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Characteristics of carbonate content and carbon and oxygen isotopic composition of northern China soil and dust aerosol and its application to tracing dust sources

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Abstract

The carbonate content and the carbon and oxygen isotopic composition of carbonate were determined for 28 surface soil samples from northern China and 30 dust aerosol samples collected at four sites in northern China: Aksu, Dunhuang, Yulin and XiAn. The carbonate content and stable C isotopic composition show distinctly different values for dust from different sources, both vary inversely with annual mean precipitation and also vary as function of grain size. The $\delta^{13}C$ value in the aerosol is lower and the $\delta^{18}O$ higher than in local bulk surface soils because the particle-size distribution in the aerosol is skew toward smaller particles. At Aksu and Dunhuang, the dust aerosol shows a higher $\delta^{13}C$ value (average-0.148‰) and also a higher carbonate content (average 10.2%) compared with Yulin. The relative contributions of remote and local sources to Yunlin aerosol was estimated by a two component isotopic mixing equation using $\delta^{13}C$ data. At XiAn, the aerosols collected during a dust storm had higher $\delta^{13}C$ value than local fugitive dusts, indicating a significant contribution from long-range transport. Stable C isotopic composition provides a useful index for identifying putative sources for ambient dust aerosol, especially when used in combination with meteorological methods such as back trajectory analysis.
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Keywords: Carbonate; Carbon and oxygen isotopes; Asian dust source

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1. Introduction

Dust storms are atmospheric phenomena that have recently been recognized as having a wide range of environmental impacts. Tropospheric aerosols, including dust, are an important component of the earth's climate system and modify climate through their direct radiative effects of scattering and absorption (Tegen and Lacis, 1996) and through their indirect effects on clouds and cloud microphysics (Rosenfield et al., 1997).

Carbonate can affect atmospheric chemical processes and aerosol characteristics because the acid neutralizing capacity of this species facilitates the heterogeneous conversion of sulfur and nitrogen oxides to particulate sulfate and nitrate (Dentener et al., 1996). In East Asia, the acid binding capacity of the suspended dust can reduce the acidity of rain (Kang and Kim, 1995).

Chemical, mineralogical and isotopic methods have been used in attempts to distinguish the source regions for atmospheric dust and eolian deposits. For example, an elemental tracer system has been used to apportion the fluxes of China-dust among sources using the ratios of Fe/Al, Mg/Al and Sc/Al (Zhang et al., 1996). The relative abundances of the clay minerals illite and kaolinite have been used to trace the regional origins of Saharan dust over the North Atlantic Ocean (Caquineau et al., 1998) and northeastern Spain (Avila et al., 1997). The particle-size distribution and oxygen-isotopic composition of quartz has been used to investigate the eolian origins of quartz in soils from the Hawaiian Islands and in Pacific pelagic sediments (Rex et al., 1969; Clayton et al., 1972). Information on oxygen isotopes in single micrometer-sized quartz grains has been used to trace the sources for Saharan dust transported long distances (Aleon et al., 2002).

Much of the early work on identifying the provenance for dust in deep-sea sediments and aerosols made use of information on strontium and neodymium isotopes (Biscaye and Dasch, 1971; Grousset et al., 1988). More recently, some research has focused on using lead isotopes to identify the sources for Asian dust. Jones et al. (2000) evaluated the importance of natural eolian Pb to the dissolved oceanic Pb budget by comparing the isotopic composition of Pb in sediment samples from the North Pacific with that of Chinese loess. Kanayama et al. (2002) estimated the contribution of Chinese desert dust to the atmospheric aerosol in Japan using Rb–Sr isotopic ratios. Nakano et al. (2004) reported that arid soils in various areas of northern China could be distinguished by using Sr–Nd isotope ratios of acid-resistant minerals and the ratios of Sr isotopes in water- and weak-acid-soluble minerals.

Carbonate is a prominent component of the soils in northwestern China where much of the Asian dust is produced (Liu, 1985; Feng et al., 2002). The carbon and oxygen isotopic composition of pedogenic carbonate is controlled by the isotopic composition of soil CO_2 and by the oxygen isotopic composition of meteoric waters (Cerling, 1984). C and O isotopes previously been used to construct climatic indices in studies of loess (Wang and Follmer, 1998; Ding and Yang, 2000), but the distribution of stable C–O isotopes in pedogenic carbonate from northern China has not been investigated in detail. The data for dust aerosols are particularly scant. Here, we report on the spatial distribution of carbonate content and the C–O isotopic composition in surface soils and dust aerosols from northern China. The dust-aerosol sources are then evaluated using geochemical methods based on these data.

2. Samples and experimental methods

2.1. Samples

Twenty-eight surface soil samples were collected in arid and semiarid areas of China including the Taklimakan Desert, Gurbantunggut Desert, Kumtag Desert, Gobi area in Hexi corridor, Badain Juran Desert, Tengger Desert, Ulan Buh Desert, Hobq Desert, Mu Us Desert, Onqin Daga sandy land, Horqin sandy land and steppe area in mid-Inner Mongolia in summer 2001. Fig. 1 shows the locations where the soil samples were collected.

