Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution

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Abstract Satellite observations have shown that the Asian Summer Monsoon strongly influences the upper troposphere and lower stratosphere (UTLS) aerosol morphology through its role in the formation of the Asian Tropopause Aerosol Layer (ATAL). Stratospheric Aerosol and Gas Experiment II solar occultation and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) lidar observations show that summertime UTLS Aerosol Optical Depth (AOD) between 13 and 18 km over Asia has increased by three times since the late 1990s. Here we present the first in situ balloon measurements of aerosol backscatter in the UTLS from Western China, which confirm high aerosol levels observed by CALIPSO since 2006. Aircraft in situ measurements suggest that aerosols at lower altitudes of the ATAL are largely composed of carbonaceous and sulfate materials (carbon/sulfur elemental ratio ranging from 2 to 10). Back trajectory analysis from Cloud-Aerosol Lidar with Orthogonal Polarization observations indicates that deep convection over the Indian subcontinent supplies the ATAL through the transport of pollution into the UTLS. Time series of deep convection occurrence, carbon monoxide, aerosol, temperature, and relative humidity suggest that secondary aerosol formation and growth in a cold, moist convective environment could play an important role in the formation of ATAL. Finally, radiative calculations show that the ATAL layer has exerted a short-term regional forcing at the top of the atmosphere of −0.1 W/m² in the past 18 years.

1. Introduction

For more than a decade, unprecedented economic growth in Southeast Asia has led to increased release of pollutants into the atmosphere, forming the so-called Atmospheric Brown Cloud (ABC), with consequent impacts on air quality and regional climate [Ramanathan and Crutzen, 2003]. The ABC occurs primarily in winter when dry conditions associated with a temperature inversion trap pollution near the surface. In summer, deep convection associated with the Asian Summer Monsoon (ASM) ventilates pollution from the boundary layer into the upper troposphere and lower stratosphere (UTLS) [Randel et al., 2010]. Satellite observations and model simulations show recurring summertime maxima of CO, for example, near 100 hPa over the Asian monsoon region [Lawrence and Lelieveld, 2010; Kar et al., 2004; Park et al., 2007]. CO, which is insoluble and has a relatively long atmospheric lifetime (~2 months), is effectively lifted by deep convection and advected horizontally in the anticyclonic circulation, which prevails in the UTLS over Asia in summer. Similar plumes of HCN, CH₃O and H₂O extend from Southeast Asia to the Eastern Mediterranean Sea during the ASM [Park et al., 2007; Li et al., 2005; Randel and Park, 2006; Randel et al., 2010].

Given the potential impacts of aerosol on the radiative and chemical balance of the UTLS, a key question is whether or not natural and anthropogenic aerosols and/or their gas-phase precursors can also be transported to the UTLS during the monsoon. There are significant impediments to this transport as, in general, aerosols and their water-soluble gas-phase precursors are effectively scavenged and removed by precipitation [Giorgi and Chameides, 1986; Balkanski et al., 1993; Mari et al., 2010]. However, recent observations of cloud and aerosol layers in the UTLS from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar [Winker et al., 2010] have shown a maximum of nonvolcanic aerosol near the tropopause during the ASM [Vernier et al., 2011], confirmed by Stratospheric Aerosol and Gas Experiment (SAGE) II observations [Thomason and Vernier, 2013],
which we call the Asian Tropopause Aerosol Layer (ATAL [Vernier et al., 2011]). This paper further investigates the optical characteristics, decadal trend, likely composition, radiative impact, and origin of the ATAL.

In section 2, we describe the satellite (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) and SAGE II) and balloon backscatter (Compact Optical Backscatter Aerosol Detector (COBALD)) measurements used in this paper and compare the satellite data with coincident in situ data obtained above Lhasa, Tibet, in August 2013. In section 3, we focus on the longer-term evolution of aerosol extinction and backscatter from SAGE II between 1996 and 2005 and from CALIPSO between 2006 and the present. We find a positive trend in the satellite measurements. We use aerosol composition measurements from aircraft to estimate the radiative forcing of the plume over the past 15 years (section 4). Finally, the mechanisms responsible for ATAL's formation and its origin are discussed (sections 5 and 6).

