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Key Points:
- A novel set of in-situ profile measurements of O₃, CO, CH₄, and particles from Tibetan Plateau during Asian summer monsoon are presented.
- Joint analyses of the profiles provide insights into transport processes controlling the northern edge of the Asian monsoon anticyclone.
- Observed CO profile maxima at 13–14 km (360–370 K) identify the level of convective transport at the upstream source regions.

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Abstract
We present data and analysis of a set of balloon-borne sounding profiles, which includes co-located O₃, CO, CH₄, and particles, over the northern Tibetan Plateau during an Asian summer monsoon (ASM) season. These novel measurements shed light on the ASM transport behavior near the northern edge of the anticyclone. Joint analyses of these species with the temperature and wind profiles and supported by back trajectory modeling identify three distinct transport processes that dominate the vertical chemical structure in the middle troposphere, upper troposphere (UT), and the tropopause region. The correlated changes in profile structures in the middle troposphere highlight the influence of the strong westerly jet. Elevated constituent concentrations in the UT identify the main level of convective transport at the upstream source regions. Observed higher altitude maxima for CH₄ characterize the airmasses’ continued ascent following convection. These data complement constituent observations from other parts of the ASM anticyclone.

Plain Language Summary
Asian summer monsoon deep convection transports surface pollutants to the stratosphere. Although satellite data have provided clear evidence of this transport, in situ measurements are critical for characterizing how monsoon is vertically re-distributing the regional emissions. We report new balloon-borne measurements over the Tibetan Plateau that provide a unique data set on the northern edge of the anticyclone, complementing other observations.

1. Introduction
The Asian Summer Monsoon (ASM) deep convection has been recognized as an important transport process for chemical composition in the UT and lower stratosphere (UTLS) and related climate impacts. The ASM convection generates a large anticyclone in the UTLS (Gill, 1980; Hoskins & Rodwell, 1995). Satellite data shows elevated mixing ratios within the UTLS anticyclone for most species with tropospheric sources (Park et al., 2007; Randel et al., 2010). Satellite data also show enhanced aerosols over the ASM, which has been referred to as the Asian Tropopause Aerosol Layer (ATAL; Vernier et al., 2011). The satellite observations, however, do not have sufficient vertical resolution and chemical composition details to characterize the chemical impacts of the transport process and the formation mechanisms of ATAL (Höpfner et al., 2019). Thus, in-situ measurements using airborne and balloon-borne platforms play an important role in obtaining necessary information for process studies.

In this letter, we present data and analysis of a balloon-borne campaign over the Tibetan Plateau (TP) in August 2020. The profile data we report have several unique characteristics. First, the location of the measurement is over the northern TP where no prior profile measurement of CO and CH₄ from the planetary boundary layer (PBL) to stratosphere are available. These data, therefore, provide complementary information to the airborne activities from other parts of the ASM system. In particular, the recent airborne campaigns, StratoClim (http://www.stratoclim.org/) project conducted high-altitude research flights over the southern flank of the ASM anticyclone (Bucci et al., 2020; Höpfner et al., 2019; Mahnke et al., 2021; von Hobe et al., 2021), and the ACCLIP campaign sampled the ASM air from the eastern edge of the anticyclone (Pan et al., 2022). These data also extend the existing sounding data from southern flank of the TP, which includes ozone, water vapor, particles, and clouds (Bian et al., 2012; Brunamonti et al., 2018; He et al., 2019; Ma et al., 2022; Yu et al., 2017). The measurements we...
present include co-located profiles of temperature, horizontal wind, O$_3$, CO, CH$_4$ mixing ratios and aerosol number concentrations from PBL to the lower stratosphere, which provide the complete vertical structure for characterizing transport behaviors at the northern edge of the ASM anticyclone. The suite of species includes two tropospheric trace gas species with different chemical lifetimes (CO and CH$_4$), and O$_3$ which is dominated by stratospheric sources. Together, they delineate the levels of convective transport and the transition from the fast convective transport to the large-scale dynamically driven slow ascent. Furthermore, the aerosol number concentration profiles near the tropopause layer provide additional information on the formation and maintenance of the ATAL.

The data analysis we present has three elements. First, the large-scale dynamical background for each selected sounding is provided for both the horizontal flow in the UTLS and vertical cross-section using reanalysis data. This step enables the identifications of the dynamical conditions responsible for correlated vertical structure changes in all profiles. The analysis of these joint changes in profile structures shed light on the dominant processes in the region's transport. Last, back-trajectories for air parcels in UT layers of enhanced CO and CH$_4$ are analyzed to investigate the influence of convection and the transport source regions. This analysis, using Lagrangian trajectory calculations, is supported by satellite data of convective cloud top and regional CO and CH$_4$ distributions for the measurement period. Together, these data and analyses provide a first chemical structure characterization of the northern edge of the anticyclone.

