Characterizing the Asian Tropopause Aerosol Layer (ATAL): balloon-borne measurements, satellite observations and modeling approaches

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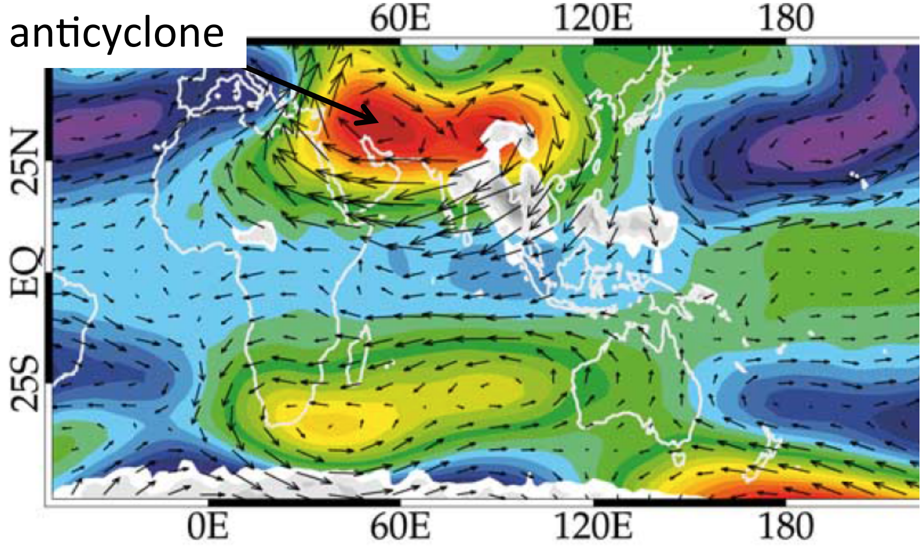
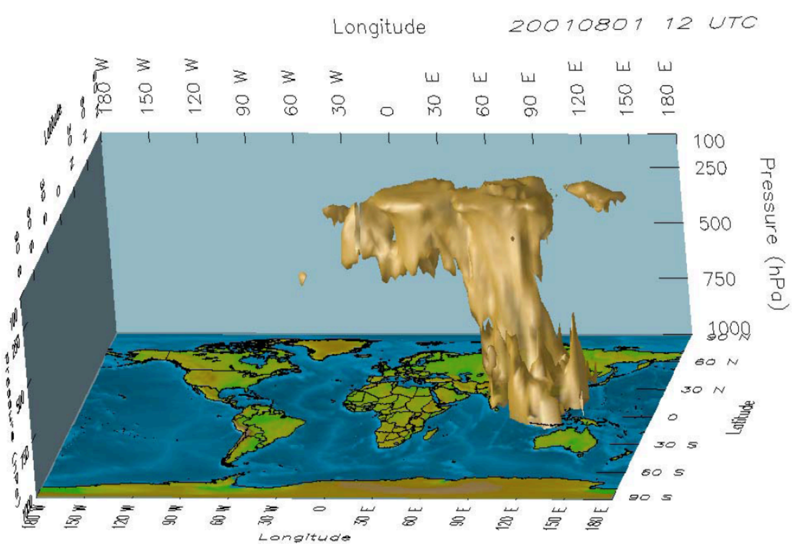
Transport of pollution into the UTLS linked to deep convection in Summer Asian Monsoon



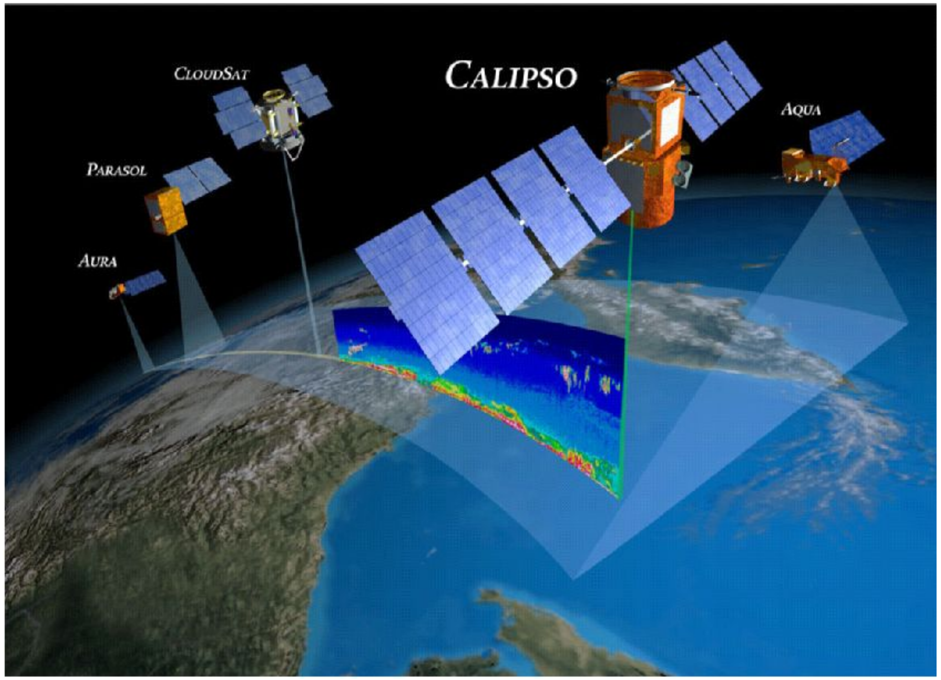
Park et al. (2007)

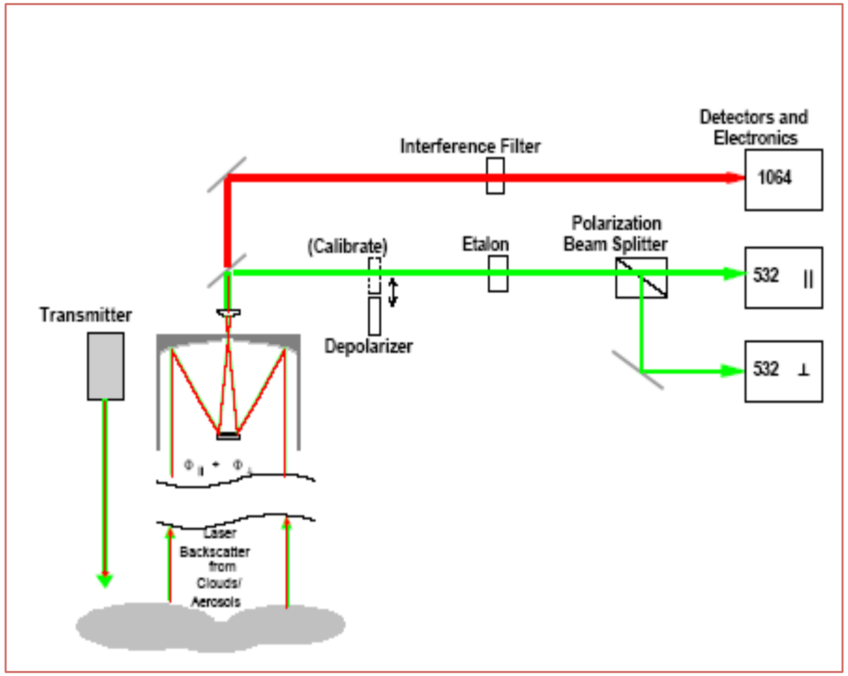


(Lawrence and Lelieveld., ACP, 2010.

- **Summer Asian Monsoon (SAM) can provide 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). Can the SAM also provide aerosols to the LS?**
- **What is the composition, size, origin, and potential climate impact of ATAL**
- **Very few *in situ* observations.**

The CALIPSO satellite lidar





CALIPSO measures attenuated backscatter at 532 and 1064 nm, and depolarization at 532 nm

Scattering Ratio $SR = \frac{\beta_{(\perp+//)532}}{\beta_m}$

~ Particle mixing ratio

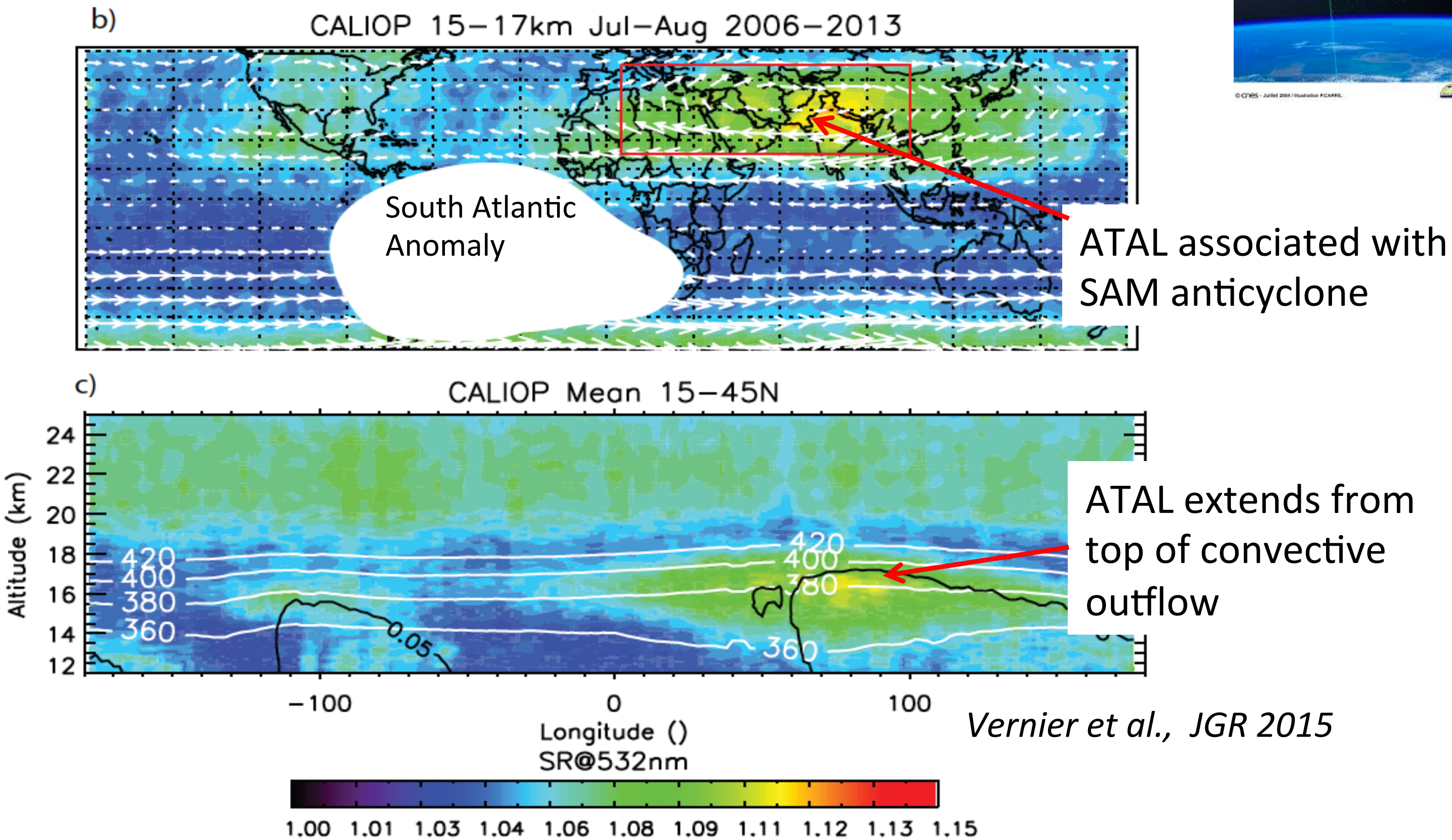
$\partial = \frac{\beta_{(\perp)532}}{\beta_{(//)532}}$ Depolarization

