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Sr-Nd isotope geochemistry of eolian dust of the arid-semiarid areas in China: Implications for loess provenance and monsoon evolution

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Abstract Minerals and rocks have distinct $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios, depending on their geological origin and ages, and these isotope ratios are less altered than elemental composition during transport in the atmosphere or after deposition as sediments, thus stable isotopes of Sr and Nd have great potential as tracers for provenance and transport of materials. During the hypergene process, Sr isotope ratios of sediments are controlled by their parent rocks, particle sizes and chemical weathering. In general, the higher the Sr isotope ratios of parent rocks, and/or the more the fine-grained fractions, and/or the stronger the chemical weathering, thus the higher the Sr isotope ratios of sediments. On the contrary, there are lower Sr isotope ratios of sediments. Nd isotope ratios of sediments, independent of their particle sizes and chemical weathering, are only associated with parent rocks. For the provenance of the Chinese Loess Plateau, different reseachers drew discordant and even contradictory conclusions by using the method of Sr-Nd isotopic tracing. From the previous Nd isotope data, it is considered that the Tarim Basin, deserts in the central and west parts of Inner Mongolia and the Tibetan Plateau are the main sources of the Chinese Loess Plateau, and are also manufacturers for eolian dust of the Far East regions, together with the Chinese Loess Plateau. Sr isotope ratios of eolian dust are solely affected by wind sorting and weathering-pedogenesis due to its homogeneous composition in the Chinese Loess Plateau. Wind sorting is related to the East Asian winter monsoon but weathering-pedogenesis is mainly associated with the East Asian summer monsoon. Studies on Sr isotopic compositions of the loess-paleosol sequence suggest that $^{87}Sr/^{86}Sr$ ratios in acid-soluble materials are an index for chemical weathering intensity of the Chinese Loess Plateau, indicating the East Asian summer monsoon variations, whereas $^{87}Sr/^{86}Sr$ ratios in acid-insoluble materials are significantly controlled by particle sizes, and can be used as a proxy indicator reflecting the East Asian winter monsoon variations. Variations of $^{87}Sr/^{86}Sr$ ratios in acid-insoluble materials in the past 2.6Ma further demonstrate that the East Asian winter monsoon gradually strengthened since the beginning of the Quaternary period. This result agrees with the prior conclusion that climate gradually cooled since the onset of the Quaternary Ice Age.

Keywords: Sr-Nd isotope, eolian dust, monsoon, loess, arid-semiarid areas.

Strontium (Sr) is a divalent alkaline earth element. Its ionic radius (0.113 nm) is slightly larger than that of calcium (0.099 nm), and Sr thus substitutes isomorphously for Ca in many minerals. Neodymium (Nd) is a rare earth element. Its chemical property and ionic radius are similar to those of samarium. Neodymium is a necessary component of such some minerals as monazite and allanite, and also substitutes isomorphously for Ca and Sr in minerals such as apatite and fluorite. $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios are greatly different in minerals and rocks with various ages, and distinct Rb/Sr and Sm/Nd ratios, but less altered than elemental composition during transport in the atmosphere or after deposition as sediments. In earth sciences, Sr and Nd isotopic tracers for provenance and transport of geological materials have been a classical means. For instance, Sr-Nd isotope geochemistry traced origins of ore deposits, rocks and minerals[1−4], crustal evolution[5,6], oil movement[7], hydrological cycle[8], source of soil materials[9−11], manufacture, transport and deposition of atmospheric dust[12−14] and paleoclimatic and paleoenvironmental evolutions[15−18].

There is a history of 2.6 Ma or longer (7 or 22 Ma) for the Chinese Loess Plateau[19,20]. The Chinese Loess Plateau has been regarded as a natural archive for studies of regional and global climate and environment changes due to continuous deposit and great thickness of eolian dust in it[19]. So far, investigations on loess and paleoclimate are involved in many science prob-

lems such as manufacture, transport and deposition of eolian dust[21,22], variation of loess provenance and development of the Chinese Loess Plateau[23—25], uplift of the Tibetan Plateau and atmospheric circulation change[26], and formation and evolution of the East Asian monsoon and its inner link to global climate change[27,28]. However, provenance of the Chinese Loess Plateau is not clear yet, and it is still necessary and urgent to study the East Asian monsoon to find new proxy indicators which can verify prior indicators. In this paper, first, the influences on Sr and Nd isotopic compositions of sediments during the hypergene process will be analyzed in detail, and second, geochemical implications of Sr and Nd isotopes will be explored for the provenance of the Chinese Loess Plateau and the evolution of the East Asian monson.

1 Influences on Sr and Nd isotopic compositions during the hypergene process

In order to examine the influences on Nd isotopic compositions, Goldstein *et al.*[29] divided a sediment sample from Amazon river into three particle-size fractions(> 45 μm, 45—2 μm, <2 μm) and determined their Nd isotopic compositions. The result showed that $\varepsilon_{Nd}(0)$ values (–9.3—–9.6) did not vary greatly with decreasing grain-size in sediments. Nakai *et al.*[30] considered that particle-size sorting did not significantly affect Nd isotopic compositions since the Nd isotopic composition of Asian loess was uniform and that of the central north Pacific was also uniform and identical to that of Asian loess. Derry and France-Lanord[31] observed differences of around 1—2ε units between grain-size fractions <2 μm and <50 μm of sediments in the Bengal fan. $\varepsilon_{Nd}(0)$ values of different grain size fractions of the moraine from the Kunlun Mountains varied from –10 to –11[32]. Eisenhauer *et al.*[33] and Tütken *et al.*[34] analyzed Nd isotopic compositions of sediments from Arctic Ocean, and found that $\varepsilon_{Nd}(0)$ values were similar in different grain size fractions of the same sample with a small variational amplitude of 0.3—0.8, independent of their corresponding grain size distribution within the statistical uncertainties. We determined $\varepsilon_{Nd}(0)$ values in different grain size fractions of eolian dust from the Duanjiapo section in the Chinese Loess Plateau (Table 1). Results showed that $\varepsilon_{Nd}(0)$ values in the samples determined ranged from –8.5 to –9.2 with a small difference of –0.7 between the maximum and the minimum, indicating the particle size effects on $\varepsilon_{Nd}(0)$ values of eolian dust were negligible.

