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Mineralogical characteristics of soil dust from source regions in northern China

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Mineral compositions of aerosol particles were investigated at four sites (Aksu, Dunhuang, Zhenbeitai, and Tongliao) in desert regions of northern China from March to May in 2001 and 2002 during the intensive field campaign period of ACE-Asia (Aerosol Characterization Experiments-Asia). The X-ray diffraction (XRD) results show the main minerals for Asian dust are illite, chlorite, kaolinite, quartz, feldspar, calcite, and dolomite. Gypsum, hornblende, and halite are also detected in several samples. Semi-quantitative mineralogical data of aerosol samples show that carbonate content decreases from western to eastern source areas; that is, soil dust collected at western source area sites of Dunhuang and Aksu are enriched with carbonate, while northeastern source area site of Tongliao is associated with low carbonate content. But the spatial distribution of feldspar exhibits a different pattern as compared to carbonate, increasing from the western to the eastern sources. The total clay content is significantly higher (73% in average) at the deposition site of Changwu than those at source areas. Air-mass back trajectory studies for the three dust storm events observed at Changwu, showed that soil dust transport pathways were as expected from carbonate content for the source identification, further demonstrating that carbonate was a useful tracer for eolian dust on regional scale in northern China.

1. Introduction

Soil dust is recognized as an important factor on climate forcing (Arimoto, 2001; Arimoto et al., 2006; Duce, 1995; Huebert et al., 2003; Li, Marlng, Savole, Voss, & Prospero, 1996; Tegen, Lacis, & Fung, 1996). According to rain-dust records in Chinese historical documents and present day observations (Arimoto, 2001; Merrill, Arnold, Margaret, & Weaver, 1994; Merrill, Uematsu, & Bleck, 1989; Zhang, 1984; Zhang, Arimoto, & An, 1997), desert regions in East Asia are considered to be the major sources for Asian dust. The annual input of mineral aerosols to the atmosphere from arid and semiarid regions of northern China is ~800 Tg (Zhang et al., 1997), contributing to about half of the global dust output (Andreae, 1995; Duce, 1995). Unfortunately, more adequate assessment of regional forcing by dust is hardly possible because of inadequate knowledge of dust optical properties and their temporal and spatial variation.

Mineral aerosol is a complex mixture of various minerals whose optical properties (extinction coefficient, single scattering albedo and asymmetry parameter) vary from mineral to mineral (Sokolik & Toon, 1999; Sokolik, Toon, & Bergstrom, 1998). For example, hematite has the largest absorption for UV and visible wavelengths (Shen et al., 2006), quartz and clay mineral have the strong absorption bands for IR, though with different major absorption band centers. The optical properties of minerals determined by the abundance of individual minerals and their aggregation are more likely correct than that calculated of the whole dust of unknown composition (Sokolik & Toon, 1999; Sokolik et al., 1998).

On the basis of chemical analysis, previous studies have shown that dusts derived from various Chinese desert regions have different elemental contents, and that elemental ratios between Al, Fe, Mg and Si are useful to distinguish the Asian dusts from different source regions (Zhang et al., 2003; Zhang, Shen, Zhang, Chen, & Liu, 1996). However, mineralogy may be more diagnostically significant for identifying aerosol source areas than elemental ratios. For instance, calcite and palygorskite have been used as indicators of dust emitted from the northern part of the Sahara Desert (Molinaroli, Guerzoni, & Rampazzo, 1993; Schütz & Sebert,

