

## Regional Sr–Nd isotopic ratios of soil minerals in northern China as Asian dust fingerprints

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### Abstract

We report that arid soils in various areas of northern China can be distinguished by using Sr–Nd isotopic ratios of acid-resistant minerals and Sr isotopic ratios of water- and weak-acid-soluble minerals. Our results show that contemporary dust falling on Beijing is transported mainly from the adjacent northwestern to western areas and is more likely to be related to desertification than dust from the remote Takla Makan desert, the southwestern Gobi desert, or the Loess Plateau. Mineral isotope fingerprinting of arid soils is a powerful tool for source identification and impact assessment of mineral dust, and can serve as a desertification index.

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### 1. Introduction

Mineral dust from the desert and loess areas of northern China and southern Mongolia (Fig. 1) affects climate, human health, and biological activity over the Northern Hemisphere on an event to geological time-scale (Duce, 1995; Chadwick et al., 1999; Wilkening et al., 2000). These arid areas have become desertified by both natural processes or human activity during the course of Chinese history. At present, desertification is accelerating and extending even farther north as a result of socio-economic pressures induced by population growth (Sheehy, 1992; Zhu et al., 1992; Zhang et al., 2001). Several studies have reported that Asian dust events, which occur throughout the year but predominantly in spring, have become more frequent in the late

20th century, particularly since 1999–2000 (Chun et al., 2001; Sun et al., 2002; Gao et al., 2003; Kurosaki and Mikami, 2003), but the relationship between desertification and recent dust events remains unresolved.

Recent increases in the amount of dust have caused serious problems, particularly in areas which are near arid regions as Beijing (Sun et al., 2002; Guo and Jiang, 2002). However, despite modeling and climatological studies (Sun et al., 2000; Husar et al., 2001; Xuan and Sokolik, 2002; Chung et al., 2003), the identification of major Asian dust sources at downwind depositional sites across the northern Pacific region by event-scale research only is difficult because the geochemical characterization of source areas is insufficient. Such identification is important not only for paleoclimate reconstruction over geological time (Biscaye et al., 1997; Asahara, 1999; Bory et al., 2002) but also for accurate environmental assessment and the planning of possible countermeasures to dust events.

Stable isotopes of Sr and Nd have potential as source-area fingerprints because minerals and rocks have

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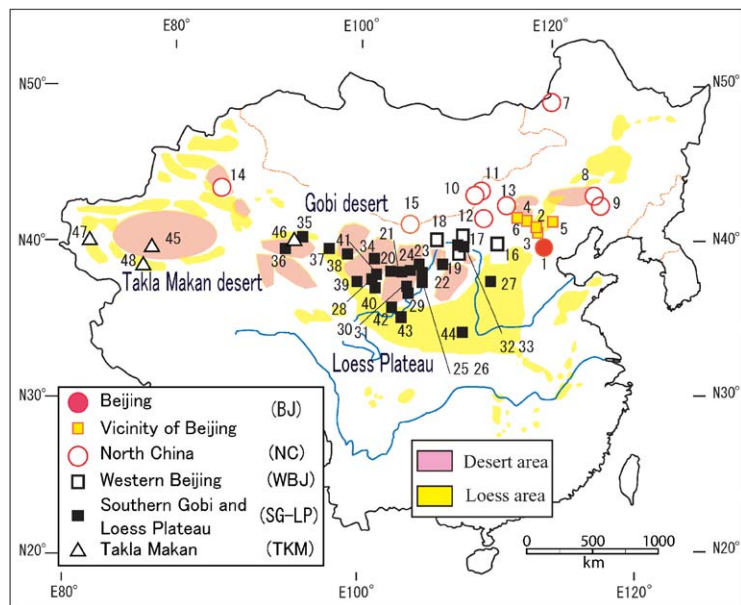


Fig. 1. Sampling points of arid and semi-arid soils in China.

