Characterizing the Asian Tropopause Aerosol Layer (ATAL) using in situ balloon measurements: the BATAL campaigns of 2014-2016

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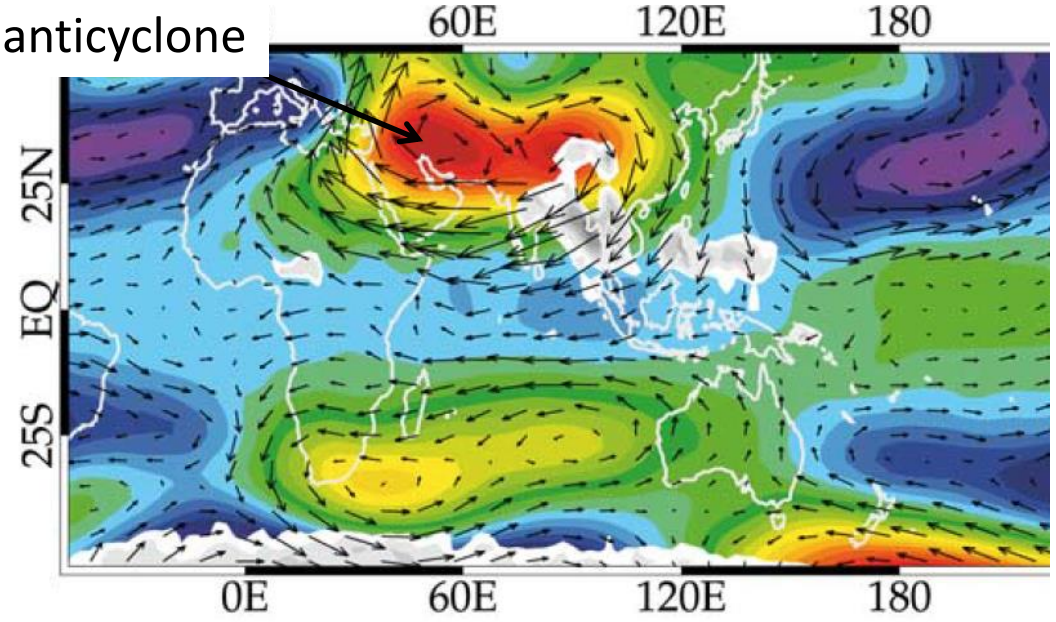
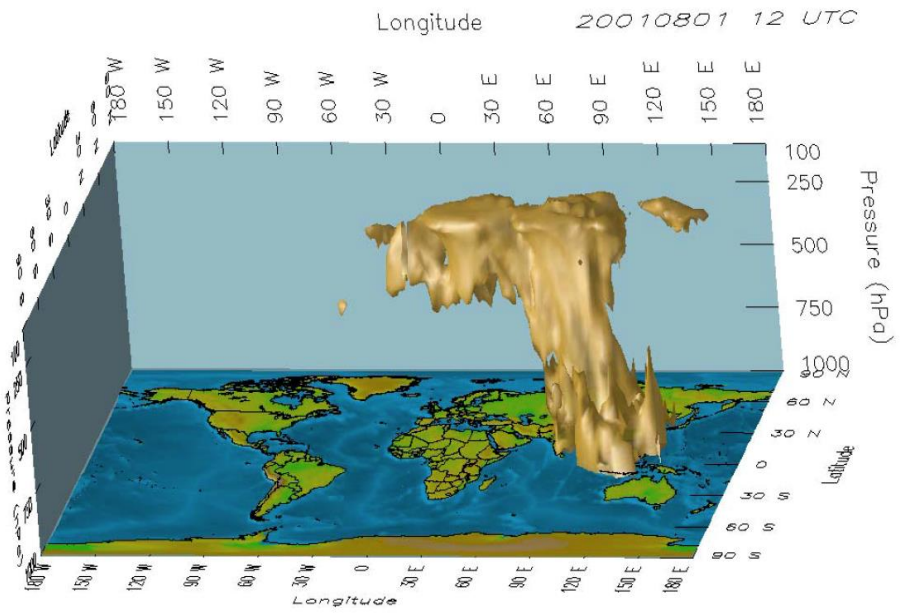
Transport of pollution into the UTLS linked to deep convection in Summer Asian Monsoon

ASM anticyclone

MLS/CO July-Aug 05 100hPa



Park et al. (2007)

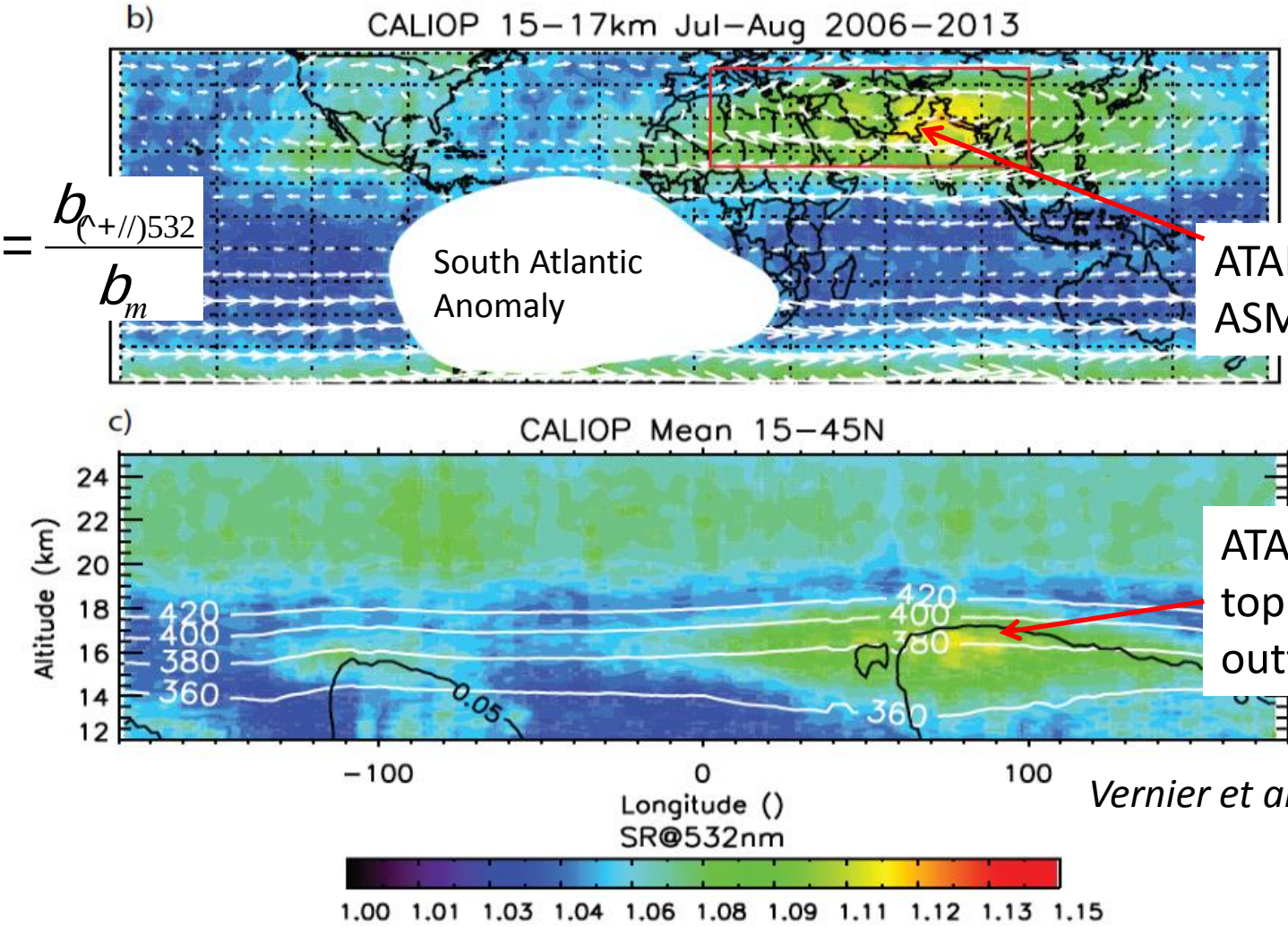


Lawrence and Lelieveld., ACP, 2010, MATCH-MPIC south Asia CO tracer

- **Asian Summer Monsoon (ASM) provides a vehicle for transport of BL gas-phase pollutants (e.g. CO, HCN, CH4) into the UTLS (Park et al., 2007; Randel et al., 2010, 2011).**
- **An Asian Tropopause Aerosol Layer (ATAL) has been found in observations (Vernier et al., 2011). Is ASM also a source of aerosols to the Lower Stratosphere?**
- **What is the origin, composition, size of ATAL aerosols, and potential climate impact?**
- **Very few *in situ* observations.**

