

Characteristics and source of black carbon aerosol over Taklimakan Desert

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Black carbon (BC) and PM₁₀ in the center of the Taklimakan Desert were online monitored in the whole year of 2007. In addition, TSP samples were also synchronously daily collected by medium-volume samplers with Whatman® 41 filters in the spring of 2007. BC in the dust aerosol was up to 1.14% of the total mass of PM₁₀. A remarkable seasonal variation of BC in the aerosol was observed in the order of winter > spring > autumn > summer. The peak value of BC appeared at midnight while the lowest one in the evening each day, which was just the reverse of that in the urban area. The contribution of BC to the total mass of PM₁₀ on non-dust storm days was ~11 times of that in dust storm. Through back trajectory and principal component analysis, it was found that BC in the dust aerosol over Taklimakan Desert might be attributed to the emission from the anthropogenic activities, including domestic heating, cooking, combustion of oil and natural gas, and the medium-range transport from those oases located in the margins of the desert. The total BC aerosol from the Taklimakan Desert to be transported to the eastward downstream was estimated to be 6.3×10^4 ton yr⁻¹.

black carbon, aerosol, the Taklimakan desert, long-range transport

1 Introduction

As the second largest desert over the world, Taklimakan is one of the two major source areas of Asian dust. In the early 1980s, it was found by means of the satellite and ground-based observation that Taklimakan Desert was the main contributor of the mineral aerosol transported to the North Pacific Ocean [1]. Half of the dust originated from the desert could be deposited onto the ocean [2], which could provide both nutrient and toxic elements/compounds to the organism in the surface seawater of the open ocean. Atmospheric iron is the limiting factor of the primary produc-

tivity in a certain ocean area [3]. The role of mineral aerosol in the global climate and environmental change has attracted much more attention than before [4–6].

The long-range transport of Asian dust would mix with the pollution aerosol on its pathway, which may have more serious and complicated impact on the global biogeochemical cycle of those elements/compounds transported, for there are much more pollution aerosol has been produced in the past two decades due to the rapid industrialization and urbanization over China. There has been heavy haze spreading over eastern and central China, especially in the Yangtze river Delta, Pearl River Delta, Beijing-Tianjin region, and Sichuan Basin, which are all on the transport pathway of Asian dust [7, 8]. Through the long-term field observation of dust aerosol on the pathway of Asian dust for the past

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eight years in our group [9, 10], it was revealed that the composition of dust aerosol had been greatly transformed during the long-range transport, which is very different from its original source and, in turn, would also change the solar radiation of dust on the earth from that it did before. Black carbon (BC), which was verified to be mainly from anthropogenic activities, has been one of the major components of the mixed aerosol, in addition to mineral aerosol, secondary water soluble aerosol, and organic carbon.

Black carbon refers to those substances that have the full light absorption as "black" substance does, which is of mostly elemental carbon produced by incompletely combustion. Black carbon is mostly derived from the anthropogenic activity, such as coal combustion, biomass burning in industry and agriculture, plus less part from the natural source, such as forest fire. Generally black carbon can remain in the air for one week or so and then be scavenged by wet and/or dry deposition. It has been estimated that the wet and dry deposition of black carbon into the global ocean were 2 and 10 Tg C yr⁻¹, respectively [11]. Bond *et al.* [12] assessed that the annual emission of black carbon was 8 Tg yr⁻¹ on a global scale, with 20% from biomass burning, 38% from fossil fuels, and the rest (42%) from the open biomass burning. Black carbon aerosol plays an important role in the global climate and environmental change, for it could heat around air by the absorption of the visible light. Unlike other components of aerosol, which have cooling effect, black carbon has become the second significant species besides CO₂ in the global climate change due to its warming effect [13, 14]. Black carbon does directly affect the balance of solar radiation [15], indirectly influence the microphysical formation of cloud by serving as cloud condensation nuclei (CCN) [16], and degrade the visibility. On the long-range transport of dust aerosol, black carbon in addition to other components in the aerosol, mixes with the pollution components and forms the atmospheric brown cloud (ABC) on the continental scale, which would decrease the solar radiation to the ground and then change the hydrological cycle on the regional and even global scale [14]. 80% of the black carbon in the aerosol over the Indian Ocean was found to mix with sulfate aerosol [17]. It was reported that the increased black carbon in the snow on the remote highest mountain, Everest, accelerated the melting of snow and changed the albedo [18]. However, so far there has been little research on the black carbon in the dust aerosol over the dust source areas. This study provides firstly the information on the black carbon collected from Taklimakan desert.

