Aerosol variations in the upper troposphere and lower stratosphere over the Tibetan Plateau

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Abstract
Satellite measurements and model simulations indicate the existence of the Asian tropopause aerosol layer (ATAL)—an enhanced aerosol layer in the upper troposphere and lower stratosphere (UTLS) associated with the Asian summer monsoon (ASM)—although it has rarely been evidenced by snapshots of balloon-borne in situ measurements. To better understand how the ATAL evolves, a portable optical particle counter (POPS) onboard a stratospheric balloon was released over the Tibetan Plateau (TP) during the ASM period of 2019. The POPS detected the ATAL in the UTLS during the ascending, descending periods, as well as during its quasi-horizontal floating periods. The aerosol number density in the ATAL showed obvious vertical variability. The peak aerosol number density in the ATAL was 180 cm$^{-3}$ around the tropopause during the ascending and descending period and the maximum aerosol number density was 290 cm$^{-3}$ around the tropopause during the floating period. And the aerosol concentration observed over the TP in the 2019 summer was approximately five times larger than that in the 2018 summer. Lagrangian simulations reveal that the minority of the observed aerosol particles were directly elevated in a region of uplift south of the Himalayas, and the majority of the particles were transported from the UTLS region situated approximately between the isentropic surfaces of 370 and 460 K. Up to 14% of the observed aerosol particles were directly influenced by the volcanic plumes from the eruption of the volcano Raikoke in June 2019.

1. Introduction
Previous studies indicated that, by creating a dome or ‘bubble’ of tropospheric air above the zonal mean tropopause, the Asian summer monsoon (ASM) may effectively transport tropospheric air into the stratosphere (Randel and Park 2006, Park et al 2007, Pan et al 2016). More importantly, the ASM coupled with deep convection-induced uplift may serve as an efficient chimney-like mechanism to vent tropospheric aerosols up into the upper troposphere and lower stratosphere (UTLS), from which these aerosols can further spread throughout the lower stratosphere of the entire Northern Hemisphere and even influence global climate change (Li et al 2005, Randel et al 2010, Vernier et al 2015, Yu et al 2015, 2017). As evidence of this hypothesis, a layer of enhanced aerosol scattering associated with the ASM referred to as the Asian tropopause aerosol layer (ATAL) has been observed by the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) (Vernier et al 2011) and by the Stratospheric Aerosol and Gas Experiment II (SAGE II) (Thomason and Vernier 2013). The ATAL generally occurs between June and August within approximately 0–160°E and 15–45°N and extends vertically from ~13 to 18 km (Vernier et al 2015). The ATAL has been reported by very few balloon-born in situ measurements. For example, (Yu et al 2017) confirmed the existence of an aerosol layer extending up to 2 km above the tropopause over the

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Yunnan-Guizhou Plateau. Recently, an ATAL was also reported over the northern edge of the TP (Zhang et al. 2019).

Previous studies have indeed provided valuable information on the ATAL from different perspectives. However, many complicated transport pathways to the ASM region, including regional-scale rapid deep convection, large-scale slow upward motions and circulations, and multiscale exchanges between the troposphere and stratosphere, are all very likely ways in which aerosols or their precursors are vented into the UTLS (Randel et al. 2010, Pan et al. 2016, Yu et al. 2017). These intricate mechanisms imply that the formation process, chemical composition, and physical and optical properties of the ATAL are highly complex and vary (Yu et al. 2015, Gu et al. 2016, Höpfner et al. 2019, Ma et al. 2019). Operational balloon observations can provide snapshots of the ATAL during its ascent but are not able to record how the ATAL evolves. Based on this key issue, a portable optical particle counter (POPS) onboard a stratospheric balloon was released atop the Qaidam TP (QDM; 37.74°N, 95.34°E; 3188 m above sea level; as shown in figure 1) on 16 August 2019. The POPS can measure particles between approximately 120 and 3400 nm in diameter using single-particle light scattering. The particle size is calculated from the pulse signal caused by each particle passing the laser beam. The instrument can provide the aerosol particle number density with a sufficiently low detection limit to capture over 60% of the ATAL particle surface area (Yu et al. 2017).

To understand the size distributions, in this study, the aerosol particles are divided into 15 size bins according to diameter boundaries of 123, 133, 145, 159, 176, 198, 225, 418, 500, 620, 930, 1274, 1591, 2067, 2649 and 3413 nm.

The remainder of this paper is organized as follows: section 2 briefly describes the observation data and the model used in this study. The main results are presented in section 3, followed by the discussion and conclusion in section 4.

2. Data and model

2.1. In situ aerosol measurements

As one of the most important in situ instruments, a POPS (Gao et al. 2016) onboard a stratospheric balloon was released atop the Qaidam TP (QDM; 37.74°N, 95.34°E; 3188 m above sea level; as shown in figure 1) on 16 August 2019. The POPS can measure particles between approximately 120 and 3400 nm in diameter using single-particle light scattering. The particle size is calculated from the pulse signal caused by each particle passing the laser beam. The instrument can provide the aerosol particle number density with a sufficiently low detection limit to capture over 60% of the ATAL particle surface area (Yu et al. 2017). To understand the size distributions, in this study, the aerosol particles are divided into 15 size bins according to diameter boundaries of 123, 133, 145, 159, 176, 198, 225, 418, 500, 620, 930, 1274, 1591, 2067, 2649 and 3413 nm.

