Background aerosol over the Himalayas and Tibetan Plateau: observed characteristics of aerosol mass loading

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Abstract

To investigate the atmospheric aerosols of the Himalayas and Tibetan Plateau (HTP), an observation network was established within the region’s various ecosystems, including at the Ngari, Qomolangma (QOMS), Nam Co, and Southeastern Tibetan (SET) stations. In this paper we illustrate aerosol mass loadings by integrating in situ measurements with satellite and ground-based remote sensing datasets for the 2011-2013 period, on both local and large scales. Mass concentrations of these surface atmospheric aerosols were relatively low and varied with land cover, showing a general tendency of Ngari and QOMS (barren sites) > Nam Co (grassland site) > SET (forest site). Daily averages of online PM$_{2.5}$ (particulates with aerodynamic diameters below 2.5 μm) at these sites were sequentially 18.2±8.9, 14.5±7.4, 11.9±4.9 and 11.7±4.7 μg m$^{-3}$. Correspondingly, the ratios of PM$_{2.5}$ to total suspended particles (TSP) were 27.4±6.65%, 22.3±10.9%, 37.3±11.1% and 54.4±6.72%. Bimodal mass distributions of size-segregated particles were found at all sites, with a relatively small peak in accumulation mode and a more notable peak in coarse mode. Diurnal variations in fine aerosol masses generally displayed a bi-peak pattern at the QOMS, Nam Co and SET stations and a single-peak pattern at the Ngari station, controlled by the effects of local geomorphology, mountain-valley breeze circulation and aerosol emissions. Mineral content in PM$_{2.1}$ samples gave fractions of 26% at the Ngari station and 29% at the QOMS station, or ~2-3 times that of reported results at human-influenced sites. Furthermore, observed evidence confirmed the existence of the aerodynamic conditions necessary for the uplift of fine particles from a barren land surface. Combining surface aerosol data and atmospheric-column aerosol optical properties, the TSP mass and aerosol optical depth (AOD) of the Multi-angle...
Imaging Spectroradiometer (MISR) generally decreased as land cover changed from barren to forest, in inverse relation to the PM$_{2.5}$ ratios. The seasonality of aerosol mass parameters was land-cover dependent. Over forest and grassland areas, TSP mass, PM$_{2.5}$ mass, MISR-AOD and fine-mode AOD were higher in spring and summer, followed by relatively lower values in autumn and winter. At the barren site (the QOMS station), there were inconsistent seasonal variations between surface TSP mass (PM$_{2.5}$ mass) and atmospheric column AOD (fine-mode AOD). Our findings implicate that, HTP aerosol masses (especially their regional characteristics and fine particle emissions) need to be treated sensitively in relation to any assessments of their climatic effect and potential role as cloud condensation nuclei and ice nuclei.
1 Introduction

Atmospheric aerosols undergo changes in their microphysical, chemical and optical properties, especially in high-altitude mountain regions. These changes primarily determine their roles in modifying regional climate, cryosphere and hydrology. This is particularly true for the Himalayas and Tibetan Plateau (HTP) region, which is surrounded by Asian dust and strong anthropogenic emissions. These natural and human-originated airborne chemicals, such as light-absorbing materials, reactive nitrogen and heavy metals can exert impacts on regional monsoon rainfall (e.g., Ramanathan et al., 2005; Lau et al., 2006), snow/ice albedo (e.g. Ming et al., 2008; Xu et al., 2009; Qu et al., 2014), nitrogen deposition (Liu et al., 2013; Liu et al., 2015) and meltwater composition (e.g., Zhang et al., 2014). Although these effects remain poorly understood, it is of first-order importance to characterize these remote atmospheric aerosols.

In the HTP, aerosol optical properties and chemical compositions have been observed almost entirely at a few specific sites. Ground-based measurements have focused on the relatively small concentrations of fine particles and total suspended particle (TSP) in the HTP’s atmospheric surface layer (Zhao et al., 2013; Xu et al., 2014). Satellite and ground-based remote sensing have also been employed and have pointed to a low aerosol optical depth (AOD) in this region (Xia et al., 2008; Xia et al., 2011; Yan et al., 2015). Mineral dust has been identified as one of the main aerosol components in the central Himalayas (Decesari et al., 2010) and the central TP (Zhang et al., 2001; Cong et al., 2007; Kang et al., 2016). Analysis of dust plumes from the surrounding deserts (the Taklimakan, Gobi and Southwest Asian deserts) and itself has indicated some potential source areas of
atmospheric particulates (Huang et al., 2007; Liu et al., 2008; Xia et al., 2008). However, these results have revealed only the somewhat pristine characteristics of HTP aerosols, dependent largely upon a significant understanding of mineral dust. Much uncertainty remains over the correct evaluation of aerosol sources, transportation and deposition, especially in relation to a much wider variety of aerosol species. Furthermore, the mountains produce extensive mountain-valley breezes, and alpine glacier/snow and stratosphere-troposphere exchanges. These conditions could in turn affect aerosol properties via transportation and chemical processes by facilitating the upward diffusion of aerosol matters (Decesari et al., 2010; Cong et al., 2015) and by changing the oxidizing capacity of the troposphere (Lin et al., 2008). Hence there are additional obstacles in understanding HTP atmospheric aerosols.