~ particle shape

- CALIPSO nadir view : 80 - 180m resolution in the UTLS

The Asian Tropopause Aerosol Layer:

Cloud-cleared aerosol scattering ratio from CALIPSO

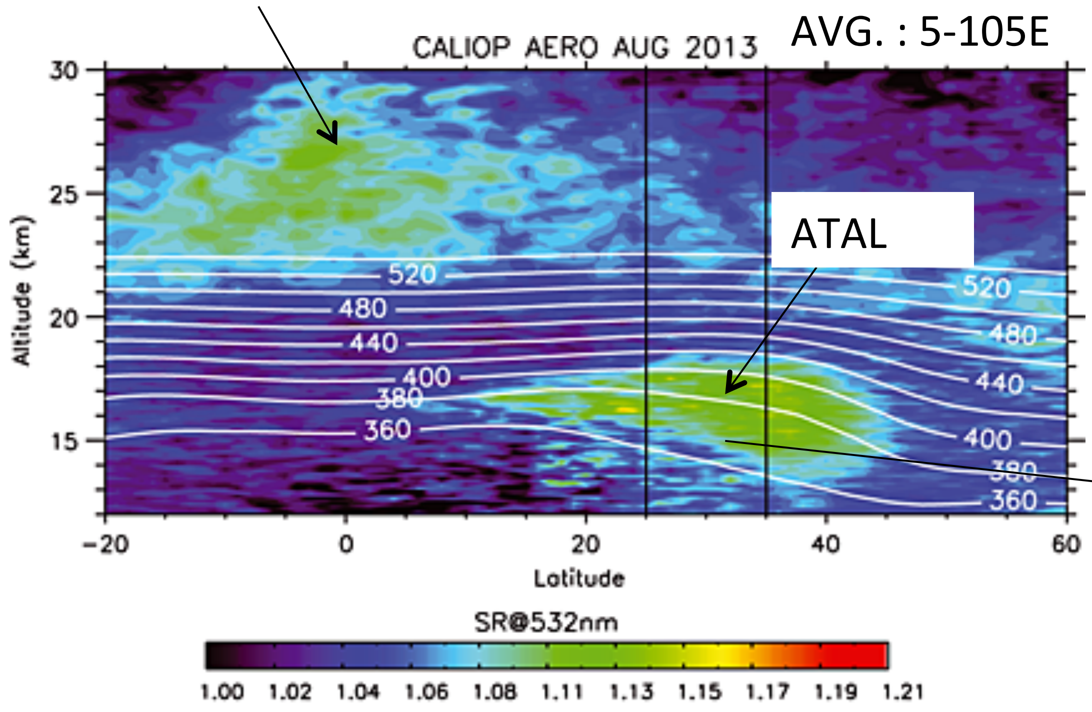


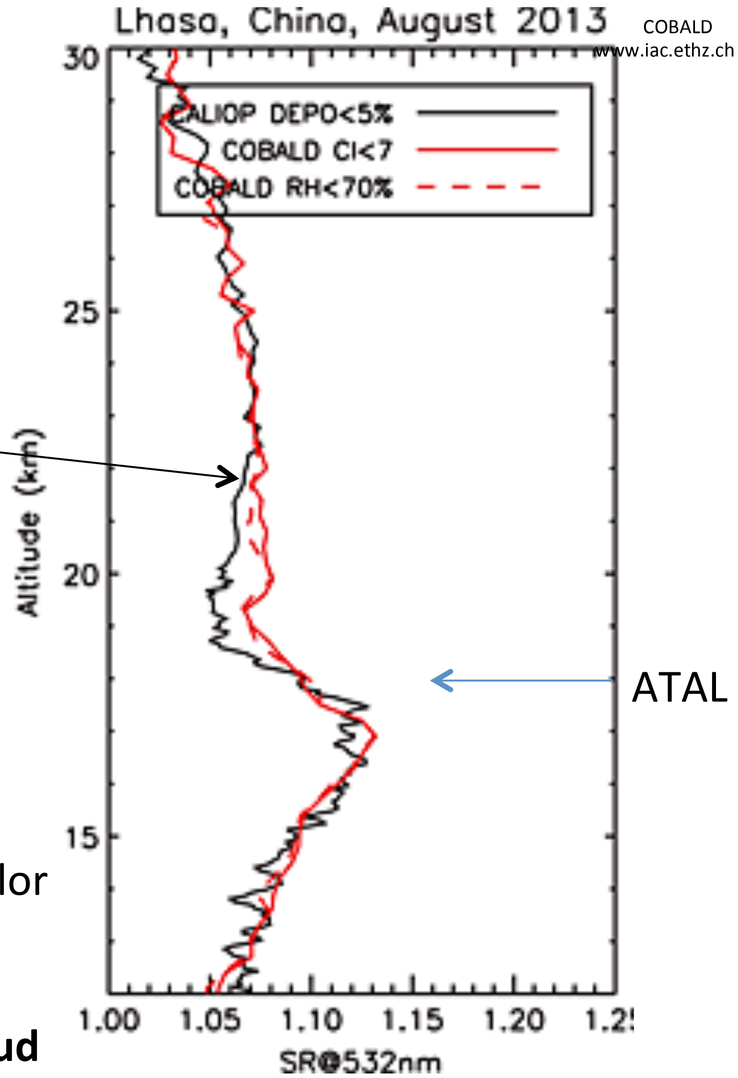
Vernier et al., JGR 2015

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

Validation of CALIPSO observations

with balloon-borne backscatter measurements

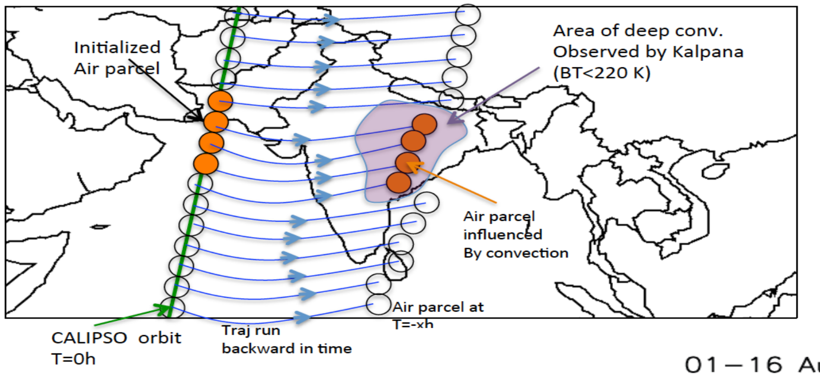




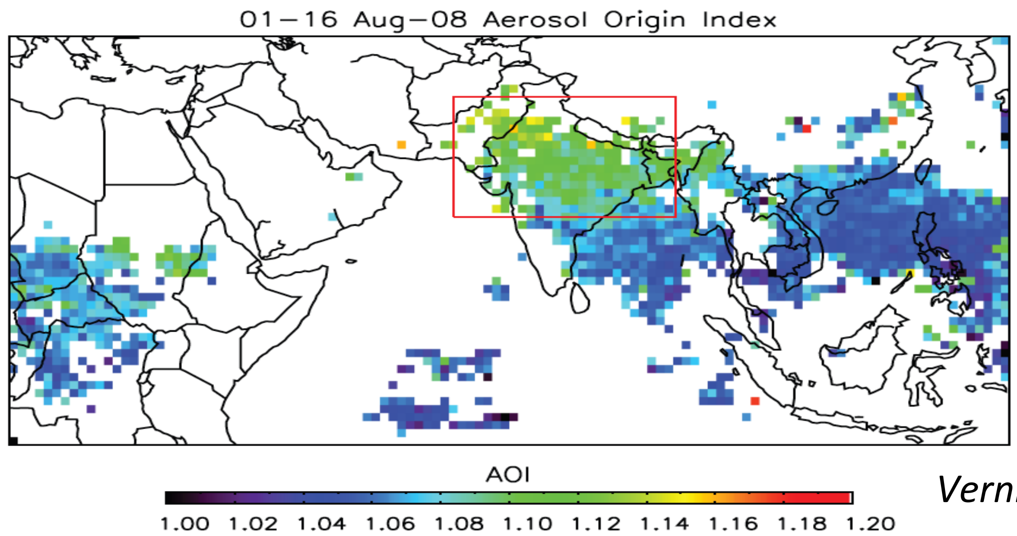
- COBALD backscatter data from Lhasa in August 2013 (**SWOP campaign, courtesy J. Bian and F. Wienhold**)
- Multiple cloud-clearing methods (using RH < 70%, Color Index < 7, Depolarization < 5%)
- Good agreement between COBALD and CALIOP
- **Confirms ATAL not the result of unfiltered cirrus cloud**

Vernier et al., JGR 2015

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



Trajectory mapping of CALIPSO observations to regions of deep convection (BT < 220K from Kalpana), 1-16 Aug, 2008

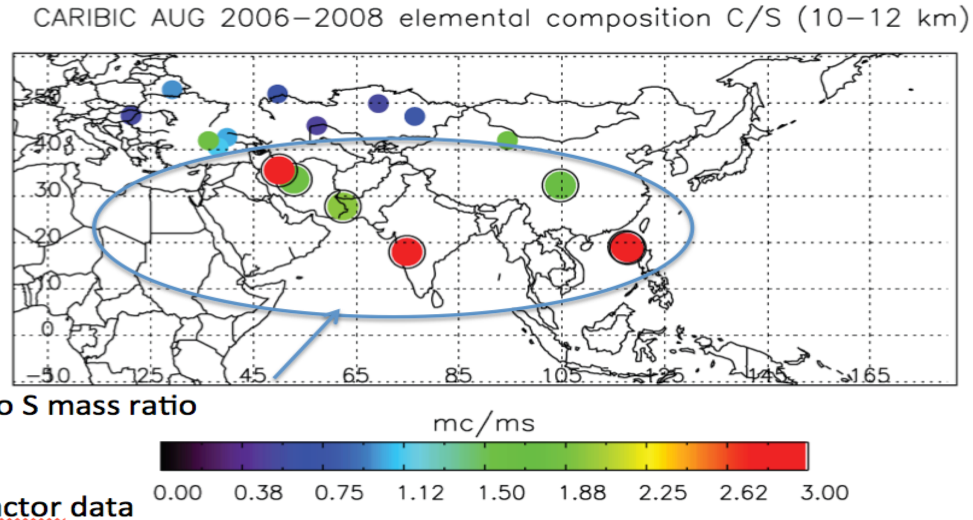


Vernier et al., JGR 2015

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

ATAL Composition: CARIBIC impactor observations from aircraft flights in August 2006-2008 (Martinsson et al., 2014)

Limited in situ observations indicate aerosol composition 10-12 km in lower ATAL mainly Sulfate + Carbonaceous