2.2. The MPTRAC model

We use the Massive-Parallel Trajectory Calculations (MPTRAC) Lagrangian particle dispersion model
Hoffmann et al (2016) to conduct backward trajectory simulations for the in situ particle observations and to conduct forward trajectory simulations for volcanic SO$_2$ plumes in this study. In the MPTRAC model, trajectories of air parcels are calculated based on numerical integration using wind fields from global meteorological reanalyses (Hoffmann et al. 2016, Rößler et al. 2018). Diffusion is modeled by uncorrelated Gaussian random displacements of the air parcels with zero mean and standard deviations $\sigma_x = \sqrt{D_x \Delta t}$ (horizontally) and $\sigma_z = \sqrt{D_z \Delta t}$ (vertically). $D_x$ and $D_z$ are the horizontal and vertical diffusivities respectively, and $\Delta t$ is the time step for the trajectory calculations. $D_x$ and $D_z$ were set to 50 m$^2$ s$^{-1}$ and 0 m$^2$ s$^{-1}$ in the troposphere and 0 m$^2$ s$^{-1}$ and 0.1 m$^2$ s$^{-1}$ in the stratosphere, respectively. In addition, sub-grid scale wind fluctuations, which are particularly important for long-range simulations, are simulated by a Markov model (Stohl et al. 2005, Hoffmann et al. 2016).

To simulate the loss processes of chemical species, SO$_2$ in case of volcanic eruptions, is simulated based on exponential decay of the mass assigned to each air parcel. The decay time of SO$_2$ varies from hours to days depending on the ambient conditions of the SO$_2$, e.g. the humidity and the concentration of OH radicals (Khokhar et al. 2005, Theys et al. 2013). After sensitivity tests, a constant half lifetime of seven days is assigned to simulate the decay of volcanic SO$_2$ emission in the UTLS region. In this study, the MPTRAC model is driven with the ERA-Interim data (Dee et al. 2011) interpolated on a $1\degree \times 1\degree$ horizontal grid on 60 model levels with the vertical range extending from the surface to 0.1 hPa. The ERA-Interim data are provided at 00, 06, 12, and 18 UTC. Outputs of the model simulations are given at a 3-hour interval.

### 3. Results

#### 3.1. Meteorological background conditions

To understand the large-scale synoptic circulations and potential aerosol transport during the flight of the stratospheric balloon, figure 2 shows the meteorological conditions on 16–17 August based on ERA5 reanalysis data. The thick black dashed contour in figure 2(a) indicates the geopotential height (GPH) of 14 320 m at the 150 hPa pressure level, which is commonly used to approximately denote the boundary of the ASM anticyclone (W J and Park 2006). On 16–17 August, as shown in figure 2, the observation site is located at the northern edge of the ASM and at the south of the subtropical jet core. The tropopause (as defined by WMO 1957) above the site is at approximately 16.5 km. Double tropopauses are observed north of the subtropical jet; these double tropopauses are usually considered an active air exchange and mixing region in the UTLS (e.g. Peevey et al. 2012). The ASM anticyclone is characterized by large dynamic variability (Garny and Randel 2013) and frequent shedding eddies (e.g. Vogel et al. 2014), as well as a horizontal transport barrier for the air masses inside and outside of the anticyclone (Ploeger et al. 2015). The measurements at this location may show the characteristics of particles under complicated transport conditions.

#### 3.2. Observational profile of the POPS

The stratospheric balloon used herein can regulate flight within the altitude layers of interest; for example, the observation unit can float within the UTLS to provide continuous measurements of the ATAL. Moreover, by controlling flight at different altitude levels where the wind direction switches between west and east, we may endeavor to achieve long-term flight within a limited and permitted airspace, as

![Figure 2. Meteorological conditions (based on ERA5 reanalysis data) averaged between 06:00 on 16 August and 14:00 on 17 August. (a) shows the zonal wind (shaded) and wind vectors at a pressure level of 150 hPa. The location of the observation site is denoted with a red triangle. The thick black dashed contour indicates the GPH of 14 320 m, and the thin gray solid contours indicate the GPHs from 14 360 to 14 480 m with an interval of 40 m. (b) shows a vertical section of the zonal wind (shaded) and contours of the potential temperature from 340 to 440 K (black solid lines) and the potential vorticity from 2 to 6 PVU (black dashed lines, 1 PVU = $10^{-6}$ K m$^2$ s$^{-1}$ kg$^{-1}$) along 95.25° E. The blue and red dots indicate the first and secondary thermal tropopauses, respectively, as defined by (WMO 1957). The red triangle indicates the latitude and altitude of the observation site, and the vertical black dashed line marks the latitude (37.74 °N).](image-url)
Figure 3. Temporal evolution of the POPS-observed aerosol number density (blue line) and balloon flight altitude (red line) during five phases (I–V, outlined by rectangles in different colors): surface measurements before flight (I), ascending flight measurements (II), level flight measurements at ∼21 km (III), hovering flight measurements within the UTLS (IV), and descending flight measurements (V).

shown by the flight path surrounding QDM through both night and day (figure 1). The relatively flat terrain of the airspace is also conducive to successful payload recovery.

