles can be highly neutralized in UTLS

- 1980s, reviews concluded that $(NH4)_2SO_4$ may weigh 10% in total stratospheric aerosol mass, but admitted that the presence of ammonia in UTLS is "an open question" – Turco, 1982, *Rev. Geophys.*
- Since 1990s, ...

Months	Location
09-10, 1991	Western Pacific
02-03, 1994	Pacific
08-09, 1996	Tropical Pacific
03-04, 1999	Tropical Pacific
02-04, 2001	Pacific
07-08, 2004	North America & Atlantic
03-05, 2006	North America & Pacific

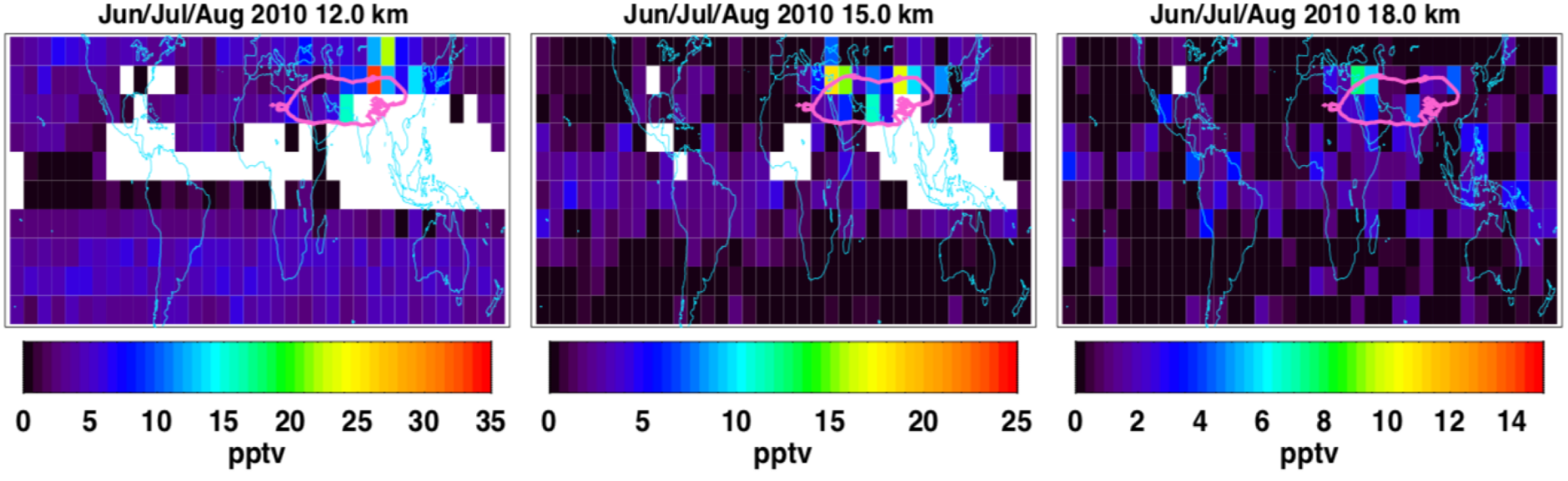


Field data show that NH_3 may not as small as what was believed.

But, how much NH_3 are there in UTLS?

