Research article

Provenance of loess deposits and stepwise expansion of the desert environment in NE China since ~1.2 Ma: Evidence from Nd-Sr isotopic composition and grain-size record

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ARTICLE INFO

Keywords:
Eolian deposits
Loess provenance
Nd and Sr isotopes
Grain size
Desert expansion
Aridification

ABSTRACT

The Quaternary wind-blown Chinese loess-paleosol sequence, which developed as a product of the intensification of Northern Hemisphere glacial-interglacial cycles, is a major source of information on aridification of the Asian interior. In northeastern (NE) China, extensive loess deposits occur that have great potential for reconstructing the long-term paleoclimatic evolution of the region. Despite existing geochemical data indicate that the probable major source areas for these loess deposits were the nearby Horqin and Otindag deserts, it is still unclear that whether there was a long-term change in the detrital source of these loess deposits. In addition, the aridity history and long-term desert evolution of NE China are currently poorly understood. In the present study, we measured the silicate Nd and Sr isotopic composition of the 28–45 μm grain-size fraction from multiple loess sections (NYZG, SBH, and TDJZ). The results show that the loess deposits of the three sites were dominantly sourced from the Otindag sandy desert and that this source remained constant for the past 1.2 Ma. Notably, the \({87Sr/86Sr}\) ratios are influenced mainly by variations in the content of clay-sized material present in the form of aggregates or adhering to the surface of coarser particles. Therefore, caution is needed in the use of \({87Sr/86Sr}\) ratios of the 28–45 μm fraction as a tracer of eolian dust, when the clay minerals are not completely removed during the pretreatment process. The grain-size variability of the three sections indicates that stepwise enhancement of aridification in the Otindag sandy desert occurred at 1.2 Ma, 0.9 Ma, and 0.7 Ma, which coincided respectively with the beginning of the Mid-Pleistocene transition (MPT), an extremely cold event at ~0.9 Ma within the MPT, and the establishment of a full glacial climate at the termination of the MPT. This finding supports the hypothesis that the growth of high-latitude ice sheets was the major driver of the long-term aridification of Asian dust source areas.

1. Introduction

Loess is a terrestrial eolian silt deposit which typically forms under an arid or semi-arid climate. It is mainly composed of silt-sized particles, and has a loose and massive structure without sedimentary bedding, and usually contains interbedded paleosols, calcareous nodules and the fossils shells of terrestrial mollusks (Liu, 1985; Pye, 1995; Muhs, 2013). The Cenozoic loess deposits in China typically consist of a Red Clay sequence formed during the late Oligocene-Pliocene (Guo et al., 2002; Lu et al., 2004, 2019; Qiang et al., 2011; Licht et al., 2014; Li et al., 2018), and a loess-paleosol sequence which formed during the Pleistocene (Liu, 1985; Kukla and An, 1989). Chinese loess deposits have been used to determine the timing of the onset and stages in the development of the aridification of the Asian interior (Guo et al., 2002; Ding et al., 2005; Lu et al., 2010), the evolution of the East Asian summer monsoon (EASM) system (An et al., 1990; An, 2000; Ding et al., 1995; Liu and Ding, 1998), and the phased uplift of the Qinghai-Tibetan Plateau (Li and Fang, 1999; An et al., 2001; Guo et al., 2002; Lu et al., 2010).
2004; Fang et al., 2019). With the establishment of a stratigraphical and chronological framework of loess deposits on the Chinese Loess Plateau (CLP), together with the development of modern analytical methods, a substantial amount of research has been conducted on loess provenance (e.g. Stevens et al., 2010, Stevens et al., 2013; Chen and Li, 2011; Nie et al., 2015; Zhang et al., 2015) and on paleoclimatic reconstruction (e.g. Da et al., 2015; Zhou et al., 2014; Yang et al., 2015; Li et al., 2017; Meng et al., 2018).

Northeastern (NE) China is an integral part of the East Asian monsoon region, and it contains extensive and thick loess deposits which represent deposition at the eastern end of the Eurasian loess belt (Fig. 1a) (Zeng et al., 2017). Long and continuous paleoclimatic records from these deposits are extremely scarce, yet there are several reasons why such records would be of high scientific value. First, the loess deposits of NE China are located downwind of the Horqin, Otindag and Hulun Buir sandy deserts (Fig. 1a), and are therefore sensitive to changes in the source area and source area aridity. Thus, loess records from the region can potentially increase our understanding of the spatiotemporal evolution of the aridification of the Asian interior during the late Cenozoic (Ding et al., 2005; Lu et al., 2010).

