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LETTER

In situ measurements and backward-trajectory analysis of high-concentration, fine-mode aerosols in the UTLS over the Tibetan Plateau

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Abstract

An aerosol layer in the upper troposphere and lower stratosphere over the Asian summer monsoon (ASM) regions, namely, the Asian tropopause aerosol layer (ATAL), has been observed based on satellite remote sensing and in situ measurements; however, its source is still under debate. In August 2018, an experimental campaign over the Tibetan Plateau at Golmud (GLM, 36.48°N, 94.93°E) was performed, during which a balloon-borne Portable Optical Particle Counter was used to measure the aerosol particle profile. Backward-trajectory simulations were conducted with the Massive-Parallel Trajectory Calculations model to investigate the possible sources and transport pathways of the observed particles. The in situ measurements showed a robust ATAL around the tropopause, 16 km above sea level, with a maximum aerosol number density of 35 cm⁻³ and a maximum aerosol mass concentration of 0.15 μg m⁻³ for particles with diameters between 0.14 and 3 μm. The aerosol particles in the ATAL are mostly smaller than 0.25 μm in diameter, accounting for 98% of all aerosol particles detected. The backward-trajectory analysis revealed that the air parcels arrived at the altitude of the ATAL through two separate pathways: (1) the uplift below the 360 K isentropic surface, where air parcels were first elevated to the upper troposphere and then joined the ASM anticyclonic circulation; and (2) the quasi-horizontal transport along the anticyclonic circulation, located approximately between the 360 and 420 K isentropic surfaces. The complex transport pathways may aggravate the challenge of analyzing the composition of the ATAL, and further observation campaigns are required to extend our knowledge.

1. Introduction

The Tibetan Plateau (TP), as the highest (average elevation >4 km above sea level: ASL) and broad (~2.5 × 10⁸ km²) plateau in the world, significantly influences the weather and climate changes in China and even worldwide via its unique land surface processes (Wu et al 2007). Westerlies, the Indian monsoon, and the Asian monsoon have a great impact on the atmospheric circulation patterns over the TP, and the thermal and dynamical processes over the TP also produce strong feedback on the monsoon system, precipitation, and the regional climate of the Northern Hemisphere (Yao et al 2012).

The Asian summer monsoon (ASM) anticyclone, existing from May–June to August–September in each year, is a dominant circulation system in the upper troposphere and lower stratosphere (UTLS) over the
TP during the summer seasons (Yanai et al 1992, Ueda and Yasunari 1998). It has been suggested that the main air source within the ASM anticyclone is from the Indian subcontinent (Bergman et al 2013, Yan and Bian 2015). Previous studies showed that the ASM may transport tropospheric air into the stratosphere effectively by creating a dome or ‘bubble’ of tropospheric air above the zonal mean tropopause (Randel and Park 2006, Park et al 2007, Pan et al 2016). It is further noted that the ASM coupled with deep convection uplifting may serve as a more efficient chimney, venting tropospheric aerosols up to the UTLS, thereby significantly influencing the high-altitude aerosol properties (Li et al 2005, Randel et al 2010, Vernier et al 2015, Yu et al 2015, 2017).

A layer of enhanced aerosol scattering associated with the ASM was observed by the Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) (Vernier et al 2011) and the Stratospheric Aerosol and Gas Experiment II (SAGE II) (Thomason and Vernier 2013). This aerosol enhancement, referred to as the Asian Tropopause Aerosol Layer (ATAL), occurs from June to August at approximately 0–160 °E and 15–45 °N. The layer extends vertically from approximately 13 to 18 km and can generate an important influence on the regional forcing at the top of the atmosphere (Vernier et al 2015). The TP is located at the core region of the ASM, thereby acting as a significant transport pathway for pollutants to enter the stratosphere (Fu et al 2006, Wright et al 2011, Bergman et al 2013, Gu et al 2016). Its function as an ‘air pump’ is particularly prominent due to the strong solar radiation heating on the high massive ‘mesa’ in the boreal summer (Xu et al 2014, Zhang et al 2018).