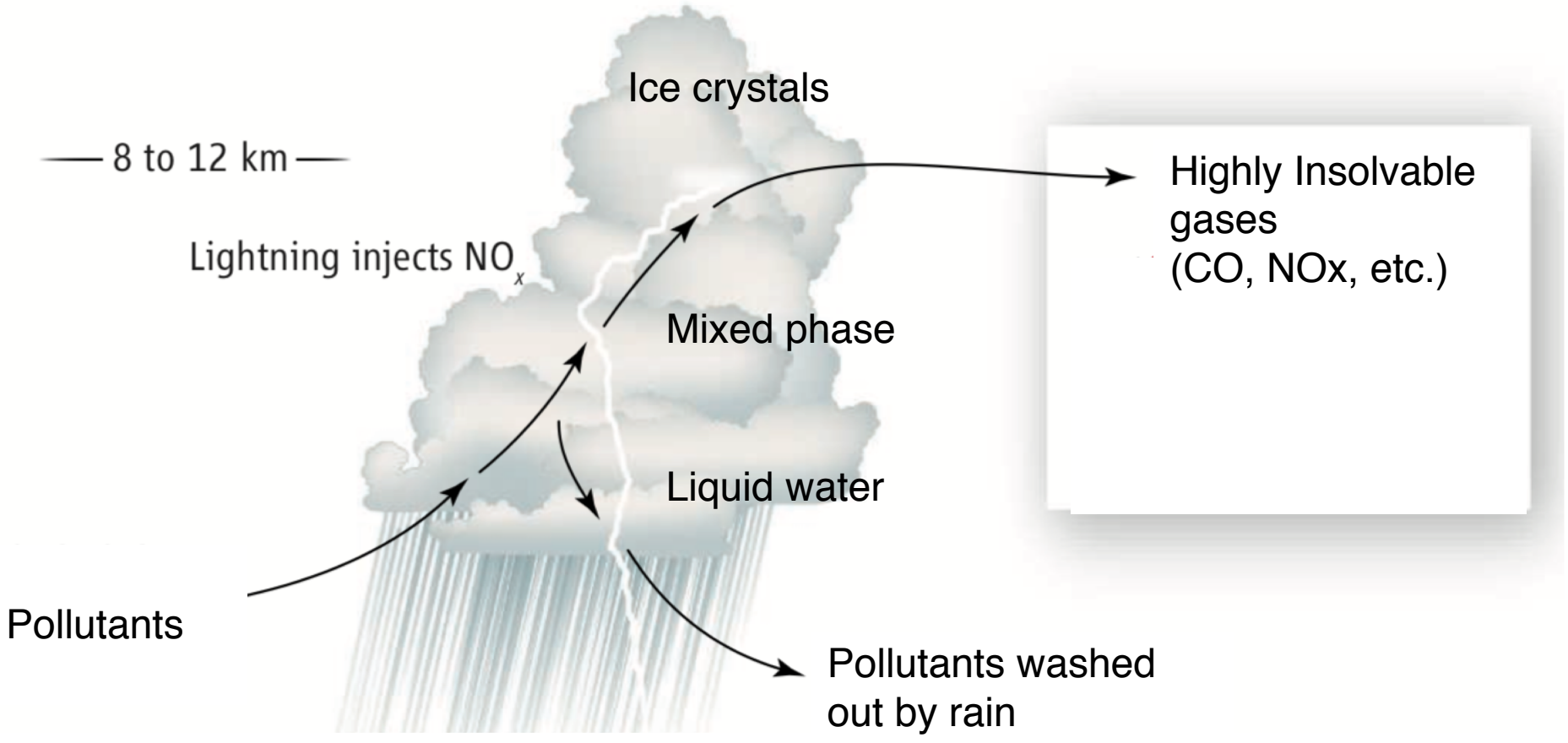
First Detection of NH_3 in UTLS from space

Höpfner et al. (ACP, 2016): "First detection of ammonia (NH3) in the Asian summer moonsoon upper troposphere". Retrieved from limb sensor – the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS)



Degree of freedoms for signal is ~ 3 above 10 km, and retrieval error is < 1 pptv.
The estimated detection limit (or variation of background) is ~3-5 pptv
There is no literature regarding locally resolved model results of NH_3 during the monsoon period.

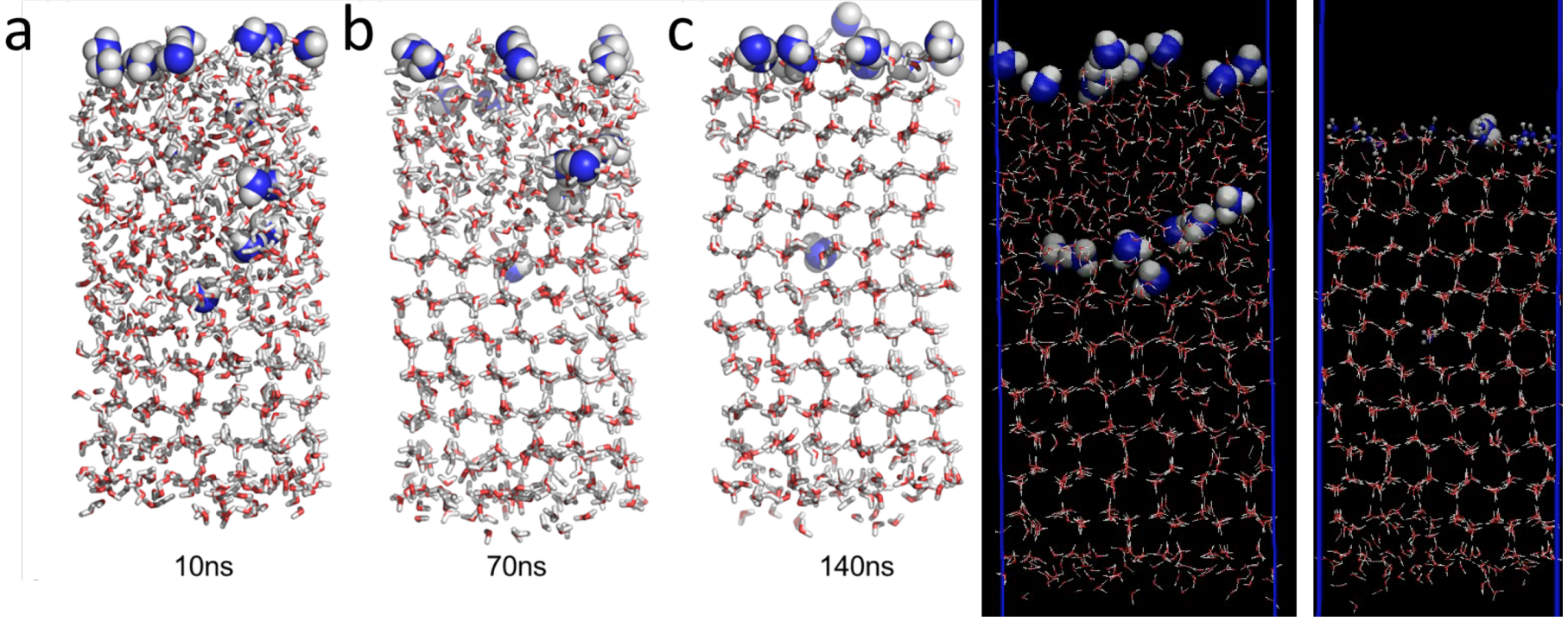
Rote of deep convection for pumping up surface air