Table 1 $\varepsilon_{Nd}(0)$ values in different grain size fractions of different layers in the Duanjiapo section[a)]

Sample No.	Bulk sample	>45 μm	45—28 μm	<28 μm	<2 μm
L_1	–8.8	–8.8	–8.5	–9.2	–8.7
S_5	–8.6	–8.5	–8.9	–9	–9.2
RC	–9.1	–9.1	–8.9	–8.7	–8.6

a) Nd isotope ratios were determined with a VG354 mass spectrometer at the Center of Modern Analysis, Nanjing University. The $^{143}Nd/^{144}Nd$ ratio of Standard Reference Material La Jolla measured was 0.511840±8 in this study (normalized values $^{146}Nd/^{144}Nd$=0.7219). The analytical blank was below 60 pg for Nd.

In addition, Yokoo *et al.*[35] determined Nd isotopic compositions of the loess and desert sands after a sequential extraction experiment and found that $\varepsilon_{Nd}(0)$ values in the loess from the Chinese Loess Plateau tended to be higher in fine-grained fractions, but their variations were small. Their results also showed that different chemical fractions of the same sample had similar $\varepsilon_{Nd}(0)$ values. Jones *et al.*[36] made leaching experiments for Chinese loess. The results exhibited that $\varepsilon_{Nd}(0)$ values of leachates and residues were fairly homogeneous. Investigations of Tacihkawa *et al.*[37] on the $\varepsilon_{Nd}(0)$ of sediments from the Mediterranean Sea revealed that the results of the HCl and HOAc leaching agreed within uncertainties.

As a result, Nd isotopic compositions of sediments were independent of particle sorting and chemical weathering, but mainly followed those of parent rocks.

Relative to Nd isotopic compositions, Sr isotopic compositions of sediments or weathering profiles were affected by more factors. They were not only influenced by Sr isotopic compositions of parent rocks but also controlled by grain size variations and chemical weathering.

Dasch[38] compared two grain size fractions from the same sample and concluded that $^{87}Sr/^{86}Sr$ ratios could increase with decreasing grain size due to the dominance of Rb-rich micas in the clay fraction. Sr isotope data in different grained fractions of Lanzhou loess (<2 μm, 0.7202; 2—20 μm, 0.7193; >20 μm, 0.7178) from Asahara *et al.*[39] displayed that in the same sample the fine fraction had a slightly higher ratio than the coarse fraction. Eisenhauer *et al.*[33] and Tütken *et al.*[34] discovered that Sr isotope ratios of Arctic Ocean sediments increased with a decrease of the grain size. Smith *et al.*[40] demonstrated that finer material transported further had higher Sr isotopic ratios.

Gallet *et al*.[41] reported that paleosols systematically showed higher $^{87}Sr/^{86}Sr$ ratios than the loess due to leaching of detrital (likely marine) carbonate with low $^{87}Sr/^{86}Sr$ ratios during pedogenesis. $^{87}Sr/^{86}Sr$ ratios of loess and paleosol samples from S_0-L_1-S_1-L_2 sequences of three sections (Xining, Xifeng and Jixian) in the Chinese Loess Plateau were determined in detail by Jahn *et al*.[42]. $^{87}Sr/^{86}Sr$ ratios varied from –0.7143 to –0.7149 for Xining section, from –0.7150 to –0.7165 for Xifeng section, and from –0.7143 to –0.7183 for Jixian section. From these data, it was inferred that there were relatively high $^{87}Sr/^{86}Sr$ ratios in places with more precipitation. Biscaye *et al*.[43] analyzed three Chinese loess samples, which were size-selected (<5 μm) and subjected to acid-removal of carbonate and other soluble matter. Results showed that the silicate fractions had very high $^{87}Sr/^{86}Sr$ ratios of 0.7264—0.7276, and confirmed that calcite exerted a significant control on Sr isotopic compositions. Kanayama *et al*.[14] further demonstrated the above conclusion with leaching experiments of loess from Yili in China. Yokoo *et al*.[35] viewed on the basis of the studies of loess and desert sands from China that the effects of weathering-pedogenesis on Sr isotope ratios could not be neglected. For the same sample, the stronger the chemical weathering, the higher the Sr isotope ratio.

We analyzed three samples from the Lishi loess, the Wuchen loess and the Tertiary red clay in the Lingtai section, respectively. Three samples were determined for Sr isotope ratios after grain-size separation and acid treatment. Sr isotope data are presented in Table 2. The samples had obviously higher $^{87}Sr/^{86}Sr$ ratios after acid treatment than before. Also, Sr isotope ratios tended to be higher in the fine fraction, especially greatly higher in the <2μm fraction than other coarser fractions. These results further agreed with previous conclusions: grain-size sorting and chemical weathering apparently influenced Sr isotope ratios, namely, the smaller the particle sizes, the higher the Sr isotope ratios, and the stronger the chemical weathering, the higher the Sr isotope ratios.

