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A preliminary study on measurements of black carbon in the atmosphere of northwest Qilian Shan

Shuyu Zhao[1], Jing Ming[1,2,*], Cunde Xiao[1], Weijun Sun[1,3], Xiang Qin[1,3]

1. State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Gansu 730000, China. E-mail: petermingjing@hotmail.com
2. China National Climate Center, Beijing 100081, China
3. Qilian Shan Station of Glaciology and Ecologic Environment, Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Gansu 730000, China

Abstract

Black carbon (BC) concentration and meteorological data are measured discontinuously from May 2009 to March 2011, at the Qilian Shan Station of Glaciology and Ecologic Environment (hereafter "QSSGEE"), located near the terminal of the Laohugou No.12 Glacier in northwestern Qilian Shan, China. We measured the daily, monthly and seasonal variations of BC concentration in the atmosphere and discussed the possible emission sources. Black carbon background concentration in this region varied in the range of 18–72 ng/m^3 with the highest in summer and the lowest in autumn. The relations between BC concentration and surface wind direction indicated that BC concentration was higher when northwest wind prevails while lower when southeast wind prevails. Air masses backward trajectories showed the potential emission sources in the northwest. Significant positive correlations between daily mean BC concentration and relative humidity indicated that BC might be one of important cloud condensation nuclei. This hypothesis needs to be confirmed further through cloud microphysical features in this region.

Key words: black carbon; Qilian Shan; cloud condensation nuclei
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Introduction

Black carbon (BC), mainly generated from incomplete-combustions of bio-fuel, fossil fuel and biomass burning (Cooke and Wilson, 1996; Andreae and Crutzen, 1997), has drawn high attentions in last few decades because of its complicate climatic and environmental significances. Black carbon could transport far away from its emission source since its atmospheric lifetime could be one week or so. Black carbon background concentration is an important parameter for estimating its dry and wet deposition fluxes, calculating the albedo reduction while BC deposits in the surface of ice and snow and simulating the radiative forcing in climate models. Therefore, a number of measurements of BC concentration have been performed in the worldwide range, such as BC vertical profiles in spring of Arctic (Spackman et al., 2010) and other aircraft measurements in western and northern Europe, Houston in the USA and over the Pacific, and even in the Antarctic (Schwarz et al., 2009, 2010; McMeeking et al., 2010).

Continuous *in situ* measurement of BC could reflect its long-term trend, and is critical to understand regional and global BC level and sources. Since 1994, BC measurement has begun in Mt. Waliguan Observatory, a baseline station of the Global Atmospheric Watch. More BC measurements have been carried out at regional background observatories and the metropolis by researchers, meteorological administrations and environmental protection departments in China, which improve our understandings of BC background concentration level in different areas and effects of BC on regional climate. However, BC measurement data are still very scarce in the hinterland of west China. Qilian Shan Station of Glaciology and Ecologic Environment (QSSGEE, since 2009) is a remote *in situ* measuring site for BC after Mt. Waliguan Observatory in 1994 and Nam Co Observatory in 2006. Here, we presented the daily, monthly and seasonal variations of BC concentration and discussed the possible emission sources and transporting paths in different seasons, according to the preliminary measurements in the atmosphere at QSSGEE.

1 Data and methods

1.1 Site description and meteorological conditions

Qilian Shan Station of Glaciology and Ecologic Environment (39°30′16″N, 96°30′22″E, 4214 m a.s.l.) located near the terminal of Laohugou No.12 Glacier in northwestern Qilian Shan, is about 104.4 km far away from the nearest industrial city (Yumen) and is a remote hinterland site (Fig. 1). Laohugou No.12 Glacier is the greatest valley glacier in Qilian Shan, and a recent measurement shows