2. Observations of ATAL From CALIPSO and COBALD

2.1. Description of Observations

The CALIOP lidar makes range-resolved measurements of elastic backscatter at 532 and 1064 nm and of linear depolarization ratios at 532 nm [Winker et al., 2010]. Due to the low signal-to-noise ratio of the CALIOP signal in the UTLS, a specific treatment of the level 1/v3.01 product was developed to infer backscatter profiles from the upper troposphere to 40 km [Vernier et al., 2009]. The data set used here is obtained following a similar algorithm, specifically (i) the total attenuated and perpendicular backscatter profiles at 532 nm are averaged every 1° latitude; (ii) the backscatter signal is corrected for the effects of attenuation by Rayleigh scattering and ozone absorption; (iii) the data are normalized using returns between 36 and 39 km rather than between 30 and 34 km altitude, used in the standard level 1 product; and (iv) cloudy pixels in the upper troposphere are removed using a volume depolarization ratio threshold of 5% [Vernier et al., 2009, 2011] and data below clouds are screened out.

Radiosondes, ozonesondes, Cryogenic Frostpoint Hygrometers (CFH), and Compact Optical Backscatter Aerosol Detector (COBALD) backscatter sondes were launched under small weather balloons from China (Lhasa and Kunming) between 2009 and 2014 [Bian et al., 2012] to study the convective outflow of the ASM. These launches provided the first in situ measurements of aerosol in the UTLS over this region. A number of backscatter profiles from COBALD sondes are shown here. COBALD downsizes the backscatter sonde of Rosen and Kjome [Rosen and Kjome, 1991] with a total mass of 540 g, suitable for operational balloon soundings; COBALD can be used to detect thin cirrus clouds [Brabec et al., 2012] and even estimate their ice water content from the backscatter signal [Cirisan et al., 2013]. Two high-power LEDs emit about 250 mW optical power each at wavelengths of 455 and 940 nm, respectively, and the light scattered back from molecules, aerosols, or ice particles is recorded by a silicon photodiode using phase-sensitive detection. The scattering ratio (SR; equation (1)) is inferred from the signal using the radiosonde air pressure and temperature to calculate the molecular backscatter.

\[
SR = \frac{\beta_{aero} + \beta_{mol}}{\beta_{mol}}
\]

where \(\beta_{aero}\) and \(\beta_{mol}\) are the aerosol and molecular backscatter at a given wavelength. SR is not quantified absolutely, but the analysis of the entire sounding profile imposes physical constraints such that SR is confined to an absolute error interval of 5%, while precision along the profile is better than 1% in the UTLS region.

2.2. CALIOP and COBALD Observations of ATAL

Figure 1 shows SR profiles from CALIOP along an orbit track on 4 August 2008 at 10:41 UTC from the Indian Ocean to India and up to Western China. The figure provides a characteristic picture of the spatial relationship of deep convective clouds and ATAL aerosols during a period undisturbed by volcanic activity. ATAL aerosols are highlighted by SR enhancements of ~1.2–1.3 over North India and Western China (~38–45°N) between 360 and 400 K potential temperature isentropic surfaces, north of the monsoon system, which is characterized by deep convective cloud (white). We note that the deep convective cloud over North India reaches the 380 K isentropic surface and penetrates the lower stratosphere [Holton et al., 1995].

Figure 2 shows a time series of CALIOP SR from July 2006 to June 2013 for the ATAL region (Figure 2a), together with a map (Figure 2b) and longitudinal cross section (Figure 2c) of SR averaged respectively in
July–August 2006–2013. Periods affected by volcanic aerosols from the eruptions of Kasatochi, Sarychev, and Nabro in summer 2008, 2009, and 2011 are excluded from the averaging. CALIOP observations show a region of elevated SR near 13–18 km extending from the Mediterranean region to eastern China (Figures 2b and 2c) and surmounted by the stratospheric aerosol (Junge) layer above 20 km (Figure 2c). The cross section also shows that ATAL extends from the region of monsoon convection (black contour delineates the cloud top) westward over the Mediterranean Sea. A substantial fraction of elevated SR values associated with ATAL is located above 380 K in the lower stratosphere (Figure 2c).