Aerosol bulk samples were collected at four sites in northwestern China from 2001 to 2003. These sites are (1) Aksu (40°16′N, 80°28′E, 1028 m above sea level, asl), located in a hyperarid area on the northern margin of Taklimakan Desert; (2) Dunhuang (40°30′N, 94°49′E, 1380 m asl), located in a hyperarid area in the Kumtag Desert; (3) Yulin (38°17′N, 109°42′E, 1135 m asl), located along the southeastern edge of Mu Us Desert; and (4) XiAn (34°14′N, 108°53′E, 440 m asl), in the southern edge of the Loess Plateau (see Fig. 1). All the samples were collected on the roofs of buildings (about 10 m high) using filter holders connected to pumps from Andersen cascade impactors (Andersen AN200) on 47-mm diameter, 1-μm pore size Teflo® membrane filters (Pall Gelman Laboratory, Ann Arbor, MI). Two fugitive dust samples were collected near ground-level beside a road in XiAn, to evaluate the composition of local aerosol sources. The collections were made during October, which is a time of year when Asian dust does not strongly affect the aerosol loadings in XiAn. One farmland soil sample, two road deposit samples and two natural soil samples in XiAn also were collected to investigate the composition of these local source materials.

2.2. Experimental

The soil samples were crushed to 200 mesh with the use of an agate sharpener. A rapid titration method (Hesse, 1972) was used to determine the carbonate content of the soils. The uncertainty in the carbonate determinations is ~1%.

The soil samples were then allowed to react with excess 100% H_3PO_4 at 75 °C for 2 h. The CO_2 generated from the samples was cryogenically trapped and analyzed with the use of a Finnigan MAT 251 mass spectrometer. Isotopic results for carbonate are
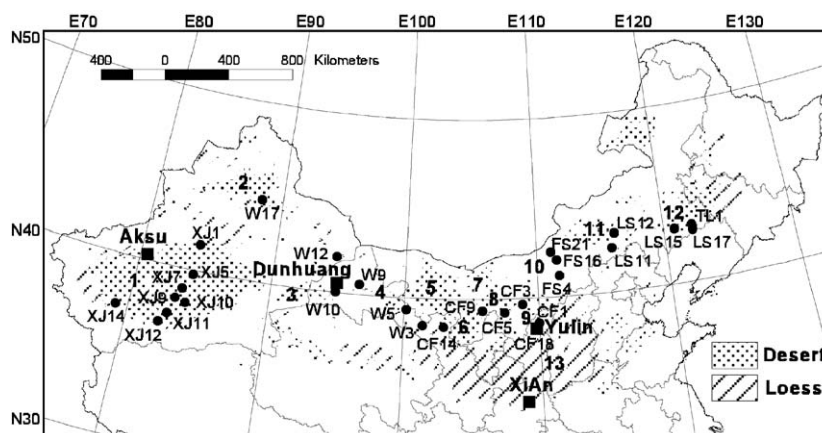
Fig. 1. The location of the samples. The dots show the locations of soil samples and the squares show the locations of aerosol samples. In this figure, 13 surface soil sampling areas are: 1, Taklimakan Desert; 2, Gurbantunggut Desert; 3, Kumtag Desert; 4, Gobi area in Hexi corridor; 5, Badain Juran Desert; 6, Tengger Desert; 7, Ulan Buh Desert; 8, Hobq Desert; 9, Mu Us Desert; 10, steppe area in mid Inner Mongolia; 11, Onqin Daga sandy land; 12, Horqin sandy land; and 13, Loess Plateau.

presented in the usual δ notation as the per mil (‰) deviation of the sample CO_2 from the Pee Dee Belemnite standard.

$$\delta = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000\text{‰}, \tag{1}$$

where R is either the isotopic carbon or oxygen ratio, ($^{13}C/^{12}C$) or ($^{18}O/^{16}O$), respectively. The standard deviation of the analysis is ± 0.1‰ for $\delta^{13}C$ and ± 0.2‰ for $\delta^{18}O$.

Three soil samples (XJ7 and XJ11 from Taklimakan Desert and W10 from Kumtag Desert) were separated into six size fractions (250–125, 125–100, 100–80, 80–63, 63–40 and $< 40 \mu m$) using a dry sieving method. The carbonate content and isotopic composition of the size-separated samples were determined using the same procedures described above for the bulk samples.

All aerosol sample filters were weighed using a Mettler Balance Model 3 (Mettler Toledo Instruments Co., Ltd., Switzerland, 1 μg resolution) to obtain the aerosol mass concentrations. The mineral composition of aerosol samples was determined by X-ray diffraction, and the carbonate contents were identified using a simple semi-quantitative approach (Biscaye, 1965; Anders et al., 2000). The results of the mineralogical analyses will be presented in a separate paper.

The stable carbon and oxygen isotope determinations for the carbonate extracted from the aerosol samples were performed on a Finnigan MAT 252 mass spectrometer fitted with an online automatic carbonate reaction system ("Kiel device"). Detailed preparation and analysis procedures have been presented by Cao et al. (2004). Isotopic results are presented using the same δ notations as is commonly done for soils. Thirty-two of the samples, including two fugitive dust samples in

XiAn, from the entire set of 86 samples had sufficient mass loadings for stable C–O isotope analyses.

3. Results and discussion

3.1. Results on soils

The results of the carbonate content and stable C–O isotopic composition of the surface soil samples are summarized in Table 1. The mean carbonate concentrations in soils from arid and semiarid areas in northern China decrease systematically from west to east (Fig. 2). Soils from the Taklimakan Desert in northwestern China had the highest carbonate content (11.8%) of the samples analyzed while the lowest value (0.3%) was observed in the soil samples from the Horqin sandy land in northeastern China (Table 1). The carbonate contents in soils from the Kumtag Desert and Hexi corridor are also high: 8.0% and 9.9%, respectively. In the soil from the Badain Juran Desert, the carbonate content was 4.6%. Comparatively low carbonate contents were found in soils from Ulan Buh Desert, Tengger Desert, Mu Us Desert and the steppe area in Inner Mongolia (1.7%, 2.6%, 0.9% and 3.0%, respectively). The geographical variability in carbonate observed here is consistent with results presented by Feng et al. (2002).