The Asian Tropopause Aerosol Layer:
Cloud-cleared aerosol scattering ratio from **CALIPSO**

$$SR = \frac{b_{(\wedge+//)532}}{b_m}$$



ATAL associated with ASM anticyclone

ATAL extends from top of convective outflow

Vernier et al., JGR 2015

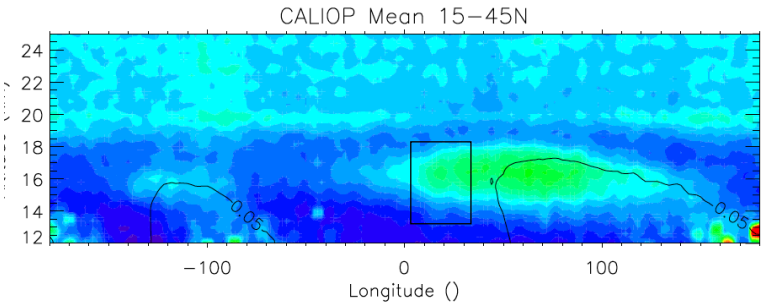
$$\P = \frac{b_{(\wedge)532}}{b_{(//)532}}$$

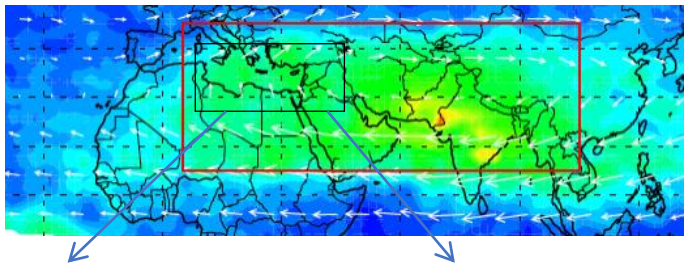
- Cloud clearing of CALIPSO backscatter using 532 nm depolarization cutoff of 5% reveals ATAL
- Buildup of enhanced aerosol associated with Asian Summer Monsoon (ASM) anticyclone, extending from the E. Med Sea to China
- Extends from top of convective outflow (black contour) in much of SE Asia

Science Questions

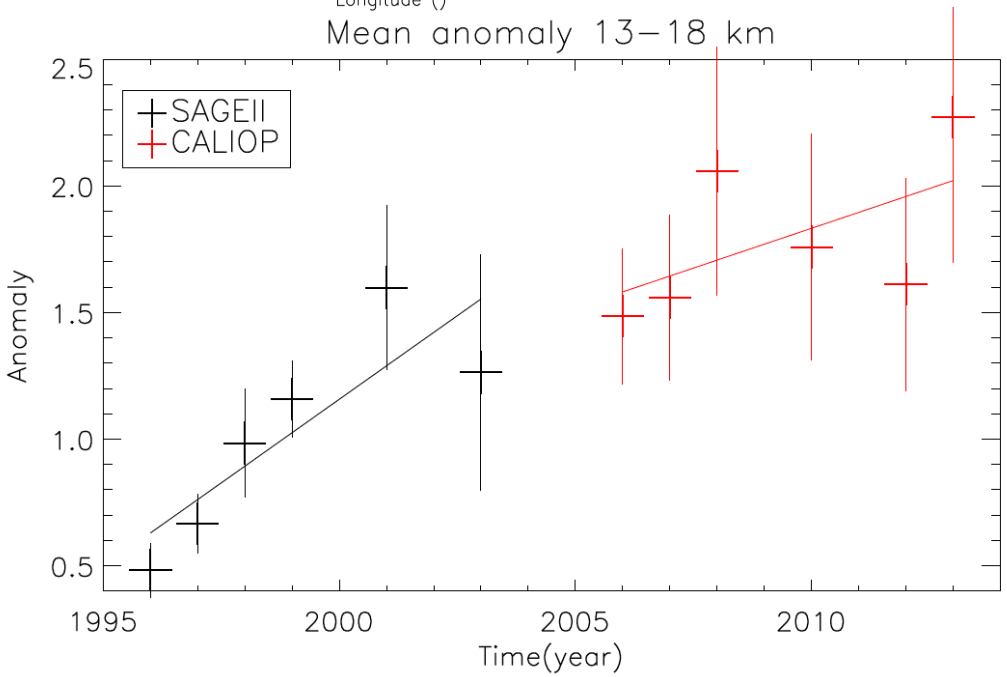
- What is the origin of the ATAL?
- What is the **size and composition** of particles within the ATAL ?
- What is the relationship between ATAL **aerosols and ice clouds**?
- Is ATAL composition well mixed throughout the ASM anticyclone?
- How do aerosol properties change along convective outflow of the India monsoon?
- What are the implications for regional climate, UTLS chemistry, and cloud properties?

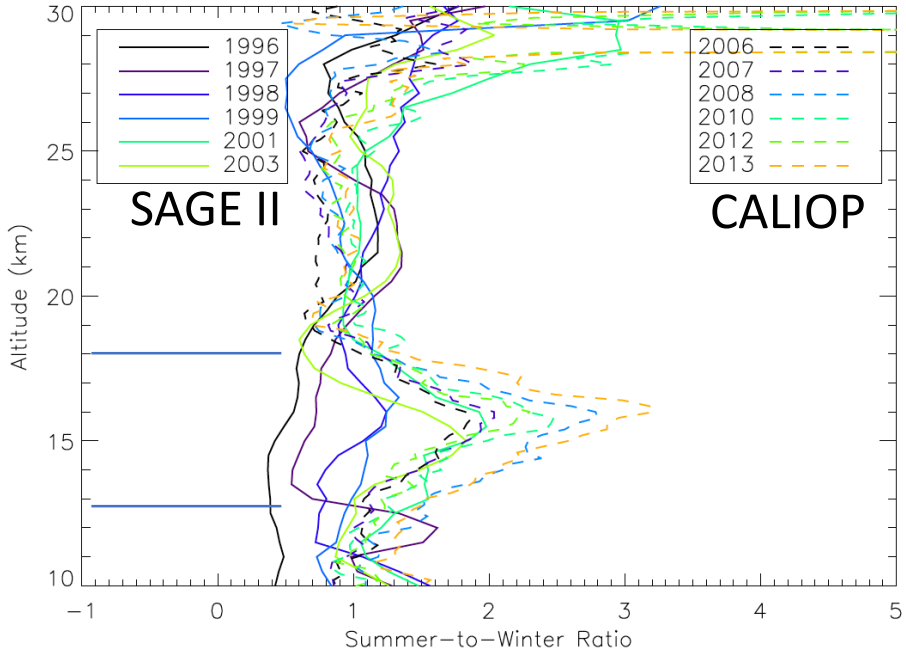
Long-term trend from satellites





30N-40N
5E-45E





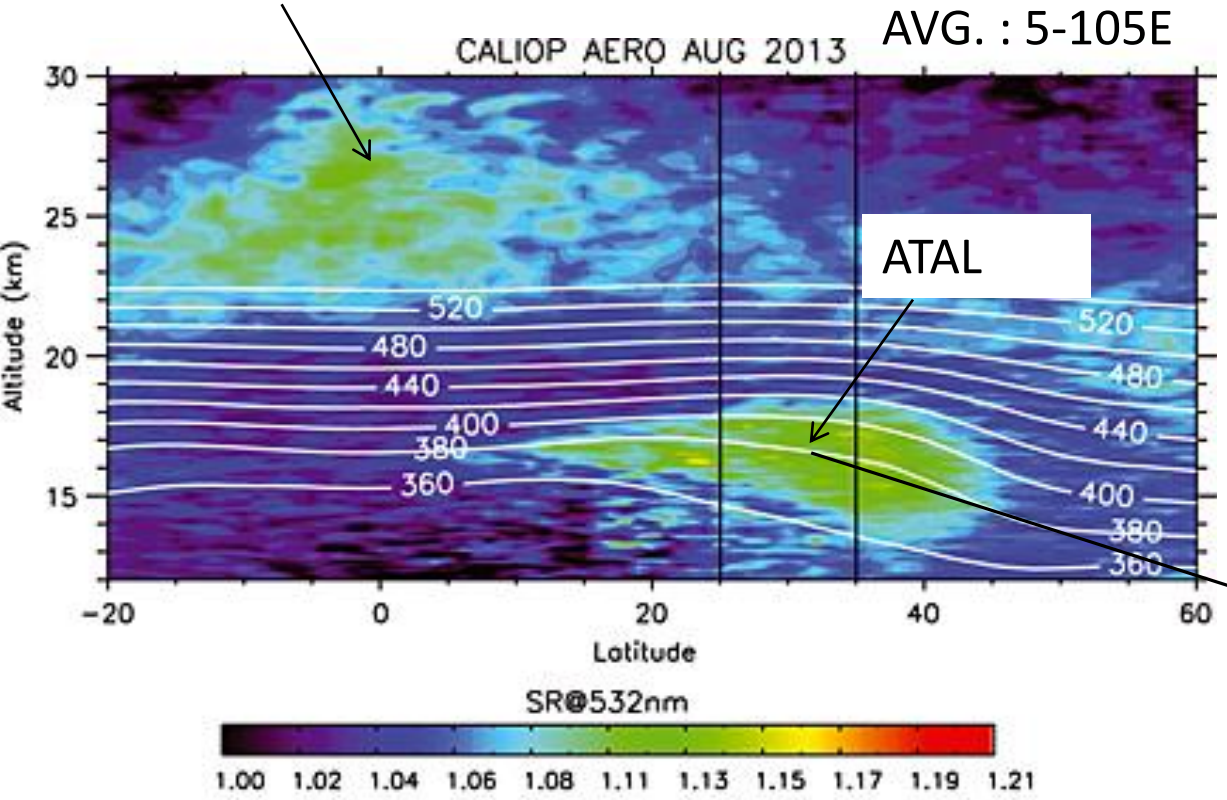
- SAGE II (extinction) and CALIOP (backscatter) summer – winter ratios (anomalies) shows increase in aerosol load over 15 years. No evidence for ATAL prior to 1998.

