



# Spatiotemporal variation of aerosol and potential long-range transport impact over the Tibetan Plateau, China

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**Abstract.** The long-term temporal–spatial variations in the aerosol optical properties over the Tibetan Plateau (TP) and the potential long-range transport from surrounding areas to the TP were analyzed in this work, by using multiple years of sun photometer measurements (CE318) at five stations in the TP, satellite aerosol products from the Moderate Resolution Imaging Spectroradiometer (MODIS) and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), back-trajectory analysis from the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLOT) and model simulations from the Goddard Earth Observing System (GEOS)-Chem chemistry transport model. The results from the ground-based observations showed that the annual aerosol optical depth (AOD) at 440 nm at most TP sites increased in recent decades with trends of  $0.001 \pm 0.003 \text{ yr}^{-1}$  at Lhasa,  $0.013 \pm 0.003 \text{ yr}^{-1}$  at Mt\_WLG,  $0.002 \pm 0.002 \text{ yr}^{-1}$  at NAM\_CO and  $0.000 \pm 0.002 \text{ yr}^{-1}$  at QOMS\_CAS. The increasing trend was also found for the aerosol extinction

Ångström exponent (EAE) at most sites with the exception of the Mt\_WLG site. Spatially, the AOD at 550 nm observed from MODIS showed negative trends at the northwest edge close to the Taklimakan Desert and to the east of the Qaidam Basin and slightly positive trends in most of the other areas of the TP. Different aerosol types and sources contributed to a polluted day (with CE318 AOD at 440 nm > 0.4) at the five sites on the TP: dust was the dominant aerosol type in Lhasa, Mt\_WLG and Muztagh with sources in the Taklimakan Desert, but fine-aerosol pollution was dominant at NAM\_CO and QOMS\_CAS with transport from South Asia. A case of aerosol pollution at Lhasa, NAM\_CO and QOMS\_CAS during 28 April–3 May 2016 revealed that the smoke aerosols from South Asia were lifted up to 10 km and transported to the TP, while the dust from the Taklimakan Desert could climb the north slope of the TP and then be transported to the central TP. The long-range transport of

aerosol thereby seriously impacted the aerosol loading over the TP.

## 1 Introduction

The heavy haze that has occurred in recent years in China has been largely attributed to the atmospheric aerosols (Zhang et al., 2015). In addition, atmospheric aerosols can affect the climate through the interactions between aerosol and radiation and between aerosol and cloud (Takemura et al., 2005; Li et al., 2017; Huang et al., 2006a, b; Liu et al., 2011, 2014), while the clouds and their precipitation are also connected to large-scale atmospheric circulations (Yang et al., 2010, 2017a). However, there is still a high level of uncertainty about the impact of aerosols on the climate, which is mostly due to the high spatiotemporal variability of aerosols (IPCC, 2013). Therefore, studying the physical and chemical properties of aerosols over different regions is essential.

The Tibetan Plateau (TP) is the largest elevated plateau in East Asia and considered one of the most pristine terrestrial regions, along with the Arctic and Antarctic. However, in the past two decades, the TP has been surrounded by an unprecedented growth of emissions of Asian air pollutants from various sources. Consequently, some studies have demonstrated that the aerosols transported from its surrounding areas (South Asia and Taklimakan Desert) have polluted the TP (Huang et al., 2007; Xia et al., 2011; Kopacz et al., 2011; Lu et al., 2012; Liu et al., 2015; Zhu et al., 2018; Jia et al., 2015, 2019). The increase in aerosols over the TP may have an important impact on the regional or global climate. Lau et al. (2006) has suggested that increased absorbing aerosols (dust and black carbon) over the TP may create a positive tropospheric temperature anomaly over the TP and adjacent regions to the south, causing the advance and enhancement of the Indian summer monsoon. Besides the impact of aerosols over the TP on the radiation budget, temperature and Indian summer monsoon, Liu et al. (2019a) reported a potential relationship may exist between the aerosol index and ice cloud properties over the TP, in which the aerosols have a more dominant influence than meteorological conditions on ice cloud properties (except for the nocturnal ice cloud droplet radius and ice water path during the daytime). Furthermore, Liu et al. (2019b) found the effect of the dust aerosols over the TP on the development of convective clouds and then movement of the some developed convective clouds could induce significant precipitation over the Yangtze River basin and North China. Attempts have been made to reveal the linkages between climate change (such as changes to glaciers and monsoons) and the air pollutants around the TP (mainly absorbing carbonaceous materials) (Qian et al., 2011; Wang et al., 2016; Lee et al., 2013; Jia et al., 2018). However, the quantitative effect of the TP aerosol on climate variability

remains largely unknown, and it is very essential to fully understand the aerosol characteristics over the TP.

A large amount of attention has been paid to aerosol characteristics over the TP (Wan et al., 2015; Tobo et al., 2007; Zhao et al., 2013; Liu et al., 2008; Du et al., 2015). Although the seasonal variations in aerosol properties over the TP have been analyzed based on ground-based observations or satellite products (Shen et al., 2015; Xia et al., 2008), analysis is needed of the long-term trends in the variation of aerosols over the TP to provide predictions and guidelines for environment policies. In past studies, spring or summer have often been studied due to the important impacts of dust and carbonaceous aerosols (Huang et al., 2007; Cong et al., 2007; Lee et al., 2013). However, most studies of the aerosol properties based on ground-based measurements have been conducted at a single site over the TP, such as NAM\_CO (Cong et al., 2009), Mt\_Yulong (Zhang et al., 2012) and Mt\_WLG (Che et al., 2011). Past studies have mostly focused on single stations or short-term variations due to the difficulty of taking a sufficient number of ground-based observations in challenging weather conditions over the remote plateau.

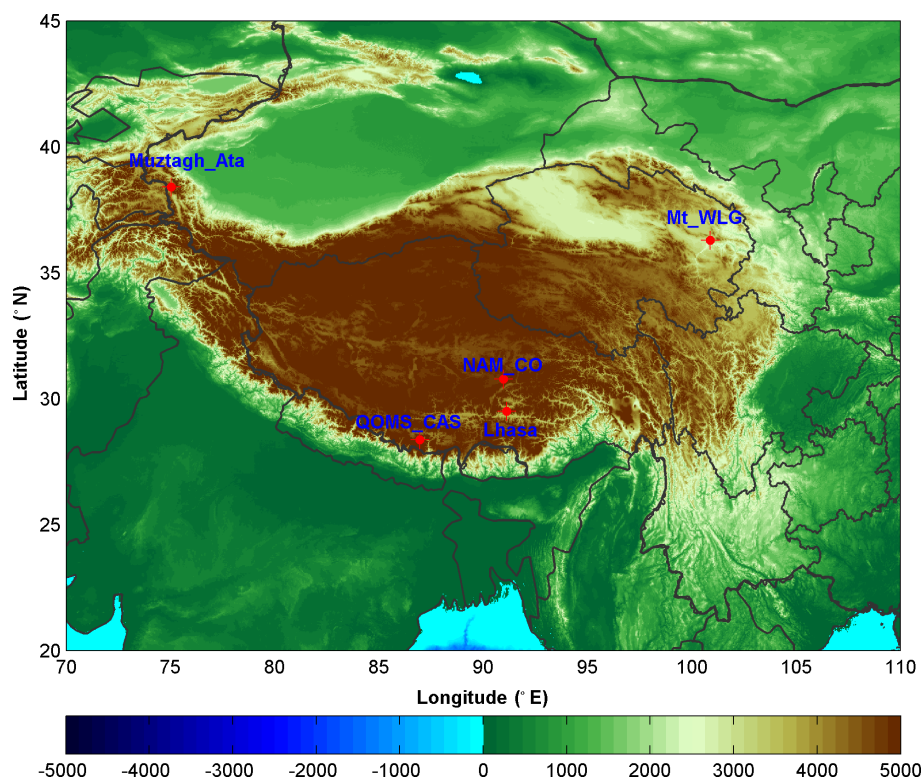
Ground-based measurements can offer more accurate data on aerosol properties, while large-scale spatial observations of aerosol optical and physical properties require satellite remote-sensing methods (Li et al., 2015, 2018; Xing et al., 2017). Thus, the long-term detection of aerosols from both ground and satellite platforms is absolutely necessary for improving our understanding of the climate effects of aerosol over the TP region. Consequently, based on multiple years of observations from five ground-based sun photometers at the TP and the MODIS aerosol optical depth product over the TP region, our work here is focused on the long-term spatiotemporal variations in the aerosol optical properties over the TP and the aerosol properties and sources during the high aerosol loading over the TP. In addition, we also combined the observation and models to study the aerosol transport process over the TP, thereby helping to reduce the uncertainties in estimating aerosol radiative forcing and aerosol sources.

In this paper, Sect. 2 describes the observation sites, data and methods. The results of the analysis of the spatiotemporal variations in aerosol properties over the TP are shown in Sect. 3. The analysis of aerosol high loading and an aerosol transport case are presented in Sects. 4 and 5, respectively. The conclusions are presented in Sect. 6.

## 2 Site, data and methodology

### 2.1 Sites

In this study, five sites in the TP equipped with sun- and sky-scanning radiometers (CE318) were used (Fig. 1). Table 1 shows the station locations and descriptions. The Lhasa station is the only urban site that suffers from local anthropogenic emissions. For the other four sites, local anthro-



**Figure 1.** Topography of the Tibetan Plateau (TP) and the five CE318 stations located in the TP (Lhasa, Mt\_WLG, Muztagh\_Ata, NAM\_CO and QOMS\_CAS).

**Table 1.** Site location and description.

Site name	Lat (° N)	Long (° E)	Site description, observation days and period
Lhasa	29.50	91.13	Urban station on the Tibetan Plateau, 3648 m a.s.l., 1554 d, May 2007–Dec 2017
Mt_WLG	36.28	100.90	Mountain, 3816 m a.s.l., 314 d, Sep 2009–Jul 2013
Muztagh_Ata	38.41	75.04	Mountain, 3674 m a.s.l., 84 d, Jun–Oct 2011
NAM_CO	30.77	90.96	Mountain, 4746 m a.s.l., 1061 d, Aug 2006–Aug 2016
QOMS_CAS	28.36	86.95	Mountain, 4276 m a.s.l., 1623 d, Oct 2009–Nov 2017

pogenic emissions are extremely rare due to the low number of human inhabitants. However, Mt\_WLG is in the north-east of the TP, where it is situated on the dust transport path from the largest desert in China (the Taklimakan Desert). The Muztagh\_Ata site is located in the northwest corner of the TP and next to the Central Asian desert and the Taklimakan Desert. NAM\_CO is located on the central Tibetan Plateau, 220 km away from Lhasa. QOMS\_CAS is located at the northern slope of Mt. Qomolangma on the border of Tibet and Nepal. Therefore, these five sites are representative of the spatial features of the TP.