Anthropogenic emissions into this region occur occasionally and are dependent on local/regional atmospheric dynamics. During the pre-monsoon period, “Atmospheric Brown Cloud” stacks up in the southern foothills of the Himalayas (Ramanathan et al., 2001). Mountain-valley breeze circulations allow these aerosols to spread upslope and then can enhance the concentrations of carbonaceous and inorganic matters in fine aerosols over the Himalayas (Decesari et al., 2010; Babu et al., 2011; Cong et al., 2015; Lüthi et al., 2015). Additionally, the South Asian summer monsoon system is one of the important atmospheric dynamics in the transportation of pollutants to the HTP region from southern and southeastern Asia (Liu et al., 2013; Sheng et al., 2013). Consequently, light-absorbing substances (such as black carbon) have received special attention. Studies have raised the hypothesis that a suppression of the Southern Asian monsoon through a weakening of the meridional surface
temperature gradient (Ramanathan et al., 2005), is likely to enhance regional monsoonal rainfall in northern India, the Himalayas, and the southern Tibetan Plateau (TP) through the “elevated-heat-pump” effect (Lau et al., 2006). Further, the post-depositional effect of decreasing snow/ice albedo is likely to lead to reductions in the HTP glaciers (Ming et al., 2008; Xu et al., 2009; Qu et al., 2014). However, the validity of the above hypothesis strongly depends on the characteristics and spatial-temporal variations in these particles (principally in mass loadings, chemical compositions, size distributions and optical properties), and their related atmospheric processes.

In general, the HTP, as a unique upland region where the relatively pristine tropospheric environment is juxtaposed with Asian anthropogenic emissions, is highly suitable for the study of background atmospheric aerosols and the interactions between natural and anthropogenic emissions, processes which may have far-reaching environmental and climatic consequences (Lawrence, 2011; Vernier et al., 2011).

It is imperative, therefore, that the first comprehensive observation of HTP atmospheric background aerosols be conducted during the 2011-2013 period, basing on four stations located in different ecosystems. Accordingly, we present in this study online PM$_{2.5}$ (particles with aerodynamic diameters ≤2.5 μm) concentrations and filter-sampled particles, as well as the size distributions of these size-segregated particles (Section 3.1). The diurnal variations in fine aerosol masses are also discussed with particular reference to local geomorphology, source emissions and meteorological settings (Section 3.2). As part of our research, we attempted to integrate these in situ observations with aerosol optical properties derived from both ground-based and satellite remote sensing, aiming to construct a topographical view of
their spatial and seasonal patterns (Section 3.3).

2 Materials and Methods

2.1 Monitoring sites and the regional environment

The HTP is the greatest upland region of the Eurasian continent in the Northern Hemisphere’s middle-low latitudes, and composes landscapes covered mainly by alpine forest, grassland/meadow, barren areas and patchy glacier/snow cover. We take ‘upland’ in the HTP region to be land above 2800 m asl; if so, this region has an upland area of ~5,000,000 km² (Fig. 1a). Four comprehensive observation platforms were established within different landscapes, including the Ngari station (79°42′E, 33°23′N, 4,264 m asl), the Qomolangma/Everest (QOMS) station (86°57′E, 28°21′N, 4,300 m asl), the Nam Co station (90°57′E, 30°46′N, 4,746 m asl), and the SouthEastern Tibet (SET) station (94°44′E, 29°46′N, 3,326 m asl) (Figs. 1 and S1). The high-altitude, inland topography produces a generally cold, arid and windy climate across most of the HTP. Additionally, the atmospheric circulation systems (including the South Asian Monsoon, the East Asian Monsoon, and the Westerlies) control the seasonal and spatial variations in precipitation patterns, i.e., winter-spring precipitation in the western HTP (Pamir areas), monsoonal rainfall in the southeastern and eastern TP and Himalayas, and sparse precipitation in the northern regions (Fig. S2).

Records of daily air pressure (P), temperature (T), relative humidity (RH), precipitation amount (PA), horizontal wind speed (WS) and wind direction (WD) observed at these stations displayed regional variability and seasonality of meteorology in the HTP during 2011-2013 (Fig. 2). Generally, the levels of P were clearly different, and decreased with ascending
altitude, showing values of 605.4± 3.7 hPa at the Ngari station, 604.6± 3.2 hPa at the QOMS station, 570.7± 4.4 hPa at the Nam Co station and 679.5± 2.9 hPa at the SET station (± 1.0 standard error). The altitude effect may have also influenced the horizontal WS values, which were 2.7±1.1, 4.3±1.6, 3.4±1.4 and 1.1±0.7 m s⁻¹ for the Ngari, QOMS, Nam Co, SET stations, respectively. The PA was controlled by Asian monsoon systems within annual ranges of 173.3-243.8 mm, 444.2-488.2 mm and 436.6-905.8 mm at the QOMS, Nam Co and SET stations, respectively. The lowest annual PA (40.9-125.3 mm) and mean RH (29.2±14.7%) were observed at the Ngari station. The greater seasonal variability in T noted at the Ngari station compared to other stations, i.e., from the lowest value (-10.6±4.8°C) in December-February to the highest value (14.0±3.1°C) in June-August, can be explained by its position far inland and its attendant climate.