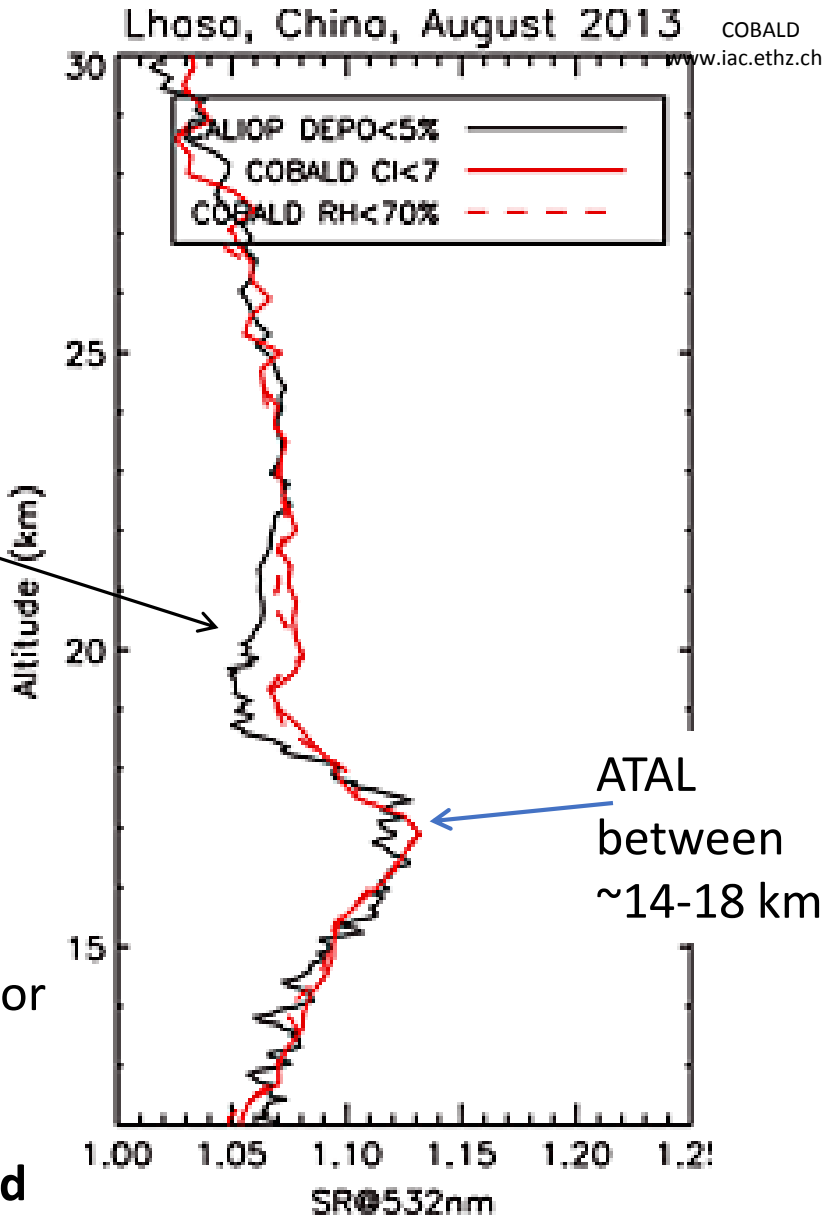
Notes: (i) different measurements, different cloud masks; (ii) selection over the Eastern Med. and Middle East with no ice clouds in the 13-18 km level; (iii) only a few profiles from SAGE II (none in 2000/2002/2005); (iv) CALIOP profiles selected during volcanic quiescent periods

Validation of CALIPSO observations

with balloon-borne backscatter measurements

Stratospheric aerosol layer





- COBALD backscatter data from Lhasa in August 2013 (**SWOP campaign, courtesy J. Bian and F. Wienhold**)
- Multiple cloud-clearing methods (using RH < 70%, Color Index < 7, Depolarization < 5%)
- Good agreement between COBALD and CALIOP
- **Confirms ATAL not the result of unfiltered cirrus cloud**

Vernier et al., JGR 2015

BATAL 2015 : Balloon-borne measurements of the ATAL

5 weeks : July-August 2015 : 30 Launches/ 4 locations/9 Institutes involved

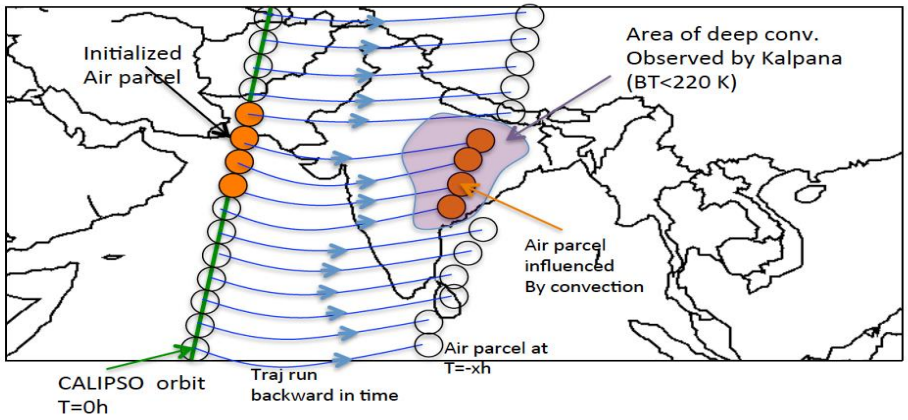
- King Abdullah University of Science and Technology (KAUST), Thuwal, Saudi Arabia, Aug 15
- 6 launches of COBALD backscatter and meteorological sondes

- 15-24 Aug 15 : Banaras Hindu University, Varanasi, India
- 7 launches of aerosol and chemical sensors

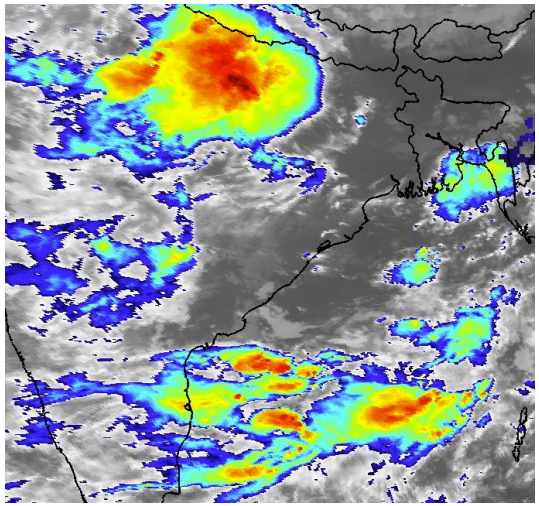
- 29 July-13 Aug 15 : Tata Institute for Fundamental Research Balloon facility, Hyderabad, India, 11 Launches of large and small aerosol, and chemical sensors

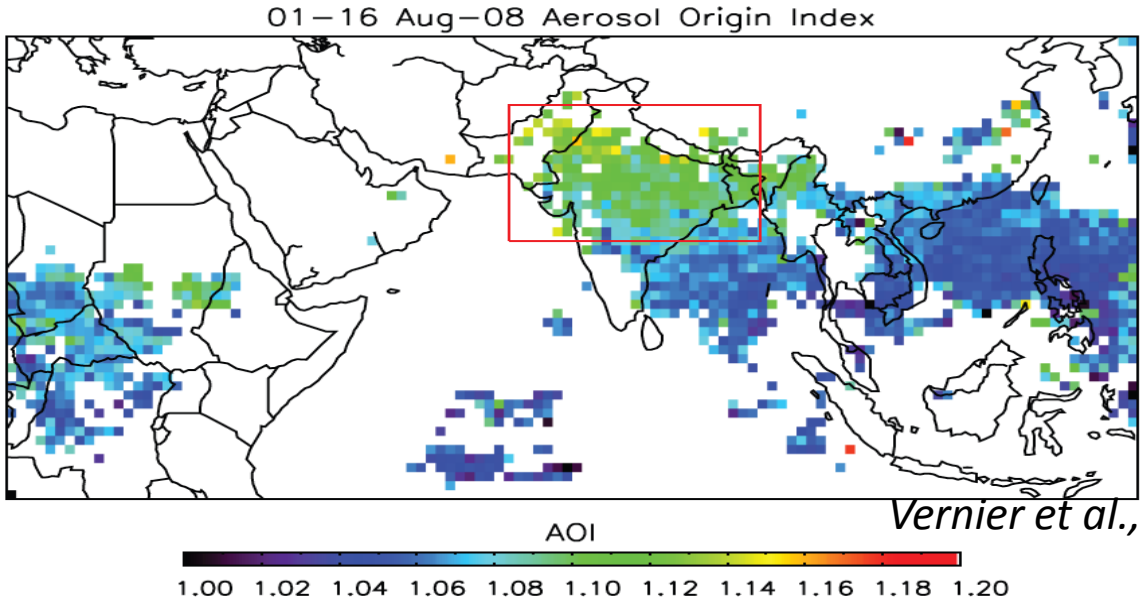
- 17-25 July 15: National Atmospheric Research Laboratory, Gadanki, India,
- 6 launches of aerosol and chemical sensors

ATAL Origin: Trajectory mapping highlights Northern India as key source for elevated aerosol in the ATAL.