2. Campaign Information and Trajectory Analysis

2.1. AirCore Campaign

AirCore is a long thin coiled stainless-steel tube with one end opened and one end closed, which was flown by a weather-balloon from surface to around 30 km. It samples air passively during the descent (Karion et al., 2010). The air samples collected by AirCore are analyzed using Cavity-Ring Down Spectrometer gas analyzer (Picarro CRDS G2401). The precisions are 1 ppb for CH$_4$ and 15 ppb for CO, as indicated by one standard deviation of the measurements at 5 s at all flow rates between 40 and 300 sccm (standard cubic centimeters per minute). We apply the method presented in Membrive et al. (2017) to place the Picarro measurements on vertical coordinates. The configuration of the tube is one of the most important factors that determine the vertical resolution. Here, we use two connected tubes with different diameters to maximize the resolution in the UTLS (Membrive et al., 2017). The first section was a 150 m long tube with 0.3175 cm (1/8 inch) outer diameter; the second section was a 50 m long tube with 0.635 cm (1/4 inch) outer diameter. With this configuration, the resolutions of the AirCore measurement are 100–400 m in the UTLS layer (10–20 km), and 200 m to 1 km in the free troposphere (3–10 km). More details of the AirCore prototype can be found in Yi et al. (2020).

The AirCore campaigns have been conducted for multiple monsoon seasons. In 2018, the AirCore system was first tested in Inner Mongolia (Yi et al., 2019). During 2019 and 2020, we deployed the AirCore system at Dachadam (DCD) (37.8 °N, 95.3 °E, elevation 3,174 m). This campaign location was selected with a strong interest in ASM transport, as well as logistic feasibility and the synergy with the activities associated with the established DCD observatory by Institute of Atmospheric Physics (IAP). As a result, Printed Optical Particle Spectrometer (POPS) and IAP-ozonesonde were co-released in addition to the AirCore. POPS can measure high-quality in situ profile of aerosol particle number density and size distribution between 140 and 3,000 nm diameter (Gao et al., 2016) and the results from the sensor's previous measurements over the TP are reported in Yu et al. (2017) and Zhang et al. (2019, 2020). The IAP-ozonesonde is developed at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS) (Zhang et al., 2014). Meteorological variables are measured by iMet radiosondes from International Met Systems (www.intermetsystems.com). Overall, every flight provides observations of O$_3$, CO, CH$_4$, particle concentrations, pressure, temperature, and wind from PBL to the lower stratosphere. In this work, we present data and analysis for four flights during the 2020 campaign.

2.2. Trajectory Calculations and Supporting Satellite Data

In support of the data analysis, back trajectory calculations are performed using the trajectory module of the three-dimensional Lagrangian chemistry transport model (CLaMS) as described in Li et al. (2020). The model run is driven by the European Center of Medium-Range Weather Forecasts (ECMWF) next-generation reanalysis (ERA5, Hersbach et al., 2020). The vertical transport scheme uses the diabatic vertical velocity derived from total heating rates. ERA5 diabatic run has been evaluated for good performance in the simulation of airmass
transportation in the UTLS region (Bucci et al., 2020). The ERA5 data used are in 1° × 1° grid, hourly, on 137 hybrid levels from the surface to 0.01 hPa, which have 400–500 m vertical resolution in the UTLS region. Here, 40-day backward trajectories are calculated for parcels initialized at each sampling altitude point and time. The vertical sampling points are separated by approximately 20 m on average.

Additional supporting information used are deep convective cloud products from Himawari 8 satellite (K. Bessho et al., 2016). Specifically, we used the data product of Cloud Top Height for the deep convective clouds in conjunction with the back trajectory analysis to identify the location of convective transport that influenced the profiles. The spatial and temporal resolution of the cloud product is 5 km and 10 min, respectively. The convective source is identified when the trajectory height is below the deep convective cloud top height. The co-location criteria used are 0.5° spatially and 1 hr temporally. We also use the averaged dry air column mixing ratio of CO (XCO) and CH₄ (XCH₄) from the TROPOspheric Monitoring Instrument (TROPOMI; Borsdorff et al., 2023; Lorente et al., 2020) satellite data as a supporting reference for the regional tropospheric CO and CH₄ distributions during the campaign period.