2.2 Observation protocols for HTP atmospheric aerosols

Detailed information of HTP aerosol measurements are presented in Table 1, and include the physical, chemical and optical properties of atmospheric aerosols at the Ngari, QOMS, Nam Co and SET stations. RP 1400 series tapered element oscillating microbalance (TEOM) machines were installed and operated at each station to collect PM₂.₅ data from the autumn on 2011 onwards. PM₂.₅ mass was weighed and quantified based on the oscillation frequency of the tapered tube (Patashnick and Rupprecht, 1991). Their values were recorded at 5-min intervals. Values ranged from 0-5 g m⁻³, with a resolution of 0.1 μg m⁻³ and a precision of ±0.5 μg m⁻³ over a 24-hour average (Xin et al., 2015). At each station, size-segregated airborne particles (with
the diameters of <0.43 μm, 0.43-0.65 μm, 0.65-1.1 μm, 1.1-2.1 μm, 2.1-3.3 μm, 3.3-4.7 μm, 4.7-5.8 μm, 5.8-9.0 μm, and >9.0 μm, respectively) were collected weekly using airborne particle nine-stage samplers (Andersen Series 20-800, USA) at a flow rate of 28.3 l min⁻¹.

Quartz filters and cellulose membranes (with diameters of 81 mm) were applied alternately for measuring different chemical species, with a collection time of 72 h per week (always over the Monday-Wednesday period). Before and after sampling, the filters were weighed using a microbalance (sensitivity ±0.01 mg) after drying for 48 h, at 25°C and 50% humidity (Xin et al., 2015). Mass concentrations of these filtered samples were in turn obtained according to the standard sampling volume.

2.3 Methods of data analysis

The baseline properties of atmospheric aerosol mass revealed a relatively stable and low aerosol loading, excluding the possible perturbations (Kaufman et al., 2001; Xia et al., 2011).

Following Kaufman et al. (2001), we calculated the median of 50 consecutive hour-average values of online PM₂.₅ masses over 2-3 day, and removed data sequences with standard deviations higher than those of the whole time series by repeatedly shifting the running medians by one measurement point. The standard deviation thresholds were 24 μg m⁻³ at the Ngari station, 13 μg m⁻³ at the QOMS station, 9 μg m⁻³ at the Nam Co station, 11.7 μg m⁻³ at the SET station. Consequently, the any remaining datasets were considered the time series of baseline PM₂.₅ masses.

We applied monthly Level 3 datasets of Multi-angle Imaging Spectroradiometer (MISR) to characterize atmospheric column AOD (at 550 nm) over the HTP for 2011-2013. Level 2.0
Aerosol Robotic Network (AERONET) datasets that at the QOMS station, and Level 1.5 datasets at the Nam Co station were also used to address fine-mode AOD (at 500 nm). Additionally, a global 0.5 km land cover climatology that derived from Moderate Resolution Imaging Spectrometer (MODIS) (Broxton et al., 2014) was converted to a 1×1 degree pixel resolution using ArcGIS software, which provided the HTP’s land cover datasets.

3 Results and discussion

3.1 Mass concentrations of online PM$_{2.5}$ and segregated particles

Figure 3 and Table 2 show the time series and statistics for online PM$_{2.5}$ measurements monitored at four HTP stations during 2011-2013. The daily mean concentrations were 18.2±8.9 μg m$^{-3}$ at the Ngari station, 14.5±7.4 μg m$^{-3}$ at the QOMS station, 11.9±4.9 μg m$^{-3}$ at the Nam Co station, and 11.7±4.7 μg m$^{-3}$ at the SET station. Fine aerosol masses were therefore generally low but variable against various background atmospheres. These results were comparable with the monitored values of 11.7±15.5 μg m$^{-3}$ at a station in the Qilian Shan Mountains in the northeastern TP (Xu et al., 2014) and 26.6±19.3 μg m$^{-3}$ at a background Himalayan site (Panwar et al., 2013).

Baseline levels of hourly PM$_{2.5}$ mass were estimated to be 11.2±3.2 μg m$^{-3}$ at the Ngari station, 9.8±3.1 μg m$^{-3}$ at the QOMS station, 9.8±3.6 μg m$^{-3}$ at the Nam Co station, and 9.2±3.0 μg m$^{-3}$ at the SET station (Table 2). The discrepancies between online PM$_{2.5}$ and their baselines were also calculated. Consequently, average percentages and concentration levels were ~22.7% and 4.2±14.0 μg m$^{-3}$ at the Ngari station, ~16.6% and 2.1±2.0 μg m$^{-3}$ at the QOMS station, ~6.8% and 0.8±5.3 μg m$^{-3}$ at the Nam Co station, and ~10.3% and 1.2±6.6 μg m$^{-3}$ at the SET station.
at the SET station (Table 2). Relatively great distinctions therefore were found at the Ngari and QOMS stations. Significant variations, indicated by their daily frequency curves, also occurred at the Ngari and QOMS stations, and were associated with episodes of high concentration events (Fig. 4). These results implied a disturbance in the high-concentration aerosol masses of inland Asia associated with possible dust impact, and dependent upon proximity to local arid and barren areas (for their typical landscapes, see Fig. S1).

