

Lidar-observed enhancement of aerosols in UTLS over the Tibetan Plateau during the Asian summer monsoon

Q. S. He¹, C. C. Li², J. Z. Ma³, H. Q. Wang⁴, X. L. Yan³, Z. R. Liang¹, and G. M. Qi⁵

¹Shanghai Meteorological Service, Shanghai, China

²Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing, China

³Chinese Academy of Meteorological Sciences, Beijing, China

⁴College of Environmental Science and Engineering, Donghua University, Shanghai, China

⁵Germu Meteorological Bureau, Qinghai, China

Correspondence to: C. Li (ccli@pku.edu.cn)

Abstract.

Vertical profiles of aerosol extinction coefficients were measured by an Micro Pulse Lidar at Naqu (31.5°N, 92.1°E, 4508m a.m.s.l.), a meteorological station located on the central part of the Tibetan Plateau during summer 2011. Observations show a persistent maximum in aerosol extinction coefficients in the upper troposphere–lower stratosphere (UTLS). These aerosol layers were generally located at an altitude of 18–19 km a.m.s.l., 1–2 km higher than the tropopause, with broad layer depth ranging approximately 3–4 km and scattering ratio of 4–9. The aerosol layers in UTLS wore off gradually with the reducing intensity of the Asian monsoon over the Tibetan Plateau at the end of August. Aerosols in UTLS over the plateau appear to be influenced by Nabro volcano eruption on 13 June 2011. Variations in these aerosols are also found to be closely related to the intensity of underlying deep convection, indicating that deep convection plays an important role in the accumulation of aerosols in UTLS over the Tibetan Plateau.

1 Introduction

Aerosols in the upper troposphere–lower stratosphere (UTLS) play an important role in the global/regional climate system and the geochemical cycle (Hanson et al.,

1 1994; Borrmann et al., 1997; Solomon et al., 1997). They also influence atmospheric
2 ozone budgets through providing surface areas for efficient heterogeneous reactions
3 (Keim et al., 1996; Solomon, 1999). Using the Stratospheric Aerosol and Gas
4 Experiment II(SAGE II) data, Li et al. (2001) found that aerosol concentrations near
5 100 hPa are higher over the Tibetan Plateau than over China's central and northern
6 regions in summer. Recent observations by balloon-borne optical particle counter
7 (Tobo et al.,2007) and aircraft-borne measurements (Keim et al., 1996; Solomon,
8 1997) showed that soot-containing liquid aerosols with the major components of fine
9 particles may also affect the aerosol layer near the tropopause. Appearance of cold
10 tropopause in the upper troposphere (possibly in the lower stratosphere also) has been
11 considered as an important factor to explain the enhancement of tropopause aerosols
12 observed in summer over the Plateau (Kim et al., 2003). This observational fact is
13 important from the point view of heterogeneous reactions on aerosol surfaces since
14 gas-to-particle conversion processes are generally more active in low temperature.
15 During summer, the elevated surface heating and rising air associated with persistent
16 deep convection over the Tibetan Plateau leads to anticyclonic circulation and
17 divergence in the UTLS(Yanai et al., 1992; Hoskins and Rodwell, 1995; Highwood
18 and Hoskins, 1998), where persistently enhanced pollutants such as aerosols, CO,
19 methane and nitrogen oxides, as well as water vapor, can be linked to the rapid
20 vertical transport of surface air from Asia, India and Indonesia in deep convection and
21 confinement by strong anticyclonic circulation (Rosenlof et al., 1997; Jackson et al.,
22 1998; Dethof et al., 1999; Park et al., 2004; Filipiak et al., 2005; Li et al., 2005a; Fu et
23 al., 2006).

24 Volcanic eruption, though as occasional event, can inject amounts of ash and
25 sulfur dioxide (SO₂) into the stratosphere, and the injected SO₂ was oxidized to
26 sulfuric acid particles through homogeneous nucleation (Wu et al., 1994). The Nabro
27 stratovolcano in Eritrea, northeastern Africa, erupted on 13 June 2011, injecting
28 approximately 1.3 teragrams of SO₂ to altitudes of 9 to 14 kilometers in the upper
29 troposphere, which resulted in a large aerosol enhancement in the
30 stratosphere(Bourassa et al., 2012). This event has been observed by lidar networks

1 such as EARLINET, MPLNET and NDACC with independent lidar groups and
2 satellite Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations
3 (CALIPSO) to track the evolution of the stratospheric aerosol layer in various parts of
4 the globe (Uchino et al., 2012; Sawamura et al., 2013), and other instruments, such as
5 the Infrared Atmospheric Sounding Interferometer (Clarisse et al., 2013) and the
6 ground-based spectrometry of twilight sky brightness (Tukiainen et al., 2013).
7 Bourassa et al. (2012) found that the aerosol enhancement built while remaining
8 confined for several weeks to the region between central Asia and the Middle East
9 after eruption of Nabro volcano using the limb scanning Optical Spectrograph and
10 Infra-Red Imaging System (OSIRIS) satellite instrument.

11 The aerosols from Nabro eruption might overlap with the background tropopause
12 aerosols in summer, changing their properties and evolution in UTLS over the
13 Plateau. A clarification of the mechanisms that aerosols transport into and disperse
14 out of the UTLS over the Plateau is an important step toward understanding
15 tropospheric influences on hydration and chemical composition in the global
16 stratosphere. Knowing the height dependence of the aerosol changes is important for
17 understanding the mechanisms responsible for the transport of aerosols from the
18 troposphere to the stratosphere over the Tibetan Plateau; however, a variety of aerosol
19 vertical distributions and optical properties over the Tibetan Plateau has not been
20 assessed in a satisfactory manner due to lack of continuous direct observations.

21 The vertical distributions of aerosol extinction coefficients were measured over
22 the Tibetan Plateau in the summer of 2011, as part of the project “Tibetan Ozone,
23 Aerosol and Radiation” (TOAR). In this study, the lidar and radiosonde measurement
24 results are presented and compared with satellite data. We find a persistent maximum
25 in aerosol extinction coefficients in the UTLS within the anticyclone, and show that
26 such aerosol accumulation can be linked to the eruption of Nabro volcano and the
27 development of the Tibetan deep convective systems. These results indicate that
28 volcanic aerosol dispersed with the weakening of Tibetan anticyclonic circulation and
29 deep convection could primarily affect aerosol and hence radiation properties near the
30 tropopause over the Tibetan Plateau.

1

2 **2 Measurements and Data**

3 **2.1 Micro Pulse Lidar**

4 An Micro Pulse Lidar (MPL-4B, Sigma Space Corp., USA) was operated at the
5 Naqu Meteorological Bureau (31.5 °N, 92.1 °E, 4508m a.m.s.l.) on the central part of
6 the Tibetan Plateau. The MPL is a backscatter lidar which uses an Nd:YLF laser with
7 an output power of 12 μJ at 532nm and 2500 Hz repetition rate. The diameter of the
8 receiving telescope is 20 cm, and the field of view is 0.1 mrad. The vertical resolution
9 of the lidar observation is 30m, and the integration time is 30 s. Data obtained on the
10 cloud-free days during nighttime were selected in order to avoid the disturbance of
11 cloud and/or rain to column-averaged lidar ratio and solar noise.