Identifying loess provenance can help discriminate the sources and transport paths of dust storms in the past and provide a reference for sustainable environmental management. In addition, because many climate proxies available for loess deposits are sensitive to changes in source materials and the dust transport path, discriminating changes in provenance is crucial for their paleoclimatic interpretation (Chen and Li, 2011). Geochemical tracers are the most effective tools for tracing the material sources of loess. They include element composition (Hao et al., 2010; Peng et al., 2016); radiogenic isotopes such as Nd, Sr, Pb, Hf (Sun, 2005; Chen et al., 2007; Chen and Li, 2013; Zhao et al., 2014; Zhang et al., 2015); zircon age distribution (Stevens et al., 2010; Stevens et al., 2013; Che and Li, 2013; Nie et al., 2014; Zhang et al., 2016); and oxygen isotopic composition of quartz grains (Rex et al., 1969; Yan et al., 2014). Among them, Nd-Sr isotopes are the most intensively studied source tracer of eolian dust, and a large amount of

Fig. 1. (a) Distribution of deserts and loess in China and the locations of the study area and the Luochuan (LC), Lingtai (LT) and Zhaojiachuan (ZJC) loess sections. The modern 400 mm annual average isohyet represents the approximate boundary between the arid zone and the East Asian monsoon zone. EASM: East Asian summer monsoon; EAWM: East Asian winter monsoon; ISM: Indian summer monsoon; CLP: Chinese Loess Plateau. (b) Topography of the study region and locations of the investigated sections, shown on the shaded relief map. (c) Spring (March–April–May, MAM) mean 850 hPa streamline and mean wind speed near the surface based on NCEP/NCAR Reanalysis for 1981–2010. (d) Nd-Sr isotopic regions of the Chinese arid zone (Chen et al., 2007; Li et al., 2009). NBC—deserts around the northern boundary of China; NMT—deserts on the northern margin of Tibetan Plateau; OD—Ordos Desert.
data has been accumulated worldwide (e.g., Sun, 2005; Chen et al., 2007; Rao et al., 2011, 2014, 2015; Chen and Li, 2013; Ben-Israel et al., 2015; Zhang et al., 2015; Zhao et al., 2015; Kumar et al., 2018; van der Does et al., 2018; Yang et al., 2017). The results of the Nd-Sr isotopic composition of the < 75 μm and < 5 μm silicate fractions of Chinese desert deposits show that the Nd-Sr isotopic composition was controlled mainly by the tectonic setting of North China, and that materials from the deserts are derived from the erosion of the rocks of the surrounding mountains (Chen et al., 2007). Changes in the Sr, Nd, and Pb isotopic composition of the < 20 μm silicate fraction at the boundary between the Red Clay and the loess-paleosol sequence on the CLP also indicate a significant shift in the dust source material at ~ 2.58 Ma (Sun, 2005). However, the Sr isotopic composition can also be affected by the highly radiogenic 87Sr/86Sr ratios of the clay mineral component (mostly < 2 μm), via the processes of eolian sorting and chemical weathering (Grousset and Biscaye, 2005; Yang et al., 2005). Chen and Li (2011) suggested that the use of the silicate Sr isotopic composition of a specific grain-size fraction (28–45 μm) could minimize grain-size effects on Sr isotopic composition, enabling it to be used as sensitive provenance tracer. Preliminary provenance studies based on bulk silicate Nd-Sr isotopic composition (Li et al., 2009; Chen and Li, 2011) and zircon U-Pb age spectrum analysis (Xie et al., 2012) demonstrate that the probable major source areas for these loess deposits in NE China were the nearby Horqin and Otindag deserts. However, the number of samples used in these previous studies was small and mainly confined to the top of the section. Thus, these previous researches are unable to demonstrate the possible occurrence of changes in sediment source in parallel with glacial-interglacial cycles and whether there was a long-term change in source, as was the case for the CLP (Chen and Li, 2013; Zhang et al., 2015). Zeng et al. (2011, 2016) conducted high-resolution magnetostратigraphic and pedostratigraphic study of three loess-paleosol sections (NYZG, SBH, and TDJZ) in the Chifeng region of Inner Mongolia (Fig. 1a). The present climate is semi-arid and controlled mainly by the tectonic setting of North China, and that materials from the deserts are derived from the erosion of the rocks of the surrounding mountains (Chen et al., 2007). Changes in the Sr, Nd, and Pb isotopic composition of the < 75 μm and < 5 μm silicate fractions of Chinese desert deposits show that the Nd-Sr isotopic composition was controlled mainly by the tectonic setting of North China, and that materials from the deserts are derived from the erosion of the rocks of the surrounding mountains (Chen et al., 2007). Changes in the Sr, Nd, and Pb isotopic composition of the < 20 μm silicate fraction at the boundary between the Red Clay and the loess-paleosol sequence on the CLP also indicate a significant shift in the dust source material at ~ 2.58 Ma (Sun, 2005). However, the Sr isotopic composition can also be affected by the highly radiogenic 87Sr/86Sr ratios of the clay mineral component (mostly < 2 μm), via the processes of eolian sorting and chemical weathering (Grousset and Biscaye, 2005; Yang et al., 2005). Chen and Li (2011) suggested that the use of the silicate Sr isotopic composition of a specific grain-size fraction (28–45 μm) could minimize grain-size effects on Sr isotopic composition, enabling it to be used as sensitive provenance tracer. Preliminary provenance studies based on bulk silicate Nd-Sr isotopic composition (Li et al., 2009; Chen and Li, 2011) and zircon U-Pb age spectrum analysis (Xie et al., 2012) demonstrate that the probable major source areas for these loess deposits in NE China were the nearby Horqin and Otindag deserts. However, the number of samples used in these previous studies was small and mainly confined to the top of the section. Thus, these previous researches are unable to demonstrate the possible occurrence of changes in sediment source in parallel with glacial-interglacial cycles and whether there was a long-term change in source, as was the case for the CLP (Chen and Li, 2013; Zhang et al., 2015). Zeng et al. (2011, 2016) conducted high-resolution magnetostратigraphic and pedostratigraphic study of three loess-paleosol sections (NYZG, SBH, and TDJZ) in the Chifeng region of Inner Mongolia (Fig. 1a). The present climate is semi-arid and controlled mainly by the dynamics of the East Asian Monsoon circulation, characterized by cold/dry winters and warm/wet summers. In winter the