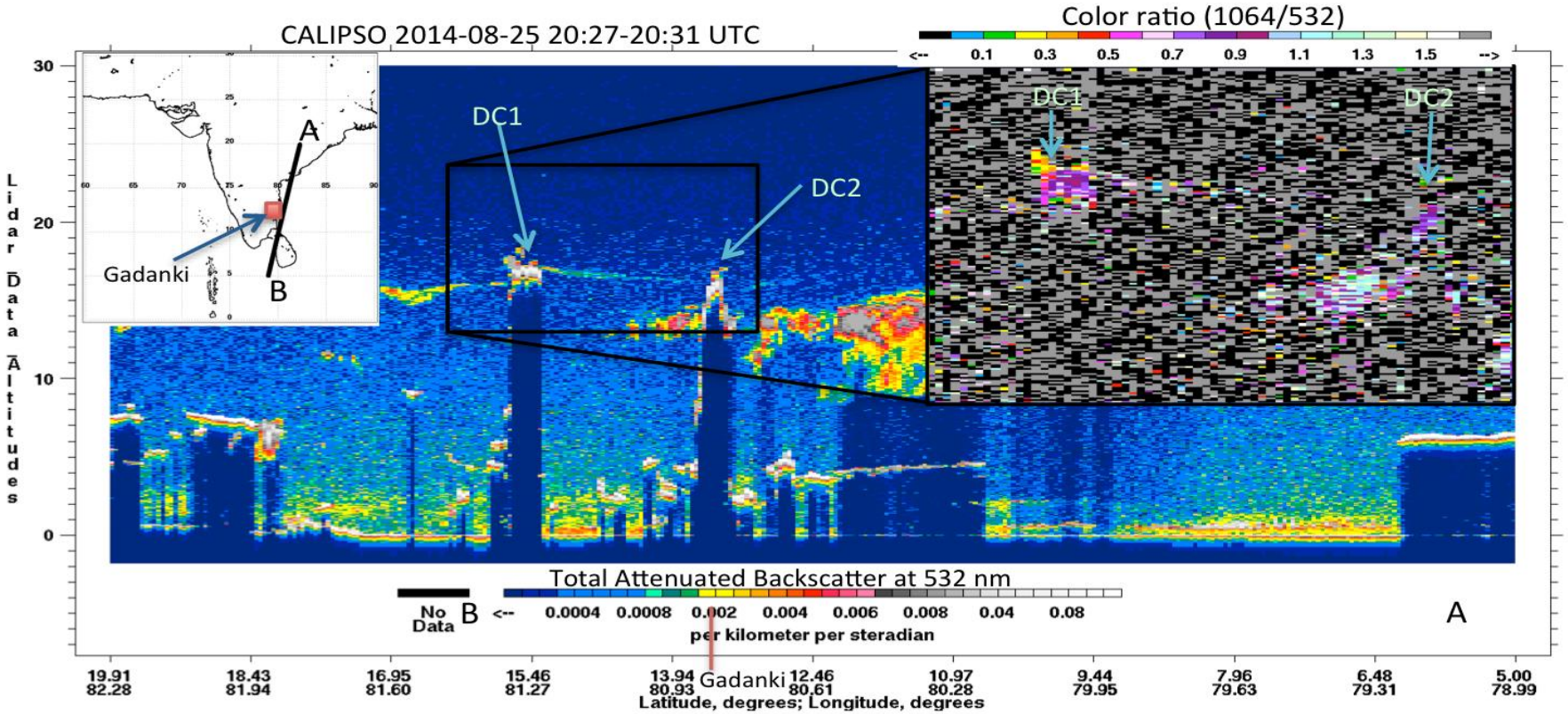
Trajectory mapping of CALIPSO elevated SR to regions of deep convection, 1-16 Aug, 2008

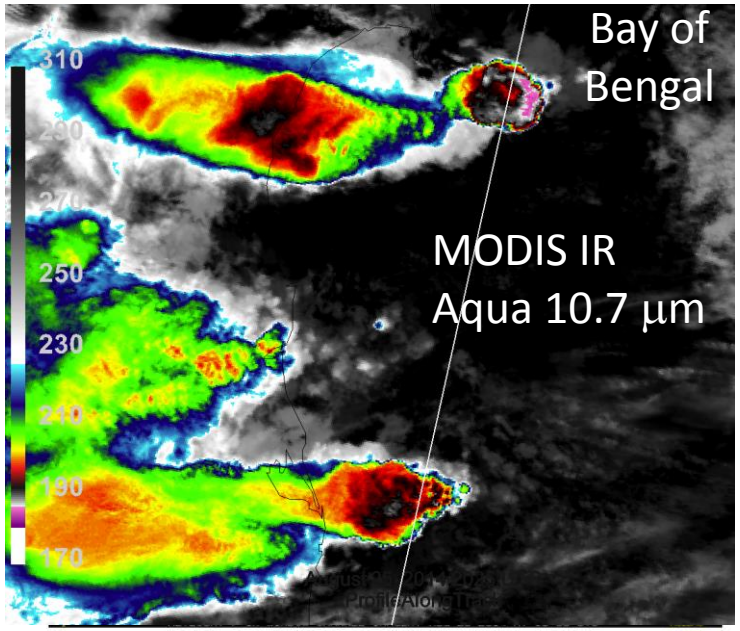


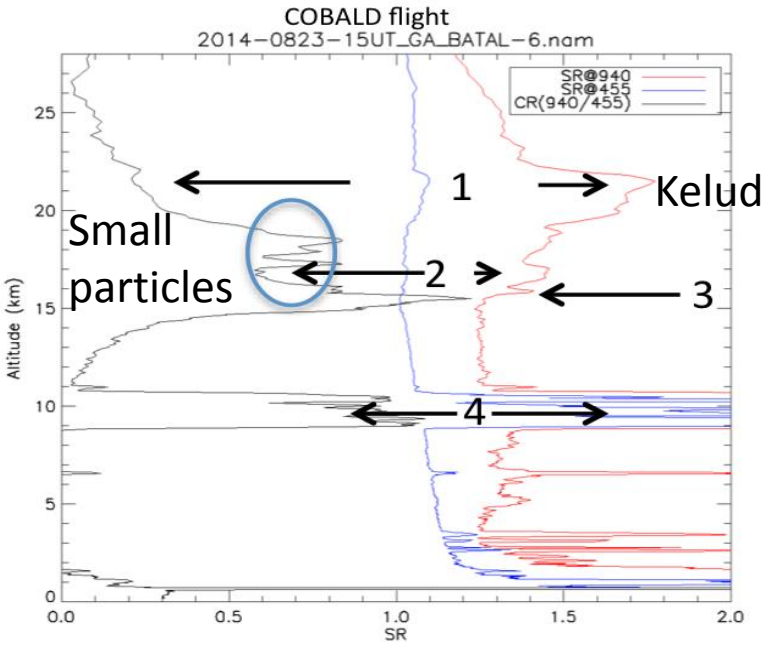


Vernier et al., JGR 2015

Bergman et al., 2013 highlight N. India, Nepal and southern Tibet as a conduit from BL to ASM A/C; **Vogel et al., 2015** show temporal variability in contributions from Indian, Chinese, and S.E. Asian contributions of inert tracers to the ASM A/C.