* Corresponding author. E-mail: petermingjing@hotmail.com

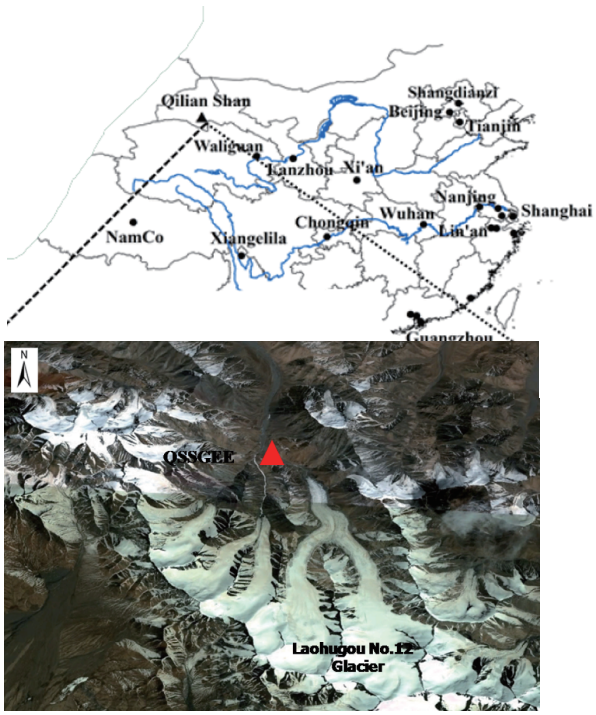

Fig. 1 Measurement sites distribution of black carbon (BC) in the atmosphere of China. The triangle location denotes QSSGEE.

that its area is about 21 km^2 with the terminal elevation of 4260 m (Liu, 2010). QSSGEE is built at the seasonal snowpack zone, above which is high-mountain snow and ice zone.

The AE-31 aethalometer is set indoor room. To minimize the influence of local personnel activities, the observation room is built at a natural moraine platform that is in the southwest of the station, and 100 m higher than the residential areas. Black carbon concentration has been measured since 2009, and meteorological data are simultaneously recorded by an automatic weather station. Controlled by the mid-latitude westerlies, the local climate is typically continental (dry and arid) with few precipitations occurring from May to September. The prevailing wind directions are SSE (33.3% in seasonal wind directions) and NNW (24.4%) in spring (March and May); NNW (35.4%) and SSE (30.0%) in summer (June-July-August); SSE (40.4%) in autumn (September-October-November) and SSE (42.9%) in winter (December-January-February) during the observation period (Fig. 2). Monthly mean air temperature, relative humidity, wind speed and mean daily precipitation during the observation period are plotted in Fig. 3. More precipitation and higher relative humidity appear in wet seasons (May-September) when precipitation is mainly caused by the mixing of air masses from the SSE and NNW directions. Monthly mean wind speed is lower (ca. 2.8 m/sec) in September/October, which increases to 3.4 m/sec in November/December.

1.2 Measurement method

Black carbon concentration is measured by AE-31 aethalometer (Magee Scientific Corporation, USA) with a seven-wavelength (370, 470, 520, 590, 660, 880 and 950

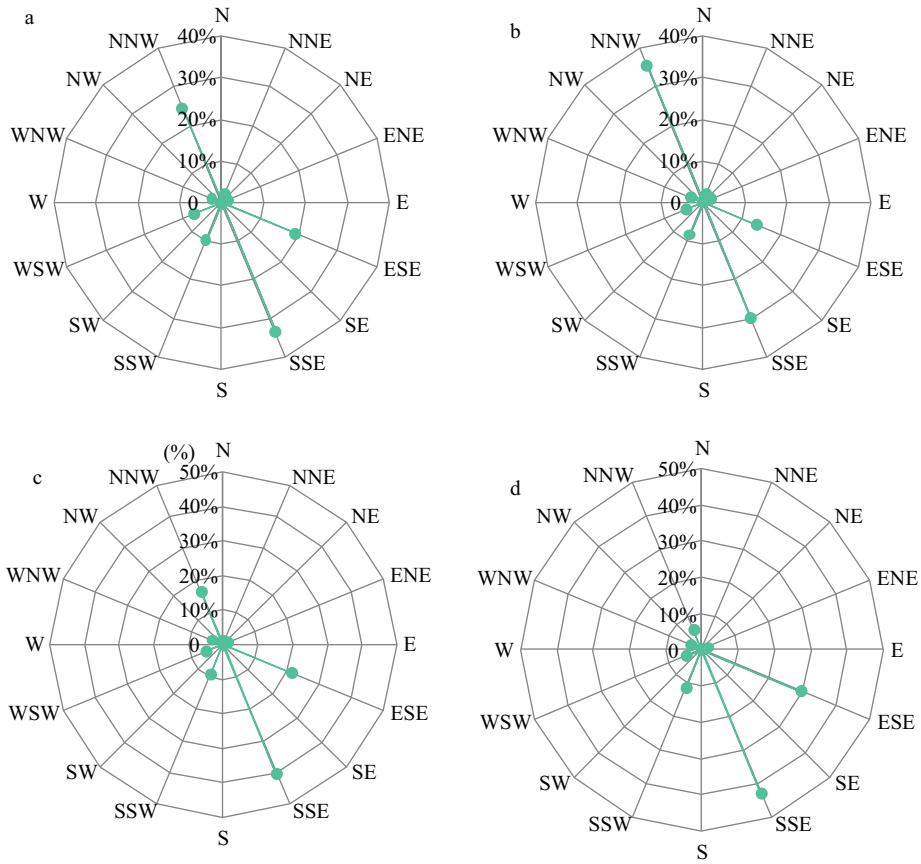

Fig. 2 Wind direction frequency in different seasons of Qilian Shan. (a) spring (March and May); (b) summer (June-July-August); (c) autumn (September-October-November); (d) winter (December-January-February).