2 Uniformity of source materials for the Chinese Loess Plateau

Thick and continuous dust deposits in the Chinese Loess Plateau were of eolian origin,which had not been invaded by groundwater since the late Tertiary[23,24]. Many proxy indicators reflecting the East Asian monsoon evolution were derived from eolian dust deposits in the Chinese Loess Plateau[27,28,44—49]. However, most proxy indicators were established on the basis of this assumption that material composition was homogenous in the Chinese Loess Plateau. Were source materials of the Chinese Loess Plateau spatio-temporally uniform?

Guo *et al*.[50] found that eolian dust varied distinctly in chemical compositions during different geological periods by investigating total Fe contents of Changwu and Yichuan sections with high resolutions, and then argued that the loess could not be regarded as a uniform material. From studies of Fe/Al ratios in the Lingtai section, Gu *et al*.[51] suggested that source materials supplying systems of eolian dust in Lingtai were unstable during the late Cenozoic except for the intervals of 4.5—3.2 Ma B.P. and 2.6—0.5 Ma B.P. Guo *et al*.[50] drew the conclusion relying on the hypothesis of negligible effects of grain sizes on Fe contents of eolian dust. Gu *et al*.[51] did not consider whether Fe/Al ratios in different grain size fractions were stable. In fact, Fe and Al contents, and Fe/Al ratios were different in different grain size fractions of eolian dust, and increased with a decrease of grain sizes[1]. Thus, more evidence will be

Table 2 Sr isotope ratios of the loess and the red clay from the Lingtai section [a]

Sample No.	Bulk sample	>45 μm	45—28 μm	28—2 μm	<2 μm
L_{10} (without acid treatment)	0.718583±6	0.715067±9	0.718440±13	0.718145±7	0.717608±12
L_{33} (without acid treatment)	0.716974±13	0.713737±29	0.712036±8	0.711129±11	0.719060±19
RC (without acid treatment)	0.716428±15	0.713277±13	0.716494±11	0.716758±26	0.717765±11
L_{10} (acid treatment)	0.720951±10	0.722665±18	0.719476±7	0.720302±10	0.726261±9
L_{33} (acid treatment)	0.720509±12	0.718122±20	0.720316±18	0.719415±13	0.729943±29
RC (acid treatment)	0.721554±18	0.719930±13	0.720479±28	0.719946±12	0.725733±9

a) Each sample with considerable weight was dipped in a Teflon container with 0.5 mol/L acetic acid solution at room temperature for more than 4 h. Then, each residue was derived by filtering and was preserved for Sr isotope analysis. Sr isotope ratios were determined with a VG 354 mass spectrometer at the Center of Modern Analysis, Nanjing University. The measured $^{87}Sr/^{86}Sr$ values were normalized to an $^{86}Sr/^{88}Sr$ value of 0.1194. The $^{87}Sr/^{86}Sr$ ratio of Standard Reference Material NBS 987 determined in this laboratory is 0.710340±40 (n=60) in recent two years. The analytical blank was below 1ng for Sr.

1) Liu L W. Mineralogy and element geochemistry of aeolian dust deposits on the Loess Plateau for the evolution of the East Asian monsoon during the last 3.6Ma. Dissertation for the Doctoral Degree. Nanjing: Nanjing University, 2002. 48—61

required to verify the conclusions mentioned above. Gallet *et al*.[41], Jahn *et al*.[42] and Yang *et al*.[52] analyzed stability of material sources in the Luochuan, Xifeng, Xining, Jixian and Lingtai sections by using Sr-Nd geochemical methods. They all made similar conclusions against the above opinion. $\varepsilon_{Nd}(0)$ and $^{87}Sr/^{86}Sr$ (–10.5—–9.2, 0.715—0.719) were rather uniform in both loess and paleosols from the L_1—L_7 sequence of the Luochuan section, and did not vary greatly with stratigraphic position. Therefore, Gallet *et al*.[41] suggested there was a uniform source for the Luochuan loess during the entire period of deposition from ~800ka ago to the present. Isotopic analyses of Jahn *et al*.[42] showed that three S_0—L_2 sequences (Xining, Xifeng and Jixian) displayed a restricted range of $\varepsilon_{Nd}(0)$ values (–10±0.5) and had typical upper crustal $^{87}Sr/^{86}Sr$ ratios (0.714—0.718), indicating that the dust source region must have undergone multiple recycling and thorough sedimentary mixing processes. The $\varepsilon_{Nd}(0)$ of eolian dust in the Lingtai section mostly varied from –8.8 to –10.6 during the last 7Ma. Thus, Yang *et al*.[52] proposed a uniform source region for the Chinese Loess Plateau as well. Although these studies were conducted at low resolutions, they still revealed chemical homogeneity of eolian dust in the Chinese Loess Plateau during various periods. Geographically, the $\varepsilon_{Nd}(0)$ was –9.6 for the bulk sample from Lanzhou[30], was –10.9 and –11.2, respectively for two typical loess samples from the Chinese Loess Plateau[25], ranged between –10.1 and –11.3 for bulk samples and residues of loess from Yinchuan and Lanzhou[35] and varied within the range of -11.6 ± 0.8 for acid-insoluble materials of loess from Yinchuan, Taiyuan, Jinchang, Zhongwei, Jiuquan, Zhangye, Wuwei and Lanzhou[53]. $\varepsilon_{Nd}(0)$ values varied less in these surface soil samples, indicating that eolian dust in the Chinese Loess Plateau was spacially uniform as well. This conclusion was in accordance with the result Wen *et al*.[54] obtained from comparisons of $\delta^{18}O$ values in the same grain size fractions of Malan loess and comparisons of REE in the clay fraction and in Malan loess between different locations in the middle reaches of Yellow River. So, source materials of the Chinese Loess Plateau were spatio-temporally uniform in chemical compositions in terms of Sr-Nd isotope geochemistry.