3. Results

3.1. Dynamical Background of the Soundings

In this work, we present the analysis of four profiles measured on 8th, 15th, 18th, and 23rd of August 2020. Using ERA5 reanalysis data, we show synoptic conditions for these four selected flights in Figure 1, where the 150 hPa Geopotential Height (GPH) field is used to indicate the location and extent of the anticyclone, the potential vorticity (PV) field is shown to highlight the separation of stratosphere and troposphere at this level. The lapse rate tropopause and the static stability are shown in the latitude-height cross section along the launch site longitude (95.3°E), which also included the zonal wind for jet stream location, isentropes, and selected PV contours (2, 4 PVU) to mark the dynamical tropopause. Both the map view and the cross-section view indicate that the balloon launches at DCD were located at the northeastern edge of the ASM anticyclone. The maximum zonal wind in the jet core locates the northern boundary of the anticyclone (Pan et al., 2016). In most cases, the upper tropospheric air mass in the launch location was inside the ASM anticyclone, equatorward of the core of subtropical jet (shown by 30 m/s wind contour).

On August 18th, the westerly jet was discontinuous in the vicinity of DCD and the high-PV airmass at the jet exit influenced soundings from DCD. This influence is shown as a tropopause-fold along the 340 K potential temperature (Θ) level (Figure 1). Although this process was the largest on the eighteenth, the stratospheric influence was shown as a “tongue” of enhanced static stability in every profile at 340 K level.

3.2. Chemical Structures of the Anticyclone Northern Edge

In this section, we present the profiles of CO, CH₄, particle number concentration, ozone mixing ratio, temperature, Θ profiles and wind fields at DCD (Figure 2). Combining the background dynamics information (Figure 1), we examine the correlated and uncorrelated structures in the profiles.

The first level of correlated changes involving all profiles is identified in the mid-troposphere (~ 9 km or 340 K). The dynamics behind this structure are the upper-level frontal zone and the associated tropopause fold below the jet core (Keyser & Shapiro, 1986), which occurs near the location of DCD (Figure 1). The influence of the frontal zone showed as a small inversion layer in the temperature profile, also small increases of ozone and static stability (indicated by the larger Θ gradient). The 340 K level also marks the sharp increase in the wind profiles, indicating the strong westerly associated with the jet core. Much larger changes are shown as the sharp decrease in three tropospheric species in the immediate layer above 340 K. The CO mixing ratios decreased from approximately 100 ppb to around 60 ppb, and CH₄ follows a similar pattern. The sharp changes of the tropospheric species indicate the influence of transport associated with the jet stream for these species, that is, the change from that dominated by local emissions to the influences from the upstream regions.

The second level of correlated changes in profiles is identified in the UT starting 13 km, where the large enhancement of CO, CH₄, and particles are observed, with August 18th as an exception. Although the three species show loosely correlated maxima in the UT indicating the influence of convectively lofted PBL pollution, their emission sources are quite different (e.g., Baker et al., 2012). With the relative short lifetime of 1–2 months (Duncan et al., 2007; Pan et al., 2022), CO is considered a good marker for convective transport. The CO maxima
near 13–14 km (360–370 K θ) indicate the level of convective outflow. Above the altitude, CO gradually decreases with altitude and reaches its typical stratospheric value around 20 km, 420–430 K in θ. The O₃ mixing ratios are less than 100 ppb below the level of CO enhancement. They begin to increase with the altitude above 13–14 km and show a rapid increase once reached the tropopause. The behavior of these two trace species is consistent with that described in von Hobe et al. (2021) in StratoClim data analysis. There, the decrease of CO and
the increase of O₃ above convective outflow was identified as the photo-chemical reaction of the airmass during the slow ascent following the rapid convective transport within the confinement of the anticyclone. Although the StratoClim measurements were around the southern flank of the anticyclone and our sounding profiles were measured near the northern edge, the behavior of these two trace species appears to be consistent. The wind profile data (Figure 2) indicate that the sounding location is under strong influence of the westerly jet in the UT between 9 and 17 km. The strong gradient in the zonal wind serves as the northern edge the anticyclone and forms a confinement layer in the UT up to the tropopause. The confinement of the anticyclone has been examined previously using idealized tracers (Randel & Park, 2006), and the strongest confinement was identified to be between 200 and 100 hPa (11–16 km).