Although the existence of ATAL has been verified, its source and composition remain controversial. For example, aircraft in situ observations suggest that the ATAL at lower altitudes is largely composed of carbonaceous and sulfate components and may have formed as the result of the transporting of surface pollutants by the monsoonal convection over the Indian subcontinent (Vernier et al 2015). By using a sectional aerosol model, the Community Aerosol and Radiation Model for Atmospheres (CARMA), coupled with the Community Earth System Model version 1 (CESM1), Yu et al (2015) indicated that the ATAL is mostly composed of sulfates, surface-emitted organics, and secondary organics. With Goddard Earth Observing System chemical transport model (GEOS-Chem) simulations, Gu et al (2016) implied that nitrate aerosol is the most dominant aerosol species in the ATAL. Höpfner et al (2019) recently suggested that ammonium nitrate particles could be transported into upper troposphere from ground ammonia sources by convection during Asian monsoons. Using the ECHAM/MESSy Atmospheric Chemistry (EMAC) model, Ma et al (2019) indicated that the ATAL over the TP, to a large extent, is caused by mineral dust emitted from the northern TP and slope areas, lofted to an altitude of at least 10 km, and accumulating within the ASM.

Most previous studies focused on ATAL formation from the transport of pollutants originating from south and southeastern Asia to the UTLS over the TP. However, the natural sources around the TP, which may also be one of the important potential sources for the formation of the ATAL, have not been reported and are still not well understood. Moreover, detailed observations and trajectory simulations are required to verify the results of the atmospheric chemistry model and to answer the questions of how the accumulated aerosol in the upper troposphere over the TP can be further transported to the lower stratosphere. Aerosol observations in the upper troposphere, either from satellites or other platforms (Qin et al 2018), have been very sparse (Vernier et al 2011). For a specific region such as the ASM region, high-quality, ongoing, direct in situ observations of aerosols will significantly reduce some limitations of the temporal and vertical resolution of satellite data and help to further verify the composition of the UTLS aerosol, especially within the thin ATAL. Given its high-altitude, unique geographical location, and consequently great challenge and high expense, in situ observations of aerosols at the TP are quite important and scarce, except, to the best of our knowledge, the following two studies. First, Tobo et al (2007) used a balloon-borne optical particle counter (OPC) (Hayashi et al 1998) to measure the vertical profile of aerosols in Lhasa, China (29.4 °N, 91.1 °E; 3650 m ASL, as shown in figure 1), in 1999. They detected relatively high number concentrations (0.7–0.8 cm$^{-3}$) of submicron-sized aerosols (0.3–1.2 μm in diameter) present near the tropopause region and inside the ASM region and were suggested to be induced by the deep convection over the TP. Later, in 2015, by analyzing measurements from the Portable Optical Particle Counter (POPS) (Gao et al 2016) in Kunming (25.01 °N, 102.65 °E; 1889 m ASL, as shown in figure 1), Yu et al (2017) confirmed the existence of a robust enhanced aerosol layer that extends up to 2 km above the tropopause. Both studies have indeed provided valuable observations about the aerosol layer during the ASM period; nevertheless, as specified later, in section 4, they also have their own limitations. Further investigations based on in situ observations are thus still urgently needed to better understand the UTLS aerosol characteristics, especially at the core areas of the TP during the ASM period. As part of the five-year (2018–2022) stratosphere–troposphere exchange experiment during the ASM (STEAM) project, a multi-location joint atmospheric campaign was performed over the TP in August 2018 (Zhang et al 2019). During the five years of STEAM project execution, multiple platforms/instruments, including long-duration stratospheric balloon, special sounding system, and ground-based instruments will be successively deployed to provide comprehensive

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Atmospheric parameters (aerosol, ozone, water vapor, CO₂, CH₄, CO, turbulence, radiation, lightning, etc). In 2018, a POPS aboard a stratospheric balloon was released at Golmud (GLM) to provide vertical aerosol measurements in the TP region. Based on the POPS measurements and model simulations, this study aims to analyze the aerosol properties and aerosol sources of the UTLS aerosol layer detected during the campaign.