In northern China, the $\delta^{13}C$ of soil carbonate (Fig. 2) varies with longitude, most notably decreasing from west to east. In the Kumtag Desert and Taklimakan Desert of northwestern China, $\delta^{13}C$ values are 2.15‰ and 0.51‰, respectively (Table 1). To the east of these sites, in the Badain Juran Desert and Hexi corridor, $\delta^{13}C$ values are −0.22‰ and −0.35‰, respectively. Farther to the east, surface soil samples from the Gurbantunggut

Table 1
Carbonate content and stable C–O isotopic composition of the surface soil samples

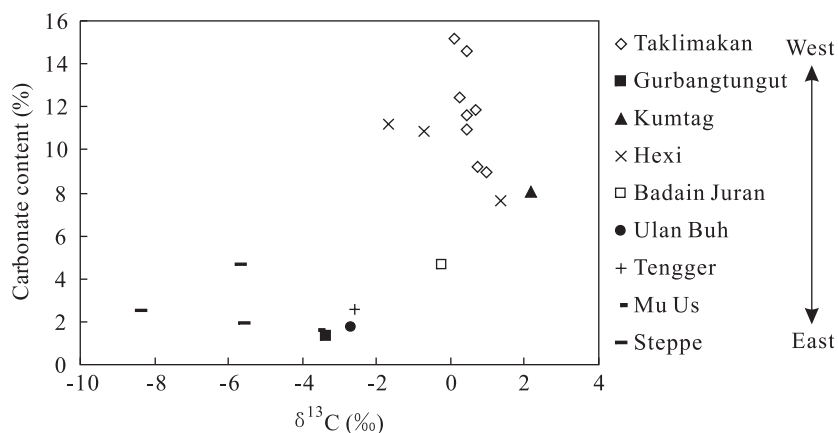| No.	Carbonate content (%)		$\delta^{13}C$ (‰)		$\delta^{18}O$ (‰)		Sampling area
	Sample	Average	Sample	Average	Sample	Average	
W17	1.3	1.3	−3.38	−3.38	−11.44	−11.44	Gurbantunggut Desert
XJ1	15.2	11.8	0.12	0.51	−7.95	−8.50	Taklimakan Desert
XJ5	11.0		0.45		−8.57		
XJ7	9.2		0.71		−8.51		
XJ9	8.9		0.96		−8.76		
XJ10	14.6		0.43		−8.56		
XJ11	11.6		0.46		−8.53		
XJ12	11.9		0.67		−8.79		
XJ14	12.4		0.24		−8.33		
W10	8.0	8.0	2.15	2.15	−10.08	−10.08	Kumtag Desert
W12	11.2	9.9	−1.68	−0.35	−8.21	−8.34	Hexi corridor
W9	7.7		1.36		−8.21		
W3	10.9		−0.72		−8.59		
W5	4.6	4.6	−0.22	−0.22	−9.29	−9.29	Badain Juran Desert
CF9	1.7	1.7	−2.69	−2.69	−10.09	−10.09	Ulan Buh Desert
CF14	2.6	2.6	−2.59	−2.59	−12.07	−12.07	Tengger Desert
CF1	0.7	0.9		−3.57		−10.09	Mu Us Desert
CF3	0.7						
CF5	1.6		−3.57		−10.09		
CF18	0.7						
FS4	2.5	3.0	−8.35	−6.53	−7.08	−8.83	Steppe area in mid-inner Mongolia
FS16	1.9		−5.58		−10.95		
FS21	4.7		−5.66		−8.47		
LS11	0.6	0.6					Onqin Daga sandy land
LS12	0.5						
LS15	0.3	0.3					Horqin sandy land
LS17	0.3						
TL1	0.4						

Fig. 2. Plot of the carbonate content vs. stable C isotopic composition of surface soil samples.

Desert, Ulan Buh Desert, Tengger Desert and Mu Us Desert have similar $\delta^{13}C$ values, ranging from $-3.57‰$ to $-2.59‰$. In the steppe area of mid-Inner Mongolia, the $\delta^{13}C$ value is $-6.53‰$. Due to low carbonate contents in the soil samples from the Onqin Daga sandy land and Horqin sandy land, the isotopic composition could not be determined.

The $\delta^{18}O$ values of surface soil samples from the western sites in Takalimakan Desert and Hexi Corridor are relatively high, with average values of $-8.50‰$ and $-8.34‰$ (Table 1). The $\delta^{18}O$ values in the Badain Juran, Kumtag, Ulan Buh and Mu Us Deserts are more negative at $-9.29‰$, $-10.08‰$, $-10.09‰$ and $-10.09‰$, respectively. The Gurbangtungut Desert and Tengger Desert have even more negative $\delta^{18}O$ values ($-11.44‰$ and $-12.07‰$) while in the steppe areas, $\delta^{18}O$ shows a considerable range from $-10.95‰$ to $-7.08‰$.