12 In general, the inversion of the LIDAR profile is based on the solution of the
13 single scattering LIDAR equation:

$$14 \quad P(r) = O_c(r)CE \frac{\beta(r)}{r^2} \exp[-2 \int_0^r \sigma(z)dz] \quad (1)$$

15 where r is the range, C is the LIDAR constant, which incorporates the transmission
16 and the detection efficiency, and E is the laser pulse energy. $\beta(r)$ represents the total
17 backscattering coefficient $\beta(r) = \beta_m(r) + \beta_a(r)$, $\sigma(r)$ is the total extinction coefficient
18 $\sigma(r) = \sigma_m(r) + \sigma_a(r)$, $\beta_a(r)$ and $\sigma_a(r)$ are aerosol backscattering and extinction
19 coefficients, respectively. $\beta_m(r)$ and $\sigma_m(r)$ are molecular contributions to the
20 backscattering and the extinction coefficients, respectively. They can be evaluated by
21 the Rayleigh-scattering theory from the Standard Atmosphere 1976 (NASA, 1976).
22 But here the molecular extinction coefficients are evaluated using temperature and
23 pressure from the radiosondes released at the lidar field site twice a day. $O_c(r)$ is the
24 overlap correction as a function of the range caused by field-of-view conflicts in the
25 transceiver system. Systematic errors of $P(r)$ were mainly observed in the lowest
26 altitudes where an incomplete overlap between the emitted laser beam and the
27 telescope field-of-view can led to an underestimation of aerosol backscatter and
28 extinction coefficients. Since the majority of aerosols are contained in the first several
29 kilometers of the atmosphere, the overlap problem must be solved. Overlap is

1 typically solved experimentally, using techniques outlined by Campbell et al. (2002).
2 The starting point is an averaged data sample where the system is pointed horizontally
3 with no obscuration. By choosing a time when the atmosphere is well mixed, such as
4 late afternoon, or, even better, when the aerosol loading is low, backscattering through
5 the layer is roughly assumed to be constant with range (i.e., the target layer is
6 assumed to be homogeneous). The similar overlap calibration was carried out at the
7 beginning of this field experiment.

8 The vertical profile of aerosol extinction coefficient σ_a is determined by a near
9 end approach in solving the lidar equation as proposed by Fernald (1984). Considered
10 the period of TOAR campaign was only two months after eruption of Nabro volcano,
11 volcanic aerosols were still freshly nucleated particles with small size. The lidar ratios
12 should therefore feature rather high (Müller et al., 2007). Sawamura et al. (2013)
13 employed the mean lidar ratio value of 50 sr at 532 nm for most groups of global lidar
14 networks to trace the evolution of the stratospheric aerosol layer from Nabro volcano
15 eruption. Therefore, a column averaged lidar ratio of 50 sr is assumed for all
16 measurement examples in this study.

17 We identify the boundaries of aerosol layer in the UTLS from the lidar extinction
18 coefficient profiles. The lowest bin with $\sigma_a=0.002 \text{ km}^{-1}$ above 18 km is identified as
19 the top of aerosol layer H_t and the bin with minimum value of σ_s between 10 km and
20 16 km as the layer base H_b . The visible optical depth of the aerosol layer is derived by
21 integrating the values of σ_a between H_b and H_t .

22 **2.2 Radiosonde Observations**

23 During the field campaigns, 76 L-band (GTS1) electronic radiosondes (Nanjing
24 Bridge Machinery Co., Ltd., China) were launched to provide vertical profiles of
25 pressure, temperature, and humidity up to 25 km to 30 km high. The radiosondes were
26 released at the lidar field site in Naqu twice a day at 0000 and 1200 UTC.

27 Eleven weather balloons with Vaisala RS92 radiosondes (Vömel et al., 2007)
28 have been launched to provide profiles of air temperature, relative humidity RH, wind
29 speed and wind direction usually up to the mid stratosphere. The RH can be measured
30 between 0 and 100% with a resolution of 1% and an accuracy of 5% at $-50 \text{ }^\circ\text{C}$

1 (Miloshevich et al., 2006; W ährn et al., 2004). While Miloshevich et al. (2009) found
2 that the RH measured by RS92 has a moist bias in the lower stratosphere (LS) and a
3 dry bias in the upper troposphere (UT). The Cryogenic Frostpoint Hygrometer (CFH)
4 is a lightweight (400 g) microprocessor-controlled instrument and operates on the
5 chilled-mirror principle using a cryogenic liquid as cooling agent. It includes several
6 improvements over the similar NOAA/CMD instrument. It is currently designed to be
7 combined with ozone sondes to provide simultaneous profiles of water vapor and
8 ozone(V ömel et al., 2007). CFH has been taken in many inter comparison experiment
9 as an absolute reference for water vapor measurements, including the validation of
10 Aura MLS water vapor products. We compared RS92 RH measurements with
11 simultaneous water vapor measurements from CFH on 13 August 2011. After
12 applying the time-lag and solar radiation bias corrections, corrected RS92 RH
13 measurements show agreement with CFH in the troposphere. The mean difference
14 between corrected RS92 RH measurements and CFH is a dry bias of 2.9% in the
15 ground layer, while the mean differences in 5-10 km, 10-15 km and tropopause
16 transition layer region are 1%, 0.6% and 1.4% moist bias, respectively. Therefore, the
17 accuracy of corrected RS92 RH measurements is comparable to the accuracy of CFH
18 in the UTLS (Yan, 2012).

19 **2.3 Satellite Observations**

20 The Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard
21 CALIPSO (Winker et al.,2003), is used to characterize aerosol extinction profiles in
22 the UTLS, which is a three-channel (532 nm parallel, 532 nm perpendicular, 1064 nm)
23 elastic lidar receiving light at the same wavelength as the emitted laser frequency.
24 CALIOP sends short and intense pulses (1064 and 532 nm) of linearly polarized laser
25 light downward towards the Earth. The atmospheric backscatter profile is retrieved at
26 60 m vertical resolution from 8–20 km with a horizontal resolution of 1 km. The
27 Level 2 aerosol extinctions at 532nm of CALIOP (version 3.0) were used to compared
28 with the ground based MPL on the Plateau, which can be obtained at
29 http://www-calipso.larc.nasa.gov/tools/data_avail/. CALIOP data are selected over a
30 300 km x 300 km square with a MPL location in its center.

1 National Oceanic and Atmospheric Administration (NOAA) satellites provide an
2 outgoing longwave radiation (OLR) product for the top of the atmosphere. OLR data
3 are calculated on a daily basis by the Climate Diagnostic Center (CDC), a division of
4 NOAA (Liebmann and Smith 1996). The horizontal resolution is 2.5° by 2.5° .
5 Missing values are computed by applying spatial and temporal interpolations. The
6 OLR in this data set is calculated by converting $10\ \mu\text{m}$ to $12\ \mu\text{m}$ channel radiances
7 measured by the Advanced High Resolution Radiometer aboard the NOAA
8 operational polar orbiting satellites. The daily mean is the average of one daytime and
9 one nighttime measurement. The OLR emitted by high, cold, deep convective clouds
10 is much lower than that by warm low clouds or by the Earth's surface. Usually, values
11 of less than $200\ \text{W m}^{-2}$ indicate deep convection (Fujiwara et al., 2009). Deep
12 convection, in turn, indicates the regions with extensive lifting of air that may act as
13 source regions for aerosol layer.

14 We used the water vapor profiles observations (version 3.3) from the Microwave
15 Limb Sounder (MLS) on the NASA Aura satellite (Waters et al., 2006). Aura MLS
16 measurements include water vapor, ozone and carbon monoxide that are useful tracers
17 of tropospheric and stratospheric air; these data have been used to document enhanced
18 levels of carbon monoxide in the upper troposphere over the Asian monsoon (Li et al.,
19 2005a; Filipiak et al., 2005) and also over the North American summer monsoon (Li
20 et al., 2005b).