2. Sampling and methods

2.1. Environmental setting and sampling

The thickest and most representative eolian loess deposits in NE China are distributed in the Chifeng region, in southeastern Inner Mongolia (Fig. 1a). The present climate is semi-arid and controlled mainly by the dynamics of the East Asian Monsoon circulation, characterized by cold/dry winters and warm/wet summers. In winter the
prevailing wind direction is northwesterly, generated by the Siberian–Mongolian high-pressure zone; while in summer the dominant direction is southeasterly, driven by the pressure gradient between the Subtropical High and the Asian Continental Low. The mean annual precipitation ranges from 600 mm in the southeast to 200 mm in the northwest, and most of the precipitation falls during the boreal summer. As illustrated in Fig. 1c, the Horqin and Otingdar sandy desert are located to the north and west of the Chifeng region, respectively.

Previous studies (Zeng et al., 2011, 2016; Yi et al., 2015, 2016) have reported the results of investigations of magnetostratigraphy (Fig. 2), magnetic susceptibility (MS), pedostratigraphy, and optically stimulated luminescence (OSL) dating of three sections from the Chifeng region: NYZG (41° 55′N, 118° 43′E, 774 m.a.s.l.), SBH (42° 18′ N, 118° 41′E, 677 m.a.s.l.) and TDJZ (42° 7′ N, 119° 0′ E, 766 m.a.s.l.). In addition, the results of grain-size analysis of the NYZG section are given in Zeng et al. (2017). These sections are located in the southwestern part of the Chifeng area (Fig. 1c), in a landscape characterized by hills composed of bedrock and valleys mantled by eolian loess deposits. The NYZG and SBH sections consist of a brickyard exposure and an underlying excavated pit, while the TDJZ section is a natural outcrop in a loess gully. The lithology of these sections can be roughly divided into two parts: an upper part consisting mainly of light brown sandy-silt loess layers interbedded with light reddish-brown paleosols or weakly-developed paleosols; and a lower part which exhibits stronger pedogenesis, with both the colour of the loess and paleosol layers becoming progressively redder with depth and with the colour contrast between them becoming weaker (Fig. 2). In addition, the texture of the lower part becomes a more homogeneous silt.