Analyses of three size-separated samples show that carbonate content of soils ranges from 4% to 17%, with values at the higher end of this range characteristic of the finer soil fractions (Fig. 3a). The $\delta^{13}C$ value for soils decreased with particle size from 80–63 to $<40\,\mu m$ (Fig. 3b) while $\delta^{18}O$ was inversely related to particle's size decreasing in the three soil samples analyzed (Fig. 3c). Dust transported over long distances usually has a mass median diameter considerably less than $10\,\mu m$ (Prospero, 1999), and so it must be noted that the large size cuts for the soil fractions analyzed limit the relevance of these data with respect to the long-range transport of aerosols. As methods for analyzing smaller quantities of material are developed, data for finer particles will become available.

3.2. Results on aerosols

The results of the carbonate content and isotopic analyses of aerosol samples are summarized in Table 2. These samples were all collected during dust storm events except sample TJ340, which was collected under moderate dust conditions.

The data for all aerosol samples from Aksu are plotted together in Fig. 4. All of the samples from this site have high percent carbonate contents ($\sim10\%$). The values for $\delta^{13}C$ during the three dust storms at Aksu vary from $-1.41‰$ to $-0.62‰$ while $\delta^{18}O$ ranges between $-8.01‰$ and $-7.49‰$. In addition, the low-dust storm sample TJ340 had a percent carbonate content and C and O isotope values similar to the dust storm samples.

The C and O isotope ratios in aerosol samples of Dunhuang varied considerably (Fig. 4). Compared with the Aksu samples, the samples collected during DDS3 and DDS4 have similar $\delta^{13}C$ values but higher $\delta^{18}O$ values. The $\delta^{13}C$ values of the other six Dunhuang samples are the highest of all samples analyzed, ranging from $0.04‰$ to $1.36‰$. These high values probably are

due to the high soil $\delta^{13}C$ value found for the Kumtag Desert which is where Dunhuang is located. The Dunhuang samples also show high carbonate content.

Except for one sample, the $\delta^{13}C$ and $\delta^{18}O$ values of the Yulin samples show a linear relationship (Fig. 4a). As discussed below, one possible explanation for the observed relationship is that the $\delta^{13}C$ and $\delta^{18}O$ values of the Yulin dust aerosols result from the mixing of two end members, one with low $\delta^{13}C$ and $\delta^{18}O$ values and the other with high $\delta^{13}C$ and $\delta^{18}O$ values. The carbonate contents of the samples vary considerably, from 2.6% to 12.1%, supporting the notion that the aerosol samples originated from different sources.

The aerosol samples collected at XiAn during three dust storm events have similar $\delta^{13}C$ values (average $-3.81‰$) but different $\delta^{18}O$ values ($-6.74‰$ to $-5.80‰$, Fig. 4a).

Although the carbonate contents of the aerosol samples are consistent with those of surface soil samples from the Taklimakan Desert and the Kumtag Desert, the aerosol samples have lower $\delta^{13}C$ values and higher $\delta^{18}O$ values than the soils (Fig. 4); this is probably due at least in part to the smaller particles in the aerosol samples. During dust storms in the deserts of northwestern China, the mass of the soil dust is dominated by particles $0.25–16\,\mu m$ in diameter accounted for $\sim69\%$ of the total mineral aerosol mass (Zhang et al., 2003a).

Although the soil data presented above do not cover particle sizes small enough to be considered representative of suspended dust, it is conceivable that the observed trends extend to smaller particle sizes even if they are not simple monotonic functions. If this should be the case, the smaller particle size distribution of the aerosol samples compared with the bulk soils may explain some of the discrepancy between the $\delta^{13}C$ and $\delta^{18}O$ values for soils and aerosols. This explanation is not entirely without support because a study of size-separated (i.e. >45, 2–45 and $<2\,\mu m$) near-surface loess samples collected from the Luochuan and Huanxian loess sections has shown that the C and O isotopes do indeed vary with particle size. More specifically, lower $\delta^{13}C$ values and higher $\delta^{18}O$ values were found in the finer fractions (Sheng et al., 2002), and these results are consistent with the proposed explanation for the differences in isotopic composition we observed for bulk soils and aerosols.