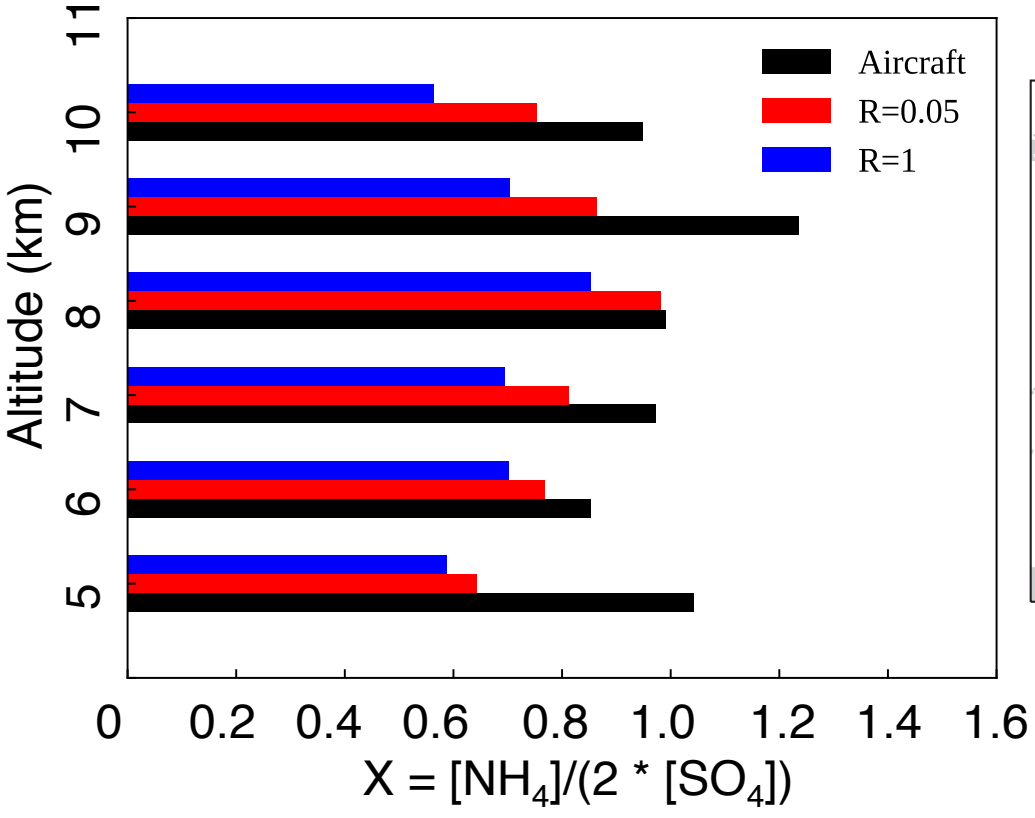
Jaeglé, 2009, *Science* with modification

Can NH_3 be retained in liquid-to-ice particle conversion?

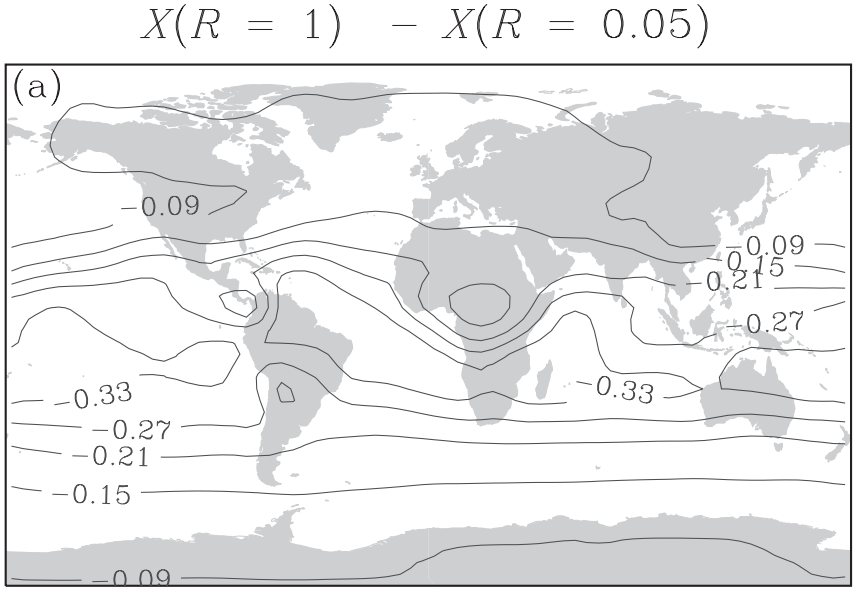
- **Lab measurements show the retention efficiency (R) : 0 -1.**
- **Molecular Dynamic Simulations:**