CARIBIC impactor data
Martinsson et al., 2014

	ng S m^{-3} (STP)	ng C m^{-3} (STP)	C/S
median	12	25	1.8
mean	21	29	2.8
Std	23	15	2.6

PV < 1 PVU; 0° < lat < 45°N;
P < 300 hPa; 20° < lon < 130°E

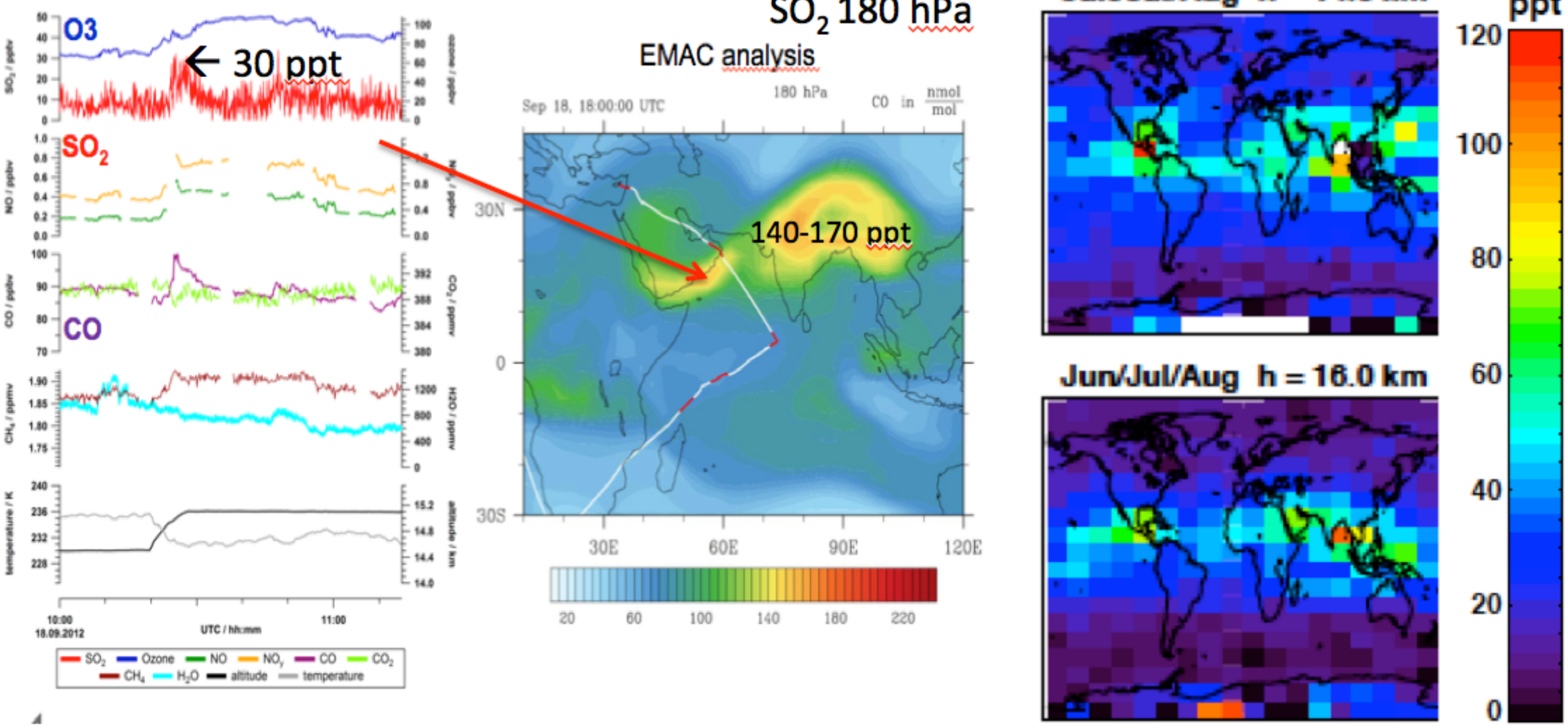
From Vernier et al. (JGR, 2015)

CESM-1 model results shown by Pengfei Yu et al., GRL 2015, estimate ATAL composed of ~60% OC and ~40% sulfate by mass

WACCM model results from Neely et al. (JGR, 2014) determined that Indian and Chinese sulfur emissions contribute only ~30% to sulfate in the ATAL

ATAL Composition: SO_2 from HALO ESMVAL and from MIPAS

SO_2 in-situ observations in Asian Monsoon outflow



HALO ESMVal flight of 18 Sept. 2012, shows up to **30 ppt SO_2 at ~15km** in UT Asian outflow.

from A. Roiger et al. presentation to SPARC SSiRC workshop, Atlanta, GA, October, 28-30, 2013.

MIPAS shows SO_2 of 50-100 ppt at 14-16 km in seasonal mean maps (2002 -2012), filtered for volcanic episodes. from M. Hoepfner et al., MIPAS SO2 in the UTLS, ACPD, 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

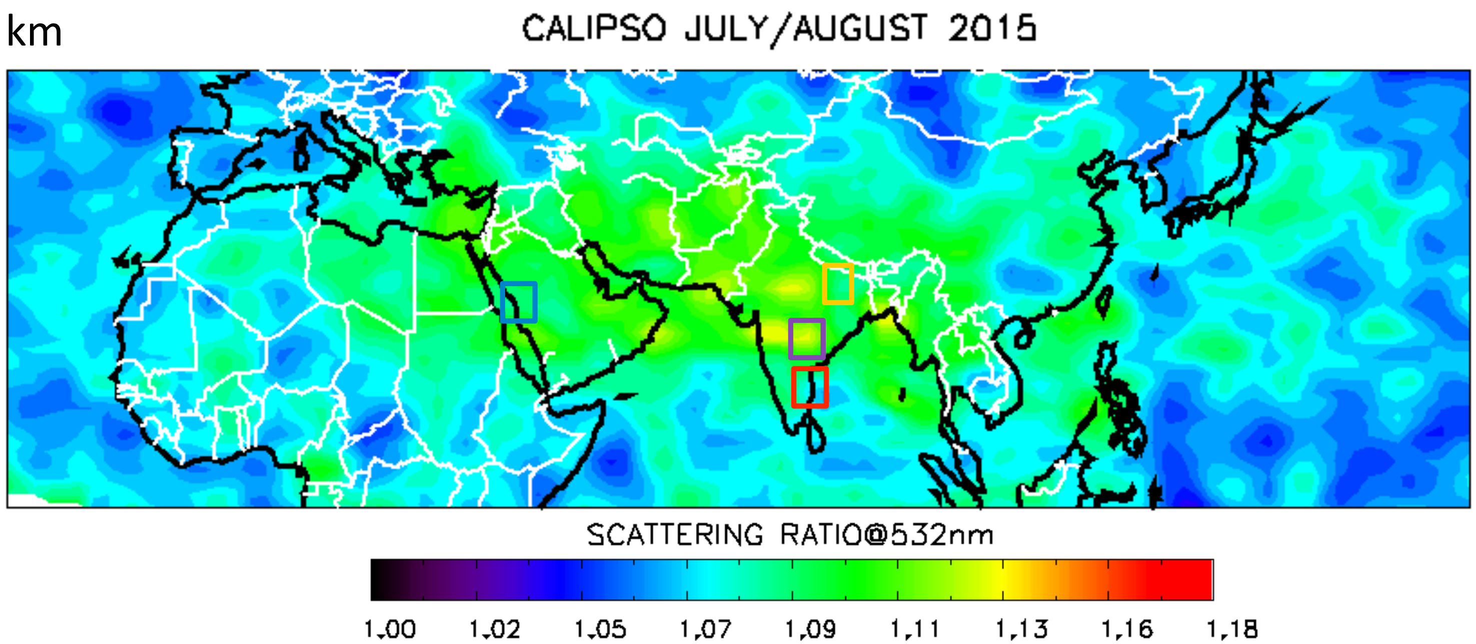
- , 15-24 Aug 15 : Banaras Hindu University, Varanasi, India
- 7 launches of aerosol and chemical sensors

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

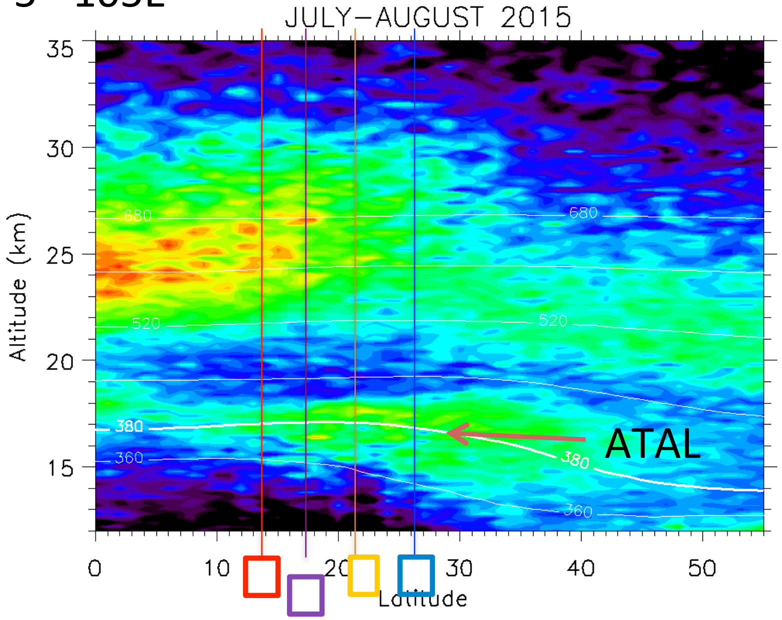
- 17-25 July 15: National Atmospheric Research Laboratory, Gadanki, India,
- 6 launches of aerosol and chemical sensors