3.3. Carbonate content and C–O isotope systems in soils

The carbonate content of surface soils from northern China varies inversely with mean annual precipitation. Calcium carbonate rapidly precipitates during drought conditions, that is, when both soil moisture and root activity are low (Schlesinger, 1985), and more generally, $CaCO_3$ typically accumulates in soils where the mean annual precipitation is less than $1000\,mm\,yr^{-1}$ (Jenny,
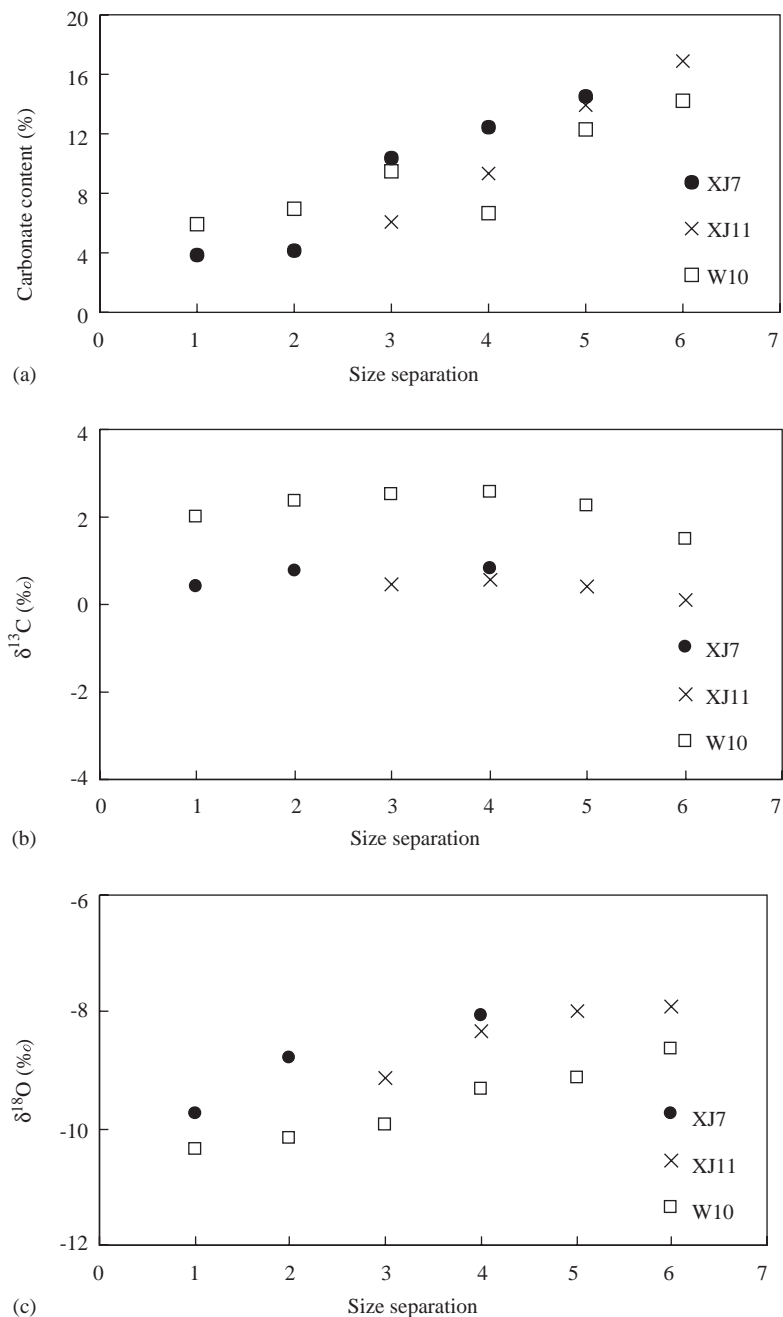
Fig. 3. The carbonate content (a), carbon isotope composition (b) and oxygen isotope composition (c) of different size soil samples. The numbers of size separation represent different size: 1, 250–125 μm; 2, 125–100 μm; 3, 100–80 μm; 4, 80–63 μm; 5, 63–40 μm and 6, <40 μm.

1980). The mean annual precipitation rates in the Chinese deserts are less than 400 mm in most cases, and precipitation tends to increase from west to east (Zhu et al., 1980). For example, the mean annual precipitation rates show the following increase from west to east: Taklimakan Desert <25 mm; inner Badain

Juran Desert, <50 mm; Alxa prefecture, 50–150 mm; and northeast sandy land, 250–400 m (Zhu et al., 1980).

Soil carbonate $\delta^{13}C$ varies due to the composition of the carbonate in the original mineral matrix combined with that produced by in situ weathering reactions. The Taklimakan Desert and surrounding areas were once

Table 2
Carbonate content and stable C–O composition of aerosol samples

Site	No.	Sampling date	Aerosol conc. (μg m^{-3})	Carbonate content (%)	δ^{13}C (‰)	δ^{18}O (‰)	Dust storm events
Aksu	TJ220	20-5-2001	3084.9	9.1	−0.86	−7.49	ADS1
	TJ221	21-5-2001	1539.9	12.2	−0.82	−7.82	ADS1
	TJ340	24-5-2001	631.0	10.1	−0.94	−7.27	
	TJ341	25-5-2001	1678.4	10.1	−0.62	−8.01	ADS2
	TJ426	31-5-2001	1092.1		−1.41	−7.91	ADS3
Dunhuang	TD296	5-5-2001	2308.1	9.3	0.71	−6.55	DDS1
	TD1237	8-4-2002	1084.2	9.5	1.36	−7.67	DDS2
	TD1238	9-4-2002	890.3	11.8	1.25	−7.99	DDS2
	TD1241	13-4-2002	1309.2	10.9	−0.76	−6.40	DDS3
	TD1247	19-4-2002	518.4	8.4	−0.91	−6.91	DDS4
	TD1652	29-4-2002	786.6	11.1	0.04	−6.48	DDS5
	TD1667	15-5-2002	912.4	7.4	0.32	−6.05	DDS6
	TD1668	16-5-2002	2177.6	12.8	0.70	−7.54	DDS6
Yulin	TY176	29-4-2001	2221.8		−3.02	−6.14	YDS1
	TY180	30-4-2001	1395.5		−0.47	−5.99	YDS1
	TY456	15-6-2001	863.6	6.2	−4.38	−7.20	YDS2
	TY543	9-2-2002	4397.3		−4.42	−7.10	YDS3
	TY546	14-3-2002	864.0		−4.33	−7.22	YDS4
	TY547	19-3-2002	11978.4		−3.50	−7.11	YDS5
	TY549	19-3-2002	4148.4	10.3	−2.41	−5.56	YDS5
	TY639	20-3-2002	2841.3	9.0	−2.43	−5.39	YDS5
	TY645	29-3-2002	1767.8	4.3	−3.09	−6.16	YDS6
	TY648	6-4-2002	874.7	4.2	−3.69	−6.79	YDS7
	TY650	7-4-2002		4.2	−3.86	−7.00	YDS7
	TY653	11-4-2002	787.3	2.6	−5.31	−8.75	YDS8
	TY654	14-4-2002	3446.8	4.0	−3.19	−6.37	YDS9
	TY658	15-4-2002	2279.8	3.8	−4.63	−7.44	YDS9
XiAn	TX1469	20-3-2002	1855.1		−3.78	−5.80	XDS1
	TX1501	22-4-2002	2494.9		−3.87	−6.74	XDS2
	TX2981	17-4-2003	2728.5		−3.78	−6.24	XDS3