To verify satellite observations of ATAL, we use balloon-borne in situ measurements of backscatter and relative humidity with respect to ice (RHi) from COBALD and CFH, respectively. COBALD/CFH measurements, obtained from Lhasa in August 2013, are shown in Figure 3. Figure 3a shows a scatter plot of color index ($\text{CI}$; equation (2)) versus SR at 940 nm, colored by RHi:

$$\text{CI} = \frac{(\text{SR}_{940} - 1)}{(\text{SR}_{455} - 1)}$$

The data have been averaged on a regular pressure grid of 1 hPa resolution. The cluster of points with high RHi ($>$70%), $\text{CI} >$~10, and SR $>$$-$2 denotes the presence of large ice particles, in contrast with the signature of aerosol with lower SR present in a relatively dry environment (RHi $<$70%). The points with low RHi, SR $<$~1.3, and CI between 2 and 20–30 suggest a wide range of aerosol size populating the UTLS. The dependence of the aerosol size on RHi is investigated further in Figure 3b by plotting SR versus RHi. The COBALD backscatter data have been converted to 532 nm for comparison with CALIPSO, using the Angstrom exponent ($\alpha$) derived from the 455 and 940 nm COBALD channels:

$$\alpha = \log\left(\frac{\text{SR}_{940} - 1}{\text{SR}_{455} - 1}\right) \log\left(\frac{940}{455}\right)$$

$$\text{SR}_{532} = 1 + (\text{SR}_{940} - 1) \times [532/940]^{\alpha}$$

Figure 3b shows that SR increases with increasing RHi, consistent with growth of aerosol size in a humid environment. A linear relationship between SR and RHi ($SR = 1.06 + 0.06RHi$, correlation coefficient $= 0.3$) has been derived for RHi $<$ 70% and SR $<$ 1.3 (aerosol domain), which is used for comparison with satellite data. The cloud-cleared median SR profile was calculated using RHi $<$ 70%, as a cloud-clearing criterion, and the 18 individual profiles obtained during the 2013 Lhasa campaign (Figure 3c). Overall, the COBALD measurements confirm an increase of SR associated with aerosol near 13–18 km as observed by CALIPSO.
In order to verify the consistency of the RHi cloud-filtering approach, we have also employed a cloud mask methodology, in which we screen out data with CI > 7 and SR_940 > 2.5 (the CI approach). This method aims to screen out large ice particles associated with high CI, from smaller aerosol. Mie calculations indicate that such thresholds should remove ice clouds effectively and retain background aerosols such as sulfate.

**Figure 2.** (a) Time series of CALIOP scattering ratio (SR: total backscatter/molecular backscatter) between June 2006 and August 2013 averaged within the red box (Figure 2b, 15–45°N; 15–105°E) at 15, 16, and 17 km within ±0.5 km. Periods influenced by volcanic plumes are pointed with back lines. (b) Map of cloud-filtered aerosol SR from CALIOP between 15 and 17 km averaged in July–August over 5 years (2006, 2007, 2008, 2010, and 2012; note the noisy data from the South Atlantic Anomaly on CALIOP are masked in white). Arrows on the map represent the wind field from the European Center for Medium-Range Weather Forecasting (ECMWF) analyses. (c) Latitudinal cross section averaged over the same period between 15°N and 45°N. The black contour on the cross section represents the mean volume depolarization contour of 0.05 taken as a proxy for cloud presence.

In summary, we show that both cloud-clearing methods present very similar results and conclude that ATAL is not a residual of ice particles but a robust aerosol feature observed in the UTLS by CALIPSO and in situ measurements. These measurements and those obtained during the 2010 and 2012 COBALD campaigns.
(not shown) in Asia provide the first in situ confirmation of the presence of enhanced aerosol levels in the UTLS during the ASM.

3. ATAL Trend From Satellites

Due to a lack of prior in situ measurements similar to those of COBALD, study of the long-term change of aerosol loadings in this region relies on the satellite observations of SAGE II and CALIOP. A recent re-analysis of past SAGE II satellite observations between the late 1990s and 2005 has confirmed the enhancement of aerosol associated with the ASM from the early 2000s [Thomason and Vernier, 2013]. Combining these records is complicated by differences in their measurement technique, however. SAGE II uses passive solar occultation to measure extinction profiles whereas CALIOP uses nadir-viewing lidar to measure aerosol backscatter profiles. Of particular importance are the different approaches used to distinguish clouds from the aerosol measurements. Both data sets need careful cloud-clearing processes since residual cloud could easily mask the presence of the ATAL feature. To help mitigate the impact of imperfect cloud clearing, we have first assembled those data sets in a cloud-free region of the Asian anticyclone.

