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Molecular markers of biomass burning, fungal spores and biogenic SOA in the Taklimakan desert aerosols

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Abstract

Biogenic primary organic aerosols (POA) and secondary organic aerosols (SOA) are important organic constituents of atmospheric particulate matter (PM). In order to better understand the atmospheric abundances, molecular compositions and sources of the desert aerosols, biomass-burning tracers (e.g. levoglucosan), primary saccharides including fungal spore tracers, and SOA tracers from the oxidation of biogenic volatile organic compounds (e.g. isoprene, monoterpenes and sesquiterpene) have been studied in ambient aerosols from the Taklimakan desert, using gas chromatography-mass spectrometry. Results showed that the total concentrations of biomass-burning tracers at Hetian (177e359 ng m 3, mean 233 ng m 3 in PM2.5) in the south rim of the desert were much higher than those at Tazhong (1.9 e8.8 ng m 3 in PM2.5 and 5.9e32 ng m 3 in TSP) in the central Taklimakan desert. Molecular markers of fungal spores were also detected in all the desert aerosols, highlighting the importance of primary bioaerosols in the Asian dust particles. A specific pattern of the dominance of 2-methylglyceric acid over 2-methyltetrols and C5-alkene triols was found in the Taklimakan desert aerosols, especially during the dust storm events, which is different from the 2-methyltetrols-dominated pattern in other ambient aerosols. Our results provide direct evidence on the biogenic POA and SOA tracers in the Taklimakan desert region, which help to better understand their impact on the aerosol chemistry in the down-wind regions.

INTRODUCTION

Atmospheric aerosols play important roles on global climate by radiative forcing, and have serious negative impacts on human health (P€oschl, 2005; Pope et al., 2009). Organic aerosols, typically a large fraction of aerosol particles, are ubiquitous in the Earth's atmosphere (de Gouw and Jimenez, 2009). They originate from a variety of sources and atmospheric processes. For example, primary organic aerosols (POA) are directly emitted from both natural sources (e.g. plant emission, soil and dust suspension, and sea spray) and anthropogenic sources (e.g. industrial emission, traffic emission and biomass burning) (Viana et al., 2013; Escudero et al., 2015). Secondary organic aerosols (SOA) are those originating from homogenous and heterogeneous reactions of volatile organic compounds (VOCs) in the atmosphere from biogenic and anthro- pogenic emissions (Claeys et al., 2004; Hallquist et al., 2009; Escudero et al., 2015).

Both POA and SOA are important components of carbonaceous aerosols in the Earth's atmosphere. However, their abundances vary among different locations. de Gouw and Jimenez (2009) reported that biomass burning and biogenic SOA formation are major sources of organic aerosols globally, especially in the tropical re- gions, whereas anthropogenic emissions are mostly located at northern midlatitudes. In addition to biomass-burning emission and biogenic SOA formation, mineral dust is one of the largest contributors to atmospheric aerosols with an estimated emission flux of 1000e3000 Tg yr ^{1} (Ginoux et al., 2001; Dentener et al., 2006; Ginoux et al., 2012). This is especially true in the “Dust Belt” of the Northern Hemisphere, which is the largest and most persistent source of dust in the world (Wu et al., 2009). In East Asia, the Gobi desert in northern China and Mongolia and the Takli- makan Desert in western China are the main source regions of the Asian dust aerosols (Ginoux et al., 2001; Sun et al., 2005). On a hemispheric to global scale, Asian dust aerosols have been shown to be the main source of dust deposited in the North Pacific and the Greenland ice sheet (Duce et al., 1980; Wu et al., 2009).

During the past decade, significant efforts have been made to understand the molecular compositions of organic aerosols in ur- ban, rural, marine, and even the Polar Regions (Graham et al., 2003; Wang et al., 2006; Fu et al., 2011, 2013; Hu et al., 2013; Ding et al., 2014). Asian dust aerosols are generally considered to be primarily composed of mixtures of quartz, clays, micas, feldspars, carbonates and other minor minerals (Sullivan et al., 2007), and have been extensively investigated in terms of their roles in atmospheric chemistry (Falkovich et al., 2004; Sullivan et al., 2007, 2010; Wang et al., 2015), cloud condensation nuclei (CCN) activities (Shi et al., 2008), radiative properties and thus environment and climate ef- fects (Buseck and Posfai, 1999; Seinfeld et al., 2004; Ginoux et al., 2012; Huang et al., 2014; DeMott et al., 2015). Mineral dust aero- sols can internally mix with anthropogenic inorganic and organic aerosols via heterogeneous reaction and/or adsorption during long- range transport (Seinfeld et al., 2004). It is important to understand the organic aerosol composition at a molecular level in relation to photochemical oxidation, long-range atmospheric transport, and coating processes on the surface of mineral dust particles (Robinson et al., 2006; Rudich et al., 2007).

Asian dust aerosols have also been reported to contain biological particles, such as fungal spores, bacteria and viruses (Wu et al., 2004; Maki et al., 2014), which are relevant to the occurrence of human diseases and public health problems associated with acute toxic effects, allergies, and asthma (Bush and Portnoy, 2001; Po€schl, 2005). Thus it would be interesting to investigate the atmospheric abundances of arabitol and mannitol, the marker compounds of airborne fungal spores (Bauer et al., 2008), in dust aerosol particles in their source regions. Furthermore, to date, there is still a lack of knowledge about important biogenic SOA tracers, such as 2- methyltetrols from the photooxidation of isoprene (Claeys et al., 2004) in the desert aerosols.

The objective of the present study is to investigate the atmo- spheric levels of molecular markers for biomass burning, fungal spores, and biogenic SOA in the Taklimakan desert aerosols. Although a small number of aerosol samples were collected in April when the dust episodes prevail over the East Asia, such a pre- liminary study can help elucidate the organic molecular composi- tion of the desert aerosols, their contributions to organic aerosol loadings in the source regions and in the downwind regions via long-range atmospheric transport.

EXPERIMENTAL SECTION

Sample collection

The Taklimakan desert is the second largest shifting sand desert in the world, located in the Tarim Basin with an area of 337,000 km^2. As much as 85% of the desert consists of shifting sand dunes (Sun and Liu, 2006). Aerosol sampling was conducted in an intensive ground-monitoring network at Tazhong (TZ, 83.67°E, 39.00°N, 1090 m a.s.l.) and Hetian (HT, 79.92°E, 37.12°N, 1450 m a.s.l.) (Fig. 1) during April 16 – 21, 2008. Tazhong is a small isolated town (about 1200 residents) located in the central Takli- makan desert, while Hetian is a city (population 320,000) located in the southwestern rim of the Taklimakan desert. Aerosol samples were collected for 24 h (normally from 8:00 a.m. to 8:00 a.m. the next day, local time) on Whatman 41 filters (Whatman Inc., Maidstone, UK) using medium-volume samplers (Mode TSP/PM$_{10}$/ PM$_{2.5}$-II, Dike, Beijing) at a flow rate of 77.6 L min^{-1}. The samplers were set up at the rooftop of a building about 10 m above the ground. Both PM$_{2.5}$ (n ¼ 5) and TSP (n ¼ 5) samples were collected simultaneously at the Tazhong site, while only PM$_{2.5}$ samples (n = 4) were available at Hetian in the present study. According to the guidelines on dust observation of the Meteorological Obser- vation Criteria issued by the National Weather Bureau of China, the sampling period at Tazhong was divided into non-dust period (ND: April 16 – 17), floating dust period (FD: April 18) and dust storm period (DS: April 19 – 21). While at Hetian, the sampling period was divided into ND (April 17 – 18) and FD (April 19 – 20). The dust ep- isodes were also revealed by the CALIOP satellite data that were available from the Atmospheric Science Data Center. Fig. 2 shows the dust aerosols prevailed over the Taklimakan desert during April 18 – 20, 2008. The samples were kept in polyethylene plastic bags immediately after sampling and then reserved in a refrigerator at – 18 °C.

Chemical measurements

Filter aliquots of the aerosol samples were analyzed for inorganic ions using ion chromatography and heavy metals using ICPAES. Detailed information on the analytical procedures can be found in a previous study (Wang et al., 2011).

Another filter cut of the aerosol samples was extracted three times with dichloromethane/methanol (2:1; v/v) under ultra- sonication for 10 min. The solvent extracts were filtered through quartz wool packed in a Pasteur pipette, concentrated using a rotary evaporator under vacuum, and then blown down to near dryness with pure nitrogen gas. The extracts were reacted with 50 µl of N, O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA, purchased from SigmaeAldrich) containing 1% trimethylsilyl chloride and 10 µl of pyridine at 70 °C for 3 h. After the reaction, derivatives were diluted by the addition of 140 µl of n-hexane containing 1.43 ng μl^{-1} of the internal standard (C_{13} n-alkane) prior to analysis by gas chromatography/mass spectrometry (GC/MS).