2 Experiment

2.1 Sampling and BC monitoring

The sampling site was located in the Tazhong (TZ) weather observation station (83.67°E, 39.00°N) in the center of the

Taklimakan Desert. Black carbon had been monitored online with an aethalometer (AE-31, Magee Scientific Co., USA) at a flow rate of 4.0 L min⁻¹ from January 2007 to February 2008. PM₁₀ was also monitored online with the TEOM series of a 1400a monitor (Rupprecht & Patashnick Co., Inc., Albany, NY, USA) at a flow rate of 16.7 L min⁻¹. Both monitors were held ~3 m above the ground. PM_{2.5} and TSP filter samples were daily collected (normally from 8:00 A.M. to 8:00 A.M. the next day) with medium-volume samplers (model: (TSP/PM₁₀/PM_{2.5})-2, flow rate: 77.59 L min⁻¹) with Whatman® 41 filters (Whatman Inc., Maidstone, UK) in springtime from March 20 to April 20 in 2007. During the heavy dust, the samples were collected for a few hours each. The samplers were held ~1.5 m above the ground. All filters were weighed before and after sampling with an analytical balance (Sartorius 25S BT, reading precision 10 µg) after stabilizing under constant temperature (20 ± 1 °C) and related humidity (40 ± 2%). All procedures were strictly quality-controlled to avoid any possible contamination of the samples. Black carbon in the sample filters was measured with an EEL smokestain reflectometer (Model 43D, Diffusion systems Ltd., London).

2.2 The monitoring method of BC

Aaethalometer (model AE-31), the BC online monitor, can simultaneously measure optical absorption at 7 wavelengths (370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm, and 950 nm) every 5 min. The principle of this monitor is to measure the attenuation of a beam of light transmitted filter with loading samples. BC concentration was calculated on the assumption that the attenuation is linearly proportional to the amount of BC on the filter. The uncertainty of measurement might originate from the multiple scattering in the filter fiber in the unloaded filter and those particles embedded in the filters. For a better result with acceptable uncertainty, the attenuation value is limited in the range of 75–125 at various wavelengths, no greater than 150. In this study the maximum attenuation was within 40–88, much less than that of the limitation, to ensure the reliability of the BC measurement.

Figure 1 shows the measured BC concentration at 7 wavelengths. The mean annual concentrations of BC at Tazhong station at 370, 470, 520, 590, 660, 880, and 950 nm were 2194.8, 1931.7, 1710.0, 1624.4, 1605.4, 1659.1, and 1653.0 ng m⁻³, respectively. It could be seen that concentrations decrease from ultraviolet to visible band (370–660 nm) and rise at infrared band (880–950 nm). Arimoto *et al.* [19] reported that the first derivative of the spectra at 555 nm and 435 nm was likely the signature of hematite and goethite in dust, for iron-oxide mineral could have absorption at such wavelengths. Also, it was found that there was a peak at 370 nm of the first derivative of spectra [20] and a noticeable absorption at infrared band (880 and 950 nm) [21], both of which were likely due to hematite. Thus, to avoid

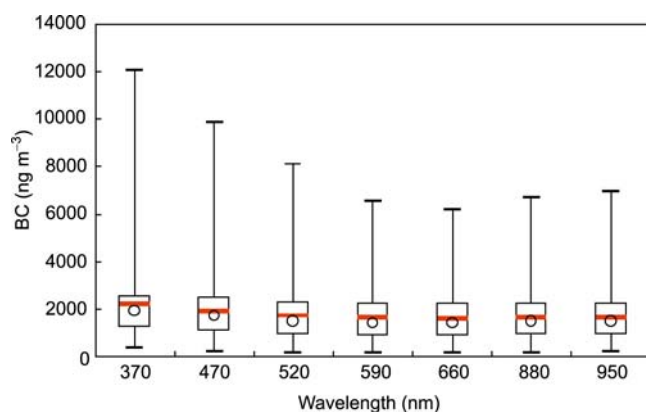


Figure 1 BC concentrations at 7 wavelengths.

the inference from iron oxides in dust, the wavelength at 660 nm was used to measure BC in this study.