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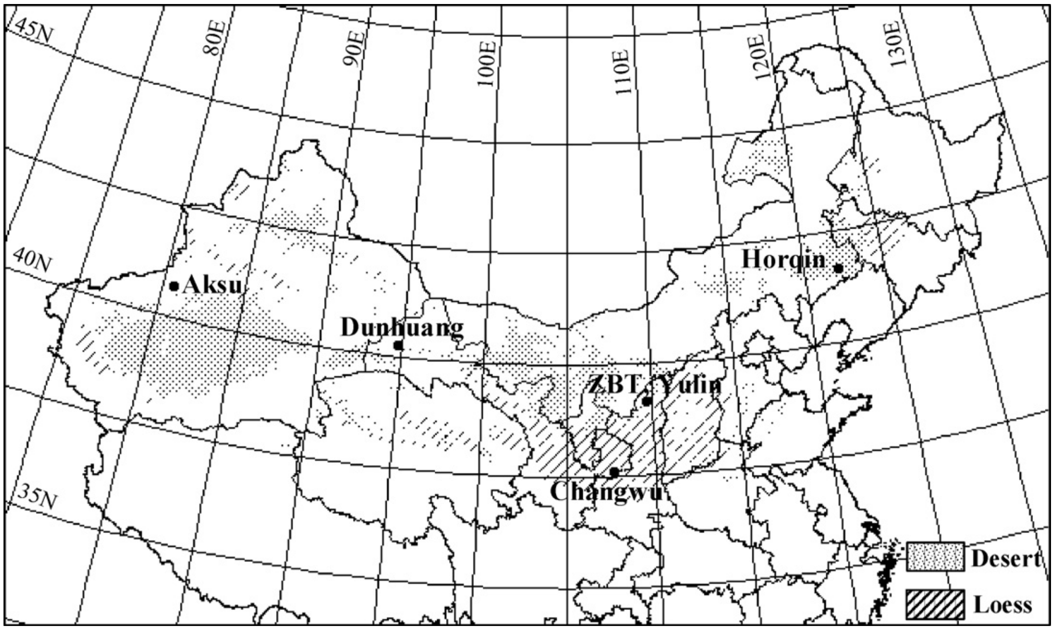

Fig. 1. Location of sampling sites.

1987). Clay mineral ratios, such as I/K ratio (I: illite, K: kaolinite) can also be used as relevant tracers of the origin of soil-derived aerosols within Saharan source regions (Caquineau, Gaudichet, Gomes, Magonthier, & Chatenet, 1998). Mineralogy of aerosols has also been used to assess the sources of eolian sediments in the North Pacific Ocean (Blank, Leinen, & Prospero, 1985; Leinen, Prospero, Arnold, & Blank, 1994; Merrill et al., 1994), in Atlantic Ocean (Biscaye, 1965) or in Greenland ice-cores (Biscaye et al., 1997; Drab, Gaudichet, Jaffrezo, & Colin, 2002) and snow pits (Bory, Biscyae, Svensson, & Grousset, 2002).

Mineralogical data on Asian dust from Chinese desert regions are not available because of limited studies in this field. In this work, we investigate spatial distribution of the mineralogy of eolian dust in Chinese desert regions (sampling sites, from west to east in northern China, were Aksu, Dunhuang, ZBT, and Tongliao) and the deposition region of Changwu, and then assess the possibility of using mineralogy as a source tracer for eolian dust by determining its origin.

2. Materials and methods

2.1. Sample collection

Bulk aerosol samples were collected at Aksu, Dunhuang, Zhenbeitai (ZBT, Yulin), Tongliao and Changwu stations (as shown in