GC/MS analyses were performed on a Hewlette–Packard model 6890 GC coupled to HewlettePackard model 5973 MSD. The GC separation was achieved on a DB-5 fused silica capillary column (30 m x 0.25 mm i.d., 0.25 µm film thickness) with the GC oven temperature programmed from 50 °C (2 min) to 120 °C at 15 °C min^{-1} and then to 300 °C at 5 °C min^{-1} with a final isotherm hold at 300 °C for 16 min. Helium was used as the carrier gas at a constant flow rate of 1.0 ml min^{-1}. The GC injector and MS ion source temperatures were maintained at 280°C and 230°C, respectively. The mass spectrometer was operated in the electron impact (EI) mode at 70 eV and scanned over the range of 50 – 650 Da. Individual compounds (TMS derivatives) were identified by comparison of mass spectra with those of authentic standards or literature data (Claeys et al., 2004; Jaoui et al., 2007; Hu et al., 2008; Lin et al., 2012). Following fragment ions were used for quantifications: m/z 217 and 204 for sugar compounds, m/z 193 for 4-hydroxybenzoic acid, m/z 194 for vanillin, m/z 297 and 312 for vanillic acid, m/z 327 for syringic acid, m/z 239 for dehydroabietic acid, m/z 219 and 203 for 2-methylglyceric acid, m/z 219 and 277 for 2-methyltetrols, m/z 231 for C_5-alkene triols, m/z 171 for pinonic and pinic acids, m/z 349 for 3-hydroxyglutaric acid, m/z 405 for 3- methyl-1, 2, 3-butanetricarboxylic acid (MBTCA), and m/z 383 for b-caryophyllinic acid. Most of the GC/MS response factors were determined using authentic standards. Because of the lack of authentic standards for some SOA tracers, 2-methylglyceric acid, 3- methyl-1, 2, 3-butanetricarboxylic acid and β-caryophyllinic acid were estimated using glyceric acid, malic acid and pinic acid as surrogate standards, respectively. Blank filters were analyzed by the procedure used for the real samples. The results showed no contamination for any target compound. Recoveries for the stan- dards were generally better than 80%. Relative standard deviation of the concentrations based on duplicate analysis was generally <10% (Fu et al., 2011).

RESULT AND DISCUSSION

Table S1 presents the concentrations of organic marker com- pounds, together with some water-soluble inorganic ions measured in the Taklimakan aerosols. Fig. 3 shows the abundances of organic compound classes in different aerosol types. In general, the total concentrations of biomass-burning tracers in $PM_{2.5}$ samples were roughly two orders of magnitude higher at Hetian (177 – 359 ng m^{-3}) than those at Tazhong (1.9 – 8.8 ng m^{-3}), indicating that biomass burning is more significant in the south rim (the Hetian site) than the central region (the Tazhong site) of the

Taklimakan desert. Sugars/sugar alcohols, monoterpene SOA tracers and the sesquiterpene-SOA tracer were also more abundant at Hetian than those at Tazhong. However, the concentrations of isoprene SOA tracers in fine samples ($PM_{2.5}$) at Tazhong were higher than those at Hetian. Total water-soluble inorganic ions in $PM_{2.5}$ samples at Tazhong were double those at Hetian (Table 1), while the concentrations of NH_4^+ and NO_3^- were higher at Hetian than those at Tazhong, probably due to the higher anthropogenic emissions (e.g., traffic emission and agricultural emission) at the urban site of Hetian.

Biomass burning tracers

Levoglucosan (1,6-anhydro-β-D-glucopyranose), a specific biomass-burning tracer formed by the pyrolysis of cellulose and hemicellulose (Simoneit et al., 1999; Simoneit, 2002), is considered as the largest source of primary, fine organic aerosols in the atmosphere (Bond et al., 2004). It is water-soluble and thus contributes to water-soluble organic carbon (WSOC) contained in aerosol particles. During the study period, the concentrations of levoglucosan in $PM_{2.5}$ samples ranged from 148 to 313 ng m^{-3} with an average of 198 ng m^{-3} at Hetian, which were consistent with those reported in fourteen Chinese megacities (Wang et al., 2006). Such a high level of levoglucosan at Hetian indicates that biomass-burning emission should be a significant source of organic aerosols in the south rim of the Taklimakan desert. Another possible source is the long-range transport of atmospheric aerosols from the regions in the Middle East and South Asia (Fig. S1) to Hetian. A recent study reported that biomass burning aerosols in these regions can even penetrate the Himalayas to its north slope (Cong et al., 2015). In the central region (Tazhong) of the Taklimakan desert, however, the concentrations of levoglucosan in $PM_{2.5}$ samples were only 0.99 –5.3 ng m^{-3} (mean 2.6 ng m^{-3}) (Tables 1 and 3). During the same sampling period, the levels of levoglucosan in TSP were 3.0 –14 ng m^{-3} (5.9 ng m^{-3}). The atmospheric levels of levoglucosan in the central regions of the Taklimakan desert are comparable with those reported in remote continental and marine regions, and one order of magnitude higher than those in the polar regions (Table 2).

Galactosan and mannosan, the isomers of levoglucosan, were identified in all the desert samples (Table S1). They are thermal decomposition products of hemicellulose, whereas levoglucosan is a thermal decomposition product of cellulose (Simoneit, 2002). Hard wood contains higher amount of cellulose than hemicellulose. Thus, the concentration ratio of levoglucosan to mannosan (L/M) can be used to differentiate hard wood (angiosperm) and soft wood (gymnosperm) burning (Simoneit, 2002; Schmidl et al., 2008; Verma et al., 2015). Previous studies have reported that L/M ratios from crop-residue burning are generally >40; L/M ratios from hardwood burning are in a range of 15 – 25 and those of softwood are 3 – 10 (Schmidl et al., 2008; Engling et al., 2009; Fu et al., 2012a; Mkoma et al., 2013). For example, high L/M ratios were reported for smoke aerosols from chamber burning of rice straw (40 – 42) (Engling et al., 2009) and high mountain aerosols affected by the severe field burning of wheat straw (40 – 46) in the North China Plain (Fu et al., 2012a). At the Hetian site, the L/M ratios ranged from 25 to 35, which were similar between FD and ND periods. At the Tazhong site, the L/M ratios varied from 23 to 59 and 33 to 60 in PM2.5 and TSP samples, respectively. Thus, high L/M ratios (23 – 60) found in the present study suggest that crop residues, herbaceous plants and hardwood are the dominant types of fuels burned in the Taklimakan region.

Phenolic compounds produced from lignin combustion were also measured in the Taklimakan aerosols, including vanillin, vanillic acid, syringic acid and dehydroabietic acid (Table S1). The burning products of hardwood lignin are enriched in vanillic acid, vanillin, and syringic acid; the burning of softwood generally produces vanillin, homovanillic acid, vanillic acid and so on; and the burning of grasses mainly produces syringic acid, vanillin and vanillic acid (Oros and Simoneit, 2001b, a; Oros et al., 2006). 4 Hydroxybenzoic acid is produced by the burning of grasses and other non-woody vegetation (Simoneit, 2002); its concentrations in the $PM_{2.5}$ samples in Hetian (6.4 – 9.9 ng m^{-3}) were about twenty times higher than those (0.11 – 0.57 ng m^{-3}) in Tazhong. Dehy- droabietic acid, a more specific biomass-burning tracer of conifer resin, was also detected in all the desert aerosol samples with higher levels at Hetian than at Tazhong (Table S1). However, ratios of vanillin to dehydroabietic acid (Vani/Dehy) in the Hetian $PM_{2.5}$ aerosols (1.2 – 3.1) are similar to those (1.1 – 2.6) in Tazhong $PM_{2.5}$ aerosols, suggesting that similar types of biofuels are widely used in the Taklimakan region.

Sugars and sugar alcohols

Primary saccharides consisting of arabinose, fructose, glucose, sucrose, and trehalose, as well as some sugar polyols including arabitol, mannitol, and inositol were detected in the Taklimakan aerosols (Table S1). Sucrose was the dominant sugar compound found in the samples, followed by glucose and fructose (Fig. 4). Fructose and glucose originate from numerous primary biogenic sources, such as terrestrial plant pollen, fruits and detritus (Speranza et al., 1997). Sucrose is the dominant component of airborne pollen grains (Pacini, 2000; Fu et al., 2012b); it plays a key role in plant flowering processes. Furthermore, their concentrations were much higher in the TSP samples than those in the fine aerosol particles (Fig. 4). In a previous study at the Gosan site, Jeju Island, Fu et al. (2012b) reported that sucrose is the dominant species among the detected saccharides with a maximum concentration up to 1390 ng m^{-3} in April.