The whole observation profiles can be divided into five phases: surface measurements before the flight, ascending flight measurements, level flight measurements at approximately 21 km, hovering flight within the UTLS, and descending flight measurements (figure 3). The surface aerosol number density varied between 280 and 350 cm$^{-3}$ during 04:40–06:42 on 16 August, when the POPS was ready for flight; note that Beijing time is used unless otherwise specified. The instrument was then released by a stratospheric balloon and provided aerosol measurements for ∼70 min during its ascent to 21 km with a mean velocity of ∼4 m s$^{-1}$. A peak aerosol concentration was observed in the UTLS during the ascending period (region II in figure 3). Several peaks occurred during the ascending period; two peaks were particularly evident. The first peak value was close to 300 cm$^{-3}$ in the middle troposphere (6–7 km). The second peak indicated the ATAL in the UTLS exhibited a strong variability in the vertical direction; the number density of the aerosol particles increased greatly from 30 cm$^{-3}$ at 15 km and reached a maximum of 180 cm$^{-3}$ around the tropopause, and from the tropopause upward, an obvious decrease in the aerosol particles number density occurred.

3.3. Characteristics of aerosol distributions in the ATAL

To clearly see the aerosol distribution within the UTLS in detail, figure 4(a) regionally magnifies figure 3 and shows the vertical aerosol variations during the ascending and descending periods. A similar thermal tropopause, i.e. 16.8 and 16.7 km, was detected by radiosonde measurements during the ascending and descending periods, respectively. The aerosol concentration decreased sharply as the balloon ascended (figure 4(a)). Several peaks occurred during the ascending period; two peaks were particularly evident. The first peak value was close to 300 cm$^{-3}$ in the middle troposphere (6–7 km). The second peak indicated the ATAL in the UTLS exhibited a strong variability in the vertical direction; the number density of the aerosol particles increased greatly from 30 cm$^{-3}$ at 15 km and reached a maximum of 180 cm$^{-3}$ around the tropopause, and from the tropopause upward, an obvious decrease in the aerosol particles number density occurred.

This ATAL feature was similarly observed by the POPS descending measurements. (Zhang et al 2019) reported the highest concentration of ∼35 cm$^{-3}$ for aerosol particles between 140 and 3000 nm in the ATAL on 16 August, the same day as in this study.
but in 2018. Although both measurements were conducted during the mature ASM period, the peak particle number in the ATAL was nearly five times greater in 2019 than in 2018 despite the slight difference between the particle detection ranges, i.e. 140–3000 nm in 2018 and 120–3400 nm in 2019. This interannual variation strongly implies that the transport pathway, formation mechanism, and accumulating particle concentration of the ATAL are highly complex and variable.

Figure 4(b) shows the aerosol number density during the hovering flight within the UTLS. The aerosol concentration increased sharply from 15 to 200 cm\(^{-3}\) as the balloon descended from 20 km to 16.5 km. During 00:00–01:30 on 17 August, the balloon performed the quasi-horizontal flight at approximately 16.2 km where most of the number density distributed between 250 and 300 cm\(^{-3}\), was detected. Compared with the peak aerosol number (180 cm\(^{-3}\)) in the ATAL during the ascending period (figure 4(a)), the POPS frequently detected > 100 cm\(^{-3}\) more aerosols during the quasi-horizontal flight (figure 4(b)). More specifically, the maximum aerosol number density was 290 cm\(^{-3}\) at 16.2 km during the quasi-horizontal flight, which was approximately twice as large as that at the same altitude during the ascending and descending periods.

To understand the aerosol size distribution, the vertical distributions of aerosol particles in 15 size bins during the ascending period are shown in figure 5. From the surface to the middle troposphere (8 km), aerosols were mostly observed in the first seven smallest particle size bins (figure 5(a)). Similar to figure 4(a), relatively high concentrations of aerosols with a diameter < 159 nm were occasionally noted between 8 and 15 km. Concentrated aerosols with a diameter < 418 nm were observed within ±1 km of the tropopause, which suggested the widespread presence of fine-mode particles in the ATAL. To closely examine the distributions of aerosols that occurred most frequently, vertical profiles of the aerosol particles in the first seven smallest bins are further displayed in figure 5(b). Below 8 km, the aerosol number decreased greatly as the particle size increased. By contrast, the concentration was similar among the first three aerosol bins in the ATAL with peak magnitudes of ~35 cm\(^{-3}\) synchronously occurring around the tropopause. Then, the concentration decreased greatly as the particle size increased from bins 4 to 7 with successive peak magnitudes of 25, 15, 10, and 5 cm\(^{-3}\). The maximum number of aerosols with diameters of 140–250 nm was ~34 cm\(^{-3}\) in the ATAL during 2018 (Zhang et al 2019); ~2.5 times more aerosols were detected in this study. In addition, many more aerosol particles of diameter > 250 nm occurred in 2019 than in 2018. The ratio of the average aerosol number density in the lower troposphere (between 4 and 8 km) and around the UTLS (between 16 and 19 km) for the smallest and the largest particle sizes in figure 5(b) were 23.4% and 4.1%, respectively.