CALIOP/BATAL-2015

15 - 18 km

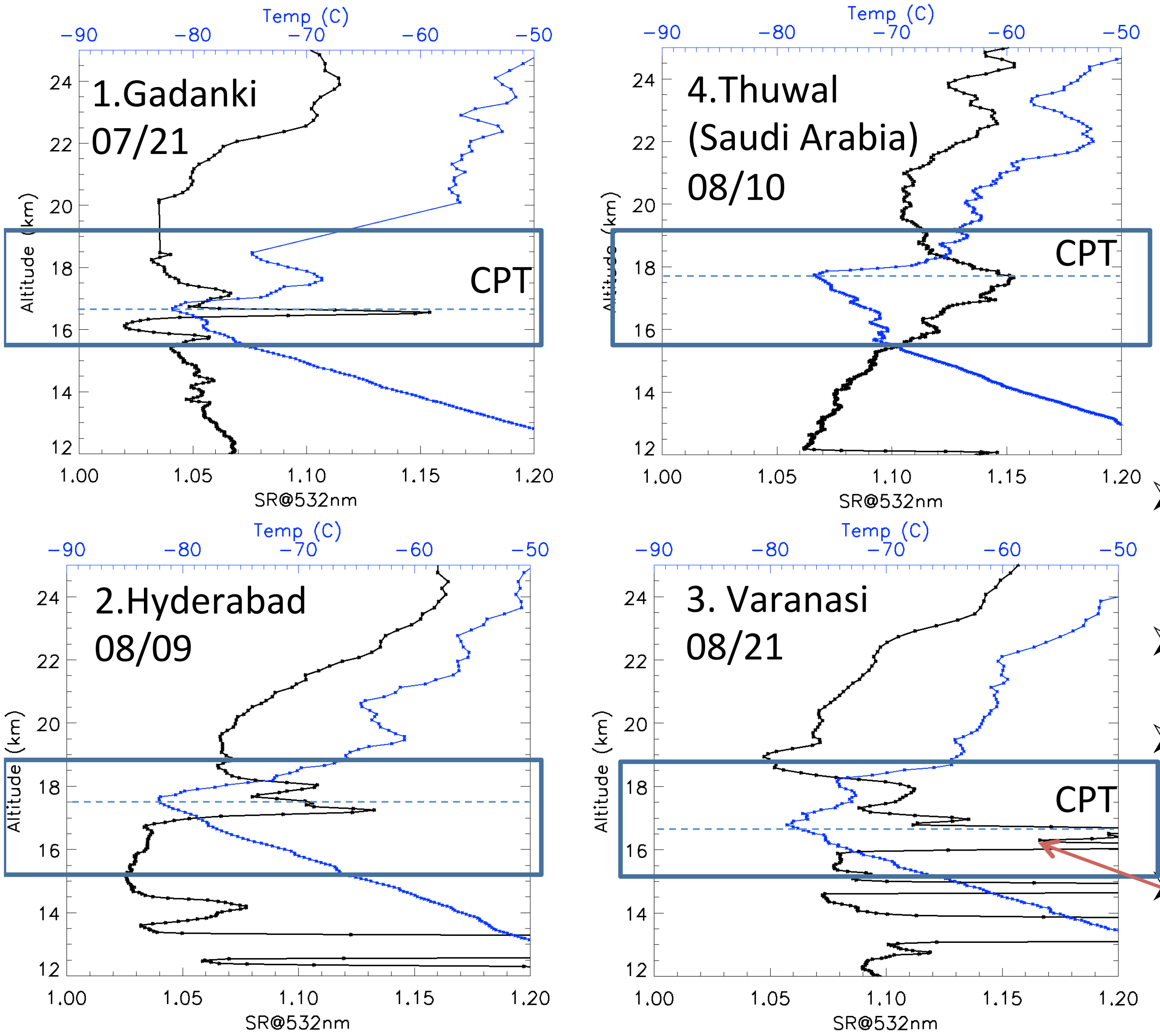


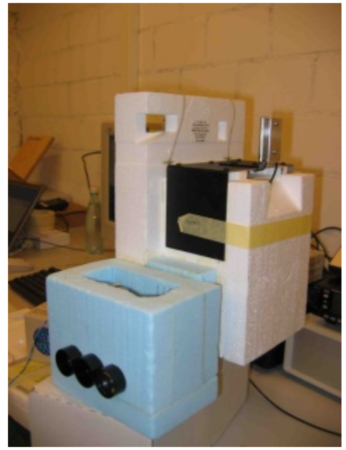
5 - 105E



- Significant enhancement of aerosol SR observed during the summer 2015
- BATAL launching locations covered a large area from the southern to the northern edge of the ATAL and the convective outflow region (Arabic Peninsula)
- Lower and middle stratosphere still influence by an old volcanic plume from Kelud eruption in Feb 2014.

BATAL 2015/COBALD flights

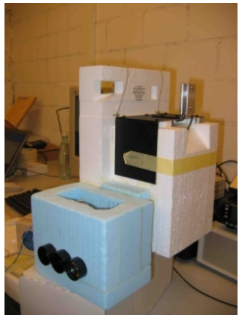




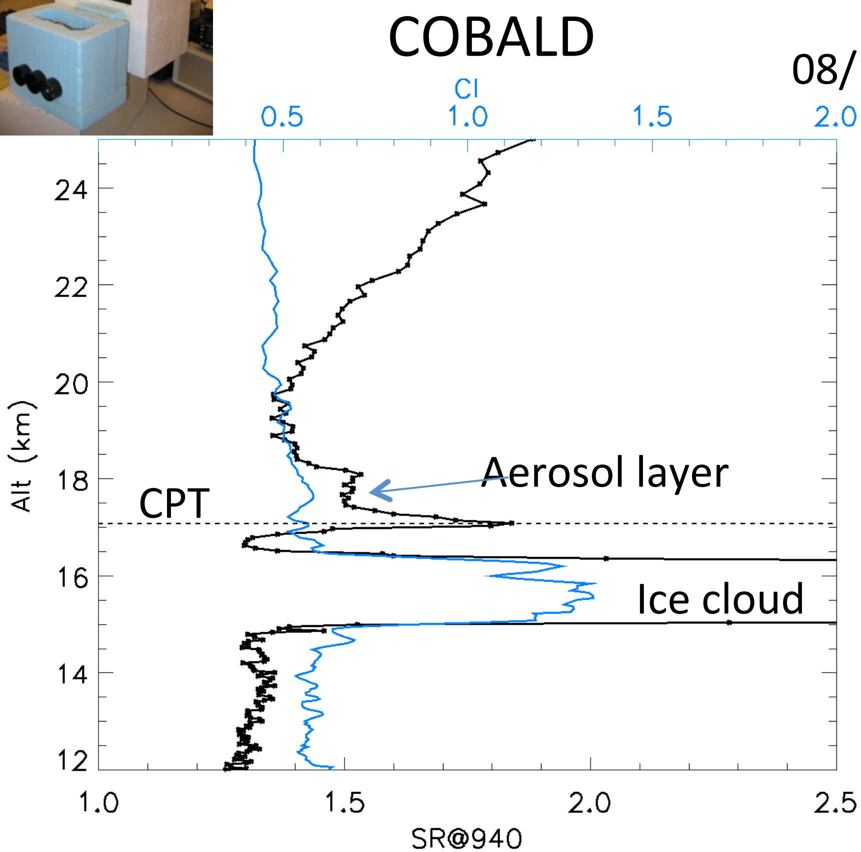
COBALD
www.iac.ethz.ch

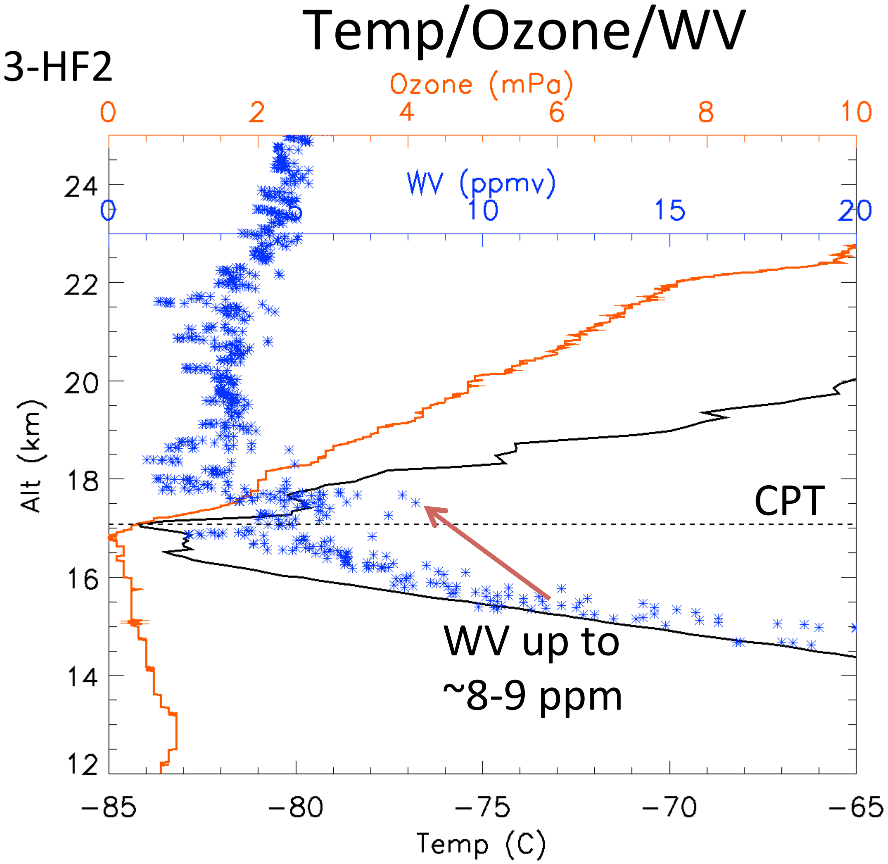
- COBALD (black) backscatter, with Temp Profiles (blue)
- Thin aerosol layers near the tropopause over India
- Contrast with broader layer observed from the Arabic Peninsula (Thuwal)
- Ice clouds near the tropopause over Varanasi

