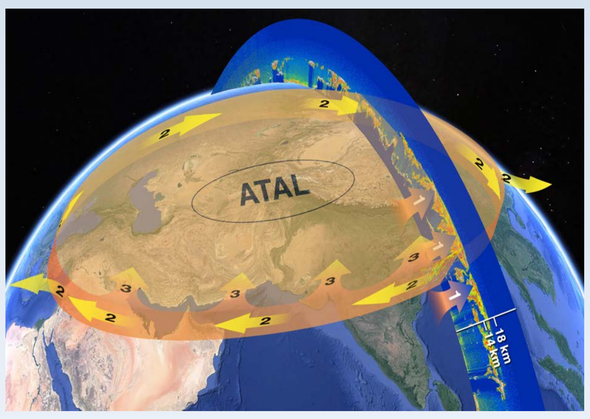
Toward the Chemical Composition of the Asian Tropopause Aerosol Layer

H. Vernier[1], N. Rastogi[2], T.D. Fairlie[3], J.-P. Vernier[4], A. Pandit[2], A. Patel[2], H. Gadhavi [2], Venkat. Ratnam[5], S. Kumar[6], V. Ravikiran[5] and F. Weinhold [7]

1 Thomas Nelson Community College, Hampton, VA, USA. 2. Physical Research Laboratory, Ahmedabad, India; 3. NASA Langley Research Center, USA; 4. Science Systems and Applications, Inc., Hampton, VA, USA; 5. National Atmospheric Research Laboratory, Gadanki, India;6. TIFR Balloon Facility, Hyderabad, India; 7. Swiss Federal Institute of Tech., Zurich, Switzerland.

1. Introduction



What is the ATAL ?

The Asian Tropopause Aerosol Layer is a recurrent aerosol feature observed in the Upper Troposphere and Lower Stratosphere over Asia during the Summer Monsoon (Vernier et al., 2011). It was discovered through satellite observations from the CALIPSO space-borne lidar and confirmed with the Stratospheric Aerosol and Gas Experiment (SAGE) II (Thomason and Vernier, 2013).

What do we know about its composition ?

The composition of the ATAL is still largely unknown. EDX analysis of aerosol samples near 10-12 km onboard commercial aircraft as a part of the CARIBIC program suggests a ratio between C and S near 2-10. Recent measurements during StratoClim highlight the presence of sulfate/nitrate above the tropopause (Rex, AGU).

Why is it important ?

The tropical UTLS is the entrance door for air to reach the global stratosphere through the Brewer-Dobson circulation. Since the ATAL represents one of the most important sources of aerosol in absence of volcanic eruptions. It has the potential to affect the earth radiative balance, stratospheric ozone chemistry and the properties of cirrus clouds.

2. Zero-pressure flights

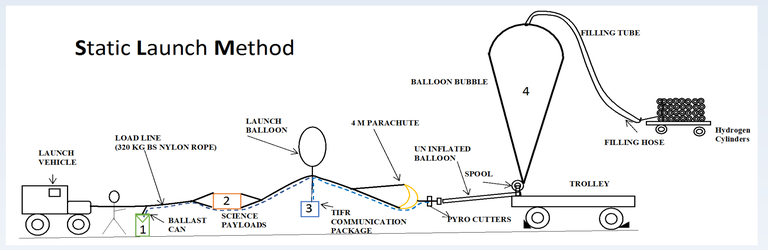
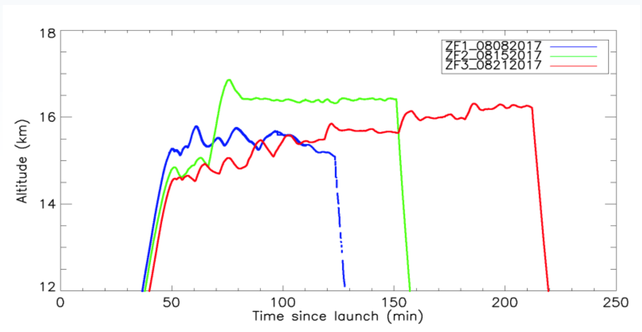
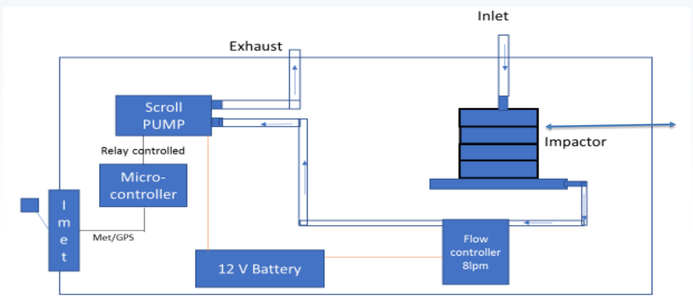