Results sensitivity: R = 1 vs. R = 0.05



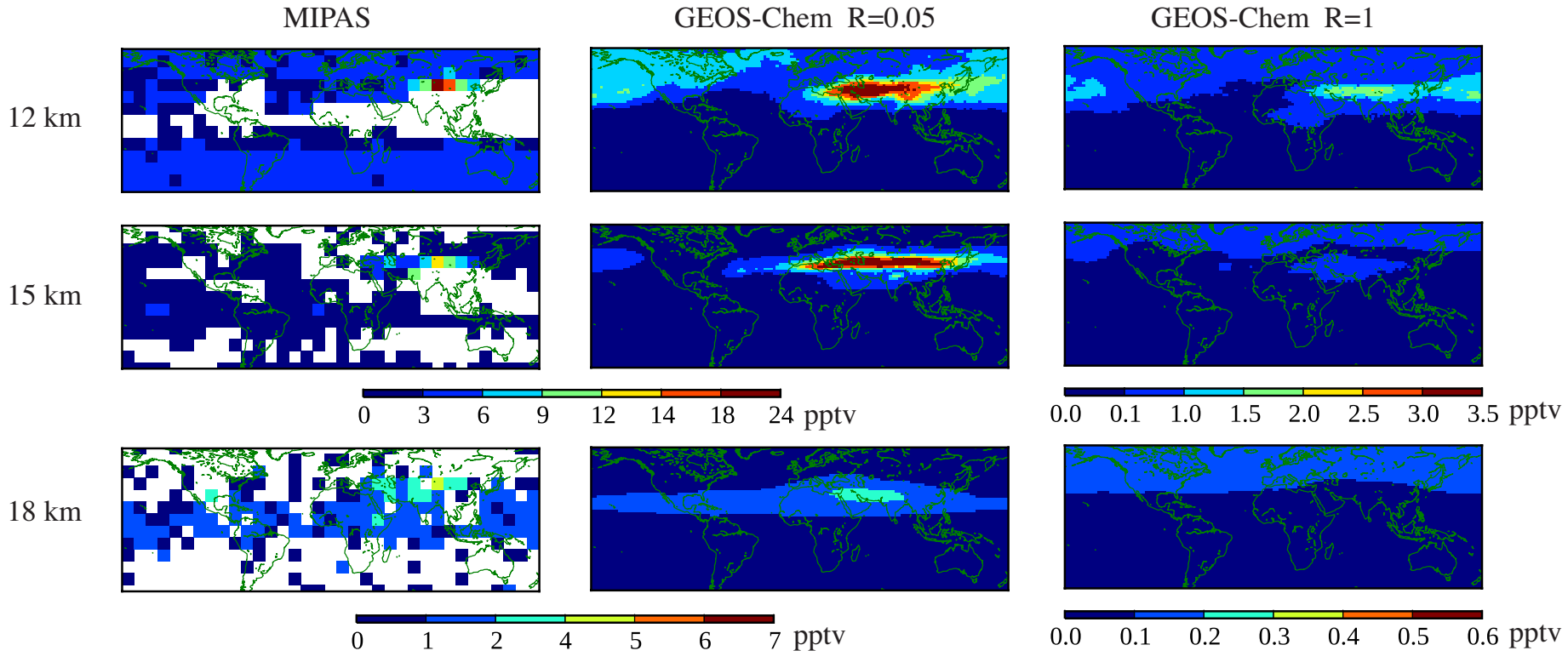
Ge et al., 2018, *PNAS,* in press.



