Soil-derived sulfate in atmospheric dust particles at Taklimakan desert

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[1] Dust-associated sulfate is believed to be a key species which can alter the physical and chemical properties of dust particles in the atmosphere. Its occurrence in the particles has usually been considered to be the consequence of particles' aging in the air although it is present in some crustal minerals. Our observation at the north and south edge of Taklimakan desert, one of the largest dust sources in the Northern Hemisphere, during a dust episode in April 2008 revealed that sulfate in atmospheric dust samples most likely originated directly from surface soil. Its TSP, PM_{10} and $PM_{2.5}$ content was proportional to samples' mass and comprised steadily about 4% in the differently sized samples, the ratio of elemental sulfur to iron was approximately constant 0.3, and no demonstrable influence of pollutants from fossil fuel combustion and biomass burning was detected. These results suggest that sulfate could be substantially derived from surface soil at the desert area and the lack of awareness of this origin may impede accurate results in any investigation of atmospheric sulfur chemistry associated with Taklimakan dust and its subsequent local, regional and global effects on the atmosphere. **Citation:** Wu, F., D. Zhang, J. Cao, H. Xu, and Z. An (2012), Soil-derived sulfate in atmospheric dust particles at Taklimakan desert, *Geophys. Res. Lett.*, *39*, L24803, doi:10.1029/2012GL054406.

1. Introduction

[2] Mineral dust particles have an important influence on the atmospheric radiation budget and climate change [*Sokolik and Toon*, 1996; *Tegen et al.*, 1996]. They also modify the chemical composition of atmospheric aerosols [*Dentener et al.*, 1996], take part in hydrological processes in the air [*Miller et al.*, 2004], link geographically distant ecosystems, and supply nutrients to remote terrestrial and marine biospheres [*Jickells et al.*, 2005].

[3] Studies of Asian dust have shown that the composition of dust particles at source regions are consistent with that of top soil and mainly composed of Mg, Al, Si, K, Ca, Fe, Ti, and Mn [*Zhang et al.*, 1993]. Suspended in the air, dust particles can be changed by surface chemical reactions and the coagulation and coalescence with other particulate matter. In the downstream marine atmosphere, they were frequently coated with sulfate and nitrate and mixed with sea salt [*Andreae et al.*, 1986; *Yamato and Tanaka*, 1994]. It has been reported that sulfate coating on dust particles can lower their effective deliquescence RH [*Semeniuk et al.*, 2007], and may modify the particles in phase and size [*Andreae et al.*, 1986]. These changes in turn feed back into many other chemical and physical processes that relate to radiative energy distribution within the atmosphere [*Bauer and Koch*, 2005]. Moreover, the presence of sulfate on mineral dust particles is able to increase the bioavailability of iron [*Solmon et al.*, 2009], which was supposed to be a key process for atmospheric dust to fertilize marine phytoplankton [*Jickells et al.*, 2005].

[4] Sulfate associated with mineral particles in the air is generally due to the formation of sulfuric acid and salts on the particle surface [*Dentener et al.*, 1996; *Li and Shao*, 2009]. It was the consequence of reactive gases (e.g., SO_2) adsorbed and oxidized on dust particles or the direct uptake of acidic gases, that was then fully or partially neutralized by alkaline components [*Ullerstam et al.*, 2002]. This has been considered extensively in the assessment of sulfate formation in associated with dust, particularly for Asian dust [e.g., *Manktelow et al.*, 2010; *Wang et al.*, 2005].

[5] Surface soil in some Chinese desert areas contains substantial sulfate [*Abuduwaili et al.*, 2008; *Zhu and Yang*, 2010]. The mass percentage of sulfate in surface soil (0–5 cm) where salt lakes developed is usually larger than 1%, and up to 28.8% in cases, such as Ebinur Lake [*Abuduwaili et al.*, 2008]. In this regard, it has been suggested that surface soil could bring sulfate into the air as dust storms occur [*Huang et al.*, 2010; *Yabuki et al.*, 2005]. S-containing dust particles were detected over the source and downwind regions of Asian dust in the continental atmosphere [*Matsuki et al.*, 2005; *Trochkine et al.*, 2003]. S in particles with S/Ca ratio close to 0.8 was regarded as gypsum and distinguished from potentially secondary sulfate in the study of *Trochkine et al.* [2003]. Unfortunately, soil-derived sulfate in dust samples at the desert areas has not been well investigated, which impeded the evaluation of the relative contribution of sulfate formed in the atmosphere.