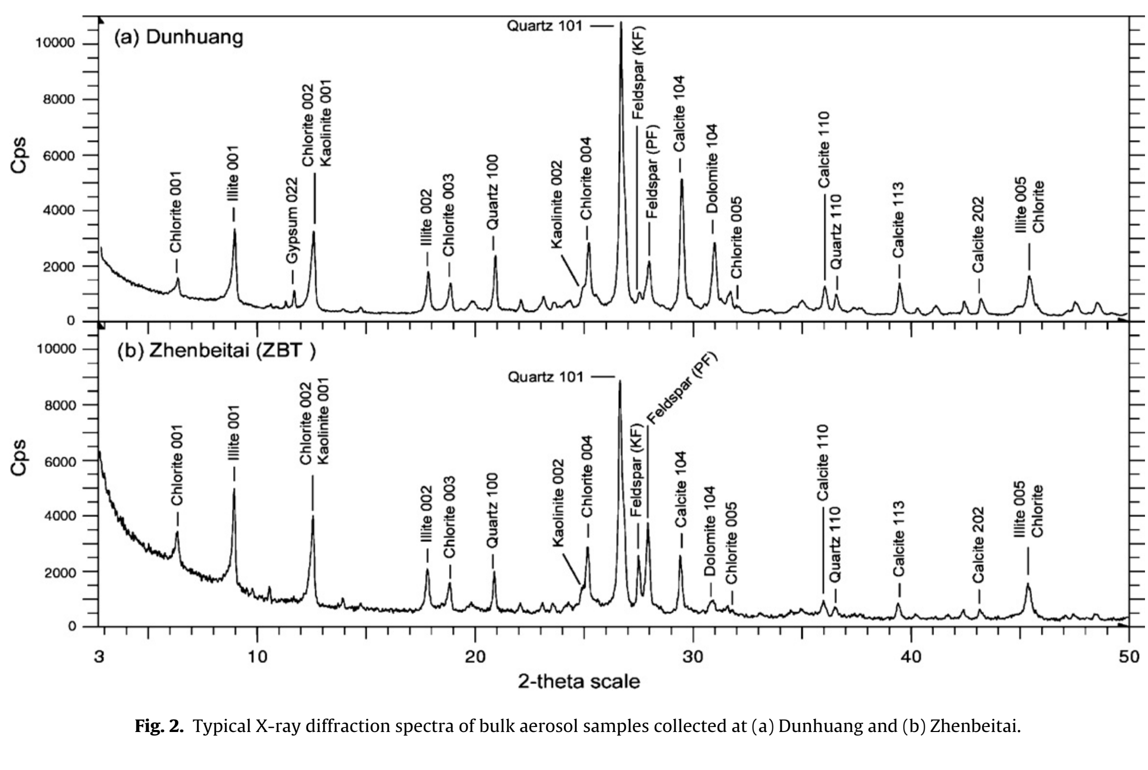

Fig. 2. Typical X-ray diffraction spectra of bulk aerosol samples collected at (a) Dunhuang and (b) Zhenbeitai.

Table 1
Replicate mineralogical analysis of Asian dust samples with arithmetic mean and standard deviation.

		Quartz	KF[a]	PF[b]	Calcite	Dolomite	Illite	Kaolinite + chlorite
Sample 1 (n = 5)	Mean	26.8	1.1	6.6	9.2	3.9	35.9	16.5
	SD	1.3	0.1	0.5	0.9	0.7	1.6	1.4
Sample 2 (n = 5)	Mean	26.4	0.9	5.2	11.4	5.8	34.4	15.9
	SD	0.6	0.2	0.3	0.7	0.5	1.1	1.0
Sample 3 (n = 5)	Mean	27.6	0.8	5.4	11.6	7.0	32.3	15.3
	SD	1.2	0.1	0.3	0.9	1.1	1.7	1.0

[a] K-feldspar.
[b] Plagioclase feldspar.

Fig. 1) that belong to the China dust storm research network of monitoring stations established in hyperarid and semiarid areas of northern China since February 2001. Dunhuang (DH) and Zhenbeitai stations also belong to the ACE-Asia ground station network. Two sampling collectors were used in the observations: RP-2025 air sampler and a bulk aerosol sampler. The RP-2025 air sampler (R&P Co., Inc., Albany, New York) has an automatic filter-changing system with a capacity of up to 16 filters (Teflon membrane, Cole-Parmer Instrument Company, Vernon Hills, Illinois) associated with an active volumetric flow-control system. It can work well under the condition of ~40 °C. Stations at ZBT, Changwu, and Tongliao were equipped with the RP-2025, and the flow rate was always 16.7 L/min. Andersen AN200 Sampler (which takes a filter holder with a pump of Andersen cascade impactor) was also used to collect the bulk aerosol samples on a Teflon membrane filter for Aksu and Dunhuang stations. The detailed description of each site and the sampling methods are summarized as follows.

Aksu station (40°16′N, 80°28′E, 1028 m above sea level) is located in a hyperarid area at the northern margin of Taklimakan Desert, 80 km east of Aksu city (Xinjiang Province, China). Bulk samples were collected on the top of a 20-m high building using Andersen AN200 sampler. The flow rate was 26.3 L/min, the sampling time was from 09:00 to 17:00 and the sampling period, from 13 May to 3 June.

Dunhuang station (40°30′N, 94°49′E, 1380 m above sea level) is situated at the hyperarid area of Kumtag Desert, 25 km southeast of Dunhuang city (Gansu province, China). Bulk samples were collected on the top of a 10-m high building using Andersen AN200 sampler, at the same flow rate of 26.3 L/min. The sampling time was from 10:00 to 16:00 and sampling period, from 29 April to 31 May 2001, and 1 March to 31 May 2002.

