Characterizing the Asian Tropopause Aerosol Layer using in-situ balloon measurements: the BATAL campaigns of 2014-2017

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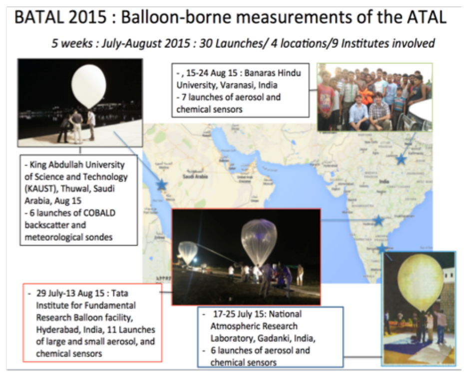
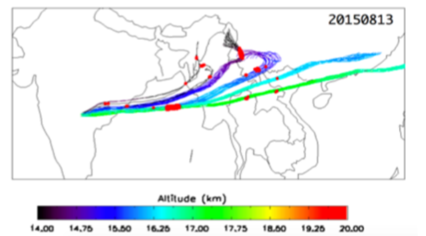
We present in situ balloon observations of the Asian Tropopause Aerosol Layer (ATAL), a summertime accumulation of aerosols in the upper troposphere and lower stratosphere (UTLS), associated with Asian Summer Monsoon (ASM). The ATAL was first revealed by CALIPSO satellite data, and has been linked with deep convection of boundary layer pollution into the UTLS. The "Balloon measurements of the Asian Tropopause Aerosol Layer (BATAL)" field campaigns to India and Saudi Arabia were designed to characterize the physical and optical properties of the ATAL, to explore its composition, and its relationship with clouds in the UTLS. We launched 55 balloon flights from 4 locations, in summers 2014-2016, and returned to make more balloon flights in summer 2017. The BATAL flights show peak aerosol backscatter near the cold point tropopause, coincident with elevated water vapor, likely from upstream convection. OPC data indicate ATAL is composed predominantly small (< 150 nm), mostly volatile (90%) particles. 2017 composition data reveal nitrate in the UTLS but no sulfate was found. CTM simulations show peaks in organic, sulfate, nitrate, and ammonium aerosol, and some gas-phase ammonia. Model results indicate N. India, E. China as dominant source regions, but with significant interannual and intraseasonal variability in relative contributions. In August 2013, simulated N. India & E. China are found to make comparable contributions (30%) to sulfate and organic aerosol in the ATAL.

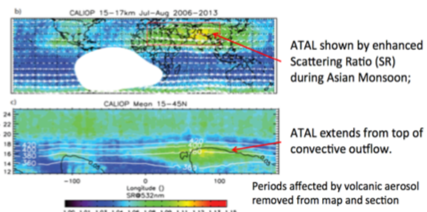


The BATAL campaign of 2015 made measurements of aerosol backscatter (COBALD), aerosol size and volatility (UW OPCs) for the ATAL using a hierarchy of balloon-borne payloads flown from sites in India (Gadanki, Hyderabad, Varanasi) and Saudi Arabia (KAUST)



Back trajectories computed from the UTLS at the flight location on 13 August show coincidences with deep convection (red dots) detected by the HIMAWARI-8 satellite

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)



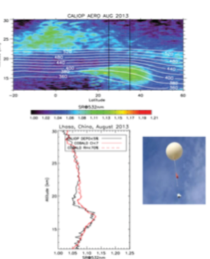
Questions: What is the origin of the ATAL? What is it made of? Are there regional climate impacts?

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

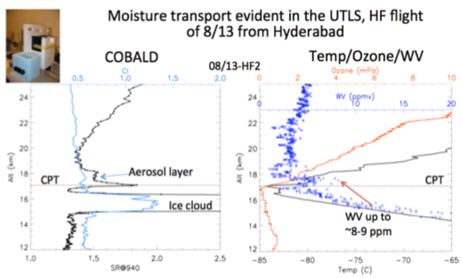
Mean (cloud-cleared) CALIOP SR, 60-120°E, shows ATAL between Pot. Temps. of 60 and 420K, extending to NH mid latitudes and tropics.