[6] We carried out observations at two sites in the Taklimakan desert in April, 2008 during a dust episode and found substantial soil-derived sulfate in the airborne particulate matters. The Taklimakan desert in northwestern China is one of the largest deserts in the world, and is the major

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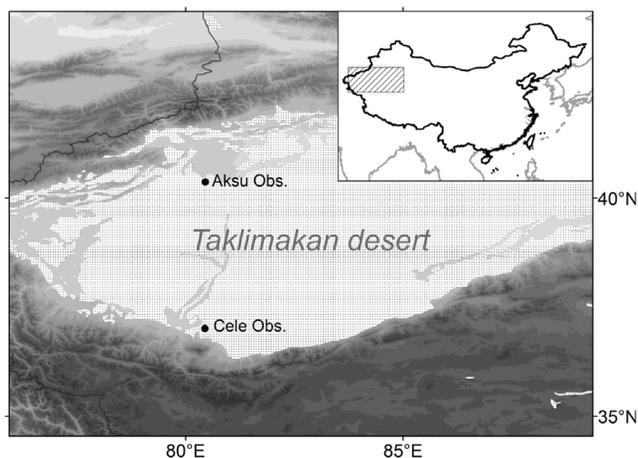
Figure 1. Location of the sampling sites.

source of airborne mineral dust in the Northern Hemisphere [*Zhang et al.*, 1997]. Because of the surrounding mountains, dust from there is usually lifted over 5000 m and transported over a long distance to the Pacific Ocean and beyond, even around the globe [*Sun*, 2002; *Uno et al.*, 2009]. The desert continuously releases mineral aerosol to elevated layers and is considered to be an important source of atmospheric background dust in free troposphere over East Asia [*Matsuki et al.*, 2003]. Therefore, understanding sulfate in dust particles at the desert areas is expected to be particularly important for discussing the sulfur chemistry associated with Taklimakan dust on a local, regional and global scale in the atmosphere. Here, we report the results about the sulfate we observed, show the evidence of its soil origin, and discuss the potential importance of the results.

2. Method

[7] The observation was carried out in the spring of 2008. Samples were collected at the Aksu observatory (40°37′N, 80°45′E; 1028 m a.s.l.) from 12 to 20 April and at the Cele observatory (37°01′N, 80°44′E; 1380 m a.s.l.) from 22 to 26 April 2008. The two observatories were set up for desert research by the Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences. Aksu observatory is located at the north edge of Taklimakan desert and Cele at the south edge (Figure 1). Dust storms from the desert frequently hit the observatories without passing non-desert areas.

[8] Meteorological conditions including wind direction, wind speed, temperature, pressure, relative humidity, precipitation and visibility were automatically recorded within the observatories. A dust storm occurred at 13:00 on 17 April (Local time) and lasted to 26 April. Both observatories recorded the whole process almost simultaneously. At Aksu, the wind speed was weak (less 2.5 m s^{-1}) and the visibility was fine (about 40 km) before 17 April. At midday of April 17, the wind speed increased rapidly at the dust storm arrival. In the following two days (18–20 April), the air at the two sites was quite turbid and the visibility was 500–5000 m. After 14:00 on 20 April, the wind became weak and the visibility increased, but lots of fine dust particles were still

suspended in the air. According to the guidelines on dust observation of the Meteorological Observing Criteria issued by the National Weather Bureau of China, the whole sampling period could be further divided into non-dust period (ND period: before 18 April), dust storm period (DS period: 18–20 April) and floating dust period (FD period: 21–26 April). It has to be noticed that the suspended particulate matters at the two observatories in all periods, including the ND period, were always dominated by mineral dust except for the large variation of the mass concentration under different dust conditions.

[9] Three mini-volume samplers (Airmetrics, Oregon, USA) were set up on the roof of buildings (about 4 m above ground) within the observatories to simultaneously collect samples of total suspension particulate matters (TSP), particle matters with aerodynamic diameters less than 10 μm (PM$_{10}$) and particulate matters with aerodynamic diameters less than 2.5 μm (PM$_{2.5}$). Aerosol particles were collected onto 47-mm quartz microfiber filters (Whatman QM/A, Maidstone, UK) at a flow rate of 5 L min^{-1}. The filters were heated at 900°C for 3 hours in order to remove possible contaminants at the filter preparation. After sample collection, the filters were placed in clean polystyrene petri dishes and stored in a refrigerator at about 4°C. Particle collection time for each sample was about 24 hours during ND periods, and 4–8 hours during DS and FD periods. Eleven sets of TSP, PM$_{10}$ and PM$_{2.5}$ samples were obtained: three ND sets and three DS sets at Aksu and five FD sets at Cele.