We further assessed mineral matter content in fine particles by analyzing elements and water-soluble inorganic ions in PM$_{3.1}$ samples with inductively coupled plasma mass spectroscopy (ICP-MS) and ion chromatography (IC). Mineral dust content was assumed to be a mixture of mainly crustal oxides, i.e. SiO$_2$, Al$_2$O$_3$, CaO, Fe$_2$O$_3$, K$_2$O, Na$_2$O and MgO. A detailed description of this approach can be found in Xin et al. (2015). Mineral content was about 26% at the Ngari station and 29% at the QOMS station. Our measurements revealed the impact of regional dust emissions, even for fine particles, over the HTP’s barren areas. Proportions were 2-3 times those of PM$_{3.1}$ (mean content 10.8%) measured at a suburban site impacted by heavy air pollutants in North China (Xin et al., 2015), and PM$_{2.0}$ (content of 14±4%) sampled at a human-influenced site in Hungary (Maenhaut et al., 2005).

Table 3 shows the statistical results of segregated-particle mass loadings according to weekly filters. These particles exhibited a general tendency of Ngari and QOMS stations (barren sites) > Nam Co station (grassland site) > SET station (forest site) in their mass levels, suggesting a potential effect associated with the HTP land cover. Furthermore, bimodal size distributions of surface-atmospheric particle masses occurred in these upland regions with an average pattern of a relatively small peak in accumulation mode and a more notable peak in
 coarse mode (Fig. 5). This represents an aerosol mass distribution pattern typical of
continental background air (Willeke and Whitby, 1975).

3.2 Diurnal variations in mass concentrations of fine aerosols

In these background atmospheres, the intensity of diurnal variabilities in PM$_{2.5}$ masses was
roughly characterized by their daytime (6:00-18:00 Local Time, LT) to nighttime (18:00-6:00
LT) ratios. Their average ratios were ~2.5 at the Ngari station, ~1.1 at the QOMS station, ~0.9
at the Nam Co station and ~1.8 at the SET station, based on hourly observations during the
2011-2013 period.

Higher ratios were found in valleys around the QOMS and SET stations, suggesting a
negative impact of mountainous valleys on the diffusion of local aerosol masses. The local
gemorphology around these sites is displayed in Figure S3. Conversely, these topographical
settings also produced mountain-valley wind circulations aligned with valley orientation, as
identified in July and August (Fig. 6). We analyzed the hourly datasets for the summer
monsoon period (July and August), as the mid-latitude westerlies are more prevalent during
the other periods and thus constrain the influence of synoptic-scale wind. Horizontal WD at
the QOMS station was consequently stronger and clearly inverse compared to that at the SET
station. Such a topographically-forced circulation can facilitate the spread of aerosols upslope
(Decesari et al., 2010; Babu et al., 2011; Cong et al., 2015). This would explain the ratio
being lower at the QOM station than at the SET station. The Ngari station is located in a
relatively open geomorphological setting, but experiences marked diurnal variations. This
phenomenon can be attributed to the dust lift from the barren land surface in the daytime, as
will be discussed below.

The overall patterns of diurnal variability in fine aerosol mass, atmospheric T and RH, as well as in horizontal WD, are shown in Figure 7. These fine particle masses begin to arise during 6:00-8:00 LT, accompanied by an increase in T and a decrease in RH. During the noontime period (10:00-14:00 LT), concentrations decreased again, shown by the trough in their diurnal curves, and coinciding with the highest T and horizontal WD values, and the lowest RH. Consequently, bi-peak patterns in diurnal variations were especially marked for the Nam Co station (whole year), and for the QOM and SET stations (autumn and winter). In contrast, the Ngari station, in the arid Asian interior, evinced a single-peak pattern in diurnal variations. Such variations are typically found in dust provenances (Mbourou et al., 1997; Stout, 2010), resulting from the atmospheric and land surface conditions prevalent during the daytime.

In the cases of April 6th-10th 2012, solar radiation (SR) imposed dramatic changes on the soil T and atmospheric T, RH and WS in the morning (Figs. 8 and 9). Here, SR was taken as the downward shortwave radiation and the soil T was the surface soil temperature at 0 cm, measured using an automated weather system at the Ngari station. Increases in atmospheric T and soil T, and a decline in RH, were synchronous from 6:00-7:00 LT, in response to solar heating. SR and soil T values rose increasingly in tandem during the 8:30-10:30 LT period, forming a very close relation ($R^2=0.96$, $P=0.98$), apparently in response to the arid and barren setting and cloud-free air at that time (Figs. 8 and 9a). Hence, soil T rose from ~ -2.5 °C to ~20 °C beyond the dew point temperature, and gradually dried out the surface moisture and uppermost layer of land (Fig. 9b). This in turn implied a reduction in the critical dust burst
threshold for barren conditions (Stout, 2010). Furthermore, the rise in morning WS created an atmospheric dynamic suited to dust suspension in the late morning, when fine materials were transported up from the land surface into the atmosphere (Fig. 8c). The combination of a declining critical dust burst threshold and favorable atmospheric fluctuation induced the increase in fine particles in the atmosphere, with a peak near noontime. During the 14:00-18:00 LT period, WS was strongest, with a range of 4-10 m s\(^{-1}\). Dependent upon its intensity, WS can dilute fine particle masses, rather than affect fine particle fluctuations between sandy surfaces and the air. In addition, a decrease in saltation activity prior to the WS dropping has frequently been observed in barren and arid continental interiors, possibly resulting from a reduction in turbulent wind fluctuations in the late afternoon (Stout, 2010). This effect can also restrict dust burst and thus its contribution to ambient fine particle content.

