Remote mineral aerosols in Westerlies and their contributions to the Chinese loess*

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Abstract The concentrations of 28 trace and rare earth elements in the aerosol particle samples were determined for a site at 4800 m above the sea level on the Qinghai-Xizang Plateau. The mass of the particulate materials in the mid-troposphere atmosphere over this site is dominated by local dust particles (70%) and remote ones in Westerlies (25%). On the basis of the main dust-derived elements (Al, Fe, Mg and Sc) and corresponding data from the Chinese deserts, an elemental tracer system has been established to proportion the dust input to the loess deposited on the center of the Loess Plateau during the last glacial cycle, which suggests that the contribution of the remote dust in Westerlies is much smaller than that of the Chinese deserts.

Keywords: atmospheric trace elements, elemental tracer, remote dust in Westerlies, loess accumulation.

Although there has been a long history of detailed investigations in paleoclimate from the Chinese loess and paleosol sequences[1, 2], sources of the loess have not been positively understood so far. Mainly supplied by the northerly Asian winter monsoon winds, the mineral dust from the Gobi and desert regions in northern and northwestern China is widely assumed to have major contributions to the loess deposited on the Loess Plateau[3—6]. The upper-level westerly winds are also considered to have possibilities for contributions of the remote sources dust to the loess. Unfortunately, the eolian dust from the monsoon winds cannot be distinguished physically from the fallout dust from the upper Westerlies, because the fallout is displaced within the low-level northerly monsoon in final transport phase.

On the basis of the elemental concentrations of the aerosol particles collected from the Qinghai-Xizang Plateau, the objectives we try to investigate here are: i) to investigate the characterization of atmospheric elements; ii) to extract the chemical signature of remote dust in Westerlies; iii) to establish an elemental tracer system for desert dust particles and remote ones associated with the data from the Chinese desert dust[7]; and iv) to proportion the dust input to the eolian loess in the center of the Loess Plateau during the last glacial cycle.

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1 Materials and methods

Forty-nine bulk aerosol samples were collected at Udaoliang (UDL, 35.2° N, 93.1° E) from 1993-09 to 1994-05. The sampling site is located on the Qinghai-Xizang Plateau about 4800 m above the sea level between the mountains of Kunlun and Tanggula. High volume GMW-2310-type sampler (Miami University, USA) was used for 10-m height ground-based sampling. The sampling intervals ranged from 5 to 24 h with the flow rate about 14 $m^3 \cdot h^{-1}$.

The aerosol samples collected on Whatman 41® filters were digested and prepared into 1% HNO_3 acid solution for elemental measuring using the inductively coupled plasma mass spectrometry (ICP-MS) at State Key Laboratory of Loess and Quaternary Geology, Chinese Academy of Sciences. With this method, we were able to determine the concentrations of 28 elements: Al, Ce, Co, Cs, Dy, Er, Eu, Fe, Gd, Hf, Ho, La, Lu, Mg, Mn, Na, Nb, Nd, Pr, Rb, Sc, Sm, Sr, Tb, Th, Ti, Tm and Yb. The concentrations were corrected by filter backgrounds, and measured by ICP-MS in 40 aliquots for two standard reference materials (GSS-2 and GSS-6) from the National Bureau of Chemical Exploration Analysis, China[8]. The quality control and quality assurance were all satisfactory for all the elements with coefficient variances of <10% and <20%, respectively.

2 Results and discussion

2.1 The chemical composition of aerosol particles

The concentrations of 28 elements in the aerosol samples from UDL are summarized in table 1. These include 7 major elements of mineral dust (Al, Fe, Mg, Na, Ti and Sc), and 14 rare earth elements (REEs). The mean concentration of Al (6 300 ng · m^{-3}) was about 60% of the annual mean of it (11 000 ng · m^{-3}) in the atmosphere over the southern margin of the Loess Plateau during the nondust storm conditions (NDSCs)[9], just equivalent to about 29% of the mean concentration of Al (22 000 ng · m^{-3}) for the 12 Chinese desert sites during the high dust season in the NDSCs[9]; and also equivalent to ~ 80 μg · m^{-3} of the mineral aerosols because mineral aerosol is approximately 8% Al in weight[10]. If we take the contributions of dust storm into account, the percentage of UDL Al relative to that over the loess and the desert regions will be lower than that mentioned above. The sum of concentrations of the 7 dust elements amounts to about 82% of the total mass attributable to all 28 elements analyzed, which suggest that these dust elements may play an important role in investigation of the mineral aerosol.