[10] Filter samples were equilibrated in a stable environment with relative humidity 35–45% and temperature 20–23°C for 24 hours to minimize particle volatilization and aerosol liquid water interference. Then they were weighted using a MC5 electronic microbalance (Sartorius, GOTTINGEN, Germany) with the sensitivity of ±0.001 mg.

[11] Water-soluble components in each sample were extracted from half of the sample filter by ultrasonic agitation in 10 mL distilled water. The extraction solution was filtered with 0.45 μm pore size microporous membranes, and then the filtrate solution was stored at 4°C in plastic tubes until subsequent analyses. An ion chromatography (Dionex DX-600) was used to determine the major anions and cations, including sulfate (SO$_4^{2-}$), nitrate (NO$_3^-$), chloride (Cl$^-$), sodium (Na$^+$), potassium (K$^+$), ammonium (NH$_4^+$), calcium (Ca^{2+}), and magnesium (Mg^{2+}). Calibration curves were constructed from the peak areas of the chromatographs produced from a series of mixed standards. Filter blanks were also prepared in the same manner and analyzed to obtain the controls of the water-soluble ionic species. Ammonium in all samples was lower than the detection limit.

[12] An energy dispersive X-Ray fluorescence (ED-XRF) spectrometry (Epsilon 5 ED-XRF, PANalytical B. V., the Netherlands) was used to quantify elements in the samples of the remained parts of sample filters. Sulfur (S), potassium (K), calcium (Ca), iron (Fe), titanium (Ti) and lead (Pb), which had low minimum detection limit and low backgrounds for quartz-fiber filters, were quantitatively determined. Analytical uncertainties, as checked by parallel analysis of the NIST standard reference material (SRM-2683), were less than 10% for most of the detected elements. However, S content was underestimated by about 20%. A fraction of the deposited material of the TSP and PM$_{10}$ samples collected on April 18 during strong dust storm

Table 1. Concentration of Sulfate ($[SO_4^{2-}]$) and Mass ($[M]$) for $PM_{2.5}$, PM_{10} and TSP Samples Under Different Dust Conditions[a]

Cases[b]	Samples	$[SO_4^{2-}]$ ($\mu g\, m^{-3}$)	$[M]$ ($\mu g\, m^{-3}$)	$[SO_4^{2-}]/[M]$	$[NO_3^-]/[M]$	$[Ca^{2+}]/[M]$	$[S]/[Fe]$
DS	$PM_{2.5}$ (n = 3)	45.0 ± 14.1	1135.2 ± 290.6	4.2 ± 2.0	0.3 ± 0.04	5.0 ± 1.5	0.30 ± 0.18
	PM_{10} (n = 3)	136.6 ± 65.6	2670.9 ± 182.9	5.2 ± 2.5	0.2 ± 0.02	3.6 ± 1.5	0.26 ± 0.11
	TSP (n = 3)	180.7 ± 62.0	3923.1 ± 306.0	4.7 ± 1.8	0.2 ± 0.04	2.9 ± 1.1	0.23 ± 0.09
FD	$PM_{2.5}$ (n = 5)	11.4 ± 2.9	305.1 ± 66.8	3.7 ± 0.4	0.8 ± 0.06	3.9 ± 0.7	0.29 ± 0.01
	PM_{10} (n = 5)	22.2 ± 6.3	548.7 ± 132.9	4.0 ± 0.2	0.6 ± 0.1	3.5 ± 0.3	0.28 ± 0.02
	TSP (n = 5)	27.3 ± 7.5	703.4 ± 177.4	3.9 ± 0.3	0.5 ± 0.1	3.3 ± 0.4	0.25 ± 0.02
ND	$PM_{2.5}$ (n = 3)	4.4 ± 0.2	125.1 ± 62.5	4.0 ± 1.5	2.5 ± 1.4	3.1 ± 1.1	0.36 ± 0.02
	PM_{10} (n = 3)	8.4 ± 0.5	194.6 ± 18.7	4.3 ± 0.5	2.1 ± 0.5	3.6 ± 0.6	0.38 ± 0.03
	TSP (n = 3)	11.5 ± 1.0	311.5 ± 73	3.8 ± 0.7	1.5 ± 0.3	3.0 ± 0.7	0.36 ± 0.04

