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

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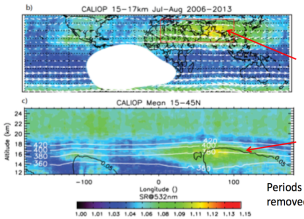
CALIPSO lidar observations have revealed a seasonal maximum in aerosol in the UTLS associated with the Asian monsoon. The ATAL is independently validated using balloon-borne backscatter sondes flown out of China (J. Bian) and India (BATAL campaigns). BATAL flights show peak aerosol backscatter near the cold point tropopause, coincident with elevated water vapor, likely from upstream convection. OPC data from BATAL (Deshler, UW) indicate ATAL is composed predominantly of small (r < 150 nm), mostly volatile (90%) particles. Limited in situ aircraft measurements suggest that the ATAL is composed primarily of carbonaceous and sulfate aerosol. CTM simulations indicate predominance of organic and sulfate aerosol, with N. India, E. China as dominant source regions. However, we also find significant interannual and intraseasonal variability in the relative contributions of principal source region. Here, we show that in August 2013, both N. India & E. China contribute to sulfate in the ATAL; simulations for 2008 found N. India dominant.

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;

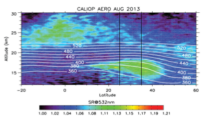
ATAL extends from top of convective outflow.

Periods affected by volcanic aerosol removed from map and section

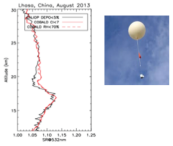
Questions: What is the origin of the ATAL? What is it made of? Are there regional climate impacts?

CALIOP obs. validated by COBALD backscatter sondes, August, 2013

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



Median SR profiles from 18 COBALD backscatter sondes launched from Lhasa, Tibet (30°N, 91°E) vs. cloud-cleared CALIOP data, +/-5°Lat., +/-30°Long. of Lhasa.



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

From Vernier et al., (JGR, 2015)

ATAL Composition

Limited in situ measurements from the CARIBIC aircraft program (Martinsson et al., 2014) indicate that aerosol composition 2-4 km below the ATAL layer is composed primarily of sulfate and carbonaceous aerosol. SO_2 as high as 50-250 ppt has been observed in ASM anticyclone, during the Oxidation Measurement Observation (OMO) airborne campaign, coincident with particles in the size range 10 nm – 2 um (H. Schlager, personal communication)

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.

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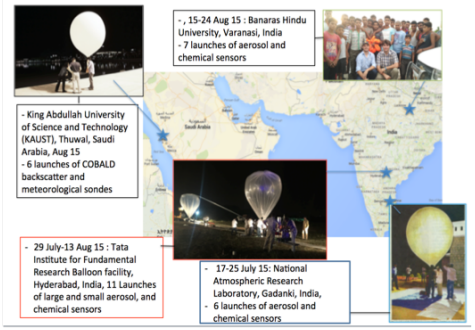
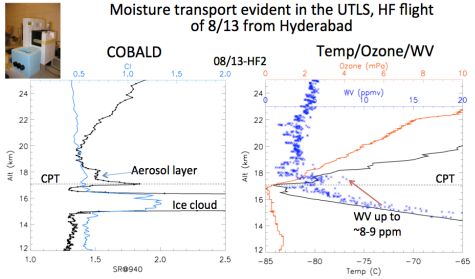
BATAL 2015 : Balloon-borne measurements of the ATAL

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



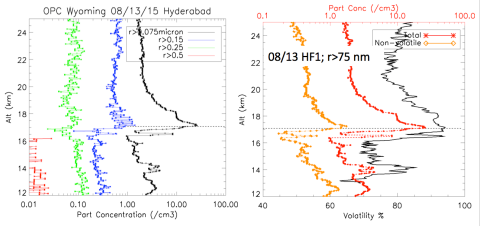
The BATAL campaign of 2015 made measurements of aerosol backscatter (COBALD), aerosol size and volatility (UW OPCs) for the ATAL using a hierarchy of balloon-borne payloads flown from sites in India (Gadanki, Hyderabad, Varanasi) and Saudi Arabia (KAUST)

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

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% particles found in the size range 0.075 < r < 0.15 µm
- Heated (180°C) and unheated inlets on OPCs indicate ~90% small, volatile particles

GEOS-Chem CTM simulations

3-D CTM for gas-phase and aerosols transport and photo-chemistry in the troposphere, driven by MERRA meteorology (www.geos-chem.org), V9.02, 2x2.5 deg. 72 levels. GEOS-Chem Version 9.02 was used.

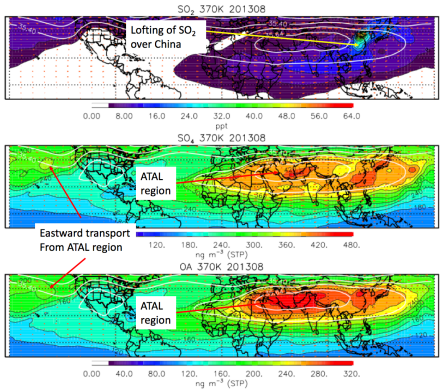
> Aerosols Components: OC, BC, SO_4-NO_3-NH_4, dust , sea-salt, limited SOA.