21

22 **3 Results**

23 Fig.1 show the vertical profiles of aerosol Scattering Ratios (SR) measured at
24 Naqu during 6–26 August 2011, along with the daily mean profiles of temperature.
25 The measurements display relatively high aerosol extinction coefficients in the UTLS,
26 which are 4-9 factors higher than those at altitudes below and close to (even higher
27 than, such as on 6 and 12 August) molecular scattering coefficients at the same
28 altitude. Compared with SR profiles of Nabro volcanic aerosol from MPLNET,
29 EARLINET, NDACC and Hefei stations during June and July (Sawamura et al.,
30 2013), the maximum SR of aerosol layers in UTLS over the Tibetan Plateau are

1 similar to that over Universitat Politècnica de Catalunya, Barcelona, Spain (41.39 N,
2 2.11 E) as one of EARLINET stations but larger than the other observations. Table 1
3 listed some statistical parameters of aerosol layer over Tibet and Shanghai (31.23 N,
4 121.53 E) for the same period. The highest aerosol extinction coefficients in the
5 UTLS over the Tibetan Plateau generally located at 18-19 km altitudes, which are 1-2
6 km higher than the tropopause. Tropopause temperatures ranged from -70 °C to
7 -80 °C, and the height of the tropopause varied from 80 hPa to 100 hPa (from 17 km
8 to 18 km), during the observational period. Moreover, such relatively high aerosol
9 extinction coefficients could extend over broad layers, ranging approximately 3-4 km.

10 The CALIOP aerosol extinction coefficients are available over UTLS for 12, 13,
11 18 and 20 August. Fig.2 compares the average extinction coefficient profile of MPL
12 with that of CALIOP and shows a good agreement between the two instruments in
13 both aerosol layers altitude and the value of extinction coefficient. In particular, the
14 MPL profiles show less standard errors at each vertical resolution altitude possibly
15 due to the good signal-to-noise ratio of MPL observed at the high altitude of the lidar
16 station and clear atmospheric environment over the Tibetan Plateau.

17 According to the period of occurrence of aerosol layers in UTLS, the continuous
18 lidar observation can be split into two stages: 6 to 12 (S1) and 22 to 26 (S2) August
19 2011 for the continuous maintenance stages of aerosol layer. Between the two stages,
20 the existence of low clouds decayed the lidar signal to the extent that no available
21 aerosol layer observed in UTLS. Additionally, some cases with cirrus in upper
22 troposphere might increase the retrieval error of extinction coefficient of above
23 aerosol layer, which are also removed from the dataset. Fig.3. shows the daily
24 variation in plateau monsoon index (PMI) and the seven-day averaged PMI time
25 series from 1 July to 31 August 2011, with an overlap of cirrus occurrence (He et al.
26 2013) and aerosol optical depth (AOD) in UTLS. PMI is an indicator of the daily
27 mean intensity of the Tibetan Plateau monsoon. A larger PMI value indicates stronger
28 monsoon in summer, which can be determined as follows (Tang et al. 1984):

29
$$\text{PMI} = H_1 + H_2 + H_3 + H_4 - 4H_0 \quad (2)$$

30 where H is the daily deviation from the monthly mean geo-potential height at 600 hPa.

1 The subscript numbers 0 to 4 indicate the location of the Center (90 °E, 32.5 °N), West
2 (80 °E, 32.5 °N), South (90 °E, 25 °N), East (100 °E, 32.5 °N) and North (90 °E, 40 °N) of
3 the Plateau, respectively.

4 Many researchers have adopted the PMI to analyze the Tibetan Plateau monsoon
5 variation. It is concluded that the index can reasonably describe the main
6 characteristics of the Tibetan Plateau monsoon (e.g., Bai et al. 2001; Bai et al. 2005;
7 Xun et al. 2011). These two stages might be caused by the different circulation
8 systems due to an apparent time interval of about 10 days with PMI undergoing a
9 substantial oscillation. During the first stage (from 6 to 12 August 2011), when the
10 AOD decreased from 6 to 7 August and increased from 8 to 12 August, the values of
11 PMI experienced an increasing trend from -20 on 6 August to 63 on 12 August. The
12 values sharply decreased to below -40 in the second stage with the low and
13 continuous decreasing AOD over UTLS from 22 to 26 August 2011. Two obvious
14 features can be found in the temporal variation of AOD: (i) AOD showed a decreasing
15 trend accompanied by decreasing PMI during the campaign period, indicating that the
16 aerosol layers wore off gradually with the reducing intensity of the Asian monsoon
17 over the Tibetan Plateau at the end of August. Bourassa et al. (2012) found that the
18 strong Asian monsoon anticyclone, which existed from June through September over
19 Asia and the Middle East, where the Nabro volcanic aerosol was observed with
20 OSIRIS, and the enhanced aerosol dispersed and quickly circulated throughout the
21 Northern Hemisphere at the end of August, when the Asian monsoon anticyclone
22 began to decay. And (ii) when the intensity of the Tibetan Plateau monsoon
23 circulation subsided to PMI less than 0, the AOD in UTLS kept persistent decline
24 regardless of the variation trend of PMI, indicating that confinement of the air in the
25 lower stratosphere induced by Asian monsoon anticyclone were destroyed to benefit
26 the enhanced aerosol dispersing to the whole Northern Hemisphere.

27 In these cases, there were interesting temporal change in maximum extinction
28 coefficients of the aerosol layer and the tropopause temperatures, as shown in Fig.4.
29 The maximum extinction coefficients appear to be anti-correlated with the tropopause
30 temperatures. The aerosol extinction coefficient usually increases with decreasing

1 tropopause temperature along the time series. The maximum extinction coefficient of
2 these cases is 12.0 m^{-1} on 12 August, when the tropopause temperature is $-76.0 \text{ }^\circ\text{C}$,
3 being the lowest compared with the other cases. The minimum extinction coefficient
4 of these cases is 4.4 m^{-1} on 26 August, corresponding to the highest tropopause
5 temperature of $-72.3 \text{ }^\circ\text{C}$.

6 There are two possible ways for the decreasing tropopause temperatures to affect
7 the enhancement of high aerosol extinction. One way is the low temperatures due to
8 adiabatic cooling of ascending air parcels induced by deep convective activities,
9 which result in the direct transportation of natural and/or anthropogenic emissions
10 from the troposphere over the Tibetan Plateau. The decreasing temperature at the
11 tropopause is generally associated with the enhancement in deep convective activities
12 over the Tibetan Plateau. The low OLR has been treated as an indicator of the
13 organized deep convective activity in the troposphere (Fujiwara et al., 2009). Fig.5
14 compares nighttime mean maximum aerosol extinction coefficients in UTLS with the
15 OLR convection proxy over the Tibetan Plateau in August 2011. The enhancement of
16 aerosols near the tropopause appears to be well correlated with the changes in OLR(r^2
17 = 0.77) without evidence of substantial time lags. Between the two stages, the
18 existence of low clouds in those days might obstruct significantly adiabatic cooling of
19 ascending air parcels and vertical transportation of aerosols in the lower troposphere,
20 resulting in different variation of aerosol layer with intensity of deep convective
21 activity in the two continuous maintenance stages of aerosol layer. The relationships
22 of the aerosol extinction coefficient and OLR in two continuous stages are also shown
23 in Fig. 5 with correlation coefficient of 0.78 and 0.86 for S1 and S2, respectively,
24 suggesting the possibility that the tropopause aerosol enhancement associates with the
25 upward transport of aerosols in deep convective systems over the Tibetan Plateau.
26 The overshooting deep convection could directly influence the aerosol concentration
27 at this level. Using daily NCEP data and monthly SAGE data, Cong et al. (2001)
28 calculated the inter annual change of the aerosol and ozone in 100 hPa, and they
29 proposed that the atmospheric masses passing over the tropopause over the Tibet
30 Plateau and its neighboring areas might possibly carry the aerosol particles of middle

1 or lower troposphere into the vicinity of the tropopause and result in an increase of the
2 aerosol loading near the tropopause. Yin et al. (2012) used a cloud resolving model
3 coupled with a spectral bin microphysical scheme to investigate the effects of deep
4 convection on the concentration and size distribution of aerosol particles within the
5 upper troposphere, and found that aerosols originating from the boundary layer can be
6 more efficiently transported upward, as compared to those from the mid-troposphere,
7 due to significantly increased vertical velocity through the reinforced homogeneous
8 freezing of droplets. Bourassa et al. (2012) also found a large stratospheric optical
9 depth enhancement after the Nabro eruption over the Tibetan Plateau located on the
10 eastern side of the Asiatic monsoon circulation, where vertical transport to
11 stratospheric altitudes is particularly effective (Park et al., 2007).