The paper is organized as follows: section 2 briefly describes the in situ observations and backward-trajectory simulations. The main results are presented in section 3, followed by a discussion in section 4 and a summary in section 5.

2. Measurements and backward-trajectory simulations

2.1. In situ aerosol measurements

On 16 August 2018, an atmospheric experiment was conducted at GLM (36.48°N, 94.93°E, shown in figure 1). GLM, with an average altitude of 2780 m, is located in the hinterland of the TP and has a plateau continental climate; it is mainly composed of the central and southern Qaidam Basin and Tanggula Mountains. This site is surrounded by wasteland with minor influences from industry emissions. A detailed introduction to the GLM campaign can be found in Zhang et al (2019). As one of the main payloads in the campaign, the POPS aboard the stratospheric balloon was launched at 6:10 local standard time (LST) on 16 August 2018. The stratospheric balloon, with the maximum volume of 4000 m³, is made of polyethylene. The aerosol measurements are uncontaminated by the balloon platform that are powered by electricity all the way. The POPS provided measurements for ~70 min during its ascent to 21 km with a mean velocity of ~5 m s⁻¹; it then collected measurements approximately 21 km until the payloads were released with a parachute at 8:55 LST. The payloads landed safely 30 min later and were retrieved. The POPS, designed with a small size and low weight, can measure particles between approximately 0.14 and 3 μm in diameter using single-particle light scattering (Gao et al 2016). The particle size is calculated from the pulse signal caused by each particle passing the laser beam. The air is pumped into the POPS from one side and is vented out on the other side, to avoid potential aerosol residual and contamination. We deployed the corrections of temperature and particles loss at the sample port when processed the observational data. The instrument is able to provide the aerosol particle number density and size distribution with a sufficiently low detection limit to capture over 60% of the ATAL particle surface area (Yu et al 2017). Additional measurements of the temperature, pressure, and relative humidity were provided by a radiosonde aboard the same balloon.

2.2. MPTRAC model

The Massive-Parallel Trajectory Calculations (MPTRAC) model is a Lagrangian particle dispersion model (Hoffmann et al 2016). In this model, trajectories for individual air parcels are calculated based on the numerical integration of the kinematic equation of motion, and simulations are driven by wind fields from global meteorological reanalyses. In this study, the MPTRAC model is driven with the ERA-Interim data (Dee et al 2011), interpolated on a 1° × 1° horizontal grid on 60 model levels with the vertical range extending from the surface to 0.1 hPa.
ERA-Interim data are provided at 00:00, 06:00, 12:00 and 18:00 UTC. The outputs of the model simulations are given at the same time interval. Winds that are more accurate will generate trajectories that are more realistic. In this study, the ERA-interim winds are chosen to drive the model for the trade-off between accuracy and speed of the calculations. For more details of the dependency of the accuracy of trajectories, please refer to Hoffmann et al. 2016. In the MPTRAC model, turbulent diffusion is modeled by uncorrelated Gaussian random displacements of the air parcels with a zero mean and standard deviations $\sigma_x = \sqrt{D_h \Delta t}$ (horizontal) and $\sigma_z = \sqrt{D_z \Delta t}$ (vertical). $D_h$ and $D_z$ are the horizontal and vertical diffusivities, respectively, and $\Delta t$ is the time step for the trajectory calculations. In this study, $D_h$ and $D_z$ are set to 50 and 0 m$^2$s$^{-1}$ in the troposphere and 0 and 0.1 m$^2$s$^{-1}$ in the stratosphere, respectively. The sub-grid-scale wind fluctuations are simulated by a Markov model (Stohl et al. 2005, Hoffmann et al. 2016). There is no chemical reaction associated with aerosols, coagulation, or sedimentation of aerosols, so in this study, we only emphasize on the trajectories but not the chemical process along with the trajectories. For more information about the MPTRAC model refer to Hoffmann et al. (2016) and Wu et al. (2017, 2018).