distinct  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios, depending on their geological derivation. Also, these isotope ratios are less altered than elemental composition during transport in the atmosphere or after deposition as sediments (Biscaye et al., 1997; Asahara, 1999; Chadwick et al., 1999; Bory et al., 2002; Grousset et al., 2003). Desert sand and loess in China are composed of a mixture of salinization minerals (formed by evaporation of soil water) and other minerals derived mainly from provenance rocks and their weathered materials; the former consist of water-soluble minerals (i.e., halite, gypsum, and anhydrite) and acetic acid (HOAc)-soluble minerals (mainly calcite), whereas the latter consist of acid-resistant minerals, especially silicates (Zhang et al., 1990). The salinization minerals of several arid soils in northern China have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios distinct from those of acid-resistant minerals (Yokoo and Nakano, 2001; Yokoo et al., 2004), and are found dissolved in spring rain in Japan (Nakano and Tanaka, 1997). In order to provide key constraints on the source-areas of Asian dust and to identify the major dust source on Beijing, we collected surface soil samples at 35 locations from the desert and loess areas of northern China and five of wind-blown origin in Beijing (Fig. 1), and determined their Sr–Nd isotopic ratios of salinization and acid-resistant minerals.

## 2. Samples and experiment

Samples were sequentially extracted with ultrapure water and 5% HOAc solution following the analytical

procedure of Yokoo et al. (2004). Bulk samples and residue splits after extraction with HOAc were digested with a  $\text{HF-HClO}_4\text{-HNO}_3$  solution. The Sr and Nd isotope ratios were determined with a Finnigan MAT262 mass spectrometer, at the Institute of Geoscience at the University of Tsukuba, Japan, following the analytical procedure of Na et al. (1995). The measured  $^{87}\text{Sr}/^{86}\text{Sr}$  values were normalized to an  $^{86}\text{Sr}/^{88}\text{Sr}$  value of 0.1194, and those of  $^{143}\text{Nd}/^{144}\text{Nd}$  to a  $^{146}\text{Nd}/^{144}\text{Nd}$  ratio of 0.7219. The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of NIST.SRM987 and the  $^{143}\text{Nd}/^{144}\text{Nd}$  ratio of the La Jolla standard determined during this study were  $0.710248 \pm 0.000021$  ( $2\sigma_{\text{mean}}$ ,  $n = 15$ ) and  $0.511845 \pm 0.000009$  ( $2\sigma_{\text{mean}}$ ,  $n = 12$ ), respectively. All  $^{87}\text{Sr}/^{86}\text{Sr}$  data were normalized to the NIST.SRM987 value of 0.710250. Analytical precisions for samples were better than  $\pm 0.000013$  ( $2\sigma_{\text{mean}}$ ) for  $^{87}\text{Sr}/^{88}\text{Sr}$  and  $\pm 0.000010$  ( $2\sigma_{\text{mean}}$ ) for  $^{143}\text{Nd}/^{144}\text{Nd}$ . Analytical results are given in Table 1.

## 3. Results and discussion

### 3.1. Regional variation of Sr–Nd isotopic ratios of minerals

Fig. 2 shows the compilation of the present and previously reported data on the HOAc-resistant residues of soils from 48 locations in northern China, demonstrating a wide variation in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.71105–0.73658) and  $\epsilon_{\text{Nd}}$  values (–28.7 to –4.0). This variation is far larger than has been reported for bulk soils (Liu

Table 1

Sr isotopic ratios of H<sub>2</sub>O- and HOAc-soluble minerals and Sr–Nd isotopic ratios of HOAc-insoluble minerals in the loess and desert sand in northern China

