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Mass Concentration and Mineralogical Characteristics of Aerosol Particles Collected at Dunhuang During ACE-Asia

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ABSTRACT

Measurements were performed in spring 2001 and 2002 to determine the characteristics of soil dust in the Chinese desert region of Dunhuang, one of the ground sites of the Asia-Pacific Regional Aerosol Characterization Experiment (ACE-Asia). The mean mass concentrations of total suspended particle matter during the spring of 2001 and 2002 were 317 μg m^{-3} and 307 μg m^{-3}, respectively. Eleven dust storm events were observed with a mean aerosol concentration of 1095 μg m^{-3}, while the non-dusty days with calm or weak wind speed had a background aerosol loading of 196 μg m^{-3} on average in the springtime. The main minerals detected in the aerosol samples by X-ray diffraction were illite, kaolinite, chlorite, quartz, feldspar, calcite and dolomite. Gypsum, halite and amphibole were also detected in a few samples. The mineralogical data also show that Asian dust is characterized by a kaolinite to chlorite (K/C) ratio lower than 1 whereas Saharan dust exhibits a K/C ratio larger than 2. Air mass back-trajectory analysis show that three families of pathways are associated with the aerosol particle transport to Dunhuang, but these have similar K/C ratios, which further demonstrates that the mineralogical characteristics of Asian dust are different from African dust.

Key words: soil dust, mass concentration, mineralogical composition, clay ratio

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1. Introduction

Soil dust is one of the important atmospheric components, not only because it has a strong effect on the urban and rural atmospheric environment (Wang et al., 2001; Zhuang et al., 2001; Cao et al., 2005; Zhang et al., 2005), but also because soil dust particles affect the Earth's radiative forcing directly by absorbing and scattering incoming solar radiation and terrestrial radiation (long wave) and indirectly by acting as cloud condensation nuclei (CCN) (Charlson et al., 1992; Li et al., 1996; Sokolik and Toon, 1996; Tegen et al., 1996). The extinction of radiant energy by mineral dust can cool or heat the atmosphere, depending on the chemical and mineralogical composition of the aerosol, the physical properties and vertical distribution of the aerosol in the atmosphere, as well as the characteristics of the ground surface. Eolian dust is typically a complicated mixture of various minerals, and the optical properties (extinction coefficient, single scattering albedo and asymmetry parameter) of the dust vary depending on mineralogical composition (Sokolik and Toon, 1999; Soklik et al., 1998). For example, hematite has the largest absorption at UV and visible wavelengths, whereas quartz and clay minerals have the strongest absorption bands at IR wavelengths.

Desert regions in East Asia are considered to be the major sources for Asian dust according to the rain-dust records in Chinese historical documents and present day observations (Zhang, 1984; Merrill et al., 1994; Xu et al., 2004;). The annual input of mineral aerosols to the atmosphere from the arid and semiarid regions of North China is estimated to be about 800 Tg (Zhang

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et al., 1997), contributing to about half of the global dust output (Andreae, 1995; Duce et al., 1995). Unfortunately, the estimation of radiative forcing by soil dust still has much uncertainty, which is mainly due to the poor knowledge of the aerosols' chemical, physical and optical properties. For example, mineralogical data of Asian dust from Chinese desert regions are not available because of the limited study in this field. Many prior studies on mineralogy have focused on the urban atmospheric particles (Chen and Xu, 2003; Liu et al., 2004; Lu et al., 2005; Shi et al., 2005).

ACE-Asia was an international program that involved sampling from aircraft, onboard ships and from a network of ground stations. One of the goals of ACE-Asia was to determine and understand the properties and controlling factors of the aerosols in the anthropogenically modified atmosphere of Eastern Asia and the Northwest Pacific and to assess their relevance for the radiative forcing of climate (Huebert et al., 2003). As part of ACE-Asia, an experimental campaign was carried out in China during the years 2001 and 2002. Here we present the results of soil dust studies conducted at the ACE-Asia ground station in Dunhuang (in Gansu province, China). The main purposes of this study were (1) to investigate the aerosol mass concentration and its temporal variation at Chinese desert regions during the intensive observation period, (2) to characterize the mineralogy of Asian dust in Chinese desert regions and to compare this with African dust, and (3) to identify the transport pathways of aerosols at Dunhuang.