[a]Also included are the mass ratios (%) of SO_4^{2-}, nitrate ($[NO_3^-]$) and water-soluble calcium ($[Ca^{2+}]$) to the mass concentration and the ratios of elemental sulfur ($[S]$) to elemental iron ($[Fe]$), with standard deviations.
[b]DS = dust storm period; FD = Floating dust period; ND = non-dust period.

period fell off the filters in the XRF analysis because of the extremely high particle loading.

3. Results and Discussion

3.1. Sulfate

[13] Table 1 is the summary of concentrations of sulfate, $PM_{2.5}$, PM_{10} and TSP and the mass ratios of sulfate, NO_3^- and Ca^{2+} to the corresponding masses under different dust conditions. Sulfate concentration in TSP, and also in PM_{10} and $PM_{2.5}$, apparently differed in different dust periods. In contrast to the concentration $11.5 \pm 1.0 \ \mu g\, m^{-3}$ of sulfate in TSP during the ND period (before the occurrence of the dust storm at Aksu), it was $180.7 \pm 62.0 \ \mu g\, m^{-3}$ during DS period (when the dust storm occurred at Aksu) and was $27.3 \pm 7.5 \ \mu g\, m^{-3}$ during the FD period (after the dust storm at Cele). Sulfate in PM_{10} and $PM_{2.5}$ showed the same trend in the different periods: highest in the DS period, lowest in the ND period and in-between in the FD period.

[14] However, the abundance of sulfate relative to the mass showed consistency in different samples of TSP, PM_{10} and $PM_{2.5}$. The ratio of sulfate concentration to the mass concentration of DS samples was in the range of 4.2–5.2 with deviations of 1.8–2.5, that of FD samples in the range of 3.7–4.0 with deviations of 0.2–0.4, and that of ND samples in the range of 3.8–4.3 with deviations of 0.5–1.5. SO_4^{2-} was thus one of the most abundant ions and it accounted steadily for 3–5% of particulate mass. These results indicated that the ratio was not size dependent and sulfate content was closely correlated with the sample mass.

[15] Ca^{2+} is a typical anion dominated by soil-derived particles in remote continental atmosphere. No considerable non-natural emission of Ca^{2+} such as road dust or construction dust was expected at the two observatories when the samples were collected. It was found that, similar to sulfate, Ca^{2+} content in the samples was proportional to the aerosol mass. The mass percentage of Ca^{2+} in all samples was similar and approximately 3–5% (Table 1). This result was consistent with the expectation that Ca^{2+} was from surface soil. Moreover, the total mole concentration of Na^+, K^+ and Mg^{2+} was much larger than that of sulfate in any samples, suggesting that sulfate in the samples could completely be in a mineral form.

[16] NO_3^- is a component that has no crustal origin and is normally produced via conversion reactions including gaseous nitrogen oxides in the air. It prefers to occur in primary particles and appears to be reversible to gaseous nitrogen

oxides under normal atmospheric conditions. In the present samples, NO_3^- accounted for 0.2–2.5% of particulate matters in mass, and was more enriched in samples of smaller size ranges. The relative contribution of NO_3^- was the largest in the $PM_{2.5}$, the smallest in the TSP and in-between in the PM_{10}. NO_3^- was apparently reduced in the DS and FD periods in comparison to that in the ND period. In the TSP, the mass percentage of NO_3^- was $1.5 \pm 0.3\%$ in the ND period, but decreased to $0.5 \pm 0.1\%$ in the FD period and $0.2 \pm 0.04\%$ in the DS period, i.e., the concentration of NO_3^- was not proportional to the concentration of aerosol mass. This trend was completely different from that of sulfate and Ca^{2+} in the samples.

3.2. Elemental Sulfur

[17] The ratio of elemental sulfur to elemental iron from the XRF analysis was also independent from size (Table 1). It was about 0.3 in all samples despite the systematic underestimate of sulfur content in the analysis. Since Fe is one of leading elements in Earth crust and no other sources of iron except surface soil were expected at the two sites under the influence of desert dust, the correlation also supports that sulfur in the samples was correlated with soil.