No.	Location	Latitude	longitude	Soil type	H <sub>2</sub> O-leachate	HOAc-leachate	HOAc-residue		
					<sup>87</sup> Sr/ <sup>86</sup> Sr	<sup>87</sup> Sr/ <sup>86</sup> Sr	<sup>87</sup> Sr/ <sup>86</sup> Sr	<sup>143</sup> Nd/ <sup>144</sup> Nd	ε <sub>Nd</sub>
<i>Beijing and Beijing vicinity area (BJ)</i>									
1	Yuhulnanlu, Beijing	39.60	116.30	loess	0.709847	0.709964	0.713431	0.512037	−11.73
	Xishan, Beijing	39.60	116.30	loess	0.710568	0.710219	0.711299	0.511893	−14.53
	Yingzizhuang, Beijing	39.60	116.30	loess	0.710162	0.710050	0.713671	0.512036	−11.74
	Taiyanggong, Beijing	39.60	116.30	loess	0.709539	0.709652	0.713760	0.512058	−11.31
	Dahongmen, Beijing	39.60	116.30	loess	0.710419	0.710428	0.712858	0.511989	−12.66
2	Xuanhua	40.50	115.00	loess	0.710583	0.710348	0.713716		
3	Kangxi, Hebei	40.20	115.48	loess	0.710724	0.710448	0.712154	0.511927	−13.87
4	Zhangbei	41.20	114.80	loess	0.709767	0.708499	0.711053		
5	Fengning, Hebei	41.20	116.50	loess	0.710180	0.710155	0.713770	0.512172	−9.09
6	Qahar Youyi Zhongqi	41.50	113.00	loess	0.711264	0.711442	0.714442	0.512024	−11.98
<i>North China (NC)</i>									
7	Hailar	49.50	119.00	sand	0.708537	0.708456	0.713582	0.512431	−4.03
8	Horqin	43.18	122.14	sand	0.710041	0.709950	0.714216	0.512280	−6.97
9	Zhangbaotun	42.10	122.43	loess	0.710276	0.710238	0.712715	0.512233	−7.90
10	Dzamin Uudo	43, 43	111.54	sand	0.709873	0.709815	0.715302	0.512388	−4.87
11	Erenhot	43.70	112.00	sand		0.709925	0.714086	0.512368	−5.26
12	Siziwang Qi	41.80	111.80	sand		0.710577	0.716017	0.512132	−9.88
13	Huade	42.80	114.50	loess	0.710568	0.710413	0.713417	0.512251	−7.55
14 <sup>a</sup>	Urumqi	43.50	87.50	loess	0.709510	0.709294	0.713984	0.512462	−3.43
15 <sup>a</sup>	Bayan Mod	41.50	105.00	loess	0.711394	0.711338	0.715528	0.512167	−9.19
<i>Western Beijing (WBJ)</i>									
16	Datong	40.20	113.20	loess	0.712357	0.711446	0.714165	0.511803	−16.29
17 <sup>a</sup>	Baotou	40.70	110.00	sand	0.711424	0.711242	0.712221	0.511559	−21.05
18	Urad Qianqi	40.43	108.40	loess		0.710617	0.716703	0.511576	−20.71
19	Wuhai, Mu Us Shamo	39.15	107.56	loess	0.711304	0.711145	0.716698	0.511824	−15.89
<i>Southern Gobi and Loess Plateau (SG–LP)</i>									
20 <sup>b</sup>	Tengger Shamo	38.50	105.00	sand	0.712685	0.712812	0.721975	0.512021	−12.04
21	Tengger Shamo	38.50	105.00	loess	0.711297	0.710893	0.718782	0.512072	−11.04
22	Mu Us Shamo	38.50	108.50	loess	0.711577	0.711082	0.718303	0.512102	−10.46
23	Alxa	38.60	105.50	loess	0.711415	0.711208	0.717645	0.512121	−10.09
24	Murengaole	38.60	105.50	loess	0.711428	0.711185	0.717579	0.512106	−10.37
25 <sup>b</sup>	Yinchuan	38.30	106.30	loess	0.711420	0.710885	0.718809	0.512101	−10.48
26	Yinchuan	38.25	105.59	loess	0.711769	0.711409	0.720741	0.512081	−10.87
27	Taiyuan	38.00	112.50	loess	0.711173	0.710863	0.717883	0.512015	−12.16
28	Jinchang	38.35	102.33	loess	0.711906	0.711837	0.721008	0.512016	−12.13
29	Zhongwei	37.46	105.53	loess	0.711419	0.711231	0.717098	0.512037	−11.72
30	Helan Shan	38.21	105.49	loess	0.711667	0.711397	0.718591	0.512061	−11.26
31	Alxa Zuoqi	38.35	105.38	loess	0.711972	0.712312	0.719411	0.512075	−10.98
32	Erdos	40.40	110.16	sand	0.711666	0.711072	0.720929	0.511918	−14.04
33	Erdos	40.00	110.00	sand	0.712461	0.711704	0.736280	0.511370	−24.74
34 <sup>a</sup>	Badain Jaran Shamo	39.00	102.50	sand	0.712782	0.712160	0.715360	0.512165	−9.23
35	Tianshijing	40.15	95.14	loess	0.711797	0.710866	0.717467	0.512003	−12.38
36	Yangwan	39.56	94.02	loess	0.712482	0.712617	0.736579	0.511169	−28.66
37	Jiuquan	39.51	97.53	loess	0.711509	0.711277	0.721424	0.512042	−11.62
38	Zhangye	39.14	99.28	loess	0.711822	0.711441	0.722161	0.512064	−11.19
39	Yuhuang	38.16	102.06	loess	0.711675	0.711377	0.722958	0.512033	−11.79
40	Wuwei	37.20	102.54	loess	0.712041	0.711544	0.719936	0.512054	−11.40
41	Badain Jaran Shamo	38.34	102.54	sand	0.712680	0.712249	0.719864	0.511984	−12.76
42	Lanzhou	36.21	103.33	loess	0.711094	0.710771	0.717111	0.512086	−10.77