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)

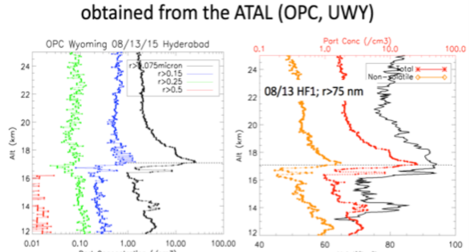


From Vernier et al., (JGR, 2015)

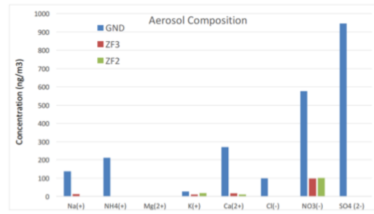


- Maximum of aerosol measured by COBALD found in vicinity of cold point tropopause
- Color Index (CI, blue line) between COBALD blue/red channels distinguishes aerosol (low values) from ice cloud (> 1, 15-16.5 km).
- Enhanced water vapor (up to 8 – 9 ppm) 17–18 km likely due to convective transport of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

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

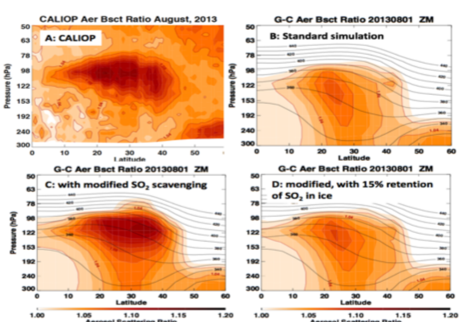


- Maximum SR coincides with peak in OPC number concentration for r > 75 nm at the cold point tropopause (data for unheated inlet shown)
- ~97% particles found in the size range 0.075 < r < 0.15 µm
- Heated (180°C) and unheated inlets on OPCs indicate ~90% small, volatile particles



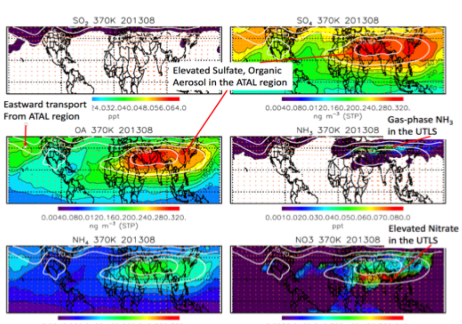
In August 2017, Aerosol composition measurements were obtained in the UTLS during zero-pressure (ZF) 3-h duration flights from Hyderabad using an Impactor, and post-flight Ion Chromatography analysis. These ZF observations reveal nitrate and some metal cations, but no sulfate was found, in contrast with ground-based (GND) observations.

Model Evaluation: The standard simulation underestimates Aerosol Backscatter measured by CALIOP in the ATAL region in August 2013. A possible explanation is excessive scavenging of aerosol precursors in convective updrafts in the model. The model tends to underestimate e.g., SO_2 in the upper troposphere, as evidenced by observations made during the SEAC4RS airborne campaign off 2013. Modified treatment of SO_2 scavenging in the model based on Henry's Law equilibrium, coupled with aqueous oxidation of SO_2 improved the agreement with CALIOP, but introduced high biases in SO_2 and sulfate compared with SEAC4RS observations. Allowing 15% retention of SO_2 in ice removed these biases but diminished agreement with CALIOP.

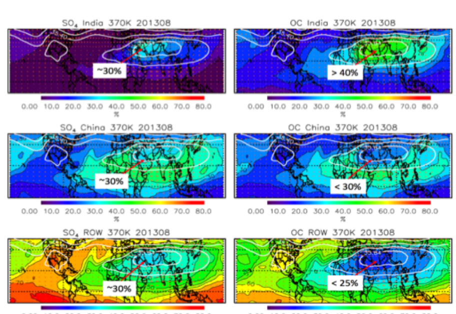


GEOS Chem CTM simulations

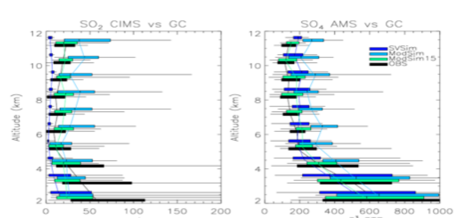
3-D CTM for gas-phase and aerosols transport and photo-chemistry in the troposphere, driven by MERRA meteorology (www.geos-chem.org), V9.02, 2x2.5 deg. 72 levels. GEOS-Chem Version 9.02 was used. Aerosols Components: OC, BC, SO_4-NO_3-NH_4, dust , sea-salt, limited SOA.



Simulated monthly mean maps at 370K of SO_2, sulfate, organic, ammonium, and nitrate aerosol, and gas-phase ammonia for August 2013 from GEOS-Chem show elevated concentrations of aerosol components in the ATAL region. Notable are elevated nitrate on the southern flank of the ATAL, and gas-phase ammonia on the northern flank.



Source apportionment: Percent contributions to UTLS sulfate and Organic Aerosol from India, China, and Rest-of-world in August 2013, from GEOS-Chem. Indian sources contribute mostly in the core of the ATAL; Chinese sources contribute in a horseshoe around the core, and to transpacific transport. Contours of Montgomery Stream function are also shown (white contours).



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 (dark blue); modified treatment of SO_2 scavenging in convective updrafts (pale blue); imposing 15% retention of SO_2 in ice cloud (green; bars span 25th – 75th percentile; whiskers 5th - 95th percentile;