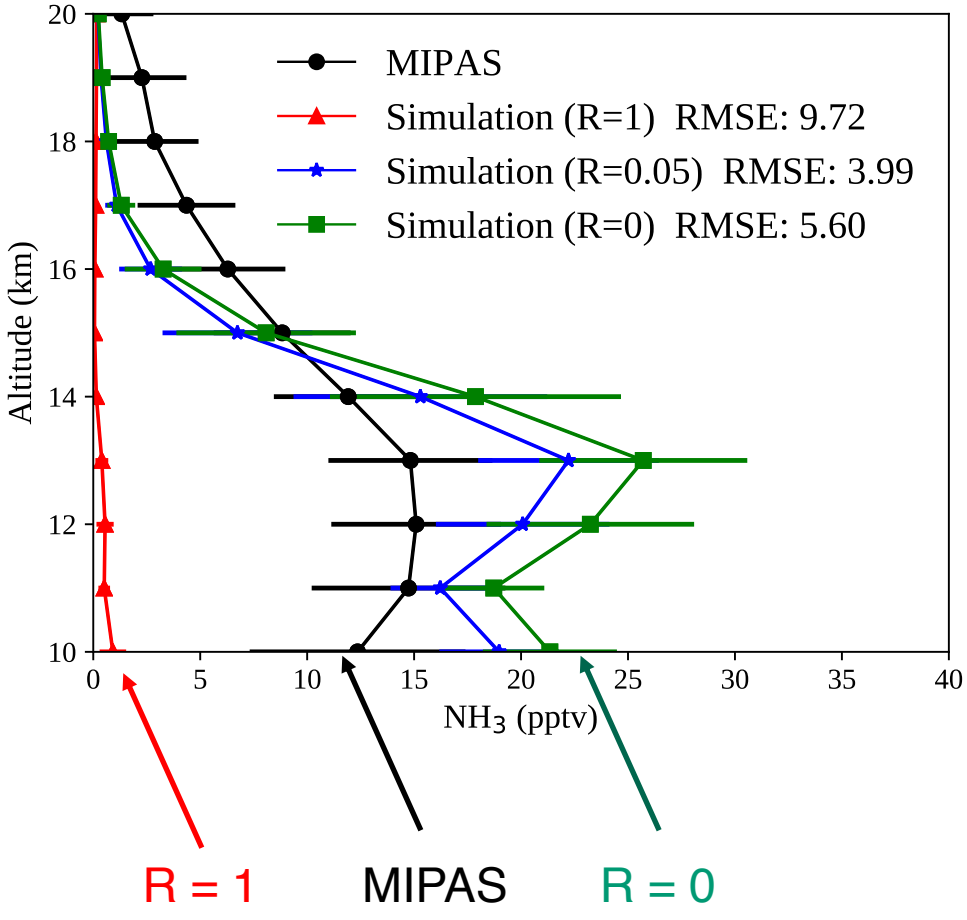
500hpa – tropopause

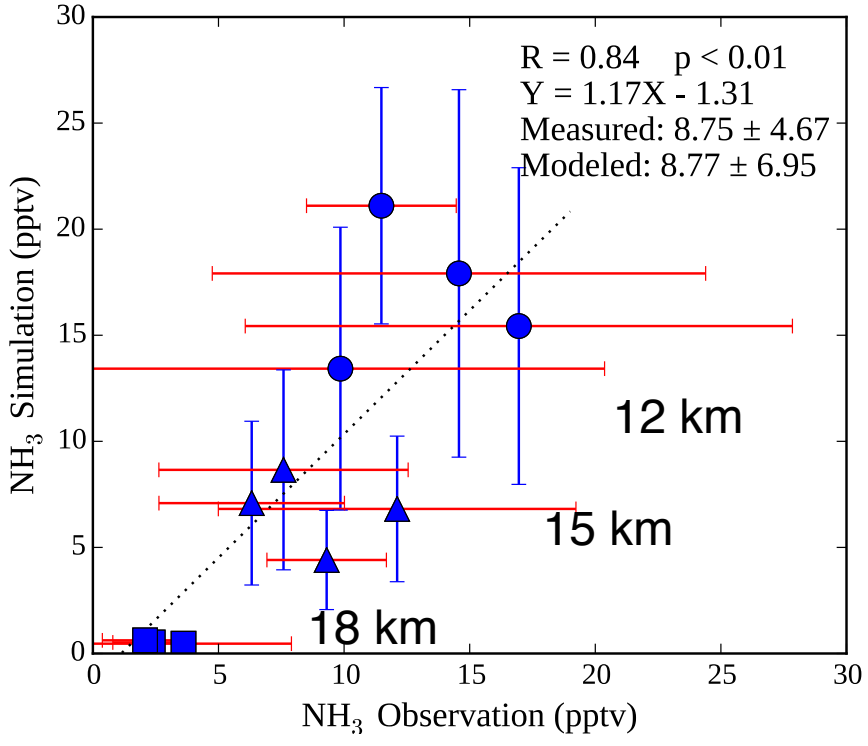
Wang, Hoffmann, Park, Jacob, Martin, 2008, *JGR.*