12 The other way for the decreasing tropopause temperatures to affect the
13 enhancement of high aerosol extinction is the enlargement of pre-existing and/or
14 vertically transported aqueous solution droplets induced by adiabatic cooling and
15 hydration of the air associated with deep convection. It has been verified that deep
16 convection over the Tibetan Plateau is likely to be a primary pathway for water vapor
17 from the maritime boundary layer (e.g., Indian Ocean, South China Sea). Dessler and
18 Sherwood (2004) have also suggested that convective transport plays a key role for
19 the accumulation of water vapor near the tropopause, resulting in an increase of H₂O
20 mixing ratio by more than 5 ppmv near the tropopause (Gettelman et al., 2002; Park et
21 al., 2004; Fu et al., 2006). But Tobo et al. (2007) used a growth model to calculate the
22 possible growth under given atmospheric conditions assuming the existence of liquid
23 solutions at equilibrium with respect to H₂O, H₂SO₄ and HNO₃, and found that
24 aerosol growth is sensitive to H₂O mixing ratios. According to the calculated growth
25 curves of liquid solutions as a function of temperature and water vapor, the high H₂O
26 mixing ratios (more than 5 ppmv) are indispensable condition for producing high
27 concentrations of fine particles near the tropopause. In fact, the H₂O mixing ratios
28 near the tropopause and aerosol layer from Vaisala RS92 radiosondes released in 6, 8
29 11 and 23 August 2011 are not more than 2 ppmv, obviously less than the previous
30 observations, as shown in Fig. 6. In consequence, the effects of gas-to-particle

1 conversion from liquid solutions would likely be secondary to the enhancement of
2 high tropopause aerosol extinction in these cases.

3 The continuous variation of water vapor distribution observed by satellite, despite
4 lower vertical resolution, might also be used to investigate the contribution of liquid
5 solutions conversion to the enhancement of high tropopause aerosol extinction. Fig.7
6 shows the time series of water vapor profile derived from MLS, tropopause level from
7 sounder temperature profiles, and the altitude of daily mean maximum aerosol
8 extinction coefficients in this region. It can be clearly seen that almost all the
9 abundant water vapor transported by deep convective systems are concentrated below
10 120 hPa altitude (about 15 km). Meanwhile, the temporal correlation of extinction
11 coefficients in aerosol layer with water vapor from day to day is weak with correlation
12 coefficient of 0.36, suggesting that it is impossible that the enhanced tropopause
13 aerosol is due to the condensation of water vapor.

14

15 **4 Conclusion**

16 In this study, we observed significantly increased aerosol extinction coefficients
17 in UTLS over the Tibetan Plateau by continuous measurements with MPL during
18 summer 2011. The retrieval of MPL showed a good agreement with CALIOP. The
19 maximum SR of aerosol layers, up to 4-9, in the UTLS generally located in 18–19 km
20 m.s.l., 1–2 km higher than the tropopause, with broad layer depth ranging
21 approximately 3–4 km.

22 Even though the aerosol layers in UTLS wore off gradually with the reducing
23 intensity of the Asian monsoon over the Tibetan Plateau at the end of August, the
24 variation of maximum extinction coefficient of aerosol layers were found to be
25 connected to the local OLR convection proxy. In addition to the eruption of Nabro
26 volcano, appearance of deep transport from the most intense convection is considered
27 to be important factors to explain the enhancement of tropopause aerosols observed in
28 summer over the Tibetan Plateau. Deficiency in water vapor in UTLS indicates that
29 the effects of gas-to-particle conversion from liquid solutions would likely be
30 secondary to the enhancement of high tropopause aerosol extinction in these cases.

1 It is must be noted that our interpretations are based on a short time observation.
2 It is difficult to conclude that either one of the two processes is dominant due to lack
3 of observations for trace gases. If further observations with more frequent soundings
4 of water vapor and trace gases can be performed to investigate a correlation of high
5 aerosol extinction with ambient temperatures, water vapor, trace gases, liquid
6 solutions and transport processes, the result will be helpful in validating origination
7 and mechanism of the enhanced aerosol extinction in UTLS.

8
9 *Acknowledgements.* This study was supported by Special Funds for Meteorological Research in the
10 Public Interest (Grant Numbers: GYHY201106023, GYHY201006047), the National Natural Science
11 Foundation of China (NSFC, Grant Numbers: 40705013, 40975012 and 41175020), the Shanghai
12 Science and Technology Committee Research Special Funds (Grant Number: 10JC1401600). We thank
13 all TOAR team members and the staff from the Tibet Meteorological Service for assisting our
14 experiment work. The authors gratefully acknowledge the NOAA/OAR/ESRL PSD, Boulder, Colorado,
15 USA, for providing the interpolated OLR data on their web site <http://www.cdc.noaa.gov/>.

16 17 **References**

- 18 Bai, H. Z., Xie, J. N., and Li, D. L.: The principal feature of Qinghai-Xizang Plateau monsoon variation in 40
19 years, *Plateau Meteor.*, 20, 22–27, 2001.
- 20 Bai, H. Z., Ma, Z. F., and Dong, W. J.: Relationship between Qinghai-Xizang Plateau region monsoon features and
21 abnormal climate in china, *Plateau Meteor.*, 16, 484–491, 2005.
- 22 Bourassa, A., Robock, A., Randel, W., Deshler, T., Rieger, L., Lloyd, N., Llewellyn, E., and Degenstein, D.: Large
23 Volcanic Aerosol Load in the Stratosphere Linked to Asian Monsoon Transport, *Science*, 337,
24 doi:10.1126/science.1219371, 2012
- 25 Borrmann, S., Solomon, S., Avallone, L., Toohey, D., and Baumgardner, D.: On the occurrence of ClO in cirrus
26 clouds and volcanic aerosol in the tropopause region, *Geophys. Res. Lett.*, 24(16), 2011–2014, 1997.
- 27 Campbell, J. R., Hlavka, D. L., Welton, E. J., Flynn, C. J., Turner, D. D., Spinhirne, J. D., Scott, V. S., and
28 Hwang, I. H.: Full-time, eye-safe cloud and aerosol lidar observation at Atmospheric Radiation Measurement
29 program sites: Instruments and data processing, *J. Atmos. Oceanic Technol.*, 19, 431 – 442,
30 doi:10.1175/1520-0426(2002)019<0431:FTESCA>2.0.CO;2, 2002.
- 31 Clarisse, L., Coheur, P. F., Theys, N., Hurtmans, D., and Clerbaux, C.: The 2011 Nabro eruption, a SO₂ plume
32 height analysis using IASI measurements, *Atmos. Chem. Phys. Discuss.*, 13, 31161–31196,
33 doi:10.5194/acpd-13-31161-2013, 2013.
- 34 Cong, C. H., Li, W. L., and Zhou, X. J.: Atmospheric mass exchange between the troposphere stratosphere over
35 the Tibetan Plateau and its neighboring regions, *Science Bulletin*, 46(22), 1914–1918, 2001.
- 36 Dessler, A. E. and Sherwood, S. C.: Effect of convection on the summertime extra tropical lower stratosphere, *J.*
37 *Geophys. Res.*, 109, D23301, doi:10.1029/2004JD005209, 2004.
- 38 Dethof, A., O'Neill, A., Slingo, J. M., and Smit, H. G. J.: A mechanism for moistening the lower stratosphere
39 involving the Asian summer monsoon, *Q. J. R. Meteorol. Soc.*, 125, 1079– 1106, 1999.
- 40 Fernald, F. G.: Analysis of atmospheric lidar observations: Some comments, *Appl. Opt.*, 23(5), 652– 653, 1984.
- 41 Filipiak, M. J., Harwood, R. S., Jiang, J. H., Li, Q., Livesey, N. J., Manney, G. L., Read, W. G., Schwartz, M. J.,