The vertical occurrence frequency of the cumulative particle number in bins 1–7 with respect to the total particle number in bins 1–15 within ±2 km relative to the tropopause is shown in figure 5(c). The percentage was >99% between −1 and 2 km; this percentage was larger by ~1% than that below −1 km. This result indicates that partly due to the effect of gravity, more aerosol particles with diameters > 418 nm were uplifted and accumulated.
Figure 5. (a) Vertical distributions of the aerosol number density in 15 particle size bins during the ascending period; the color bar represents the values of the aerosol number density. (b) Vertical profiles of the aerosol number density for the first seven particle size bins occurring most frequently. (c) Vertical occurrence frequency of the cumulative aerosol particle number in bins 1–7 with respect to the total particle number in bins 1–15; altitudes ± 2 km relative to the tropopause are shown. (d) Occurrence frequency of aerosols in 15 bins within ± 2 km relative to the tropopause during the ascending and descending periods (blue bars) and the hovering flight period within the UTLS (red bars). The horizontal dashed lines in (a) and (b) represent the thermal tropopause calculated by sounding measurements.

at lower altitudes than at higher altitudes in the ATAL. Additionally, larger particles also tend to be activated in cloud updrafts and can be removed more efficiently by precipitation. The statistics of the aerosol occurrence frequency within ± 2 km relative to the tropopause are shown for all 15 bins during the ascending and descending periods and the hovering flight period within the UTLS in figure 5(d). The maximum occurrence frequency was ∼23% for aerosols of diameter < 133 nm; a distinct decreasing pattern was then revealed as the aerosol particle size increased. Most of the aerosols in the ATAL had diameters of < 418 nm, accounting for ∼99% of all particles detected. At a more detailed level, a larger percentage of aerosol particles > 176 nm (bins 5–15) was observed during the hovering flight period within the UTLS than during the ascending and descending periods, which might be explained as follows. Compared to the altitudes during the ascending and descending periods, most measurements during the hovering flight period were collected at a lower altitude (approximately 16.2 km), resulting in the detection of more aerosol particles with large diameters because of the effect of gravity and the other factors mentioned above.

3.4. Source regions of the POPs-observed aerosol particles in the ATAL
To investigate the possible source regions of the POPs-observed particles in the ATAL, in the first step, we retrieve the 3D locations, i.e. the longitudes, latitudes, and altitudes of the observations with particle number densities > 50 cm⁻³ between the altitudes of 15 and 19 km, assign one air parcel to each of these locations to represent the aerosol particles, and then trace these air parcels backward for approximately 55 days (from the observation time backward to 18 UTC on 21 June 2019) using the MPTRAC model. There are in total 836 air parcels released in this step. For the sake of clarity, figures 6(a) and (b) respectively show the uniform selections of one-half of the trajectories originating from the boundary layer, and
Figure 6. Latitude versus the potential temperature (unit: K) of the 50 d backward trajectories colored according to the day reversed from 16 August 2019. (a) Shows the backward trajectories that originated from the surface, while (b) shows the backward trajectories that terminated at the observation site via quasi-horizontal transport. The latitudes and potential temperatures of the initial air parcels of the backward trajectories are denoted by black dots, and the latitude of the observation site (QDM) is denoted by the gray dashed line.

1/30 of trajectories transported from the UTLS till the end of the simulation, colored by days reversed from the observation time. During the simulation time period, a minority of the trajectories were elevated in the region of uplift south of the Himalayas approximately 20 to 35 d before the observations were conducted. There was no particle observed during our campaign coming from the boundary layer after 21 UTC on 26 July 2019 (approximately 20 d before the observations). The majority of the observed particles were directly transported from the UTLS region situated approximately between the isentropic surfaces of 370 and 460 K, which indicated that the aerosols transported to, or directly formed in the UTLS region between the isentropic surfaces of 370 and 460 K had a large impact on the aerosol observed during the campaign. These two separate transport pathways are generally consistent with those revealed by the trajectory simulations in (Zhang et al. 2019). However, the highest concentration of aerosol particles in the UTLS observed in this study is approximately five times as large as that described in (Zhang et al. 2019). Excluding the influences of the transport pathways, other factors, e.g. differences in the observation site locations, local meteorological conditions, and variations in the occurrence of deep convections, should be further considered.

To analyze the possible source regions in the UTLS for the majority of observed particles that are transported through the quasi-horizontal pathway from a statistical perspective, an ensemble of 100 000 air parcels was assigned to the locations of the selected observations mentioned above. In this step, we made the aerosol particles evenly distributed in all air parcels. We used 100 000 air parcels to wrap all the aerosols, so at the 3D locations where observations show larger particle number density, we assign more air parcels there, i.e. the number of air parcels assigned to each of the 3D locations of the observations was linearly proportional to the particle number density measurements there. Then, the backward trajectories initialized by the 100 000 air parcels were traced backward for the same time period of 55 d. ‘Snapshots’ of the proportion of air parcels between the isentropic surfaces of 370 and 460 K every 10 d are shown in the left panels of
Figure 7. Left panels: percentage of air parcels in proportional to total air parcels for the backward trajectories initialized by the in situ measurements. Right panels: same as left panels but for the forward trajectories initialized by the volcanic SO$_2$ emissions. The results are binned every 2$^\circ$ longitude and 1$^\circ$ latitude and only for the air parcels between the isentropic surfaces of 370 and 460 K. The figures are shown at 00 UTC on each date (please refer to the title of each figure for the date). The thick black contours denote the 14320 m geopotential height on the 150 hPa pressure surface. The black triangle denotes the location where the observation campaign was conducted, and the red triangle denotes the location of the Raikoke volcano.
Because the location of the observation site was at the boundary of the ASM, in most cases, the source region of the observed particles was inside the ASM anticyclone. However, because of the dynamical variabilities of the ASM (Ploeger et al. 2015), the observation site was sometimes exposed to the air outside of the ASM. Thus, part of the particles also originated from regions other than inside of the ASM.