## 2.2 Data

### 2.2.1 CE318 aerosol optical properties

The column-integrated aerosol properties over the five TP sites are derived from CE318 measurements. Table 1 shows the observation period. The CE318 instrument measures direct solar spectral radiation and the angular distribution of sky radiance. These spectral radiances can be used to retrieve aerosol optical parameters (such as aerosol optical depth – AOD) based on Beer's law and aerosol microphysical properties (such as volume size distribution) and the radiative forcing through radiation transfer theory (Dubovik and King, 2000; Dubovik et al., 2006). The instruments were periodically calibrated using the Langley method at AERONET global calibration sites (Izaña, Spain; or Mauna Loa, USA) or using the intercomparison calibra-

tion method at the Beijing-CAMS site (Che et al., 2015). The cloud-screened and quality-controlled data of AOD, extinction Ångström exponent (EAE), and aerosol volume size distribution ( $dV(r)/d\ln r$ ) are used in this work (Giles et al., 2019). Eck et al. (1999) showed that the uncertainty of the AOD was approximately 0.01 to 0.02. The EAE was calculated from the AOD at 440 and 870 nm. The errors of retrieval for  $dV(r)/d\ln r$  were less than 10 % in the maxima of the  $dV(r)/d\ln r$  and may increase up to 35 % for the minimum values of  $dV(r)/d\ln r$  within the radius range between 0.1 and 7  $\mu\text{m}$ ; for the edges of the retrieval size, the errors increased apparently but did not significantly affect the derivation of the main feature of  $dV(r)/d\ln r$  (Dubovik et al., 2002).

### 2.2.2 The MODIS AOD product

The Moderate Resolution Imaging Spectroradiometer (MODIS) instrument is a multispectral sensor with a wide spectral range from 0.4 to 14.4  $\mu\text{m}$  in 36 wavelength bands, on board the Terra (morning descending direction) and Aqua (afternoon ascending direction) satellites in polar orbit. Its broad swath of 2330 km permits retrieval aerosol products to cover the global word within 1–2 d. In this study, both the Terra and Aqua MODIS Collection 6 Deep Blue (DB) and Dark Target (DT) combined AOD at 550 nm products with 10 km spatial resolution (Levy et al., 2013) from 2006 to 2017 were used. The combined MODIS DT and DB AOD at 550 nm (MODIS\_AOD) merges the products from the two algorithms based on the normalized difference vegetation index (NDVI) statistics as follows: (1) the DT AOD data are used for  $\text{NDVI} > 0.3$ , (2) the DB AOD data are used for  $\text{NDVI} < 0.2$ , and (3) the mean of both the algorithms or AOD data with a high-quality flag is used for  $0.2 \leq \text{NDVI} \leq 0.3$ . The MODIS\_AOD had been validated in the global or regional areas (Bilal et al., 2018; Ma et al., 2016; Sayer et al., 2014). The root-mean-square error (RMSE) of MODIS\_AOD was approximately 0.13, and the percentage of MODIS\_AOD data within the expected error was more than 71 % at the Kunming site, which is near the TP (Zhu et al., 2016).

### 2.2.3 The CALIOP profile data

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), the primary instrument on board the CALIPSO satellite, is a near-nadir-viewing two-wavelength (532 and 1064 nm) polarization-sensitive lidar that performs global vertical profile measurements of aerosols and clouds (Winker et al., 2010). It provides three primary calibrated and geolocated profile products: total attenuated backscatter at 532 and 1064 nm and the perpendicular polarization component at 532 nm. The CALIOP products (version 4.10) used in this study include the attenuated backscattering coefficient profiles from Level 1B and the vertical feature

mask data products of aerosol subtype from Level 2 products under 15 km altitude, which were downloaded from the Langley Atmospheric Science Data Center (ASDC). Kumar et al. (2018) had shown that the AOD from CALIOP version 4.10 agreed with the ground-based CE318 observation at a site in the central Himalayas with a correlation  $> 0.9$ , and  $\sim 87\%$  of the matchup data were within the expected error limits.

## 2.3 Methodology

The ground-based CE318 observations and MODIS AOD products were analyzed to show the spatiotemporal variations in aerosol properties in the TP.

The CE318-observed AOD at 440 nm with values larger than 0.4 at each site was specially analyzed to study the aerosol properties of the high aerosol loading over the TP. The value of 0.4 was selected because the mean annual values of AOD observed by CE318 instruments at the TP sites were less than  $\sim 0.1$  in the past studies (Xia et al., 2016; Cong et al., 2009), and this value is normally regarded as the high aerosol loading (Eck et al., 2010; Giles et al., 2012). Back trajectories were used for the aerosol source analysis in the TP. The back trajectories on the high aerosol loading days were calculated by using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model which was driven by the  $1^\circ$  horizontal resolution archived meteorological fields (Draxler and Hess, 1998). Back trajectories of 72 h ending at the five sites at 10 m a.g.l. (above ground level) at 12:00 UTC on the days with high aerosol loading (CE318 AOD at 440 nm  $> 0.4$ ) were used to identify the air mass sources.

A case of long-range aerosol transport to the TP was selected based on the ground CE318 observations over Lhasa, NAM\_CO and QOMS\_CAS. The HYSPLIT back trajectories and the MODIS and CALIOP products were used to show the potential aerosol sources, spatial aerosol loading and the vertical features of the aerosol over the TP during the case period. In addition, the Goddard Earth Observing System (GEOS)-Chem chemistry transport model was used to simulate the AOD and its components (dust and carbon aerosols) during the case period, which may reflect the change in aerosol type during the case period.

The GEOS-Chem chemical transport model (version 11-01) coupled with the online radiative transfer calculations (RRTMG) at  $0.5^\circ \times 0.667^\circ$  horizontal resolution over the East Asia domain (Bey et al., 2001; Wang et al., 2004) was used. The model was driving by the Global Modeling and Assimilation Office (GMAO) MERRA-2 meteorology with the temporal resolution of 3 h for meteorological parameters and 1 h for surface fields. The simulation type of full chemistry in the troposphere was selected. The implementation of RRTMG in GEOS-Chem was described in Heald et al. (2014). The AOD was calculated according to Martin et al. (2003). The default global anthropogenic emissions were



**Table 2.** Seasonal aerosol optical depth (AOD<sub>440nm</sub>) and extinction Ångström exponent (EAE<sub>440–870nm</sub>) from CE318 at the five sites in the TP.

Site	AOD				EAE			
	MAM	JJA	SON	DJF	MAM	JJA	SON	DJF
Lhasa	0.16 + 0.10	0.12 + 0.08	0.10 + 0.18	0.09 + 0.08	0.72 + 0.37	0.97 + 0.40	1.11 + 0.38	0.91 + 0.52
Mt_WLG	0.13 + 0.16	0.14 + 0.07	0.08 + 0.11	0.08 + 0.07	0.37 + 0.38	0.65 + 0.40	1.04 + 0.80	0.58 + 0.69
Muztagh_Ata	NaN	0.14 + 0.06	0.14 + 0.05	NaN	NaN	0.73 + 0.30	0.64 + 0.27	NaN
NAM_CO	0.07 + 0.07	0.06 + 0.04	0.03 + 0.05	0.03 + 0.01	0.63 + 0.44	0.62 + 0.45	0.65 + 0.32	0.78 + 0.43
QOMS_CAS	0.08 + 0.06	0.06 + 0.04	0.03 + 0.01	0.03 + 0.02	1.04 + 0.38	0.76 + 0.43	0.85 + 0.51	1.10 + 0.67

overwritten over East Asia by the MIX inventory from Li et al. (2014). The Global Fire Emission Database (GFED) (van der Werf et al., 2010) has been used to specify emissions from fire. More details on the model and the other emissions data used and the evaluation of AOD in the east and south of the TP were shown in Zhu et al. (2017).

In this study, the AOD values from the CE318, MODIS, and GEOS-Chem model were used. For convenience, CE318\_AOD, MODIS\_AOD and Model\_AOD stand for the AOD observed by CE318, MODIS and the AOD simulated by the GEOS-Chem model, respectively. For CE318\_AOD, the 440 nm wavelength is often studied, while MODIS\_AOD and Model\_AOD generally use the data at 550 nm wavelength. Thus, unless otherwise specified, CE318\_AOD, MODIS\_AOD and Model\_AOD hereinafter represent the ones at 440, 550 and 550 nm, respectively.