Photo of the launch preparation during BATAL 2018 (credit NARL/TIFR). Scheme of the Static Launch Method and the zero-pressure flight train developed for the BATAL campaign. From left to right : Ballast module, science payloads, TIFR communication, parachute and plastic balloons

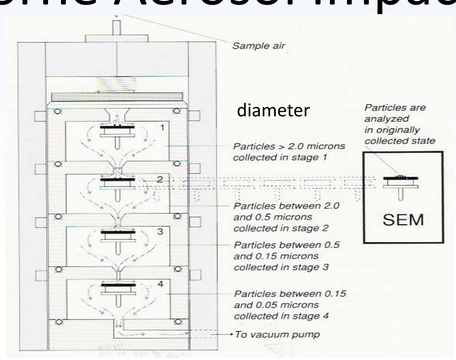


Time series of the 3 zero-pressure balloon flights GPS altitude during BATAL 2017. Release of ballast during the flights allows to stay aloft and compensate for the cooling of the gas inside the balloon and the reduction in buoyancy force. With this technique, the maximum duration flight was 2h50 min (ZF3). Extending the duration of balloon flights near the tropopause is used to sample aerosols with the balloon-borne impactor.

3. Description of the Balloon-borne Aerosol Impactor



Schematic diagram of the balloon-borne impactor

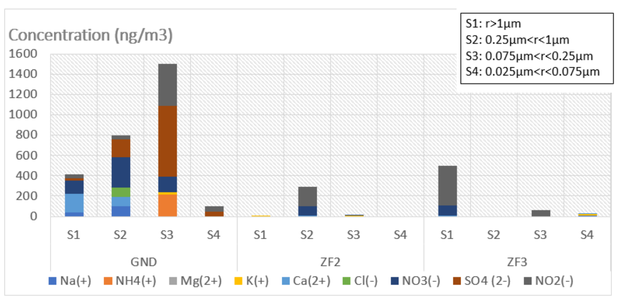


4-stage impactor

Loading of the impactor in the clean room at the TIFR balloon facility

A Balloon-borne aerosol Impactor (BAI) was developed to sample aerosols near the tropopause along zero-pressure balloon flights conducted at the Balloon facility of TIFR in Hyderabad during BATAL 2017. The 4-stage impactor is connected to a vacuum pump from which air is pulled from outside. A controller is connected to the vacuum pump to control a flow of 7 lpm. The pump is connected and controlled to an electronic system to operate at the desired altitude. For the ZF flights, the pump was switch on between 150 hPa and 70 hPa to collect particles through the impactor. PTFE filters were used and loaded in the impactor.