3.5. Influence of a volcanic eruption on the observed aerosols in the UTLS

On 21 June 2019, the Raikoke volcano erupted and injected approximately 1.45 Tg of SO$_2$ into the atmosphere (Global Volcanism Program 2019); the majority of SO$_2$ was ejected to the UTLS at altitudes of approximately 13 km, and the maximum plume altitude was approximately 16 km (Global Volcanism Program 2019). Usually, the SO$_2$ in a volcanic plume will transform into sulfate aerosols in a matter of hours to days (Khokhar et al. 2005, Theys et al. 2013). Therefore, the eastward transport of a volcanic plume rich in SO$_2$ will probably influence the aerosol loading in the mid-latitudes, including the UTLS over the TP. Using SO$_2$ observations from the Atmospheric Infrared Sounder (AIRS) and a backward trajectory approach using the MPTRAC model, we retrieved the SO$_2$ emissions of the Raikoke volcanic eruption and traced the SO$_2$ plume forward to investigate its dispersion and possible influence on the composition of the observed aerosols during the QDM campaign. Please refer to appendix 1 for the details about the inverse modeling method of the volcanic sulfur emission and the validation. This emission retrieval approach also has been explained and tested in detail in a variety of studies regarding the transport of volcanic plumes (e.g. Hoffmann et al. 2016, Wu et al. 2017, 2018).

The left panels and right panels of figure 7 show the movements of air parcels for the backward trajectories initialized by our observations and the forward trajectories initialized by the volcanic sulfur emissions, respectively. In figure 7, we first divide the globe into bins of $2^\circ$ in longitude and $1^\circ$ in latitude, and then at a 3 h time interval during the 55 d trajectory simulation, we calculate the percentage of the air parcels between the isentropic surfaces of 370 and 460 K in each bin in proportional to the total number of air parcels. The same is done both for the backward trajectories initialized by our observations (on the left panels) and the forward trajectories initialized by the volcanic sulfur emissions (on the right panels). In this way, we aim to show the movement of the air parcels with time from a statistical perspective. As shown in the right panels of figure 7, after the eruption, the volcanic plume was transported eastward and diffused throughout the Northern Hemisphere; the distribution was largely influenced by the transport barrier of the subtropical jet and the ASM and the majority of the volcanic plumes are confined outside of the ASM.

The volcanic plume may have had an effect on the observed aerosols by directly dispersing over the observation site and influencing the composition of the air through which the instrument passed. Alternatively, the plume may have influenced the composition of the air upwind of the observation site; i.e. the air parcels may have been contaminated before being transported to the location of the instrument. To roughly estimate the proportion of the observed aerosol particles that may have been influenced by the volcanic plume, we selected the bins in the left panels that are overlapped with the bins in the right panels of figure 7 at a time interval of 3 h during the 55 d simulations, and then added up the percentage of the selected bins for the backward trajectories. It was calculated that there were up to 14% of the released air parcels influenced by the Raikoke volcanic eruption. As described in section 3.4, we assigned a total number of 100 000 air parcels to represent the total observed aerosol particles, and the number of air parcels assigned to each location of the observations was linearly proportional to the particle number density measured there, i.e. the aerosol particles were evenly distributed in all air parcels. So we estimate that up to 14% of the observed particles may have been influenced by the Raikoke volcanic eruption.

Please note that while the MPTRAC model can estimate the conversion of SO$_2$ into sulfate aerosols during the transport process, it cannot resolve the chemical processes of aerosol coagulation and deposition. However, we can assume that the minuscule sulfate aerosols that formed in the volcanic plume remained in the plume in the mid- and high-latitude upper troposphere and stratosphere, where the atmosphere is relatively dry and clean compared with the lower troposphere, and these sulfate aerosols had a reduced possibility of interacting with clouds or of being washed out. Therefore, the estimation of the proportion of observed particles influenced by the Raikoke volcanic plume is limited to the UTLS region, and the influence is only from the sulfur in the volcanic plume, whereas other particles in the volcanic plume, e.g. volcanic ash, are not included.

4. Discussion and conclusions

By using measurements from a POPS onboard a stratospheric balloon and model simulations, this study presented the detailed properties of the ATAL over the TP during the ASM period in 2019. The main conclusions are summarized as follows.

The ATAL with a thickness of about 4 km was detected by the POPS, and the ATAL exhibited large variations in the vertical direction as shown by the POPS measurements during the ascending and descending periods; The aerosol particles density in the ATAL increased up to a maxima of 180 cm$^{-3}$ around the tropopause and decreased from the tropopause upwards. The maximum aerosol number was
290 cm$^{-3}$ at 16.2 km during the quasi-horizontal flight, which was approximately twice as large as that at the same altitude during the ascending and descending periods. During the quasi-horizontal flight of the balloon, the maximum aerosol number was 250–300 cm$^{-3}$ at night, which was $\sim$100 cm$^{-3}$ greater than that around the tropopause during the daytime. Compared with observations acquired in the 2018 summer, a much higher aerosol density was detected in the ATAL in the 2019 summer, although both experiments were conducted during the mature period of the ASM, with similar transport pathways and source regions of the observed aerosol particles.

Based on the backward-trajectory simulation, we found that a minority of the aerosol particles observed in the tropopause layer were elevated in the region of uplift south of the Himalayas approximately 20 to 35 d before the observations were conducted. And the majority of the observed particles were from the UTLS region approximately between the isentropic surfaces of 370 and 460 K during the 55-day backward trajectory simulation. However, it is challenging to define all the direct origins of the aerosol between the 370 and 460 K. The aerosols in the UTLS may be directly transported from the boundary layer by the convections associated with the ASM, formed out of precursors during the vertical transport process (e.g. Park et al 2007, Randel et al 2010, Yu et al 2017), formed from the sulfur dioxide out of volcanic eruptions (Vernier et al 2011, Hoffmann et al 2016, Wu et al 2017), or elevated from the upper troposphere via the ASM spiral (Ma et al 2019, Vogel et al 2019, Zhang et al 2019). Our results show that any possible mechanism that contributes to the Northern Hemisphere UTLS aerosols may have an influence on the source and composition of the ATAL.