3 Source areas of eolian dust in the Chinese Loess Plateau

Northern China is characterized by an extensive distribution of deserts, gobi areas and deteriorated lands. These regions are chiefly controlled by the upper-level westerly and the low-level East Asian monsoon. Northerly and northwesterly winds of the East Asian winter monsoon are the main driving forces to produce and carry eolian dust. In a broad sense, the gobi regions and deserts adjacent to the northern part of the Chinese Loess Plateau and northwestern inland basins in China could be the main sources of the Chinese Loess Plateau[19]. Where on earth did eolian dust in the Chinese Loess Plateau come from? Bowler *et al*.[55] argued that the Qaidam Basin has been an important dust source for the Chinese Loess Plateau during the geological past. Derbyshire *et al*.[56] concluded that the huge piedmont alluvial fans along the northern Qilian Mts. were the main sources for loess deposits in the western Loess Plateau. Fang *et al*.[57] considered that the Tibetan Plateau was an important dust sources for the vicinity of its east and the Far East – Pacific Ocean regions. Wang *et al*.[58] pointed out that lacustrine sediments, shrub dunes, gobi deserts and deteriorated lands were the most potential geogenic dust contributors, but mobile dunes were not. Different researchers drew distinct conclusions from different angles. Apparently, sources of the Chinese Loess Plateau were extremely complicated.

In recent years, some researchers explored inner relations between deserts and the loess from the angle of Sr-Nd isotope geochemistry. For example, Liu *et al*.[25] concluded that the loess on the Chinese Loess Plateau might have a source region in the Tarim Basin but clearly not in the North Tianshan by comparing Sr-Nd isotope geochemistry of sediments of various types from the Tarim Basin and the North Tianshan with those of loess deposits from the Chinese Loess Plateau. Sun[59] suggested that the gobi and deserts in central and west parts of Mongolia and the adjoining gobi and deserts in China were the dominant source areas of the Chinese Loess Plateau by analyzing isotopic, chemical and mineralogical characteristics of the <20μm fraction of loess from the three northwestern inland basins and the Chinese Loess Plateau, together with data of modern meteorology. Investigations of Yokoo *et al*.[35] implied that the Taklimakan Desert was not a main source of the Chinese Loess Plateau, but deserts in the Alxa and Ordos plateaus were. Nakano *et al*.[53] determined mineralogical, elemental and Sr-Nd isotopic composi-

tions of acid-soluble and acid-insoluble materials in desert sands from Tengger Desert, Mu Us Desert and in the loess from the Chinese Loess Plateau, and pointed out that the loess of the Chinese Loess Plateau was composed of the fine-grained fraction of desert sand that was transported to the southeast in the atmosphere. These studies were carried out on the basis of the viewpoint that eolian dust in the Chinese Loess Plateau came from deserts in China, but identical ideas were not obtained yet.

Sr-Nd isotopic compositions were distinct in various rocks and minerals, and were less altered during the hypergene transport process, these isotopes thus had great potentials as provenance tracers for rocks and minerals. However, as we discussed in section 1, for eolian dust, chemical weathering and grain size effect prominently influenced Sr isotopic compositions although they had not a great impact on Nd isotopic compositions. Sr isotope ratios of whole samples only reflected mixed information if whole samples were used as testing objects because whole samples consisted of different minerals. An obvious progress was made in the pre-treatment procedure if a certain-grained fraction of the whole sample was determined as a testing object for Sr isotope ratios, but different components with different Sr isotope ratios in a certain-grained fraction were not separated yet. If bulk samples after chemical treatment were selected as testing objects, Sr isotope ratios also recorded mixed information from different grain size fractions. Northern eolian dust is typically a sedimentary compound, of which coarse-grained fractions probably originated from local sources whereas fine-grained fractions could come from remote sources, also perhaps subject to weathering-pedogenesis during the transport in the atmosphere or the accumulation in the Chinese Loess Plateau. Consequently, it is necessary for excavating more authentic information on loess provenance to select a suitable certain-grained fraction without interferences of other grained fractions and apply reasonable chemical pre-treatments avoiding the influences of chemical weathering on eolian dust in the Chinese Loess Pleateau when the relationship between the loess and deserts is discussed by using Sr-Nd isotope methods. Besides, identical ages of samples determined and systemic investigations should be importantly considered.

On the basis of previous data, we plotted a map of the arid-semiarid areas in China with Nd isotopic compositions for loess, modern dust and other sediments (Fig. 1). As Fig. 1 displayed, $\varepsilon_{Nd}(0)$ values varied mainly from −12.8 to −9.2 for loess, surface soils, sands and other sediments from the Tarim Basin and surrounding areas, deserts in the central and western Inner Mongolia, the Tibetan Plateau, and were similar to those for eolian dust in the Chinese Loess Plateau.