3. Results

3.1. Meteorological background conditions at GLM

Figure 2 shows the meteorological background at 01:00 UTC (09:00 LST) on 16 August 2018 based on the ERA5 reanalysis. Note that the in situ observations were conducted from 6:10 to ~9:20 LST. The thick black dashed contour in figure 2(a) indicates the geopotential height (GPH) of 14320 m at the 150 hPa pressure level, denoting a commonly used boundary of the ASM anticyclone (Randel and Park 2006). As shown in figure 2(a), the GLM site is located at the north of the ASM and at the southern edge of the subtropical jet, which indicates that the westerly winds and the anticyclonic circulation around the ASM may play a key role in the origin and transport of the observed particles presented in the following section. The vertical section (figure 2(b)) shows the thermal tropopause (WMO 1957), 16 km, which was calculated from the temperature profile provided by the radiosonde GPS attached to the POPS.

3.2. Aerosol measurements over the GLM site

Figure 3 shows the vertical distribution of the aerosol number density and mass concentration measured by the POPS during the GLM campaign. Note that the POPS’s power was cut off at ~5 km ASL during the descending period to protect the instrument. The aerosol measurements generally agreed well during the ascending and descending periods. Notably, this POPS went through the ATAL both upward and downward and indicated a clear aerosol peak at 14–18 km, i.e. ±2 km around the tropopause (16 km). At a more detailed level, as specified in the follows, a certain quantitative discrepancy existed between the ascending and descending periods, due to the temporal and spatial variations of the aerosol distributions. The aerosol peak, with a maximum aerosol number density of 27 cm$^{-3}$ and a maximum aerosol mass concentration of 0.13 μg m$^{-3}$, occurred almost at the tropopause during the ascending period; more aerosols tended to be detected at the tropopause during the descending period, with the above two values of 35 cm$^{-3}$ and 0.15 μg m$^{-3}$, respectively.

To better understand the vertical distribution for different aerosol sizes, the aerosols are further divided into 27 particle size bins; their number densities during the ascending period are shown in figure 4(a). This figure indicates that the majority of the POPS-based
aerosols in the UTLS were fine-mode particles that occurred most frequently at the first five smallest particle size bins (i.e. 0.140–0.157, 0.157–0.176, 0.176–0.198, 0.198–0.222, and 0.222–0.249 μm in figure 4), accounting for 98% of all particle numbers in the observed UTLS aerosol layer. Few aerosols within 0.249–3.0 μm (bins from 5 to 27) were detected. To clearly see the distributions of the first five smallest particle size bins that occurred most frequently, their vertical profiles are further displayed in figure 4(b). The peak aerosol concentrations synchronously occurred at the tropopause for these five bins. The number density was at a maximum (10.6 cm⁻³) for aerosol sizes within 0.140–0.157 μm; it then decreased greatly in the rest of the four bins with peak magnitudes of 7.5, 4.4, 2.6 and 0.8 cm⁻³, respectively.

3.3. Backward-trajectory simulations
To investigate the possible source of the aerosols observed by the POPS, we selected the observed particles with particle number densities larger than 5 cm⁻³ and then traced these particles backward for one month (from 16 August 2018 backward to 16 July 2018). Figures 5(a), (b) shows the longitude and latitude of the 20 representative trajectories, colored by days reversed from 16 August 2018 and the potential temperature, respectively. Figures 5(c)–(f) demonstrates the vertical cross sections corresponding to the trajectories shown in figure 5(a). As shown in figures 5(a) and (c), during the initial 10 days, the backward trajectories were mainly driven by the anticyclocnic circulation around the ASM; the majority of the trajectories came immediately from the west.
between isentropic surfaces ranging from 365 to 420 K, while the minority of the trajectories came from the east approximately 5 d before the observation and ranged from 360 to 370 K. As seen in figures 5(b)–(f), the hourly trajectory positions are plotted, and large distances between single dots indicate rapid uplift. The longitude and latitude of the origins of the 30 d backward trajectories are denoted by black dots in (a), (b) and by gray dashed lines in (c)–(f).