Under the influence of the Raikoke volcanic eruption in June 2019, we estimated that up to 14% of the observed particles were contaminated by the volcanic plume. The percentage that 14% of the observed aerosol particles influenced by the Raikoke eruption seems small considering the large aerosol loading in the UTLS contributed by the volcanic eruption, but actually reasonable. Because the locations where the observations were made were often inside of the ASM boundary when the volcanic plume dispersed to the north of the ASM. The ASM boundary usually acts as a slightly leaky transport barrier in the upper troposphere that isolates the air inside of the ASM from that outside of the ASM (e.g. Ploeger et al 2015, Wu et al 2017), and this barrier has protected the very thin ATAL from being totally concealed by the volcanic aerosol plume and made it distinguishable by the POPS. However, if a volcanic eruption injected aerosol to the UTLS inside of the ASM, it would be impossible to separate the ATAL from the thick volcanic aerosol layer created by, for instance, the Nabro eruption in 2011 (Vernier et al 2015).

The ATAL is originally defined as a thin layer of aerosol that regularly appears during the ASM season. And it is probably formed by the mechanism that the convections associated with the ASM elevating the surface pollutions up to the height of the Asian tropopause every summer (Vernier et al 2011, 2015, Yu et al 2017). While volcanic eruptions are not considered as the regular source of the ATAL and also the contribution of volcanic aerosols may not explain the large increase in the particle concentration observed in 2019 than that observed in 2018 (Zhang et al 2019). By depicting the detailed spatial variations in the aerosol distribution in the ATAL, this study is expected to assist future research on the composition and formation mechanism of the ATAL and to benefit the evaluation and parameterization of aerosol model simulations. Moreover, experiments with POPS instruments onboard a stratospheric balloon and a latex balloon will be synchronously conducted at 5–7 representative regions over the TP to comprehensively explore the properties of the ATAL during various periods of the ASM.

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Appendix 1:

The inverse modeling of the volcanic SO$_2$ emission and simulation of the transport of volcanic plumes

1. The AIRS SO$_2$ measurements

AIRS (Aumann et al 2003) is an infrared nadir sounder with across-track scanning capabilities aboard the National Aeronautics and Space Administration’s (NASA’s) Aqua satellite. Aqua was launched in 2002 and operates in a nearly polar Sun-synchronous orbit at about 710 km with a period of 98 min. AIRS provides nearly continuous measurement coverage with 14.5 orbits per day and with a swath width of 1780 km and it covers the globe almost twice a day. The AIRS footprint size is...
13.5 km × 13.5 km at nadir and 41 km × 21.4 km for the outermost scan angles respectively. The along-track distance between two adjacent scans is 18 km. The AIRS observations provide good horizontal resolution and make it ideal for observing the fine filamentary structures of volcanic SO$_2$ plumes.

In this study, we use an optimized SO$_2$ index (SI, unit: K) to estimate the amount of SO$_2$ injected into the atmosphere by the Raikoke eruption 2019. The SI is defined as the brightness temperature differences in the 7.3 µm SO$_2$ waveband:

$$SI = BT(1412.87 \text{ cm}^{-1}) - BT(1371.52 \text{ cm}^{-1})$$  \hspace{1cm} (1)

where BT is the brightness temperature. This SI is more sensitive to low concentrations and performs better on suppressing background interfering signals than the SI provided in the AIRS operational data products. It is an improvement of the SI definition given by (Hoffmann et al 2014) by means of a better choice of the background channel (selecting 1412.87 cm$^{-1}$ rather than 1407.2 cm$^{-1}$). The SI increases with increasing SO$_2$ column density and it is most sensitive to SO$_2$ in the upper troposphere and stratosphere. SO$_2$ injections into the lower troposphere are usually not detectable in the infrared spectral region because the atmosphere gets opaque due to the water vapor continuum. A detection threshold of 3 K was used in this study to identify the Raikoke SO$_2$ injections. AIRS detected the Raikoke SO$_2$ cloud from its initial eruption on 22 June 2019.

2. Reconstruction of the Raikoke SO$_2$ emission time series

To reconstruct the altitude-resolved SO$_2$ emission time series, we follow the approach of (Hoffmann et al 2016) and use backward trajectories and AIRS SO$_2$ measurements. Since AIRS measurements do not provide altitude information, we establish a column of air parcels at each location of an AIRS SO$_2$ detection. The vertical range of the column is 0–25 km, covering the possible vertical dispersion range of the SO$_2$ plume in the first few days. The AIRS footprint size varies between 14 and 41 km, so in the horizontal direction, we choose an average of 30 km as the full width at half maximum (FWHM) for the Gaussian scatter of the air parcels. In our simulation, a fixed number of 100 000 air parcels is assigned to all air columns and the number of air parcels in each column is linearly proportional to the SO$_2$ index. Then backward trajectories were calculated for all air parcels, and trajectories that are at least 2 d but no more than 5 d long and that have passed the volcano domain are recorded as emissions of Raikoke. The volcano domain is defined to be within a radius of 75 km to the location of the Raikoke and 0–20 km in the vertical direction, covering all possible injection heights. Sensitivity experiments have been conducted to optimize these pre-assigned parameters to obtain the best simulation results.