[18] Sulfate produced via SO_2 conversion on particle surface is due to the presence of SO_2 in the ambient air. Anthropogenic SO_2 in the atmosphere is usually from fossil fuel combustion and vegetation burning [*Chuang et al.*, 1997]. To investigate if there was a substantial contribution of fossil fuel combustion and vegetation burning in the samples, K (a typical element in aerosols from biomass burning) and Pb (a typical element in aerosols from fossil fuel combustion) in the aerosol samples of the ND period were investigated with their enrichment factors relative to surface soil using iron as the reference element. Because there is not a uniform reference sample for soil of the Taklimakan desert, a surface soil sample which we collected from sand dunes near Cele site was applied as reference surface soil. The soil was suspended in a chamber (the Desert Research Institute re-suspension chamber) and samples in different size ranges were collected from the chamber through TSP, PM_{10} and $PM_{2.5}$ inlets onto filters [*Wu et al.*, 2011]. The elements in the TSP, PM_{10} and $PM_{2.5}$ surface soil samples were analyzed using the ED-XRF in this study.

[19] The enrichment factors of K and Pb and also those of S are summarized in Table 2. The factors of K and Pb in any size range were 1–2. Both K and Pb had a good correlation with Fe during the whole sampling period. The correlation

Table 2. Enrichment Factors of Pb, K and S for the ND Period

	K		Pb		S	
Samples	Range	Average	Range	Average	Range	Average
PM$_{2.5}$ (n = 3)	1.22–1.29	1.26 ± 0.03	0.26–1.84	1.14 ± 0.81	15.2–16.7	16.14 ± 0.79
PM$_{10}$ (n = 3)	1.34–1.59	1.44 ± 0.14	0.95–3.02	2.23 ± 1.12	14.5–16.9	15.95 ± 1.21
TSP (n = 3)	1.27–1.34	1.32 ± 0.04	0.44–1.39	1.06 ± 0.54	15.4–18.4	16.52 ± 1.68

coefficient was 0.99 for K and 0.81 for Pb (Figure S1 in the auxiliary material), indicating that K and Pb in the samples were derived most likely from the surface soil as Fe, and no demonstrable influence of pollutants from biomass burning and fossil fuel combustion was expected.[1]

[20] S in aerosol samples was somewhat enriched with the enrichment factors of 15–17. This could be due to lower concentration of S in the reference sample. The S content in the topsoil of the dust source region was affected by the parent material composition, climatic conditions as well as landform types. Previous studies revealed that the S/Fe ratio of the soil in the Taklimakan desert was generally less than 0.04, but in the regions where salt lake developed, such as Turpan Basin, the ratio was very large −up to 0.37 [*Suzuki et al.*, 1993]. The S/Fe of the soil samples (sand dunes near Cele observatory) was 0.022, which was close to the lowest value of surface soil at the desert areas, but much smaller than that of dry salt lake soil. We consider that our samples represented a mixture of sand dune dust and dry salt lake sediments. This hypothesis can explain the large enrichment factors of S and is not against the fact that the factors were size-independent and showed little change along with dust concentration in the air.

[21] In the light of the above results, we conclude that the sulfate in the dust samples was unlikely produced via atmospheric processes but originated with dust itself, i.e. the sulfate was more likely brought into the air by the dust. The Taklimakan area was a paleo-ocean 5.3 million years ago [*Sun and Liu*, 2006] and there are many developed and weathered salt lake areas which formed at the marine transgression in the late Cretaceous and early Tertiary periods. The Taklimakan is a closed basin without outlets for runoff and accumulating salts. Because of the dry climate and strong evaporation, a heavy accumulation of salts, most containing chlorides, sulfates or carbonates, has formed in the basin [*Yabuki et al.*, 2005]. Surface soil of the desert is abundant in sulfate minerals in some areas, such as gypsum, anhydrite and thenardite [*Yabuki et al.*, 1998].