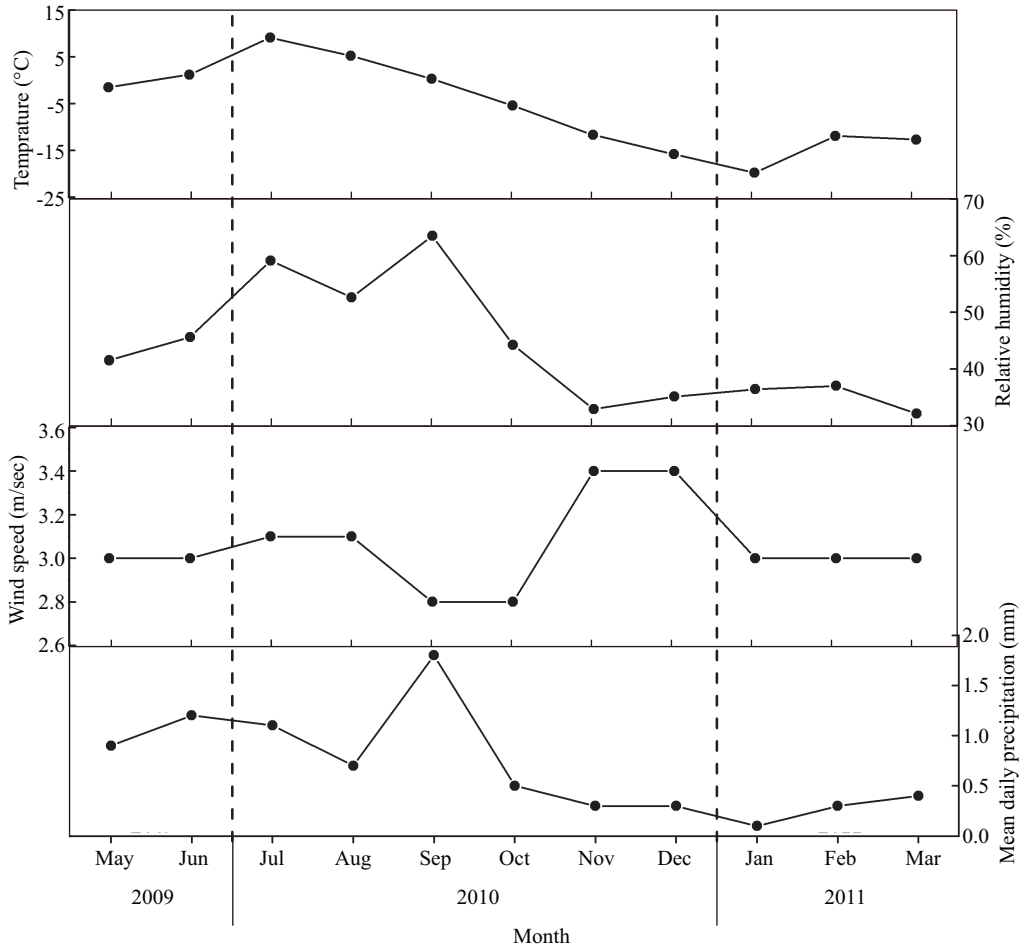

Fig. 3 Monthly mean air temperature, relative humidity, wind speed and mean daily precipitation during the BC measurement period in the region of Qilian Shan.

nm) which is composed of air sampling system, optical system, filter fiber-advance structure and data collection and processing system. The aethalometer is installed at a level of 3 m above the ground using an inlet tube and pump which is operated at a stable airflow rate about 3.9 L/min for 24 hr/day with a time resolution of 5 min. The power supply of the instrument is solar panels.