2.2 Sources of trace elements

The concentrations for 4 factors resolved by absolute principal component analysis (APCA) of the 28 elements in the 49 UDL aerosol samples are also given in table 1.

Table 1 Observed and APCA estimated concentrations[a] of atmospheric trace elements over UDL

	Concentration/ng · m^{-3} (1993-09—1994-05, n=49)												Commu-nality
	observed		sum of estimated contributions	mean percentage of total estimated concentration									
	range	mean		Factor 1		Factor 2		Factor 3		Factor 4			
Al	730—17200	6300	6200	1900	(30)	380	(6.1)	3900	(63)	81	(1.3)		0.95
Ce	0.03—45	11	10	6.9	(66)	1.2	(11)	2.3	(22)	0.05	(0.46)		0.96
Co	0.05—15	3.8	3.3	1.9	(56)	0.39	(12)	1.0	(32)	0.02	(0.66)		0.80
Cs	0.13—6.1	1.4	1.4	0.86	(61)	0.12	(8.8)	0.43	(30)	0.01	(0.63)		0.95
Dy	0.05—1.7	0.64	0.39	0.15	(38)	0.20	(52)	0.04	(10)	0.00	(0.20)		0.70
Er	0.01—3.3	0.51	0.47	0.04	(9.4)	0.35	(74)	0.08	(16)	0.00	(0.34)		0.93
Eu	0.00—0.65	0.23	0.08	0.07	(81)	0.01	(10)	–	(–)	–	(–)		0.62
Fe	620—10000	4100	3400	990	(29)	240	(7.1)	2100	(62)	43	(1.3)		0.88
Gd	0.01—3.1	0.73	0.66	0.20	(30)	0.34	(52)	0.11	(17)	0.00	(0.36)		0.87
Hf	0.02—2.4	0.64	0.39	0.14	(37)	0.31	(80)	–	(–)	–	(–)		0.91
Ho	0.02—1.0	0.33	0.15	0.12	(80)	0.06	(40)	–	(–)	–	(–)		0.89
La	0.50—20	5.4	4.8	3.2	(67)	0.44	(9.0)	1.1	(24)	0.02	(0.49)		0.96
Lu	0.01—1.4	0.17	0.15	–	(–)	0.14	(99)	0.00	(2.0)	0.00	(0.04)		0.92
Mg	170—4600	1600	1500	340	(23)	56	(3.7)	1000	(67)	22	(1.4)		0.94
Mn	7.9—200	66	60	19	(32)	8.1	(13)	32	(54)	0.67	91.1)		0.89
Na	73—7400	2500	2200	[illegible]		–	(–)	2300	(100)	47	(2.2)		0.72
Nb	0.15—6.8	1.7	1.7	0.89	(53)	0.24	(14)	0.54	(32)	0.01	(0.67)		0.93
Nd	0.53—19	5.0	13	2.7	(21)	1.1	(8.3)	8.9	(69)	0.19	(1.4)		0.93
Pr	0.13—3.9	1.1	0.96	0.62	(64)	0.16	(17)	0.18	(19)	0.00	(0.39)		0.96
Rb	0.29—130	25	10	1.6	(15)	3.5	(33)	5.2	(50)	0.11	(1.04)		0.91
Sc	0.10—10	2.1	0.25	0.07	(28)	0.14	(56)	0.04	(16)	0.00	(0.33)		0.80
Sm	0.01—4.4	1.1	1.0	0.29	(28)	0.49	(47)	0.26	(25)	0.01	(0.51)		0.86
Sr	2.7—570	88	96	62	(65)	16	(16)	18	(19)	0.37	(0.39)		0.84
Tb	0.00—1.2	0.15	0.16	0.02	(13)	0.12	(73)	0.02	(13)	0.00	(0.27)		0.89
Th	0.15—6.6	1.6	1.5	0.99	(67)	0.24	(16)	0.25	(17)	0.01	(0.35)		0.88
Ti	38—960	310	280	89	(32)	10	(3.6)	180	(64)	3.7	(1.34)		0.92
Tm	0.00—1.4	0.14	0.14	0.01	(6.4)	0.14	(99)	–	(–)	–	(–)		0.96
Yb	0.02—5.8	0.66	0.72	0.05	(7.1)	0.62	(86)	0.05	(6.8)	0.00	(0.14)		0.93

a) Total matrix sampling adequacy: 0.85.