In this study, a total of 1895 bulk samples were taken at 5-cm intervals from the SBH and TDJZ sections for grain-size measurements. In addition, 23 samples were collected from the NYZG section, 15 samples from the TDJZ section, and 6 samples from the SBH section, which were used for Nd-Sr isotopic measurements of the 28–45 μm silicate fraction; the sampling was based on the loess and paleosol unit alternations (Fig. 2). It was hoped that this sampling strategy would identify possible changes in eolian dust sources on both the glacial-interglacial scale, as well as a possible long-term source shift over the last 1.2 Ma. In addition, in order to assess possible grain-size effects on the 87Sr/86Sr ratios of the 28–45 μm silicate fraction, this fraction was also used for grain-size and microscopic analysis.

2.2. Extraction of silicate fractions and Nd-Sr isotopic measurements

Primary soil/paleosol particles generally occur as bonded aggregates, and organic matter and secondary carbonates are two important non-silicate types of cement that form soil aggregates. Thus, in order to obtain dispersed mineral particles, the organic matter and carbonate content of samples was removed using 10% H2O2 and 10% HCl, respectively, followed by dispersal with an ultrasonic vibrator for about 10 min to further disperse the particles, the samples were sieved to isolate the 28–45 μm silicate fraction, the sampling was based on the loess and paleosol unit alternations (Fig. 2). It was hoped that this sampling strategy would identify possible changes in eolian dust sources on both the glacial-interglacial scale, as well as a possible long-term source shift over the last 1.2 Ma. In addition, in order to assess possible grain-size effects on the 87Sr/86Sr ratios of the 28–45 μm silicate fraction, this fraction was also used for grain-size and microscopic analysis.

High-resolution magnetostratigraphic results suggest that the basal age of the NYZG section is ~1.02 Ma, the basal age of the SBH section is ~1.02 Ma, and the top and basal ages of the TDJZ loess sequence are ~0.63 and ~1.22 Ma, respectively (Zeng et al., 2016). The TDJZ section represents the oldest known loess deposits in NE China. The chronology of the NYZG section was reported by Zeng et al. (2017). In the present study, the chronology of the SBH and TDJZ sections were established using the same approach as for the NYZG section: the age control points were derived from the correlation of Chinese loess-paleosol sequences with the benthic δ18O record (Ding et al., 1994; Lu et al., 1999; Harol et al., 2000; Wu et al., 2005; Hao et al., 2012). The age control points the NYZG, SBH and TDJZ sections are shown in Fig. S1-S3. A grain-size sedimentation-rate age model was then used to interpolate the age at each sampling level between these age control points (Porter and An, 1995).

Fig. 3 shows that the MS time series for the three sections in NE China are consistently in-phase with the CLP records as well as with the benthic δ18O stack (Lisiecki and Raymo, 2005); in addition, the overall structures of the MS curves are well correlated on a glacial-interglacial timescale. This suggests that our chronology for these sections is valid on orbital timescales and that it can be used for comparison with other palaeoclimatic records.

3. Results and discussion

3.1. Constancy of the provenance of loess deposits from NE China since 1.2 Ma

A previous Nd-Sr isotopic investigation has been conducted of the < 75 μm silicate fraction of surface materials (including eolian and fluvial sand) from all the deserts in North China (Chen et al., 2007; Li et al., 2009). The results indicated that these deserts could be classified
into three isotopic regions (Fig. 1d, Fig. 4), which are controlled by the tectonic setting and the mountain ranges in North China (Chen et al., 2007). The regions are: (1) deserts around the northern boundary of China (NBC), which have the highest $\varepsilon_{\text{Nd}}$ values and the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratios; (2) deserts on the northern margin of the Tibetan Plateau (NMTP), with moderate $\varepsilon_{\text{Nd}}$ values and the highest $^{87}\text{Sr}/^{86}\text{Sr}$ ratios; and (3) deserts on the Ordos Plateau (OD), with the lowest $\varepsilon_{\text{Nd}}$ values and moderate $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. As shown in Fig. 4, the loess deposits (28–45-μm fraction) from the three sections in NE China have high $\varepsilon_{\text{Nd}}$ values with the narrow range of −9.7 to −7.0, which is consistent with the results of the bulk silicate fraction (Li et al., 2009). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are relatively low, with a slightly larger range of variation (0.710–0.715) than the bulk silicate fraction (Li et al., 2009).