5. IC analysis results

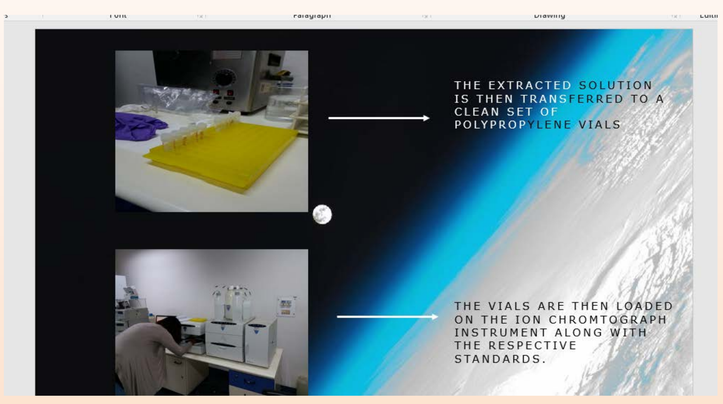


Major ions were measured with dual channel Ion Chromatograph (DIONEX, ICS-5000 DC).

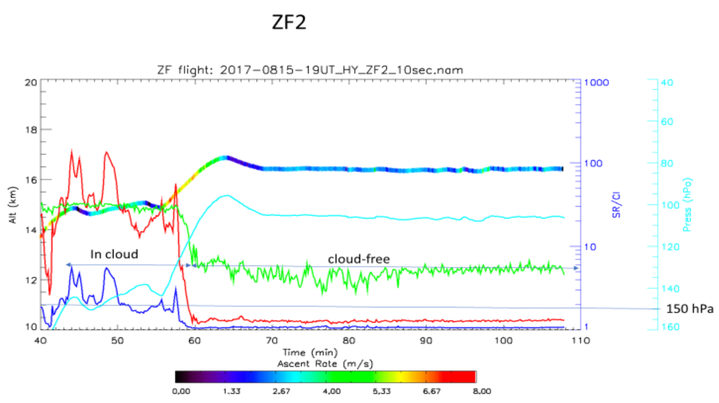
Ground: Among cations, Na^+ & Ca^{2+} were seen only on stages 1 &2 of ground samples (GND) with corresponding anions (NO_3^-, SO_4^{2-}, & NO^{2-}) co-existing at the same stage. NH_4^+ was seen only on stage 3 with a conc. of 212ng/m3. K^+ was also seen on stage 3 (fine mode) and it was likely from biomass burning. City pollution from Hyderabad is likely the source of those aerosols.

Flight ZF2 & ZF3. Significant amount of NO_3^- and NO_2^- was observed with traceable amount of the proxies of mineral dust (Ca^{2+}) and biomass burning (K^+). Other species were below 5 (for cations) to 10 (for anions) ng/m3 (detection limit). K^+ was seen on stage 3 of ZF2 together with the only existing anion NO_3^- thus suggesting the most likely form of KNO3 on this stage. Ca^{2+} was always associated with anions NO_3^- & NO_2^- on the same stages (ZF2/Stage 2, ZF3/Stage 1, thus depicting the probable existence of Ca^{2+} in the form of $Ca(NO_3)2$, & $Ca(NO_2)2$. However, NO_3^- and $NO2^-$ are in large excess compared to Ca^{2+} and thus they may be existing in other forms. Nitric acid Trihydrate (NAT, $HNO_3.3(H2O)$ could be another form in which NO_3^- may be present in the tropical UTLS (C. Voigt et al., 2008, ACP). Overall, Nitrate and Nitrite were the dominating population of aerosols near the tropopause during those flights.