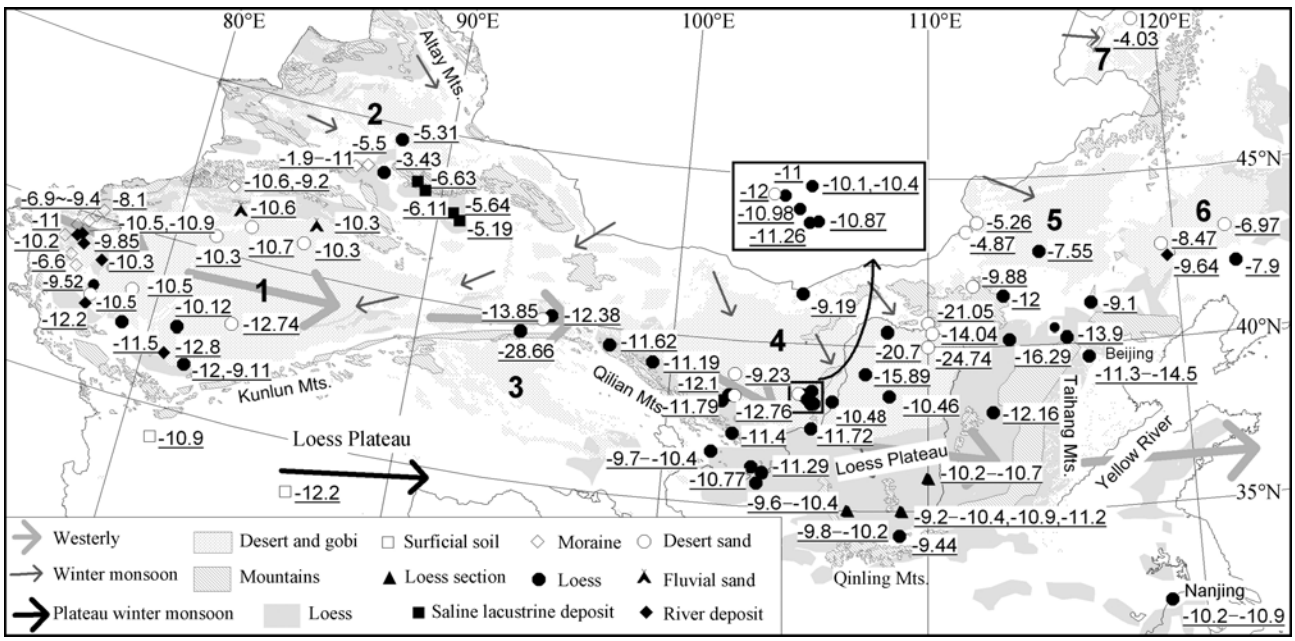

Fig. 1. Map of the arid-semiarid areas in China with Nd isotopic compositions. 1. Tarim Basin; 2. Zhungar Basin; 3. Qaidam Basin; 4. Deserts in the central and western Inner Mongolia; 5. Otindag sandy land; 6. Horqin sandy land; 7. Hulun Buir sandy land. $\varepsilon_{Nd}(0)$ data are from refs. [25, 29, 30, 32, 35, 36, 41, 42, 53].

But $\varepsilon_{Nd}(0)$ values for sediments from other areas were quite distinct from those for eolian dust in the Chinese Loess Plateau. Therefore, deserts in the Tarim Basin and in the central and western Inner Mongolia and the Tibetan Plateau were probably dominant source areas of the Chinese Loess Plateau.

4 Long range transport of northern eolian dust

There are several great dust source areas in the world[12,60－62]. Asian regions, particularly the arid-semiarid areas in China, are one of the greatest atmospheric dust sources. According to evaluation from Zhang *et al*.[62], annual average release flux of Asian dust reached 800Tg, accounting for about half the global dust yearly production. Meteorologic observations[63,64] showed that Asian dust was transported for a long range downwind by the westerly to Korea, Japan, the Pacific Ocean, northern America and even Greenland. Making up a large fraction of the atmospheric aerosol, the mineral dust led to the decrease of atmosphere environment quality, harmed human health and impacted on climate change. On the other hand, it neutralized acid rain in the atmosphere, facilitated biogeochemical cycle of ocean and soils to improve productivities. Eolian dust played an important role in the Earth environment system, thus attracting increasing attention from people.

Over the past 20 years, researchers obtained evidence for long range transport and deposit of northern eolian dust by using Sr-Nd isotope geochemical tracers. Nakai *et al*.[30] investigated in detail provenance of dust in the Pacific Ocean. Their results revealed that samples from the north central Pacific had $\varepsilon_{Nd}(0)$ of −10 and $^{87}Sr/^{86}Sr$ of >0.715 which were virtually identical to the values for Asian loess. Jones *et al*.[36] reported that sediments from a central North Pacific province were characterized by unradiogenic and remarkably homogeneous $\varepsilon_{Nd}(0)(-10.2\pm5)$, of which the silicate fraction was exclusively eolian and originated from Chinese loess. Asahara *et al*.[40] agreed by comparison of $^{87}Sr/^{86}Sr$ ratios between sediments from the Pacific Ocean and loess from Lanzhou and Nanjing that a lot of loess from the Asian continent was carried by middle-latitude westerlies into the north central Pacific, and then led to high $^{87}Sr/^{86}Sr$ ratios (0.711—0.719) in the north central Pacific. Investigations of Biscaye *et al*.[43] on GISP2 confirmed that dust in ice core came from Asia. Svensson *et al*.[65] also recognized dust in ice core of Asian inland origin. Asahara *et al*.[66] proposed that the Asian continental materials and the volcanic materials from island-arc volcanos were two main end members controlling Rb-Sr isotopic systematics of sediments from the Pacific Ocean. Kanayama *et al*.[14] analyzed Sr isotopic compositions of particles over Japan and then concluded that particles over Japan came from inland Asia (Mongolia and Inner Mongolia of China) during the "Kosa" event. Kanayama *et al*.[13] analyzed "Kosa" particles extracted from "Red snow" that fell on Yamagata Prefecture in January and March 2001 for mineralogy, chemistry and Sr isotopes. They claimed that "Kosa" particles were of inland Asian origin, and the dust source for the event in January was located in Mongolia and the northern part of China (40—45°N, 100—110°E), whereas the source in March was from a slightly lower latitude (35—45°N, 100—115°E). Pettke *et al*.[67] determined $\varepsilon_{Nd}(0)$ values for the silicate fractions of sediments at ODP Sites 885/886 in the past 11 Ma. Yang *et al*.[52] analyzed variation in $\varepsilon_{Nd}(0)$ values for eolian dust in the Lingtai section during the last 7Ma. Except for high $\varepsilon_{Nd}(0)$ values influenced by the volcanic material which was considered by authors, two curves of $\varepsilon_{Nd}(0)$ values varied principally within a similar and narrow range in the past 7Ma (Fig. 2), indicating that the silicate fractions of sediments in the central north Pacific Ocean had a similar source to eolian dust in the Lingtai section.