Arabitol and mannitol are tracers for airborne fungal spores (Bauer et al., 2008). Their concentrations were higher at Hetian than those at Tazhong (Fig. 4), suggesting that primary bioaerosols such as fungal spores are relatively minor in the central Taklimakan region. By comparison with those reported in previous studies, the atmospheric levels of arabitol and mannitol in the Taklimakan region were similar to those in remote marine aerosols (Table 2), representing the background level of fungal spores in the Earth's atmosphere. Interestingly, higher concentrations of arabitol than mannitol were observed in the present study; such a pattern was also observed in the atmospheric aerosols at Alert in the Canadian High Arctic (Fu et al., 2009b). In the marine aerosols collected during a round-the-world cruise, the levels of arabitol were slightly lower than or similar to those of mannitol (Fu et al., 2011). At the Chichi-jima Island in the western North Pacific, the atmospheric concentrations of arabitol were lower than those of mannitol, with a seasonal pattern characterized by higher concentrations in summer than in winter (Chen et al., 2013). In the tropospheric aerosols collected at the summit of Mt. Fuji (3776 m a.s.l.), the concentrations of mannitol were double those of arabitol (Fu et al., 2014). Although both arabitol and mannitol are molecular marker compounds of fungal spores, their abundances may change among different species. Fungi are widely distributed in all terrestrial ecosystems and the kingdom Fungi is one of the most diverse groups of organisms on Earth (Tedersoo et al., 2014). In addition to soil fungi, a recent study (Fro€hlich-Nowoisky et al., 2012) has proven the high fungal diversity in the atmosphere over land and oceans.

The concentrations of disaccharide trehalose in TSP samples (1.2 – 14 ng m^{-3}, mean 6.4 ng m^{-3}) were four times higher than those (0.36 – 3.4 ng m^{-3}, 1.6 ng m^{-3}) in $PM_{2.5}$ samples at Tazhong. Higher concentrations of trehalose were also observed during the DS period than FD and ND periods (Table 3). Trehalose is present in a variety of microorganisms (fungi, bacteria and yeast), and a few species of higher plants and invertebrates (Medeiros et al., 2006). As a fungal metabolite, the enrichment of trehalose in aerosol particles is frequently accompanied by the resuspension of soil particles (Simoneit et al., 2004; Feng and Simpson, 2007) and un- paved road dust (Simoneit et al., 2004). High abundance of trehalose in marine aerosols observed at the Gosan site in March – April was related to the Asian dust events (Fu et al., 2012b), indicating the importance of soil resuspension/dust aerosols as the source.

The biogeographic distribution of primary biological aerosols are relevant for the Earth system, climate, and pubic health on local, regional and global scales (Fro€hlich-Nowoisky et al., 2012). Here, the atmospheric concentrations of molecular markers for primary bioaerosols over the Taklimakan desert are comparable to those over marine regions, which highlights their importance in the process of cloud formation and precipitation over the desert region because fungal spores and other bioaerosols are considered to be relevant as ice nuclei or giant cloud condensation nuclei (Christner et al., 2008; Pratt et al., 2009). Furthermore, dust aerosols containing biological particles such as pollen could be recorded in ice cores of inland glaciers via the long-range atmospheric transport and deposition, which provides useful information on the dust events together with the regional biological diversity in the history.

Biogenic SOA tracers

Went (1960) reported that the oxidation of VOCs emitted from forests could lead to the formation of organic aerosols. Since then, considerable efforts have been devoted to understanding the formation processes of SOA from biogenic VOCs, including isoprene, monoterpenes, sesquiterpenes, and oxygenated hydrocarbons (e.g. alcohols, aldehydes and ketones) through field observations and chamber and modeling studies (Hallquist et al., 2009 and references therein). Despite its large flux, isoprene had not been generally considered to be an SOA precursor due to the high volatility of its known reaction products. Claeys et al. (2004) first identified two diastereoisomeric 2-methyltetrols as oxidation products of isoprene in the Amazonian rain forest aerosols. Since then, these compounds have been detected in ambient aerosol samples from different geographical locations in the world (Kleindienst et al., 2007; Hu et al., 2008; Fu et al., 2009a; Hallquist et al., 2009; Kourtchev et al., 2009; Hu et al., 2013). However to date, little is known about the biogenic SOA tracers including isoprene oxidation products in desert regions.

Six compounds were identified as isoprene SOA tracers in the Taklimakan desert aerosols, i.e., 2-methylglyceric acid, three C5- alkene triols, and two 2-methyltetrols (2-methylthreitol and 2-methylerythritol). 3-Methyltetrahydrofuran-3, 4-diols, which have been proposed to be formed through the acid-catalyzed intra- molecular rearrangement of isoprene epoxydiols under low-NOx conditions (Lin et al., 2012), were not detectable in these samples. The concentrations of 2-methylthreitol and 2-methylerythritol were very low in the $PM_{2.5}$ samples at Tazhong, ranging from 0.005 to 0.017 ng m^{-3} (mean 0.009 ng m^{-3}) and 0.007 – 0.036 ng m^{-3} (0.017 ng m^{-3}), respectively. This is reasonable because the Taklimakan desert is located in the hinterland of the Asian continent and has little water; very few oasis towns are located in the north, east and south regions of the desert including Hetian, where the concentrations of 2-methyltetrols (the sum of 2-methylthreitol and 2-methylerythritol, 0.22 ng m^{-3}) were about ten times higher than those at Tazhong (0.025 ng m^{-3}) in the central Taklimakan desert. In addition, relatively high concentrations of 2-methylthreitol and 2-methylerythritol were found at Hetian with average concentrations of 0.078 ng m^{-3} and 0.15 ng m^{-3}, respectively. The concentrations of 2-methyltetrols are similar to those (mean 0.074 ng m^{-3}) reported in Canadian High Arctic aerosols collected at Alert during winter and spring (Fu et al., 2009a), and are 1 – 3 orders of magnitude lower than those reported in urban and forest aerosols in low-to mid-latitudes (Wang et al., 2005; Kleindienst et al., 2007; Ding et al., 2014).

C_5-alkene triols, which are reported as photooxidation products of isoprene (Wang et al., 2005), were detected in all the samples with the concentration range of 0.003 – 0.071 ng m^{-3} (Table S1). 2-Methylglyceric acid (2-MGA) is possibly formed by further oxida- tion of methacrolein and methacrylic acid. The concentrations of 2-MGA in the desert aerosols were much higher at Tazhong than at Hetian (Table S1).

The detected monoterpene oxidation products include pinonic acid, pinic acid, 3-hydroxyglutaric acid (3-HGA), and 3-methyl-1, 2, 3-butanetricarboxylic acid (MBTCA). Total concentrations of monoterpene SOA tracers in $PM_{2.5}$ samples ranged from 0.18 to 0.99 ng m^{-3} (0.52 ng m^{-3}) at Tazhong versus 0.75 – 1.1 ng m^{-3} (0.87 ng m^{-3}) at Hetian. The formation of MBTCA can be explained by further reaction of cis-pinonic acid with OH radical (Szmigielski et al., 2007), thus it has been considered as a higher-generation product. However, MBTCA (mean 0.001 ng m^{-3}) was only detect- able at a trace level among the TSP samples at Tazhong. In remote marine aerosols over the Pacific and Atlantic Oceans, MBTCA was found to be less significant than those in the coastal aerosols (Fu et al., 2011), which suggests that MBTCA is not a dominant species in the pristine environments.

The emission of β-caryophyllene, one of the dominant sesqui- terpenes, varies significantly between vegetation species (Duhl et al., 2008). Concentrations of β-caryophyllinic acid, formed either by ozonolysis or photo-oxidation of β-caryophyllene rapidly in the atmosphere, were found to be up to 0.32 ng m^{-3} at Hetian, which are several times higher than those at Tazhong, being similar to the results of other biogenic SOA tracers. Such a temporal pattern can be explained by the stronger influence of biogenic VOCs emitted from vegetation near Hetian rather than Tazhong. The three-day back trajectories also showed that the air masses mainly originated from the southwest via mountain regions to the Hetian site (Fig. S1).