In general, the Nd-Sr isotopic composition of our samples falls within the range of the isotopic values characteristic of the NBC, which is distinctly different from that of the loess deposits of the CLP. It has been suggested that the Sr isotopic composition can be affected by the highly radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the clay minerals (Yang et al., 2005). Thus, the Nd-Sr isotopic comparison between < 75 μm silicate fraction of surface materials from all of the deserts in North China and the 28–45 μm fraction of loess 28–45 μm fraction from NE China could be potentially influenced by the difference of clay mineral content. However, it is noteworthy that the Nd-Sr isotopic comparison is not significantly influenced by the clay mineral content (Fig. 4). We infer that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the desert materials would have decreased after removal of the clay-sized component by the eolian sorting process, and therefore the loess samples still fall within the range of the isotopic region of the NBC. Part of the Nd-Sr isotopic distribution is located outside the range of the potential sources, possibly because of grain-size effects on the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the loess samples used in this study, or because of the inadequate sample size used for classification of potential sources. Thus, the loess deposits from the three sections are characterized by a high $\varepsilon_{\text{Nd}}$–low $^{87}\text{Sr}/^{86}\text{Sr}$ signature, indicating the dominant contribution of dust from the isotopic region of the NBC. The NBC consists of the Gurbantunggut, Hunlun Buir, Otindag, and Horqin sandy deserts, which are located in the Central Asian Orogenic Belt (CAOB), with the highest $\varepsilon_{\text{Nd}}$ values resulting from the widely distributed paleo-ocean crust and basalts (Buchan et al., 2002). However, it is noteworthy that the $\varepsilon_{\text{Nd}}$ values of the loess deposits in NE China are concentrated at the low-value end of the NBC distribution, and mainly fall within the range of the Otindag sandy desert (Fig. 4b). Detrital zircon U-Pb age spectra and Hf isotopic composition analysis indicated that the sand in the Otindag sandy desert was derived from both the CAOB and the mountains of the North China Craton (NCC) which have the lowest $\varepsilon_{\text{Nd}}$ values (Xie et al., 2007). Thus, the $\varepsilon_{\text{Nd}}$ values of sand in the Otindag and Horqin sandy deserts are substantially lower than those of the Hunlun Buir and Gurbantunggut sandy desert which are completely surrounded by the CAOB (Chen and Li, 2011). Therefore, it can be concluded that the loess deposits of the NYZG, SBH, and TDZJ sections are derived from the Otindag sandy desert, which is located a relatively short distance upwind. Moreover, the modern spring (March–April–May) mean 850 hPa streamlines and mean wind-speed distribution (Fig. 1b) also indicate that the prevailing wind direction in the Chifeng area is northwesterly, and that the Otindag sandy desert has one of the most vigorous wind regimes in China, which favors dust generation and transportation by spring dust storms. Thus, our results provide new evidence for the hypothesis that the Chinese loess deposits are mostly derived from the adjacent deserts on the upper wind direction with a short distance of transportation (Chen and Li, 2011).