2. Methods

2.1 *Aerosol sampling*

Aerosol samples for this study were collected in spring 2001 and 2002 at Dunhuang (Gansu Province, China). Dunhuang station (40°30′N, 94°49′E, 1380 m above sea level), which was established at the end of April 2001, is situated in the hyper-arid area of Kumtag Desert (mean annual precipitation of about 50 mm), 25 km southeast of Dunhuang city (Gansu province, China). Bulk samples were collected daily on the top of a 10-m-high building using Teflon® membrane filters (Cole-Parmer Instrument Company, Vernon Hills, Illinois). The Dunhuang station was equipped with an Andersen AN200 Sampler fitted with a total suspended particle (TSP) inlet which operated at a flow rate of 26.3 L min^{-1}. Ths sampling period is from 29 April to 31 May 2001 and 1 March to 31 May 2002, between 1000 and 1800 LST. A total of 108 samples are available in this research.

2.2 *Instrumental analyses*

Prior to mineralogical analysis by X-ray diffraction (XRD), chemical analyses were performed at Beijing Normal University on bulk samples using a proton-induced X-ray emission (PIXE) method (Zhu and Wang, 1998).

For mineralogical analysis, sample pre-treatment was done following the method developed by Caquineau et al. (1997) for low-mass mineral aerosol samples. Particles were first extracted from the filter with deionised water, then concentrated and finally deposited on a pure silicon slide, such a sample-holder exhibiting a very low and smooth XRD background (Queralt et al., 2001).

XRD analysis was performed at the Research Institute of Petroleum Exploration and Development (Beijing, China). The analysis was carried out on a Rigaku D/max-2500 diffractometer with CuKα radiation at 50 kV and 100 mA. Scans were performed from 2.6° to 50°2θ at a rate of 1°2θ per minute. After that, a high resolution scan from 24° to 26°2θ at a rate of 0.1°2θ per minute was performed to determine the relative contribution of kaolinite and chlorite to the 7Å peak. Peak areas were converted to weight percentages using the weighting factors of Biscaye (1965) and Svensson et al. (2000). The content of clay minerals was determined using the weighting factors of Biscaye (1965). Likewise, the relative abundance of quartz, feldspar, carbonates and clay minerals were estimated using the following relationships:

AllMin=Qz101 + (KF + PF) + Cal104 + Dol104 +AllClays,

Quartz=Qz101/AllMin,

K-Feldspar=KF/AllMin.

P-Feldspar=PF/AllMin

Calcite=Cal104/AllMin,

Dolomite=Dol104/AllMin,

Clay=AllClay/AllMin.

It is a simple semi-quantitative approach. Although the obtained mineral percentages do not represent the real weight composition of the sample, these mineralogical data are very useful to compare samples to each others, provided that they have been analysed in the same way (Biscaye et al., 1965; Svensson et al., 2000).

3. Results and discussion

3.1 *Temporal variation of TSP mass concentration*

Observed TSP mass concentrations are plotted in Fig. 1. The average TSP concentrations during the spring of 2001 and 2002 are 317 μg m^{-3} and 307 μg m^{-3}, respectively, higher than the ACE-Asia supersite Zhenbeitai (ZBT, Yulin, Shaanxi province) with an average value of of 260 μg m^{-3} during spring 2001, but lower than the high dust loading of 926 μg m^{-3}