covered by seawater before the Tertiary, and lake sediments deposited in this area after the seawater disappeared. As a result, the original carbonates in the northwestern Chinese deserts are mainly paleo-marine carbonate and lacustrine carbonate with the δ^{13}C values around 0‰ (Wen, 1989). There is little vegetation in the Taklimakan Desert due to its hyper-arid climate, and therefore the δ^{13}C value there is dominated by this original carbonate. Soil CO_2, which is involved in the formation of pedogenic carbonate, in turn is affected by vegetation, and in regions vegetated with C_3 and C_4 plants, the average soil δ^{13}C values of soil CO_2 are −22.2‰ and −8.5‰ (Cerling, 1984). The influences of the soil CO_2 in regions less arid than the Taklimakan and where there is more vegetative cover, such as Gurbantunggut Desert and Mu Us Desert, tend to drive δ^{13}C values toward more negative values, and this explains the observed east/west variability.

Compared with δ^{13}C, the spatial pattern in the oxygen isotope composition of pedogenic carbonate is consider-ably more complex, and the observed spatial distribu-tion of δ^{18}O is not easily explained due to the multiplicity of factors that can affect O isotopes. δ^{18}O in soil carbonates depends primarily on the isotopic composition of soil water, and this in turn is related to the proportions of ^{16}O and ^{18}O in local meteoric water (Cerling and Quade, 1993). The oxygen isotope ratios in precipitation are mainly affected by the source of the water and the degree of rainout (Rozanski et al., 1993), but it also has been shown that variations in δ^{18}O in recent precipitation is related to air temperature (Dansgaard, 1964; Rozanski et al., 1993).

The preliminary analysis of possible precursor materi-als for mineral aerosol presented above suggests that the combination of data for carbonate concentrations and stable C isotopes provides a new approach for investi-gating dust source areas complementary to the more
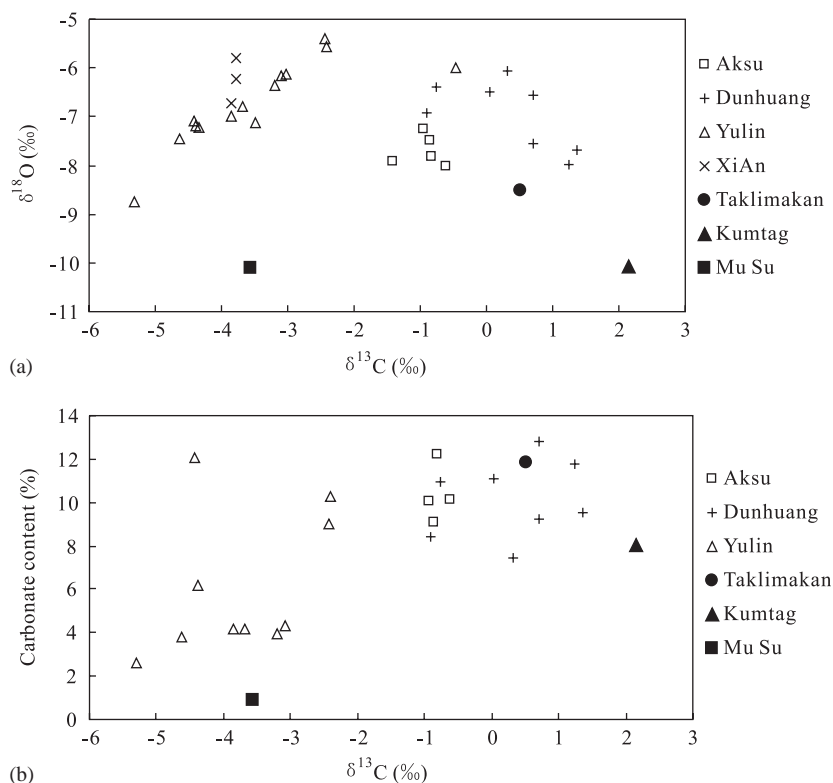
Fig. 4. Plots of the carbonate content and stable C–O isotopic composition of aerosol samples and the average values of soil samples in the Taklimakan, Kumutag and Mu Su Deserts: (a) Stable O isotopic composition vs. stable C isotopic composition; (b) Carbonate content vs. stable C isotopic composition.

established strategies for determining provenance. These studies also show that the data for the stable C isotopic composition of carbonate in soils is more likely to be a useful tool for discriminating among dust source regions than the stable O isotopic composition of soil carbonate.

3.4. Implication for tracing source of aerosols

The data we have generated for carbonate content and stable C–O isotopic composition of surface soils and aerosol samples described above, suggests that C isotopic composition may provide a useful index for tracing the sources of the dust aerosols while the carbonate content and O isotopic composition may be useful as auxiliary indices. To investigate the power of stable C isotopes as tracers, we calculated three-dimensional 3-day back trajectories (Fig. 5) for the dust events at four sampling sites using the NOAA HYS-PLIT4 trajectory model (Draxler and Hess, 1998).