Black carbon concentration calculated by Eqs. (1) and (2) (Hansen, 2005a) is derived from the linear relationship between BC concentration and light attenuation absorption coefficient. Here, we should note that BC concentration represents all light-absorbing particle matter concentration in the atmosphere, because, compared with BC, light attenuation absorption contribution of other light-absorbing particle matter (e.g., dust) could be neglected (Warren and Wiscombe, 1980; Yang et al., 2009):

$$M_{\mathrm{BC}} = \frac{A \times \mathrm{ATN}(\lambda)}{kQ(t_1 - t_0)} \tag{1}$$

$$\mathrm{ATN}(\lambda) = 100 \ln(I_0/I_1) \tag{2}$$

where, I_0 and I_1 are separately transmission light intensities of the reference and the sample beams at time t_0 and t_1 (sec), A (m^2) is the area of the sampling spot on the filter, and ATN is attenuation absorption coefficient. Q (L/min) is the sampling airflow rate and k (m^2/g) is absorption efficiency. The light beam transmits through the blank spot, also called "referenced spot" and the sampling spot, respectively, using photoelectric diode to detect the transmitting of the light, then converting light attenuation absorption coefficient into BC concentration with absorption efficiency which varies as a function of wavelength. In general, BC concentration calculated at 880 nm with absorption efficient of 16.6 m^2/g (Hansen, 2005) is regarded as the real BC concentration in the atmosphere (Bodhaine, 1995).

The number of days on which BC concentration is observed is totally 245 during the observation period. The instrument stopped working on June 15 in 2009 due to the power failure. Up to July 14 in 2010, the instrument is recovered to measure BC concentration after it is repaired and calibrated in the factory. The numbers of days on which BC concentration measurements have been made in May and June of 2009, July of 2010 and February and March of 2011 correspond to 15 days or so.

2 Results and discussion

2.1 BC concentration variation in the atmosphere

Figure 4 shows that the variations of daily mean BC concentrations are generally less than 150 ng/m^3 except

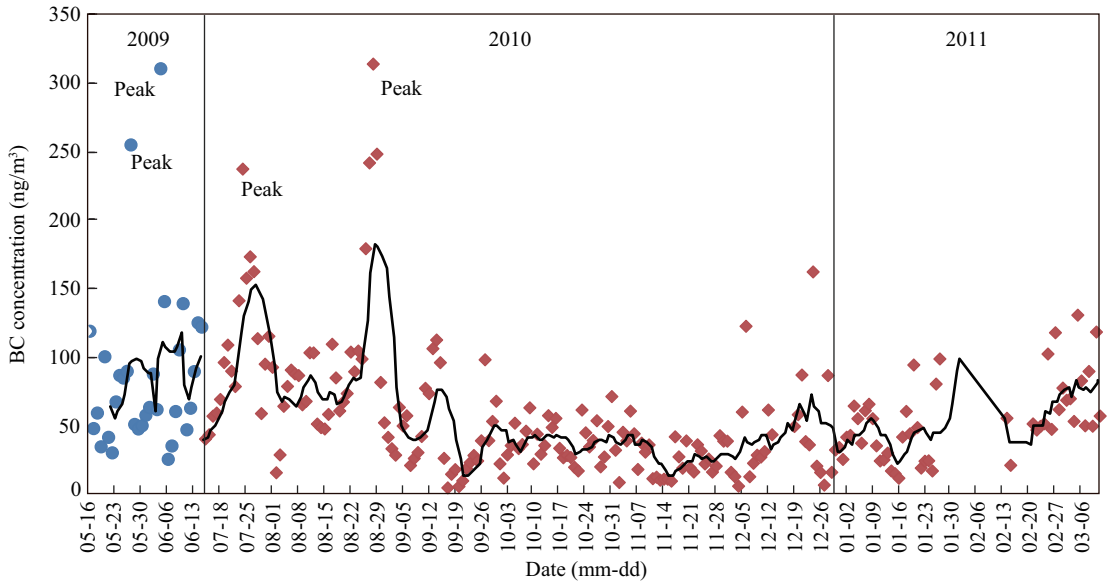

Fig. 4 Daily mean BC concentration during the measurement period in the region of Qilian Shan, the dark solid line is the seven-day smooth average of BC concentration.

several peak values that are higher than 200 ng/m^3, occurring on May 27 and June 4 in 2009 and July 24 and August 28 in 2010. Daily mean BC concentration in 2010 varies more largely than that in 2009, especially in July and August. Seven-day smooth average of BC concentration varies mainly in the range of 50–120 ng/m^3 in 2009 and changes from 28 ng/m^3 (November) to 150 ng/m^3 (August) in 2010. The daily mean BC concentration is usually less than 150 ng/m^3, and this low BC concentration level reflects a weakly local emission. Black carbon background concentration, defined as the concentration of BC in a pristine air mass when the contribution from anthropogenic sources is absent, varies in the range of 18–72 ng/m^3 in Qilian Shan, which is close to the background concentration in Mt. Waliguan (50–130 ng/m^3) (Tang et al., 1999). Mean daily BC concentration measured in Qilian Shan during the measurement period is 48 ng/m^3, and it is slightly lower than that measured in the Nam Co Observatory in the central Tibetan Plateau (Ming et al., 2010). Above all, *in situ* measurements of BC background concentration in the three sites over the Tibetan Plateau are comparable.