Specific distribution patterns of isoprene SOA tracers in the Taklimakan aerosols

Previous field measurements have often shown the predominance of 2-methyltetrols among the isoprene SOA tracers. For example, in a recent study on the spatial distributions of biogenic SOA tracers over China in summer, Ding et al. (2014) reported a 2-methyltetrols-dominated pattern with the ratios of 2-methylglyceric acid to 2-methyltetrols (2-MGA/2-MTs) to be lower than 1, which is consistent with field observations in other places (Kleindienst et al., 2007; Kourtchev et al., 2009). However, Fig. 5 clearly shows that 2-MGA was by far the dominant species in the Tazhong aerosol samples, followed by 2-methyltetrols and C_5-alkene triols. Furthermore, the relative abundance of 2-MGA was even enhanced during DS; the contribution of 2-methylglyceric acid to the sum of isoprene SOA tracers was up to 99% during DS. By contrast, the pattern of relative abundance at Hetian in the south rim of the desert differ from that at Tazhong during ND, where 2- methyltetrols are the dominant species, followed by 2-MGA and C5-alkene triols, being similar to those in marine aerosols collected during a round-the-world cruise (Fu et al., 2011). In addition, the relative abundance of 2-MGA was also strongly enhanced at Hetian during FD (Fig. 5).

The predominance of 2-MGA in the Taklimakan desert region, especially during FD and DS periods, suggests that its formation processes and/or atmospheric fates are different from other locations. Chamber experiments have demonstrated that 2-MGA is formed under high-NO_x conditions, while 2-methyltetrols are mainly generated under low-NO_x conditions (Surratt et al., 2006, 2010). In general, anthropogenic activities such as fossil fuel combustion and biomass burning can emit a large amount of NO_x and result in high concentrations of nitrate in polluted urban regions. In the Taklimakan desert region, the atmospheric level of NO_x are low due to weak anthropogenic activities (Wu et al., 2014). In situ production of NO_x from lightning and electoral storms may also contribute to the atmospheric level of NO_x (Price et al., 1997). However, such a potential emission source should be minor because lightning activities rarely occur in the desert region (Wu et al., 2014). Although it has been reported that the surface dust hosts a certain amount of nitrate in the Taklimakan desert (Wu et al., 2014), the concentrations of nitrate in TSP at Tazhong showed relatively little variation (1.4 – 6.2 µg m^{-3}) under different dust conditions compared with the large aerosol masses ranging from 108 µg m^{-3} during ND to more than 10 mg µg m^{-3} during DS (Table S1). This indicates that the atmospheric nitrate in the desert atmosphere is independent from desert soil (Wu et al., 2014).

In addition, heterogeneous reactions of nitrogen oxides (including HNO_3) on the preexisting dust particle are one of the main contributors to the production of nitrate (Sullivan et al., 2007). However, this process is not efficient in dry desert atmosphere because the liquid water is limited, which is the key for the heterogeneous reactions (Usher et al., 2003). In the present study, the concentrations of nitrate were higher in TSP samples (1.4 – 6.2 µg m^{-3}, mean 3.4 µg m 3) than in $PM_{2.5}$ samples (0.07 – 1.3 µg m^{-3}, 0.59 µg m^{-3}) at Tazhong. The level of nitrate in the present study is consistent with those reported by Wu et al. (2014), who proposed that the background nitrate in the Taklimakan desert is present in the form of nitric acid due to gas-phase conversion of NO_x on desert soil surface. In a thermodynamic equilibrium modeling study, Turpin et al. (1997) proposed that nitrate in the hot desert air should be in the form of nitric acid.

Recent studies (Surratt et al., 2010; Lin et al., 2012) have shown that IEPOX (isoprene epoxydiols, IEPOX = β-IEPOX + α-IEPOX) reactively uptakes onto sulfate aerosols to yield 2-methyltetrols, C_5-alkene triols, dimers, and IEPOX-derived organosulfates. At Tazhong, significantly higher levels of sulfate in TSP were observed during DS than those during FD and ND (Table 1).

Interestingly, the abundances of sulfate relative to the aerosol mass (2.5 – 5.8%) were almost constant. Such results indicate that the sulfate content was closely correlated with the mass loading of the desert aerosols. Using Fe, one of the leading elements in Earth crust as a typical tracer of desert surface soil, Wu et al. (2012) reported that the sulfate in the Taklimakan desert aerosols was not likely produced via atmospheric processes but originated from dust resuspension. Here, the ratio of sulfate to elemental Al, another specific tracer of soil-derived particles, also showed consistency in different samples of TSP and $PM_{2.5}$ at Tazhong and Hetian, further supporting the idea that the surface soil should be the dominant source of sulfate in the Taklimakan aerosols. Thus, the high content of sulfate in aerosols may favor the formation of 2-MGA.

In addition, laboratory studies have shown that relative humidity (RH) plays an important role on isoprene SOA formation: the formation of 2-methyltetrols was similar in both low- and high-RH conditions, while an enhancement of 2-methylglyceric acid was found in the low-RH conditions (Zhang et al., 2011). The dominance of 2-MGA over 2-methyltetrols in the aerosols collected at Tazhong, which is located in the central Taklimakan desert with an extreme continental-arid climate, is consistent with the idea that the low- RH leads to the enhanced formation of 2-methylglyceric acid in the atmosphere.

Surratt et al. (2010) reported that RH can affect the concentration ratio of C_5-alkene triols to 2-methyltetrols in the absence of NO_x. The ratios of C_5-alkene triols to 2-methyltetrols were found to be <0.10 in marine aerosols over the open oceans (Fu et al., 2011), while they were much higher (up to 0.66) over the coastal regions, such as off the California Coast, in the Indian Ocean near India and the South China Sea. The lower ratios in the pristine open oceans are in agreement with the low ratios (<0.10) obtained in laboratory experiments for the isoprene photooxidation in the absence of NO_x (Kleindienst et al., 2009). Here, the ratios of C_5-alkene triols to 2- methyltetrols were 0.10 – 0.23 in $PM_{2.5}$ at Hetian, which were lower than those (0.25 – 0.85) in the more arid region (Tazhong) in the central Taklimakan desert (Table 4).

CONCLUSION

Molecular markers of biomass burning, fungal spores, and biogenic secondary organic aerosols were studied for aerosol samples collected at Tazhong (TSP and $PM_{2.5}$) and Hetian ($PM_{2.5}$) in the Taklimakan desert in April 2008. Total concentrations of biomass-burning tracers at Hetian (177 – 359 ng m^{-3}, mean 233 ng m^{-3}) in the south rim of the desert were much higher than those (1.9 – 8.8 ng m^{-3} in $PM_{2.5}$ versus 5.9 – 32 ng m^{-3} in TSP) at Tazhong in the central Taklimakan desert. This study demonstrated that biomass-burning activities in the Asian arid region are on a background level. The concentrations of molecular markers for primary bioaerosols such as fungal spores in the Taklimakan desert aerosols are lower than or similar to those reported in remote marine regions. Total concentrations of biogenic SOA tracers in the $PM_{2.5}$ samples from Hetian (1.2 – 2.1 ng m^{-3}, mean 1.5 ng m^{-3}) were lower than those (0.71 – 9.2 ng m^{-3}, 2.7 ng m^{-3}) at Tazhong. Among the isoprene SOA tracers, the concentrations of 2-methylglyceric acid were more abundant than those of 2-methyltetrols and C_5-alkene triols in the Taklimakan desert aerosols. Such a predomi- nance of 2-methylglyceric acid in the desert aerosols was different from the 2-methyltetrols-dominated pattern in ambient aerosols collected from other geographical locations. Furthermore, the abundance of 2-methylglyceric acid relative to 2-methyltetrols was enhanced during the dust storm period. Our results strongly sug- gest the unique formation processes of isoprene SOA tracers in the desert region.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.10.087.