1 Waters, J. W., and Wu, D. L.: Carbon monoxide measured by the EOS Microwave Limb Sounder on Aura: First
2 results, *Geophys. Res. Lett.*, 32, L14825, doi:10.1029/2005GL022765, 2005.

3 Fu, R., Hu, Y., Wright, J. S., Jiang, J. H., Dickinson, R. E., Chen, M., Filipiak, M., Read, W. G., Waters, J. W., and
4 Wu, D.: Short circuit of water vapor and polluted air to the global stratosphere by convective transport over the
5 Tibetan Plateau, *Proc. Natl. Acad. Sci. USA*, 103,5664–5669, 2006.

6 Fujiwara, M., Iwasaki, S., Shimizu, A., Inai, Y., Shiotani, M., Hasebe, F., Matsui, I., Sugimoto, N., Okamoto, H.,
7 Nishi, N., Hamada, A., Sakazaki, T., and Yoneyama, K.: Cirrus observations in the tropical tropopause layer over
8 the western Pacific, *J. Geophys. Res.*, 114, D09304, doi:10.1029/2008JD011040, 2009.

9 Gettelman, A., Salby, M. L., and Sassi, F.: Distribution and influence of convection in the tropical tropopause
10 region, *J. Geophys. Res.*, 107(D10), 4080, doi:10.1029/2001JD001048, 2002.

11 Hanson, D. R., Ravishankara, A. R., Solomon, S.: Heterogeneous reactions in sulfuric acid aerosols: A framework
12 for model calculations, *J. Geophys. Res.*, 99(D2), 3615- 3629, 1994.

13 He, Q., C., Li, J., Ma, H., Wang, G., Shi, Z., Liang, Q., Luan, F., Geng, and X., Zhou: The properties and
14 formation of cirrus clouds over the Tibetan Plateau based on summertime lidar measurements. *J. Atmos. Sci.*
15 doi:10.1175/JAS-D-12-0171.1, 70, 901-915, 2013.

16 Highwood, E. J. and Hoskins, B. J.: The tropical tropopause, *Q. J. R. Meteorol. Soc.*, 124, 1579– 1604 ,1998.

17 Hoskins, B. J. and Rodwell, M. J.: A model of the Asian summer monsoon. Part I: The global scale, *J. Atmos. Sci.*,
18 52, 1329– 1340, 1995.

19 Jackson, D. R., Driscoll, S. J., Highwood, E. J., Harries, J. E., and Russell, J. M.: Troposphere to stratosphere
20 transport at low latitudes as studied using HALOE observations of water vapour 1992–1997, *Q. J. R. Meteorol.*
21 *Soc.*, 124, 169– 192, 1998.

22 Keim, E. R., Fahey, D. W., Delnegro, L. A., Woodbridge, E. L., Gao, R. S., Wennberg, P. O., Cohen, R. C.,
23 Stimpfle, R. M., Kelly, K. K., Hints, E. J., Wilson, J. C., Jonsson, H. H., Dye, J. E., Baumgardner, D., Kaw, S. R.,
24 Salawitch, R. J., Proffitt, M. H., Loewenstein, M., Podolske, J. R., and Chan, K. R.: Observations of large
25 reductions in the NO/NO_y ratio near the mid-latitude tropopause and the role of heterogeneous chemistry,
26 *Geophys. Res. Lett.*, 23, 3223-3226, 1996.

27 Kim, Y. S., Shibata, T., Iwasaka, Y., Shj, G., Zhou, X., Tamuraa, K., and Ohashi, T.: Enhancement of Aerosols
28 near The Cold Tropopause in Summer over Tibetan Plateau: Lidar and Balloon-borne measurements in1999 at
29 Lhasa, Tibet, China, in: *Lidar Remote Sensing for Industry and Environment Monitoring III*, edited by: Singh U.
30 N., Itabe, T., and Liu, Z., *Proceedings of SPIE, Hangzhou, China* , 4893, 496-503, 2003.

31 Li, Q., Jiang, J., Wu, D., Read, W., Livesey, N., Waters, J., Zhang, Y., Wang, B., Filipiak, M., Davis,
32 C., Turquety, S., and Wu, S.: Convective outflow of South Asian pollution: A global CTM simulation compared
33 with EOS MLS observations, *Geophys. Res. Lett.*, 32, L14826, doi:10.1029/2005GL022762, 2005a

34 Li, Q., Jacob, D., Park, R., Wang, Y., Heald, C., Hudman, R., Yantosca, R., Martin, R., and Evans, M.: North
35 American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone, *J.*
36 *Geophys. Res.*, 110, D10301 doi:10.1029/2004JD005039, 2005b.

37 Li, W. L. and Yu, S. M.: The characteristics of aerosol spatial and temporal distribution, radiation forcing and
38 climate effect by numerical simulation over the Tibetan Plateau, *Sci. in China (Series D)*, 31(Supp.), 300-307,
39 2001.

40 Liebmann, B. and Smith, C. A.: Description of a complete (interpolated) outgoing longwave radiation dataset, *Bull.*
41 *Am. Meteorol. Soc.*, 77, 1275–1277, 1996.

42 Miloshevich, L. M., Vömel, H., Whiteman, D. N., Lesht, B. M., Schmidlin, F. J., and Russo, F.: Absolute accuracy
43 of water vapor measurements from six operational radiosonde types launched during AWEX-G and implications
44 for AIRS validation, *J. Geophys. Res.*, 111, D09S10, doi: 10.1029/2005JD006083, 2006.

1 Miloshevich, L., Vömel, H., Whiteman, D. N., and Leblanc, T.: Accuracy assessment and correction of Vaisala
2 RS92 radiosonde water vapor measurements, *J. Geophys. Res.*, 114(D11), doi: 10.1029/2008JD011565, 2009.

3 Müller, D., Ansmann, A., Mattis, I., Tesche, M., Wandinger, U., Althausen, D., and Pisani, G.:
4 Aerosol-type-dependent lidar ratios observed with Raman lidar, *J. Geophys. Res.*, 112, D16202,
5 doi:10.1029/2006JD008292, 2007.

6 NASA: U.S. Standard Atmosphere Supplements, U.S. Govt. Print. Off., Washington, D.C., 1976.

7 Park, M., Randel, W. J., Gettelman, A., Massie, S. T., and Jiang, J. H.: Transport above the Asian summer
8 monsoon anticyclone inferred from Aura Microwave Limb Sounder tracers, *J. Geophys. Res.*, 112, D16309,
9 doi:10.1029/2006JD008294, 2007.

10 Park, M., Randel, W. J., Kinnison, D. E., Garcia, R. R., and Choi, W.: Seasonal variation of methane, water vapor,
11 and nitrogen oxides near the tropopause: Satellite observations and model simulations, *J. Geophys. Res.*, 109,
12 D03302, doi:10.1029/2003JD003706, 2004.