The procedure of the APCA was described in details in refs. [9, 11]. The accuracy of the data, the percentage of each element as well as the agreement between the sum of contributions derived from the APCa and measured concentrations all indicate good fits of elements to the factors. In most cases, only small amount of the variance in the trace element concentrations were left unexplained (table 1).

Factor 1 (F1) was highly loaded with REEs and trace elements (Ce, Cs, La, Ho, Nd, Pr, Sr and Th); 7 dust elements also had some loading in this factor. Factor 2 (F2) was dominated by other REEs and trace elements (Dy, Er, Gd, Hf, Lu, Sc, Sm, Tb, Tm and Yb). Factor 3 (F3) was mainly loaded with major dust elements (Al, Fe, Mg, Mn, Na and Ti). There was no higher loading for all the elements in Factor 4 (F4). The UDL

particles appear to be mainly composed of mineral dust aerosols, because the elemental concentrations in the bulk sample resemble the pattern of the concentrations for average crustal rock[10]. This is demonstrated by calculation of enrichment factors relative to average crustal rock ($EF_{crust}=(X/Al)_{air}/(X/Al)_{crust}$) in fig. 1. For the concentrations in each factor, the EF_{crust} values of the highly loaded elements (HLEs) in F1 were generally lower than 5, except for Cs, suggesting that the particles in F1 were crust-derived (fig. 1). The concentrations of HLEs in F2 were enriched by factors 10 to 80, suggesting non-crustal enrichments (fig. 1). The EF_{crust} for HLEs in F3 were all smaller than 5, except for Nd, also reflecting the existence of the mineral dust (fig. 1). The F4 particles were quite similar to F3. Because the sum of elemental concentrations in F4 did not exceed 3% of that in all the factors (fig. 1, table 1), one can neglect the contributions of F4 particles to the bulk ones. The 7 dust elemental concentrations were all similar to the average crustal rock for all the factors as well as bulk samples, exhibiting that the chemical differences of the aerosol particles emitted from various sources were significant only for the REEs or other trace elements at the site.

For further identification of the sources of the aerosol particles, the REE distribution models of all the factors are given in fig. 2. The REE model for F1 resembles most to that of the average crustal rock[10], suggesting that remote dust existing in F_1 has characteristics of the large-scale and remote sources (fig. 2). Comparing F1, F3 and F4 shows relatively large difference from crustal rock, reflecting that the dust in two factors

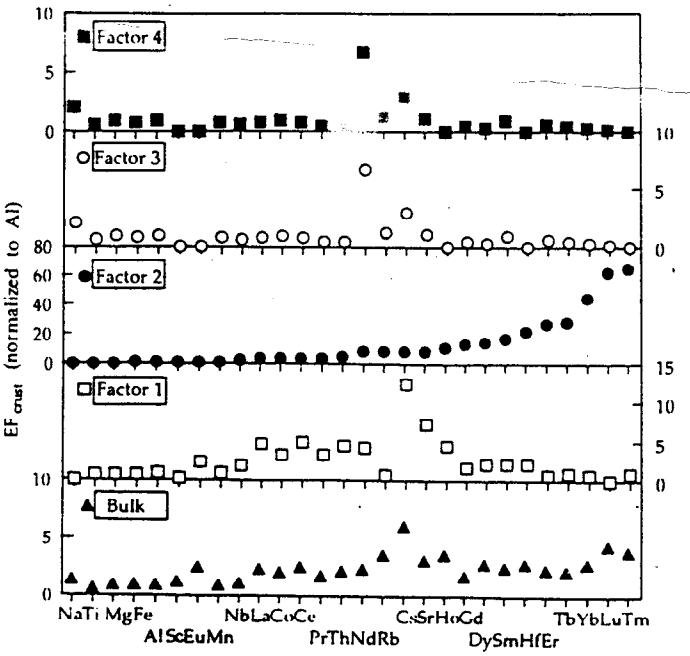

Fig. 1. The enrichment factors relative to average crustal rock of 28 elemental concentrations in each factors and bulk aerosol samples at UDL.