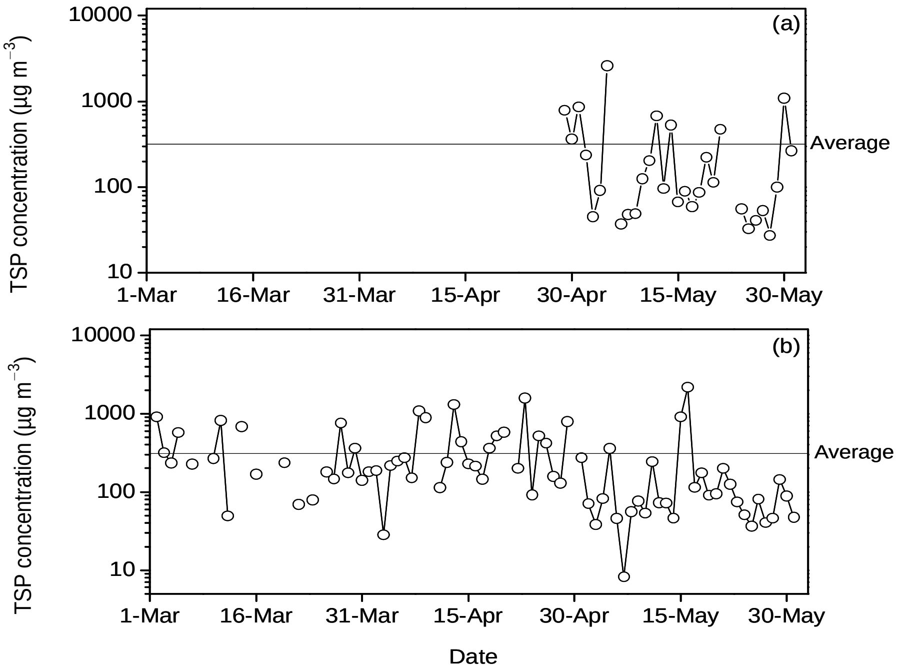

Fig. 1. TSP mass concentration in spring (a) 2001 and (b) 2002 at the Dunhuang site.

at the Taklimakan desert station of Aksu (Zhang et al., 2003). Prior studies have shown that typical dust loadings at downwind depositional areas during spring 2001 were 210 μg m^{-3} at Changwu (Shaanxi province) and 300 μg m^{-3} at Qingdao (Zhang et al., 2003; Zhang et al., 2004). Therefore, high dust loadings were observed in the desert regions.

During spring 2002, the average TSP concentrations of March, April and May were 336, 418, and 193 μg m^{-3}, respectively at the Dunhuang site. The TSP concentration of May 2001 was 299 μg m^{-3}, higher than the TSP loading of May 2002. The observations of aerosol particles in other desert regions and dust depositional areas also show that high dust concentrations often occurred in April (Niu et al., 2001; Zhang et al., 2003, 2004), implying that April has a high frequency of dust storm occurrence. Urban airborne particle concentrations in Beijing during April, August, October and December in 2002 are 509, 245, 319 and 281 μg m^{-3} on average, respectively (Zhang et al., 2004), which shows that even in the non dust storm seasons, the aerosol particle loading in the urban atmospherie are higher than in Asian dust source regions. There is no doubt that natural soil dust can contribute to the city atmospheric particle pollution, but anthropogenic aerosols also make an important contribution to the city air pollution.

During the sampling period, two kinds of meteorological conditions prevailed: dust storm (DS) conditions characterized by high wind speed and low visibility, and non dust storm (N-DS) conditions corresponding with calm and weak wind. During the whole of spring 2001 (only May) and 2002, eleven DS episodes were observed at Dunhuang station with TSP levels (Table 1) substantially higher than the springtime average concentration of 307 μg m^{-3}. The average TSP concentration of the DS episodes was 1095 μg m^{-3}. During the sampling period, three strong dust storms were observed: 5 May 2001 (called DS 1), 20 March 2002, and 16 May 2002 (DS10). We lost the data of the DS on 20 March 2002 due to a power failure, but a high Particulate Matter (PM) concentration was observed in other regions (Sugimoto et al., 2003; Cao et al., 2005; Zhang et al., 2005). The TSP loadings for DS1 and DS10 are 2591 and 2177 μg m^{-3} at the Dunhuang site, respectively. Meteorological observation also shows that most of the DS events at Dunhuang were associated with high wind speed and low visibility. For example, during DS1, the average wind speed was 11 m s^{-1}, and the visibility was only 200 m.

In comparison with the DS events, the non-dusty days with calm or weak wind speed had a background aerosol loading average of 196 μg m^{-3} in springtime, and 274 μg m^{-3} in March, 261 μg m^{-3} in April, and 91 μg m^{-3} in May in 2002. In spring 2001, we only have one month of samples, and the results show that the aerosol mass concentration on non-dusty days was 131 μg m^{-3} in May. At the ZBT station, the aerosol loadings on non-dusty days were 300 μg m^{-3} in March and 430 μg m^{-3} in May, which are higher than at Dunhuang station (Zhang et al., 2003). This difference

Table 1. Observed of TSP concentration and mineralogical composition during eleven dust storms in spring 2001 and 2002 at Dunhuang, China.