### 3 Temporal–spatial variations in aerosol properties

#### 3.1 Aerosol properties observed by the CE318 instruments

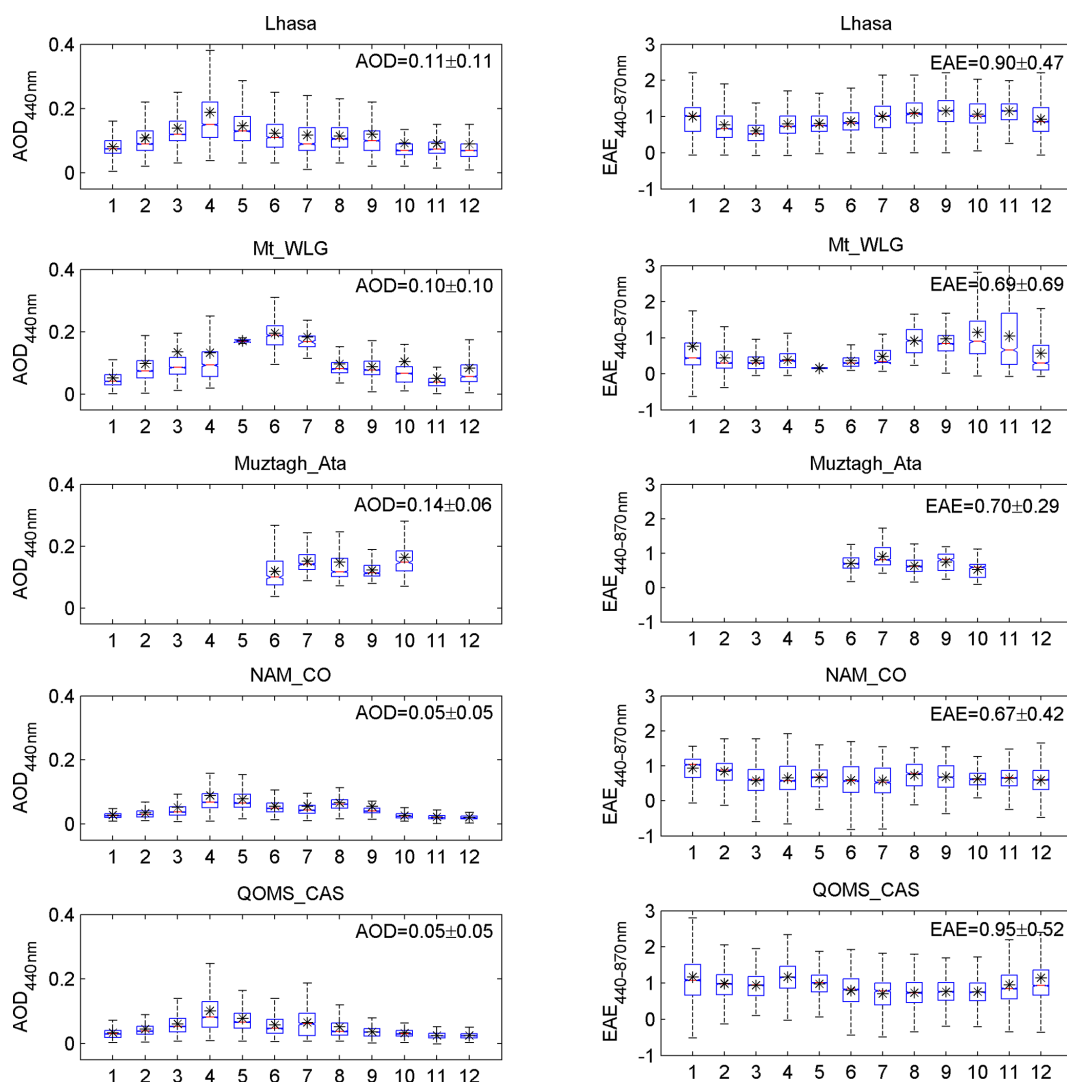
The monthly, seasonal, and annual variations in aerosol properties observed from the CE318 instruments at the five TP sites were analyzed.

The monthly variations in CE318\_AOD and EAE at the five sites over the TP are shown in Fig. 2. The monthly mean CE318\_AOD was highest in April at the Lhasa (0.19), NAM\_CO (0.09) and QOMS\_CAS (0.10) sites, while the value at Mt\_WLG was highest in June (0.20). The monthly mean CE318\_AOD rapidly increased from January to April and then slightly decreased until December at the Lhasa, NAM\_CO and QOMS\_CAS sites. However, the monthly mean CE318\_AOD at Mt\_WLG was nearly symmetrical from January to December. The monthly variation in EAE was different from the AOD. The highest monthly mean values of EAE occurred in September at Lhasa (1.15), October at Mt\_WLG (1.15), and in January at the NAM\_CO (0.93) and QOMS\_CAS (1.17) sites. The EAE at QOMS\_CAS also showed a high value of 1.17 in April, which may be caused by the smoke aerosol transported from South Asia during this period. The monthly mean EAE first decreased from January

to March and then increased until September at Lhasa. The monthly mean EAE values at NAM\_CO also decreased from January to March but did not increase apparently in the following months. The EAE at Mt\_WLG decreased from January to May and then increased obviously from May to October. The Lhasa, NAM\_CO, and QOMS\_CAS sites are near and located in the south of the TP. Thus, the variations in the aerosol properties at these three sites were similar. The Mt\_WLG site is located in the northeast of the TP, which is different from the southern sites. The Muztagh\_Ata is in the northwest of the TP and is the closest site to the Taklimakan Desert, which causes the high AOD there (a few observed data may be another reason). Looking at the monthly CE318\_AOD and EAE values together, high CE318\_AOD was often accompanied by low EAE at Lhasa, Mt\_WLG and NAM\_CO, indicating that these sites suffered from coarse aerosols such as dust (Huang et al., 2007; Liu et al., 2015; Zhang et al., 2001). However, the QOMS\_CAS site showed high CE318\_AOD and high EAE in April, which may be related to smoke aerosols transported from South Asia.

Table 2 shows the seasonal statistics of CE318\_AOD and EAE. A distinct seasonal variation in CE318\_AOD and EAE can be found over the TP sites. The CE318\_AOD mean values in fall (SON) and winter (DJF) were lower at all sites except Muztagh. Muztagh\_Ata showed high CE318\_AOD in both observed seasons. Except for that in Muztagh, the maximum seasonal CE318\_AOD was observed in spring (MAM) (Lhasa, NAM\_CO and QOMS\_CAS) or in summer (JJA) (Mt\_WLG). The minimum seasonal EAE occurred in spring (Lhasa and Mt\_WLG) or summer (NAM\_CO and QOMS\_CAS), while the maximum EAE values were mostly observed in fall (Lhasa and Mt\_WLG) and winter (NAM\_CO and QOMS\_CAS). These indicated frequent dust events over the TP in the spring and summer period. Mt\_WLG is situated on the dust transport path from the Taklimakan Desert, which causes the high CE318\_AOD observed in spring and summer at this site.

The seasonal size distributions of the five sites in Fig. 3 also demonstrated that coarse-mode aerosol was dominant at the five TP sites in almost all seasons, which was different from those in the eastern pollution regions of China, such as the Yangtze River Delta, where fine-mode aerosol

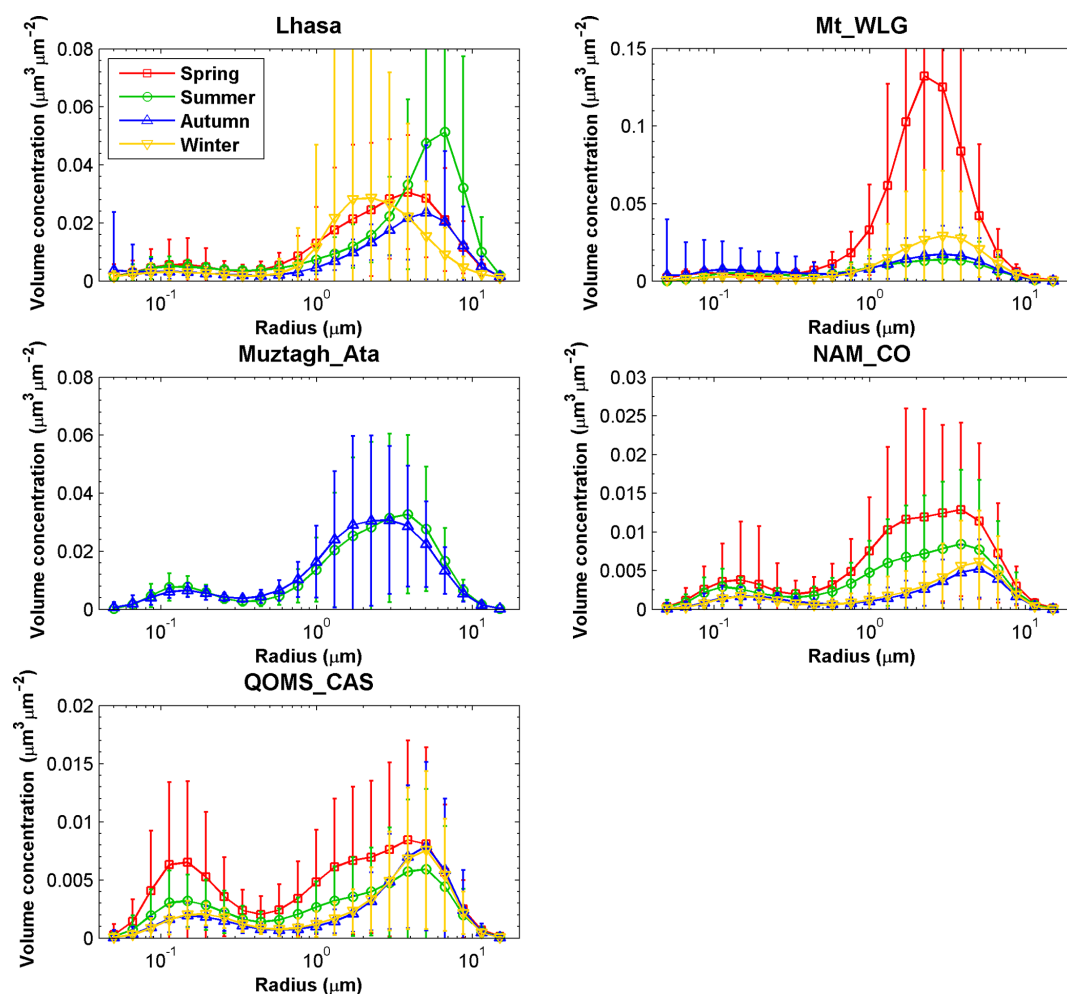


**Figure 2.** Box plots of the monthly aerosol optical depth (AOD) and extinction Ångström exponent (EAE) from CE318 at the five sites located on the Tibetan Plateau, i.e., Lhasa, Mt\_WLG, Muztagh\_Ata, NAM\_CO and QOMS\_CAS. In each box, the red line in the center is the median, and the lower and upper limits are the first and the third quartiles, respectively. The lines extending vertically from the box indicate the spread of the distribution with the length being 1.5 times the difference between the first and the third quartiles. The asterisk symbols indicate the geometric means in each month. The annual mean values and errors are also shown in each subgraph.

was dominant (Zhuang et al., 2018). This size distribution explained the relatively low annual averages of EAE at the five sites (all annual EAE in Fig. 2 are less than 1.0), compared to those at the inland urban and suburban sites in China (Xin et al., 2007), such as Beijing (1.19) (Fan et al., 2006), Nanjing (1.20) (Zhuang et al., 2017, 2018), Kunming (1.25) (Zhu et al., 2016) and Chengdu (1.09) (Che et al., 2015). In addition, spring was the season with a high volume concentration of coarse-mode aerosol. Among the five sites, the southernmost site, QOMS\_CAS, showed the highest annual mean EAE, and the size distribution was distinctly bimodal, especially in spring. This was also because of the frequent biomass burning activity in India and Nepal, which can transport the fine aerosol to the QOMS\_CAS site.

The annual averages of CE318\_AOD (shown in Fig. 2) were 0.05–0.14 over TP sites. These average values were lower than those in other regional background sites, such as Longfengshan (0.35) in northeast China (Wang et al., 2010), Xinglong (0.28) in the North China Plain (Zhu et al., 2014), Lin'an (0.89) in East China (Pan et al., 2010) and Dinghushan (0.91) in South China (Chen et al., 2014). The low aerosol loading over the five TP sites indicates excellent air quality over the TP region.