Peak values of daily mean BC concentration in Fig. 4 are mainly because of the prevailing wind directions that transport BC from emission sources or no precipitations occurring for those days.

2.2 Relationship between BC concentration and surface wind fields

Seasonal variation of BC concentration in Qilian Shan, different from Mt. Waliguan and urban sites in China, shows the highest of 100 ng/m^3 in summer and the lowest of 37 ng/m^3 in autumn (47 and 78 ng/m^3 for winter and spring, respectively) (Fig. 5). The BC concentration in Mt. Waliguan influenced mainly by seasonal wind direction shows the highest in spring and the lowest in winter; wherein coal burning in spring also contributes to the highest BC concentration (Tang et al., 1999). In urban sites, residents living heating leads to the highest BC

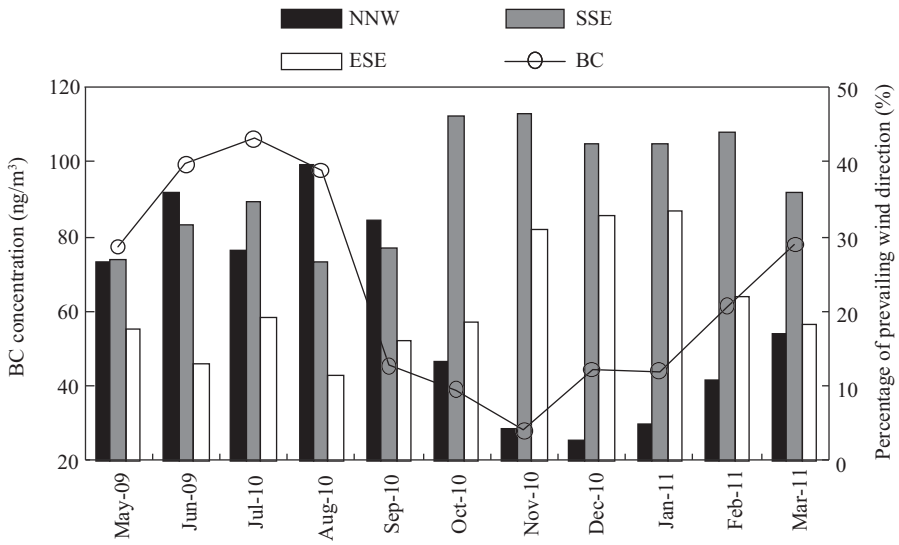

Fig. 5 Co-variation of BC concentration and the prevailing wind direction in the region of Qilian Shan during the measurement period.

concentration in winter (Cao et al., 2007, 2009). As for QSSGEE, no local emissions contribute BC concentration after the personnel leave the station at the end of October, so we ignore local emission contribution to seasonal variations of BC concentration. Considering key factors influencing BC concentration such as emission sources, general circulation transport, local topography, dry and wet removing processes and dynamic effects in boundary layer (Huang et al., 2010; Ramachandran and Sumita, 2010), we suggest that BC in Qilian Shan is mainly from emission sources of neighboring areas by long-range atmospheric transport.

Seasonal variations of wind direction and wind speed are recorded by the automatic weather station installed at the station. Hourly-averaged surface prevailing wind directions are SSE and NNW, accounting for the percentage of 27.0% and 26.5%, respectively. In summer, NNW, amount to 35.4%, becomes the prevailing wind, and SSE (30.0%) is secondary. In autumn, the wind directions shift from NNW (32.2%) and SSE (28.5%) in September, SSE (46.2%) in October to SSE (46.4%) and ESE (30.8%) in November. However, ESE (42.9%) prevails in winter, and SSE (29.7%) is secondary. Co-variation between monthly mean BC concentration and wind direction suggests that BC concentration is higher while NNW prevails with correlation coefficient of 0.754 (n = 11), exceeding 99% confidence level. When SSE and ESE prevail, BC concentration is lower with the correlation coefficient of –0.672 and –0.680, respectively, exceeding 95% confidence level (Fig. 5). The peak of daily mean BC concentration is also closely related to surface wind direction, and the peak values usually correspond to prevailing NNW on that day or before. Therefore, emission sources in the north and northwest of Qilian Shan could influence BC concentration in this region. In addition, BC concentration decreases rapidly after the precipitation occurs; this suggests that wet removing processes have significant influences on BC concentration in Qilian Shan.