From comparisons of $\varepsilon_{Nd}(0)$ values between eolian dust of northern China (Fig. 1) and sediments in the north Pacific Ocean[36, 67], it was inferred that the Tibetan Plateau, the Tarim Basin, deserts in the central and western inner Mongolia and the Chinese Loess Plateau were the main sources for eolian dust transported over long distances to Korea, Japan, the north Pacific Ocean and northern America. The westerly was perhaps both an inner driver and a carrier for eolian dust on the Tibetan Plateau which was transported over long distances. However, the westerly may solely serve as a carrier for eolian dust in other areas of northern China to transport over long distances but the East Asian winter monsoon was an inner driver controlling eolian dust in these regions transported far east.

5 Sr isotope tracing the East Asian monsoon evolution

5.1 $^{87}Sr/^{86}Sr$ ratios in acid-soluble materials reflecting East Asian summer monsoon variations

Chen *et al*.[68] drew a conclusion based on studies of

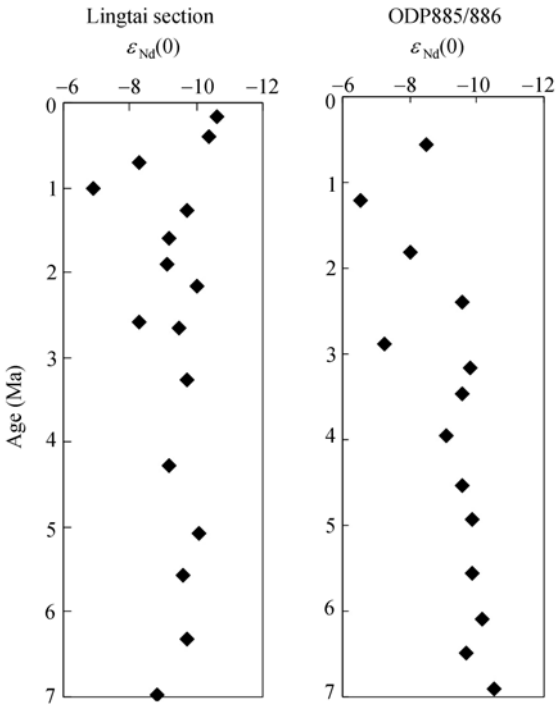

Fig. 2. Comparison of $\varepsilon_{Nd}(0)$ values between the Lingtai section[52] and ODP 885/886[67].

the Luochuan section that there was a rising trend of $^{87}Sr/^{86}Sr$ ratios in the following order: loess calcite→weakly pedogenetic, paleosol calcite→palaeosol calcite→pure secondary calcite, and further pointed out that chemical weathering was an important influence on variations in $^{87}Sr/^{86}Sr$ ratios of these materials. Liu *et al.*[69] considered that variations in $^{87}Sr/^{86}Sr$ ratios of carbonates (0.8 mol/L HCl solution) from Luochuan loess section indicated the intensity of chemical weathering and paleoclimatic change. Yang *et al.*[18] investigated Sr isotope ratios at high resolutions for the S_0-L_1-S_1 sequence of the Heimugou section in Luochuan, and obtained a proxy for chemical weathering associated with the East Asian summer monsoon-$^{87}Sr/^{86}Sr$ ratios in calcite. Although these researchers had approximately similar conclusions, they used different chemical pre-treatment methods for samples determined. Of these pretreatment methods, the method of Yang *et al.*[18] was the most reasonable. They suggested through detailed experiments that 0.5 mol/L acetic acid solution was the most available medium to dissolve calcites in loess deposits but not the detrital minerals. We analyzed Sr isotopic characteristics in acid-soluble materials of the loess-paleosol sequence in Luochuan during the last 800ka by applying the chemical pretreatment method of Yang *et al.*[18] (Fig. 3). Results