However, the aerosol loading at the TP sites presented interannual changes. The annual variations in CE318\_AOD and EAE over the TP at the four sites, i.e., Lhasa, Mt\_WLG, NAM\_CO and QOMS\_CAS, are shown in Fig. 4. The data for the CE318 observations at the Muztagh\_Ata site are only



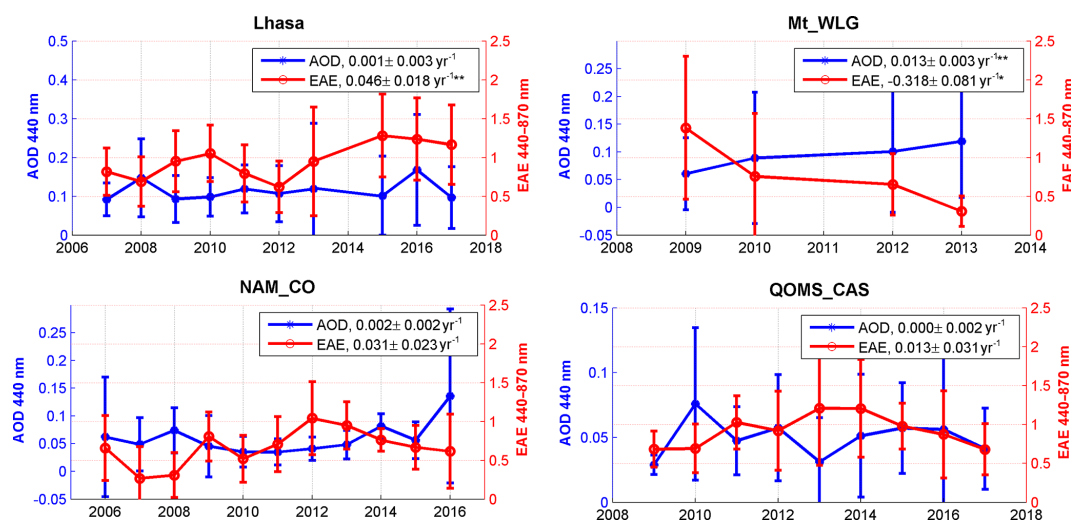
**Figure 3.** Seasonal variation in aerosol size distribution at the five sites located in the Tibetan Plateau.

available for 2010; thus, the annual variation at this site is not shown here. The annual CE318\_AOD showed increasing trends of  $0.001 \pm 0.003 \text{ yr}^{-1}$  at Lhasa,  $0.013 \pm 0.003 \text{ yr}^{-1}$  at Mt\_WLG and  $0.002 \pm 0.002 \text{ yr}^{-1}$  at NAM\_CO during the CE318 observation period. The Mt\_WLG site showed the most obvious increase in CE318\_AOD during 2009–2013. These results indicated an increase in aerosol loading at the three sites. The long-term annual variation of CE318\_AOD at QOMS\_CAS was very small ( $0.000 \pm 0.002 \text{ yr}^{-1}$ ), but there were still short-term annual variations (the values decreased from 2010 to 2013 and increased from 2013 to 2016). The annual trends of EAEs were more evident than the CE318\_AOD at these four sites. Most sites showed an increasing tendency in the average annual EAE except for the Mt\_WLG site, which showed a large decreasing trend of  $-0.318 \pm 0.081 \text{ yr}^{-1}$ . This showed that the size of aerosol at the Mt\_WLG site increased, while the size of the aerosol decreased in the other three sites. Looking at the CE318\_AOD and EAE values together, the positive trend of CE318\_AOD and the positive trend of EAE in the long-term variation at

most sites over the TP indicated the addition of fine-mode aerosol which may be related to the anthropogenic impact or long-distance transport of polluted dust to the TP. However, in the short term, the increase in the average annual CE318\_AOD was often associated with the decrease in EAE over the TP, which suggested the addition of coarse-mode aerosol during the CE318 observation period.

### 3.2 Aerosol properties from MODIS

Ground-based observations can offer accurate aerosol optical properties at point locations but lack spatial coverage. The MODIS aerosol product can provide the spatial variation in AOD over the TP. Thus, we evaluated the MODIS\_AOD using the ground-based observation CE318\_AOD at 550 nm over the TP sites. The CE318\_AOD at 550 nm was interpolated from 440, 675, 870 and 1020 nm by using an established fitting method from Ångström (1929). The matchup method was that the CE318 data within 1 h of the MODIS overpass were compared with the MODIS data within a

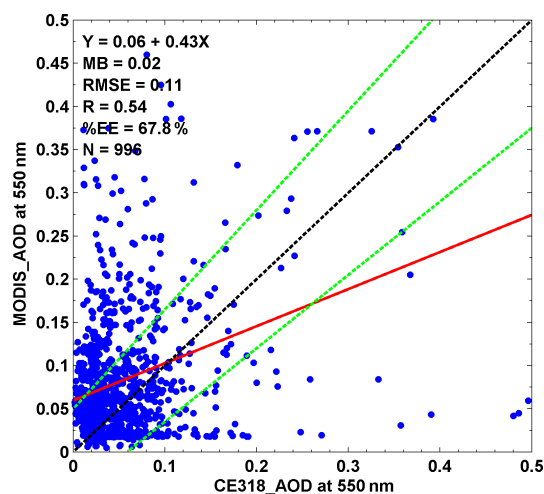


**Figure 4.** Annual averages of and trends in AOD and EAE from CE318 at four sites located in the Tibetan Plateau.

25 km radius of the ground-based site. The minimum requirement for a matchup was at least 3 pixels from MODIS.

Figure 5 shows the results of MODIS\_AOD compared to the collocated ground CE318 observations over the TP. There were 996 instantaneous matchups of Terra and Aqua MODIS during the CE318 instrument measurement period at the five TP sites. The MODIS\_AOD overestimated the AOD at 550 nm with a positive mean bias of 0.02 and a RMSE of 0.11. The RMSE value was lower than that of the North China Plain sites ( $\sim 0.25$ ) (Bilal et al., 2019). The slope and intercept of the best-fit equation between the MODIS\_AOD and CE318\_AOD at 550 nm were 0.46 and 0.06, respectively, with a correlation coefficient ( $R$ ) of 0.54. There were 67.8 % of the compared AODs within the expected error envelope of  $0.05 + 0.15 \text{ AOD}$  (% EE). The  $R$  value was lower than that in the global assessment statistics, while the % EE was higher than that in the global evaluation (Bilal and Qiu, 2018). Overall, the results suggest that the MODIS\_AOD product can be used to study the aerosol spatial variation over the TP region.

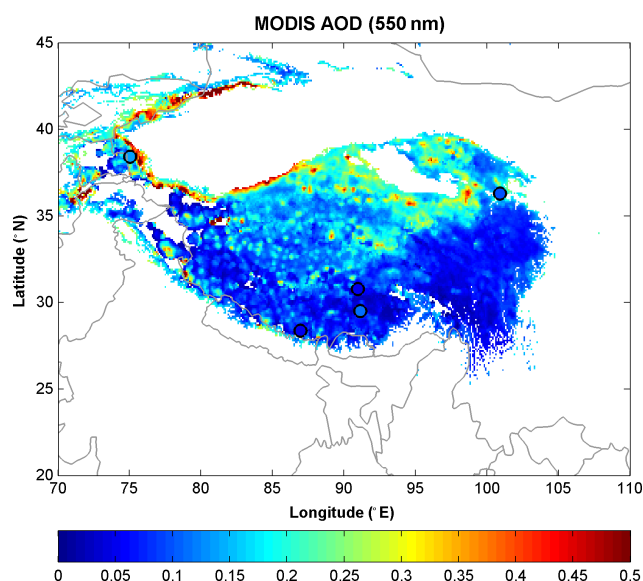
The spatial distribution of the annual MODIS\_AOD is shown in Fig. 6. The MODIS\_AOD agreed with the CE318\_AOD at 550 nm at the five TP sites. The northwest area around the Taklimakan Desert and the northern part of the TP on the transport path of the Taklimakan Desert dust showed high MODIS\_AOD ( $> 0.25$ ) in recent decades. In addition, the southern edge showed slightly high MODIS\_AOD ( $0.2\text{--}0.25$ ) influenced by the aerosol transport from South Asia. There were some small areas with high MODIS\_AOD ( $\sim 0.2$ ) in the center of the TP, and the southeast region showed low MODIS\_AOD ( $\sim 0.1$ ), which may be attributed to the aerosol transport and surface features such as vegetation cover, since there are few inhabitants. The seasonal departure of MODIS\_AOD (Fig. 7) showed that high positive MODIS\_AOD departure often appeared in spring, especially for the northwest edge, northern area and



**Figure 5.** Comparisons of the 550 nm AOD measured by the CE318 instrument (CE318\_AOD) over Tibetan Plateau stations with the MODIS retrieval Deep Blue and Dark Target combined AOD of 10 km spatial resolutions (MODIS\_AOD). The statistical parameters in this figure include the number of matchup data ( $N$ ), the slope and intercept at the y axis of linear regression (read line), the mean bias (MB), the root-mean-squared error (RMSE), the correlation coefficient ( $R$ ) and the percentage of data within the expected error  $0.05 + 0.15 \text{ AOD}$  (% EE) which is used as the MODIS AOD expected uncertainty over land (green lines).

southern edge of TP, which may be a result of aerosol transport from the frequent dust events in the Taklimakan Desert and the fire activities in South Asia in spring.

A linear regression analysis of the trends in annual MODIS\_AOD over the TP from 2006 to 2017 was conducted using the least-squares method. The spatial distribution of the annual trends in MODIS\_AOD during 2006–2017 is illustrated in Fig. 8. There were no statistically significant



**Figure 6.** Spatial distribution of MODIS\_AOD at 550 nm over the Tibetan Plateau (only the altitude > 3000 m) during 2006–2017. The color-filled circles are the CE318 observation AOD averages at TP sites.

trends in most areas during 2006–2017. The MODIS\_AOD showed negative trends in the northwest edge close to the Taklimakan Desert and to the east of the Qaidam Basin and slightly positive trends in most of the other areas. The areas where MODIS\_AOD decreased were mainly located near the desert or on the transport path of the desert dust. This descending trend may be related to the significant reduction in dust emissions caused by the decline in wind speed in recent years (Yang et al., 2017b). The positive trend in most other areas may be due to the rapid increase in human activities, such as the expansion of tourism to the TP and biomass burning in South Asia.