In this layer above the convective outflow, the CH₄ and particle profiles are not correlated with CO. While the CO mixing ratio decreases and the O₃ mixing ratio increases above 13–14 km, CH₄ remains elevated, shows additional maxima 2 km above the lower peak values, and decreases sharply above the tropopause. The layer of elevated CH₄ in the UT in all three cases (with exception of 08–18) is approximately 4 km deep between the level of convective outflow (13 km) to the tropopause (17 km). This vertical structure of CH₄ is consistent with its much longer chemical lifetime (8–10 years; Lelieveld et al., 2016), which allows the continued ascent of elevated CH₄ following the convective outflow. Based on the modeling study of Vogel et al. (2019), the airmass’ slow ascent in an upward spiraling way over the Asian monsoon convection from 360 to 460 K has a rate of 1–1.5 K/day. Applying the rate here, the elevated CH₄ layer located between 360 and 390 K corresponds to a transit time scale of one month. The longer lifetime also makes it possible for the CH₄ profile observations to include contributions of multiple convection events and sources. Further examination of this process will be shown using trajectory model in Section 3.3.

The particle profiles give the clearest indication that the air mass in the confinement layer is non-local. As marked in Figure 2, there is a sharp change at the bottom of the JetStream where the local PBL airmass meets the much cleaner airmass from the frontal zone. In the UT, the particle profiles show a broad layer of elevated concentrations with a multi-maxima structure, similar to that in CH₄ profiles. The correlated change in the vertical structure between the particle and CH₄ profiles suggests the contributions of multiple convective events. The broader layer structure is consistent with the understanding that this layer of aerosols, as part of ATAL, is dominated by the formation of secondary aerosols. This point was the highlight of several studies in StratoClim data analysis. In particular, Höppner et al. (2019) reported the observed connection between the ammonium nitrate aerosol layer and the substantial amount of ammonia transported from surface into the UT. Appel et al. (2022) shows that the ATAL originates from the conversion of inorganic and organic gas-phase precursors, rather than from the uplift of primary particles from below. Our observations of the elevated particle concentration extending to 3 km above the level of convective outflow and up to the tropopause level support the concept that the particles continue to form and grow within the confinement of the ASM anticyclone, following the rapid convective transport of precursor gases to the UT.

Finally, changes are observed in all profiles across the tropopause, consistent with the transition from the UT to the lower stratosphere. In particular, the changes in the wind profiles, both the wind speed and wind directions,
Figure 3. Back trajectory analysis of convective transport sources. The regional CO (a) and CH$_4$ (d) distributions are shown as monthly mean of column average mixing ratios (XCO and XCH$_4$) using TROPOMI data. White space on the map are missing data. (b) Map of the back-trajectories calculated for the measurement points near the CO maximum on 2020-08-15, colored by trajectory transit time (see Figure 2e). Black crosses (●) indicate the locations of convective events that influenced the sounding airmasses, and the green star shows the measurement location. The same analyses for 2020-08-08 and 2020-08-23 are also shown in gray, with pink crosses marking identified convective influence. (c) The air parcels back-trajectories as in (b) in $\theta$ versus transit time space and colored by longitude. (e) and (f) are the same as in (b) and (c), respectively, but for CH$_4$ on 2020-08-23.

marks the tropopause level as the top of confinement in this location. This is consistent with the rapid increase of O$_3$ and decrease of CH$_4$ across the tropopause.

Note that the profile from August 18th 2020 has distinct structure among the four soundings, with weaker maxima in CO, CH$_4$ and particles at upper levels (Figure 2e). UTLS back trajectories for this profile do not follow the anticyclonic flow but originate farther to the west (not shown here), and hence the composition reflects impacts from airmasses outside of the anticyclone.

3.3. Back Trajectory Analysis of Convective Transport Source Region for CO and CH$_4$

In this section, we investigate the convective influence observed in the sounding profiles and the source regions of the elevated upper tropospheric CO and CH$_4$ (Figure 2). These elevated layers were measured under the conditions of strong westerly winds (Figure 2), which supports the hypothesis that the elevated pollutants are not from local sources but resulted from horizontal transport following convective uplifting events upstream. We verify the hypothesis using back trajectory analysis to establish relationships between the observed layer of high CO or CH$_4$ and upstream deep convection events, which are identified using satellite data of convective cloud top height (Section 2.2).