DS numbers	Date	TSP[a]	Illite[b]	K	C	Quartz	KF	PF	Calcite	Dolomite	K/C	I/K
DS1	5 May 2001	2591.0	69	12	18	21	1	4	9	4	0.66	6
DS2	12 May 2001	677.0	70	10	19	19	2	6	4	2	0.54	7
DS3	30 May 2001	1085.9	70	8	22	19	1	3	6	3	0.38	8
DS4	2 Mar 2002	908.9	70	11	19	26	2	6	11	5	0.55	6
DS5	11 Mar 2002	817.1	75	6	19	21	1	5	8	4	0.31	13
DS6	8–9 Apr 2002	987.3	70	12	18	20	1	4	11	6	0.67	6
DS7	13 Apr 2002	1309.2	66	10	24	25	1	10	11	4	0.40	7
DS8	23 Apr 2002	1579.9	68	11	21	26	1	6	12	6	0.55	6
DS9	25–26 Apr 2002	470.1	73	8	19	18	1	5	5	5	0.40	10
DS10	5 May 2002	357.7	69	9	22	20	1	6	8	4	0.40	8
DS11	15–16 May 2002	1545.0	67	13	20	24	1	5	10	5	0.64	5

[a] mass concentration in μg per standard cubic meter; [b]mineral relative content in pencentage.

may reflect the significant contributions of the anthropogenic sources to the ZBT station. Because ZBT is close to Yulin County, pollution aerosols are easily entrained by the southerly wind and mixed with the mineral dust (Alfaro et al., 2003; Zhang et al., 2003; Arimoto et al., 2004). At the deposition area of the Chinese Loess Plateau station of Changwu, the TSP loading during N-DS was 173 μg m^{-3}, similar to the desert region station as observed at Dunhuang.

3.2 *Mineralogical composition of Asian dust at Dunhuang station*

XRD experiments show that illit, kaolinite (Kao), chlorite (Chl), quartz, potassium feldspar (KF), plagioclase feldspar (PF), and carbonate (generally Calcite and dolomite) are common minerals in Asian dust (Table 1). Gypsum, halite and amphibole only occurred in a few samples. This result is consistent with the research by Trochkine et al. (2003) using a scanning electron microscope with an energy dispersive X-ray analyzer. Figure 2 is a typical XRD pattern of low mass aerosol samples. The major peaks are indexed, allowing us to recognise the main minerals as mentioned above. In a qualitative view, all the samples collected at Dunhuang have the same mineralogical composition. In fact, the difference in the mineral composition between the DS and N-DS conditions are slight at the Chinese desert region station of Dunhuang (Table 2). This result implies that soil dust in both DS and N-DS conditions has similar sources.

Calcite content is 9% on average, and varies from 3% to 15%. Another carbonate mineral, dolomite, is 4% on average and varies from 2% to 8%. This carbonate content is higher than the aerosol collected at Changwu during spring 2001, in which the calcite content is only 4% (Shen et al., 2004). An air mass back-trajectory analysis also proved that northern desert source areas are a major source of mineral dust in Changwu. Investigation of the soil dust mineralogy shows that carbonate content decreases from the western to eastern source areas (Shen et al., 2004). Recently, several studies have also found a variation in carbonate content or cations of Ca^{2+} in surface soil samples or Asian dust aerosols in Chinese deserts (Arimoto et al., 2004; Zhang et al., 2003; Wang et al., 2005). But one should be careful when using carbonate to trace eolian dust origins, because sedimentation processes occurring during atmospheric transport principally affect coarser particles like carbonate. Two other coarse-sized minerals, quartz (20% on average, varying from 12% to 27%) and plagioclase feldspar (5% on average, ranging from 3% to 10%), at Dunhuang are also slightly higher than the aerosols at Changwu.

The clay minerals of the Dunhuang samples are characterized by a high illite content (66%–75%) and rather low chlorite (17%–24%) and kaolinite (6%–15%) contents. The average contents of illite, chlorite, and kaolinite are 70%, 20%, and 10%, respectively (Table 1). Illite systematically dominates the clay fraction, and smectite is not detected in the samples. The clay mineral content is similar to the aerosols at Changwu (Shen et al., 2004) since sedimentation processes during atmospheric transport have little effect on the fine-sized particles.

The K/C (kaolinite/chlorite) and I/K (illite/kaolinite) ratios are also given in Table 1. The K/C ratio is 0.48 on average, ranging from 0.31 to 0.75 (Fig. 3), while the I/K ratio is 7.71 on average, varying between 4.5 and 12.5. Aerosols collected at other Chinese desert regions and depositional areas also show similar K/C ratios, ranging from 0.13 to 0.96 (Shen et al., 2005). Such values of the K/C ratio are in good agreement with that of 0.45 measured by

Table 2. Summary statistics for the TSP concentration and mineralogical composition during dust storms and non dust storm conditions in spring 2001 and 2002 at Dunhuang, China.