Results from Aksu aerosol samples indicate that the Taklimakan Desert was the main source for the dust during all three dust storm events (Table 2). The back trajectories for the samples TD1241 and TD1247 from Dunhuang, passed through the eastern Taklimakan

Desert (Fig. 5a). The $\delta^{13}C$ values for those samples were −0.76‰ and −0.91‰, respectively (Table 2), and these were similar to the values of the northern Taklimakan as seen in Aksu aerosol samples. Thus, the isotopic data and the transport models both indicate that the dust in these two samples originated in the Taklimakan. The back trajectories of the other aerosol samples originated from other directions, and their $\delta^{13}C$ values were higher, suggesting stronger impacts of more regional sources on these aerosol samples.

As noted above, the $\delta^{13}C$ and $\delta^{18}O$ values of the aerosol samples from Yulin show a linear relationship except for sample TY180 (Fig. 4a). The stable C–O isotopic composition of local surface soils from the Mu Us Desert are −3.57‰ and −10.09‰. Among the aerosol samples, TY653 may represent the local aerosol with a $\delta^{13}C$ value of −5.31‰ and −8.75‰ for $\delta^{18}O$ because the short and meandering back trajectory for this sample suggests that the aerosol was mainly from local sources. Analyses of the air mass back trajectories shows that all air masses sampled at Yulin followed routes coming from the west or from the northwest (Fig. 5b). The dust sources along the route such as Kumtag, Taklimakan, Hexi Corridor and Badain Juran
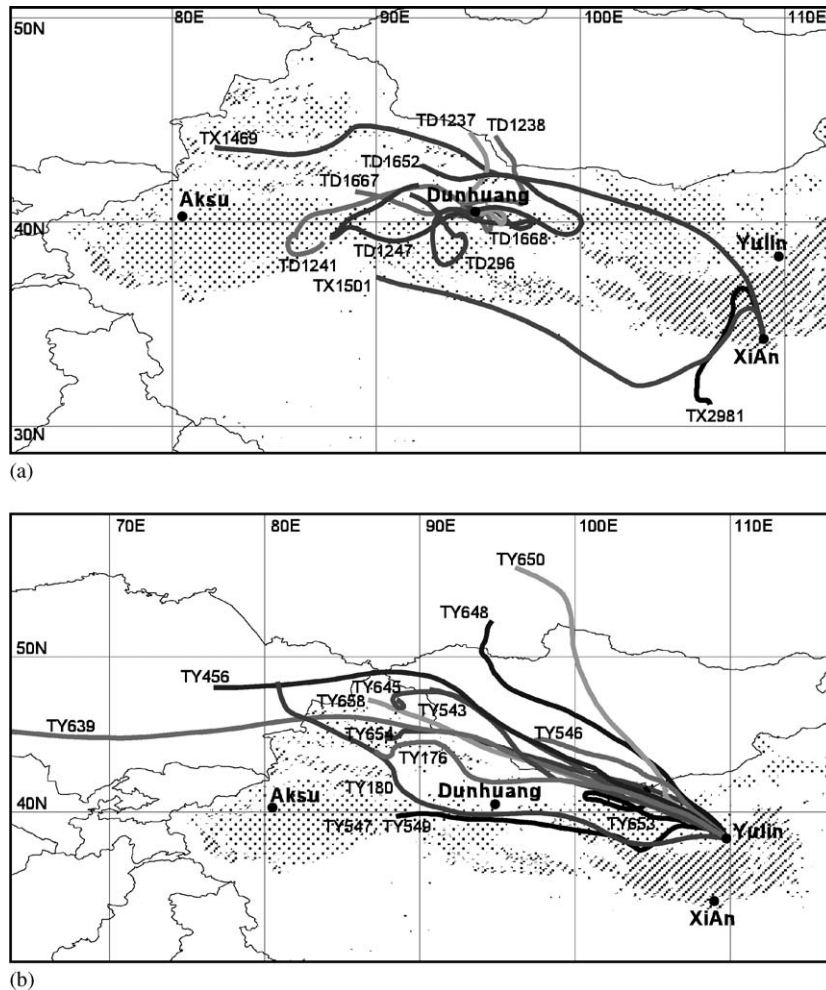
Fig. 5. 3-day back trajectories of the dust storm events associated with the aerosol samples in (a) Dunghuang and XiAn and (b) Yulin.

have high $\delta^{13}C$ values, which would result in moderate $\delta^{13}C$ value if mixed with the lower $\delta^{13}C$ of local dust. Some back trajectories to Yulin passed over the Gurbantunggut Desert, which has a relatively low $\delta^{13}C$ value, but in general this desert is not an important dust emission source (Zhang et al., 2003b). Although no data are presently available for southern and western deserts and semi-desert area of Mongolia, we could predict its $\delta^{13}C$ value would be higher than that at Yulin owing to the low annual precipitation and observed the moderate $\delta^{13}C$ value in the aerosol samples that had back trajectories that passed over this region.

From the two component Langmuir isotope mixing equation (Langmuir et al., 1978), the relative contributions of remote and local sources can be apportioned based on the $\delta^{13}C$ data as follows:

$$\delta A = X_R \delta_R + X_L \delta_L, \tag{2}$$

where δA denotes the $\delta^{13}C$ of ambient aerosol samples, δ_R denotes the $\delta^{13}C$ of remote source (we set $\delta^{13}C$ to $-0.15‰$ which is the average value of the aerosol samples at Aksu and Dunhuang), δ_L denotes the $\delta^{13}C$ of local source based on observations ($-5.31‰$), X_R and X_L are the relative percent contributions of the remote and local sources which sum to 100%.