Based on satellite measurements, the (Global Volcanism Program 2019) estimated that about 1.45 Tg of SO$_2$ were injected into the UTLS during the explosive eruption period. So a total mass of 1.45 Tg is assigned to the 100 000 air parcels that represent the SO$_2$ plume. To simulate the loss processes of chemical species, SO$_2$ in our case is simulated based on the exponential decay of the SO$_2$ mass assigned to each air parcel with a constant half lifetime of seven days. 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. The ERA-Interim data are provided at 00, 06, 12, and 18 UTC. Outputs of the model simulations are given at a 3 h interval.

Figure A1 shows the distribution of the SO$_2$ emission within the 75 km radius to the location of
Raikoke with time and altitude, which is considered as the volcanic SO$_2$ emission of Raikoke in 2019. During its explosive eruption period, the Raikoke released SO$_2$ almost without interruption in about 30 h, but with large variations in height and magnitude. The initial eruption began at about 18:00 UTC on 21 June 2019 followed by continuous eruptions on 22 June, ranging from 4 km to about 16 km. The tropopause at the location of Raikoke on 21 and 22 June 2019 is at about 9 km, so the majority of SO$_2$ was injected directly into the mid-latitude lower stratosphere, and the largest SO$_2$ injection occurred around 13 km. This timeline of the eruptions and the plume heights are consistent with the observation and analysis reported by the (Global Volcanism Program 2019). The derived SO$_2$ emission time series are the basis of the simulations of SO$_2$ plume in the following forward simulations of the volcanic SO$_2$ plumes.

3. Simulation and validation of the Raikoke plume dispersion

The Raikoke volcano erupted and injected approximately 1.45 Tg of SO$_2$ into the atmosphere. A new set of 100 000 air parcels is assigned to the derived SO$_2$ emission shown in figure A1, with 14 500 kg of SO$_2$ for each of the air parcels. The trajectories initialized with this SO$_2$ emission time series are calculated with the MPTRAC model from 18:00 UTC 12 June 2019 (first eruption) to 00UTC 16
August 2019 (when the in situ observation campaign was made).

In order to validate the simulation, and also to indirectly validate the reconstructed emission time series, we compare the simulations of plume dispersion with AIRS SO₂ measurements in figure A2. The simulated evolution of the SO₂ plume is shown in figure A2 (left column) and compared with the AIRS SO₂ measurements 6 h before and after the model output time (right column). The simulation outputs are given every 3 h, but only results at selected time and dates are shown. In order to make a direct comparison of the simulations and the AIRS measurements, the SO₂ index from AIRS detections was converted into SO₂ column density using the correlation function described in (Hoffmann et al 2014), which was obtained by radiative transfer calculations. Only SO₂ column density values larger than 3 Dobson units (DU) are shown in figure A2.

Generally, the simulations agree well with the AIRS measurements in position and diffusion and the simulations can provide more information on the SO₂ distribution than the AIRS measurements alone. The differences between the SO₂ plumes are partly attributed to a mismatch in time of the AIRS SO₂ measurements and the simulation outputs. In magnitude, the SO₂ column density from AIRS is slightly smaller than that from MPTRAC simulations, and the SO₂ maxima are found in slightly different locations. This was also found by (Hoffmann et al 2016) and (Wu et al 2017) for other eruption events and this was attributed to the fact that the inverse modeling approach is optimized to reproduce the spatial extent of the plume but not the absolute emissions. Except for the discrepancy between the times of different data, the remaining difference may also be attributed to the initial setting of the total SO₂ mass, the SO₂ lifetime and the uncertainties of the ERA-Interim winds.

The SO₂ in the volcanic plume was converted into sulfate aerosol during while it disperses, so the SO₂ column density decreases gradually with time. In this study, we assume that in the UTLS region between the isentropic surfaces of 370 K and 460 K, the sulfur in the form of sulfate aerosol remains in the volcanic plumes that represented by the air parcels released. This assumption is tested to be practical in the UTLS region. In the mid-latitude UTLS, the atmosphere is relatively dry and clean compared with the lower troposphere, so the sulfate aerosols may have a much smaller chance to interact with clouds or to be washed out. The sulfate aerosols in the plume could also be affected by other mechanisms that speed up the loss of sulfur, for example, coagulation in the volcanic plume, the absorption of sulfur onto fine ash particles, etc. But for a moderate eruption, such as the Raikoke eruption, sulphuric particle growth may not be as significant as it is in a large volcanic eruption, so the scavenging efficiency of sulfur should be low.