COBALD flight encountered ice, and small aerosol near the cold point tropopause

- Heavy plastic balloon flights

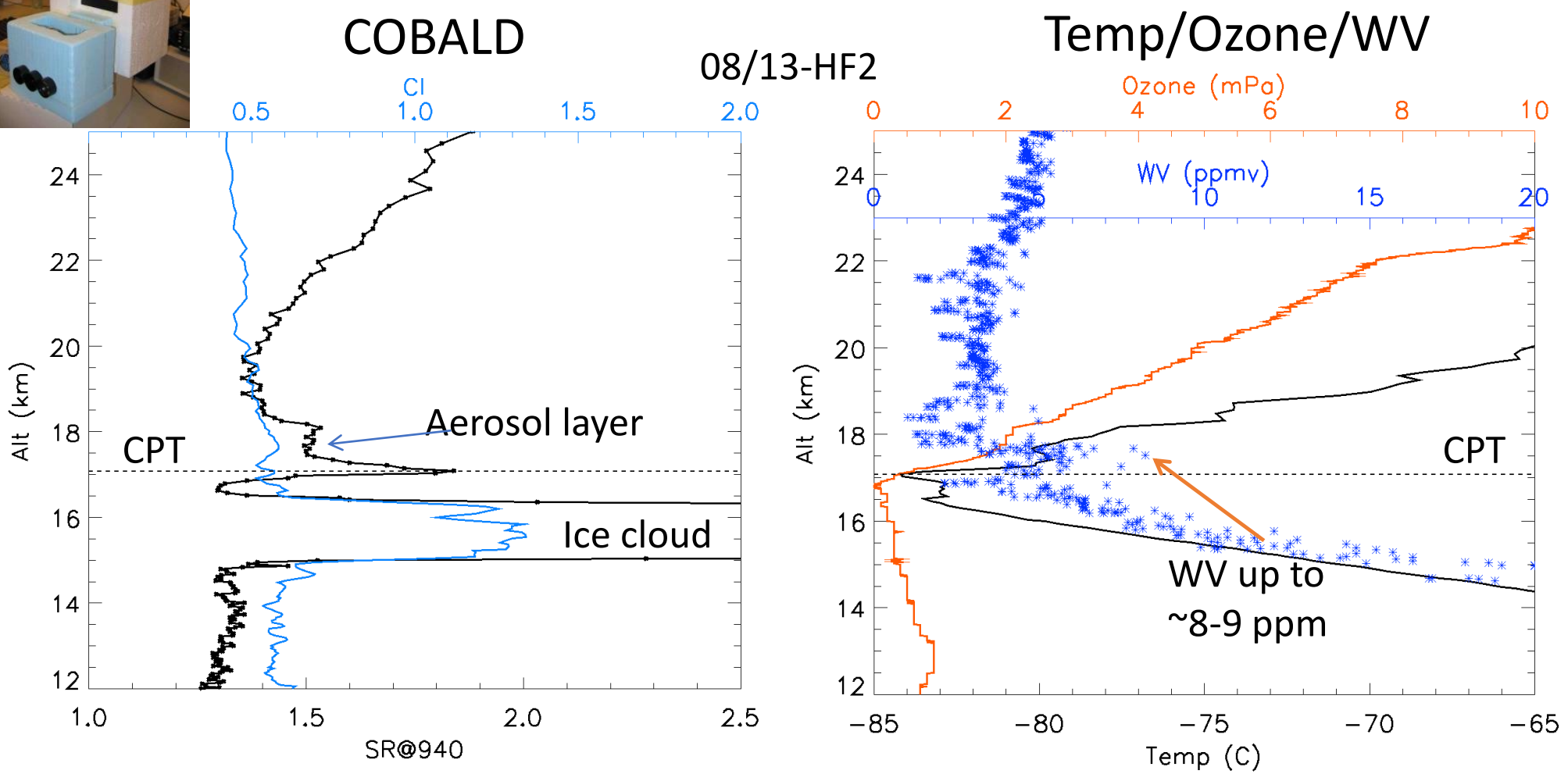
- Regular latex balloon flights (1200g-3000g)

Zero pressure flight near tropopause

- Extended UTLS balloon flights (Boomerang)

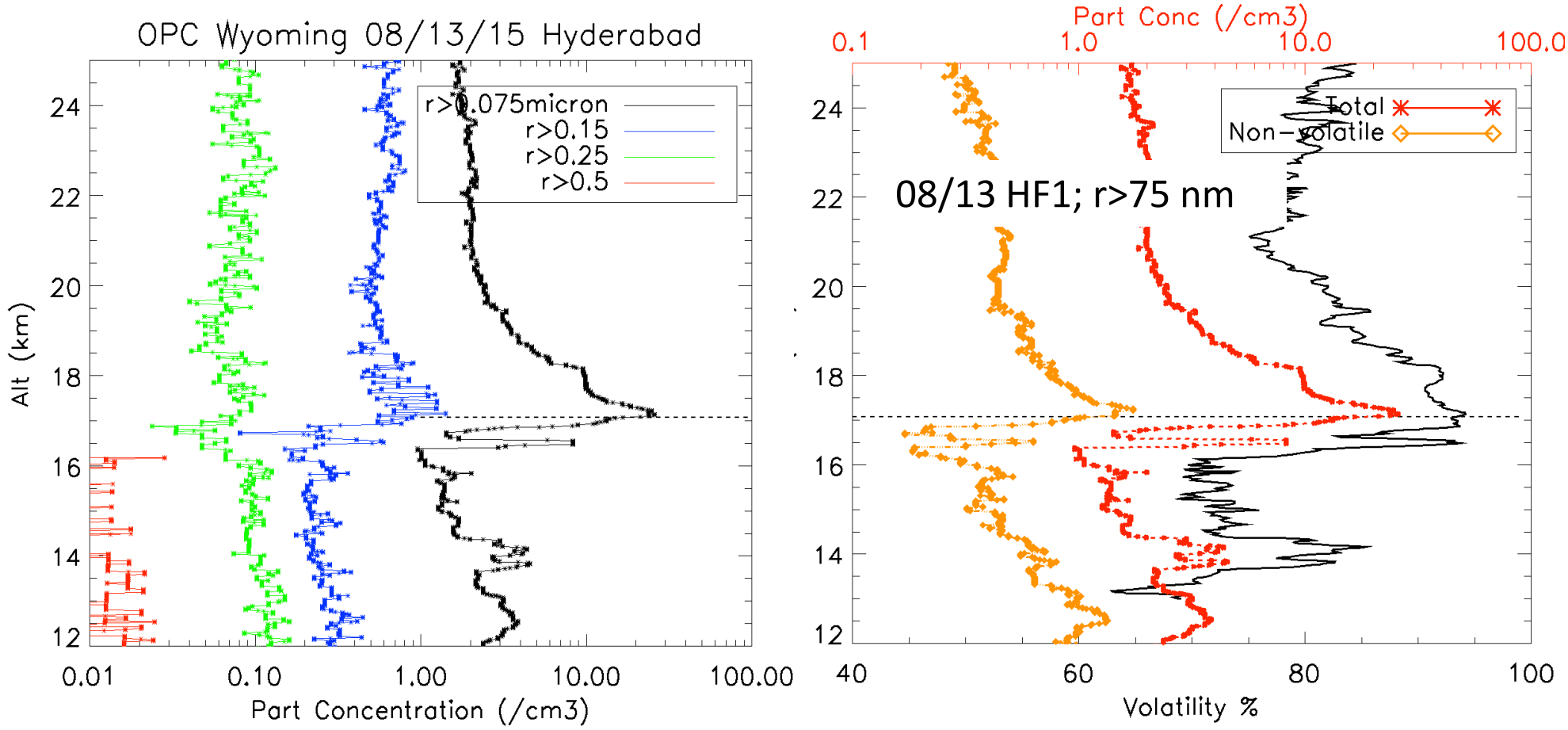
Flight type	Approx. Payload weight	Balloon type	Measurements
Heavy flight (HF)	~50 kg	H_2 filled 3000 m^3 Polyethylene	*Particle size distribution, volatility, ozone, water vapour, particle backscatter and meteorological parameters*
Zero-pressure Flight (ZF)	~26 kg	H_2 filled 300 m^3 Polyethylene	*extended aerosol measurements near tropopause*
Medium Flight (MF)	~8 kg	H_2 filled 2000g/ 3000g Latex	*Combined Particle backscatter/ozone/water vapour/particle size distribution*
Light Flight (LF)	<1.5kg	H_2 filled 1200g Latex	*Particle backscatter/ meteorological parameters*
Boomerang Flight (BF)	~8 kg	H_2/He filled, tandem balloons, 2000g / 1200 g	*Particle sampling using impactor with controlled ascent rate*

Moisture transport evident in the UTLS, HF flight of 8/13/2015 from Hyderabad



- Maximum of aerosol measured by COBALD found in vicinity of cold point tropopause
- Color Index (CI, blue line) between COBALD blue/red channels distinguishes aerosol (0.5 - 0.6) from ice cloud (> 1, 15-16.5 km).
- Enhanced water vapor (up to 8 – 9 ppm) 17–18 km likely due to convective transport of moisture upstream

First size distribution and volatility measurements obtained from the ATAL (OPC, UWY)



- Maximum SR coincides with peak in OPC number concentration for r > 75 nm at the cold point tropopause (data for unheated inlet shown)
- ~97% of particles found in the size range 0.075 < r < 0.15 µm
- Heated (180°C) and unheated inlets on OPC instruments indicate >90% small, volatile particles

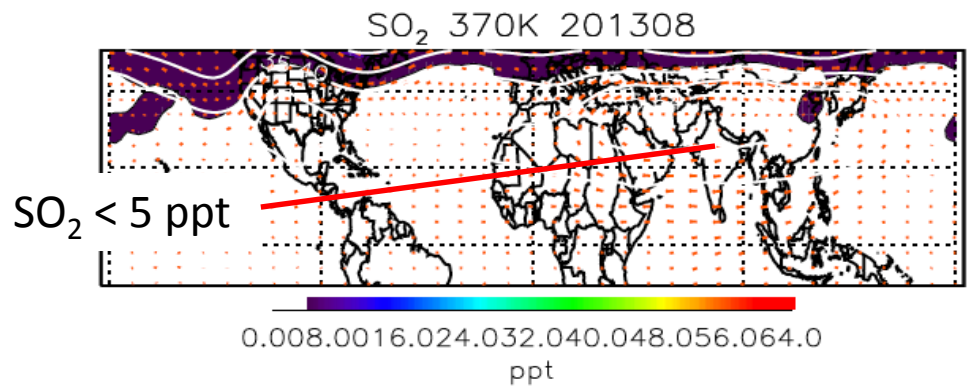
Further pursuit of composition and origin: GEOS-Chem simulations

3-D CTM for gas-phase and aerosols transport and photochemistry in the troposphere, driven by GEOS-5 meteorology (www.geos-chem.org), V9.02, 2x2.5 deg. 72 levels.