Zhenbeitai station (38°17′23″N, 109°42′18″E, 1135 m above sea level) is located at the southeastern edge of Mu Us desert, 10 km north of Yulin city (Shanxi Province, China). The areas to the north of ZBT belong to the vast hyperarid region of northern China. Samples were collected under northerly wind from 6:00 to 18:00 each day using RP-2025 sampler on the top of a 20-m-tall tower. Sampling period was from March to May 2001 and 2002.

Tongliao station (43°14′20″N, 122°14′E, 226 m above sea level) is located at the center of the Horqin sandy land, 20 km away from the Tongliao city. Samples were collected by using RP-2025 sampler placed at the top of a 10-m-tall tower. Sampling conditions were the same as ZBT. Sampling period was from March to May 2002.

The only deposition station was Changwu (35°12′N, 107°40′30″E, 1220 m above sea level), situated midwest of the Chinese Loess Plateau, 100 km from Xi'an city. Samples were also collected using RP-2025 sampler on top of a 10-m-high building. Sampling time consisted of two phases each day, 6:00 to 8:00 and 11:00 to 13:00. Sampling episode was from March to May 2001.

2.2. Mineralogical analysis by XRD

Samples were pre-treated according to the method developed by Caquineau, Magonthier, Gaudichet, and Gomes (1997) for low mineral-content aerosol samples. Particles were first extracted from the filter with deionised water (pH ~ 7.1), then concentrated by centrifugation (50,000 rpm for 30 min) and finally deposited on a pure silicon slide sample-holder characterized by very low and smooth XRD background (Queralt, sanfeliu, Gomez, & Alvarez, 2001).

X-ray experiment was carried out using Ragaku D/max-2500 diffractometer with Cu Kα-radiation at 50 kV and 100 mA. Scans were performed from 2.6° to 50° (2θ) at a rate of 1° (2θ) per minute. The major peaks were indexed, to permit recognition of the minerals: clays such as illite and chlorite + kaolinite; quartz; both K-feldspar (KF) and plagioclase (PF); carbonate (calcite and dolomite) and gypsum. Typical X-ray diffraction spectra of two Asian dust samples, collected respectively at Dunhuang and Zhenbeitai (ZBT), are shown in Fig. 2.

A semi-quantitative approach was used to evaluate the relative abundance of each mineral (mainly illite, and kaolinite + chlorite) as determined using the weighting factors of Biscaye (1965). Likewise, relative abundance of quartz, feldspar, carbonate and clay minerals was estimated using the following relationships:

- AllMin = Qz101 + (KF + PF) + Cal104 + Dol104 + AllClays,
- Quartz = Qz101/AllMin,
- K-feldspar = KF/AllMin.
- P-feldspar = PF/AllMin

Table 2
Semi-quantitative mineralogical composition of Asian dust collected in source and deposition areas as determined by XRD.

	Quartz	KF[a]	PF[b]	Calcite	Dolomite	Illite	Kaolinite + chlorite
Aksu (n = 11)	24 (3)	2 (1)	7 (2)	11 (1)	3 (1)	38 (3)	15 (2)
Dunhuang (n = 35)	19 (4)	1 (1)	6 (2)	9 (3)	4 (1)	43 (2)	18 (2)
ZBT (n = 37)	20 (5)	3 (1)	8 (2)	5 (3)	2 (1)	43 (5)	20 (2)
Tongliao (n = 11)	26 (6)	6 (2)	15 (8)	1 (1)	0 (1)	37 (7)	13 (2)
Changwu (n = 16)	15 (3)	1 (1)	4 (2)	5 (2)	2 (1)	50 (3)	23 (2)

( ) Standard deviation.
[a] K-feldspar.
[b] Plagioclase feldspar.