To verify the reliability of the measurement, both BC concentrations measured online with aethalometer and the BC in the filter samples measured with smokestain reflectometer were compared with each other. The correlation coefficient of the two measurements with two instruments was greater than 0.72, indicating both measurements were agreeable. In addition, the precision of BC online measurement is greatly dependant on the stability of the flow rate. The flow rate was stable and kept within a small range of 3.8–4.1 L min⁻¹ in this study, thus, the results could be reliable.

2.3 Chemical analysis

2.3.1 Element analysis

19 total elements in the aerosol samples (Al, Fe, Mn, Mg, Ti, Sc, Na, Sr, Ca, Co, Cr, Ni, Cu, Pb, Zn, Cd, V, S, and As) were measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES, Model: ULTIMA, JOBIN-YVON Company, France). The detailed analytical procedures are given elsewhere [22, 23].

2.3.2 Ion analysis

6 inorganic anions (SO₄²⁻, NO₃⁻, Cl⁻, F⁻, PO₄³⁻, NO₂⁻), 5 organic acid anions (formic, acetic, oxalic, melonic, and methylsulfonic acid (MSA)) and 5 cations (NH₄⁺, Ca²⁺, K⁺, Mg²⁺, Na⁺) were analyzed by Ion Chromatography (IC, Dionex ICS 3000, USA). The aerosol was leached via deionized water and the pH of the filtrate measured by a pH meter was used as a parameter to directly denote aerosol acidity. Detailed information was given elsewhere [24].

2.2.4 The meteorological factor

The meteorological factors, including temperature, pressure, related humidity, wind direction, and wind speed, were observed at the Tazhong weather station.

2.2.5 Back trajectory

The National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used to compute forward and backward air trajectories at model vertical velocity using GDAS archived meteorological data set [25].

3 Results and discussion

3.1 General description of BC aerosol over the Taklimakan desert

Figure 2 shows the variation of daily BC concentration from Jan. 1, 2007 to Feb. 28, 2008. During the period of monitoring, BC concentration was in the range of 173.5–6191.9 ng m⁻³ with the annual average of 1605 ng m⁻³, which is one order of magnitude higher than that in Waliguan (130–300 ng m⁻³), a background observation station of the global air-pollution. The BC concentrations in 42.5% of the days in the whole year were over the average. The ratio of BC to PM₁₀ on annual average was 1.14%, indicating that the dust over the Taklimakan Desert has been polluted by the anthropogenic activity. On May 9, 2007, the highest peak of 6191 ng m⁻³ in the whole year occurred at the desert, which was even higher than the average BC in winter of 2006 in the urban mega-city, Shanghai. Such high BC concentration would suggest that even in the remote Taklimakan desert the air has been polluted by the anthropogenic activities. The great range of the daily BC variation was due to the great variation of the daily dust aerosol in Taklimakan desert. Taklimakan desert is enclosed by the surrounding high mountains, which would be in favor of the accumulation of BC aerosol produced in the surrounding oasis area and then transported in the local, regional, and even the long-range areas. On dusty days the black carbon, which adhered to the particle and deposited once on the ground, could be re-suspended into the air and led to the dramatic increase of BC in the dust aerosol.

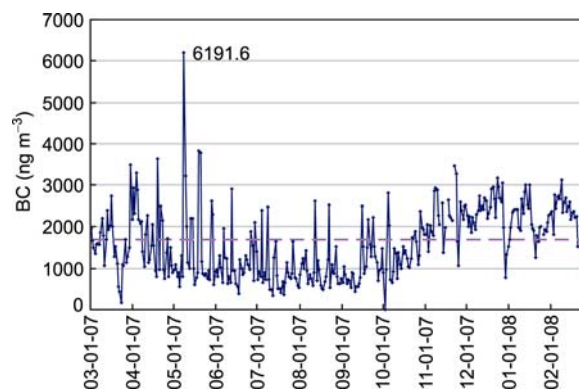


Figure 2 Variation of the daily BC concentration from Jan. 1, 2007 to Feb. 28, 2008.