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LIST OF FUGURES AND TABLES

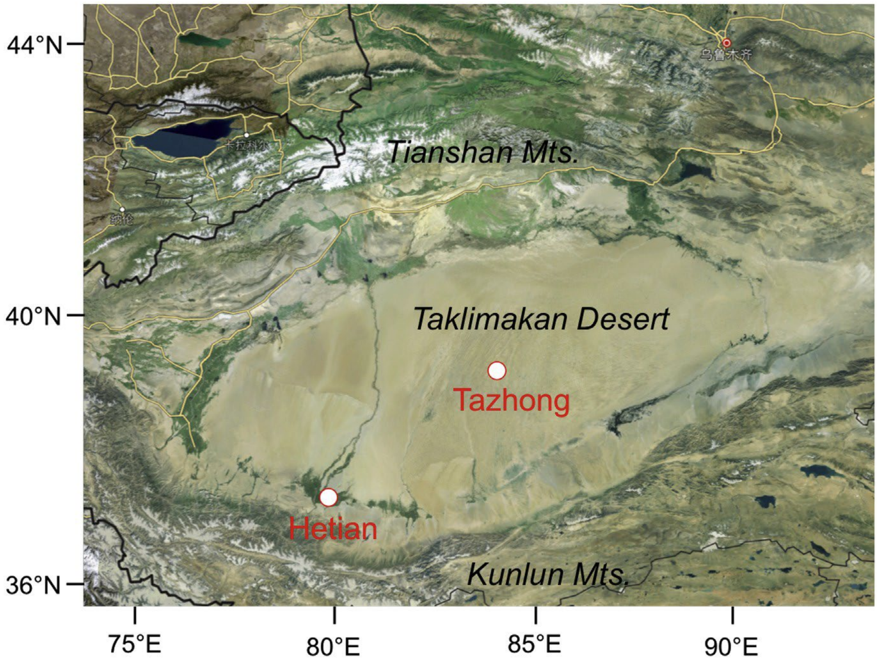


Fig. 1. A map showing the sampling sites of Tazhong (TZ) and Hetian (HT) in the Taklimakan Desert, China.

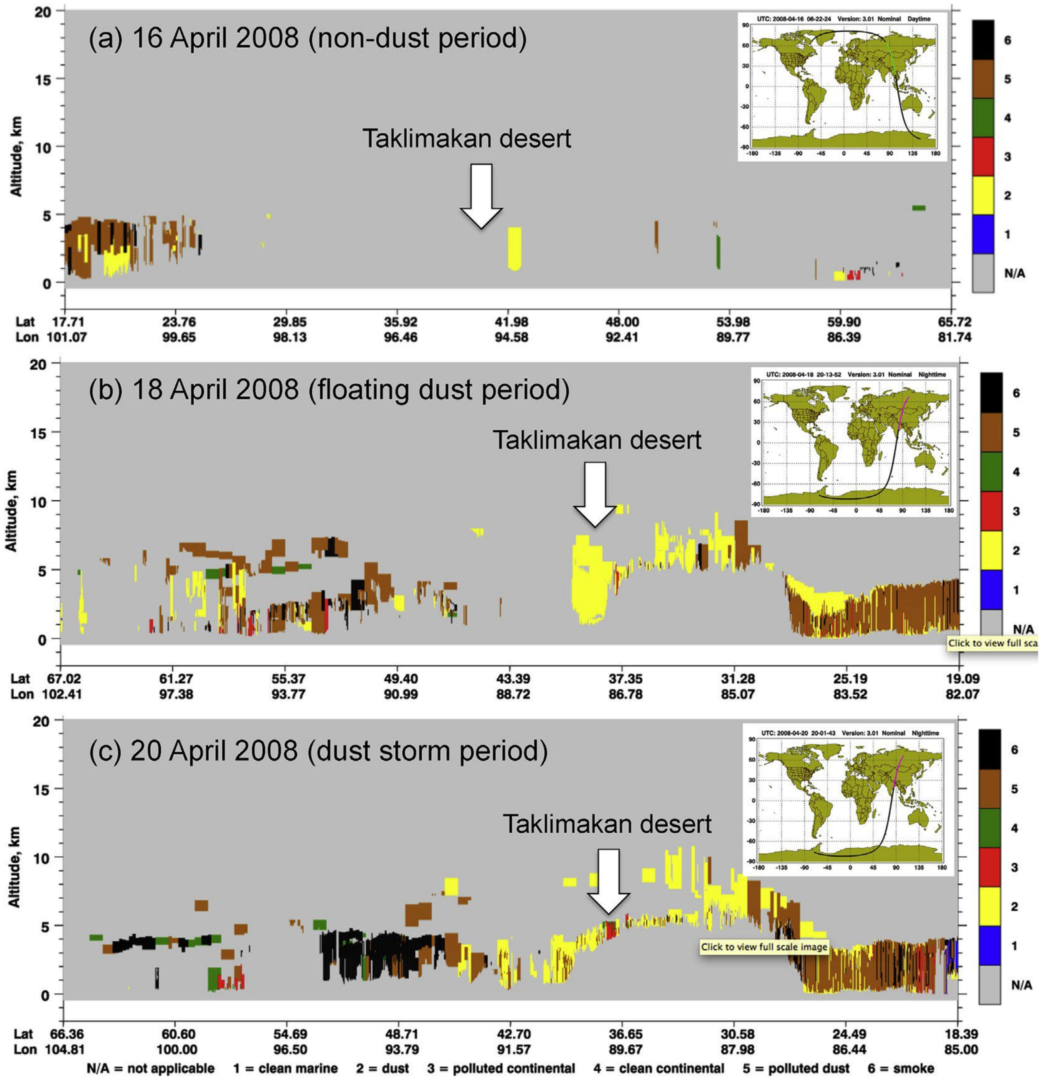


Fig. 2. CALIOP lidar transects across the Taklimakan desert during the sampling period (on 16 April, 18 April, and 20 April 2008, respectively) show the main aerosol types derived from the CALIOP observation. The inner panels in the figures show the orbit tracks across the sampling region and adjacent areas.

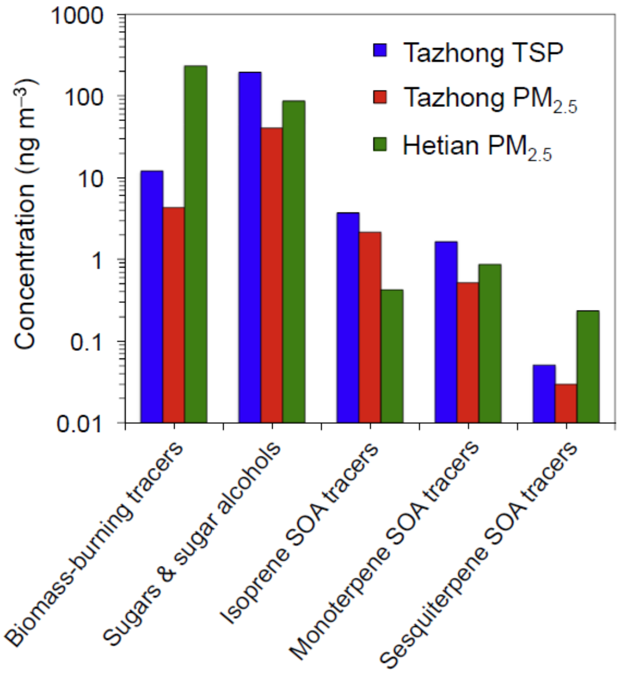


Fig. 3. Concentrations of organic compound classes measured in the Taklimakan desert aerosols.

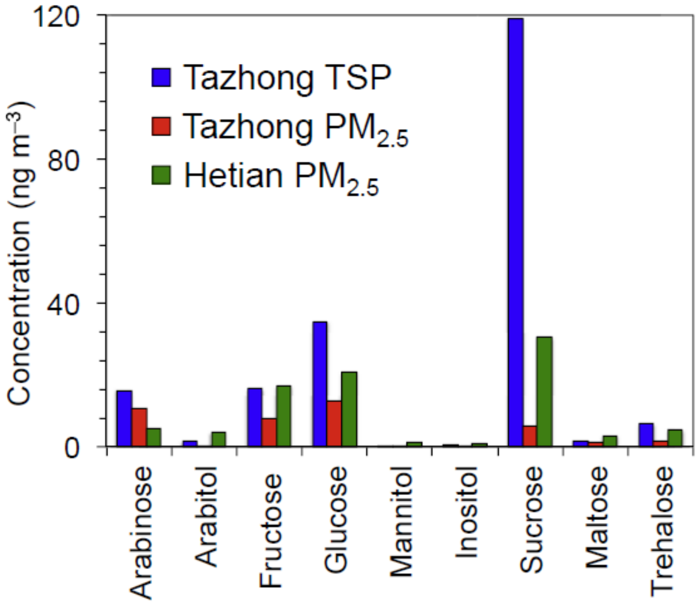


Fig. 4. Molecular distributions of sugars and sugar alcohols measured in the Takli- makan desert aerosols.