3.3. Inter-comparison From Taklimakan Desert to the Downstream Areas

[22] Sulfate in aerosol particles at the Taklimakan desert was documented in some published literature. For example, *Huang et al.* [2010] investigated the sulfate in dust samples at Tazhong (TZ) in the Taklimakan desert. They found that the average mass fractions of sulfate in PM$_{2.5}$ and TSP were 3.53% and 2.53%, respectively, and the fractions were similar under DS and NDS conditions. They also confirmed correlation of sulfate with Al, Na$^+$, Cl$^-$ and Ca^{2+}. These results are consistent with the present study and indicate the crustal origin of substantial sulfate at the desert.

[23] There are few published papers in which sulfate associated with long-range transported Taklimakan dust in the downstream areas can be confirmed. The lack of observation is likely due to the elevated transport route of the dust, which was usually encountered only at high-altitude sampling sites or aircraft missions. The top of Mt. Fuji (3776 m a.s.l.) is an isolated place and is under the influence of free troposphere flow in the downstream of Asian continent [*Tsutsumi et al.*, 1994], where dust from the Taklimakan desert frequently occurred [*Matsuki et al.*, 2003]. *Suzuki et al.* [2008] reported that the average concentrations of sulfate and Ca^{2+} at the top of Mt. Fuji when Asian dust appeared in March-September 2002 were 64 μg m^{-3} and 103 μg m^{-3}, respectively, and they attributed the Ca^{2+} to desert dust. The mass percentage of sulfate in dust aerosol was estimated to be ∼2.45% if Ca^{2+} was assumed to be the indicator of natural dust and accounted for ∼4% of dust mass. It is very similar to the results of this study, suggesting that the sulfate was possibly governed by soil-derived one.

[24] One trans-Pacific dust events observed at Whistler, British Columbia during INTEX-B 2006 was traced back to the Taklimakan desert by backward trajectory analysis and model simulations [*McKendry et al.*, 2008; *Leaitch et al.*, 2009]. The sulfate in coarse dust particles (diameter > 3 μm) collected using a non-rotating MOUDI sampler was closely correlated with Ca^{2+}, and was estimated to account for ∼3.3% of mineral dust fraction in aerosol mass if both of them originated from desert dust. This result is also very similar to the results of this study, suggesting that the sulfate of coarse particles in the MOUDI results could be accounted for by soil-derived sulfate. If this is true, sulfate formation on coarse particles must be very slow and not as efficient as expected. Notice that this is completely different from the sulfate in fine particles (diameter < 1 μm) which had a higher SO$_4^{2-}$/Ca^{2+} mass ratio (16.2) and a much higher mass percentage of sulfate (>30%) than the coarse particles in the same case. It has been proven that there were usually a large number of sulfur-containing calcium-rich particles in the fine size range in long-range transported Asian dust plumes [*Matsuki et al.*, 2005; *Zhang and Iwasaka*, 2006].

[25] There is no information on soil-derived sulfate in long-range transported dust at the downstream areas of Asian continent to the extent of our knowledge. In contrast to that secondary sulfate in dust particles should be present in principle in the surface layer of dust particles, soil-derived sulfate in dust particles could be inside of the particles. This hypothesis is consistent with the result that sulfate was rarely detected on the surface of individual natural dust particles in the continental atmosphere [*Zhang and Iwasaka*, 1999; *Zhang et al.*, 2005] whereas usually and substantially detected in integrated samples [*Wang et al.*, 2005]. *Levin et al.* [1996] suggested investigating the correlation of S content with particle surface area and volume to identify if S

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[1]Auxiliary materials are available in the HTML. doi:10.1029/2012GL054406.

appeared on the surface of or in dust particles. It was likely a probable approach for an extensive investigation of soil-derived sulfate in dust particles.

[26] Processing of dust particles in association with sulfate formation in the atmosphere is critical to their climatic and environmental effects. The results of this study warrant that sulfate in mineral dust particles at the downstream areas of the Asian continent could be from surface soil if the particles are from the Taklimakan desert. Thus it is not safe to attribute sulfate in atmospheric dust samples totally to secondary species in the elevated levels over the Northern Pacific and its marginal areas. The production of sulfate via particle surface reactions might be largely overestimated without evaluating its soil-derived part although how to investigate this issue is still a difficult task at the current stage. Since dust from the Taklimakan desert contributes largely to mineral particles in the free troposphere and frequently has an environmental effect from local to global scale, the lack of awareness of this origin may impede accurate results in any investigation of the sulfur chemistry associated with Taklimakan dust and the subsequent local, regional and global effects on the atmosphere.

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