3.2 Seasonal variation of BC in Taklimakan desert

BC aerosol over the desert was characterized by the obvious seasonal variation. BC concentrations in spring, summer, fall, and winter were 1621.9, 992.2, 1543.5, and 2261.7 ng m^{-3} , respectively, with the order of winter > spring > fall > summer. The highest concentration of BC in winter (in the range of 775–3117 ng m^{-3}), as compared to BC over the Musu desert (range: 900–6700 ng m^{-3}) [26], indicated that even the air over the desert has been polluted by BC. The higher concentration of BC in winter and spring must be mainly ascribed to the coal combustion for domestic heating. In spring BC was not only from the contribution of coal combustion, but also from the re-suspension of those particles containing BC, which were transported from those pollutant sources and deposited in the desert ever before. These particles containing BC were re-entrained into the air in the dust season with the strong wind. The lower BC in summer might be related to the wash-out by rainfall, though the amount of precipitation was extremely small. In fall, BC aerosol concentration rose with the increase of coal heating.

Figure 3 shows the monthly BC concentration over the centre of the desert in 2007. The maximum concentration among the twelve months was 2735.2 ng m^{-3} in January, and BC in November and December were 2238.2 and 2340.1 ng m^{-3} , respectively. In the period of dust, i.e., in March, April, and May, BC were 1596.2, 1732.0, and 1541.1 ng m^{-3} , respectively, which were lower than those in winter. In July BC was in the minimum of 877.5 ng m^{-3} . The variation of monthly BC was similar to that of BC in the urban city [27], which indicated clearly that BC in the desert was directly influenced by the anthropogenic activities, i.e., the emission from the industrial and agricultural activities at the surrounding oasis.

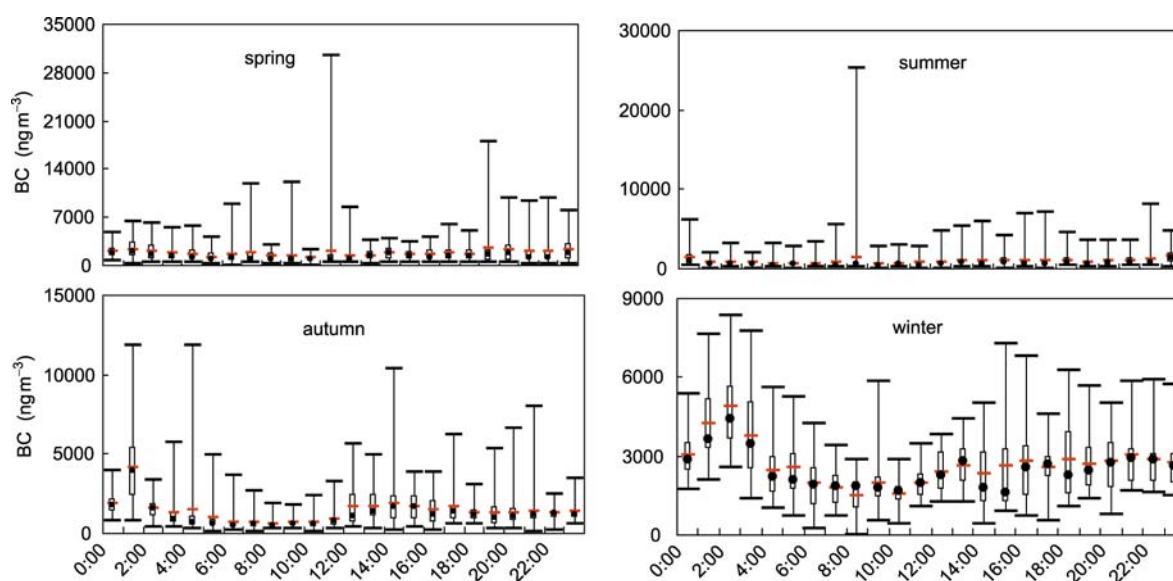


Figure 4 Variation of the hourly mean concentration of BC in four seasons.