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



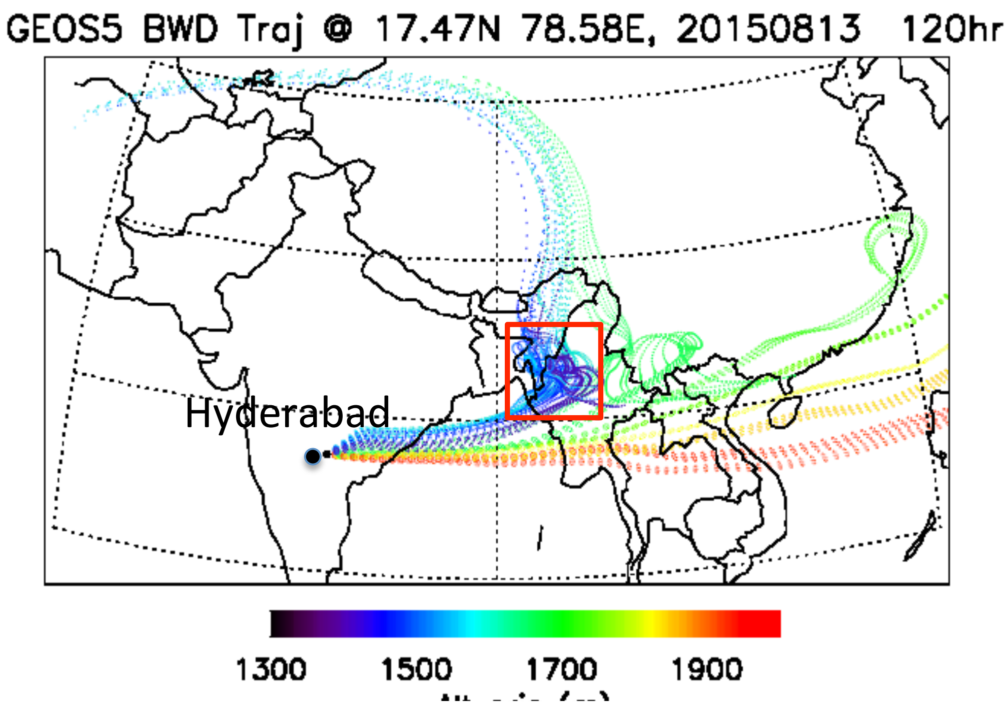
08/13-HF2

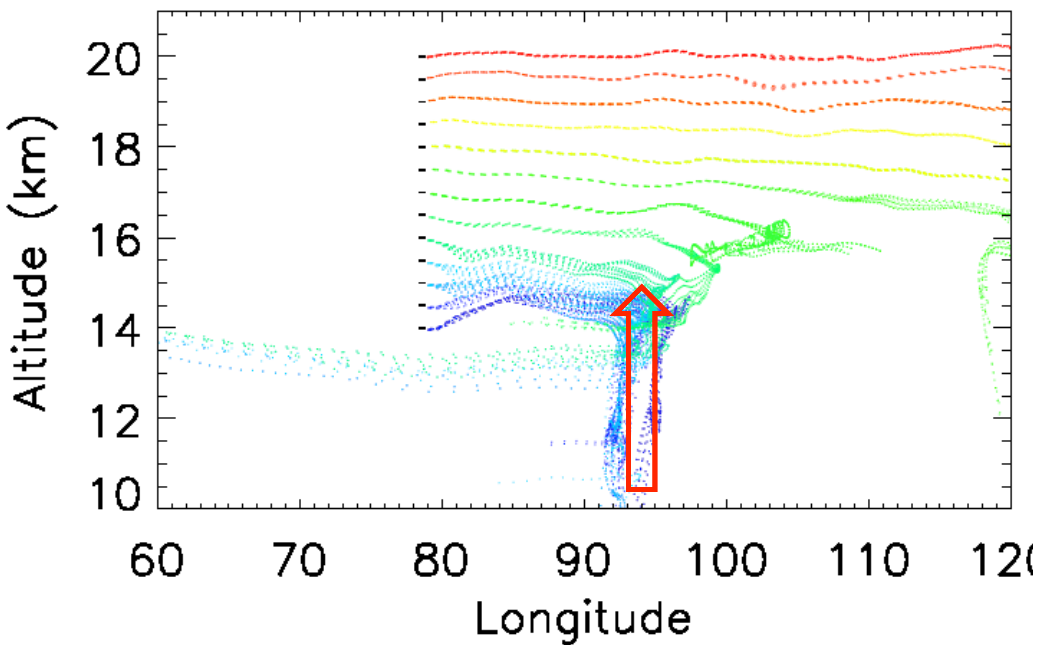




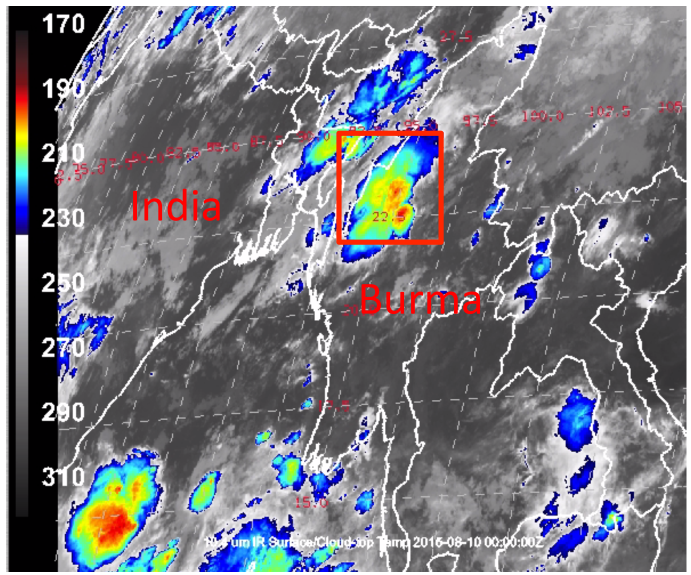
- Maximum of aerosol measured by COBALD found in vicinity of cold point tropopause
- Color Index (CI, blue line) between COBALD blue/red channels distinguishes aerosol (low values) from ice cloud (> 1, 15-16.5 km).
- Enhanced water vapor (up to 8 – 9 ppm) 17–18 km likely due to convective transport of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

Air masses origin

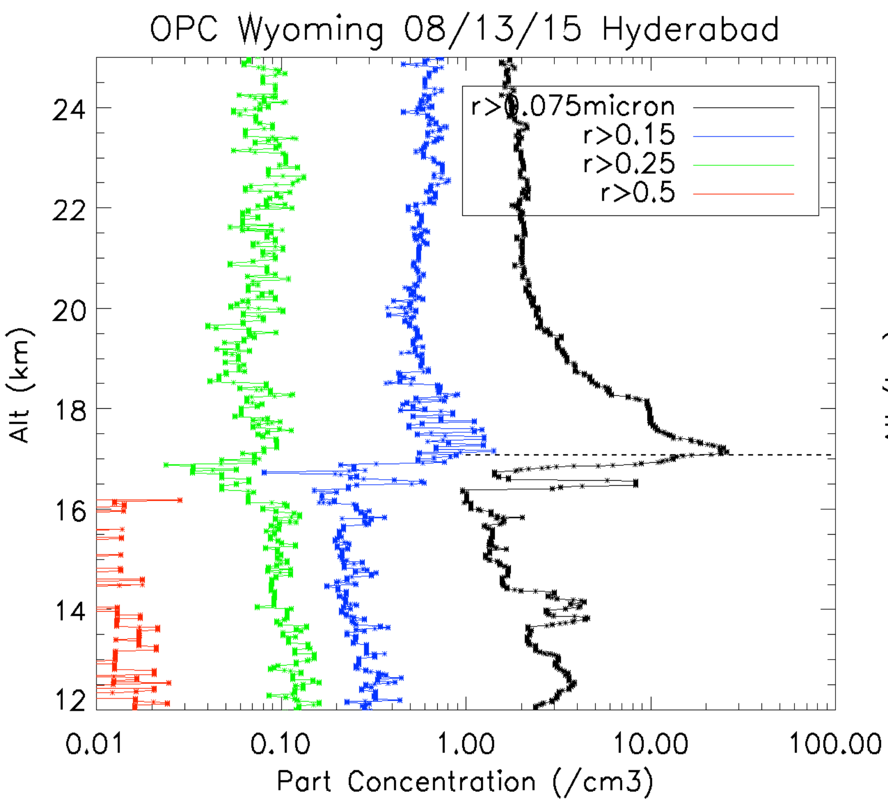


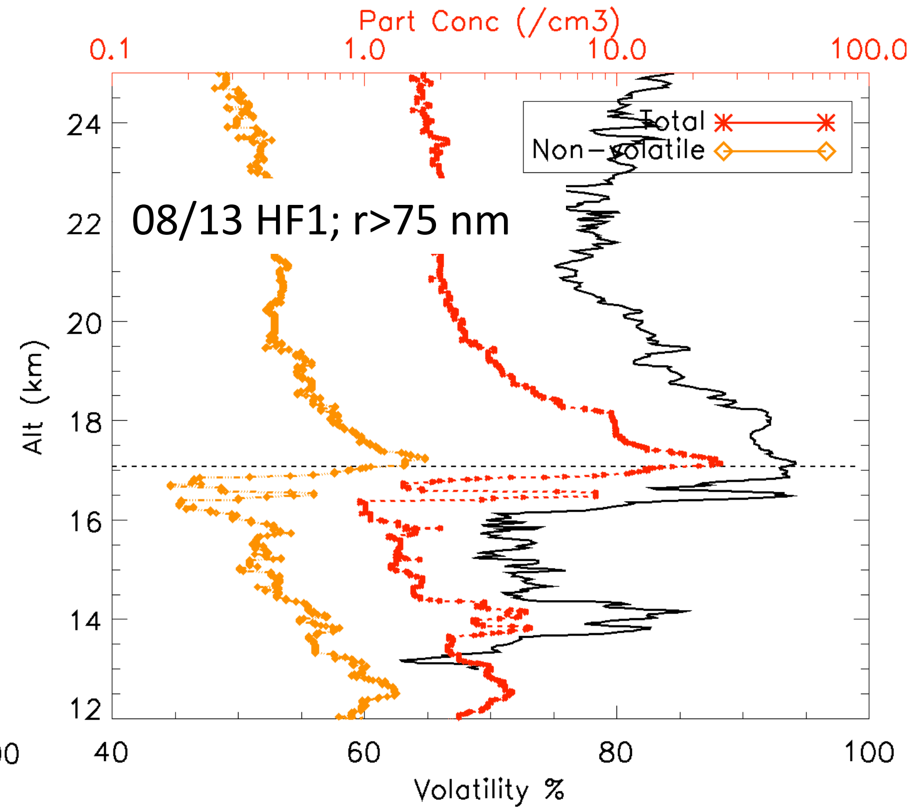


- Back-trajectories from air masses sampled by the 08/13 HF balloon flight from Hyderabad
- May have been Influenced by deep convection over Western Burma and Eastern India previous 48-72 h.



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

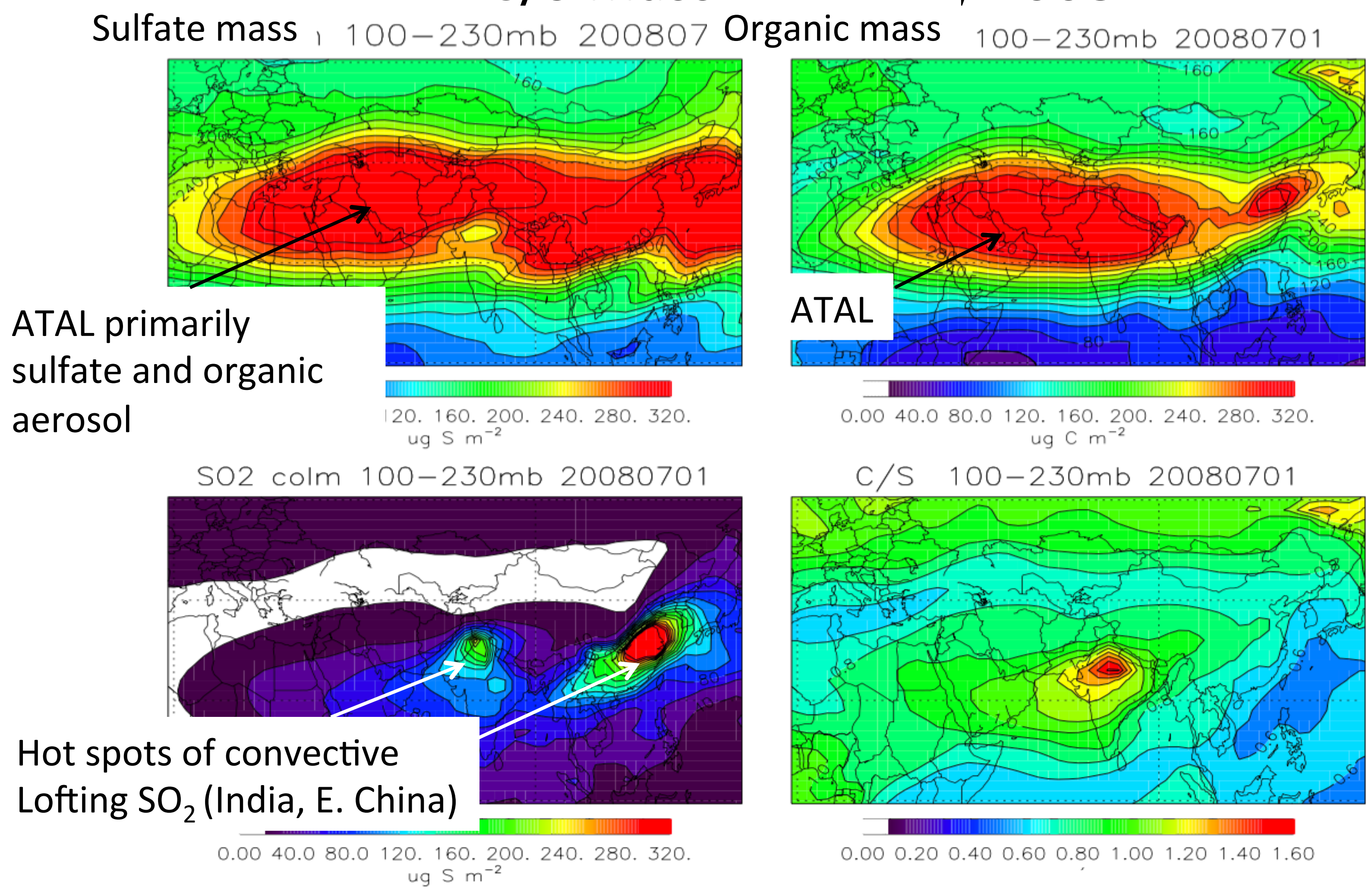
3-D CTM for gas-phase and aerosols transport and photochemistry in the troposphere, driven by GEOS-5 meteorology (www.geos-chem.org), V9.02, 2x2.5 deg. 72 levels.