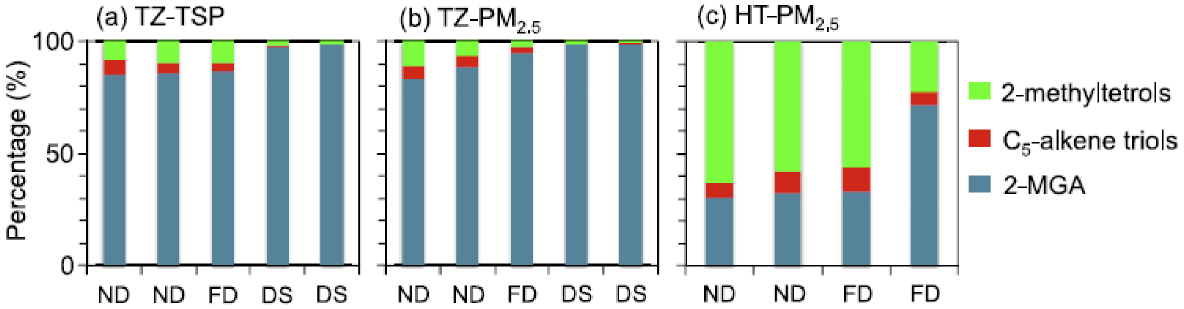


Fig. 5. Relative abundances of isoprene SOA tracers in the Taklimakan desert aerosols. ND ¼ non-dust period; FD ¼ floating dust period; DS ¼ dust storm period.

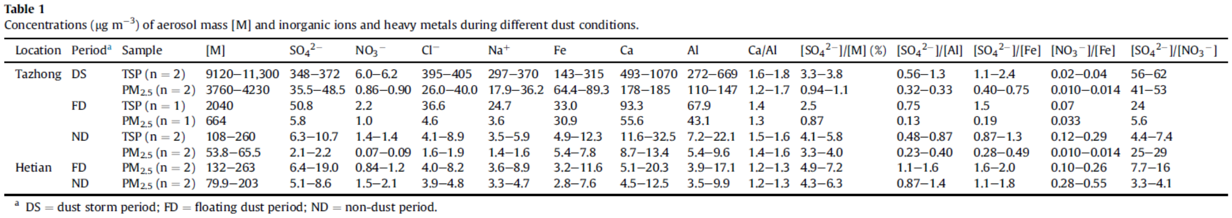


Table 1
Concentrations (μg m^{-3}) of aerosol mass [M] and inorganic ions and heavy metals during different dust conditions.

Location	Period[a]	Sample	[M]	SO_4^{2-}	NO_3^-	Cl^-	Na^+	Fe	Ca	Al	Ca/Al	$[SO_4^{2-}]/[M]$ (%)	$[SO_4^{2-}]/[Al]$	$[SO_4^{2-}]/[Fe]$	$[NO_3^-]/[Fe]$	$[SO_4^{2-}]/[NO_3^-]$
Tazhong	DS	TSP (n = 2)	9120–11,300	348–372	6.0–6.2	395–405	297–370	143–315	493–1070	272–669	1.6–1.8	3.3–3.8	0.56–1.3	1.1–2.4	0.02–0.04	56–62
		$PM_{2.5}$ (n = 2)	3760–4230	35.5–48.5	0.86–0.90	26.0–40.0	17.9–36.2	64.4–89.3	178–185	110–147	1.2–1.7	0.94–1.1	0.32–0.33	0.40–0.75	0.010–0.014	41–53
	FD	TSP (n = 1)	2040	50.8	2.2	36.6	24.7	33.0	93.3	67.9	1.4	2.5	0.75	1.5	0.07	24
		$PM_{2.5}$ (n = 1)	664	5.8	1.0	4.6	3.6	30.9	55.6	43.1	1.3	0.87	0.13	0.19	0.033	5.6
	ND	TSP (n = 2)	108–260	6.3–10.7	1.4–1.4	4.1–8.9	3.5–5.9	4.9–12.3	11.6–32.5	7.2–22.1	1.5–1.6	4.1–5.8	0.48–0.87	0.87–1.3	0.12–0.29	4.4–7.4
		$PM_{2.5}$ (n = 2)	53.8–65.5	2.1–2.2	0.07–0.09	1.6–1.9	1.4–1.6	5.4–7.8	8.7–13.4	5.4–9.6	1.4–1.6	3.3–4.0	0.23–0.40	0.28–0.49	0.010–0.014	25–29
Hetian	FD	$PM_{2.5}$ (n = 2)	132–263	6.4–19.0	0.84–1.2	4.0–8.2	3.6–8.9	3.2–11.6	5.1–20.3	3.9–17.1	1.2–1.3	4.9–7.2	1.1–1.6	1.6–2.0	0.10–0.26	7.7–16
	ND	$PM_{2.5}$ (n = 2)	79.9–203	5.1–8.6	1.5–2.1	3.9–4.8	3.3–4.7	2.8–7.6	4.5–12.5	3.5–9.9	1.2–1.3	4.3–6.3	0.87–1.4	1.1–1.8	0.28–0.55	3.3–4.1

[a] DS = dust storm period; FD = floating dust period; ND = non-dust period.

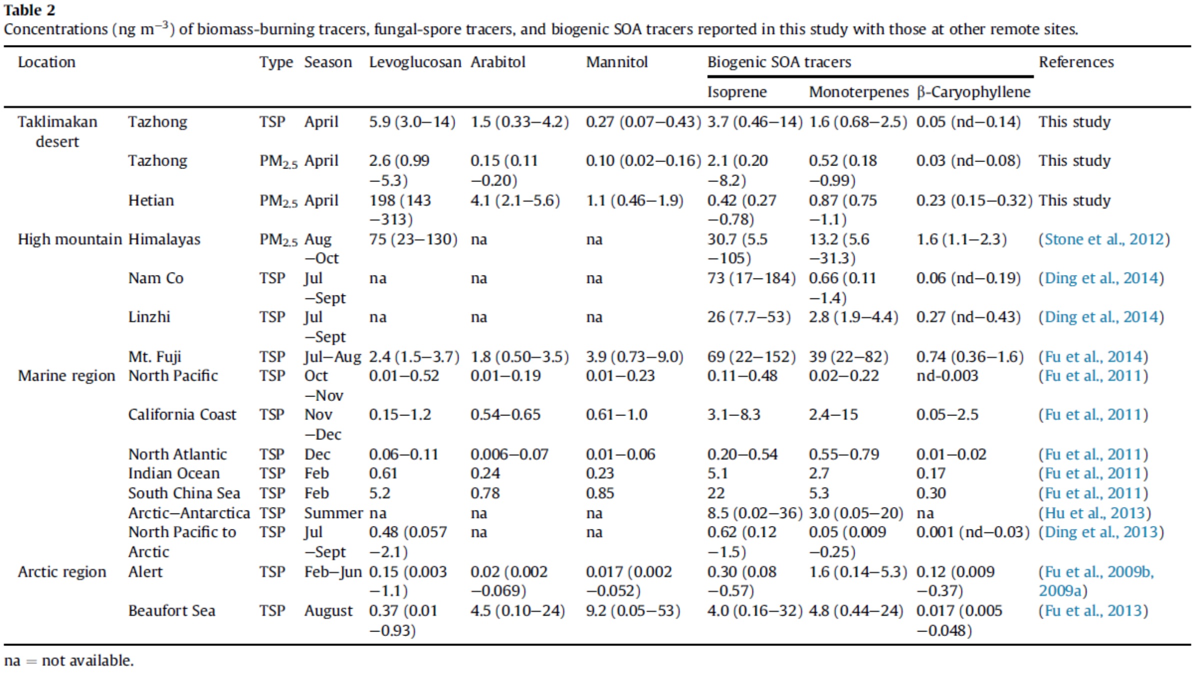
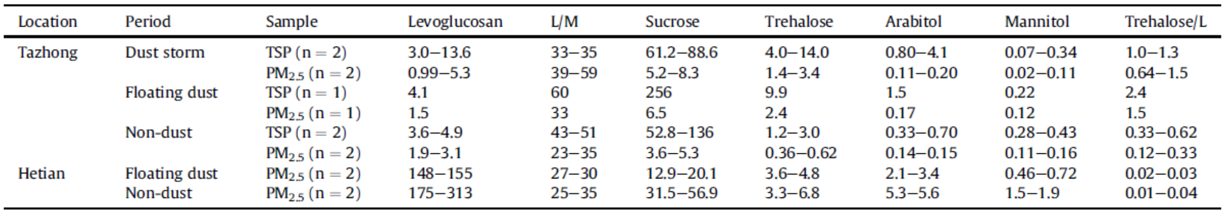


Table 2
Concentrations (ng m^{-3}) of biomass-burning tracers, fungal-spore tracers, and biogenic SOA tracers reported in this study with those at other remote sites.