Data availability

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

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References

Aumann H H et al 2003 AIRS/AMSU/HSB on the Aqua mission: design, science objectives, data products, and processing systems IEEE Trans. Geosci. Remote Sens. 41 253–64
Dee D P et al 2011 The ERA-Interim reanalysis: configuration and performance of the data assimilation system Q. J. R. Meteorol. Soc. 137 553–97
Gao R S et al 2016 Alight-weight, high-sensitivity particle spectrometer for PM2.5 aerosol measurements Aerosol Sci. Technol. 50 88–99
Garney H and Randel W J 2013 Dynamic variability of the Asian monsoon anticyclone observed in potential vorticity and correlations with tracer distributions J. Geophys. Res.-Atmos. 118 13421–33
Global Volcanism Program 2019 Report on Raikoke (Russia) eds Crafford and E Venzke Bulletin of the Global Volcanism Network 448
Gu Y, Liao H and Bian J 2016 Summertime nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer monsoon region Atmos. Chem. Phys. 16 6641–63
Hoffmann L, Griesbach S and C J M 2014 Volcanic emissions from AIRS observations: detection methods, case study, and statistical analysis Proc. SPIE 9242
Hoffmann L, Rößler T, Griesbach S, Heng Y and Stein O 2016 Lagrangian transport simulations of volcanic sulfur dioxide emissions: impact of meteorological data products J. Geophys. Res.-Atmos. 121 4651–73
Höpfner M et al 2019 Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons Nat. Geosci. 12 608–12
Khokhar M F, Frankenberg C, Van Roozendael M, Beirle S, Kühl S, Richter A, Platt U and Wagner T 2005 Satellite observations of atmospheric SO2 from volcanic eruptions during the time-period of 1996–2002 Adv Space Res 36 879–87
Li Q et al 2005 Convective outflow of South Asian pollution: a global CTM simulation compared with EOS MLS observations Geophys. Res. Lett. 32 L14826
Ma J et al 2019 Modeling the aerosol chemical composition of the tropopause over the Tibetan Plateau during the Asian summer monsoon Atmos. Chem. Phys. 19 11587–612
Pan L L et al 2016 Transport of chemical tracers from the boundary layer to stratosphere associated with the dynamics of the Asian summer monsoon J. Geophys. Res.-Atmos. 121 14159–74
Park M, Randel W J, Gettelman A, Massie S T and Jiang J H 2007 Transport above the Asian summer monsoon anticyclone inferred from Aura Microwave Limb Sounder tracers J. Geophys. Res.-Atmos. 112 D16309
Peevey T R, Gillett C J, Randall C E and Kunz A 2012 Investigation of double tropopause spatial and temporal global variability utilizing high resolution dynamics limb sounder temperature observations J. Geophys. Res.-Atmos. 117 D16105
Ploeger F et al 2015 A potential vorticity-based determination of the transport barrier in the Asian summer monsoon anticyclone Atmos. Chem. Phys. 15 13145–59
Randel W, Park M, Emmons L, Bernath P, Walker K A, Boone C and Pumphrey H 2010 Asian monsoon transport of pollution to the stratosphere Science 328 611–3
Rößler T, Stein O, Heng Y, Baumeister P and Hoffmann L 2018 Trajectory errors of different numerical integration schemes diagnosed with the MPTRAC advection module driven by ECMWF operational analyses Geosci. Model Dev. 11 575–92
Stohl A, Forster C, Frank A, Selbert P and Wotawa G 2005 Technical note: the Lagrangian particle dispersion model FLEXPART version 6.2 Atmos. Chem. Phys. 5 2461–74
Theys N et al 2013 Volcanic SO2 fluxes derived from satellite data: a survey using OMI, GOME-2, IASI and MODIS Atmos. Chem. Phys. 13 5945–88
Thomason L W and Vernier J P 2013 Improved SAGE II cloud/aerosol categorization and observations of the Asian tropopause aerosol layer: 1989–2005 Atmos. Chem. Phys. 13 4605–16
Vernier J P et al 2015 Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution J. Geophys. Res.: Atmos. 120 1608–19
Vernier J P, Thomason L W and Kar J 2011 CALIPSO detection of an Asian tropopause aerosol layer Geophys. Res. Lett. 38 L07804
Vogel B, Günther G, Müller R, Grooß J-U, Hoor P, Krämer M, Müller S, Zahn A and Riese M 2014 Fast transport from Southeast Asia boundary layer sources to northern Europe: rapid uplift in typhoons and eastward eddy shedding of the Asian monsoon anticyclone Atmos. Chem. Phys. 14 12745–62
Vogel B, Müller R, Günther G, Spang R, Hanumanthu S, Li D, Riese M and Stiller G P 2019 Lagrangian simulations of the transport of young air masses to the top of the Asian monsoon anticyclone and into the tropical pipe Atmos. Chem. Phys. 19 6007–34
W J R and Park M 2006 Deep convective influence on the Asian summer monsoon anticyclone and associated tracer variability observed with Atmospheric Infrared Sounder (AIRS) J. Geophys. Res.: Atmos. 111 D12314
WMO 1957 A three-dimensional science: second session of the Commission for Aerology WMO Bulletin 4 134–8
Wu X, Griessbach S and Hoffmann L 2017 Equatorward dispersion of a high-latitude volcanic plume and its relation to the Asian summer monsoon: a case study of the Sarychev eruption in 2009 Atmos. Chem. Phys. 17 13439–55
Wu X, Griessbach S and Hoffmann L 2018 Long-range transport of volcanic aerosol from the 2010 Merapi tropical eruption to Antarctica Atmos. Chem. Phys. 18 15859–77
Yu P et al 2017 Efficient transport of tropospheric aerosol into the stratosphere via the Asian summer monsoon anticyclone Proc. Natl Acad. Sci. USA 114 6972–7
Yu P, Toon O B, Bardeen C G, Mills M J, Fan T, English J M and Neely R R 2015 Evaluations of tropospheric aerosol properties simulated by the community earth system model with a sectional aerosol microphysics scheme J. Adv. Model Earth Syst. 7 865–914
Zhang J, Wu X, Liu S, Bai Z, Xia X, Chen B, Zong X and Bian J 2019 In situ measurements and backward-trajectory analysis of high-concentration, fine-mode aerosols in the UTLS over the Tibetan Plateau Environ. Res. Lett. 14 124068