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, SO4, dust , NO3, limited SOA in current run.

Update to wet scavenging of SO_2 in convective updrafts:
fraction of SO_2 subject to scavenging limited by Effective Henry's Law equilibrium and aqueous oxidation by H2O2

Series of 6 month simulations (1 Apr. 2008 – 1 Oct. 2008)

Simulated columns (100-230 hPa) of SO4, OC, SO2, with C/S mass ratio July 2008

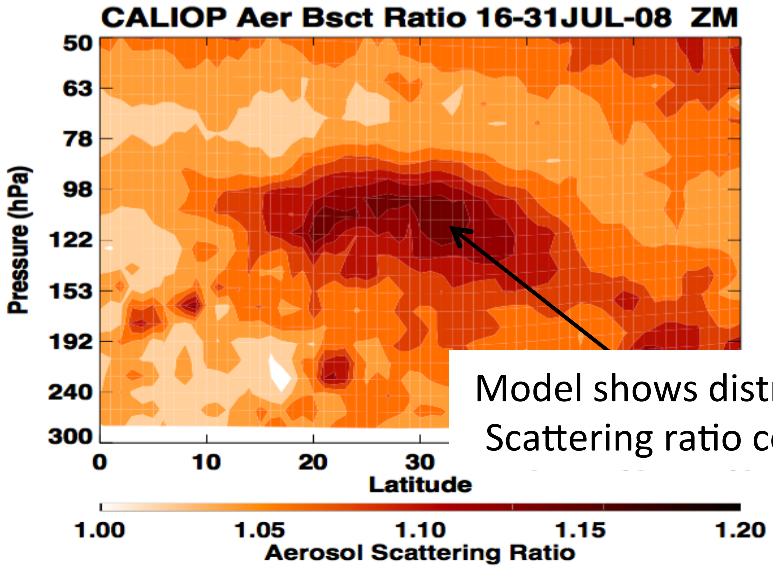


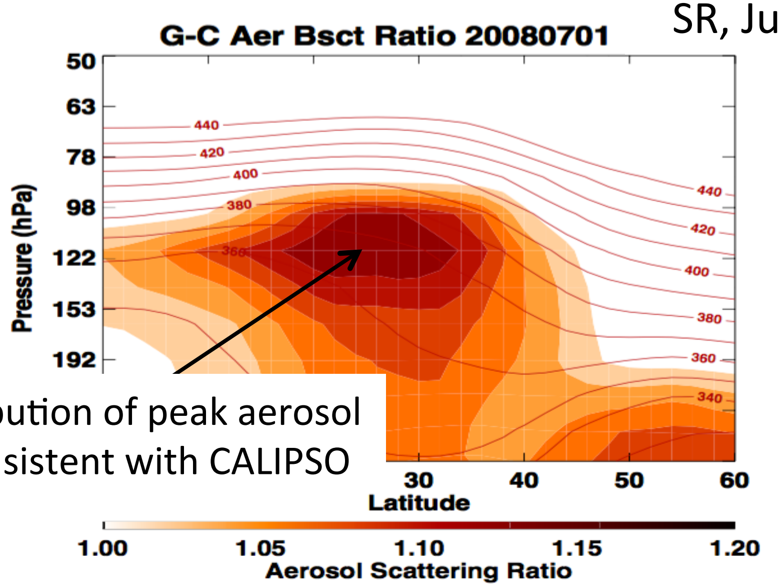
CESM-1 model results shown by Pengfei Yu et al., GRL 2015, estimate ATAL composed of ~60% OC and ~40% sulfate by mass

C/S ratios > 1 in ATAL generally consistent with data from CARIBIC observations at lower altitudes

CALIOP vs. simulated Scat.Ratio, July, 2008

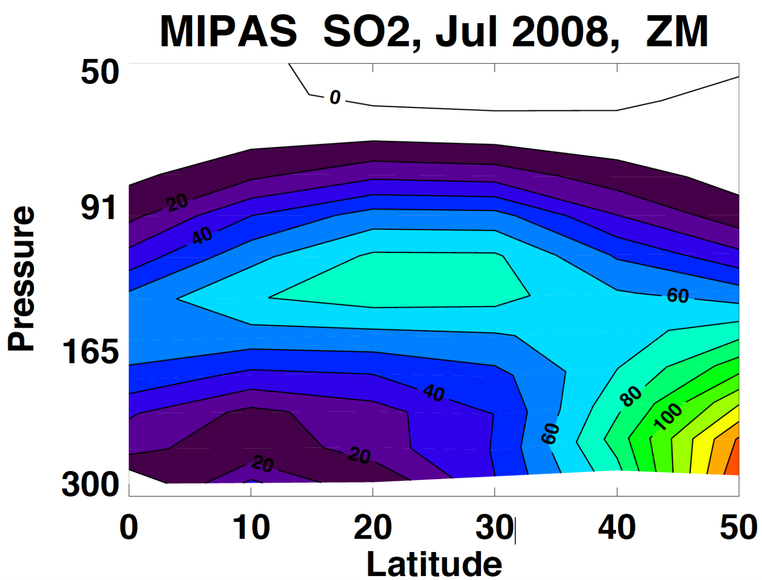
30-105°E Means
SR, July, 2008

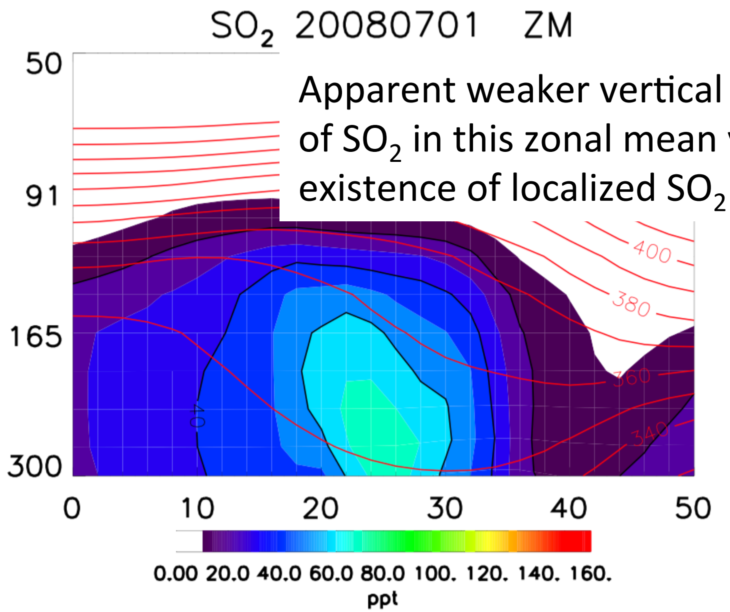




Model shows distribution of peak aerosol Scattering ratio consistent with CALIPSO