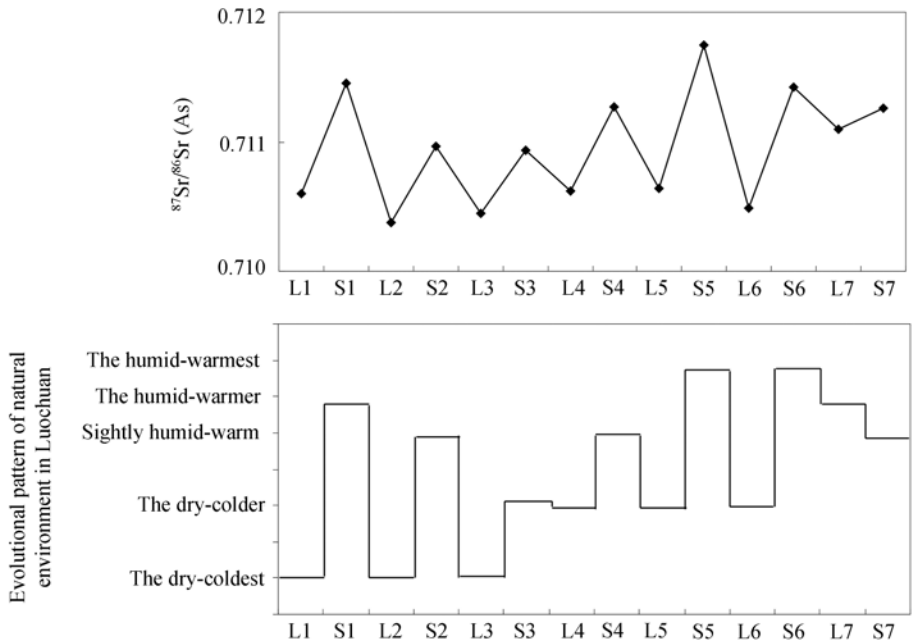

Fig. 3. Sr isotope ratios of acid-soluble materials and evolutional pattern of natural environments[71] in the loess-paleosol sequence of the Luochuan section. Acid treatment procedure: Each sample with considerable weight was taken and dipped in a Teflon container with 0.5 mol/L acetic acid solution at room temperature for more than 4 h. Then, each leachate was derived by using a quantitative filter paper (ϕ9cm) and was preserved for Sr isotopic analysis. Sr isotope ratios were determined with a VG 354 mass spectrometer at the Center of Modern Analysis, Nanjing University. The measured $^{87}Sr/^{86}Sr$ values were normalized to an $^{86}Sr/^{88}Sr$ value of 0.1194. The $^{87}Sr/^{86}Sr$ ratio of Standard Reference Material NBS 987 determined in this laboratory is 0.710340±40 (n = 60) in recent two years. The analytical blank was below 1ng for Sr. Standard deviation of Sr isotopic ratios for the same sample with multiple determinations was less than ±0.00005 [18].

showed that Sr isotope ratios were higher in paleosols whereas lower in the loess with a regular fluctuation between high and low values, and also gradually decreased upwards with strata.

Eolian dust in the Chinese Loess Plateau was similar in mineralogical composition although different in content[19]. Different minerals had different resistant ability to chemical weathering. Minerals of eolian dust could be classified in the order of weatherproof intensity from weak to strong: halite and gypsum→calcite→dolomite→phosphate→silicate minerals (feldspar and clay minerals)→Quartz. Primary minerals of eolian dust in the Chinese Loess Plateau were chemically destroyed and secondary minerals formed synchronously due to influences of chemical weathering. There were basically three types of primary minerals: first, quartz without Sr and Rb was not responsible for $^{87}Sr/^{86}Sr$ ratios of eolian dust; second, silicate minerals with rich Rb and poor Sr had high $^{87}Sr/^{86}Sr$ ratios; last, calcite, dolomite and anorthite with rich Sr and poor Rb had low $^{87}Sr/^{86}Sr$ ratios. Chemical weathering of the Chinese Loess Plateau was at an initial stage during which labile elements such as Ca and Na were merely leached out of the Chinese Loess Plateau[70]. Chemical weathering brought about dissolution of most primary carbonates which Sr with low $^{87}Sr/^{86}Sr$ ratios released from partly entered secondary carbonates and was partly leached out of the Chinese Loess Plateau. On the other hand, chemical weathering destroyed silicate minerals such as feldspar and clay minerals with rich Rb and poor Sr which Sr with high $^{87}Sr/^{86}Sr$ ratios released from was partly leached out and partly entered secondary carbonates and /or water-soluble fractions to result in the increase of $^{87}Sr/^{86}Sr$ ratios in secondary carbonates and water-soluble fractions. The greater the chemical weathering, the higher the $^{87}Sr/^{86}Sr$ ratios of secondary carbonates and water-soluble fractions of eolian dust. Contrarily, there were lower $^{87}Sr/^{86}Sr$ ratios of secondary carbonates and water-soluble fractions of eolian dust.

The loess formed under relatively arid and cold climates. During this period, chemical weathering weakened as the East Asian winter monsoon reigned, and $^{87}Sr/^{86}Sr$ ratios of acid-soluble materials for the loess thus were low. Paleosols developed under relatively warm and humid climates. During this period, chemical weathering strengthened as the summer monsoon prevailed, so paleosols had high $^{87}Sr/^{86}Sr$ ratios of acid-soluble materials. There was different chemical weathering intensity for various loess and paleosol layers. As Fig.3 shows, variations between high and low ratios of Sr isotope in acid-soluble materials in the L_1—S_7 sequence of the Luochuan section were similar to variations of the natural environments Cheng *et al.*[71] derived through calculations with mathmatic methods, indicating a close relationship between variations in chemical weathering intensity and warm-humid degrees of the natural environments. The East Asian summer monsoon was remarkably characterized by both humid and warm climates. Thus, variations in Sr istopoe ratios of acid soluble materials in the L_1-S_7 sequence reflected characteristics of the East Asian summer monsoon evolution with gradual decrease in its intensity from 800 ka.B.P. to the present. In addition, high resolutional Sr isotope data of Yang *et al.*[18] showed that Sr isotope ratios were higher in the loess L_1SS_1 developed during the interstade of the last glacial period during which it was more humid and cooler than in the loess L_1LL_1 and L_1LL_2 formed during the last stage and the early stage of the glacial period respectively during which there was dry and cold climate. Additionally, there were slightly lower Sr isotope ratios of acid-soluble materials in the top of the paleosol S_1 developed during the late period of the last interglacial period with a temperate and humid climate than in the middle and bottom of the paleosol S_1 developed during the early and middle periods of the last interglacial period with a warm and humid climate. These results revealed that Sr isotope ratios in acid-soluble materials could not only distinguish cold-dry climates from humid-warm climates but also discern humid-cool and humid-temperate climates. Due to clear genesis and definite climatic implication, Sr isotope ratio can be used as a good proxy for the East Asian summer monsoon evolution.

5.2 $^{87}Sr/^{86}Sr$ ratios in acid-insoluble materials indicating East Asian winter monsoon variations

Yang *et al.*[17] thought that $^{87}Sr/^{86}Sr$ ratios in acid-insoluble materials indicated chemical weathering intensity of source areas through investigations on the Luochuan section. Chen *et al.*[72] also discussed the possibility that $^{87}Sr/^{86}Sr$ ratios in acid-insoluble materials reflected chemical weathering in source areas of eolian dust. Yang *et al.*[73] determined $^{87}Sr/^{86}Sr$ ratios in the Lingtai section and further recognized that continental chemical weathering intensity had a decreasing trend whereas physical weathering intensity gradually increased since 2.5 MaB.P.