13 Rosenlof, K. H., Tuck, A. F., Kelly, K. K., Russell III, J. M., and McCormick, M. P.: Hemispheric asymmetries in
14 water vapor and inferences about transport in the lower stratosphere, *J. Geophys. Res.*, 102, 13,213– 13,234, 1997.

15 Sawamura, P., Vernier, J. P., Barnes, J. E., Berkoff, T. A., Welton, E. J., Arboledas, L. A., Guzmán, F. N.,
16 Pappalardo, G., Mona, L., Madonna, F., Lange, D., Sicard, M., Beekmann, S. G., Payen, G., Wang, Z., Hu, S.,
17 Tripathi, S. N., Jabonero, C. C., and Hoff, R. M.: Stratospheric AOD after the 2011 eruption of Nabro volcano
18 measured by lidars over the Northern Hemisphere, *Environ. Res. Lett.*, 7(3), doi:10.1088/1748-9326/7/3/034013,
19 2013

20 Solomon, S.: Stratospheric ozone depletion: a review of concept and history, *Rev. Geophys.*, 37, 275-316, 1999.

21 Solomon, S., Borrmann, S., Garcia, R. R., Portmann, R., Thomason, L., Poole, L. R., Winker, D., and McCormick,
22 M. P.: Heterogeneous chlorine chemistry in the tropopause region, *J. Geophys. Res.*, 102(D17), 21411- 21429,
23 1997.

24 Tang, M. C., Liang, J., Shao, M. J., and Shi, G.: Preliminary analysis on the yearly variation of Tibetan Plateau
25 monsoon, *Plateau Meteor.*, 3, 76–82, 1984.

26 Tobo, Y., Zhang, D., Iwasaka, Y., Shi, G., Kim, Y., Ohashi, T., Tamura, K., and Zhang, D.: Balloon-borne
27 observations of high aerosol concentrations near the summertime tropopause over the Tibetan Plateau, *Atmos. Res.*,
28 84, 233-241, 2007.

29 Tukiainen, S., Kujanpää J., Bingen, C., Robert, C., Tóard, C., and Dekemper, E.: Nabro volcano aerosol in the
30 stratosphere over Georgia, South Caucasus from ground based spectrometry of twilight sky brightness, *Atmos.*
31 *Meas. Tech.*, 6, 2563-2576, doi:10.5194/amt-6-2563-2013, 2013

32 Uchino, O., Sakai, T., Nagai, T., Nakamae, K., Morino, I., Arai, K., Okumura, H., Takubo, S., Kawasaki, T., Mano,
33 Y., Matsunaga, T., and Yokota, T.: On recent(2008–2012) stratospheric aerosols observed by lidar over Japan,
34 *Atmos. Chem. Phys.*, 12, 11975-11984, doi:10.5194/acp-12-11975-2012, 2012.

35 Vömel, H. H., Selkirk, L., Miloshevich, J., Valverde-Canossa, J., Valdes, J., and Diaz, J.: Radiation Dry Bias of
36 the Vaisala RS92 Humidity Sensor, *J. Atmos. Ocean. Tech.*, 24, 953–963, 2007.

37 Währn, J., Oyj, V., Reikioski, I., Jauhainen, H., and Hirvensalo, J.: New Vaisala Radiosonde RS92: Testing and
38 Results from the Field, Eighth Symposium on Integrated Observing and Assimilation Systems for Atmosphere,
39 Oceans, and Land Surface, Seattle, USA, 13 January 2004, 2004.

40 Waters, J. W., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R.
41 E., Filipiak, M. J., Flower, D. A., Holden, J. R., Lau, G. K., Livesey, N. J., Manney, G. L., Pumphrey, H. C.,
42 Santee, M. L., Wu, D. L., Cuddy, D. T., Lay, R. R., Loo, M. S., Perun, V. S., Schwartz, M. J., Stek, P. C.,
43 Thurstans, R. P., Boyles, M. A., Chandra, K. M., Chavez, M. C., Chen, G. S., Chudasama, B. V., Dodge, R., Fuller,
44 R. A., Girard, M. A., Jiang, J. H., Jiang, Y., Knosp, B. W., LaBelle, R. C., Lam, J. C., Lee, K. A., Miller, D.,

1 Oswald, J. E., Patel, N. C., Pukala, D. M., Quintero, O., Scaff, D. M., Van Snyder, W., Tope, M. C., Wagner, P. A.,
2 and Walch, M. J.: The Earth Observing System microwave limb sounder (EOS MLS) on the Aura satellite, *IEEE T.*
3 *Geosci. Remote*, 44, 1075– 1092, 2006.

4 Winker, D. M., Pelon, J., and McCormick, M. P.: The CALIPSO mission: Space borne lidar for observation of
5 aerosols and clouds, *Proc. SPIE*, 4893, 1–11, 2003.

6 Wu, P.M., Okada, K., Tanaka, T., Sasaki, T., Nagai, T., Fujimoto, T., and Uchino, O.: Balloon observation of
7 stratospheric aerosols over Tsukuba, Japan Two years after the Pinatubo volcanic eruption, *J. Meteor. Soc. Jpn.*, 72,
8 475–480, 1994.

9 Xun, X. Y., Hu, Z. Y., Cui, G. F., He, H. G., Sun, J., Hao, L., and Gu, L. L.: Change of monsoon in
10 Qinghai-Xizang Plateau and its correlation with summer precipitation of Ordos Plateau, *J. Arid Land Resour.*
11 *Environ.*, 25 (4), 79–83, 2011.

12 Yan, X. L.: The observation and study on the upper troposphere and lower stratosphere water vapor and ozone
13 over Tibetan Plateau and its adjoint regions, master's thesis, Beijing: Chinese Academy of Meteorological Sciences,
14 2012.

15 Yanai, M., Li, C., and Song, Z.: Seasonal heating of the Tibetan Plateau and its effects on the evolution of the
16 Asian summer monsoon, *J. Meteorol. Sci. Jpn.*, 70, 319– 351, 1992.

