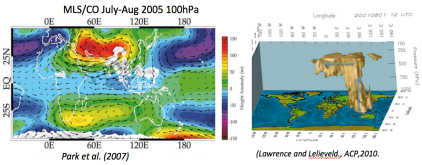
Characterizing the Asian Tropopause Aerosol Layer (ATAL) using satellite observations, balloon measurements and a chemical trasnport model

T. D. Fairlie[1], J.-P. Vernier[2], H. Liu[3], T. Deshler[4], M. Natarajan[1], K. Bedka[1], T. Wegner[1], N. Baker[1], H. Gadhavi[5], M.V. Ratnam[5], A. Jayaraman[5], A. Pandit[5], A. Raj[5], H. Kumar[5],S. Kumar[6], A. Singh[7], G. Stenchikov[8], F. Wienhold[9] and J. Bian[10]

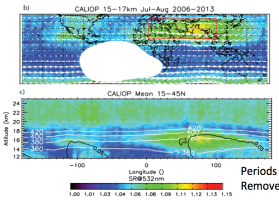
1. NASA Langley Research Center, USA; 2. Science Systems and Applications, USA; 3. National Institute of Aerospace, USA; 4. University of Wyoming, USA; 5. National Atmospheric Research Laboratory, Gadanki, India; 6. National balloon facility, TIFR, Hyderabad, India; 7. Banaras Hindu University, India; 8. King Abdullah University of Science and Tech., Saudi Arabia; 9. Swiss Federal Institute of Tech., Zurich, Switzerland; 10. LAGEO, Inst. of Atmos. Phys., Chinese Acad. Sci., Beijing, China

Transport of pollution in the Upper Troposphere by Asian Monsoon



- Pollutant tracers (e.g. CO, HCN, CH_4) accumulate in the UTLS Asian summer anticyclone; linked to deep convection in the Asian Monsoon (Park et al., 2007; Randel et al., 2006, 2010).

Asian Tropopause Aerosol Layer (ATAL) detected in CALIPSO and SAGE II observations (Vernier et al., 2011; Thomason and Vernier, 2013), predicted in model studies (Q. Li et al., 2005).



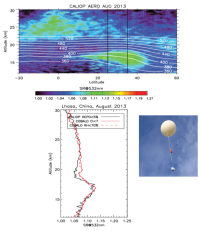
ATAL shown by enhanced Scattering Ratio (SR) during Asian Monsoon;

ATAL extends from top of convective outflow.

Periods affected by volcanic aerosol Removed from map and section

Questions: What is the origin of the ATAL? What is it made of?
Are there regional climate impacts?
There are very few *in situ* observations.

CALIOP obs. validated by COBALD backscattersondes, August, 2013



Mean (cloud-cleared) CALIOP SR, 60-120°E, shows ATAL between θ of 60 and 420K, extending to NH mid latitudes and tropics.

Median SR profiles from 18 COBALD backscattersondes launched from Lhasa, Tibet* vs. cloud-cleared CALIOP data, +/-5°Lat., +/-30°Long. of Lhasa.

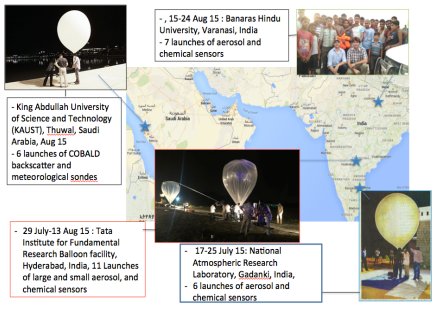
*(located, 30°N,91°E)

COBALD data from J. Bian (CAS, Beijing, China), F. Wienhold (ETH-Zurich)

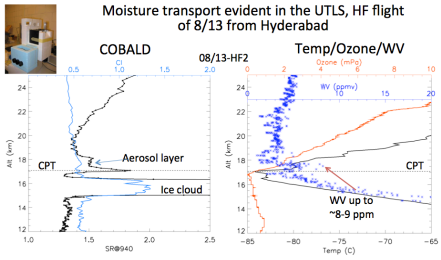
From Vernier et al., (JGR, 2015)

BATAL 2015 : Balloon-borne measurements of the ATAL

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

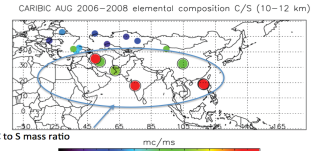


Moisture transport evident in the UTLS, HF flight of 8/13 from Hyderabad



- 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 (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

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



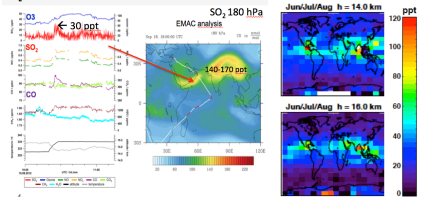
CARIBIC impactor data Martinsson et al., 2014

	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)

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, ACP, 15, 7017-7037, 2015.

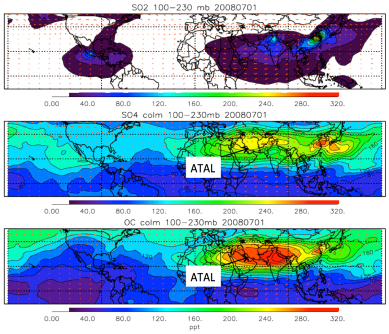
GEOS-Chem simulations

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 ➔ 47 levels.
Aerosols Components: OC, BC, SO_4-NO_3-NH_4, dust ,sea salt, limited SOA in current runs.