Table 1 (continued)

No.	Location	Latitude	longitude	Soil type	H <sub>2</sub> O-leachate <sup>87</sup> Sr/ <sup>86</sup> Sr	HOAc-leachate <sup>87</sup> Sr/ <sup>86</sup> Sr	HOAc-residue		
							<sup>87</sup> Sr/ <sup>86</sup> Sr	<sup>143</sup> Nd/ <sup>144</sup> Nd	ε <sub>Nd</sub>
43 <sup>b</sup>	Lanzhou	36.00	103.80	loess	0.711297	0.710733	0.719438	0.512059	−11.29
44 <sup>b</sup>	Xian	34.20	108.80	loess	0.711198	0.710914	0.718321	0.512154	−9.44
<i>Takla Makan (TKM)</i>									
45 <sup>a</sup>	Taklimakan Shamo	38.00	83.00	sand	0.711906	0.713050	0.715670	0.511985	−12.74
46 <sup>a</sup>	Dunhuang	40.00	94.80	sand	0.711958	0.712770	0.715097	0.511928	−13.85
47 <sup>a</sup>	Ye Cheng	38.00	77.50	loess	0.710902	0.709906	0.716434	0.512150	−9.52
48 <sup>a</sup>	Qira	37.50	81.00	loess	0.711133	0.709905	0.716774	0.512119	−10.12

No: Sampling number is the same as in Fig. 1.

<sup>a</sup>Sr isotope ratio data are after Yokoo et al. (2001).

<sup>b</sup>Data are after Yokoo et al. (2004).

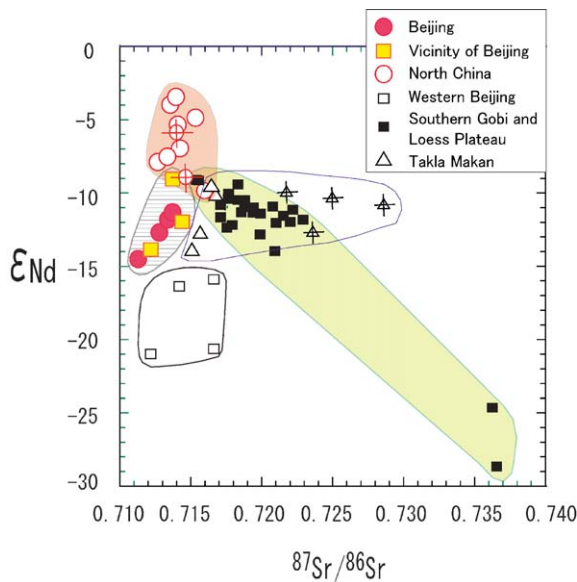


Fig. 2. <sup>87</sup>Sr/<sup>86</sup>Sr vs. ε<sub>Nd</sub> values of residual minerals after HOAc extraction. Symbols are the same as in Fig. 1. Several data points are from Yokoo and Nakano (2001) and Yokoo et al. (2004). Data for fine-grained (<5 μm) minerals from the Gobi and TKM deserts (indicated by a cross over a circle or triangle) are from Biscaye et al. (1997) and Bory et al. (2002), respectively.

et al., 1994; Gallet et al., 1996; Yokoo et al., 2004), indicating that Chinese arid soils are more heterogeneous than previously thought and are derived from multiple sources (Bory et al., 2002; Grousset et al., 2003). On the basis of the isotopic data, we can classify the arid soils of northern China into five regions (Figs. 1 and 2): north China (NC), at latitudes north of about

42°N; the Takla Makan Desert and the surrounding area (TKM); the southwestern Gobi Desert and Loess Plateau (SG–LP); Beijing and areas immediately to its northwest (BJ); and the western region, the area between about 200 and 1000 km west of Beijing (WBJ).