Results of the mixing models indicate that remote sources accounted for 13–94% in ambient aerosol during the dust storm events at Yulin. The carbonate content of local aerosol is low, only 2.6%, but the carbonate concentrations were high in the two aerosol samples whose carbon isotopes apparently were strongly influenced by remote sources (samples TY549 and TY639). The isotope-mixing model indicates that remote sources contributed about 56% of the aerosol in these samples, and this is consistent with the high carbonate content due to impacts from remote sources.

Table 3
Carbonate content and stable C–O composition of aerosol and soil samples in XiAn

No.	Sampling date	Carbonate content (%)	$\delta^{13}C$ (‰)	$\delta^{18}O$ (‰)	Description
Q405	27-10-2003		−14.53	−11.80	Fugitive dust beside the road
Q406	27-10-2003		−13.00	−12.08	Fugitive dust beside the road
H-2	25-10-2003	3.0	−10.43	−11.36	Farmland soil in suburb of XiAn
CA-1	27-10-2003	5.6	−7.16	−6.88	Exposed soil beside the road
CA-2	27-10-2003	1.9	−7.05	−3.11	Soil dug from beside the road
CA-3	27-10-2003	6.1	−14.70	−12.33	Soil deposited beside the road
CA-4	27-10-2003	7.6	−14.27	−11.54	Soil deposited beside the road

Only three aerosol samples from XiAn were analyzed, and although they have similar $\delta^{13}C$ values, back trajectories indicate that the dust came from different sources (Fig. 5a). In XDS1, the observed $\delta^{13}C$ value may be the result from the mixing of dust from Gurban-tunggut Desert, Gobi area, the deserts north of the Loess Plateau and the Loess Plateau. The air masses sampled in XDS2 passed over the Taklimakan Desert (when the back trajectory extends to 4 days), then passed via the desert in Qaidam Basin and the Loess Plateau to XiAn. In contrast, XDS3 came from the area south of XiAn, passing over the loess Plateau before XiAn, and in this case, local material from Loess Plateau was the likely source for the dust. The $\delta^{13}C$ values of both soils and aerosol samples from XiAn are strongly negative (Table 3). The average $\delta^{13}C$ value of the two roadside aerosol samples was −13.77‰ while that for the exposed soil was about −7‰. In contrast, during the three dust storms, the average $\delta^{13}C$ value for the aerosol samples was −3.8‰, indicating significant contribution from a remote source with higher $\delta^{13}C$. Samples XDS1 and YDS5 were collected during the same dust storm (Table 2), but the $\delta^{13}C$ value of XDS1 is lower than YDS5, which shows the stronger impact from local sources.

4. Conclusions

The mean carbonate contents in soils from various arid and semiarid areas in northern China decrease systematically from west to east as the increasing trend in mean annual precipitation increases soil moisture and root activity. Of the sites from which soils were collected, the Taklimakan Desert in northwestern China had the highest carbonate content (11.8%) while the lowest value, 0.3%, was found at the Horqin sandy land in northeastern China. The carbonate content of the soil samples also varies inversely with grain size.

Combining information on stable C isotopic composition with carbonate concentrations, provides a new approach for distinguishing among the dust source areas

that complements other methods for investigating provenance. The stable C isotopic composition of soil carbonate, measured as $\delta^{13}C$, also decreases from west to east in northern China, and arid soils in various areas of northern China have distinctly different $\delta^{13}C$ value. The stable C isotopic composition of carbonate is a more useful indicator than stable O isotopes for differentiating among dust source areas. In surface soil, the $\delta^{13}C$ value decreases with particle sizes from 80–63 to $<40\,\mu m$, and the $\delta^{18}O$ value increases slightly in smaller particles.

The stable isotopic compositions of the aerosol samples at Aksu reflect the characteristics of the Taklimakan dust source, with the $\delta^{13}C$ value ranging from −1.41‰ to −0.62‰ and $\delta^{18}O$ from 8.01‰ to −7.27‰. The particle size distribution of the mineral dust aerosol is much smaller than bulk surface soil, and this is a possible explanation for why the dust aerosol has lower $\delta^{13}C$ value and higher $\delta^{18}O$ value compared with bulk soils. Further studies are needed to evaluate the variations in $\delta^{13}C$ with particle size because this important issue relates directly to the evolution of the chemical and isotopic signals during transport.

The high average $\delta^{13}C$ value (−0.15‰) and high carbonate content of the aerosol samples at Aksu and Dunhuang reflects the characteristics of northwestern deserts. The $\delta^{13}C$ values in aerosol samples from Dunhuang are slightly higher than in those from Aksu except for two samples, which showed clear evidence for transport from the Taklimakan and had $\delta^{13}C$ values similar to Aksu. The aerosol samples collected at Yulin also showed evidence of material that had been transported from the northwestern deserts. Similarly, the average $\delta^{13}C$ value for the two roadside aerosol samples from XiAn was more negative than that determined for aerosol samples collected during three dust storms; this is evidence for a significant contribution from northwest deserts.

The C isotopic composition of carbonate is a useful index to trace the sources of the dust aerosols, especially when used in combination with data for carbonate contents and back trajectory analysis. The O isotopic

composition provides supplemental information, but the spatial pattern seen for soil O isotopes is considerably more complex. The stable C and O isotopic tracers of dust composition provide another means for evaluating the provenance of desert dust collected far from the source or sources.

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