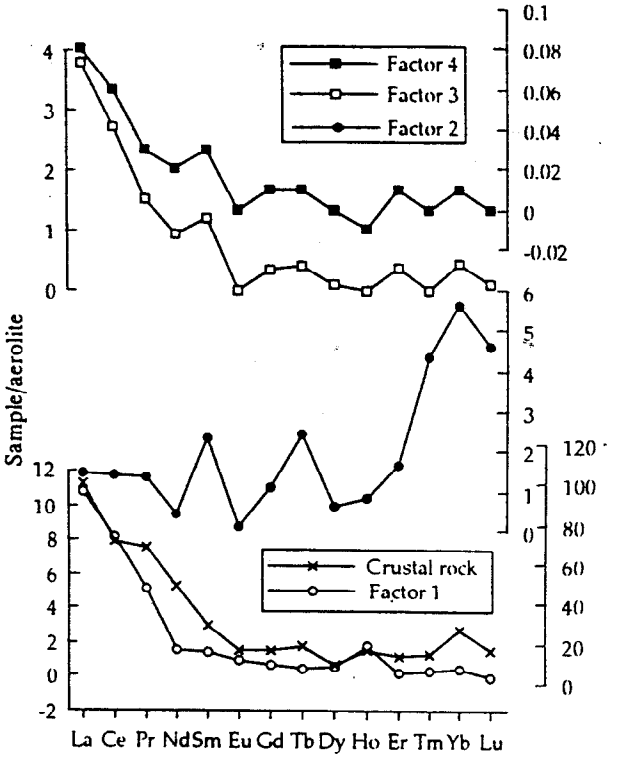

Fig. 2. REE distribution models of all the factors and bulk aerosol samples at UDL.

appear to be representative of the particles from the local sources on the Qinghai-Xizang Platear (fig. 2). The model for F2 shows bigger difference from the crustal rock. One can consider that the particles in F2 are dominated by the non-crustal sources no matter by the EF_{crust} calculation or the REE model (figs. 1, 2).

The correlations of each factor score with the sum of 28 elemental concentrations demonstrate that the dust particles in F3 ($r^2=0.58$) and F1 ($r^2=0.38$) are the major components of the aerosol particles at UDL, and that the particles represented by F2 and F4 give minor contributions to the aerosols. We further calculated the percentage of the sum of all the elemental concentrations in each factor relative to that of the bulk samples, the average of each factor for 49 samples suggests that the remote dust particles in Westerlies (F1) were about 25% of the total aerosol particles, and local dust particles from the plateau (F3 plus F4) account for about 70% of the total. Only about 5% of the particles might be attributable to the noncrustal aerosol particles (F2).

2.3 A four-element tracer system for dust aerosol

Atmospheric mineral dust is composed of various elements; no special elements are unique to specific source regions. Some elements may vary with the source regions because different regions have different mixes of the minerals. The two keys to derive the source signatures for tracing the origins of elements are finding the right elements and handling the data with the appropriate statistical techniques. Constructing signatures from too many available elements will add too much noise. The best approach seems to limit the signatures to a few elements with the greatest tracer power.

Several requirements should be met by elements and signatures before they can be used in a source tracer system, namely, the elements should be crust-derived, evenly sampled and accurately measured, stably and homogeneously emitted in each region; and each signature should be distinguished and remain recognizable during transport.