The seasonal trends in MODIS\_AOD at 550 nm over the TP during 2006–2017 are presented in Fig. 9. The spring showed the most obvious decline in MODIS\_AOD ( $\sim 0.02 \text{ yr}^{-1}$ ) at the northern edges and northeast part of the TP during 2006–2017, which may also be related to the reduction in dust emissions as the trend in the annual MODIS\_AOD (seen in Fig. 8). In summer, the positive trend in MODIS\_AOD over the TP was relatively apparent, and most sporadic higher positive values of  $\sim 0.01$  occurred in the central and southern part of the TP. Summer is the tourist season in the TP, and tourism has developed in past decades, which may be one of the reasons for the higher positive trend in summer in the TP. The positive trends in autumn and winter were relatively lower than those in summer, and the most positive trends were located at the northern TP. The reason for this phenomenon needs to be explored.

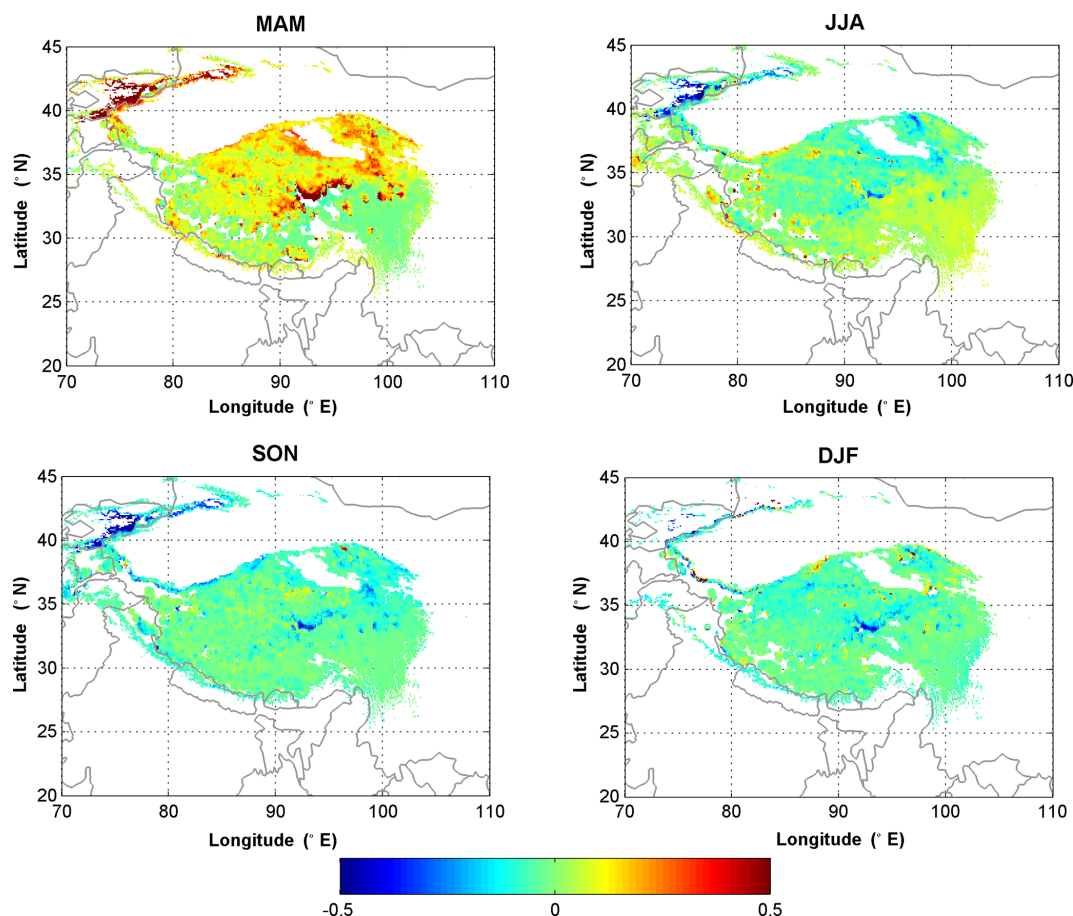
#### 4 Aerosol properties and potential sources during high aerosol loading

The annual mean AOD in the TP was normally low due to the few human inhabitants and high altitude. However, some high CE318\_AOD values larger than 0.4, which is normally regarded as high aerosol loading (Eck et al., 2010; Giles et al., 2012), were observed at the five sites in the TP by CE318. Thus, the CE318\_AOD larger than 0.4 over TP can be considered the aerosol pollution. The frequencies of high aerosol loading (CE318\_AOD > 0.4) during the CE318 measurements were 1.57 %, 1.79 %, 0.21 %, 0.42 % and 0.11 % at the Lhasa, Mt\_WLG, Muztagh\_Ata, NAM\_CO and QOMS\_CAS sites, respectively. The aerosol properties and sources during high aerosol loading in the TP need to be studied.

Figure 10 shows the CE318\_AOD with values larger than 0.4 versus EAE observed by CE318 at the five sites in the TP. Except for the Lhasa and Mt\_WLG sites, almost all values of CE318\_AOD were less than 1.0, which reflected the relatively clear environment over the TP. The EAE showed two centers at  $\sim 0.1$  and  $\sim 1.5$ . The low EAE ( $\sim 0.1$ ) center was related to dust events, which can cause higher concentrations of coarse particles in the atmosphere. Besides, most values of the low EAE ( $< 0.5$ ) area were less than 0.2 (only a few EAEs between 0.2 and 0.5 were observed at Lhasa and Mt\_WLG), indicating that the pure-dust-type aerosols were more common than the polluted-dust-type aerosols in the TP according to Eck et al. (2010). The high EAE center at  $\sim 1.5$  indicated mainly small sub-micron radius particles, which can be attributed to anthropogenic emissions. The values of EAE > 1.0 at the NAM\_CO and QOMS\_CAS sites were generally higher than those at the Lhasa and Mt\_WLG sites. According to past studies, the EAE of biomass burning aerosol is generally higher than the urban/industry aerosol (Giles et al., 2012; Eck et al., 2010), which may cause the higher EAE at NAM\_CO and QOMS\_CAS (more biomass burning aerosol) than at Lhasa and Mt\_WLG (more urban/industry aerosol) for the high EAE area. On the other hand, the values in the middle range of 0.5–1.0 were rare, indicating a smaller mix of natural and human sources. The percentage of EAE bins to the number of CE318\_AOD > 0.4 was distinct (Table 3). The percentage of EAE < 0.5 was higher than that of EAE > 1.0 at Lhasa, Mt\_WLG and Muztagh\_Ata, indicating more nature dust pollution than the anthropogenic pollution at these three sites. However, a greater number of high EAE values (> 1.0) were observed than EAE < 0.5 values at the NAM\_CO and QOMS\_CAS sites, suggesting that anthropogenic pollution was more than natural dust pollution at these two sites.

Figure 11 shows the aerosol size distribution binned by CE318\_AOD at the five sites in the TP. The volume concentration of coarse-mode particles increased more apparently than fine mode at Lhasa, Mt\_WLG and Muztagh sites when the values of CE318\_AOD increased. However, the size dis-





**Figure 7.** The seasonal departure of MODIS\_AOD over the Tibetan Plateau (altitude > 3000 m).

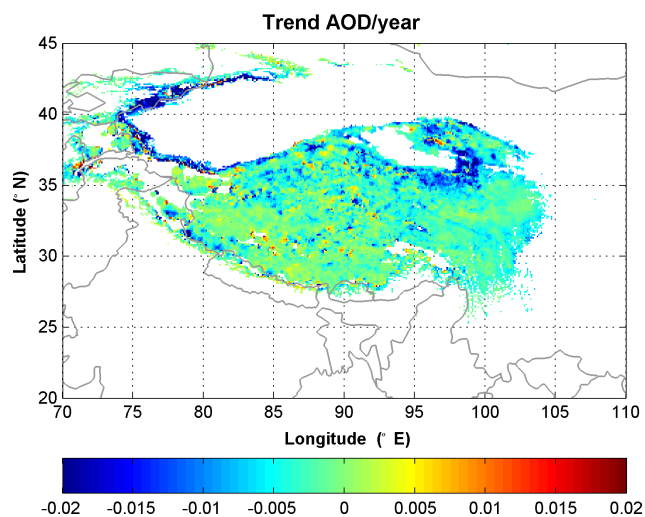
**Table 3.** The percentages of EAE < 0.5, 0.5–1.0, and > 1.0 for high AOD observations at the five sites.

Site	<i>N</i> of AOD > 0.4	% EAE < 0.5/ <i>N</i>	% 0.5 < EAE < 1.0/ <i>N</i>	% EAE > 1.0/ <i>N</i>
Lhasa	655	60.6	3.4	36.0
Mt_WLG	290	73.4	0	26.6
Muztagh_Ata	5	100	0	0
NAM_CO	140	27.9	2.8	69.3
QOMS_CAS	59	23.7	0	76.3

tribution at NAM\_CO and QOMS\_CAS showed the dominant increase in fine-mode aerosol. These indicate the different aerosol pollution type in these five sites: dust dominant in Lhasa, Mt\_WLG and Muztagh\_Ata and fine-mode aerosol (mainly biomass burning aerosol) pollution dominant at NAM\_CO and QOMS\_CAS.

The dominant aerosol pollution type showed obvious distinctions among the five sites on the TP, so what was the distinct aerosol pollution source at each site? We used the HYSPLIT back-trajectory model and the MODIS\_AOD on the day with aerosol pollution (CE318\_AOD > 0.4) to show the aerosol source for the pollution day at each site. Figure 12 shows the 72 h back trajectories ending at the five sites

(10 m a.g.l.) in the TP overlaid with the mean MODIS\_AOD on the aerosol pollution day which was observed by the ground-based CE318 (CE318\_AOD > 0.4). The CE318 instruments observed 78, 20, 2, 15 and 14 d at Lhasa, Mt\_WLG, Muztagh\_Ata, NAM\_CO and QOMS\_CAS, respectively, with instantaneous CE318\_AOD > 0.4. The aerosol pollution days at Lhasa, Mt\_WLG and Muztagh\_Ata observed by CE318 often had low EAE (black lines). The airflows ending at the Lhasa site on the polluted days were mainly from the northwest and southwest. The MODIS\_AOD around Lhasa in the area of the back trajectories with CE318 EAE < 0.5 passing did not show significantly high values, especially in the Taklimakan Desert,



**Figure 8.** Trend in the MODIS\_AOD at 550 nm during 2006–2017.

which indicated that the dust pollution at Lhasa was mainly from local or surrounding dust events rather than transport from the Taklimakan Desert. The Mt\_WLG showed that the air mass on the pollution days comes from the west and east and the path of back trajectories had high MODIS\_AOD. The high values of MODIS\_AOD showed two transport paths of dust aerosol to Mt\_WLG: one was through the Qaidam Basin and the other was through the northeast edge of the TP. The two polluted days observed by CE318 at the Muztagh\_Ata showed the easterly airflows originating from the Taklimakan Desert. The direction of the back trajectories of EAE < 0.5 that ended at NAM\_CO was similar to Lhasa, while the southerly air flows with high EAE (red trajectories) originated from Nepal, where frequent biomass burning happened and caused the high MODIS\_AOD values. The trajectories ending at QOMS\_CAS and the high MODIS\_AOD of the path revealed the transport of finer aerosol from South Asia to this site.