Update to wet scavenging of SO_2 in convective updrafts:

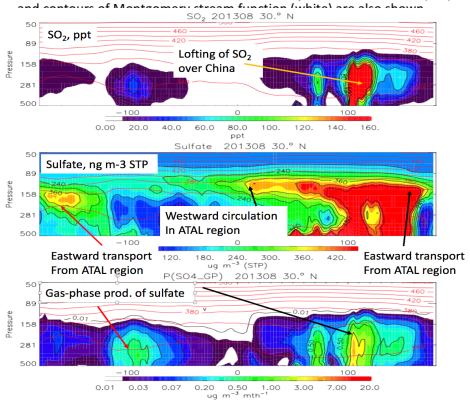
Standard code assumes instantaneous titration of SO_2 or H_2O_2. Modified code combines an Effective Henry's Law equilibrium with aqueous oxidation of SO_2 by H_2O_2 and O_3. 15% retention of SO_2 in cold cloud imposed to best match SEAC4RS aircraft observations (see right).

Series of 6 month simulations (1 Apr.– 1 Oct., 2008 and 2013)

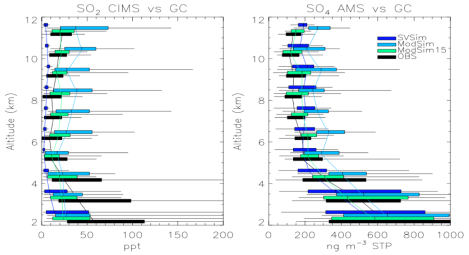
1. All emissions turned on;
2. No Fossil-Fuel (FF) nor biofuel (BF) emissions from China
3. No Fossil-Fuel (FF) nor biofuel (BF) emissions from India



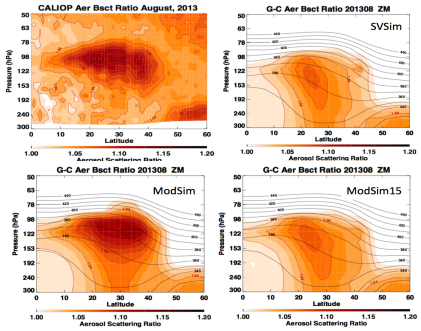
Maps of simulated monthly mean SO_2, sulfate and organic aerosol at 370K for August 2013, using the modified version of the model. Maps show convection of SO_2 over China; circulation in the ATAL; eastward and westward detrainment from ATAL region. Units are ppt for SO_2, ng m^{-3} STP for sulfate and OA. OA/OC=2.1 assumd. Monthly-mean wind vectors (red) and contours of Montgomery stream function (white) are also shown.



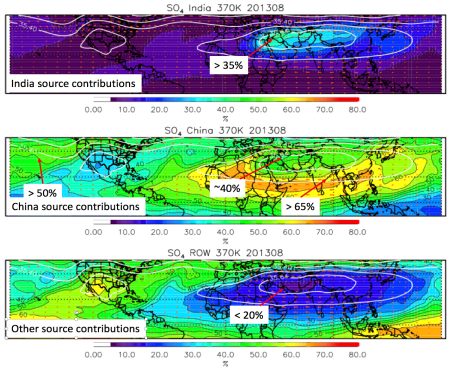
Simulated monthly mean SO, and sulfate aerosol cross-sections for August 2013, using the modified version of the model. Illustrates convection of SO_2 over E. China, westward circulation around the ATAL anticyclone, eastward detrainment from ATAL region. Sulfate maximum in the upper troposphere associated with gas-phase oxidation of convectively lofted SO_2.



Observed vs. simulated SO_2 and sulfate from DC8 flights in SEAC4RS airborne campaign (Aug.-Sep., 2013). Observations (**black**) are of CIMS SO_2 (Huey group) and AMS submicron sulfate (Jimenez group); Standard model results (SVSim, **dark blue**) underestimate SO_2; modified treatment of SO_2 scavenging in convective updrafts (ModSim, **pale blue**) overestimates the observations; imposing 15% retention of SO_2 in ice cloud (ModSim15, **green**) improves simulation; bars span 25th – 75th percentile; whiskers 5th –



Cross-section of CALIOP aerosol scattering ratio (SR) averaged over longitudes (30 – 120°E) for August 2013; (SVSim) Standard model results underestimate observed SR; simulations with modified treatment of SO_2 without (ModSim) and with (ModSim15) 15% retention of SO_2 in ice cloud overestimate and underestimate observed CALIPSO SR, respectively. Lidar ratios of 70 sr assumed for both sulfate and OC.



Source apportionment: Percent contributions to UTLS sulfate from Indian subcontinent, China, and Rest-of-world August 2013, from the ModSim15 simulation. India contributes to the core of the ATAL anticyclone at this time. China sources contribute to the flank of the anticyclone, or are detrained eastward in the subtropical jet, or westward across the Atlantic. ROW contributions to ATAL are small (~25%) in this case.