One such region is the Eastern Mediterranean Sea and the Middle East where the westward flow near 100 hPa associated with the Asian Anticyclone enables the transport of air masses from Southeast Asia [Lawrence and Lelieveld, 2010]. Ice clouds are not sustained in this region because of low relative humidity [Sassen et al., 2008]. We have analyzed 10 years of SAGE II data (1996–2005) and 8 years of CALIOP data (2006–2013) in this region. Figure 5 shows the ratio of summer (July–August) to winter (January–February) aerosol extinction and backscatter profiles and their averages between 13 and 18 km. Periods affected by volcanic eruptions in the SAGE II (Ruang/Raventador in 2002 and Manam in 2005) and CALIPSO data sets (Tavurvur in 2006, Sarychev in 2009 and Nabro in 2011) are excluded from the analysis. Consequently, we use data in winter 2008 and 2013 to calculate the CALIOP summer-to-winter ratios for 2006–2008 and 2010–2013, respectively.
In 1996 and 1997, the SAGE II summer-to-winter ratio of aerosol extinction (Figure 5, top) is below 1 from 13 km to 18 km, confirming a minimum of aerosol [Thomason and Vernier, 2013]. A peak in summer to winter aerosol appears in 1998 at 17 km, which increases to ~2.0 in the CALIPSO data for 2008. The mean summer-to-winter ratio calculated between 13 and 18 km from 1996 to 2013 increases by a factor of 3–4. In order to compare the SAGE II and CALIOP data sets in Figure 5, we assumed that the extinction-to-backscatter (lidar) ratio is similar in winter and summer. This implies the same aerosol size distribution and composition of UTLS aerosol in winter and summer. In Figure 6, we use an alternative approach, comparing integrated aerosol extinction from SAGE II between 13 and 18 km with integrated extinction obtained from CALIPSO.
aerosol backscatter, assuming a range of lidar ratio (40 and 70 sr$^{-1}$), consistent with liquid sulfate aerosol [Jäger et al., 1995]. Figure 6 (top) shows the resultant summertime Aerosol Optical Depth (AOD) time series for the Mediterranean region. To verify if this trend can be extrapolated to the entire ATAL region, we cloud-cleared the SAGE II data by eliminating all measurements with CR $>$ 3 (CR: 1020-to-525 extinction ratio) [Thomason and Vernier, 2013], a conservative approach which is effective in removing ice clouds and air masses with a cloud-aerosol mixture. Figure 6 (bottom) shows a similar trend in AOD over the entire ATAL region, as for the Mediterranean Sea (top). AOD values are shown to have increased by a factor 3 in the past 15 years. Despite the lack of an overlapping period between both instruments, which limits our ability to infer a trend, the overall good continuity between SAGE II and CALIPSO suggests an increase of aerosol extinction during the past 15 years over the entire ATAL region.

4. ATAL Composition and Radiative Impact

4.1. ATAL Composition

Direct evidence of aerosol composition from lower levels of the ATAL is available from a few in situ aerosol samples of the upper troposphere, collected as part of the Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC) program [Nguyen et al., 2006; Nguyen and Martinsson, 2007; Martinsson et al., 2014]. Aerosol samples were collected for ~2 h (over ~1000 km) using impactors on commercial aircraft and subjected to elemental composition analysis. Figure 7 shows estimates of carbon-to-sulfur mass ratio (C/S) obtained during seven flights between Europe and Asia in August 2006, 2007, and 2008. Potential vorticity (PV) was obtained for the CARIBIC data points using meteorological fields from the European Center for Medium-Range Weather Forecasting (ECMWF). Observations made in the Asian upper troposphere (altitude above 250 hPa and PV $<$ 1 PVU) are encircled with a black line and made large in Figure 7. On average, C/S in the Asian upper troposphere is 4.0 ($\pm$3.2 std) from which the carbon and sulfur concentration are respectively 36 and 13 ng/m$^3$ ($\pm$49%). Including observations from July 2006, 2007, and 2008, the number of observations within the ATAL region increases to 14 with C/S of 2.8 ($\pm$2.5 std) on average. Figure 7 shows a decrease in C/S from Asia to Europe, marking a transition between tropospheric and stratospheric air masses at 10–12 km along the flight tracks. Since the most likely form of S from the CARIBIC measurements is primarily sulfuric acid aerosol (H$_2$SO$_4$), the mass of sulfate is 3 times higher than the mass of S. The form of C is unknown, however, so the mass ratio between sulfate and carbonaceous aerosol cannot be determined easily. Nevertheless, those results highlight that aerosol at lower levels of the ATAL is mainly composed of carbonaceous aerosol and sulfate.