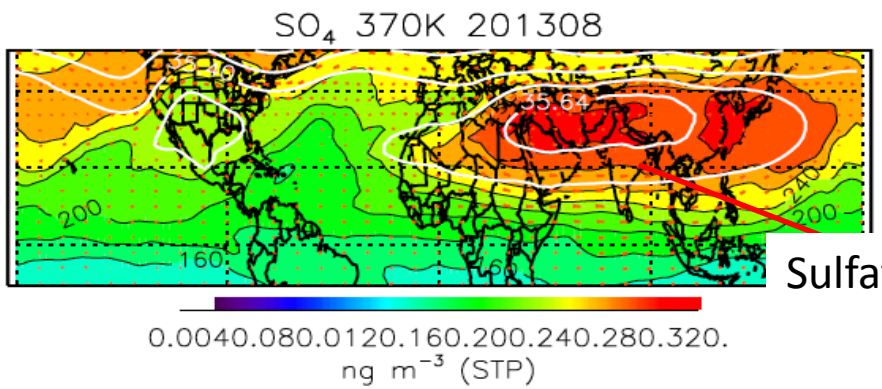
Emissions:
Fossil fuel: EDGAR, with regional options, e.g. Streets (S.E. Asia);
Carbonaceous aerosol: Bond (2007)
Biofuel: Logan and Yevich (2003), with regional options
Biogenic: MEGAN
Biomass Burning: GFED3 (daily)
Volcanic: (SO_2 from AeroCom project)
Aerosols: OC, BC, SO_4, dust , NO_3, limited SOA in current run.

Series of 6 month simulations (1 Apr. 2013 – 1 Oct. 2013)

Simulated ATAL composition: GEOS-Chem SO_2, NH_3, Sulfate, Organic, Ammonium and Nitrate aerosols 370K, August 2013

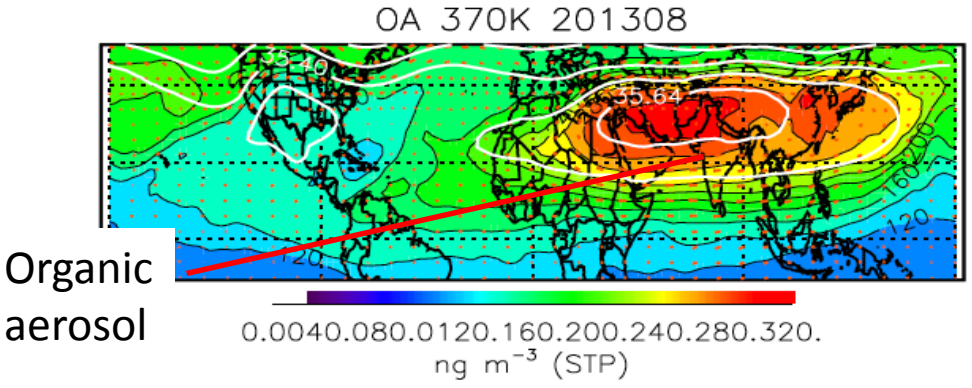


SO_2 < 5 ppt

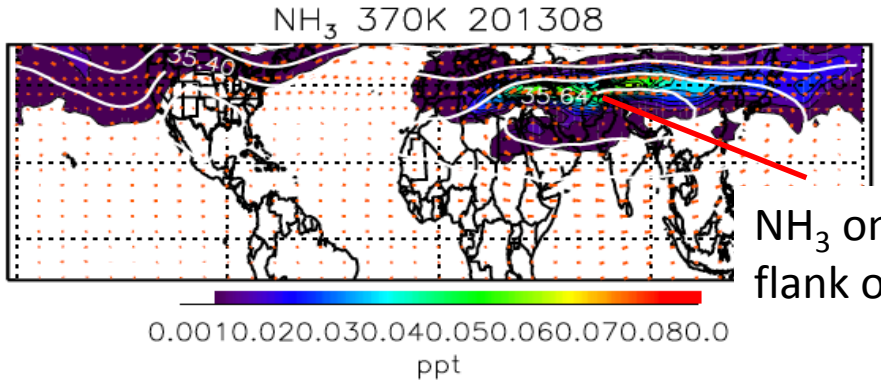


Sulfate

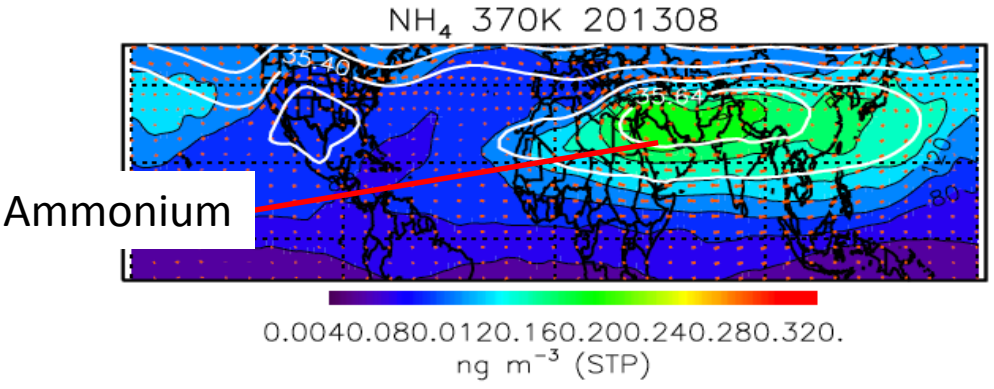
White contours: Montgomery stream function



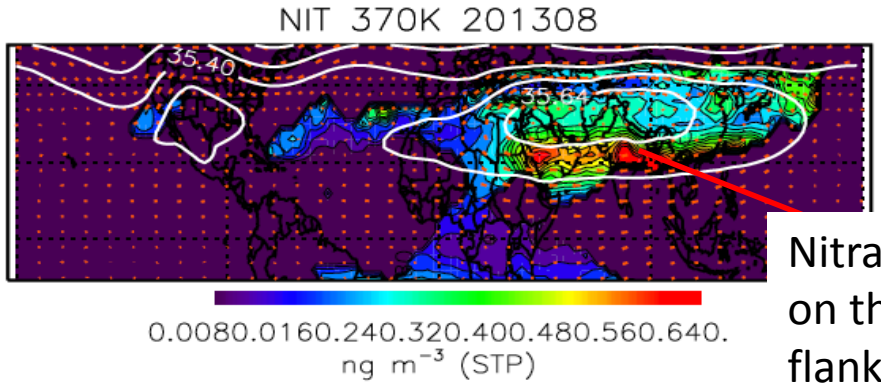
Organic aerosol



NH_3 on N flank of A/C



Ammonium



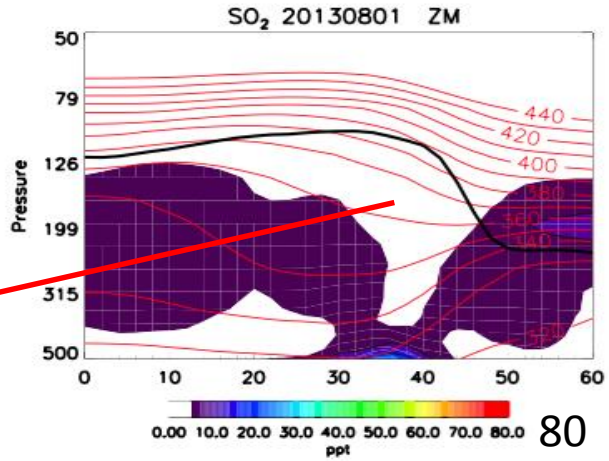
Nitrate peak on the S flank of A/C

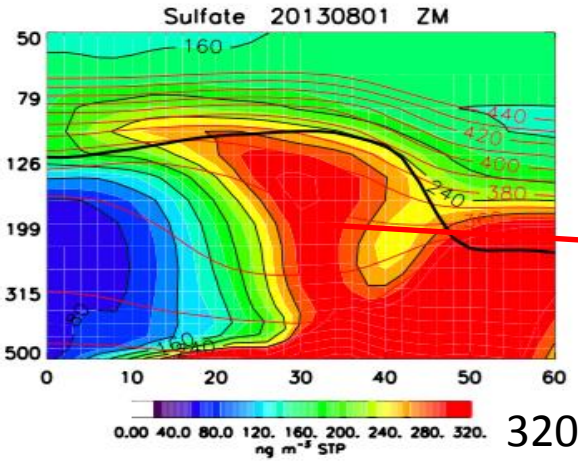
Gu et al. (2016) used GEOS-Chem to deduce nitrate as "the dominant aerosol species" in the ATAL region