Comparison with MIPAS



Quantitative comparison



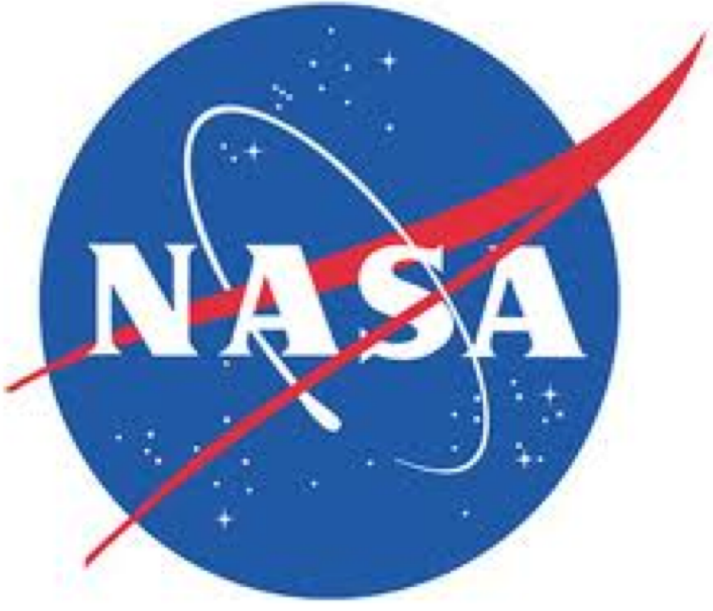


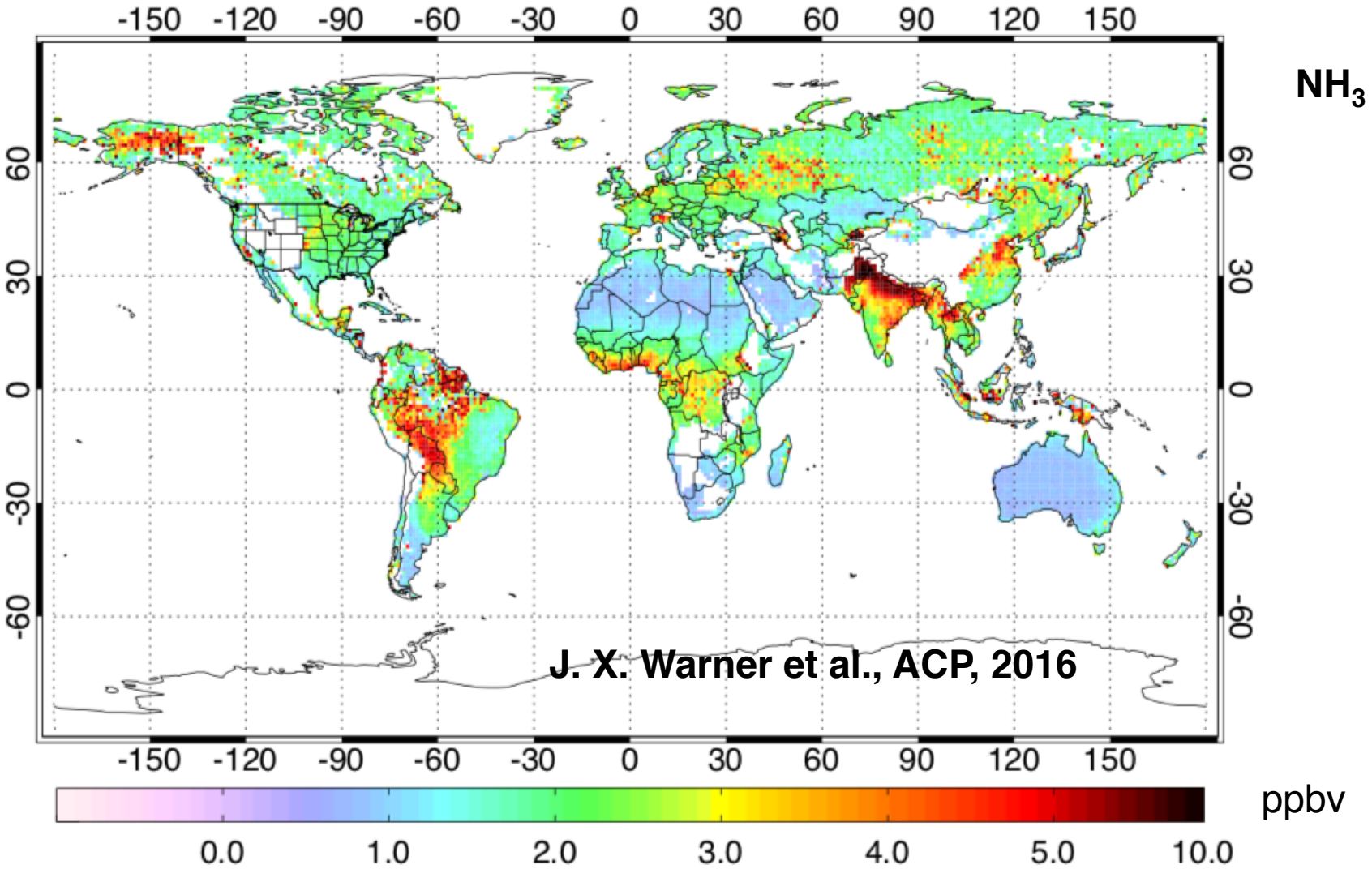
Results are from 2003 and 2007-2011

Summary and Discussion

- **The retentions of gases upon freezing of water from available studies are conflicting.**
- **Using molecular dynamic simulations (MDS), we have revealed that the retention efficiency of NH_3 upon freezing cloud is close to 0 rather than 1 during deep convection.**
- **Implementation of SMD results in GEOS-Chem yield a much better agreement with aircraft and MIPAS data for NH3 in UTLS available in the past 20+ years.**
- **Because NH3 emission is not regulated in most countries and its future increase is likely persistent from agricultural growth and the warmer climate, the ef- fect of NH3 on composition and phase of aerosol particles in the UTLS can be significant, which in turn can affect cirrus cloud formation, radiation, and the budgets of NOx and O3.**
- **Methodologically, MDS demonstrated as a tool for improving parameterization of interactions between trace gases and cloud (including ice) particles in global atmospheric models.**