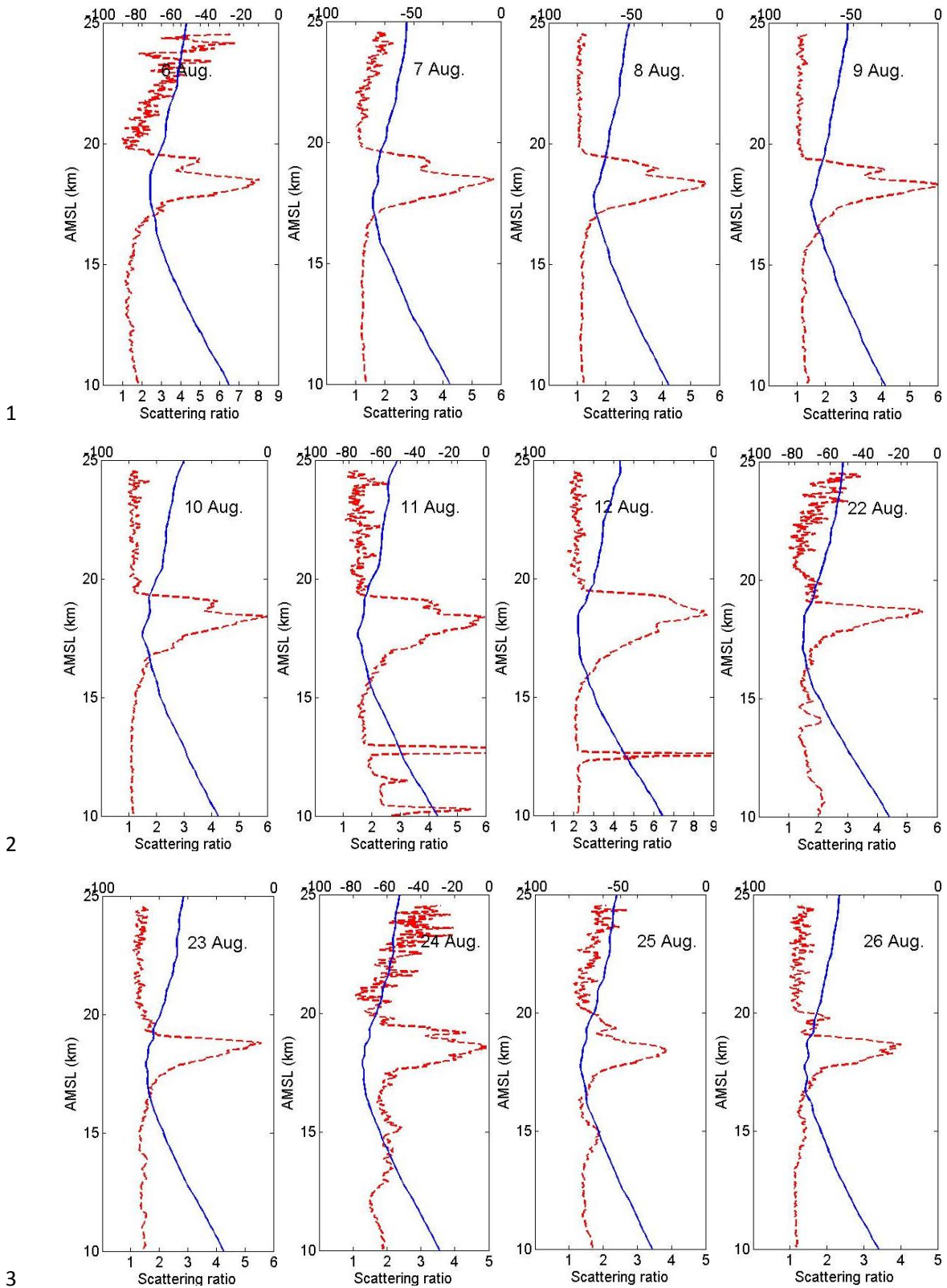
17 Yin, Y., Chen, Q., Jin, L., Chen, B., Zhu, S., and Zhang, X.: The effects of deep convection on the concentration
18 and size distribution of aerosol particles within the upper troposphere: A case study, *J. Geophys. Res.*, 117,
19 D22202, doi:10.1029/2012JD017827, 2012.

20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44

1 **Table 1** Statistical parameters of aerosol layer over Tibet and Shanghai. Maximum extinction
 2 coefficient (EC_{max}), Averaged extinction coefficient (EC_{ave}), aerosol layer depth(ALD), aerosol layer
 3 height over sea level and aerosol optical depth (AOD) of the aerosol layer from 20:00 to 06:00 local
 4 standard time (LST). The numbers in parenthesis correspond to the standard deviations.

	$EC_{max}(km^{-1})$	$EC_{ave}(km^{-1})$	ALD(km)	ALH(km)	AOD
Tibet	0.007	0.002(0.002)	3.604(1.626)	18.492(0.248)	0.016(0.002)
Shanghai	0.010	0.006(0.003)	4.380(0.764)	16.860(0.839)	0.027(0.006)

5
 6
 7
 8
 9
 10
 11
 12
 13
 14
 15
 16
 17
 18
 19
 20
 21



1

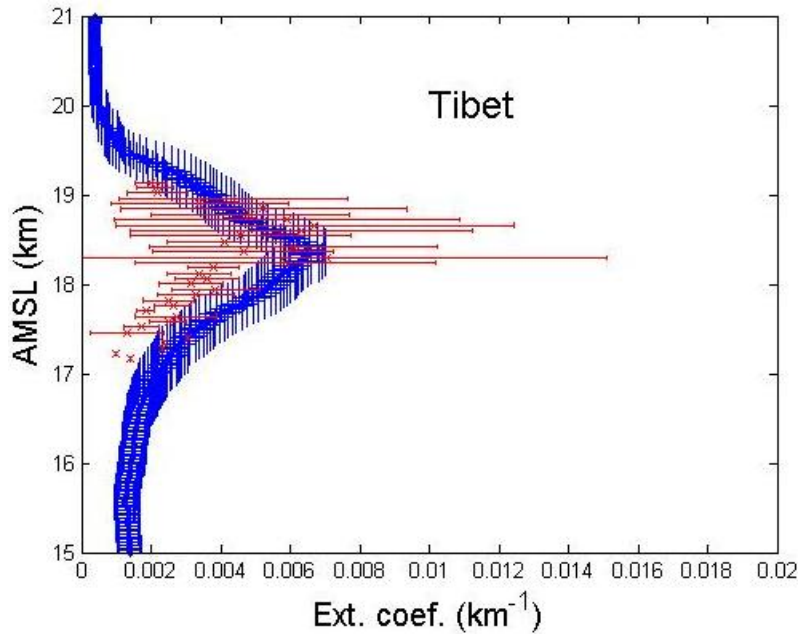
2

3

4 **Fig. 1.** The nighttime mean aerosol scattering ratio profiles (dashed line) from MPL. The daily mean
 5 profiles of temperature (solid line °C) from the two radiosondes each day are overlaid to indicate the
 6 altitude of the tropopause (~18 km m.s.l.).