Hoepfner et al. (2016) found ~30 ppt NH_3 in MIPAS observations of the ASM anticyclone

ATAL composition: simulated (GEOS-Chem) SO_2, NH_3, Sulfate, Organic, Ammonium and Nitrate aerosols, 30-105°E, August 2013

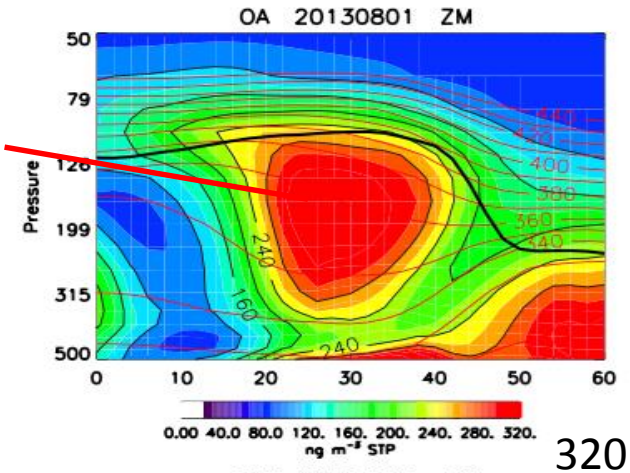
SO_2 < 5 ppt

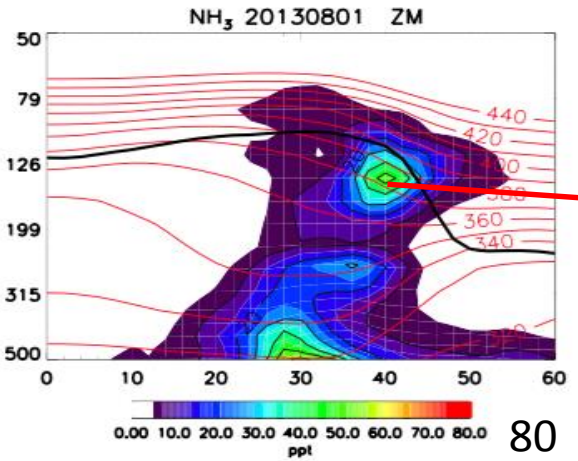




Sulfate

Organic aerosol

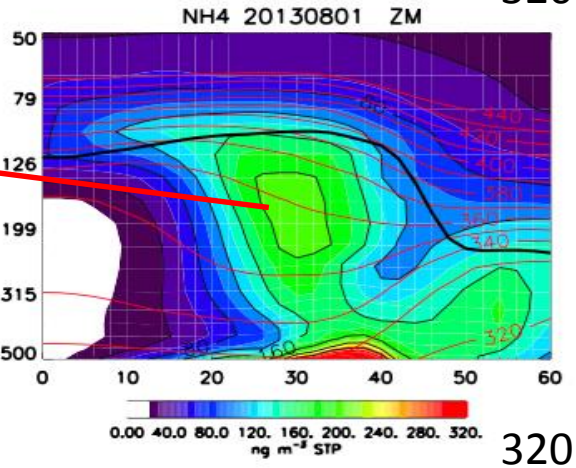


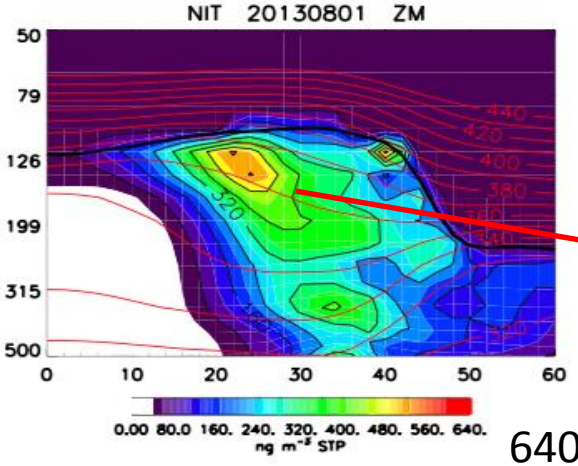


NH_3 in the ATAL

Units: Gas: ppt
Aer: ng m^{-3} STP

Ammonium aerosol

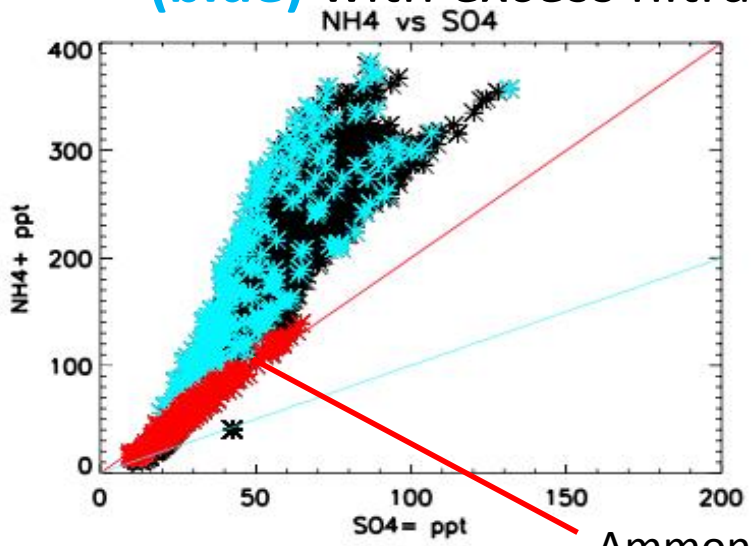


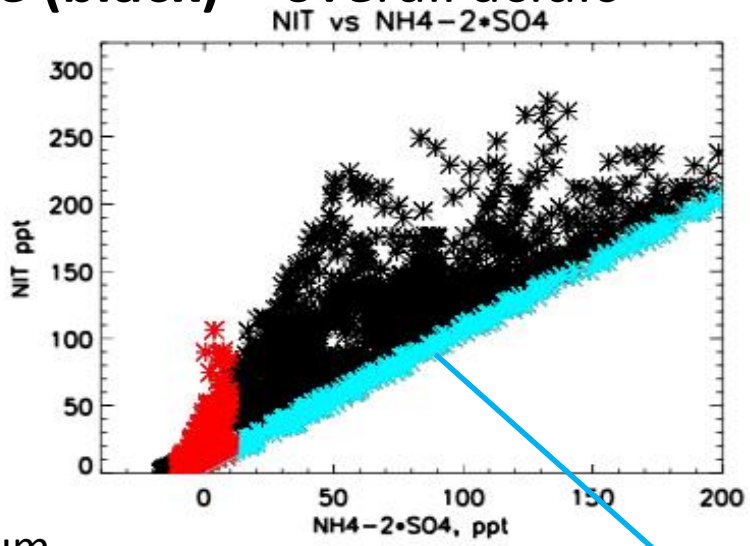


Nitrate aerosol

Simulated ATAL composition: GEOS-Chem, (30-105°E, 15-40°N), 300 hPa – tropopause, August 2013

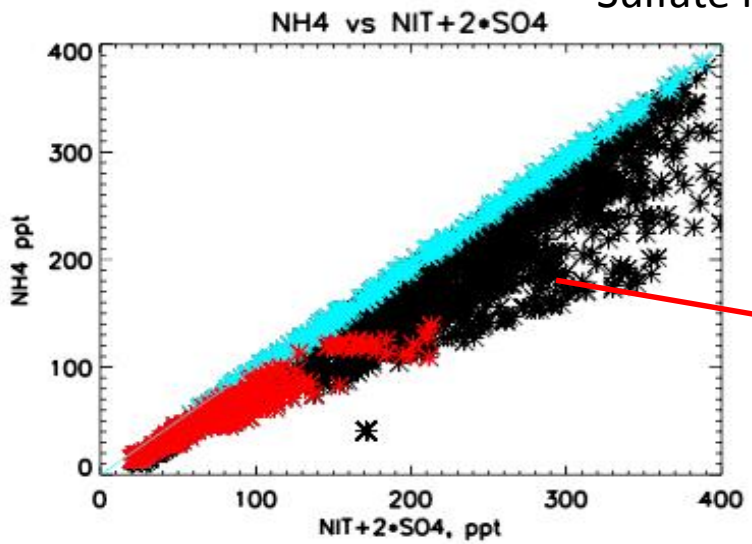
ISORROPIA II yields sufficient ammonium to take up available sulfate, **$(NH_4)_2SO_4$**, **(red)** with excess taken up as ammonium nitrate, **NH_4NO_3**, **(blue)** with excess nitrate **(black)** – overall acidic





Ammonium Sulfate line (**red points**)