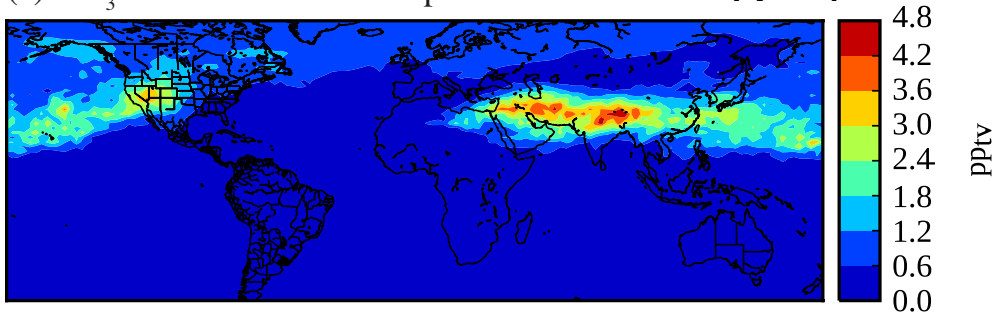
Thank You!



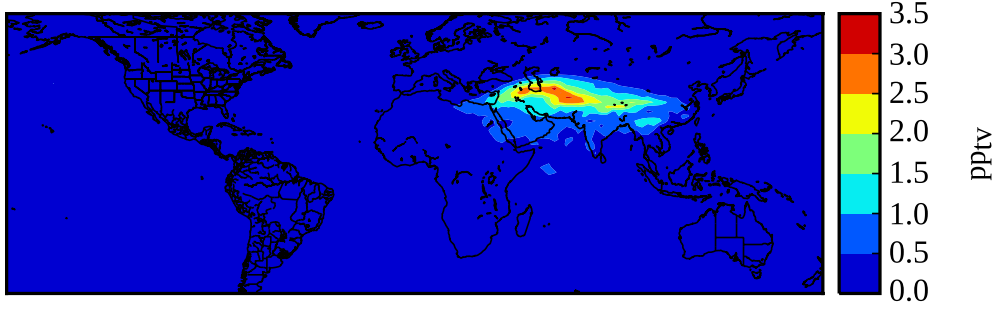

-150 -120 -90 -60 -30 0 30 60 90 120 150
60
30
0
-30
-60
NH3
J. X. Warner et al., ACP, 2016
ppbv
0.0 1.0 2.0 3.0 4.0 5.0 10.0