Wind speed is another important factor that has impacts on BC concentration in the atmosphere because the larger wind speed favors the stronger air advection and the transport and diffusion of aerosols. In Qilian Shan, strongly wind developing in winter favors the advection and diffusion of aerosols. On the other hand, the atmosphere in winter is in a stable state with stratification motions, so aerosols transported by general circulation in upper troposphere could have less influence on BC concentration except weakly dry deposition. Therefore, wind speed also contributes to the low BC concentration in winter of Qilian Shan.

2.3 Relationship between BC concentration and relative humidity

Generally, the size of BC is about 200 nm, and belongs to nuclei modal; however, its size would vary with source types and distance to the emission sources (Schwarz et al., 2009b; McMeeking et al., 2010). Zhang et al. (2000) analyze aerosol size characteristics in Beijing and suggest that aerosol number concentration is positive correlated to relative humidity while relative humidity is lower; wherein relative humidity is lower than 75%, number concentration of fine aerosol particles increases with relative humidity while relative humidity is higher than 75%, number concentration of fine aerosol particles decreases. Therefore, BC could accumulate and mix with hydrophilic aerosols and experience hygroscopic growth while relative humidity is lower. However, precipitation in Qilian Shan mainly occurs in July, August and September when relative humidity is higher and the cloud cover is intensified. Formation of clouds is closely related to the number concentration of cloud condensation nuclei (CCN), and that whether CCN would be activated is mainly influenced by aerosol size, composition and airflow speed in the clouds (Koch et al., 2011). Although hydrophobic BC could also be as CCN while relative humidity is higher, larger hygroscopic aerosols (e.g., OC, sulfate and nitrate) could experience hygroscopic growth to activate CCN more easily and become cloud droplets and rain droplets. Meanwhile, the light-absorption capacity of aged BC weakens and subsequent calculated BC concentration also decreases while relative humidity is higher, so BC concentration is usually weakly negative correlated with relative humidity in wet seasons. When monthly mean relative humidity (July-August-September) is higher than 50%, daily mean BC concentration has weakly negative correlations with relative humidity, and this might be caused by remaining time and hygroscopicity of BC in the atmosphere of Qilian Shan region. This weakly negative correlation in main wet seasons in Qilian Shan suggest that higher BC concentration might have reduced the number of CCN activated and subsequent cloud cover and precipitation, although BC itself could be as CCN and relative humidity is much higher in wet seasons. Maybe, BC near the surface has influenced convective precipitation in summer, so further studies about meteorological observation and BC vertical profile in boundary layer at QSSGEE are needed.

Carrico et al. (2003) indicate that the single-scattering albedo of anthropogenic aerosols increases from 0.91 to 0.96 corresponding to relative humidity increasing from 40% to 85%, which suggests an obvious hygroscopic growth while relative humidity is becoming higher. Noh et al. (2011a) also discover that long-range industrial/urban haze and forest fire plumes particles could grow larger when relative humidity increases from 30% to 85% with a hygroscopic growth factor of 1.49 ± 0.46. Therefore, aerosols including BC could experience hygroscopic growth to some different extent during the long-range transport. Large particles emitted from biomass burning (mainly OC) could be activated to CCN more easily than small particles emitted from diesel burning (mainly BC), and soot could become carriers of dry deposition of hygroscopic and secondary aerosols which should be activated to CCN and then form clouds (Noh et al., 2011b). In other words, BC itself could be as CCN to increase CCN number concentration while it would decrease CCN number concentration by collision and coalescence with hygroscopic aerosols.

Daily mean BC concentration and relative humidity

in Qilian Shan have significant positive correlations with coefficient of 0.284 (n = 239) > 0.231 (n = 200, t-test threshold), exceeding 99.9% confidence level (Fig. 6). Although BC aerosols are hydrophobic, they could act with hydrophilic aerosols (e.g., sulfate, nitrate) and become mixing nuclei during the long-range transport (Jacobson, 2001). The significant positive correlations between daily mean BC concentration and relative humidity during the measurement period indicate that BC might be as one of main CCN to influence precipitable water and precipitation intensity in this region. While relative humidity is lower than the threshold (ca. 50% for Qilian Shan) below which CCN could grow by nuclei activation, the outer of BC could experience hygroscopic growth by mixing with hygroscopic aerosols and also be as carriers where water vapors experience condensation growth with increasing relative humidity. In condensation processes, BC concentration could cumulate in several days in the atmosphere, but it would decrease rapidly by wet removing once the precipitation occurs. Therefore, we speculate that BC particles may be as one of important CCN in Qilian Shan region. Nevertheless, only through further studies on BC mixing states, deposition fluxes, deposition types and content in precipitation, can we understand more about BC microphysical properties and its effect on cloud formation and precipitation in this region.