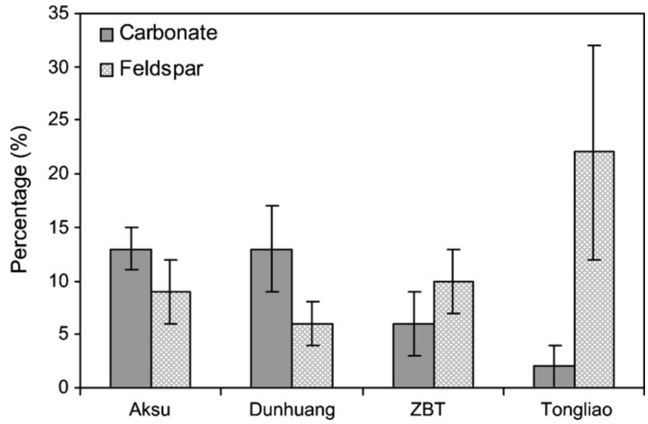

Fig. 3. Total carbonate (calcite plus dolomite) and feldspar (potassium plus plagioclase ones) average composition for dust samples collected in source areas.

- Calcite = Cal104/AllMin,
- Dolomite = Dol104/AllMin,
- Clay = AllClay/AllMin.

It should be noticed that the mineral composition thus obtained does not represent real weight percentages of the minerals, though such semi-quantitative approach is very useful for comparing samples with each other, provided that they have been analyzed in the same way.

We estimated the variability due to sample preparation by preparing and analyzing samples five successive times. Arithmetic means and standard deviation obtained for three dust samples are presented in Table 1. For non-clay minerals, the lowest variability observed is for quartz (less than 5%) and the highest for potassium feldspar (up to 19%). However, one should note that potassium feldspar is present in very small quantities in the replicate samples (around 1%). For clay minerals, very low variability is observed for illite (around 2%) and for kaolinite + chlorite (around 5%).

3. Results and discussion

3.1. Mineralogical characteristics of eolian dust at source regions

The X-ray diffraction data of Table 2 show that illite, kaolinite, chlorite, quartz, alkali feldspar (KF), plagioclase feldspar (PF), calcite, and dolomite are common minerals in Asian dust. Gypsum, halite and amphibole only occurred in few samples. From a qualitative point of view, all the samples have the same mineralogical composition. During all sampling periods, two types of meteorological conditions prevailed: dust storms (DS) as characterized by average wind speeds as high as 7.6 m/s, and non dust storm (N-DS), corresponding to days with calm or weak winds. For each sampling site, dust collected during both DS and N-DS events displays similar mineralogical compositions, that is, hardly any distinction. Table 2 shows the average mineral composition of dust samples at four desert regions.

Comparison of the mineralogical compositions of mineral dusts at four source regions, reveal certain quantitative differences, e.g., carbonate and feldspar contents are variable between sampling sites as shown in Fig. 3. The calcite content is similar for Aksu and Dunhuang (11% and 9%, respectively), though remarkably different at ZBT (5%), especially at Tongliao (1%). The distribution of dolomite content is similar as calcite at the four sampling sites, showing that the carbonate content in eolian dust decreases from west to east in northern China. However, the spatial distribution of the feldspar content shows some difference for carbonate minerals, varying from 9%, 6%, and 10% at Aksu, Dunhuang and ZBT, to 22% at Tongliao, that is, more than twice than other regions. As a result, the soil dust at Tongliao is characterized by relatively high feldspar and low carbonate content than the other three stations.

Elemental characteristics are also consistent with the mineralogical characteristics of Asia dust in different region sites. Prior study of elemental composition at Aksu, Dunhuang, ZBT, and Tongliao showed that Ca concentration at Aksu and Dunhuang (which represents the western source area) was higher than that at ZBT, which represents the northern source region (Zhang et al., 2003), and lowest Ca concentration was found at Tongliao, which represents the northeast source regions (Shen et al., 2007). The coincidence between element Ca concentration and carbonate content further supports the XRD experiments.