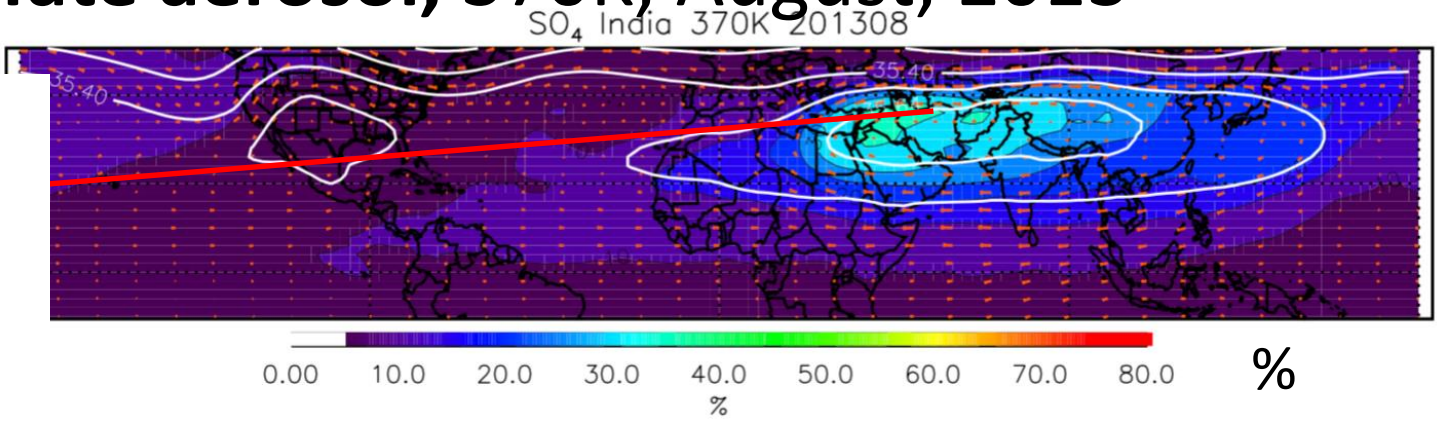
Ammonium Nitrate line (**blue points**)



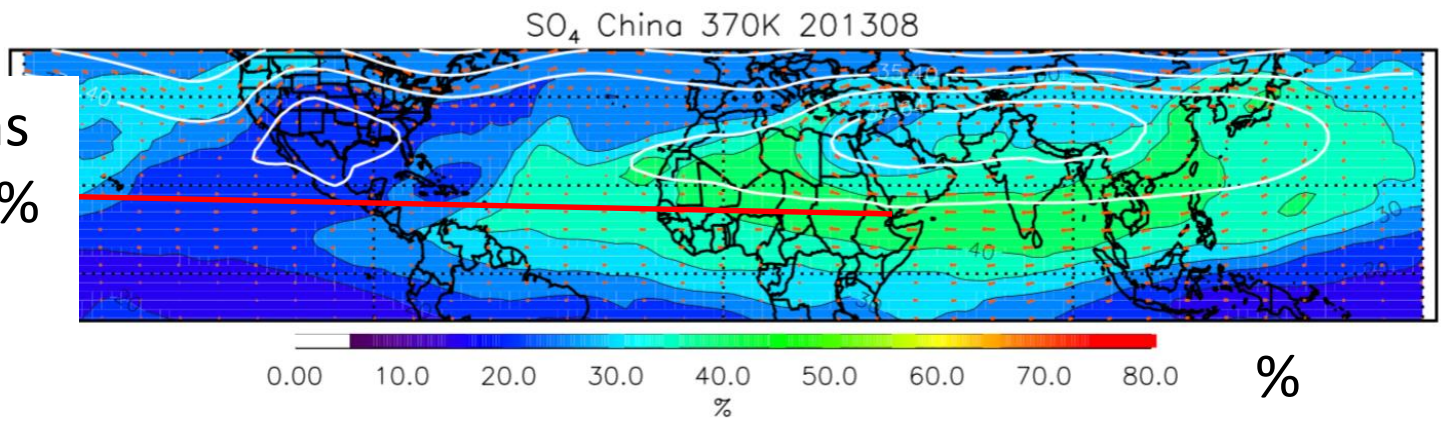
Acidic conditions

Simulated (GEOS-Chem) Source Apportionment of **Sulfate aerosol,** 370K, August, 2013

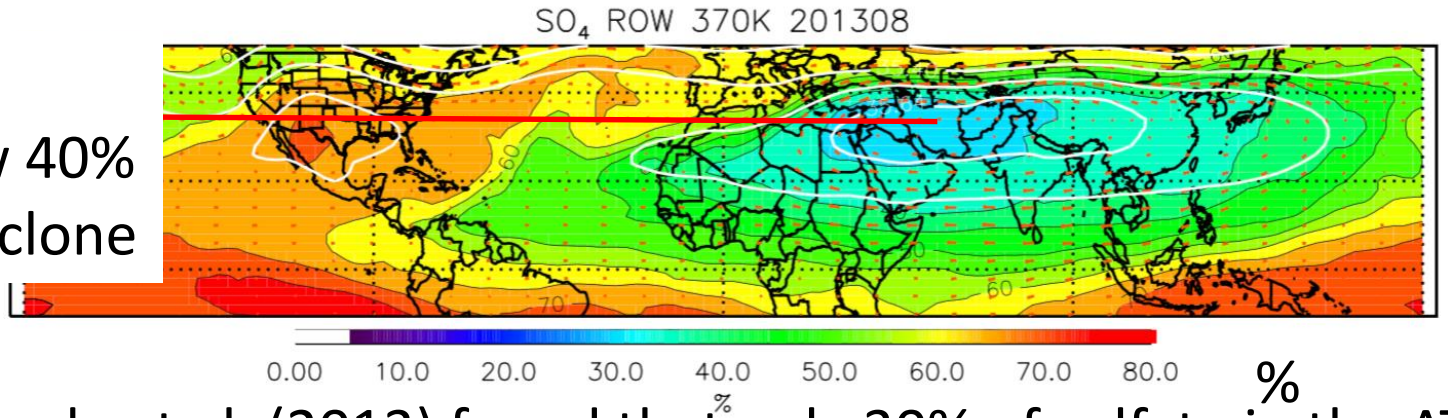
Indian emissions Contribute up to 40% in core of the ASM A/C.



Chinese emissions Contribute 40-50% in a horseshoe around ASM A/C



Rest of the world contributes below 40% In the ASM anticyclone



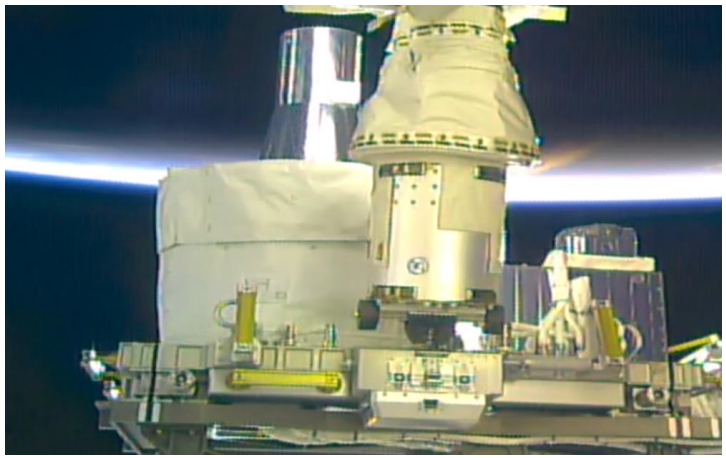
In contrast with Neely et al. (2013) found that only 30% of sulfate in the ATAL due to regional sources

Concluding remarks

- The Asian Tropopause Aerosol Layer is a recurring feature of the UTLS anticyclone associated with the Asian Monsoon, revealed by CALIPSO data
- Our balloon observations show ATAL aerosols in the vicinity of the cold point tropopause in proximity with ice clouds
- Optical Particle Counters indicate that ATAL particles are mostly small (97% D < 300 nm) and mostly volatile (~90%)
- Composition remains uncertain, but GEOS-Chem suggests sulfate-nitrate-ammonium, and organic aerosol
- Modeling studies indicate that sources in E. China and N. India are important contributors to ATAL. Significant interannual variability
- Importance of regional sources is component dependent
- Microphysical simulations (Crumeyrolle) indicate conditions favorable for new particle formation and growth

Next

- BATAL returns to India in 2017
- Fly an **aerosol impactor**, on a "Zero-pressure" flight; combined with ion chromatography to inform composition.
- Fly **Optical Particle Counters** for ranges [0.1 – 5 μm; 5 – 100 μm], and a **Condensation Particle Counter** measuring to a few nm to inform on aerosol and ice cloud particle size.
- **Excited about SAGE III on ISS**, providing continuity with the CALIPSO era,
  measuring O_3, aerosol extinction in nine channels, H_2O, NO_2



- Wish StratoClim 2017 success!
- Established agreements/collaboration with NARL/ISRO (India) present an opportunity for possible airborne campaign in India


Thanks for your
attention