These in situ observations established that land surface and low-layer atmosphere are the key physical controls of the diurnal PM\(_{2.5}\) mass cycle at the Ngari station. They also confirmed that regional dust emissions contributed to the chemical composition of fine aerosols.

Bi-peak like diurnal variations in the PM\(_{2.5}\) masses at the Nam Co station, located in a grassland site near the great Nam Co Lake (Fig. S3), are shown in Figure 10. The planetary boundary layer height (PBLH) was derived from National Center for Environmental Prediction reanalysis data which used a 1×1 degree pixel and a 3 h temporal resolution (http://www.arl.noaa.gov/gdas1.php). In response to increasing T, the PBLH rose during the daytime from <100 m to >2500 m, associated with a rise in WS (Fig. 10). This combination of factors resulted in a marked diffusion of fine particles, shown by the trough in PM\(_{2.5}\) concentrations between 10:00 LT and 16:00 LT. This also accounted for the <1 daytime to
nighttime ratio.

### 3.3 Spatial and seasonal patterns in atmospheric aerosol masses

The monthly mean MISR-AOD values for 2011-2013 suggested HTP atmospheric aerosol masses were generally isolated from surrounding emissions (Fig. 11a). The integrated results of surface-atmospheric aerosol parameters and atmospheric-column aerosol optical properties yielded spatial distributions which suggested that TSP concentrations and MISR-AOD values decreased as land cover varied from barren land, through grassland, to forest (Fig. 11a).

The mean fraction of PM$_{2.5}$ to TSP was 27.4±6.65%, 22.3±10.9%, 37.3±11.1% and 54.4±6.72% for the Ngari station, QOMS station, Nam Co station, and SET station, respectively (Fig. 11b). These values increased from barren to forest areas, inversely to TSP masses. A time-average map of the aerosol fine-mode fraction (at 550 nm) for 2011-2013 was also constructed using monthly MODIS Terra (version 5.1) Level 3 values. Its spatial distribution was clearly consistent with the ground-based results recorded at various sites (marked by circles with various colors in Fig. 11b).

Figure 12 shows how MISR-AOD values varied along two cross-sections in different months. These results further confirmed a general decline in AOD from northwest to southeast crossing typical plateau landscapes (Section A), and from north to south in the eastern TP (Section B). Furthermore, such a spatial pattern was more notable for April-August, coinciding with the appearance of the Asian tropospheric aerosol layer during this period (Vernier et al., 2011). This may imply the significance of the development of the Asian tropospheric aerosol layer in modulating the AOD level over this plateau.
TSP mass and MISR-AOD values over HTP forest (SET station) and grassland (Nam Co station) sites shared a common seasonal pattern, with relatively higher values in spring and summer, followed by relatively lower values in autumn and winter (Fig. 13a). At the barren site (QOM station), there were inconsistent seasonal variations between surface-atmospheric TSP (PM$_{2.5}$) and atmospheric column AOD (fine-mode AOD) (Figs. 13a, b). Furthermore, there was no correlation between hourly surface PM$_{2.5}$ mass and fine-mode AOD (at 500 nm) at this site (Fig. S4). Using the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), Huang et al. (2007) detected frequent dust plumes in the lower atmosphere (~4-7 km asl) of the western HTP. These dust plumes possibly impacted the vertical distribution of aerosol masses over these barren areas.

PM$_{2.5}$ concentrations and fine-mode AOD values were higher in spring and summer than in autumn and winter at HTP forest and grassland sites, but not at the barren site (Fig. 13b). Ratios of PM$_{2.5}$ to TSP were apparently higher at the SET and Nam Co stations compared to QOM station, with a more marked difference in summer and autumn (Fig. 13c). In a background continental atmosphere, fine aerosols mainly originate from biogenic or wildfire emissions. Wild fire was were extremely rare in the HTP region, and fire-related emissions from the Asian Brown Cloud occurred only during the winter and spring, as measured in the Himalayan region (Cong et al., 2015; Decesari et al., 2010). Therefore, biogenic emissions and related products may be essential sources of fine aerosols over the HTP’s forest and grassland areas. In the southeastern TP, strong monoterpene emissions were reported, as there are a great number of alpine forest species (Wang et al., 2007); biogenic emissions were identified as the main precursors of atmospheric low-weight organic acids (Liu et al., 2014).
In the central TP, biogenic contributions to secondary organic carbon were estimated to be 
\( \approx 75\% \); biogenic aerosol tracer concentrations were also higher in summer than in winter 
(Shen et al., 2015).

4 Summary and conclusions

We studied aerosol mass loadings for the period 2011-2013 over the highland region of the 
HTP on both local and regional scales, through integrating multi-station measurements with 
satellite and ground-based remoting sensing. We found that mass concentrations of these 
surface atmospheric aerosols were relatively low and varied with land cover, with the general 
tendency of Ngari and QOMS (barren sites) > Nam Co (grassland site) > SET (forest site). 
PM\(_{2.5}\) concentrations at these sites were 18.2±8.9, 14.5±7.4, 11.9±4.9 and 11.7±4.7 μg m\(^{-3}\), 
respectively. Correspondingly, their fractions (to TSP) were 27.4±6.65%, 22.3±10.9%, 
37.3±11.1% and 54.4±6.72%. Bimodal mass distributions of size-segregated particles were 
found at all sites, with a relatively small peak in accumulation mode and a more marked peak 
in coarse mode. Diurnal variations in fine aerosol masses generally displayed a bi-peak 
pattern at the QOMS, Nam Co and SET stations, and a single-peak pattern at the Ngari station, 
controlled by the effects of local geomorphology, mountain-valley breeze circulations and 
aerosol emissions. Minerals matter content in PM\(_{2.5}\) samples was 26% at the Ngari station and 
29% at QOMS, or ~2-3 times that of reported results at human-influenced sites. Furthermore, 
our observations confirmed that land surface and boundary layer settings create a dynamic for 
these fine particles to be lifted from the barren land surface into the atmosphere.