Furthermore, such a variability of the carbonate content in dust samples follows the same geographical distribution of carbonate in northern China soils. Indeed, Wang, Zhang, Arimoto, Cao, and Shen (2005) collected soils from western to eastern regions of northern China in areas close to our sampling sites and measured the carbonate content by a titration method. These authors showed that the Taklimakan Desert (northwestern China) had the highest carbonate content (∼12%) while the Horqin sandy land (northeastern China) displayed the lowest (<1%). The authors related this consistent decrease of carbonate soil content from west to east to climate variation, namely a strong increase of mean annual precipitation and a decrease in aridity from west to east of China. Feng, Endo, and Cheng (2002) also observed that the calcium carbonate content in northern China soils increased significantly with increasing mean temperature and decreasing annual precipitation. Recently, detrital dolomite was also used as a tracer for the source regions of Asian dust (Li et al., 2007). This study detected detrital dolomite only in the soil samples collected from the regions on the northern margin of Tibetan Plateau (NMTP), including Taklimakan Desert, Kumtag Desert, Qaidam Desert, Sanjiangyuan, Hexi Corridor, Badain Jaran Desert and their adjacent areas, but not in the samples from the deserts of northeastern China, such as Horqin sandy land and Onqin Daga desert. Thus mineral dust from the NMTP can be indicated by the presence of detrital dolomite. Consequently, carbonate and feldspar can be considered as regional dust tracers for the origin of Asian dust. Western source areas (represented by Aksu and Dunhuang stations) are characterized by high carbonate and low feldspar contents while northeastern source area (represented by Tongliao) displays low carbonate and high feldspar (especially PF) contents.

Quartz content at Aksu, Dunhuang and ZBT is lower than Tongliao, but the difference is not significant compared to carbonate or PF content. The clay mineralogy of dust is characterised by a high illite content (37–43%) and rather low chlorite + kaolinite contents (13–20%). Smectite has not been detected in the samples. Illite content at Tongliao is somewhat lower than those at Aksu, Dunhuang and ZBT, but the difference is negligible. As shown in Table 2, the chlorite + kaolinite content varied more significantly than illite at the four sampling sites.

As a result, we can summarize the mineralogical characteristics of dust aerosols collected at the source regions. Soil dust derived from western source regions (represent by Aksu and Dunhuang) is characterized by high carbonate content, which discriminates it from dust originating from northern and northeastern source regions. Soil dust derived from northeastern source regions (represented by Tongliao) is characterized by low carbonate and high feldspar (especially PF) contents, which differentiate it from dust from western source and northern source regions.

There is no specific mineral for each of the source areas, but the mineralogical characteristics of three source regions are different from one another. As mentioned above, the content of both calcite and PF changed uniquely for mineral dust and for surface soil originating from different sources. This difference calls for consideration while choosing the mineralogical signature of Asian dust on regional scale. PF belongs to coarse size particles (Blatt, Middleton, & Murray, 1980), which has short residence time in the atmosphere since it subsides readily during the transport process, thus leaving carbonate (including calcite and dolomite) as an appropriate tracer for eolian dust on regional scale.

3.2. Mineralogical characteristics of eolian dust at a deposition station: Changwu

Located more than 400 km far away from potential source regions, Changwu is assumed to be representative of deposited Asian dust after atmospheric transport, and particularly of the Chinese Loess Plateau. Table 2 also shows average mineralogical composition of mineral dust collected at the deposition site of Changwu. The same major minerals as for the other sampling sites are detected, e.g., clay minerals occur in the same relative proportion which means high illite content and low chlorite and kaolinite contents. Nevertheless, it can be noticed that the total clay content is significantly higher (73% in average) than in dust collected at the four source areas. This noticeable enrichment in clay minerals reflects a shift of the size distribution of the dust towards finer particles, possibly due to sedimentation processes during atmospheric transport which principally affect coarse particles. The average wind speed during dust storm events at Changwu is only 0.9 m/s, which is lower than those of 7.6 m/s at Aksu, 6.6 m/s at Dunhuang, 5.4 m/s at ZBT, and 7.4 m/s at Tongliao. Low wind speed is beneficial for large particles landing. The observed clay enrichment leads us to consider that local dust emission did not significantly affect the mineralogical composition of the dust collected at Changwu. The contents for quartz and PF are lower than those at source regions. Carbonate content is at the same level as dust collected at northern source area site of ZBT. It seems that soil dust collected at Changwu comes more likely from the northern source area provided we consider carbonate as tracer candidate for soil dust on a regional scale. The three big dust storm events at Changwu, which occurred on March 15 (DS1), April 8–10 (DS2), and April 29 (DS3) in 2001, can be used to assess the usefulness of carbonate in tracing Asian dust sources. The calcite content for these three dust storm events were 8.4%, 5.2%, and 4.1%, respectively. High calcite content indicates that DS1 most likely originated from Western source areas, while the other two DS events seemingly from northern source areas. Assuming that the mineralogy of the dust collected during DS at Changwu was not biased by local emission, we attempted to determine whether or not the dust originated from specific source areas.