DS numbers	Date	TSP	Illite	K	C	Quartz	KF	PF	Calcite	Dolomite	K/C	I/K
Average DS[a]	14	1147.1	70	10	20	22	1	5	9	4	0.50	7
SD[b]		643.5	2.6	2.3	1.9	3.2	0.5	1.8	2.9	1.8	0.1	2.1
Average Non-DS[c]	95	194.0	71	9	20	19	2	5	9	4	0.45	8
SD		190.0	1.9	1.4	1.3	3.5	0.7	1.7	2.8	1.0	0.1	2
Total samples	109	310.0	70	10	20	20	1	5	9	4	0.48	8
SD		419.0	2.2	1.9	1.5	3.5	0.6	1.7	2.7	1.3	0.1	1.8

[a]Dust storm envent; [b]Standard deviation; [c]Non dust strom

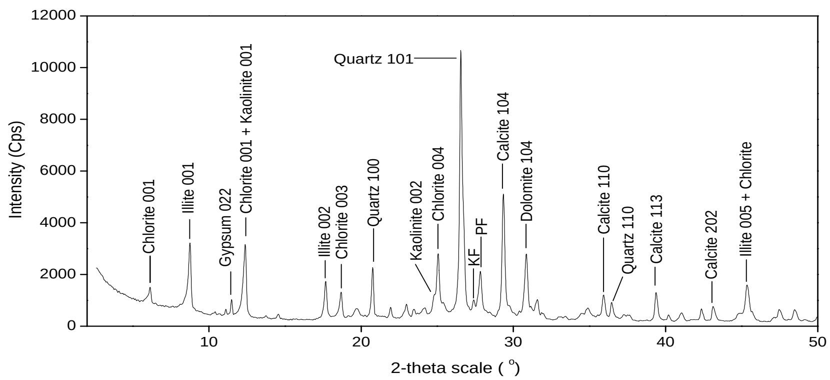

Fig. 2. Typical XRD pattern of a soil dust sample at Dunhuang.

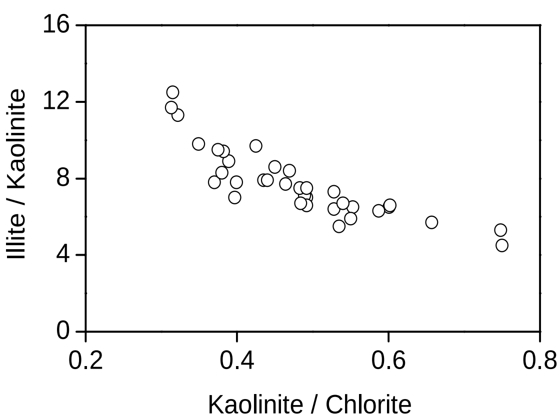

Fig. 3. Illite-to-kaolinite ratio (I/K) versus kaolinite-to-chlorite ratio (K/C) for bulk aerosol samples collected at Dunhang.

Sun et al. (2000) in soil dust collected in Beijing during a very strong dust storm in April 1998 and proved to originate from the Gobi area of southern Mongolia and the Gobi and desert areas of northern China.

Moreover, this K/C ratio is often used to distinguish between the low latitude desert sources such as Sahara (rich in kaolinite, e.g. Chester et al., 1972; Drees et al., 1993) and high latitude regions in Asia (rich in Chlorite, e.g., Heath and Pisias, 1979) when studying dust areas (Biscaye et al., 1997; Svensson et al., 2000; Drab et al., 2002). For instance, in studying the surface soil and modern aerosol samples [called "possible source area" (PSA) samples] in order to compare them with Last Glacial Maximum (LGM) mineral dust, Biscaye et al. (1997) and Svensson et al. (2000) obtained measured K/C ratios between 0.35 and 0.91 for Asian PSA samples (collected in northern China and southern Mongolia). Soil dust representative of the modern conditions in Greenland snow pits and ice cores also displays a similar mineralogical signature (Bory et al., 2002; Drab et al., 2002). This clay mineral ratio can also be compared with African dust. Several studies of Saharan dust clearly point out that kaolinite is rather abundant compared to chlorite, leading to a K/C ratio generally larger than 2 (Glaccum and Prospero, 1980; Chester et al., 1984; Molinaroli, 1996; Avila et al., 1997) or even the absence of chlorite (Caquineau et al., 1998).

The result shows that Asian dust has a special clay mineral composition, characterized by a K/C ratio lower than 1, while the K/C ratio in Sahara dust is larger than 2. This obvious difference between Asian dust and African dust suggests that the K/C ratio provides a good signature to trace the soil dust origin on a hemispheric scale.