The Nd-Sr isotopic compositions of the three sections, plotted on the refined timescale, are illustrated in Fig. 5. There is a negative correlation between $\varepsilon_{\text{Nd}}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ for the loess deposits of NE China; however, there is no significant correlation between variations in Nd-Sr isotopic composition and loess-paleosol alternations, which suggests...
Fig. 4. (a) Comparison of the Nd-Sr isotopic composition of the NYZG, SBH and TDJZ sections with the isotopic ranges of potential sources, desert sand, loess deposits of the Chinese Loess Plateau (ZJC section) and sites from NE China analyzed in a previous study (a). NBC—deserts around the northern boundary of China; NMTP—deserts on the northern margin of the Tibetan Plateau; OD—Ordos Desert. (b) Comparison of the Nd-Sr isotopic composition of loess deposits from NE China and desert sand of the NBC (Gurbantunggut, Hunlun Buir, Otindag, and Horqin sandy deserts). The loess samples in this study are divided into two intervals based on the obvious differences of the grain-size variability before and after 700 ka, illustrated in Fig. 5. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 5. Magnetic susceptibility (MS), grain-size > 63 μm and Nd-Sr isotopic composition of the NYZG, SBH and TDJZ sections (using the refined chronology). Light-green horizontal bars indicate major paleosol layers. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
that there are no significant shifts in dust source on a glacial-interglacial scale. Thus, we propose that the observed irregular fluctuations in Nd-Sr isotopic composition on the glacial-interglacial scale may reflect the heterogeneity of the source materials in the Otindag sandy desert rather than changes in dust source. In addition, the $^{87}$Sr/$^{46}$Sr variability is characterized by a generally long-term decreasing trend since 1.2 Ma, with an abrupt decrease at 0.7 Ma, which is in accord with the grain-size analysis results of the loess deposits (Fig. 4b, Fig. 5). Unlike the Sr isotopes record, the eNd record shows no obvious trend and abrupt decrease at 0.7 Ma (Fig. 4b, Fig. 5). It has been suggested that silicate Nd isotopes are a reliable source tracer which is little affected by grain-size sorting (Grousson and Biscaye, 2005). Thus, we raise a possibility that the $^{87}$Sr/$^{46}$Sr ratios of the 28–45 μm fraction might have been influenced by the grain-size sorting. This seems to be true, especially our Nd isotope results demonstrated that the dust source of the loess deposits in the Otindag sandy desert has remained constant since at least 1.2 Ma, implying that the prevailing wind direction and the tectonic-topographic pattern has remained unchanged in the Otindag sandy desert during this interval.

3.2. Possible grain-size effect on the $^{87}$Sr/$^{46}$Sr ratios of the extracted silicate fraction

Due to the extremely high $^{87}$Sr/$^{46}$Sr ratios of clay minerals (which are enriched in the < 2 μm fraction), it has been suggested that grain-size sorting is the dominant control on the $^{87}$Sr/$^{46}$Sr isotopic ratios of loess deposits (Yang et al., 2005). Thus, the Sr isotopic composition of bulk silicate samples is mainly controlled by the size of the < 2 μm fraction or by the proportion of clay minerals (Chen et al., 2007). Chen and Li (2013) suggested that measuring the silicate Sr isotopic composition of the 28–45 μm fraction of loess deposits can eliminate the influence of grain-size sorting. However, the $^{87}$Sr/$^{46}$Sr ratio of the 28–45 μm grain-size fraction in our study indicates a grain-size effect. As illustrated in Fig. 6, the Sr isotopic composition of the 28–45 μm fraction generally tracks the variations of the clay content of the 28–45 μm fraction extracted by wet sieving, and there is a strong positive correlation between them. The grain-size distribution curves also support the occurrence of clay-sized particles in the 28–45 μm fraction, indicated by the presence of peaks at the fine end of the curves (Fig. 6e). Thus, the presence of clay minerals within the 28–45 μm fraction is the likely primary cause of the observed grain-size effect on the $^{87}$Sr/$^{46}$Sr ratios. SEM observations showed that clay-sized particles were present as aggregates or adhering to the surface of quartz and/or feldspar grains (Fig. 7). We therefore propose that pretreatment using an ultrasonic vibrator alone, without chemical dispersants such as (NaPO₃)₆, has failed to ensure the complete dispersion of the samples, due to flocculation and adhesion of clay-sized particles. Although a few samples contained only a few clay-sized particles after wet sieving, the clay content of the extracted 28–45 μm fraction is generally related to the clay-sized content of the bulk samples (Fig. 6f). In addition, it is noteworthy that the clay-sized content of several samples actually increased after wet sieving. Thus, a more effective approach for extracting the 28–45 μm fraction should be developed, and caution is needed in inferring source changes of loess deposits based on the Sr isotopic composition of the 28–45 μm fraction, in the absence of Nd isotopic evidence. However, although the Sr isotopic compositions in this study are affected by grain size, the range of Sr isotopic composition has not been significantly altered, most of which still lies within the isotopic ranges of the potential sources (Fig. 4).