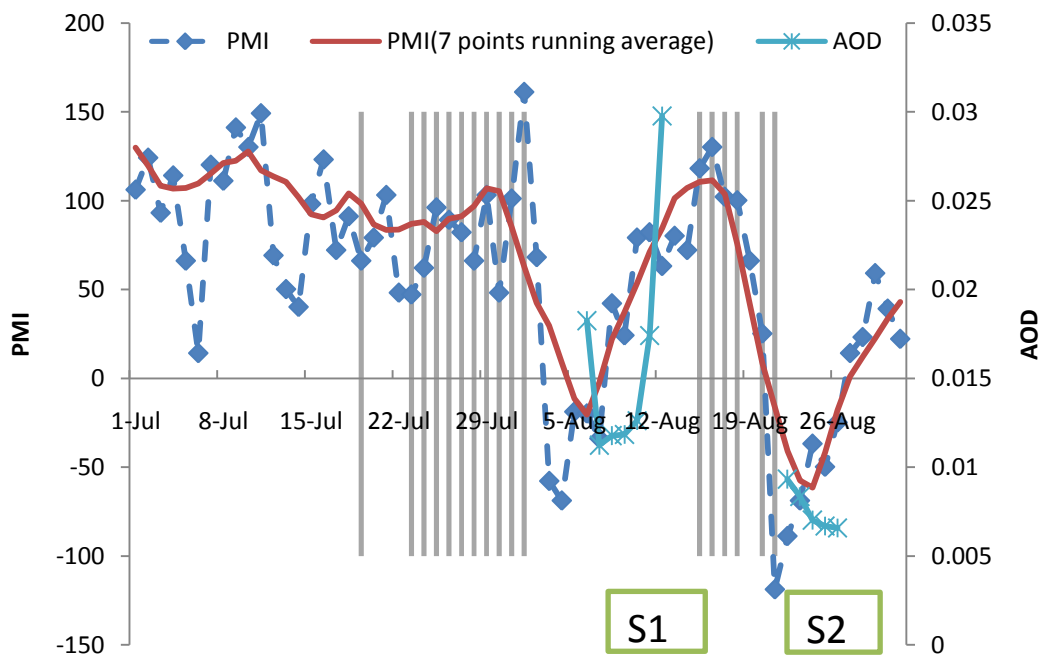
7

8



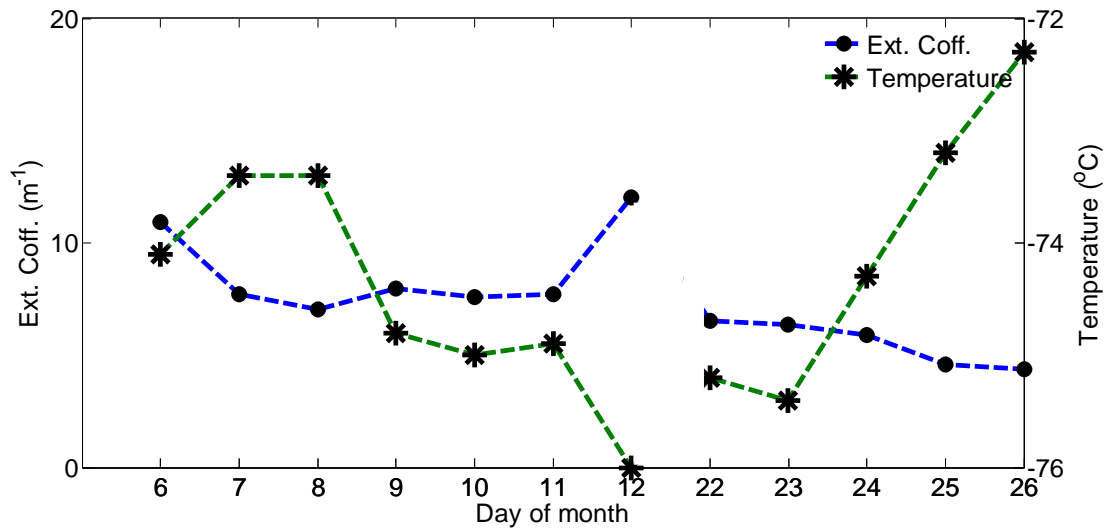
1
2
3
4
5

Fig.2. The average extinction coefficient profile of MPL (blue solid line) and the average extinction coefficient at each layer from CALIOP (red stars) during the whole observation period. The standard errors are marked as the error bar.



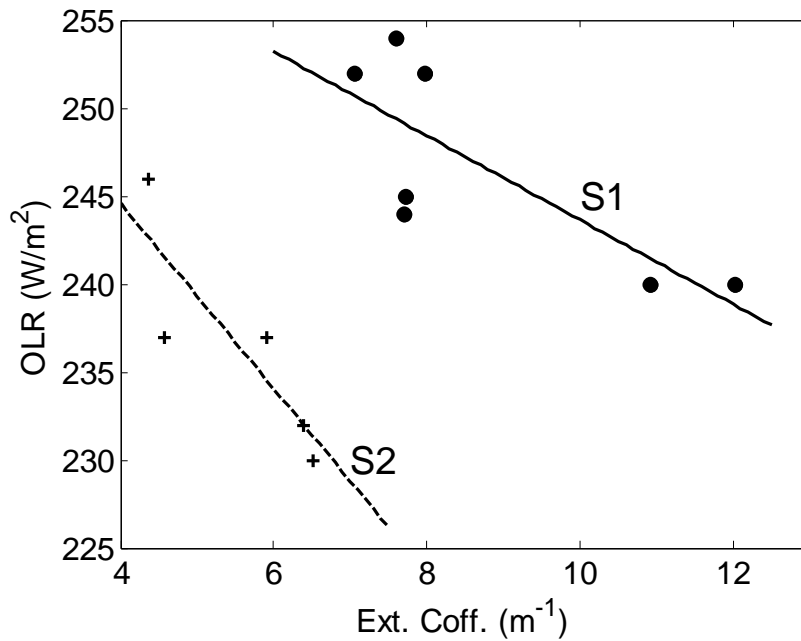
6
7
8
9
10
11

Fig. 3. Daily variation of PMI and the 7-day-averaged PMI time series from 1 Jul to 31 Aug 2011 and AOD in UTLS retrieved from MPL over the Tibetan Plateau. The days with cirrus occurrence are shaded (He et al. 2013).



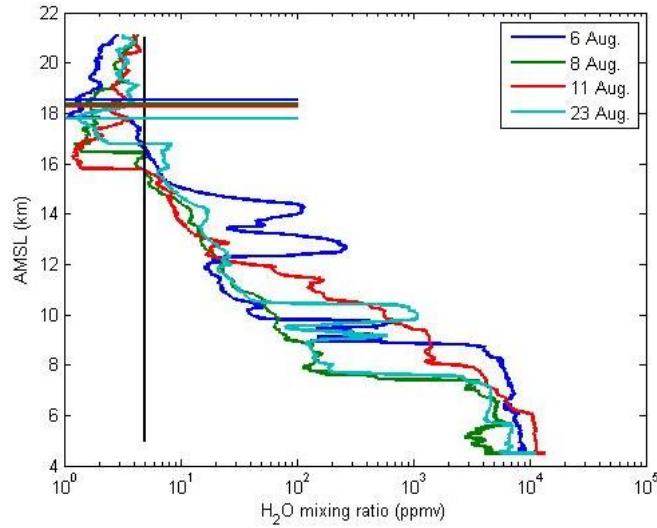
1
2
3
4
5

Fig. 4. The time series of maximum extinction coefficient in the aerosol layers and temperature at the tropopause over the Tibetan Plateau day-on-day in August 2011.

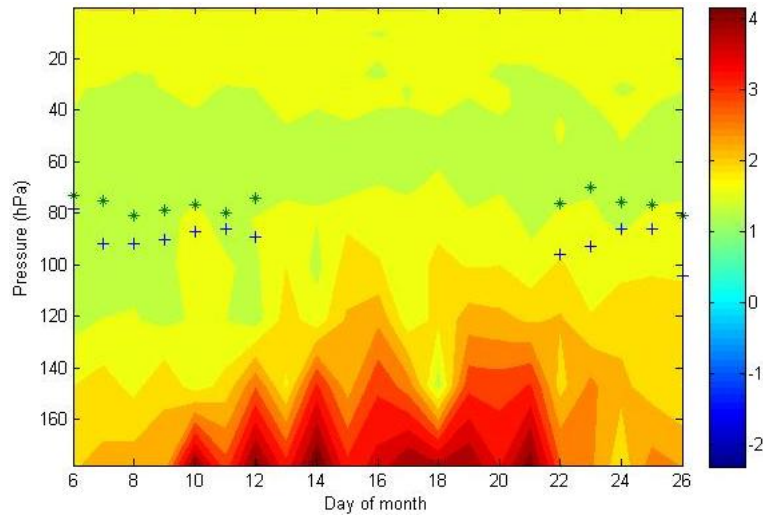


6
7
8
9
10

Fig. 5. Nighttime mean maximum extinction coefficient in the aerosol layer vs. OLR. The dots represent the data for 6–12 August (S1) and the pluses for 22–26 August 2011 (S2).



1
 2 **Fig. 6.** Vertical profiles of water vapor from Vaisala RS92 radiosondes released in 6, 8, 11 and 23
 3 August 2011, respectively. The black line along y axis represents the 5 ppmv of water vapor mixing
 4 ratio. The color lines along x axis are the altitudes with the maximum extinction coefficient of aerosol
 5 layers for each day.
 6



7
 8 **Fig. 7.** Altitude-time distributions of MLS water vapor (ppmv, color bar in natural logarithm) from 6 to
 9 26 August 2011. Stars indicate the layer with nighttime mean maximum extinction coefficient and
 10 pluses stand for the tropopause level of each day, respectively.
 11