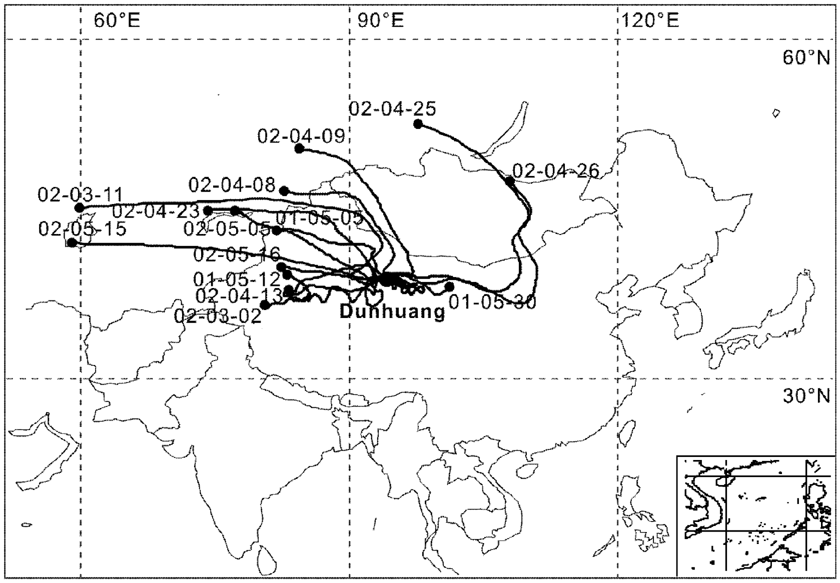

Fig. 4. Air mass back-trajectory during a duststorm (DS) period at Dunhuang.

3.3 *Transport pathways of dust storms at Dunhuang station*

Five-day back trajectories arriving at 1100 m above ground level (a.g.l.) (0600 UTC) were calculated for Dunhuang using the NOAA HYSPLIT4 trajectory model to investigate the soil dust transport pathways. Figure 4 only shows the air mass back-trajectories of the eleven dust events at Dunhuang station. Three general pathways of surface trajectories that passed over Dunhuang during spring 2002 were identified. The first pathway family shows that an air mass passed through the southern part of Xinjiang Province (Taklimakan Desert), moving east to Dunhuang. DS2 (12 May 2001), DS4 (2 March 2002), DS7 (13 April 2002), and DS11 (16 May 2002) belong to this pathway family. The second family of pathways mainly came from Kazakhstan and then passed through the Gobi and desert region (Gurbantunggut Desert) of the northern part of Xinjiang Province or the southwestern desert and desert-margin of Mongolia to Dunhuang. This family of pathways includes DS1 (5 May 2001), DS5 (11 March 2002), DS6 (8–9 April 2002), DS8 (23 April 2002), and DS10 (5 May 2002). In the third family of pathways [DS3 (30 May 2001), DS9 (25–26 April 2002)], the air masses mainly came from the high latitudes and passed through northern and southern Mongolia into the central regions of Inner Mongolia (China), and finally turned west and passed through the Gobi and desert regions of the western part of Inner Mongolia to Dunhuang.

The air mass back-trajectory results also prove that the Asian dust originating from different source regions has a similar clay mineral composition (focused on K/C ratio). This K/C ratio can supply another tool to distinguish between Asian dust and African dust.

4. Conclusions

Measurements were performed in spring 2001 and 2002 to determine the characteristics of soil dust at the Chinese desert region of Dunhuang. The mean TSP mass concentrations in spring 2001 and 2002 were 317 μg m^{-3} and 307 μg m^{-3}, respectively. TSP loading during DS episodes was 1095 μg m^{-3} on average, higher than the background aerosol loading during N-DS conditions of 196 μg m^{-3}. On a regional scale, Asian dust shows a similar K/C ratio varying from 0.13 to 0.96. On a global scale, we have shown that Asian dust displays a very specific K/C ratio lower than 1, whereas Saharan dust displays a K/C ratio larger than 2. Moreover, these results can be used to identify the origin of soil dust in Greenland ice cores, either for LGM conditions or modern ones. An air mass back-trajectory analysis shows three families of pathways are associated with the aerosol particle transport to Dunhuang. Regardless of the air mass transport pathway, a similar K/C ratio was observed, which further demonstrated this mineralogical characteristic of Asian dust.

Our research paid more attention to mass concentration and mineralogical characteristics of the Total Suspended Particle matter, whereas the detection of fine size particle properties of Asian dust, for example, $PM_{2.5}$, needs further work.

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