To statistically analyze the possible origins of the observed large particle number density, a new set of 100 000 air parcels was assigned to the selected observations in the GLM campaign, and the number of the air parcels assigned to each of the observations is linearly proportional to the particle number density of the observations. Then, the backward trajectories were initialized by the 100 000 air parcels and traced backward for the same time period (from 16 August 2018 backward to 16 July 2018). The altitudes of the air parcels are shown in figure 6. It is observed that on 11 August 2018, 5 days before the observation, all the air parcels were from the northwest and southwest, moving clockwise along the western part of the anticyclonic circulation, and the altitude range was approximately 14–15 km. On 6 August 2018, the air parcels mainly from the west and south were at approximately 15 km, but a small fraction, particularly the part in the east, arose from 10 to 13 km. From 1 August 2018 backward, the air parcels were from all directions around the observation site, and a larger
portion of the air parcels was elevated from the lower troposphere.

As demonstrated in figure 5, the backward trajectories revealed that the air parcels went through two separate pathways to arrive at the location of the observations. One pathway is the uplift below the 360 K isentropic surface, where the air parcels were first elevated to the upper troposphere and then joined the anticyclonic circulation before going to the observation location. The other pathway is the quasi-horizontal transport along the anticyclonic circulation. Figure 7 shows the proportion of air parcels between the 360 and 420 K isentropic surfaces. Figure 8 shows the proportion of air parcels below the 350 K isentropic surface. Comparing the panels in figure 7 with the panels for the same date in figure 8, it is clear that before 1 August 2018, almost all the air parcels took the second pathway (the quasi-horizontal transport along the anticyclonic circulation) to the observation location. The air parcels that took the quasi-horizontal transport pathway became more dispersive in July 2018 (see figure 7). However, 20 d before the observation on 27 July 2018, the convection at the south of the Himalayas played a significant role in elevating the air parcels, and the panels for other dates in July (figure 8) revealed the surface origin of these elevated air parcels: the Southern Hemisphere tropics caused by the Somali jet, the southern Asian continent, and the West Pacific.

4. Discussion

4.1. The progress and the limitation of the current in situ observation

As mentioned earlier, two studies, i.e. those by Tobo et al. (2007) and Yu et al. (2017) performed in situ aerosol observations over the TP. The first study used an OPC to measure the number concentrations of aerosol particles for five size bins (i.e. diameter ≥ 0.3, 0.5, 0.8, 1.2 and 3.6 μm) at Lhasa, which is located in the southern part of the TP. By deploying the newly developed and more advanced POPS, Yu et al. (2017) confirmed a robust ATAL in Kunming over the TP. While valuable knowledge has been presented by both studies, they still have limitations. For example, the instrument used in the first study was not sufficiently sensitive to provide a detailed size distribution. Note that the minimum detection limit of the OPC was...
0.3 μm, indicating that it will not detect most fine aerosol particles in the ATAL. The second study can provide more detailed aerosol properties between 0.14 and 3 μm, but it was performed at the peripheral region of the TP, and thus, it might be difficult to deduce the ATAL properties over the core TP areas, especially for the short-term local convective transport.

To observe the aerosol number density and mass concentration at GLM in this study, the POPS, to a certain extent, is able to compensate for the limitations in the previous in situ observations. However, it is still subject to limitations. For example, the collected samples in this single campaign may introduce the problem of representativeness, so further verification is needed by conducting more in situ observations at the GLM station and, if possible, a few other stations in the TP. Provided that the altitude range of the ATAL at GLM is acquired in advance of our campaign in 2018, by regulating the stratospheric balloon platform, we can control the POPS within the ATAL for a long period to provide more detailed horizontal and vertical variations. This observational mode will be achieved in a future campaign. Moreover, the direct measurements of the chemical composition of the ATAL are still unavailable at the moment and are limited by the current payloads aboard the balloon.