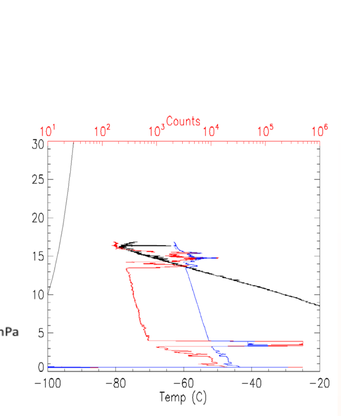
4. Lab experiment



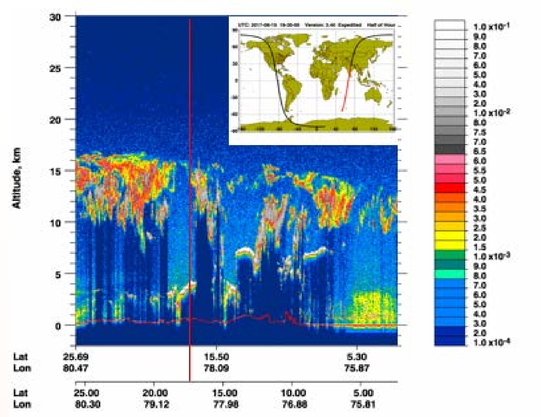
6. COBALD/CALIPSO measurements along ZF2



Time series of GPS and COBALD data.



COBALD vertical profile uncalibrated

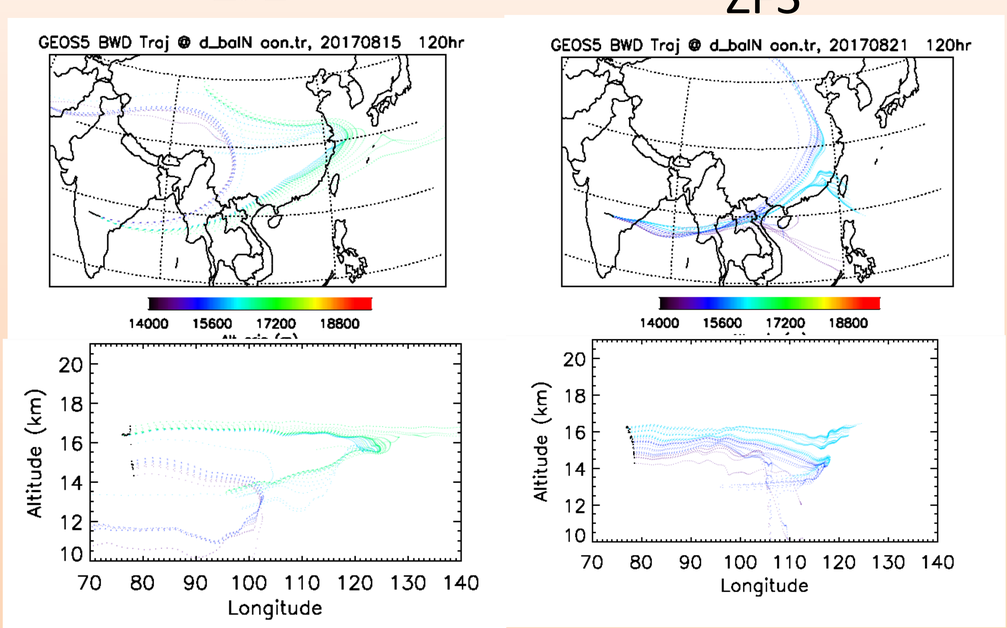


CALIPSO curtain passing near Hyderabad (Red line)

Time evolution of GPS altitude during ZF2 together with COBALD Scattering Ratio data at 455nm (blue), 970 nm (red) and Color Index (455/970; green). ZF2 was quasi-collected (within 50 km and 2h) of a CALIPSO overpass (see right). The position of the balloon flight is shown by the red line. The flight crossed a thin cirrus cloud between ~13-15 km.

7. Discussion: Origin of air masses

ZF2 ZF3



- Back trajectory analysis from ZF2/ZF3 using the LaTM (GEOS-5 winds) model show that air samples during both flights travelled along the South-Eastern side of the Asian Anticyclone with potential influence of convection from Eastern China.

8. Conclusions

We show :

- A new balloon-borne method for sampling aerosols in the UTLS using zero-pressure balloon flights
- The First Ion Chromatography analysis of UTLS aerosols collected during the Summer Asian Monsoon
- The Dominant presence of nitrate/nitrite aerosols within the ATAL region. Nitrate could be in the form of liquid nitrate and/or solid Nitric Acid Trihydrate (NAT) particles. More research is needed to understand the presence of nitrite.