3.3 Day and night variation of BC

Figure 4 displays the hourly variation on average of BC on a whole day in January, April, July, and October, as the representative day in winter, spring, summer, and fall. Figures 4(a), (b), (c), and (d) all show a peak at 0:00–2:00 and a vale at 8:00–11:00, which was just the reverse of that in the urban area, where the maximum occurred within 8:00–11:00 in a day. The strong wind with high speed and the active convection in daytime bring BC from the surrounding oasis into the desert; while at nighttime the stable atmospheric structure with lower wind speed keeps BC to be accumulated and reach the peak at midnight. The ratios of BC at night to BC in the day (N/D) were all larger than the unity (Figure 5), suggesting that BC at nighttime was greater than that in daytime. In most months, the ratios were within 1.17–1.50, which verified that there was a remarkable accumulation of BC at night. In April, May, June, and July, the ratios were less than those in other months, which could be due to the dust storm season, when the strong wind

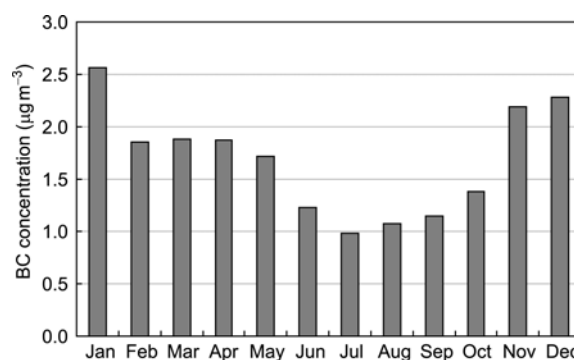


Figure 3 Variation of the monthly mean concentration of BC over Taklimakan Desert in 2007.

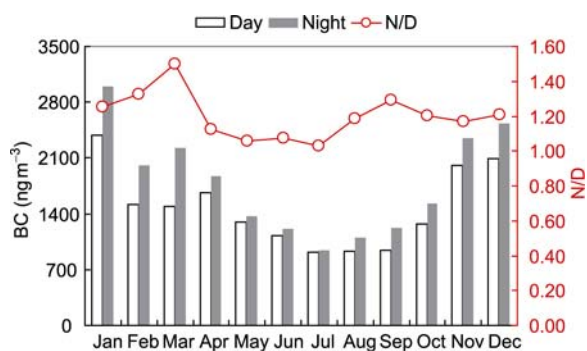


Figure 5 The monthly mean concentration of PM_{10} and the ratios of BC to PM_{10} .

would blow at both day and night and there would be no big difference between day and night.

3.4 Sources of BC over Taklimakan

3.4.1 Correlation analysis and primary factor analysis of BC with other components in dust aerosol

Table 1 lists the correlation coefficients of BC with other components in dust aerosol collected over Taklimakan Desert in the spring of 2007. The correlation coefficients of BC with As, Pb, and Cd reached 0.62, suggesting that BC was mainly from anthropogenic activities as it was well known that As, Pb, and Cd in aerosol were derived from the anthropogenic pollution. Table 2 shows the result of the primary factor analysis of BC with other elements and ions in aerosol. The four factors could explain 94.7% of the total variance. Factor 1, which could explain 83.3% of the variance and included almost all of the species except NH_4^+ , could represent all of those species which were from the surface soil of the desert and lifted by the strong wind and entrained to the air. BC was one among those species and it was from the accumulation of the deposited aerosol that was transported in the local, regional, and even long-range areas from other pollution sources. As mentioned above, in the surrounding areas of the desert there are oasis areas, where the anthropogenic activities could produce those pollutants,

Table 1 Correlation coefficients (r) of BC with other species in the aerosol at Taklimakan Desert