(a) NH_3 12 km 90% wet deposition

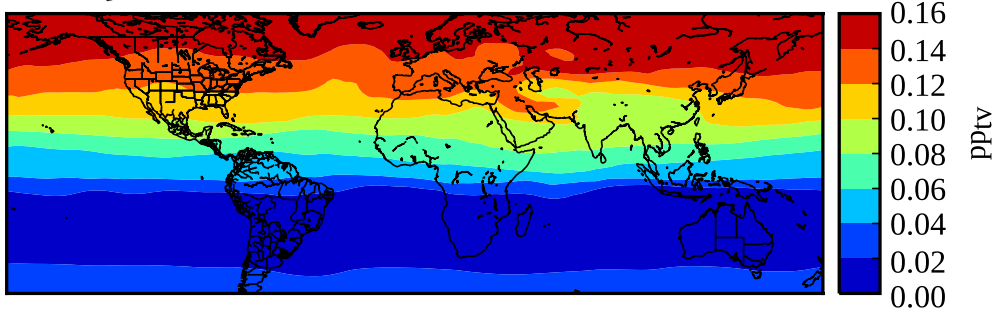
R = 1



(b) NH_3 15 km 90% wet deposition

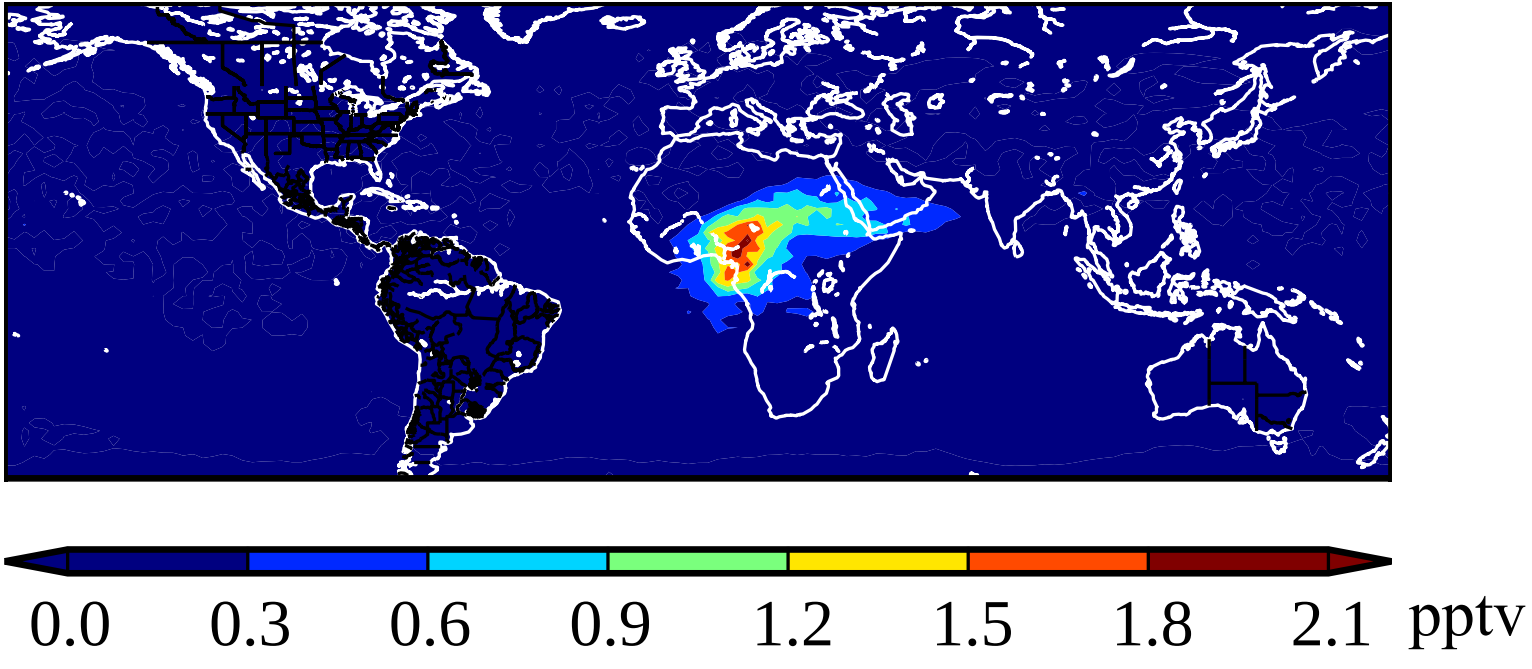


(c) NH_3 18 km 90% wet deposition



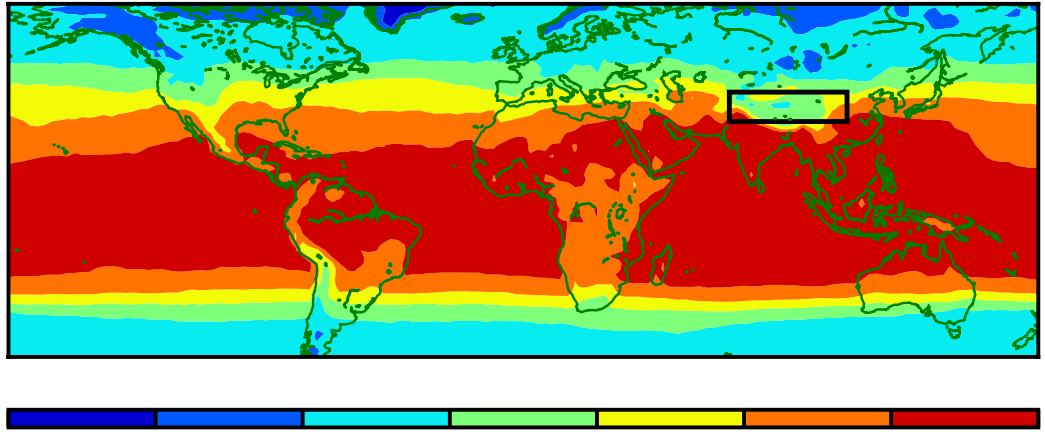
Summer of 2005

$NH_3(R=0.05) - NH_3(R=1)$ 12 km



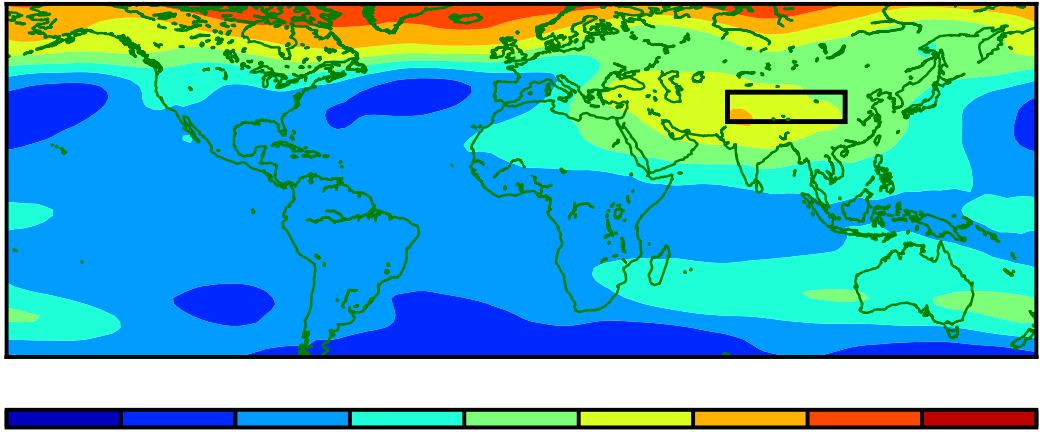
Boreal winter (December, January, February) for the year of 2005.

(a) tropopause height



6.0 7.5 9.0 10.5 12.0 13.5 15.0 16.5 km

(b) air temperature 12 km



212 214 216 218 220 222 224 226 228 230 k