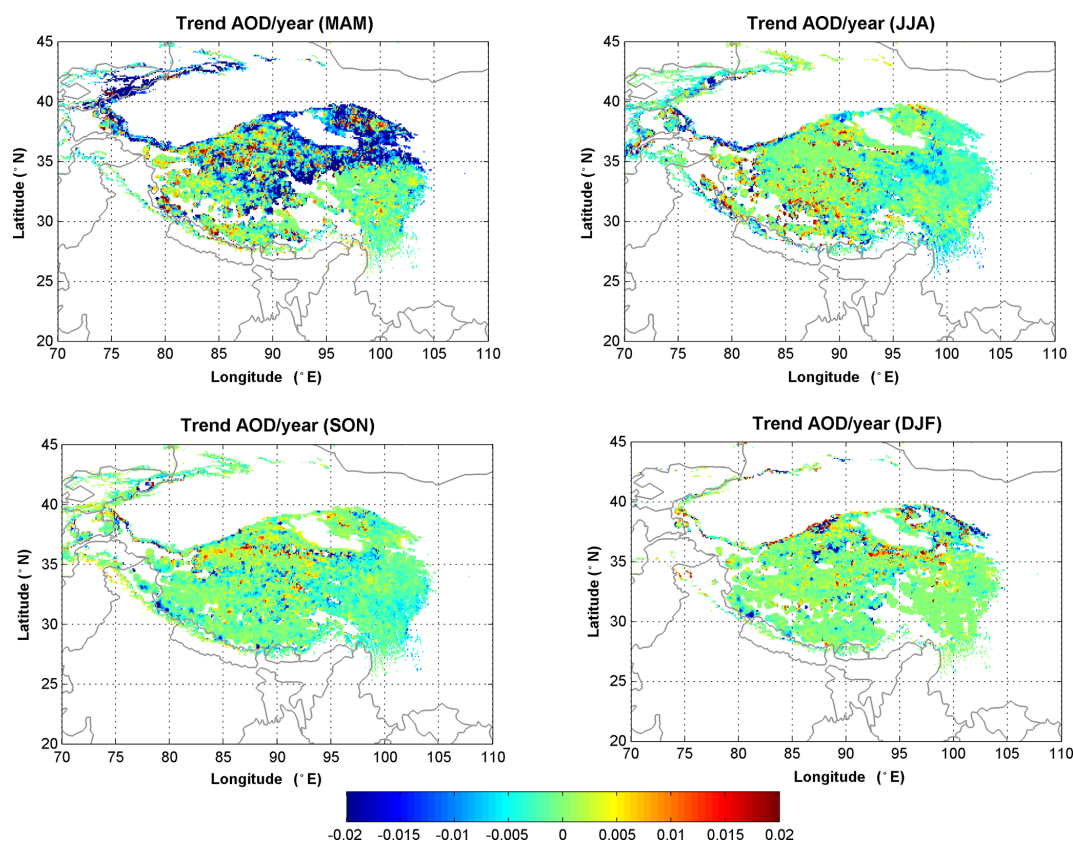
## 5 Case study of long-range transport to the TP

The long-range transport of aerosol can cause the aerosol pollution and affect the long-term variation in aerosol over the TP. In addition, the dominant aerosol type may change at the TP sites during a case of aerosol transport. Thus, a specific case of aerosol pollution during 27 April–3 May 2016 was analyzed further. This case was selected based on the observations from the CE318 instruments. During 28 April–1 May, the instantaneous CE318\_AOD at the Lhasa, NAM\_CO and QOMS\_CAS sites showed the values larger than 0.4, which reached up to more than 3 times the mean values of CE318\_AOD of each site (0.11 at Lhasa, 0.05 at NAM\_CO and QOMS\_CAS). This was indicative of aerosol pollution at the three sites. Then, how about the aerosol prop-

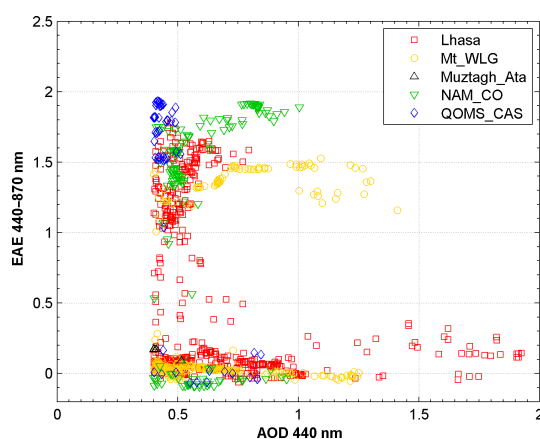
erties of this period and where did the polluted aerosol originate?

Figure 13 shows the daily CE318\_AOD and EAE during 27 April–3 May at the three sites. The mean values of CE318\_AOD were 0.45, 0.38 and 0.23 at Lhasa, NAM\_CO and QOMS\_CAS, respectively. These even reached 4 times the annual mean CE318\_AOD at each site. The mean EAEs were 0.98, 1.22 and 1.44 at Lhasa, NAM\_CO and QOMS\_CAS, respectively, which were higher than the annual averages and suggested the arrival of fine aerosols. There were CE318\_AOD peaks at the three sites during 27 April–3 May. Lhasa showed an increase in CE318\_AOD from 0.30 on 27 April to 0.51 on 28 April and maintained a high CE318\_AOD to a value of 0.54 on 1 May, after which it decreased to 0.34 on 2 May. NAM\_CO also showed an increase in CE318\_AOD during the first 2 d of the period but decreased after 29 April. QOMS\_CAM showed a slight increase in CE318\_AOD from 27 April to 30 April, which was later than those of the other two sites. Combining the EAE on these days, fine-mode aerosol was brought to Lhasa and NAM\_CO during 27–29 April, and then coarse aerosol began to occur on 30 April and even became the dominant aerosol in the following several days. The fine aerosol at the QOMS\_CAM site was maintained for an additional day after those at the two sites, and then the coarse aerosol increased.

The GEOS-Chem model simulation also supported the above results. Figure 14 shows the comparison between the Model\_AOD ( $0.5^\circ \times 0.667^\circ$ ) and CE318\_AOD at 550 nm and the ratios of the model-simulated aerosol types (dust, both organic carbon (OC) and black carbon (BC) aerosol) to the total Model\_AOD during this case period at the three sites. The evaluation results showed that the model underestimated the daily AOD at the three sites during this period, with negative mean biases from  $-0.28$  to  $-0.08$ . However, the Model\_AOD was relatively highly correlated with the CE318\_AOD at 550 nm, with the  $R$  values of 0.61 at Lhasa, 0.89 at NAM\_CO and 0.86 at QOMS\_CAS. These  $R$  values were higher than the model evaluation in South China and the Indo-China Plain ( $\sim 0.5$ ) (Zhu et al., 2017). Thus, the variation trend from Model\_AOD agreed well with that measured by the CE318 instruments during these days. During the first 4 d of the case period (27 April to 30 April), the ratios of different aerosols to the total Model\_AOD showed that the sums of OC and BC aerosols were higher than those of dust aerosol at all three sites. Besides, the sums of OC and BC at Lhasa and QOMS\_CAS were higher than that of NAM\_CO. These indicated that the smoke aerosol affected the three sites more severely than dust during the first 4 d, and Lhasa and QOMS\_CAS sites were nearer to smoke sources than NAM\_CO. After 30 April, the sum of BC and OC decreased while dust increased, and the increase in dust at the three sites was NAM\_CO > Lhasa > QOMS\_CAS. Therefore, the major aerosol source was changed and the NAM\_CO site was closer to the dust source after 30 April. This phenomenon had continued until 2 May at NAM\_CO and Lhasa and until



**Figure 9.** Trends in the MODIS\_AOD at 550 nm during 2006–2017 in each season.



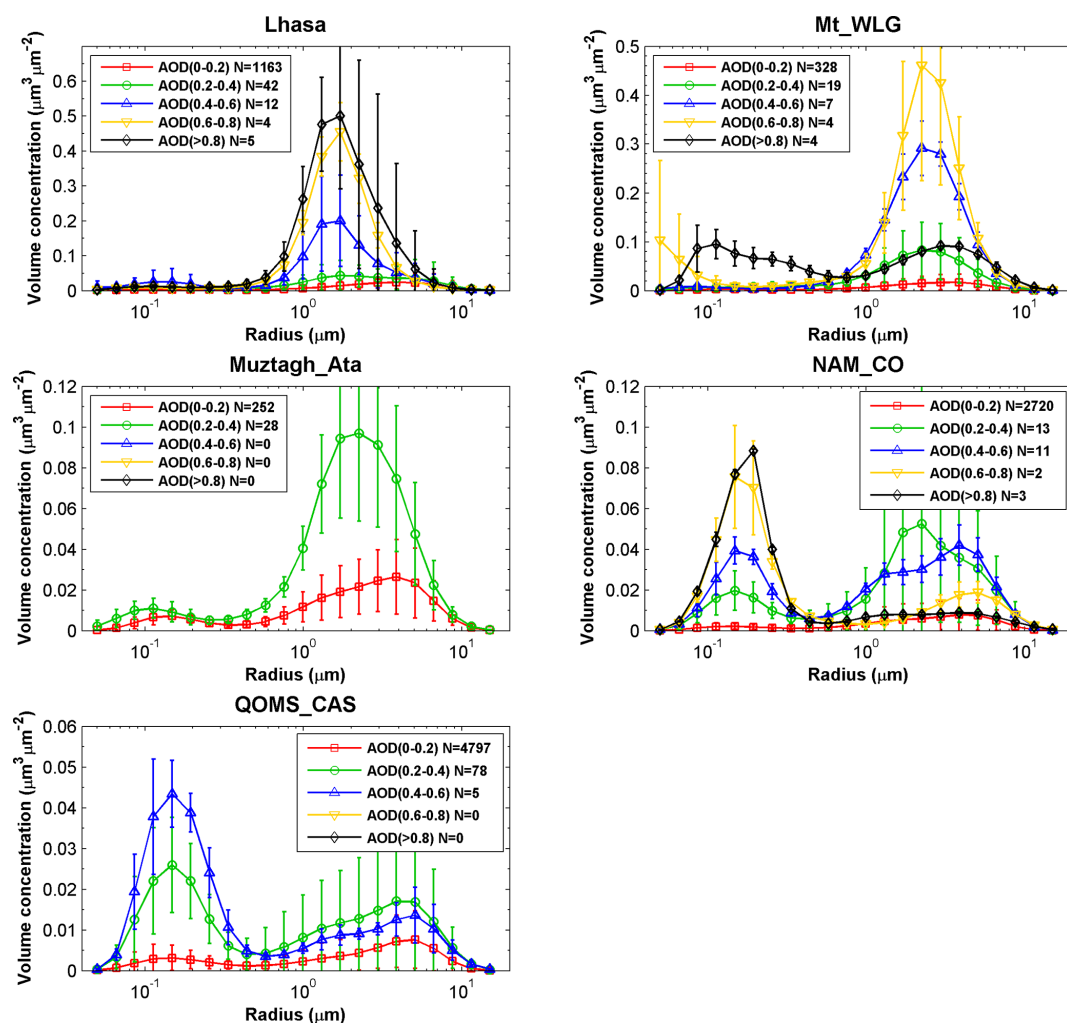
**Figure 10.** AOD vs. EAE (only CE318\_AOD at 440 nm > 0.4 was considered) observed by CE318 at the five sites on the Tibetan Plateau.