Species	Mass	F ⁻	HCOO ⁻	Cl ⁻	NO ₂	NO ₃ ⁻	C ₃
r	0.52	0.56	0.20	0.39	0.12	0.44	0.60
species	SO ₄ ²⁻	C ₂ O ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺
r	0.50	0.25	0.41	-0.01	0.54	0.60	0.56
species	Al	As	Ca	Cd	Co	Cr	Cu
r	0.55	0.63	0.53	0.60	0.66	0.61	0.67
species	Fe	Mg	Mn	Na	Ni	P	Pb
r	0.56	0.60	0.66	0.40	0.61	0.59	0.62
species	Sr	Ti	V	Zn	S		
r	0.59	0.55	0.66	0.69	0.48		

Table 2 Principle component factor analysis of the components in the aerosol at Taklimakan Desert

	Factor 1	Factor 2	Factor 3	Factor 4
Mass	0.96	-0.07	0.15	-0.02
BC	0.58	0.54	-0.12	0.21
Cl ⁻	0.83	-0.21	0.03	0.06
NO ₃ ⁻	0.51	0.36	0.71	-0.29
SO ₄ ²⁻	0.97	-0.18	0.09	0.10
C ₂ O ₄ ²⁻	0.75	-0.19	0.01	0.58
Na ⁺	0.94	-0.28	0.04	0.14
NH ₄ ⁺	-0.33	0.72	0.24	0.48
K ⁺	0.98	-0.12	0.12	0.02
Mg ²⁺	0.93	0.07	0.26	-0.09
Ca ²⁺	0.96	-0.12	0.01	0.10
Al	0.98	-0.01	0.06	-0.05
As	0.74	0.41	-0.43	-0.16
Ca	0.99	-0.07	0.04	-0.01
Cd	0.98	0.03	-0.14	-0.01
Co	0.99	0.09	-0.06	-0.05
Cr	0.99	0.04	-0.04	0.00
Cu	0.96	0.21	-0.10	-0.06
Fe	0.98	0.01	0.04	-0.03
Mg	0.99	0.03	0.05	-0.04
Mn	0.99	0.08	-0.08	-0.03
Na	0.95	-0.25	0.04	0.09
Ni	0.99	0.04	-0.05	0.00
P	1.00	-0.03	-0.04	-0.01
Pb	0.85	0.27	-0.17	-0.11
Sr	0.99	-0.03	-0.07	0.00
Ti	0.99	-0.03	0.04	-0.05
V	0.98	0.12	-0.09	-0.05
Zn	0.91	0.13	-0.13	-0.07
S	0.97	-0.16	0.09	0.05
% of variance	83.3	5.3	3.3	2.7
cumulative %	83.3	88.6	91.9	94.7

including BC. Besides, the exposed sedimentary rocks distributed in those areas around the Tarim Basin could also form BC through weathering. Factor 2 included three species, BC, NH_4^+ , and As. NH_4^+ and As were well known to be from pollution sources, and BC was further verified to be from those anthropogenic activities, such as coal combustion and traffic vehicle. All of the results mentioned above demonstrated that BC in the air over the remote Taklimakan Desert originated from the anthropogenic activity.

3.4.2 Source analysis by comparison of BC in dust storm days (DS) with that in non-dust storm days (NDS)

Figure 6 displays the variation of PM_{10} and the ratio of BC/ PM_{10} , which denoted the contribution of BC to the total mass of PM_{10} . Here, DS was operationally defined as those days with $PM_{10} > 500 \mu g m^{-3}$, and NDS with $PM_{10} <$

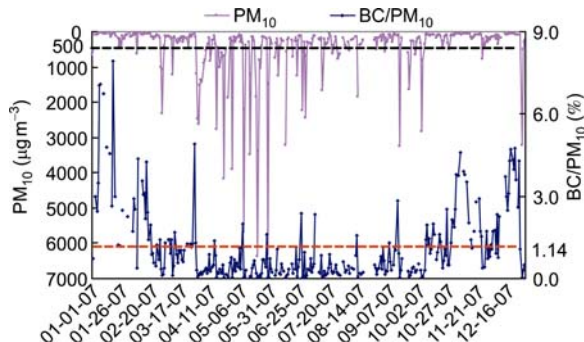


Figure 6 The ratio of BC to PM₁₀.