2.4 Source analyses of atmospheric BC

We calculate three-day typical air masses backward trajectories to influence BC concentration in different seasons of Qilian Shan by HYSPLIT model provided by Air Resources Laboratory of NOAA, combined with reanalysis grid meteorological data of 2.5°×2.5° provided by Climate Diagnostics Center NCEP/NCAR I (CDC-I). Based on the altitude of QSSGEE (4124 m), air masses heights are set at 3800 m, 4200 m and 5500 m, respectively. Approximate transport paths in different seasons (spring and summer in 2009, summer and autumn in 2010) of Qilian Shan are shown in Fig. 7. In late spring (May in 2009), air masses mainly come from the northwest of the measurement site, northwest airflows at mid-troposphere (5500 m, 500 hPa) and surface are nearly the same, consisting with the most frequency of NNW recorded by local automatic weather station. This indicates that BC emission sources in northwest China, especially in the region of northwest Gansu Province and Xinjiang Province, would influence BC concentration in this region. In early summer (June in 2009), BC emission sources indicated by leading airflow in mid-troposphere and surface airflow have horizontal differences, but they still locate in the northwest of the measurement site, resulting in higher BC concentration. During mid-summer (July in 2010), leading airflow in mid-troposphere shifts to NW and surface airflow shifts to SSE, later turning to E, roughly consisting with SSE recorded by local automatic weather station. Meanwhile, air temperature and relative humidity are both higher while precipitation is relatively less; therefore, accumulative effects of aerosols result in the highest BC concentration, from which we suggest that washout effects could also have remarkable contribution to BC concentration in this region, but quantitative contribution proportion needs to be determined further. While in autumn (October in 2010), air masses mainly come from the west, and leading and surface airflows indicated by models are nearly the same, obviously different from SSE recorded by local automatic weather station, which might suggest the reliability of reanalysis data in different seasons or in mountain regions of west China. However, the lowest BC concentration in autumn is likely to reveal that BC emission sources mainly locate among the northwest and north of the measurement site, and air masses from the west and the south are relatively clean to influence less on BC concentration in Qilian Shan region.

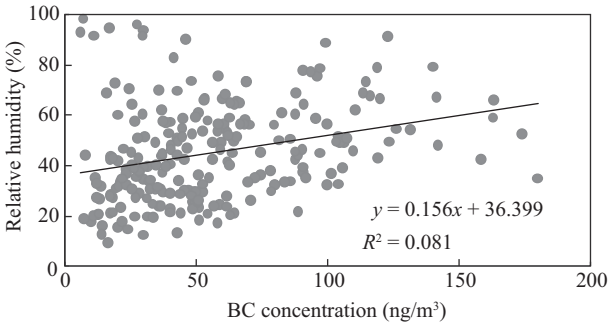

Fig. 6 Scatter plot distributions of daily mean BC concentration and relative humidity.

Based on three-day typical air masses backward trajectories analysis, we conclude that BC in the atmosphere of Qilian Shan mainly come from atmospheric long-range transports at mid-troposphere, and shorter ground paths contribute less, which reflects large-scale terrain could significantly influence air masses transport and regional aerosol concentrations. Therefore, while analyzing BC emission sources in high-elevated mountain regions in China, we must consider the large-scale terrain which could decrease surface wind speed by friction and shorten the source distance. As for clean atmosphere in regions like Qilian Shan, BC mainly come from atmospheric long-range transport at the height of local free atmosphere, so height settings of influencing air masses need to be determined by the altitude of the measurement site before source analyses.

3 Conclusions

Black carbon concentration in the atmosphere of Qilian Shan is the highest of 100 ng/m^3 in summer and the lowest of 37 ng/m^3 in autumn. Monthly mean BC concentration is the highest of 106 ng/m^3 (July) and the lowest of 28 ng/m^3 (November). BC background concentration varies in the range of 18–72 ng/m^3, comparable to that measured in Mt. Waliguan and Nam Co Observatory. Seasonal variation of BC concentrations in Qilian Shan is obviously influenced by surface wind directions; BC concentration is strikingly higher while NNW prevails, lower when SSE prevails. Daily mean BC concentration is positively correlated to relative humidity with correlation coefficient of 0.284 (n= 239), which indicates that BC could be one of important