1 May at QOMS\_CAS. In the last 1 or 2 d, the dust decreased while the BC and OC obviously increased, which could cause the mixture of different aerosols.

Then, how was the spatial aerosol loading around the TP and the vertical feature of aerosol transported to the TP? Figure 15 shows the MODIS\_AOD and 72 h back trajectories

at Lhasa (the first row); the CALIOP-derived vertical profile of total attenuated backscatter at 532 nm (the second row); and the vertical feature mask of aerosol (the third row) on 28 April, 1 May and 3 May during the case study period. The MODIS\_AOD showed high values in the south (South Asia) and north (Taklimakan Desert) on the 3 d. The high values in South Asia were caused by anthropogenic aerosols (such as biomass burning) or dust polluted by anthropogenic aerosols, while the high MODIS\_AOD in the Taklimakan Desert resulted from dust. The values and areas of the high MODIS\_AOD in South Asia and the Taklimakan Desert on 1 and 3 May were higher and larger than those on 28 April. The back trajectories ending at Lhasa on the 3 d were different. On 28 April, the air flows originated from the southwest (South Asia region). However, the air masses on 1 and 3 May were from the northwest (Taklimakan Desert).

The CALIPSO ground tracks across the TP and through South Asia and the Taklimakan Desert were chosen to show the aerosol transport to the TP sites. On 28 April, the Level 1 attenuated backscatter at 532 nm derived from CALIOP (the second row) showed apparent aerosol layers in the southern area (Bhutan and northeast India), and this aerosol layer even extended to an altitude of  $\sim 10$  km over the TP along the southern slope of the TP. On 1 May, the CALIOP attenuated backscatter not only showed the deep aerosol layers south



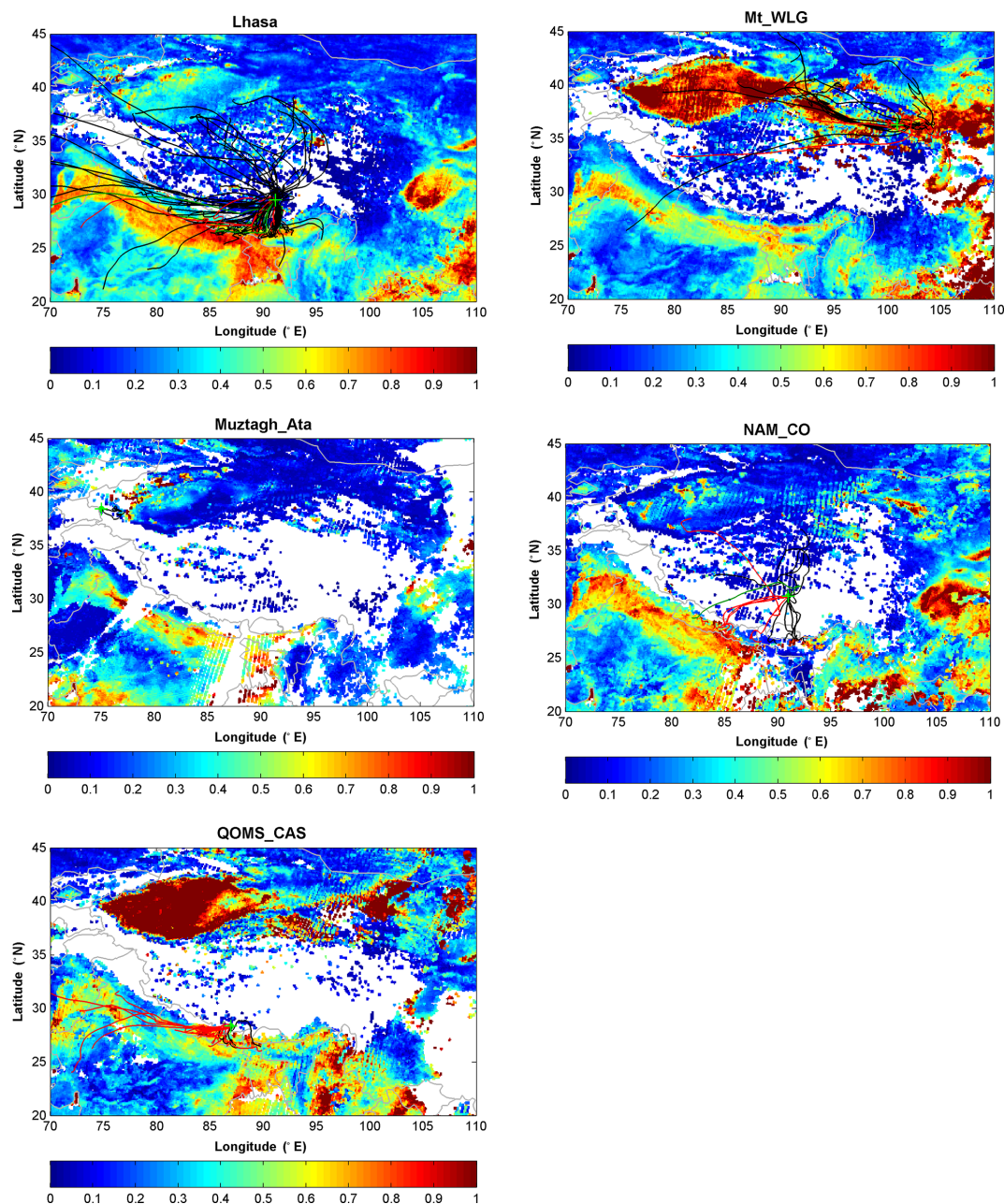
**Figure 11.** Aerosol size distribution binned by CE318\_AOD at the five sites on the Tibetan Plateau.

of the TP but also showed stronger aerosol layers north of the TP (Taklimakan Desert area). Besides, the north aerosol layers also climbed into the air over the TP but not as high as the southern aerosol layer. On 3 May, there were also aerosol layers in the south and north of the TP that were both transported to above the TP, but the aerosol loading over the TP was lower than that on 28 April and 1 May (the values of attenuated backscatter on 3 May were lower), which corresponded to the lower CE318\_AOD on this day than those on 28 April and 1 May at the three TP sites (Fig. 13).

The vertical feature mask of the aerosol from CALIOP (the third row) showed the aerosol types on the 3 d. On 28 April, the aerosol layer in the north ( $\sim 35^\circ$  N) and above the TP was mainly the smoke aerosol and was even near to 10 km. The back trajectories ending at Lhasa also showed that the southern airflow brought the smoke aerosol and polluted dust from South Asia to the center of the TP. On 1 May, the aerosol layer on the southern slope of the TP was also smoke aerosol and polluted dust, while the aerosol layers

in the northern TP and above the TP were almost all dust aerosol, which could be explained by the northwest airflows carrying the dust aerosol from the Taklimakan Desert. These may be the result of the lower EAE values at Lhasa and NAM\_CO than that at QOMS\_CAM (Fig. 13). On 3 May, the aerosol type above the central TP and the southern TP was occupied by polluted dust aerosol, and the EAE at NAM\_CO and QOMS\_CAM also showed a slight increase on 3 May. These results agree with the aerosol simulation from GEOS-Chem. Jia et al. (2015) has shown that the dust from India polluted by anthropogenic aerosols can be transported to the TP, but the back trajectories on 1 and 3 May illustrated that the airflows that ended at Lhasa were from the north or northwest rather than the south, indicating that the polluted dust over the TP on 3 May was more likely the mixing result of dust and smoke aerosol. In addition, the lengths of the back trajectories (especially the back trajectories at 10 m and 500 m a.g.l.) on 1 May showed that the airflows moved slowly, which allowed the possibility of aerosol mixture over





**Figure 12.** The back trajectories ending at the five sites (10 m a.g.l.) on the Tibetan Plateau overlaid with the mean MODIS\_AOD at 550 nm on the high aerosol loading day observed by ground-based CE318 ( $\text{CE318\_AOD} > 0.4$ ). Red stands for  $\text{EAE} > 1.0$ , black for  $\text{EAE} < 0.5$ , and green for  $\text{EAE}$  within 0.5–1.0.

the TP. The observations and model simulations illustrated the following scene: first, the smoke aerosol in South Asia was lifted up to 10 km, contaminating the TP sites, and transported to the center of the TP; then, the dust from the Taklimakan Desert could climb the north slope of the TP and be transported to the TP; finally, the dust and smoke aerosol over the TP were mixed. This case of aerosol pollution shows that the anthropogenic aerosols (mainly smoke) in South Asia and dust in the Taklimakan Desert could be transported to the

center of the TP and they both can even cause mixed aerosol pollution above the TP. The past case studies of aerosol transport to the TP are mostly individual dust or smoke aerosol, while this case of aerosol pollution over the TP showed the mixing pollution during the last 2 d of the case period.