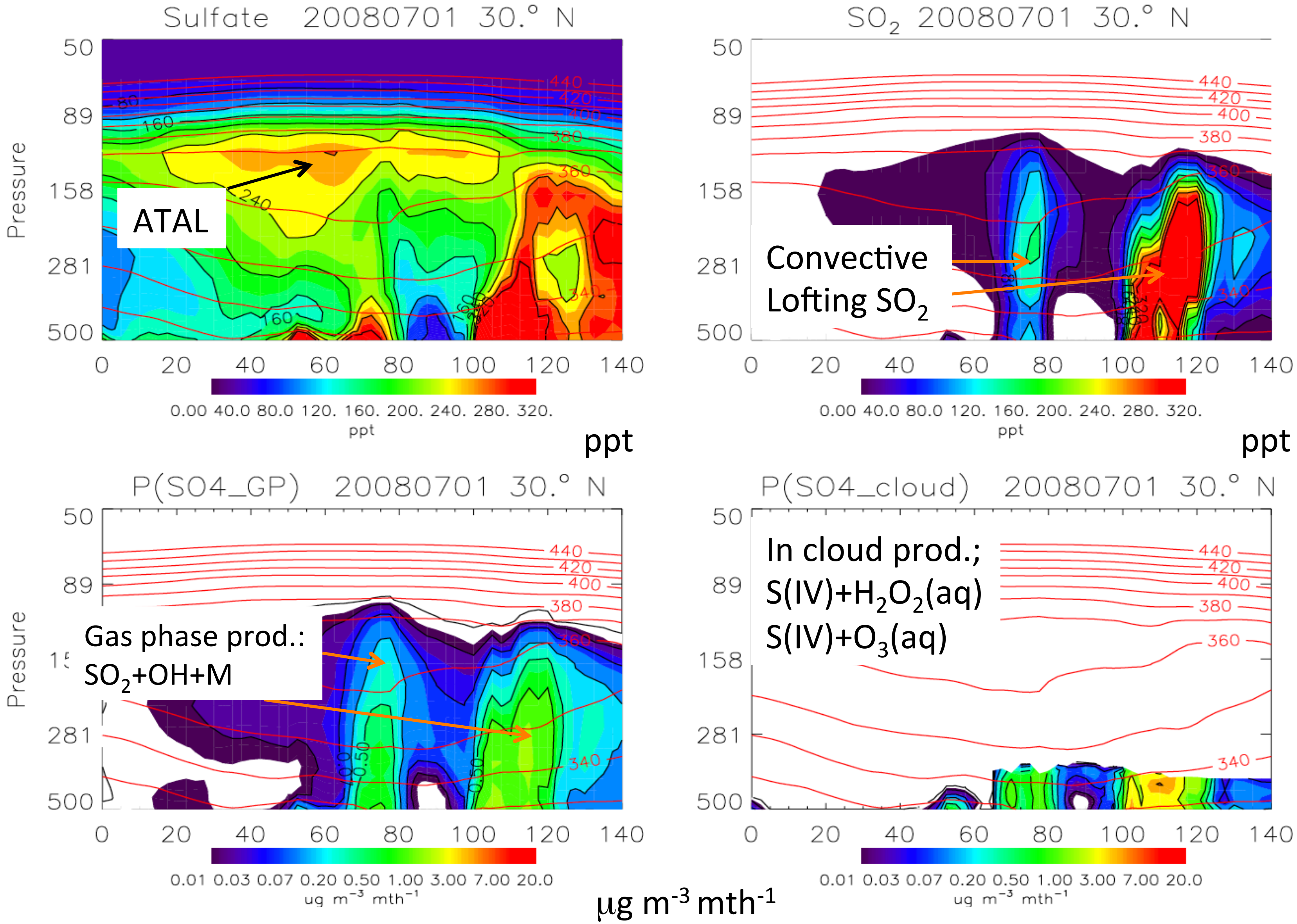
MIPAS vs G-C SO_2 July 2008



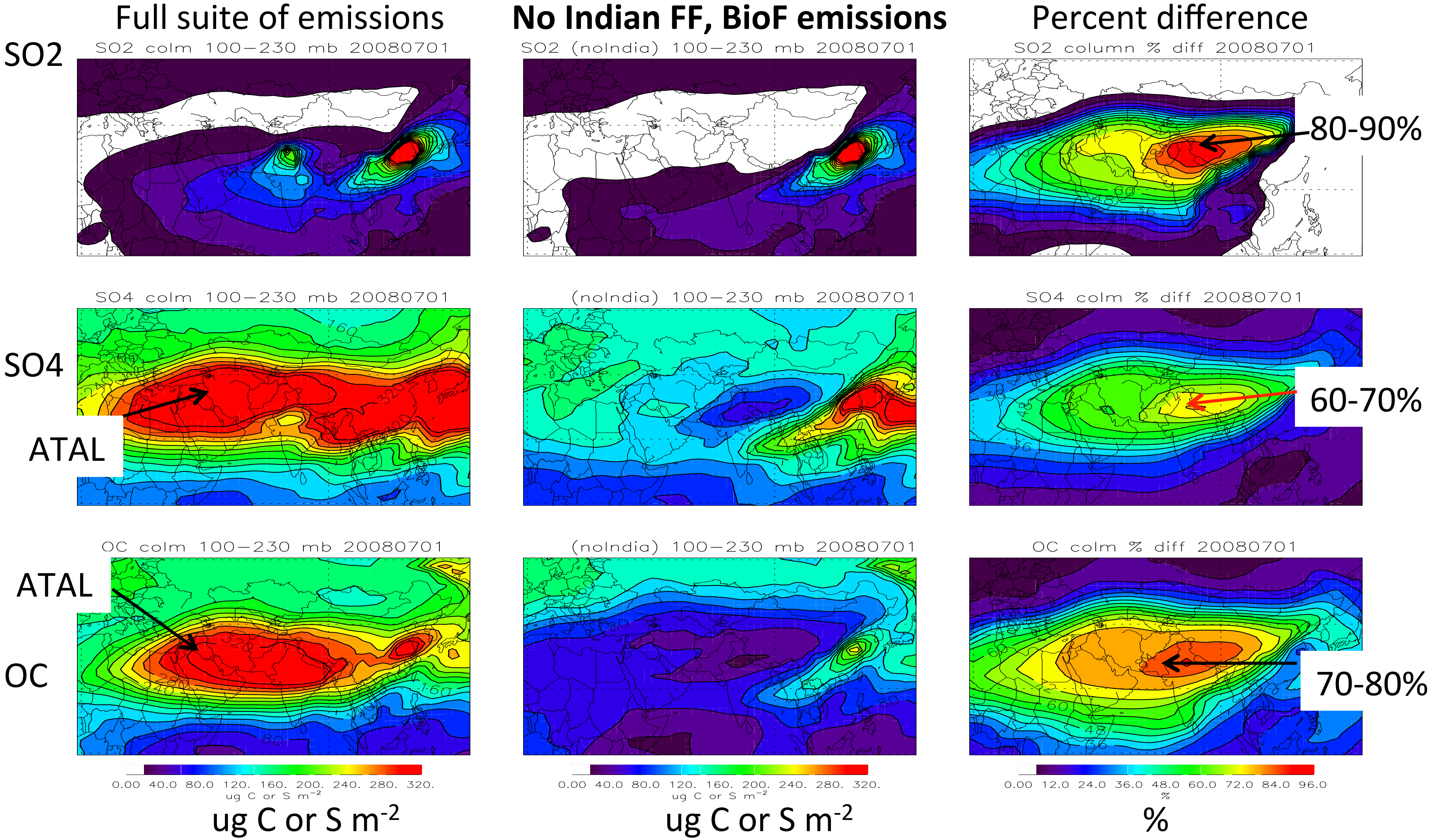


Apparent weaker vertical penetration of SO_2 in this zonal mean view masks existence of localized SO_2 towers

Model indicates ATAL sulfate sustained by convective lofting of SO_2 combined gas-phase conversion (July, 2008, 30°N)



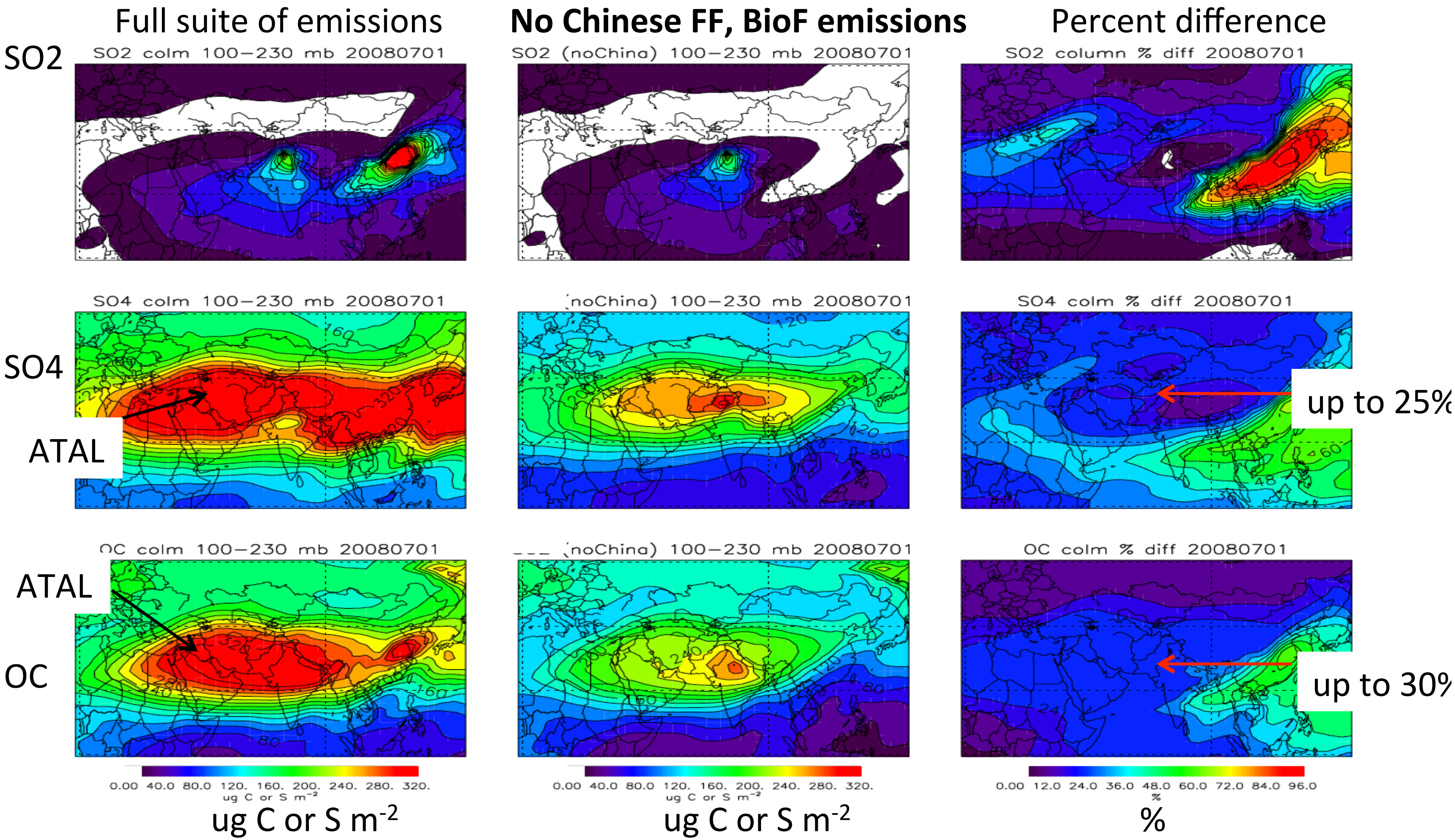
G-C **columns** (100-230 mb), **July 2008 mean**
Contribution of India emissions



Contribution from Indian emissions alone to the ATAL (July, 2008) show up to 70% (SO_4) and 80% (OC) contributions. Neely et al (JGR, 2014) determined that Indian and Chinese sulfur emissions contribute only ~30% to ATAL.

G-C **columns** (100-230 mb), **July 2008 mean**

Contribution of Chinese emissions



Contributions from Chinese emissions alone to the ATAL (July, 2008) show up to 25% (SO4) and 30% (OC) contributions. Neely et al (JGR, 2014) determined that Indian and Chinese sulfur emissions contribute only ~30% to ATAL.

Conclusions

- Indian Sub-continent key place to understand ATAL's origin:
- Key results of the BATAL campaign includes :
  - ❑ First size distribution of the ATAL : Made of very small/volatile particles of less than 0.2 micron.
  - ❑ Strongly correlated with Cold Point Temperature
  - ❑ Influenced by convective moisture.
  - ❑ Likely resulting from New Particle Formation (sulfate or SOA ?)

- Our modeling studies indicate:
  - ❑ ATAL composition a combination of sulfate and organic carbon
  - ❑ Sulfur contributions to ATAL shift from India to E. China from early –> late summer.
  - ❑ Regional emissions make dominant contribution to ATAL
  - ❑ Results sensitive to parameterized treatment of precursors in convective updrafts

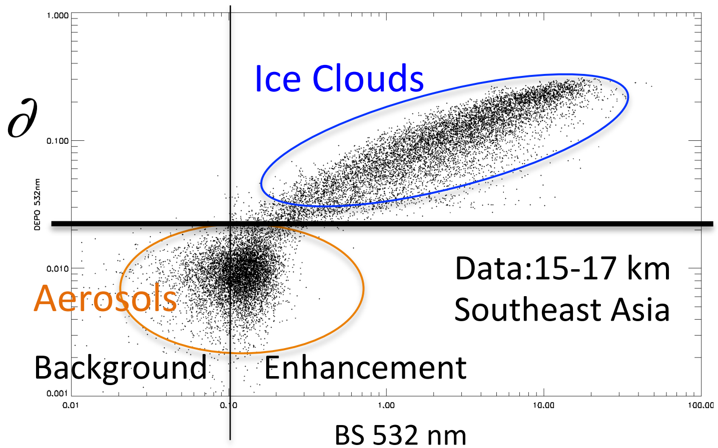
What’s next?