To investigate the dust sources and transport pathways to Changwu, 2-day air-mass back trajectories arriving at 500 m AGL at 06:00 UTC were calculated using the NOAA HYSPLIT 4 trajectory model. Fig. 4 shows that the air masses ending at Changwu for DS1 originated from north-western areas, such as Taklimakan Desert and passing over the Hexi corridor. For DS2 and DS3, air mass transported nearly as the same pathway, which was associated with air masses coming directly from the north, i.e., from Mongolia and passing over Gobi Desert of central Mongolia and then to Changwu. Prior study of eolian dust collected at Xi'an also showed that mineral dust originating from western source area carried abundant carbonate (Cao et al., 2005; Shen et al., 2009). Therefore, carbonate may serve as a useful tracer for mineral dust on a regional scale. Sun, Zhang, and Liu (2001) reported that numerous case studies indicated that the dust deposited in proximal region, such as the Loess Plateau and offshore regions, originated dominantly from the Gobi Deserts in Mongolia and northern China rather than from the Taklimakan Desert. This was due to atmospheric transport that occurred at altitude lower than 3000 m for dust originating from Gobi Desert, and higher than 5000 m when originating from Taklimakan Desert. In the former case, dust deposit occurred in proximal region while, in the latter case, dust could be transported for very long distances.

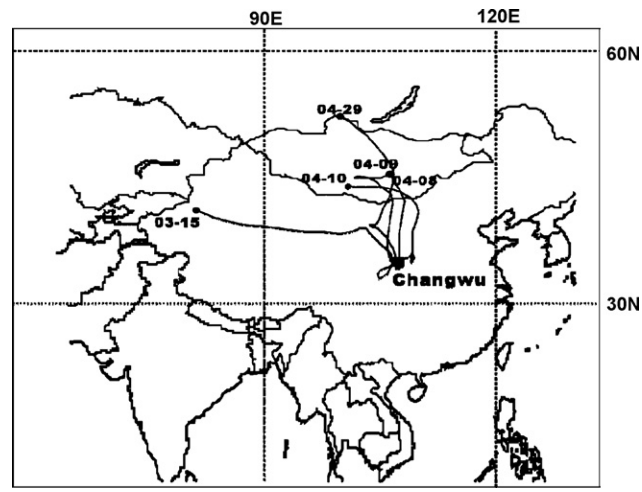

Fig. 4. 2-day air-mass back trajectories for three dust storm events at Changwu.

In the present study, contribution of the Taklimakan Desert cannot be definitively excluded (Cao et al., 2005). If air masses have carried dust emitted by more than one source, this dust could then have been mixed together during atmospheric transport before reaching Changwu. Such a hypothesis cannot be ruled out because of the similarities in clay mineralogy of the potential sources areas. Air-mass trajectory has proved that northern desert path predominates during spring 2001 at Changwu (Zhang et al., 2003).

Inasmuch as the source identified by carbonate content was consistent with the air-mass back trajectory results, such conclusion could be drawn that carbonate served as a good tracer for eolian dust on regional scale in northern China.

4. Conclusions

The mineralogical composition of bulk aerosol samples collected in four distinct source areas has been investigated. The main mineralogical difference in dust composition is its carbonate (especially calcite) and feldspar contents. Northwestern sources such as Taklimakan and Kuntag Deserts are characterised by their highest calcite contents while the Horqin sandy land (in northeastern China) displays the lowest. The mineralogical characteristics are consistent with the elemental composition for dusts from the different source regions. This finding also well reflects the variability of the carbonate composition of underlying soils. Feldspar minerals vary in the opposite direction of carbonate, the highest content being observed in northeastern source areas. On the other hand, the clay mineralogy of the dust displays little variation within the source regions with high illite and low kaolinite + chlorite contents. The mineralogical composition of dust collected in the deposition area of the Chinese Loess Plateau (Changwu) was enriched by clay minerals. Back air-mass trajectories for the three dust storm events revealed that the specific calcite signature of Asian dust was not modified after atmospheric transport, even if significant enrichment in total clay content was observed as compared to dust collected in source areas. Finally, carbonate content provides a useful tool for studying provenance of long-range atmospheric transported Asian dust on regional source.

Acknowledgments

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