500 µg m⁻³. In spring and summer DS occurs frequently, accounting for >80% of DS in the whole year. BC aerosol exists mainly in fine particles, as it was reported that BC was mostly aggregated in a spherical grain with a diameter of ~50 nm [28]. The average ratio of BC/PM₁₀ in NDS was 1.78%, 11 times of that in DS (0.16%). In 67% of DS days

in the whole year the ratio of BC/PM₁₀ was less than the average of 0.16%, while in >50% of NDS days, the ratio of BC/PM₁₀ was higher than the average of 1.78%. Particularly, in January, the monthly average ratio of BC/PM₁₀ reached 7.9%. For January is in the heating period, the high BC contribution to the total mass of PM₁₀ would further indicate that BC in the air over the desert was mainly from the anthropogenic activity, such as the domestic heating with coal combustion. It is worthy to note that on a day with DS the ratio of BC/PM₁₀ was as high as 4.7%, which was found by further investigation to be related to an accident of the leakage of natural gas, which would again reveal that BC could be mainly from the anthropogenic activity.

3.4.3 Source of BC with back trajectory analysis

BC frequently acted as the tracer of anthropogenic activity for its inert chemical property. It could remain nearly one week in the air and be transported over a long distance even in the desert. Figure 7 shows the computed typical back

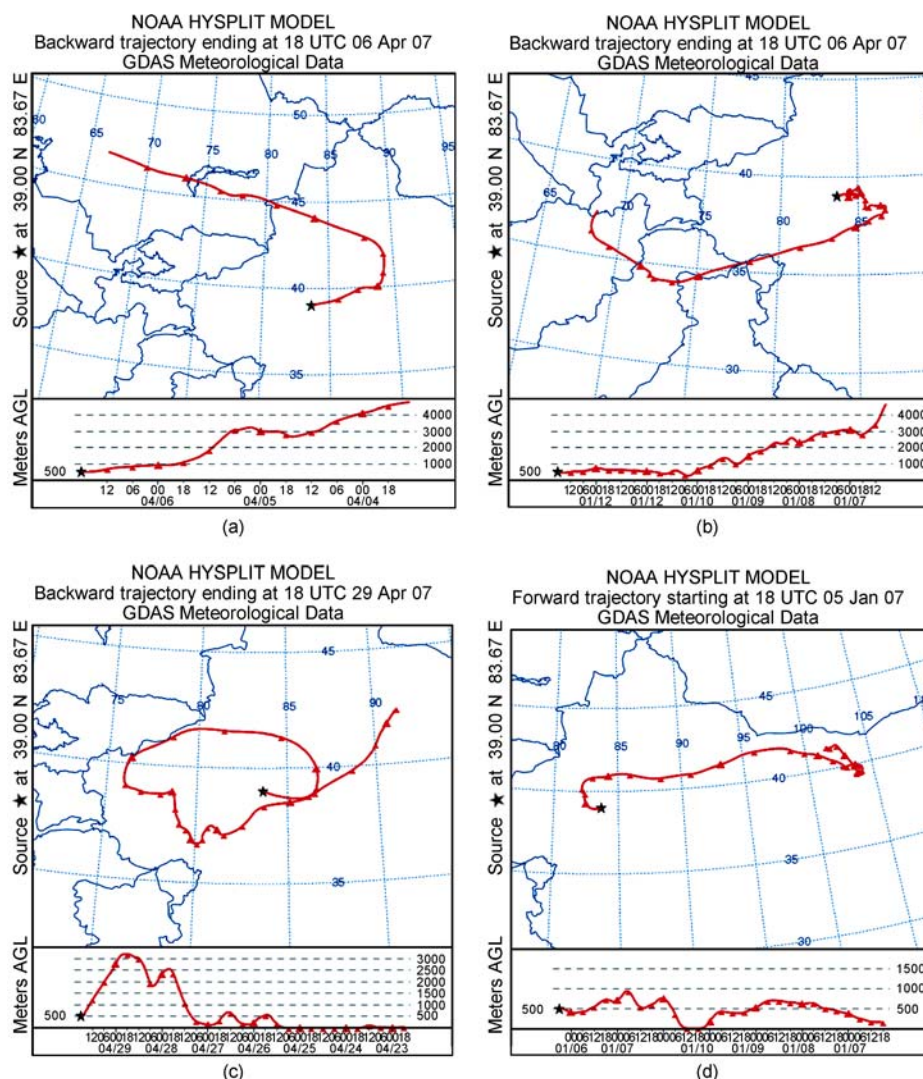


Figure 7 Types of back trajectories for long-range transport of dust aerosol in the Tazhong area. (a) NW; (b) NS; (c) local; (d) east.