- BATAL 2016
  - ❑ Coincident with StratoClim campaign
  - ❑ In situ measurements of ATAL composition (impactor)
  - ❑ Characterization of and temporal changes in aerosol size
- Modeling studies:
  - ❑ StratoClim observations will provide invaluable model constraints
  - ❑ GEOS-Chem UCX – sulfur simulation of the troposphere and stratosphere

Thanks

The Asian Tropopause Aerosol Layer

Cloud clearing
(Vernier et al., 2009)

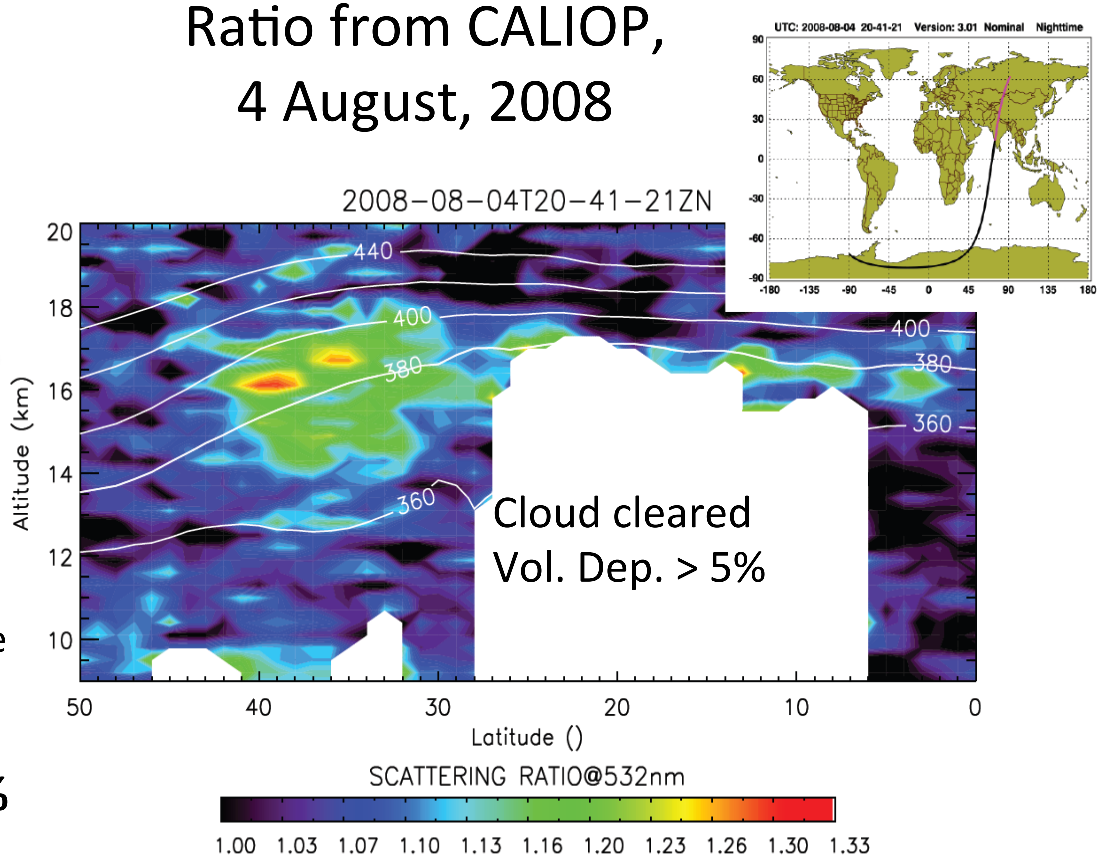


CALIOP depolarization ∂ used to separate Ice from(low depolarizing) aerosols.

Clearing criterion: Vol. Dep. > 5%

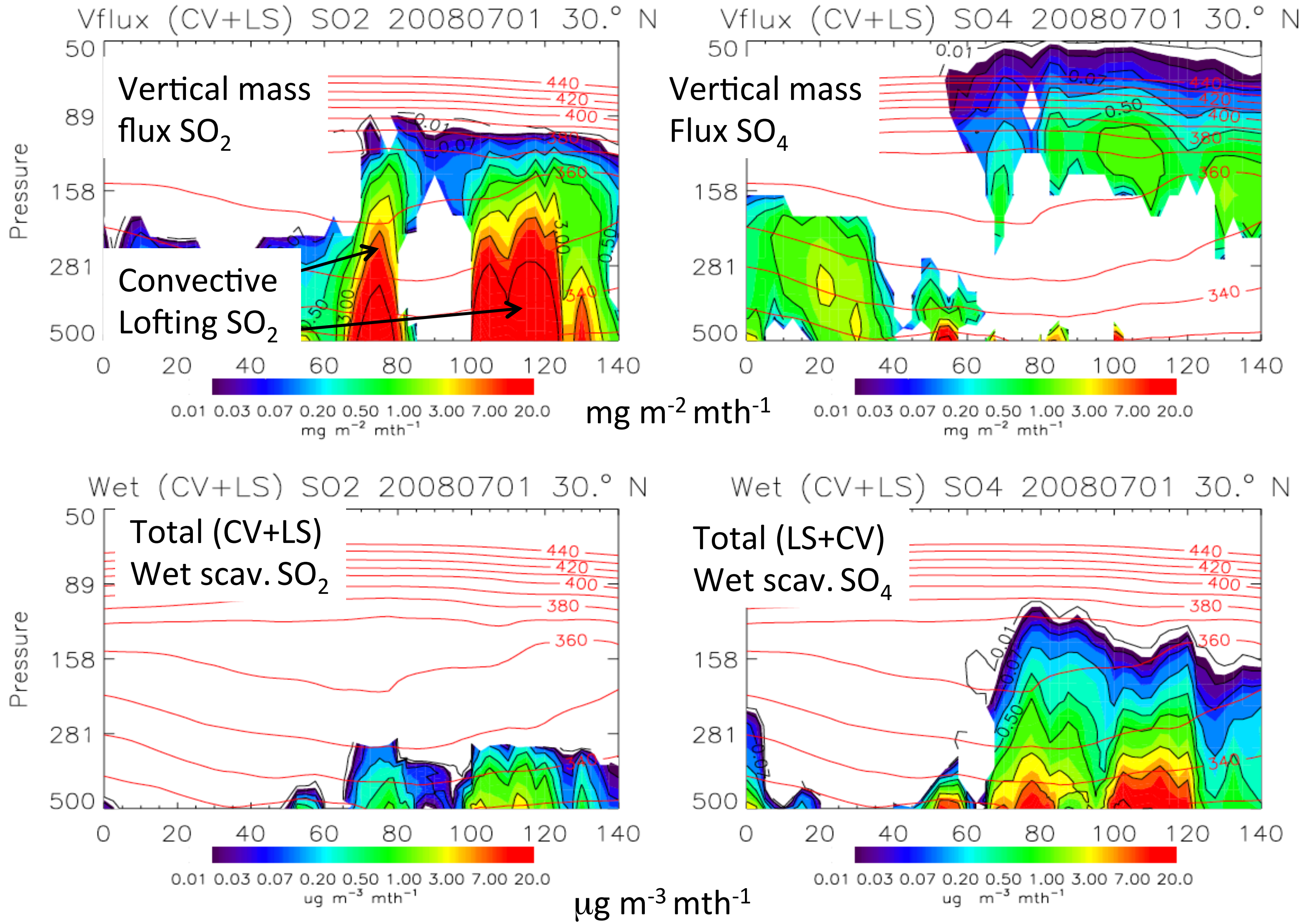
Low depolarization – spherical particles.

Cloud-cleared Scattering Ratio from CALIOP, 4 August, 2008

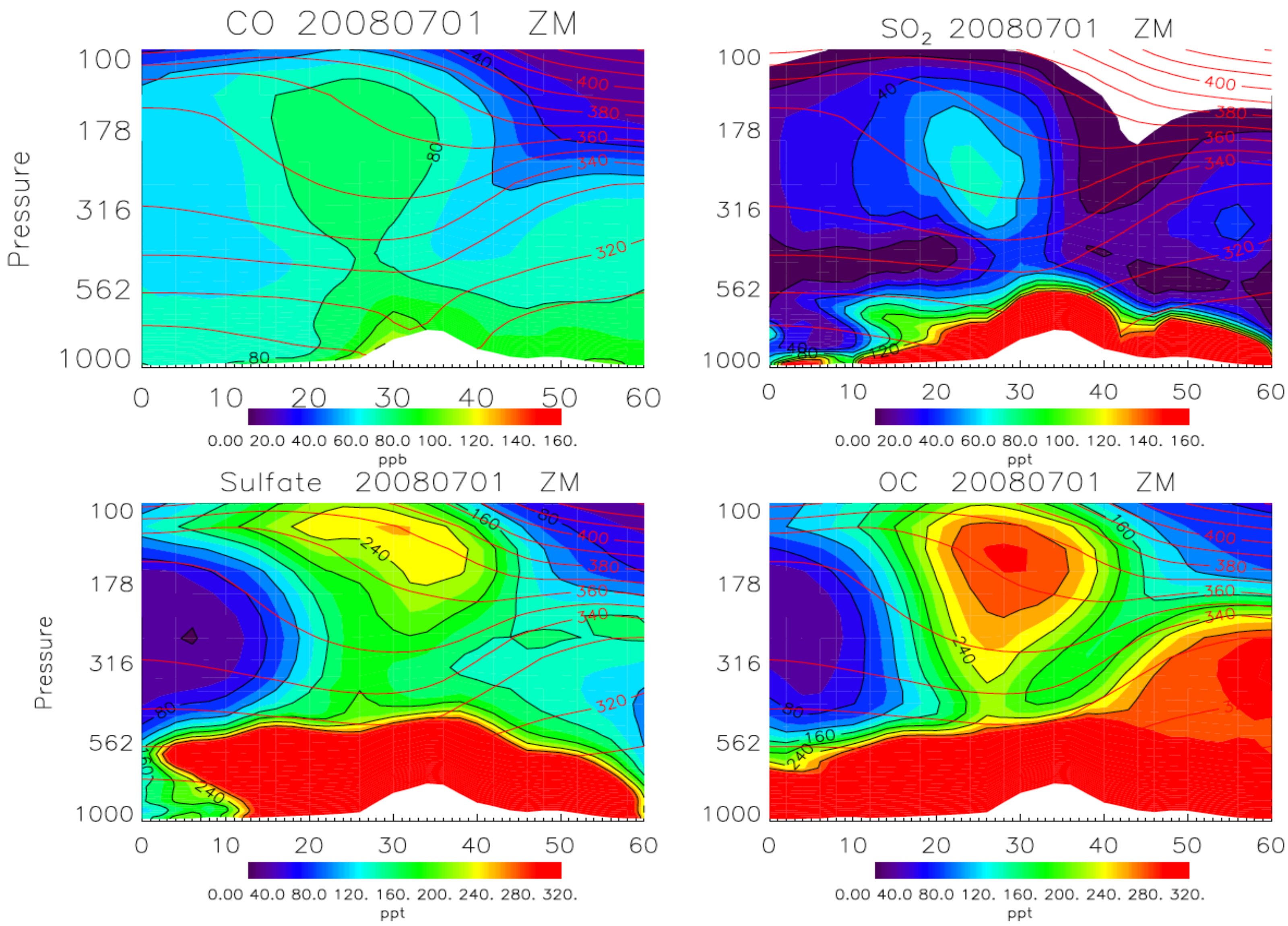


From Vernier et al., (JGR, 2015)

Convective lofting of SO_2 extends into the upper troposphere while lofting of sulfate is undercut by scavenging by precip.



Simulated Latitude X-sections 30-105°E mean, CO, SO2, Sulfate and Organic aerosol, July, 2008



Standard Version of code with instantaneous titration of SO2 in wet convective updrafts shows no ATAL