The SG–LP covers a large geographical region, roughly between 35°N and 40°N and between 90°E and 110°E, and is composed of many deserts (Badain Jaran, Tengger, Ulan Buh, Hobq, Mu Us), collectively called here the southern Gobi Desert, and the loess areas to the south and southeast, known as the Central Loess Plateau. The HOAc-insoluble materials from the SG–LP are characterized by high <sup>87</sup>Sr/<sup>86</sup>Sr ratios (>0.717, except for two samples with ratios of 0.715) and moderate ε<sub>Nd</sub> (mean, −11.2 ± 2.8, excluding two samples with values of −28.7 and −24.7). The relatively homogeneous ε<sub>Nd</sub> values are similar to those reported by Gallet et al. (1996) for bulk loess and paleosol samples from the Loess Plateau (mean, −10 ± 0.5), showing that most Nd in arid soils is contained in HOAc-insoluble silicates and phosphates (Yokoo et al., 2004). The HOAc-insoluble minerals of TKM have similar ε<sub>Nd</sub> values to those of SG–LP, but the former has lower <sup>87</sup>Sr/<sup>86</sup>Sr values than the latter. However, our TKM <sup>87</sup>Sr/<sup>86</sup>Sr values are lower than those reported by Bory et al. (2002) for the fine-grained soil minerals from the Takla Makan Desert (mean 0.725 ± 3), but have similar ε<sub>Nd</sub> values (Fig. 2). This disagreement may suggest that the Sr–Nd isotopic ratios of TKM soils are heterogeneous and overlap those of SG–LP. Other soils from NC, BJ, and WBJ are characterized by <sup>87</sup>Sr/<sup>86</sup>Sr values (0.711 to 0.717) lower than those of SG–LP. Of these, the HOAc-insoluble minerals of NC are characterized by high ε<sub>Nd</sub> values (−9.9 to −3.4), whereas those of WBJ are low (−21.1 to −15.9). The low <sup>87</sup>Sr/<sup>86</sup>Sr values and high ε<sub>Nd</sub> values of NC are consistent with those reported values (Biscaye et al., 1997) for three

soils from Mongolia, which indicates that the soil isotopic features of NC extend to south Mongolia. The Sr–Nd isotopic ratios of their HOAc-insoluble minerals show that BJ soils are intermediate between NC and WBJ soils, indicating that the dust falling on Beijing is mainly derived from these two regions.

The H<sub>2</sub>O- and HOAc-extracted minerals had similar  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, which were distinctly lower than those of the residual minerals resistant to HOAc (Yokoo et al., 2004). This result strongly suggests that these minerals were not derived from provenance materials but were formed in each case by similar processes of salinization. These salinization minerals showed a regional Sr isotopic variation; most BJ and NC samples had  $^{87}\text{Sr}/^{86}\text{Sr}$  values lower than 0.711, whereas those from other areas had  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios higher than this value (Fig. 3). This result is consistent with the Sr–Nd isotopic constraints from the HOAc-insoluble minerals, but shows a larger contribution of NC soils to BJ dust. The H<sub>2</sub>O-leachates of most samples (40 in 49 samples) had slightly higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios than their HOAc-equivalents (Table 1, Fig. 2). Yokoo et al. (2004) reported that Sr in the H<sub>2</sub>O-extracted minerals (halite, sylvite, and anhydrite) of several soil samples in the SG–LP was <0.1% in the bulk samples, and 50% of the Sr was distributed in the HOAc-extracted minerals (carbonate). Accordingly, the higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in the H<sub>2</sub>O-leachates may be ascribed to a contribution of labile Sr on the surface of silicate minerals with high  $^{87}\text{Sr}/^{86}\text{Sr}$  in addition to Sr from salinization minerals. The contribution of the silicate Sr would become

negligible in the HOAc-leachates because of the dominant Sr in carbonate.