Figure 5. (top) Profiles of summer-to-winter ratio (also called anomaly) of aerosol extinction (SAGE II, 1996–2005) and backscatter (CALIOP, 2006–2013), averaged between 30°N–40°N and 5°E–45°E near the Eastern Mediterranean region. Periods affected by volcanic eruptions are excluded. (bottom) Time series of average anomaly profiles between 13 and 18 km, together with the 1 standard deviation.
4.2. Radiative Impact

Regional summertime aerosol optical depth associated with the ATAL has increased from ~0.002 to ~0.006 over the last 18 years as inferred from satellite observations by SAGE II and CALIOP (Figure 6). The radiative impact of the ATAL during the ASM is assessed with a stand-alone radiative transfer model \cite{Natarajan2011}. Aerosol effects include scattering (short-wave (SW) and long-wave (LW)), absorption (SW and LW), and emission (LW). The aerosol optical properties are adopted from \cite{Hess1998}. The input data for these calculations include pressure, temperature, water vapor, ozone, cloud optical depth, and cloud fraction profiles adopted from the Global Modeling Initiative model simulations and Model Era Retrospective-Analysis (MERRA) analyses. The radiative transfer model uses vertical profiles of aerosol extinction due to sulfate and organic carbon, which have very similar optical properties (refractive index, size distribution), as suggested by the CARIBIC measurements.

Model output consists of net radiative fluxes at the top of the atmosphere (TOA), and at the surface; vertical flux profiles and net heating rates are also obtained. Differences in the net flux with and without an aerosol enhancement represent the radiative forcing. For this calculation, we have used extinction profiles derived from CALIOP for December 2007 to January 2008 (winter) and July to August 2008 (summer) to compute the enhancement associated with ATAL and its associated radiative forcing estimate. Using two sets of radiative simulations with sulfate/organics, we found a mean radiative forcing at the TOA of \(-0.12\) and \(-0.09\) W/m\(^2\) for clear and total sky, respectively. Total sky calculations show a reduction of the absolute SW RF over the monsoon region due to cloudiness, while negative SW RF values in cloud-free, water-covered regions (e.g., east Mediterranean Sea), which contribute significantly to the overall negative SW RF, are relatively unchanged between clear and total sky calculations.

The global mean radiative forcing at the TOA from increasing CO\(_2\) between 2000 and 2010 has been estimated to be \(-0.3\) W/m\(^2\) \cite{Solomon2011}. While CO\(_2\) is a long-lived and well-mixed gas, ATAL occurs only regionally over Asia during the

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure6.png}
\caption{Mean summertime AOD from SAGE II and CALIPSO over the Mediterranean Sea (5–35°E; 30–40°N) and the entire ATAL region (5–105°E; 15–45°N). Note that the mean AOD from CALIOP was calculated using a lidar ratio from 40 to 70 sr.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure7.png}
\caption{Carbon-to-sulfur mass ratio (colored circles) obtained from elemental composition analysis from seven flights between Europe and Asia near 11–12 km in August 2006, 2007, and 2008. Measurements within the upper troposphere are encircled in black using the dynamical criteria of potential vorticity <1 PVU.}
\end{figure}
monsoon; nevertheless, ATAL’s RF appears comparable to that of greenhouse gases and should therefore be considered in regional surface temperature changes at decadal scale. Using intermediary and fully coupled ocean–atmosphere climate models, Solomon et al. (2011) and Fyfe et al. (2013) found that global increase of stratospheric aerosol levels between 2000 and 2010 led to a surface cooling of around 0.07 K. While it is premature to affirm that ATAL could lead to a similar cooling without a climate model investigation, these results suggest that ATAL could have contributed to summertime reductions in surface temperature over Asia in the past 15 years.