Acid insoluble materials were the residues after secondary calcite was dissolved and free Sr on the surface of clay minerals was leached by 0.5 mol/L acetic acid solution. Chemical weathering of the Chinese Loess Plateau was at an initial stage with only leaching of soluble elements such as Ca and Na. It was possible that very small quantities of silicate fractions were destroyed by chemical weathering. Although these small quantities of silicate fractions destroyed by chemical weathering can obviously affect variations of Sr isotope ratios in acid-soluble materials, they can be neglected relative to silicate fractions in the residues. Thus, the residues can be regarded to inherit $^{87}Sr/^{86}Sr$ ratios of source areas. Experiments demonstrated that different grained fractions after weak acid treatment had distinct $^{87}Sr/^{86}Sr$ ratios (Table 2): the <2μm fraction had higher $^{87}Sr/^{86}Sr$ ratios than other coarser grained fractions. Due to broadly homogeneous compositions of source materials of the Chinese Loess Plateau, $^{87}Sr/^{86}Sr$ ratios in the acid-insoluble materials were primarily controlled by wind sorting. Wind sorting was a main pattern influencing variations in grain sizes of eolian dust. Eolian dust accumulated in the Chinese Loess Plateau had more coarse-grained fractions with relatively little minerals enriched in Rb when wind sorting was stronger, thus $^{87}Sr/^{86}Sr$ ratios in eolian dust were lower. On the contrary, there were higher $^{87}Sr/^{86}Sr$ ratios in eolian dust. The loess formed under relatively arid and cold climates. During this period, there was a reign of the East Asian winter monsoon, wind sorting was strong and initial eolian dust accumulated into loess contained more coarse particle fractions, thus $^{87}Sr/^{86}Sr$ ratios were low. Paleosols were developed under relatively warm and humid climates. During this period, the summer monsoon strengthened, wind sorting was weak and initial eolian dust developed into paleosols contained more fine particle fractions, so paleosols had high $^{87}Sr/^{86}Sr$ ratios of acid-soluble materials. In this sense, $^{87}Sr/^{86}Sr$ ratios of the acid-insoluble materials in the loess -paleosol sequence varied periodically, and further reflected the periodical evolution of the East Asian winter monsoon during the Quaternary (Fig. 4). $^{87}Sr/^{86}Sr$ ratios of the acid-insoluble materials gradually decreased from 2.6 Ma B.P. to the present, indicating that the East Asian winter monsoon intensity gradually increased since the beginning of the Quaternary period, which was in accordance with gradual cooling of the Quaternary climate since the onset of the Quaternary Ice Age. Therefore, $^{87}Sr/^{86}Sr$ ratios of the acid-insoluble materials can be viewed as a good proxy for the East Asian winter monsoon variations.

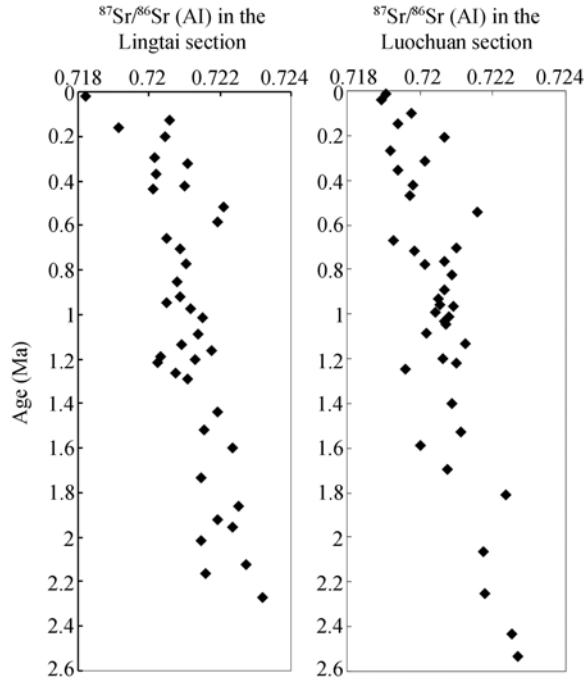

Fig. 4. Variations in $^{87}Sr/^{86}Sr$ ratios of acid-insoluble materials (AI) in the Lingtai and Luochuan sections with time[17, 73].

6 Conclusions

By analyzing Sr-Nd isotope geochemistry of eolian dust from northern China, we draw several conclusions as follows:

(1) Sr isotope ratios of sediments are controlled by their parent rocks, particle sizes and chemical weathering. In general, the higher the Sr isotope ratios of parent rocks, and/or the more the fine-grained fractions, and/or the stronger the chemical weathering, thus the higher the Sr isotope ratios of sediments. On the contrary, there are lower Sr isotope ratios of sediments. Nd isotope ratios of sediments, independent of their particle sizes and chemical weathering, are only associated with parent rocks.

(2) Geographical distributions of Nd isotope in the arid-semiarid areas of China reveal that the Tarim Basin, deserts in the central and west of Inner Mongolia and the Tibetan Plateau are the main sources of the Chinese Loess Plateau, and are also manufacturers for eolian dust of the Far East regions, together with the Chinese Loess Plateau.

(3) Sr isotope ratios in acid-soluble materials of eolian dust recorded information on the East Asian summer monsoon evolution, whereas $^{87}Sr/^{86}Sr$ ratios in

the acid-insoluble materials reflected the variation characteristics of the East Asian winter monsoon.

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