trajectories of air mass at the center of Taklimakan Desert in four seasons using Model HYSPLIT 4.0 developed by atmospheric research laboratory of NOAA, USA. All of those back trajectories computed on each day in January, April, July, and October, to be the representatives of the four seasons, were grouped into 3 typical categories: Northwest (NW), Southwest (SW), and local directions. Figures 7(a), (b), (c), and (d) denoted the air mass from NW, WS, local, and East, respectively. To sum up the results, it was found that 70% of the back trajectories in the four seasons were from the west, in particular, from the northwest, while ~30% were from the local, and less than 1% were from the east. On the pathway of the air mass from the northwest and west direction, there were agriculture oasis belts with more industries, such as coal mines, cement factories, and oil refining factories, which would emit more fine particles with BC, and the air mass would carry them into the enclosed basin. In addition, a great number of fire spots were found in the north part of Kazakstan and the south part of Russia (<http://rapidfire.sci.gsfc.nasa.gov/>), which might be from biomass burning and/or natural fire. The results further verified that BC aerosol over the Taklimakan Desert originated from the uncontrolled anthropogenic activity with the westerlies.

4 The long-range transport of BC aerosol

Figure 8 shows that dust aerosol over the Taklimakan Desert

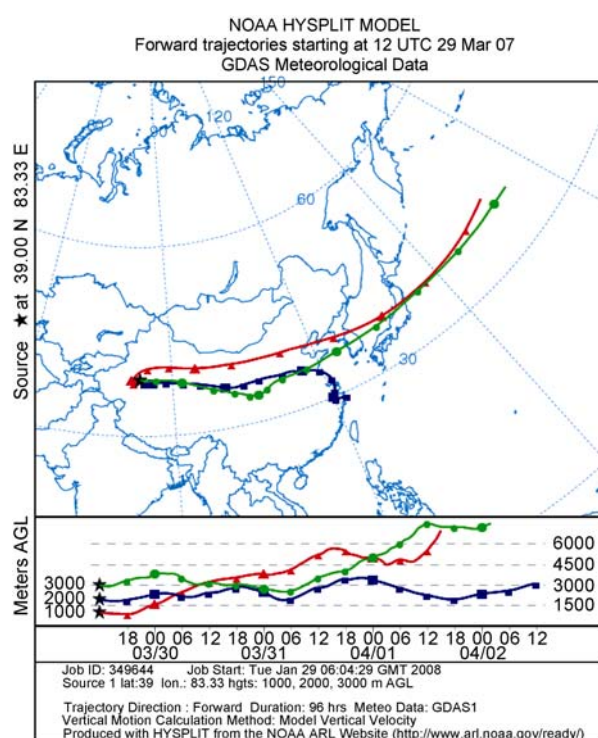


Figure 8 Transport of dust aerosol in the Taklimakan Desert on the regional and global scale.

could be transported through the center of China, and have an impact on the coastal areas of China, and even travel to the North Pacific Ocean. As mentioned above, although the ratio of BC to dust aerosol was small, the effect of BC on the radiative forcing was significant, for BC was the only component in aerosol to have the warming effect. It has been reported that if the ratio of BC to TSP could reach 6%, the optical depth could increase by 11%, the atmospheric radiation would reduce by 35%, and the atmospheric forcing would rise by 50% [29]. The annual emission of Asian dust was 10.4×10^6 ton (by PM_{10}) [30], and the emission of dust from the Taklimakan Desert was estimated to be 5.4×10^6 ton. With the average ratio of BC/PM_{10} , it was estimated that Asian dust could transport 6.3×10^4 ton of BC to the remote ocean, which would have an impact on the atmospheric radiative forcing and the global climate change.

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