Combining surface aerosol and atmospheric-column aerosol optical property data, we
found that TSP masses and MISR-AOD values generally decreased as land cover varied from barren to forest, inversely to PM$_{2.5}$ ratios. The seasonality of aerosol mass parameters was land-cover dependent. Over forest and grassland areas, TSP mass, PM$_{2.5}$ mass, MISR-AOD and fine-mode AOD values were higher in spring and summer and relatively lower in autumn and winter. Such spatial and seasonal variations were possibly associated with regional biogenic emissions and related aerosol products. At QOMS, there were inconsistent seasonal variations between surface TSP mass (PM$_{2.5}$ mass) and atmospheric column AOD (fine-mode AOD).

This study provides new insights on understanding the mass properties of HTP atmospheric aerosols. HTP aerosol masses (especially their regional characteristics and fine particle emissions) need to be treated sensitively in relation to assessments of their climatic effect and potential role as cloud condensation nuclei and ice nuclei.

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Table 1. Geographical conditions and aerosol observations at HTP background sites (Ngari, QOMS, Nam Co and SET stations).

<table>
<thead>
<tr>
<th>Station</th>
<th>Location</th>
<th>Altitude (asl)</th>
<th>Description</th>
<th>Research content</th>
<th>Observation</th>
<th>Instrumentation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ngari</td>
<td>79°42′E</td>
<td>4,264 m</td>
<td>Semi-arid area, western TP</td>
<td>1. Online and size distribution of aerosol masses;</td>
<td>1. PM_{2.5} (5 min) and nine-stage aerosol mass</td>
<td>1. TEOM RP1400 and nine-stage Anderson samplers;</td>
</tr>
<tr>
<td>station</td>
<td>33°23′N</td>
<td></td>
<td></td>
<td>2. Chemical composition and matter closure of size-segregated aerosols;</td>
<td>(weekly);</td>
<td>2. IC, ICP-MS and thermal optical carbon analyzer;</td>
</tr>
<tr>
<td>QOMS</td>
<td>86°57′E</td>
<td>4,300 m</td>
<td>North slope of the central Himalaya</td>
<td>3. Aerosol optical properties</td>
<td>3. Aerosol optical depth and Angstrom exponent (hourly);</td>
<td>3. Microtops II sunphotometer at Ngari and SETS stations, *CIMEL sunphotometer at QOMS and Nam Co stations</td>
</tr>
<tr>
<td>station</td>
<td>28°21′N</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nam Co</td>
<td>90°57′E</td>
<td>4,746 m</td>
<td>Alpine grassland, central TP</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>station</td>
<td>30°46′N</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>SET</td>
<td>94°44′E</td>
<td>3,326 m</td>
<td>Alpine forest, southeastern TP</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>station</td>
<td>29°46′N</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*The QOMS and Nam Co stations are also Aerosol Robotic Network (AERONET) sites (http://aeronet.gsfc.nasa.gov/). The abbreviations IC, ICP-MS and TEOM stand for ion chromatography, inductively coupled plasma mass spectroscopy and tapered element oscillating microbalance, respectively.
Table 2. Concentrations (µg m\(^{-3}\)) of hourly, daily and baseline PM\(_{2.5}\) and differences (µg m\(^{-3}\)) between online and baseline PM\(_{2.5}\) at four HTP stations for 2011-2013. Num and S.D. stand for number and standard deviation, respectively.