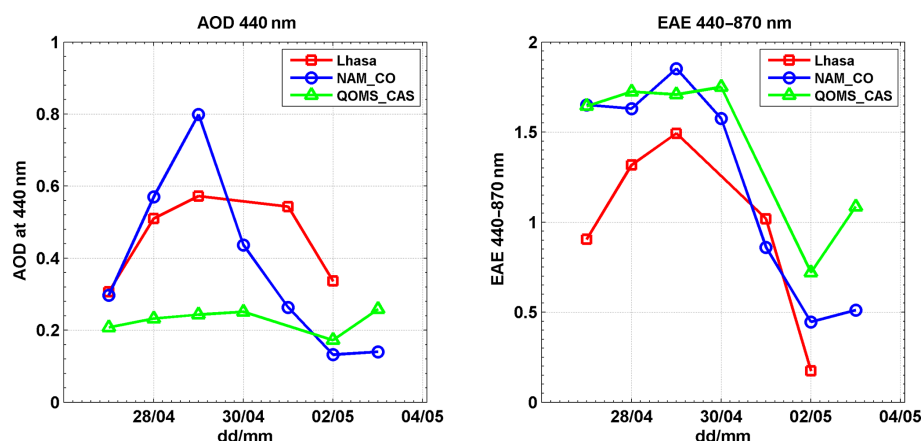


Figure 13. CE318-observed daily AOD and EAE during 27 April–3 May 2016 at Lhasa, NAM\_CO and QOMS\_CAS.

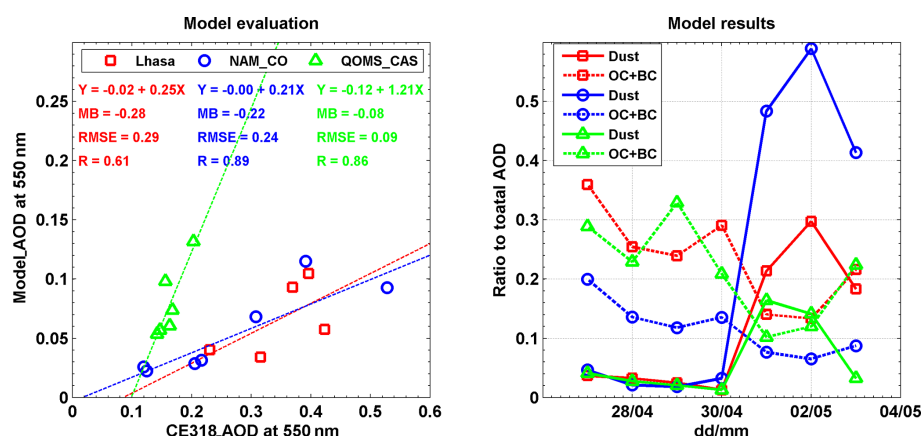


Figure 14. The model evaluation of the GEOS-Chem model simulated the daily average AOD (Model\_AOD) by using the CE318-observed daily AOD (CE318\_AOD) at 550 nm and the model results of the ratios of dust or organic carbon (OC) and black carbon (BC) aerosol to the total AOD during 27 April–3 May 2016 at Lhasa, NAM\_CO and QOMS\_CAS. The statistical parameters used in the modal evaluation are the same as those in Fig. 5.

## 6 Conclusion

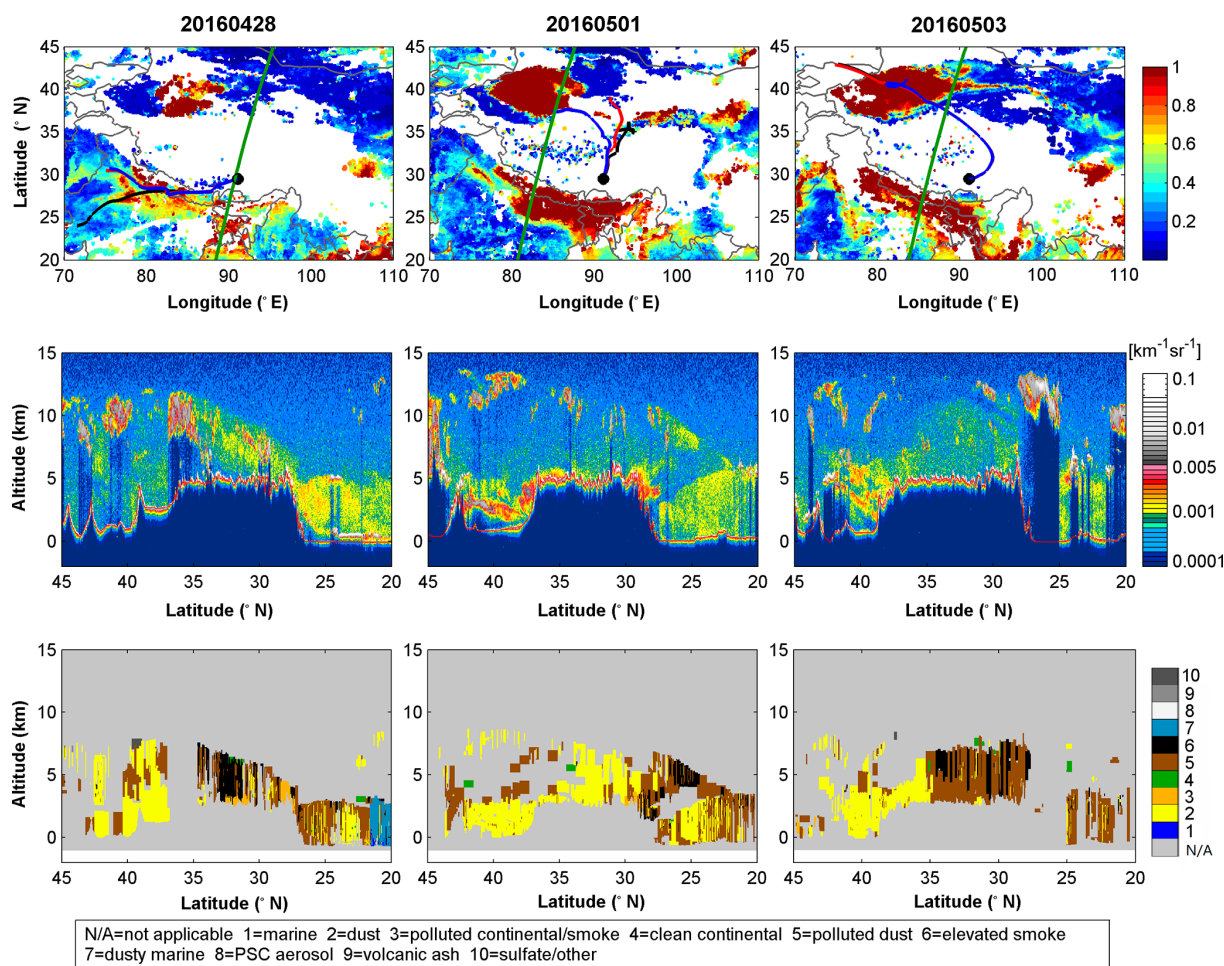
The long-term spatiotemporal variations in the aerosol optical properties and the impacts of the long-range aerosol transport over the TP were analyzed by using a combination of ground-based and satellite remote-sensing aerosol products as well as model simulations. The major conclusions are drawn as follows:

1. The annual CE318\_AOD at most TP sites showed increasing trends ( $0\text{--}0.013\text{ yr}^{-1}$ ) during the past decade. Increasing tendencies in the annual averaged EAE were also found at most TP sites. Spatially, the MODIS\_AOD showed negative trends in the northwest edge close to the Taklimakan Desert and the east of Qaidam Basin and slightly positive trends in most of the other areas of the TP.
2. Different aerosol types and sources contributed to the high aerosol loading at the five sites: dust was domi-

nant in Lhasa, Mt\_WLG and Muztagh with sources in the Taklimakan Desert, but fine-aerosol pollution was dominant at NAM\_CO and QOMS\_CAS with transport from South Asia.

3. A case of smoke followed by dust pollution at Lhasa, NAM\_CO and QOMS\_CAS during 28 April–3 May 2016 showed that the smoke aerosol in South Asia was first uplifted to 10 km and transported to the center of TP. Then, the dust from the Taklimakan Desert could climb the northern slope of the TP and be transported to the TP, allowing the dust and smoke aerosol over the TP to mix.

There are some limitations to this study. First, ground-based remote-sensing and MODIS\_AOD products may have had missing data due to cloud interference. Second, only half of a year of observations at the Muztagh\_Ata station may not be sufficient to fully reveal pollution days in the northwest TP region, which could have affected the statistics to



**Figure 15.** The MODIS\_AOD at 550 nm and 72 h back trajectories ending at Lhasa at three heights above the ground level (10 m in black, 500 m in red and 1000 m in blue lines) (the first row); the CALIOP-derived vertical profile of total attenuated backscatter at 532 nm (the second row); and the vertical feature mask of aerosol on 28 April, 1 May and 3 May 2016 (the third row). The green lines in the panels of the first row stand for the CALIOP ground tracks.

some extent. More long-term in situ observations are needed in the TP. However, due to the remoteness and challenging weather conditions over the plateau, maintaining long-term in situ observation stations over the TP is very difficult. The numerical model simulation is more practically feasible to study the aerosol properties over the TP, but the model accuracy is far from ideal over the TP. Thus, long-term numerical model simulation coupled with satellite observations and intensive short-term field campaigns should be used to analyze the aerosol properties over the TP in the future.

**Data availability.** The data for the four sites (Mt\_WLG, Muztagh\_Ata, NAM\_CO and QOMS\_CAS) are available from the AERONET website (<https://aeronet.gsfc.nasa.gov/>, last access: 30 November 2019). The dataset of Lhasa used in the study can be requested by contacting the corresponding author. The MODIS aerosol products are available from <http://ladsweb.nascom.nasa.gov> (last access: 30 November 2019). The HYSPLIT model and meteo-

rological fields' data can be from <https://www.arl.noaa.gov/hysplit/> (last access: 30 November 2019). The CALIPSO data are from <https://eosweb.larc.nasa.gov> (last access: 30 November 2019). The GEOS-Chem model code and share data can be obtained from <http://wiki.seas.harvard.edu/geos-chem> (last access: 30 November 2019).

**Author contributions.** All authors helped to shape the ideas and review this paper. JZ, XX and HC designed and wrote the manuscript; JZ, XX, HC and JW helped to analyze the data; HC, XZ, SK and ZC carried out the sun photometer observations; and JW, ZC, SK, TZ, XY and YZ provided constructive comments on this study.

**Competing interests.** The authors declare that they have no conflict of interest.

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