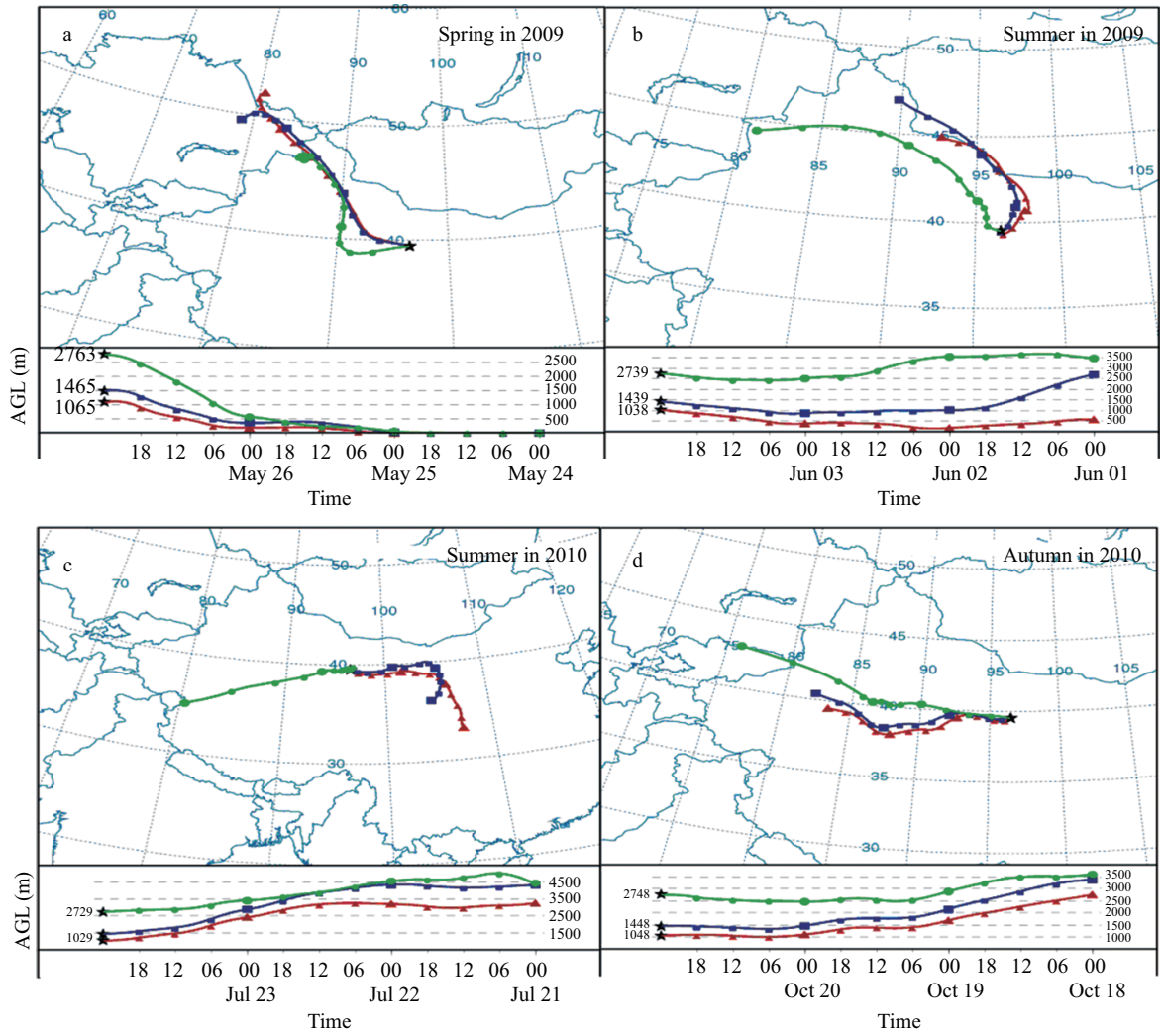

Fig. 7 Three-day typical air masses backward trajectories at different heights for different seasons in QSSGEE. (a) late spring, (b) early summer in 2009; (c) mid-summer, (d) mid-autumn in 2010. http://ready.arl.noaa.gov/HYSPLIT_traj.php.

CCN that involve in the formation of clouds. Nevertheless, this needs to be confirmed further through cloud microphysical features (cloud droplets modal, CCN size and composition). Furthermore, effects of BC on cloud cover, precipitable water and precipitation intensity need detailed studies. Finally, three-day typical air masses backward trajectories calculated by HYSPLIT model show that BC in the atmosphere mainly come from the emission sources located among the northwest and north region of Qilian Shan. Therefore, industrial activities, urbanization and residents living emissions in north and northwest of Qilian Shan would influence BC background concentration.

Acknowledgments

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