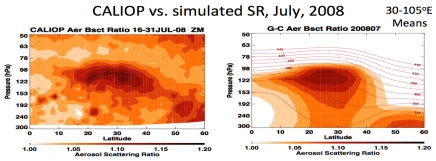
Update to wet scavenging of SO_2 in convective updrafts: combines an Effective Henry's Law equilibrium with aqueous oxidation by H_2O_2

Series of 6 month simulations (1 Apr. 2008 – 1 Oct. 2008)
1. All emissions turned on;
2. No Fossil-Fuel (FF) nor biofuel (BF) emissions from China
3. No Fossil-Fuel (FF) nor biofuel (BF) emissions from India

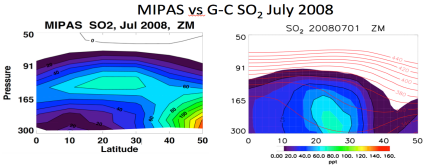
Upper tropospheric components, all emissions



CALIOP vs. simulated SR, July, 2008 30-105°E Means

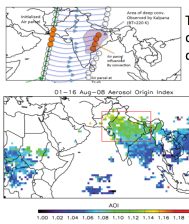


MIPAS vs G-C SO_2 July 2008



Model comparison with MIPAS SO_2 and CALIOP scattering ratio (SR) improved with updated treatment for SO_2 scavenging in convective updrafts

ATAL origin

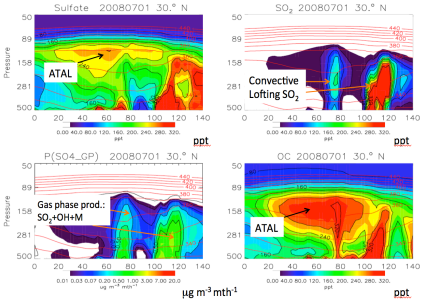


Trajectory mapping of CALIPSO observations to regions of deep convection (BT<220K from Kalpana);

SO_2 emissions, July, mg m^{-2} EDGAR with Streets (SE Asia)

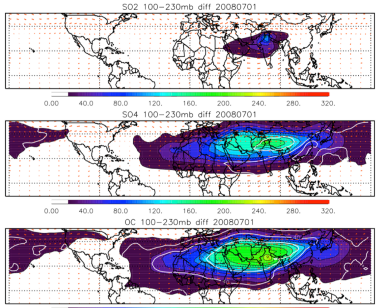
Map of mean trajectory-mapped CALIPSO SR (AOI) sourced to deep convection, 1-16 Aug., 2008, indicates Northern India as key deep convective source for elevated aerosol in the ATAL.

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

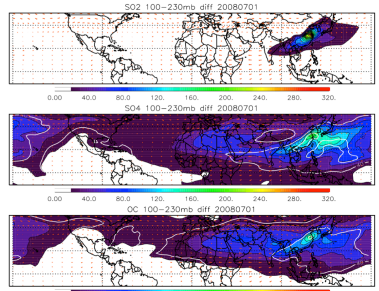


Source apportionment studies: Impact of Indian and Chinese emissions

Upper tropospheric components, contribution of Indian emissions



Upper tropospheric components, contribution of Chinese emissions



Contributions from Indian and Chines sources, with % contributions (white contours). Model indicates a dominant (>60%) contribution from Indian emissions to ATAL in July, 2008; Chinese emissions (20-30%) remain largely outside the anticyclone in this episode; rest-of-world emissions (not shown) found to contribute <20% to ATAL. These contributions change with transience of the ATAL anticyclone.

Summary

- CALIOP observations have revealed a seasonal maximum of aerosol in the UTLS associated with the Asian monsoon. The Asian Tropopause Aerosol Layer (ATAL) has been independently validated using backscatter sondes flown out of China and from India.
- Limited in situ measurements of composition (CARIBIC) indicate that the ATAL is composed primarily of carbonaceous and sulfate aerosol. Elevated SO_2 (~30 ppt) found in monsoon outflow in the UTLS (in HALO ESMVal campaign).

Summary continued

- Balloon observations (BATAL, 2015) reveal ATAL aerosols near the cold point tropopause, often in vicinity of ice cloud and elevated water vapor
- GEOS-Chem simulation shows improved comparison with MIPAS SO_2 and CALIOP backscatter with updated treatment of wet scavenging of SO_2 in deep convective updrafts.
- Model indicates dominant contribution of regional sources of SO_2 and OC (70-80%) in ATAL composition, compared with rest-of-world contributions.

Reference: Vernier, J.-P., T. D. Fairlie, M. Natarajan, F. G. Wienhold, J. Bian, B. G. Martinsson, S. Crumeyrolle, L. W. Thomason, and K. M. Bedka (2015), *Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution*, J. Geophys. Res. Atmos., 120, 1608–1619, doi: 10.1002/2014JD022372., and references therein.

T.D.Fairlie@nasa.gov; Jeanpaul.Vernier@nasa.gov; Hongyu.Liu-1@nasa.gov