Specifically, 40-day back trajectories initialized at sampling points in a selected layer near the maximum CO (CH$_4$) are analyzed to identify the space-time co-location of the air parcels' back trajectories with the convection events. The contributing convection events are identified when the co-located air parcel's trajectories are found below the satellite data based convective cloud top. The results shown in Figure 3 are primarily focused on the elevated CO layer on 2020-08-15 and the elevated CH$_4$ layer measured on 2020-08-23. To give a perspective of
regional CO and CH₄ background in August 2020, we have also included monthly mean CO and CH₄ distributions in the region derived from TROPOMI satellite data column average mixing ratios.

The 2020-08-15 CO profile is highlighted, because the CO maximum mixing ratio in that profile is the highest among the four cases. Using CO > 105.6 ppbv (97% of maximum) as the criterion, a total of 29 trajectories are initialized in the layer 13.3 0.2 km. Among which 17 trajectories identified convective influence. The analysis indicates that the convective events contributing to the upper tropospheric CO maximum are along the east coast of China (Figure 3b), which is part of a high tropospheric CO region in the monthly mean XCO map from TROPOMI (Figure 3a). These column average mixing ratios are strongly weighted by the contribution of the lower troposphere (Borsdorff et al., 2023). The trajectory map highlights that the transport pathway in the UT from the top of the convection was dominated by the anticyclonic flow. The sounding sampled convective transport event approximately 1 week before from the east coast of China (Figure 3b). This transport pathway is quasi-isentropic with a weak ascending motion. The 0 change of 2 K in the 7 days indicates that the convective outflow (>360 K) is above the level of zero radiative heating (Fueglistaller et al., 2009; von Hobe et al., 2021). Although not the focus of our analysis, we have included the back trajectories for the air parcels in the near maximum layer in the map for the 2020-08-15 and 2020-08-23 soundings (Figure 3b). In these two cases, the source regions are near the southeastern edge of the TP and the head of Bay of Bengal. The transport following the convection events are similar in the three cases with nearly one loop around the anticyclone.

The back trajectory analysis for the 2020-08-23 CH₄ profile maximum is shown in Figures 3e and 3f. Using CH₄ > 1,995.2 ppbv as the criterion (99% of the maximum), 38 trajectories are initialized in the layer 15.3 0.5 km. Among which, 31 identified convective influence. The analysis reveals two major clusters of deep convection events for the maximum CH₄ layer: one cluster is near the southeastern edge of the TP with around 10 days in back trajectory transit time, the other is over southern Pakistan with 16 days of back trajectory transit time. The transport followed the anticyclonic flow for approximately two loops. These convection events were deeper than the CO events, at near 365 370 K 0. The back trajectories show strong ascending motion in the 2 weeks after convection before the sounding measurements, with 1 K/day ascent rate. This strong ascent is broadly consistent with higher radiative heating rates in this region and 0 range (von Hobe et al., 2021). Both clusters of convective transport regions are associated with high tropospheric CH₄ from the TROPOMI monthly mean. The primary source region with high XCH₄ mixing ratio observed in southern Pakistan is consistent with more than 2 ppmv of CH₄ in the sounding maximum.

The similarity and differences between the CO and CH₄ cases provide several insights into the transport behavior within the ASM anticyclone. The slow ascending motion in an anticyclonic pattern following rapid convective lifting demonstrate the two-step transport process in the ASM (Vogel et al., 2019). The large difference in their lifetimes explains the deeper layer of elevated CH₄ while the structure of short-lived CO serves to mark the level of convective outflow. The analysis also shows that the day-to-day variability of the anticyclone bringing convectively lofted PBL air into the confinement from a large region, from the east coast of China to the southern Pakistan in these examples.

4. Conclusions and Discussions

High vertical resolution profile measurements of CO, CH₄ mixing ratios, using the AirCore technique, along with co-located ozonesonde and POPS measurements from the northern TP during August 2020 are presented. These co-located profiles from surface to 25 km provide rich information for characterizing the role of large-scale circulation and the ASM convection in controlling the vertical chemical structure near the northern edge of the anticyclone. Our analysis of the co-located profiles with the support of dynamical background from reanalysis data and back trajectory modeling lead to a set of key findings. They are summarized below.

1. The atmosphere vertical structure from surface of the TP (3 km above sea level) to the lower stratosphere are largely controlled by the ASM anticyclone. The chemical structure is divided into three layers. The local PBL influence is largely terminated at the bottom of the anticyclone (340 K) where the air mass has different degrees of stratospheric influence. The vertical structure in the anticyclone confinement layer (9–17 km) is dominated by a layer of elevated PBL pollutants in the UT (maxima 13–15 km). The layer is not contributed by the local PBL but from upstream convective transport sources. The top of the confinement layer is
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Data Availability Statement

Data is available at Cai (2024).

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