### 3.2. Origin of mineral dust falling on Beijing

The isotopic signatures of the salinization- and provenance-derived minerals clearly show that the contribution of SG–LP and TKM soils to the BJ dust was smaller than that of the NC and WBJ soils. Many modeling and air-mass trajectory studies (Zhang et al., 2001; Xuan and Sokolik, 2002) show a small emission of mineral dust from the Loess Plateau but a high potential for dust emission and frequent observations of dust from the southern Gobi and Takla Makan deserts. This apparent contradiction can be explained by examining wind direction and the distance between Beijing and the dust emission areas. The direction of the jet stream over the middle latitudes of China, estimated by the distribution of mean geopotential height, is northwesterly to westnorthwesterly during most seasons, but it is westerly in summer when Asian dust activity is weak (NMC, 1997). This leads to the generally accepted conclusion that the source of dust from the loess of the Loess Plateau is the fine-grained fraction of desert sand which has been sieved and graded during atmospheric transport from the northwest to the southeast (Liu, 1985; Biscaye et al., 1997; Sun et al., 2002). This conclusion can account for the Sr–Nd isotopic similarity of the HOAc-insoluble minerals between the Loess Plateau and the deserts to its north because their provenance materials are similar. This wind direction suggests that the amount of dust from western to southwestern regions (TKM, SG–LP) falling on Beijing would be less than that from the northwestern region (NC). Further, the prevailing near-surface wind direction in the TKM is easterly and northeasterly, and dust from there is lofted to high elevations (>5000 m) and moves north and northwestward (Sun et al., 2002). As a result, most TKM-derived dust is thought to be transported directly to the Pacific region without large deposition on the Chinese mainland.

The distance between the emission and deposition areas is also an important factor because the total deposition of Asian dust decreases exponentially in the Pacific region with distance from the source (Mori et al., 2002). The distance at which the concentrations are reduced to one-half is estimated to be about 450 km. Hence, the proportion of total dust deposited at Beijing generated in the Takla Makan and southern Gobi deserts is estimated to be 0.8–1.6% and 6–13%, respectively, whereas the proportion generated in the WBJ is 0.25–0.5. Thus, the large proportion of the NC- and WBJ-derived dust in the BJ samples can be attributed to the short distance between these areas and Beijing and the prevailing northwesterly movement of the dust-carrying air mass.

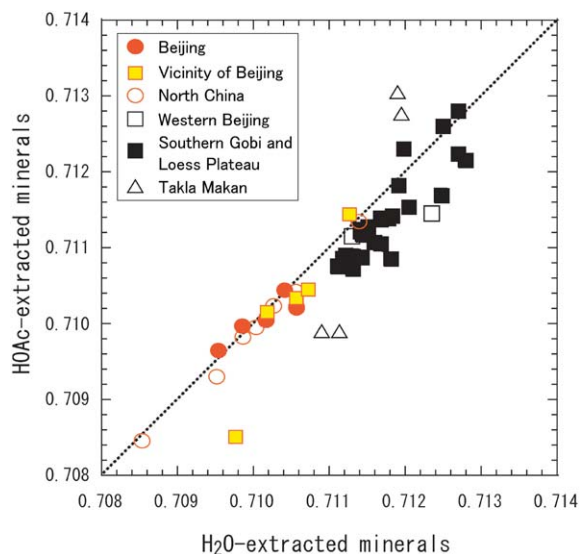


Fig. 3. Relationship between  $^{87}\text{Sr}/^{86}\text{Sr}$  values in H<sub>2</sub>O- and HOAc-extracted minerals. Symbols are the same as in Figs. 1 and 2.

The steppe lands in Inner Mongolia in China to southern Mongolia have been desertified as a result of overgrazing, inappropriate cultivation practice, and overuse of irrigation (Sheehy, 1992), in addition to a decrease in precipitation (Yatagai and Yasunari, 1995). In these areas, lakes and ponds have dried up, and the fine-grained sediments thus exposed are easily picked up by strong winds. Consequently, fine-grained sediments exposed by desertification may be a major source of dust falling on Beijing. Therefore, detailed research in these areas is essential for elucidating Asian dust sources as well as assessing desertification and other relevant environmental problems.

#### 4. Conclusions

Our Sr–Nd isotopic studies for surface soils in the desert and loess areas in northern China clearly show the presence of regional variation in the acid-resistant and salinization minerals, and suggest the dominant transportation of recent mineral dust to Beijing from its adjacent northwestern to western areas where the desertification is extending. Isotopic fingerprinting using two kinds of minerals is a powerful tool for constraining models of atmospheric circulation and dust transport as well as for identifying dust sources on event to glacial-interglacial timescales.

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