<table>
<thead>
<tr>
<th></th>
<th>Ngari station</th>
<th>QOMS station</th>
<th>Nam Co station</th>
<th>SET station</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM(_{2.5})</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hourly</td>
<td>1963</td>
<td>2049</td>
<td>11067</td>
<td>6871</td>
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<tr>
<td>Num</td>
<td>0.2-267.4</td>
<td>0.1-99.7</td>
<td>0.2-98.8</td>
<td>0.1-78.5</td>
</tr>
<tr>
<td>Range</td>
<td>18.5±24.3</td>
<td>13.8±12.3</td>
<td>11.8±8.1</td>
<td>11.7±10.0</td>
</tr>
<tr>
<td>Mean±S.D.</td>
<td>4.2±14.0</td>
<td>2.1±2.0</td>
<td>0.8±5.3</td>
<td>1.2±6.6</td>
</tr>
<tr>
<td>Daily</td>
<td>88</td>
<td>236</td>
<td>480</td>
<td>351</td>
</tr>
<tr>
<td>Num</td>
<td>7.1-77.3</td>
<td>2.6-48.1</td>
<td>3.9-43.9</td>
<td>2.8-28.7</td>
</tr>
<tr>
<td>Range</td>
<td>18.2±8.9</td>
<td>14.5±7.4</td>
<td>11.9±4.9</td>
<td>11.7±4.7</td>
</tr>
<tr>
<td>Mean±S.D.</td>
<td>11.2±3.2</td>
<td>9.8±3.1</td>
<td>9.8±3.6</td>
<td>9.2±3.0</td>
</tr>
<tr>
<td>Baseline</td>
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<td>1658</td>
<td>8880</td>
<td>4032</td>
</tr>
<tr>
<td>Num</td>
<td>4.9-24.4</td>
<td>2.6-18.4</td>
<td>3.3-28.4</td>
<td>2.7-20.3</td>
</tr>
<tr>
<td>Range</td>
<td>11.2±3.2</td>
<td>9.8±3.1</td>
<td>9.8±3.6</td>
<td>9.2±3.0</td>
</tr>
<tr>
<td>Mean±S.D.</td>
<td>11.2±3.2</td>
<td>9.8±3.1</td>
<td>9.8±3.6</td>
<td>9.2±3.0</td>
</tr>
<tr>
<td>Online-Baseline</td>
<td>1477</td>
<td>1431</td>
<td>8590</td>
<td>3554</td>
</tr>
<tr>
<td>Num</td>
<td>-16.3-109.7</td>
<td>-15.7-53.7</td>
<td>-17.7-33.0</td>
<td>-13.9-49.4</td>
</tr>
<tr>
<td>Range</td>
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<td>2.1±2.0</td>
<td>0.8±5.3</td>
<td>1.2±6.6</td>
</tr>
<tr>
<td>Mean±S.D.</td>
<td>11.2±3.2</td>
<td>9.8±3.1</td>
<td>9.8±3.6</td>
<td>9.2±3.0</td>
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</table>
Table 3. Average mass concentrations ± standard deviations (µg m\(^{-3}\)) for size-segregated particles (at various µm) and PM\(_{1.1}\), PM\(_{2.1}\), PM\(_{9}\) and TSP sampled from the HTP surface atmosphere during 2011-2013. Num stands for number of samples.

<table>
<thead>
<tr>
<th>Species</th>
<th>Ngari station</th>
<th>QOMS station</th>
<th>Nam Co station</th>
<th>SET station</th>
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<tbody>
<tr>
<td>Num</td>
<td>54</td>
<td>89</td>
<td>65</td>
<td>66</td>
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<tr>
<td>&lt;0.43</td>
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<td>5.8±6.0</td>
<td>4.1±4.3</td>
<td>2.6±2.6</td>
</tr>
<tr>
<td>0.43-0.65</td>
<td>8.8±5.8</td>
<td>6.6±6.2</td>
<td>3.7±3.4</td>
<td>2.4±2.1</td>
</tr>
<tr>
<td>0.65-1.1</td>
<td>7.5±3.6</td>
<td>6.5±5.0</td>
<td>3.6±4.5</td>
<td>2.6±2.2</td>
</tr>
<tr>
<td>1.1-2.1</td>
<td>7.0±3.8</td>
<td>7.6±6.8</td>
<td>3.2±3.1</td>
<td>2.3±2.0</td>
</tr>
<tr>
<td>2.1-3.3</td>
<td>8.1±5.5</td>
<td>7.1±5.5</td>
<td>3.3±3.5</td>
<td>2.3±2.1</td>
</tr>
<tr>
<td>3.3-4.7</td>
<td>7.2±3.6</td>
<td>8.3±11.1</td>
<td>3.4±3.8</td>
<td>2.5±2.0</td>
</tr>
<tr>
<td>4.7-5.8</td>
<td>7.2±3.4</td>
<td>8.3±10.5</td>
<td>3.4±4.0</td>
<td>2.4±2.2</td>
</tr>
<tr>
<td>5.8-9</td>
<td>7.8±7.1</td>
<td>7.7±5.9</td>
<td>3.7±4.9</td>
<td>2.0±1.6</td>
</tr>
<tr>
<td>&gt;9</td>
<td>5.6±8.2</td>
<td>7.8±6.6</td>
<td>3.5±4.1</td>
<td>2.3±3.5</td>
</tr>
<tr>
<td>PM(_{1.1})</td>
<td>24±14.5</td>
<td>18.9±17.3</td>
<td>11.3±12.2</td>
<td>7.7±6.8</td>
</tr>
<tr>
<td>PM(_{2.1})</td>
<td>30.6±14.2</td>
<td>26.3±20.6</td>
<td>14.5±12.9</td>
<td>10.0±8.2</td>
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<tr>
<td>PM(_{9})</td>
<td>60.9±27.5</td>
<td>57.5±45.4</td>
<td>28.4±25.9</td>
<td>19.2±15.0</td>
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<tr>
<td>TSP</td>
<td>66.4±29.6</td>
<td>65.1±50.9</td>
<td>31.9±29.0</td>
<td>21.5±18.0</td>
</tr>
</tbody>
</table>
Figures