Location		Type	Season	Levoglucosan	Arabitol	Mannitol	Biogenic SOA tracers			References
							Isoprene	Monoterpenes	β-Caryophyllene	
Taklimakan desert	Tazhong	TSP	April	5.9 (3.0–14)	1.5 (0.33–4.2)	0.27 (0.07–0.43)	3.7 (0.46–14)	1.6 (0.68–2.5)	0.05 (nd–0.14)	This study
	Tazhong	$PM_{2.5}$	April	2.6 (0.99–5.3)	0.15 (0.11–0.20)	0.10 (0.02–0.16)	2.1 (0.20–8.2)	0.52 (0.18–0.99)	0.03 (nd–0.08)	This study
	Hetian	$PM_{2.5}$	April	198 (143–313)	4.1 (2.1–5.6)	1.1 (0.46–1.9)	0.42 (0.27–0.78)	0.87 (0.75–1.1)	0.23 (0.15–0.32)	This study
High mountain	Himalayas	$PM_{2.5}$	Aug–Oct	75 (23–130)	na	na	30.7 (5.5–105)	13.2 (5.6–31.3)	1.6 (1.1–2.3)	(Stone et al., 2012)
	Nam Co	TSP	Jul–Sept	na	na	na	73 (17–184)	0.66 (0.11–1.4)	0.06 (nd–0.19)	(Ding et al., 2014)
	Linzhi	TSP	Jul–Sept	na	na	na	26 (7.7–53)	2.8 (1.9–4.4)	0.27 (nd–0.43)	(Ding et al., 2014)
	Mt. Fuji	TSP	Jul–Aug	2.4 (1.5–3.7)	1.8 (0.50–3.5)	3.9 (0.73–9.0)	69 (22–152)	39 (22–82)	0.74 (0.36–1.6)	(Fu et al., 2014)
Marine region	North Pacific	TSP	Oct–Nov	0.01–0.52	0.01–0.19	0.01–0.23	0.11–0.48	0.02–0.22	nd-0.003	(Fu et al., 2011)
	California Coast	TSP	Nov–Dec	0.15–1.2	0.54–0.65	0.61–1.0	3.1–8.3	2.4–15	0.05–2.5	(Fu et al., 2011)
	North Atlantic	TSP	Dec	0.06–0.11	0.006–0.07	0.01–0.06	0.20–0.54	0.55–0.79	0.01–0.02	(Fu et al., 2011)
	Indian Ocean	TSP	Feb	0.61	0.24	0.23	5.1	2.7	0.17	(Fu et al., 2011)
	South China Sea	TSP	Feb	5.2	0.78	0.85	22	5.3	0.30	(Fu et al., 2011)
	Arctic–Antarctica	TSP	Summer	na	na	na	8.5 (0.02–36)	3.0 (0.05–20)	na	(Hu et al., 2013)
	North Pacific to Arctic	TSP	Jul–Sept	0.48 (0.057–2.1)	na	na	0.62 (0.12–1.5)	0.05 (0.009–0.25)	0.001 (nd–0.03)	(Ding et al., 2013)
Arctic region	Alert	TSP	Feb–Jun	0.15 (0.003–1.1)	0.02 (0.002–0.069)	0.017 (0.002–0.052)	0.30 (0.08–0.57)	1.6 (0.14–5.3)	0.12 (0.009–0.37)	(Fu et al., 2009b, 2009a)
	Beaufort Sea	TSP	August	0.37 (0.01–0.93)	4.5 (0.10–24)	9.2 (0.05–53)	4.0 (0.16–32)	4.8 (0.44–24)	0.017 (0.005–0.048)	(Fu et al., 2013)

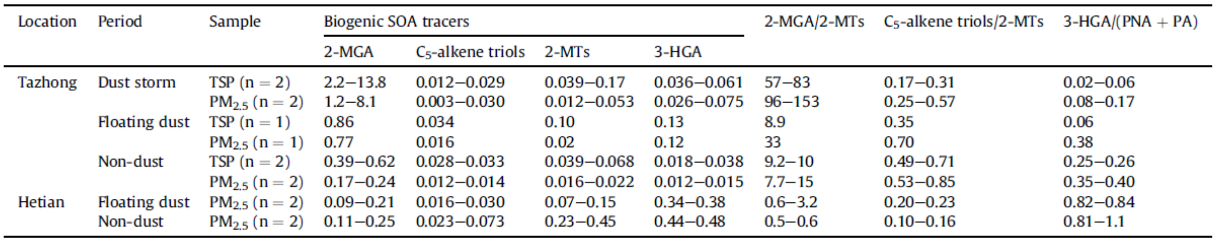
na = not available.

Table 3
Concentrations of primary organic marker compounds (ng m^{-3}) and their concentration ratios during different dust conditions.



Location	Period	Sample	Levoglucosan	L/M	Sucrose	Trehalose	Arabitol	Mannitol	Trehalose/L
Tazhong	Dust storm	TSP (n = 2)	3.0–13.6	33–35	61.2–88.6	4.0–14.0	0.80–4.1	0.07–0.34	1.0–1.3
		$PM_{2.5}$ (n = 2)	0.99–5.3	39–59	5.2–8.3	1.4–3.4	0.11–0.20	0.02–0.11	0.64–1.5
	Floating dust	TSP (n = 1)	4.1	60	256	9.9	1.5	0.22	2.4
		$PM_{2.5}$ (n = 1)	1.5	33	6.5	2.4	0.17	0.12	1.5
	Non-dust	TSP (n = 2)	3.6–4.9	43–51	52.8–136	1.2–3.0	0.33–0.70	0.28–0.43	0.33–0.62
		$PM_{2.5}$ (n = 2)	1.9–3.1	23–35	3.6–5.3	0.36–0.62	0.14–0.15	0.11–0.16	0.12–0.33
Hetian	Floating dust	$PM_{2.5}$ (n = 2)	148–155	27–30	12.9–20.1	3.6–4.8	2.1–3.4	0.46–0.72	0.02–0.03
	Non-dust	$PM_{2.5}$ (n = 2)	175–313	25–35	31.5–56.9	3.3–6.8	5.3–5.6	1.5–1.9	0.01–0.04

Table 4
Concentrations of secondary organic marker compounds (ng m^{-3}) and their concentration ratios during different dust conditions.



Location	Period	Sample	Biogenic SOA tracers				2-MGA/2-MTs	C_5-alkene triols/2-MTs	3-HGA/(PNA + PA)
			2-MGA	C_5-alkene triols	2-MTs	3-HGA			
Tazhong	Dust storm	TSP (n = 2)	2.2–13.8	0.012–0.029	0.039–0.17	0.036–0.061	57–83	0.17–0.31	0.02–0.06
		$PM_{2.5}$ (n = 2)	1.2–8.1	0.003–0.030	0.012–0.053	0.026–0.075	96–153	0.25–0.57	0.08–0.17
	Floating dust	TSP (n = 1)	0.86	0.034	0.10	0.13	8.9	0.35	0.06
		$PM_{2.5}$ (n = 1)	0.77	0.016	0.02	0.12	33	0.70	0.38
	Non-dust	TSP (n = 2)	0.39–0.62	0.028–0.033	0.039–0.068	0.018–0.038	9.2–10	0.49–0.71	0.25–0.26
		$PM_{2.5}$ (n = 2)	0.17–0.24	0.012–0.014	0.016–0.022	0.012–0.015	7.7–15	0.53–0.85	0.35–0.40
Hetian	Floating dust	$PM_{2.5}$ (n = 2)	0.09–0.21	0.016–0.030	0.07–0.15	0.34–0.38	0.6–3.2	0.20–0.23	0.82–0.84
	Non-dust	$PM_{2.5}$ (n = 2)	0.11–0.25	0.023–0.073	0.23–0.45	0.44–0.48	0.5–0.6	0.10–0.16	0.81–1.1

Table S1. Concentrations of biogenic primary and secondary organic compounds and water-soluble inorganic ions in aerosol samples collected at Tazhong (TZ) and Hetian (HT) in the Taklimakan Desert, China.