5. Origin of ATAL

We investigate the source of ATAL by combining cloud top brightness temperature (BT) from the KALPANA geostationary (GEO) satellite, which we use as a proxy of deep convection (Devasthale and Fueglistaler, 2010), with back trajectory calculations for air initialized at CALIPSO observation points. The trajectories were computed using the NASA Langley trajectory model (Fairlie et al., 2014) and examined for coincidence with deep convection. Figure 8 illustrates the trajectory approach. Air parcels are initialized at the location of CALIOP observations between 14 and 18 km (green line) and back trajectories are computed in search of intersection with deep convection. Air parcels are tagged as convectively influenced (circle filled in orange), when the BT observed by KALPANA along their back trajectories falls below 220 K (Devasthale and Fueglistaler, 2010). Air parcels are also tagged with SR observed by CALIPSO at initialization; air parcel SR is assumed approximately conserved along the back trajectories. We initialized more than 50,000 air parcels between 1 and 16 August 2008. BT along those trajectories was extracted from the KALPANA satellite (1 h time resolution) and only air parcels experiencing deep convection (BT < 220 K), and with trajectories of less than 10 days, are retained.

Figure 9 shows a gridded 1° × 1° map of the aerosol origin index (AOI) for 1–16 August 2008. We have defined AOI as the average air parcel SR at the location where those air parcels (at least five) have encountered deep convection:

\[
\text{AOI}(i,j) = \frac{\sum_{n=0}^{n} \text{SR}_n}{n}
\]

with \( n > 5 \).

Figure 9 suggests that convection over the northern India subcontinent makes a major contribution to the ATAL. High AOI values are observed over the northern Indian subcontinent where the colocation of deep convection and high levels of ground pollution could explain ATAL’s formation. On the other hand,
convection over ocean, including the South China Sea and the southern Bay of Bengal, seems to have a limited impact on aerosol loadings observed within the Asia Monsoon anticyclone. We investigated the dependence of the AOI on the time between observation by CALIPSO and corresponding trajectory encounter with deep convection but could not identify a statistically significant relationship.

6. Relationships of SR With Humidity, Temperature, and CO in the ATAL

While black carbon (BC) and organic carbon (OC) emissions have grown only slightly over the past two decades [Ohara et al., 2007], Asian emissions of gas precursors for aerosol formation such as SO2 and non-methane hydrocarbon carbons (NMHC) have increased by 25% and 60%, respectively [Ohara et al., 2007]. The CARIBIC experiment has revealed that air parcels in the UT during the ASM were marked with high levels of NMHC from biofuel burning and transportation-related emission [Baker et al., 2011]. Given their low saturation pressures and chemical lifetimes of up to 10 days [Baker et al., 2011], NMHCs are a potential important source of gas precursors for secondary organic aerosol formation in the upper troposphere. During the CARIBIC flights, NMHC and CO were shown to be highly correlated. Thus, we expect to see a relationship between the temporal evolution of CO and SR in the ATAL observations.

Figure 10 shows the temporal evolution from June 2006 to August 2008 of CO and RHi from the Microwave Limb Sounding (MLS, V2.21) at 100 hPa, together with the number of mesoscale convective systems (MCSs) reaching at least 14 km taken from Tropical Rainfall Measurement Mission (TRMM) observations, and the mean temperature and SR between 14 and 18 km from radio occultation of the Constellation Observing System for Meteorology, Ionosphere, and Climate (COSMIC) and CALIOP, respectively. Given the relative broad vertical resolution of the MLS data at 100 hPa (~±2 km), we have averaged the vertically resolved data (CALIOP and COSMIC) between 14 and 18 km. The aerosol peak observed in August each year lags the peak in CO by ~1 month and is in phase with the temperature minimum observed in the UT. RHi and temperature are anticorrelated during the dry season suggesting that temperature is the primary factor controlling RHi in the absence of convective sources of moisture. When the convective season starts (June), CO and RHi increase together as increased occurrence of MCSs brings moisture and pollution to the upper troposphere. At the end of the convective season (September/October), the decrease in RHi lags the drop in CO by ~1 month, while the peak in RHi appears to be prolonged by sustained low temperatures. Simultaneous drops in SR and RHi at the end of the convective period are consistent with the positive correlation found...
between SR and RHi in the balloon-borne in situ measurements at Lhasa (Figure 3b), and with the spatial gradient in SR (Figure 2) and RHi (not shown) between the Asian convective region and the Eastern Mediterranean Sea. These observations suggest that RHi exerts a control on aerosol size in the ATAL.