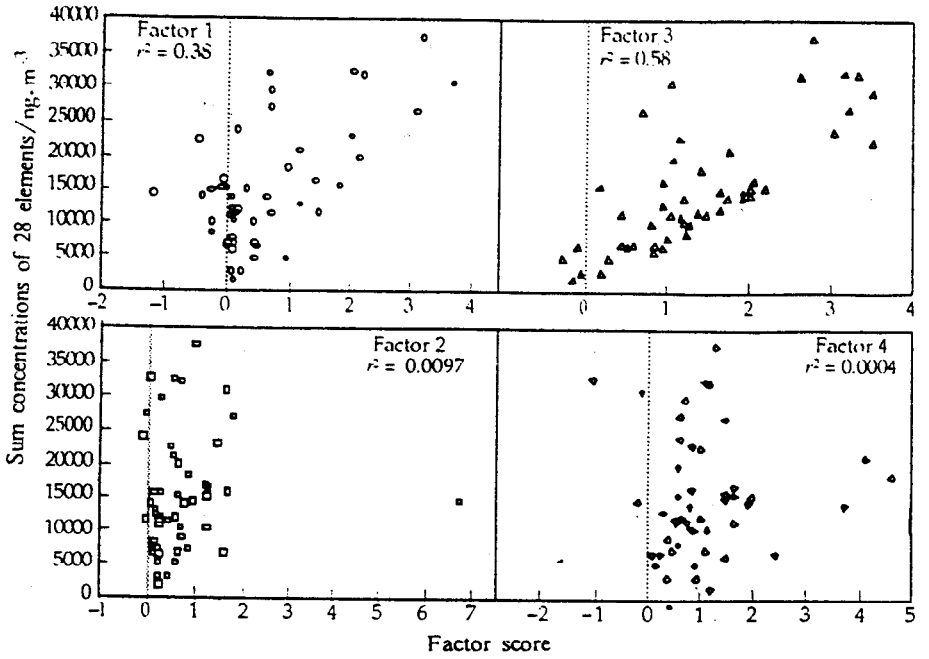

Fig. 3. Relationships between the sum of 28 elemental concentrations in aerosol samples at UDL and factor scores for each factor.

Based on the data of the Chinese deserts[7], we are planning to use only 4 elements (Al, Fe, Mg and Sc) to establish a tracer system. The 4 tracer elements have proved to be crust-derived by the enrichment factor analysis (fig. 1), and to have relatively lower uncertainties for determination with ICP-MS. Because of the absence of the corresponding data and the existence of the higher measuring uncertainties for Na, Ti, and Mn in the data set of the loess, we did not choose them in our tracer system even though these elements are fairly good tracer elements as well. Being of low concentrations, low tracing power and relatively higher measuring uncertainties, the other crust-derived elements, Eu, La, Co, Ce, Pr, Th, Nd and Rb, were not included into the system.

Our source signatures consist of 4 elemental ratios to Al. The ratios are used to normalize for the various effects such as the strength of source dust emission in different time, dispersion and removal during the transport[13]. Aluminum is used in the denominator because it is a general dust found at similar concentrations in diverse source regions and hence will not bias the rations toward any particular regions. To resolve the mixed dust into source components, the chemical element balance (CEB) procedure[14, 15] is used. This approach is essentially a weighted multiple-linear regression with the elements in the sample as dependent variable and the elements in the signatures as independent variables:

$$C_i = \sum_{j=1}^{p} X_{ij} S_j, \quad (1)$$

where C_i is the concentration of the ith element in an ambient sample scaled to Al, X_{ij} is the corresponding data of the ith element in the jth source signature, S_j is the derived strength or "source coefficient" of the jth source signature, p is the assumed number of source signatures.

Table 2 Source signatures for Chinese dust

	X/Al		
	NW deserts ($n=32$)	N deserts ($n=58$)	Remote dust in Westerlies ($n=49$)
Al	1.0	1.0	1.0
Fe	0.83	0.52	0.24
Mg	0.19	0.24	0.13
Sc ($\times 10^{-4}$)	2.7	3.4	1.4

Geometric mean Al ($ng \cdot m^{-3}$): 23000 for NW deserts, 19000 for N deserts, 370 (mode) for remote dust in Westerlies.

Table 3 Results of the Fisher's Protected Least Significant Difference for source signatures among the major sources of Chinese dust[a]

Source	Mean difference	p-value
NW deserts vs. remote dust in Westerlies	0.16	0.03
N deserts vs. remote dust in Westerlies	0.10	0.003
NW deserts vs. N deserts	0.06	0.003

a) Significance level: 5%.

The source signatures of Chinese dust derived from the 4 crustal elements are given in table 2. The signature of Chinese northwestern desert includes data for Taklimakan desert and Kumutage desert; the signature of Chinese northern deserts were derived from the data over the deserts in the northern Inner Mongolia and northwestern margin of the Loess Plateau[7].

To eliminate the effects of dust transport among the desert sites, the signature for individual desert was calculated on the basis of mode concentration of frequency distribution, and the source signatures were the geometric mean of the individual ones[7]. The signature of the remote dust in Westerlies was also derived from mode value to further erase the influence of the dust transport.