Fig. 1. The main landscapes (1×1 degree pixel resolution) (a) and the aerosol observation sites in the HTP (b, c, d, e). The highland HTP region is taken as land above 2800 m asl but the thresholds are 1500 m asl for areas 92-97°E and 26-34°N, and 2000 m asl for areas 98-104°E and 24-34°E, accounting for the regional deviations caused by the extremely steep topography. The classification of landscapes, according to MODIS land cover classification (Broxton et al., 2014), suggests different land covers at these stations (here the forest areas comprise evergreen, mixed, and deciduous forests).
Fig. 2. Time series for hourly air temperature (T), relative humidity (RH), pressure (P), precipitation amount (PA), wind speed (WS) and wind direction (WD) in the HTP during 2011-2013, at the Ngari station (black), the QOMS station (red), the Nam Co station (blue), and the SETS station (cyan), respectively.
Fig. 3. Hourly and daily mean concentrations (μg m⁻³) of PM₂.₅ at the Ngari station (Oct. 2011-Oct. 2012), the QOMS station (Mar. 2012-Dec. 2013), the Nam Co station (Oct. 2011-Dec. 2013), and the SET station (May. 2011-Dec. 2013) in the HTP. Periods with no data were due to power supply problems or equipment breakdown. A daily mean was calculated only when at least eight hourly means were available during that day.
Fig. 4. Frequency distributions of daily PM$_{2.5}$ concentrations over the HTP observed during the 2011-2013 period. High-concentration peaks around the range of 12.5-20 μg m$^{-3}$ occurred in the frequency curves of the Ngari and QOMS stations, as indicated by the grey shading. The maximum PM$_{2.5}$ bin concentration was set to 50 μg m$^{-3}$, although a small fraction existed at higher concentrations.
Fig. 5. Size distributions of mass aerosol particles in the background surface atmosphere of the HTP (a: Ngari station, b: QOMS station, c: Nam Co station, d: SET station) as observed over the 2011-2013 period. Boxes show the percentile values (25, 50, 75) and whisker plots show maximum and minimum of non-outliers numbers, and the small blue circles behind the boxes are the distribution points.
Fig. 6. Wind rose plots for afternoon (12:00-16:00 LT) and nighttime (00:00-04:00 LT) in July and August at the QOMS station (a, b) and the SET station (c, d). An hourly horizontal wind direction (WD) was used, with its radii values expressed as percentages for wind blowing from particular directions.
Fig. 7. Seasonal diurnal variations in PM$_{2.5}$ concentrations, air T and RH over the 2011-2013 period at four background HTP sites (the Ngari station, the QOMS station, the Nam Co station and the SET station). The local time (LT) was used at each site, according to longitudinal position.
Fig. 8. Diurnal variations in PM$_{2.5}$ masses and related environmental factors for April 6th-10th 2012 at the Ngari station (located in a typical barren and arid area of inland Asia). SR is downward shortwave radiation and soil T is the surface soil temperature at 0 cm. The local time (LT) and a 30 min mean were used.
Fig. 9. Correlations between SR and soil T (a), soil T and PM$_{2.5}$ mass (b), and WS and
PM$_{2.5}$ mass (c) during the morning (8:30-10:30 LT) at the Ngari station, for April 6th-10th 2012. The smaller inserts show all recorded points within the measured timeframe. Note that the fit line of Figure 9a is for April 6th-9th 2012, because there was a rainfall event (~8:00-11:00 LT) on April 10th 2012, as indicated in Figure 8. However, even if the dataset for April 10th 2012 is included, the fit line remains more or less consistent, with $R^2=0.61$ and $P=0.79$. The local time (LT) and a 30 min mean were used.

Fig. 10. Diurnal variations in PM$_{2.5}$ concentrations and related environmental factors for October 8th-12th 2012 at the Nam Co station. 30 min mean datasets were used, based on local time (LT).
Fig. 11. Spatial patterns in AOD and TSP mass (a) and aerosol fine-mode fraction and the ratio of PM$_{2.5}$ to TSP (b) over the HTP. Figure 11a shows mean MISR AOD (at 550 nm) for 2011-2013 as derived from monthly Level 3 datasets. Figure 11b shows a time-average map of the MODIS fine-mode fraction (at 550 nm) for 2011-2013, according to monthly Terra (version 5.1) Level 3 values. Ground-based observations are average values sampled in 2011-2013.
Fig. 12. Mean MISR AOD (at 550 nm) for two cross-sections during various months in the 2011-2013 period. Missing datasets are plotted in white. The longitudinal Section A is from the southeast (100°E, 25.5°N-102°E, 31.5°N) to the northwest (79.5°E, 32.5°N-81.5°E, 38.5°N); the latitudinal Section B is from the south (95°E, 28°N-101°E, 28°N) to the north (95°E, 39°N-101°E, 39°N). J-D stands for the months of January-December.
Fig. 13. Seasonal characteristics of landscape-classified aerosol masses in the HTP, based on in situ observations and remote sensing datasets. The MISR-AOD (at 550 nm) values in Figure 13a are monthly Level 3 datasets for the 2011-2013 period over the HTP, and were classified based on landscape. The fine-mode AOD (at 500 nm)
data for barren and grassland sites were obtained from AERONET results at the QOMS station and the Nam Co station, respectively (Fig. 13b). Fine-mode AOD (at 550 nm) data for the forest area in Figure 13b were estimated, based on monthly MODIS Terra (version 5.1) Level 3 results, using the formula fine-mode AOD (at 550 nm) = AOD (at 550 nm) * fine-mode fraction (at 550 nm). Site land cover classifications are: alpine forest at the SETS station; alpine grassland at the Nam Co station; and barren land cover at the QOMS station. Boxes show the percentile values (25, 50, 75) and whisker plots show maximum and minimum of non-outliers numbers, and the small point within each box is the mean value. The abbreviations are March–May: MAM; June–August: JJA; September–November: SON; and December–February: DJF.