Nevertheless, the 1 month phase lag between aerosol SR and RHi at the beginning of the convective period (May/June) suggests RHi does not fully explain the SR variation. Recent field campaigns have shown high concentrations of nanometric particles near and below the tropopause (350–370 K) above Africa, Brazil, and Australia [Weigel et al., 2011]. Growth of such particles through condensation and coagulation could contribute to the development of the ATAL. The time for such particles to grow to a size detectable by remote sensors such as CALIOP and SAGE II may explain the time lag between CO/RHi and SR (Figure 10).

7. Conclusions

We have analyzed satellite, balloon, and aircraft observations of aerosol optical properties and composition in the UTLS over Asia to better understand the decadal trend, likely composition, origin, and radiative impact of the Asian Tropopause Aerosol Layer. Balloon-borne backscatter measurements from China confirm the summertime aerosol enhancement near the tropopause observed by the CALIPSO space-borne lidar since 2006. ATAL is a robust aerosol feature observed up to 420 K into the lower stratosphere from satellites (SAGE, CALIPSO) as well as in situ measurements. We found a positive trend in summertime UTLS aerosol optical depth of about 0.004 resulting in a regional total sky radiative forcing of around −0.1 W/m² over the past 18 years. This comparison to published RF estimates of 0.3 W/m², associated with the global increase in CO₂, a climate model investigation is required to study the impact of ATAL on tropospheric temperature.

Aircraft measurements from the CARIBIC experiment suggest that sulfate and carbonaceous aerosols dominate the ATAL. The time delay of a month between peaks of CO and aerosol within the ATAL region, together with recent observations of newly formed particles in the tropics (e.g., SCOUT-AMMA in West Africa and SCOUT-O3 in Darwin), suggests the potential importance of secondary aerosol formation. We found a positive correlation between SR and RHi in satellite and in situ data, which also suggest that RHi exerts a control on aerosol size in the ATAL.

We used back trajectories initialized from CALIPSO to investigate the potential origin of ATAL by searching for coincidence with deep convection. Our analysis suggests that monsoon convection over the Indian subcontinent combined with high levels of pollution from the ground is mainly responsible for ATAL’s formation and impact the global UTLS aerosol budget.

Acknowledgments

All data and codes used to produce this study can be obtained by contacting Jean-Paul Vernier (jeanpaul.vernier@nasa.gov) and will be shared through a dropbox system (https://www.dropbox.com/home) under the folder JGR-ATAL-2014 (https://www.dropbox.com/sh/k3v1mgocj3tpz4/ AAC25oBOkjYE_3cxedZS7mTAa?dl=0). We thank Mian Chin for the discussion on the comparison between CALIPSO and the GOCART model (not shown in this paper). We also thank Robaidek from the University of Wisconsin and ISRO for making available the KALPANA satellite data via the McIDAS-V system (http://www.ssec.wisc.edu/data/!). The following satellite data used in this study are publically available at CALIPSO, https://eosweb.larc.nasa.gov/project/calipso/calipso_table; SAGE, https://eosweb.larc.nasa.gov/project/sage2/sage2_table; MLS, http://mirador.gsfc.nasa.gov/cgi-bin/mirador/homepageAlt.pl?keyword=MLS; TRMM, http://mirador.gsfc.nasa.gov/cgi-bin/mirador/homepageAlt.pl?keyword=TRMM; and Cosmic GPS: http://www.cosmic.ucar.edu/data.html. The COBALD balloon campaign in Lhasa was supported by National Natural Science Foundation of China (grants 91337214 and 41175040).

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