A critical assumption of least-squares apportionment is that signatures are statistically independent. The more similar the two or more signatures are, the less reliable when they are used to apportion elemental concentrations. Through a testing with the Fisher's Protected Least Significant Difference, one can see that the significant differences for the signatures do exist among the 3 dust sources (table 3).

Due to the large-scale sources and good mixing during transport of the remote dust in Westerlies, the source signature of the dust based on the elemental ratios is less influenced

by the source change and can be considered generally consistent during the last glacial cycle. The signatures for desert sources are also assumed to be stable since the last 130000 a B.P., because the general structure of modern Chinese desert was formed before that time[16]. The desert dust has not been affected by any re-working processes according to the fact that no greater than 10% of the post-depositional loess in the Loess Plateau is attributable to the re-working processes even during the interglaciation[12].

The results of CEB analysis between the source signatures and the data of loess-paleosol in the center of the Loess Plateau during the last glacial cycle are shown in table 4. The data in the different layers are derived from the average concentration of the individual samples with a 10 cm interval (about the duration of 1000 a)[12]. Although the contributions of each source differ in different elements, the average contributions to the 4 tracer elements from the three source regions suggest that the northern desert source dominated the dust input during the Holocene (S0), and in the L1-LL1 the contributions from the northwestern and northern deserts are about 46% and 34%, respectively. During the stage of L1-SS1 and L1-LL2 the dust depositions on the Loess Plateau were considerably

Table 4 Contributions of the three sources to crustal elements in eolian loess samples in the center of the Loess Plateau during the last glacial cycle

Source contribution	Source coefficient	Al	Fg	Mg	Sc	Mean
Luochuan, Heimugou S0						
% NW desertes	0.01	1.0	1.7	0.71	1.5	1.2
% N deserts	0.91	91	96	82	100	92
% Remote dust in Westerlies	0.08	8.0	3.8	3.8	0.60	4.0
% Others						2.6
L1-LL1						
% NW deserts	0.49	49	56	28	49	46
% N deserts	0.47	47	34	34	59	43
% Remote dust in Westerlies	0.04	4.0	1.3	1.5	0.21	1.8
% Others						9.3
L1-SS1						
% NW deserts	0.37	37	45	22	40	36
% N deserts	0.60	60	46	44	82	58
% Remote dust in Westerlies	0.03	3.0	1.0	1.1	0.17	1.3
% Others						4.8
L1-LL2						
% NW deserts	0.39	39	47	22	41	37
% N deserts	0.57	57	43	40	75	54
% Remote dust in Westerlies	0.04	4.0	1.4	1.5	0.22	1.8
% Others						7.7
S1						
% NW deserts	0.97	97	91	60	79	82
% N deserts						
% Remote dust in Westerlies						
% Others						18

attributable to the northern desert source, 58% and 54%, respectively. In contrast, the dust mainly derived from the northwestern desert source (82%) dominated the input to the loess during the last interglaciation. It is most important that the dust input attributable to the remote dust in Westerlies did not exceed 4% of the total mass during the different stages spanning the last glacial cycle. This appears to suggest that the major sources for eolian dust on the Loess Plateau are the deserts in North and northwest China.

The contributions of the northwestern deserts to Mg are generally smaller than that to other elements (table 4), and the contributions of the northern deserts to Sc are normally larger than that to other elements. This may reflect that there are also other sources affecting the concentrations of the two elements, or that the two are not the best tracer elements. Although we could not use the other potential tracer elements such as Si, Ti, Na and Mn because they are not commonly obtained for desert dust, remote dust and eolian loess with the different measuring methods, the remote dust obtained in Westerlies is much affected by local dust and non-crustal aerosols, the results derived from our preliminary tracer system still give us some valuable information.

3 Conclusions

The investigations of the multi-elements in the aerosol particles on the Qinghai-Xizang Plateau demonstrate that the collected aerosol particles are mainly composed of local and remote dust, and the contributions of non-crustal dust are not much.

An elemental tracer system for dust particles has been established by combining the data from the remote dust in Westerlies with the Chinese desert dust, which suggests that the dust input derived from the remote dust during the different stages since the last glacial cycle was much lower than that from the desert sources. The climatic information with a global significance recorded in the loess-paleosol sequence is probably not easily observed in the mass exchange from the remote dust in Westerlies.

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