4.2. The complicated composition of the ATAL
There has been disagreement on the main composition of the ATAL. As shown by the backward-trajectory analysis in section 3.3, in the 30 d before the particles were observed near the tropopause, some of them originated from near the earth’s surface and ventured upward towards the UTLS region by convection. However, the majority of the particles were transported along the ASM anticyclonic circulation, and the trajectories covered a large horizontal area (see figures 6 and 7). The types of aerosols or their precursors that existed along the trajectories together with the background meteorological conditions and chemical components play a role in modulating the ATAL composition. Therefore, the ATAL has a complex composition, such as the already proposed components of sulfate aerosols, nitrate aerosols, carbonaceous materials, and mineral dust (Yu et al 2015, Gu et al 2016, Höpfner et al 2019, Ma et al 2019).

Figure 7. Distribution of the air parcels (in the percentage of the total number of air parcels) between the 360 and 420 K isentropic surfaces on days 5, 10, 15, 20, 25, and 30 after the release of the backward trajectories. The values are binned every 1° in longitude and 0.5° in latitude. The geographical locations of the origins of the 30 d backward trajectories are denoted with red triangles.
The pathway of the large-scale convection associated with the ASM that transport surface anthropogenic pollutants to the ATAL has been verified by Yu et al. (2017). Meanwhile, any aerosol or its precursor that are transported to or produced in the vertical range of the horizontal trajectories by any mechanism have the potential to join the ATAL, including the anthropogenic sources, carbonaceous, sulfates, secondary organics, and nitrate aerosol etc. Our current in situ observations have shown that the size of most particles in the UTLS is relatively small (<0.25 μm). Further direct measurements of the chemical compositions of the ATAL, which is also our main objective in future observations, will resolve the above disagreement.

5. Summary and outlook

Our present understanding of the ATAL is largely based on satellite retrievals and model simulations. Direct observations are important but rare because of the challenging geographical location of the TP. To conduct in situ observations of the ATAL over the TP, a POPS aboard a stratospheric balloon was launched over GLM during the ASM period in 2018.

The measurements showed a robust aerosol layer in the UTLS at approximately 14–18 km, i.e. ±2 km around the tropopause at 16 km. The maximum aerosol number density and aerosol mass concentration were 35 cm$^{-3}$ and 0.15 μg m$^{-3}$, respectively. Aerosols at the observed UTLS aerosol layer existed most frequently in the five smallest particle size bins: 0.140–0.157, 0.157–0.176, 0.176–0.198, 0.198–0.222, and 0.222–0.249 μm, accounting for 98% of all particle numbers. The number density was at a maximum (10.6 cm$^{-3}$) for aerosols within 0.140–0.157 μm and decreased greatly as the particle size increased. The backward trajectories showed that the air parcels can arrive at the observed aerosol layer by two separate pathways. One pathway is the uplift below the 360 K isentropic surface south of the Himalayas, in which air parcels are first elevated to the upper troposphere and then are joined the anticyclonic circulation before going to the observation location. Air parcels can also accumulate in the ASM anticyclone by quasi-horizontal transport along the anticyclonic circulation, approximately between the 360 and 420 K isentropic surfaces.

A similar observational mode as that in the GLM campaign is planned. Various instruments, such as a...
radiometer (shortwave, longwave, and UV), will be deployed on schedule to fly within the ATAL for a long period. The horizontal level is vital for the accurate radiation observation, which kept well during the 2018 flight. Moreover, spatial and temporal variations of aerosol chemical compositions in ATAL are also expected, provided that the instruments under development are available during the next campaign. Studies based on these comprehensive observations and model simulations are likely to further improve our understanding of the properties of the ATAL and its radiative impacts in the UTLS over the TP during the ASM period. These studies may also expand our knowledge of the mechanism of the stratosphere and troposphere exchange over the TP and reduce the uncertainties in the estimates of the regional weather and climate.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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