Marker compounds	Tazhong (TSP)		Tazhong ($PM_{2.5}$)		Hetian ($PM_{2.5}$)	
	mean	range	mean	range	mean	range
aerosol mass (μg m^{-3})	4570	108–11300	1760	53.8–4230	169	79.9–263
Biomass-burning tracers (ng m^{-3})						
levoglucosan	5.9	3.0–14	2.6	0.99–5.3	198	148–313
galactosan	0.26	0.11–0.57	0.05	0.02–0.07	5.3	3.5–9.1
mannosan	0.15	0.07–0.39	0.07	0.02–0.14	6.6	5.2–9.0
4-hydroxybenzoic acid	0.87	0.34–2.2	0.25	0.11–0.57	7.4	6.4–9.9
vanillin	1.2	0.76–2.8	0.67	0.32–1.5	6.0	5.1–7.5
vanillic acid	0.60	0.15–1.7	0.15	0.09–0.37	2.6	1.6–3.9
syringic acid	2.1	0.27–8.8	0.22	0.08–0.31	3.4	1.9–5.1
dehydroabietic acid	0.94	0.28–2.4	0.36	0.18–0.56	3.4	1.8–4.8
subtotal	12	5.9–32	4.3	1.9–8.8	233	177–359
Sugars and sugar alcohols (ng m^{-3})						
arabinose	16	4.5–40	11	6.0–13	5.2	4.3–6.8
arabitol	1.5	0.33–4.2	0.15	0.11–0.20	4.1	2.1–5.6
fructose	16	4.6–22	7.7	6.1–11	17	7.7–27
glucose	35	14–70	13	10–21	21	9.5–34
mannitol	0.27	0.07–0.43	0.10	0.02–0.16	1.1	0.46–1.9
inositol	0.49	0.07–0.96	0.11	0.03–0.16	0.96	0.37–1.4
sucrose	119	53–256	5.8	3.6–8.3	30	13–57
maltose	1.7	0.59–3.9	1.4	0.53–3.3	3.1	2.2–3.9
trehalose	6.4	1.2–14	1.6	0.36–3.4	4.6	3.3–6.8
subtotal	196	85–334	40	33–58	87	45–142
Isoprene SOA tracers (ng m^{-3})						
2-methylglyceric acid (2-MGA)	3.6	0.39–14	2.1	0.17–8.1	0.17	0.089–0.24
ΣC_5-alkene triols [a]	0.027	0.012–0.034	0.015	0.003–0.030	0.035	0.016–0.071
2-methylthreitol	0.025	0.013–0.054	0.009	0.005–0.017	0.078	0.025–0.14
2-methylerythritol	0.057	0.024–0.11	0.017	0.007–0.036	0.15	0.042–0.31
sum of 2-methyltetrols	0.082	0.039–0.17	0.025	0.012–0.053	0.22	0.067–0.45
subtotal	3.7	0.46–14	2.1	0.20–8.2	0.42	0.27–0.78

α/β-pinene SOA tracers (ng m^{-3})						
3-hydroxyglutaric acid (3-HGA)	0.16	0.036–0.38	0.10	0.026–0.15	0.41	0.34–0.48
pinonic acid	0.95	0.46–1.8	0.17	0.049–0.44	0.24	0.16–0.35
pinic acid	0.53	0.15–1.4	0.25	0.10–0.47	0.23	0.20–0.25
MBTCA [b]	0.001	nd [c]–0.002	nd	nd	nd	nd
subtotal	1.6	0.68–2.5	0.52	0.18–0.99	0.87	0.75–1.1
β-caryophyllene SOA tracer (ng m^{-3})						
β-caryophyllinic acid	0.05	nd–0.14	0.03	nd–0.08	0.23	0.15–0.32
subtotal of SOA tracers	5.4	1.4–17	2.7	0.71–9.2	1.5	1.2–2.1
Water-soluble inorganic ions (μg m^{-3})						
Cl^-	170	4.1–405	15	1.6–40	5.2	3.8–8.2
NO_3^-	3.4	1.4–6.2	0.59	0.07–1.0	1.4	0.84–2.1
SO_4^{2-}	158	6.3–372	19	2.1–48	9.8	5.0–19
Na^+	140	3.5–370	12	1.4–36	5.1	3.3–8.9
NH_4^+	0.13	nd–0.33	0.07	nd–0.20	0.60	0.54–0.65
K^+	6.2	0.24–16	0.82	0.13–2.4	0.73	0.65–0.86
Mg^{2+}	8.5	0.41–22	1.4	0.27–4.0	0.49	0.35–0.75
Ca^{2+}	58	8.0–158	17	5.9–43	8.3	6.9–10
subtotal	544	24–1310	65	12–175	32	23–50

[a] C_5-alkene triols: the sum of cis-2-methyl-1, 3, 4-trihydroxy-1-butene, trans-2-methyl-1, 3, 4-trihydroxy-1-butene, and 3-methyl-2, 3, 4-trihydroxy-1-butene. [b] MBTCA: 3-methyl-1, 2, 3-butanetricarboxylic acid. [c] nd = not detected.

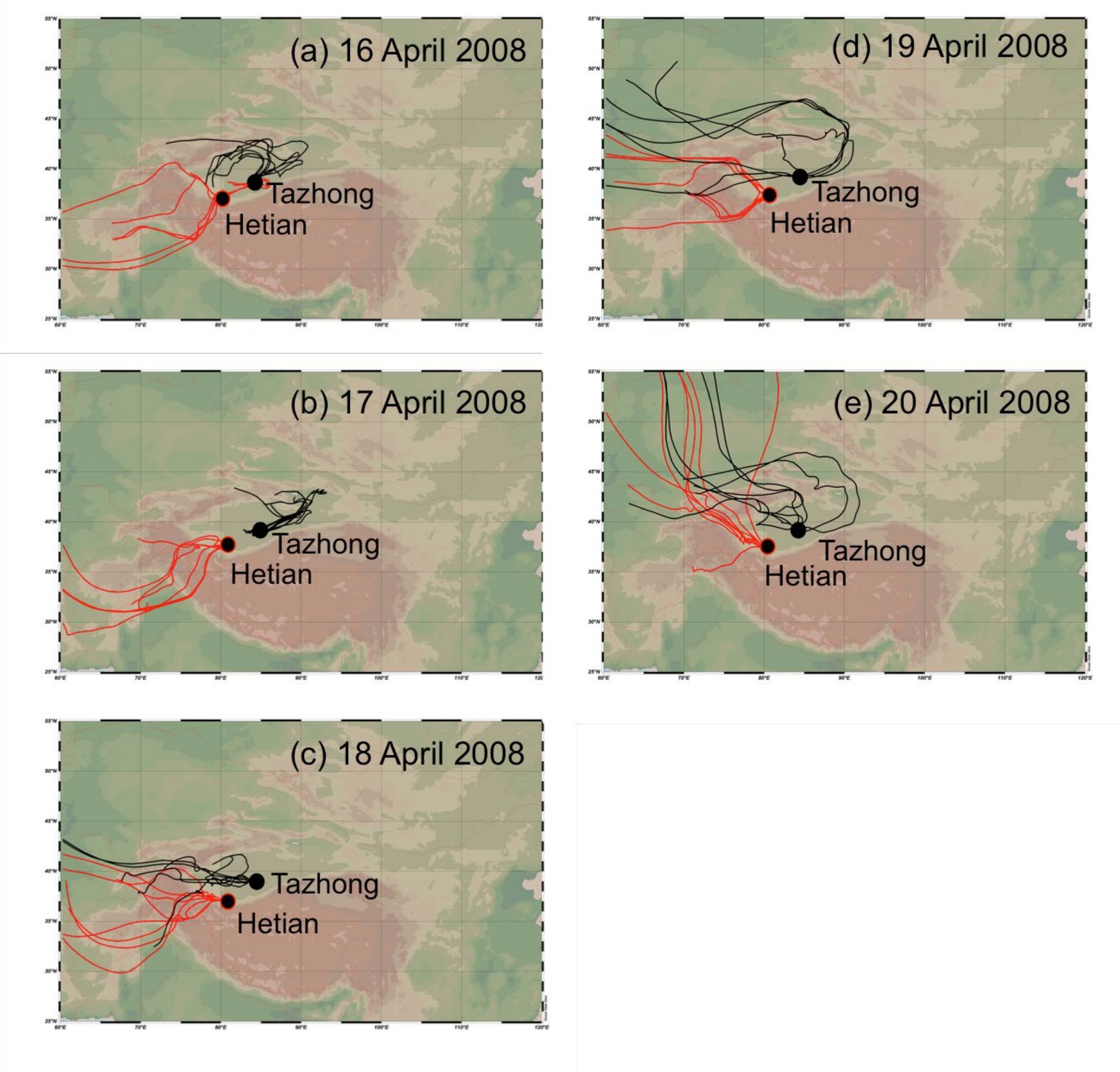


Fig. S1. Three-day back trajectories of air masses arrived at the Tazhong site in the central Taklimakan desert and the Hetian site at the south rim of the Taklimakan desert.