3.3. Stepwise expansion of the Otindag sandy desert during the MPT and linkage to the growth of high-latitude ice sheets

3.3.1. Evidence for the desert migrations from the grain-size variability

As is evident in Fig. 5, the grain size (% > 63 μm) of the NYZG, SBH and TDJZ sections is related to their distance from the Otindag sandy desert: The SBH section is the closest to the southeastern margin of the Otindag sandy desert, and its grain size is the coarsest; and the TDJZ and NYZG sections are located to the southeast of the SBH section, and their grain size is similar during the interval of 1.0–0.7 Ma. The grain-size variability of both the SBH and NYZG sections exhibits a long-term coarsening trend with a substantial coarsening event at 0.7 Ma. In addition, the SBH section exhibits a less substantial coarsening event at 0.9 Ma. Previous research suggested that the variation of the sand fraction (% > 63-μm) in loess deposits near the desert margin was sensitive to the expansion and contraction of areas of mobile sand dunes in northern China (Ding et al., 2005). Since our results of Nd-Sr isotopic composition demonstrate that the dust source of the loess deposits in the study area dominantly sourced from the Otindag sandy desert and that this source remained constant for the past 1.2 Ma. (see Section 3.1, above), implying that the sand-sized fraction in the loess deposits of NE China is a useful proxy of the migration of the desert margin and of wet-dry climatic oscillations in the Otindag sandy desert (Zeng et al., 2017). The grain-size variability of the three sections indicates that the stepwise enhancement of aridification in the Otindag sandy desert since 1.2 Ma can be divided into two major intervals: Interval I: 1.2–0.7 Ma, and Interval II: 0.7–0 Ma, which are separated by a substantial drying event at 0.7 Ma. Since the SBH section is much closer to the margin of the Otindag sandy desert, its grain-size should be more sensitive to the migration of the desert margin and therefore it is more likely to have recorded the low amplitude drying event at 0.9 Ma.

3.3.2. Possible causes of the stepwise expansion of the Otindag sandy during the MPT

The marine oxygen isotope record ($δ^{18}$O) is a proxy for changes in global ice volume (Zachos et al., 2001). Spectral analysis of the marine $δ^{18}$O record suggests that global ice volume dynamics underwent a fundamental change between 1.25 and 0.7 Ma, namely the mid-Pleistocene transition (MPT), during which the dominant periodicity changed from 41 kyr to 100 kyr, and enhanced significantly after 0.7 Ma (Fig. 8a) (Ruddiman et al., 1986; Clark et al., 2006; Song et al., 2014; Chalk et al., 2017; Wang et al., 2017; Sun et al., 2019). Despite certain discrepancies occur particularly within some thin loess-paleosol units before 0.7 Ma, the grain-size time series (% > 63 μm) of the NYZG, SBH and TDJZ sections for the major units are readily matched from one section to another at their overlapping parts (Fig. 5). Thus, in order to examine periodicities in the frequency domain of the loess record in NE China, it is crucial to stack a continuous and complete grain-size record since 1.2 Ma, owing to the different age of the three sections. The composite grain-size record was obtained following the procedures by Ding et al., (2002). Notably, both the rhythms and amplitude of stacked grain-size record are almost identical to the LR04 benthic $δ^{18}$O stack (Fig. 8c) (Lisiecki and Raymo, 2005). The wavelet transforms of grain-size record were performed using MATLAB software package to examine the variation of frequency space (Gristed et al., 2004). The wavelet spectra of the stacked grain-size record and the LR04 benthic $δ^{18}$O stack (Lisiecki and Raymo, 2005) are highly coherent, both displaying a mixing of 23-, 41- and 100- kyr cycles since 1.2 Ma, and exhibiting a continuous and significant 100- kyr periodicity after ~0.7 Ma. Notably, both frequency variability of the two records experienced a 100-kyr lull from ~1.0 Ma to ~0.7 Ma, which is a marked feature of the MPT (Clark et al., 2006). The oxygen isotope records from Chinese caves have demonstrated that the Asian monsoon over the past 0.64 Ma exhibited its dominant periodicity in precession cycles (23- kyr), which indicates that the insolation changes caused by the Earth’s precession drove the low-latitude hydrological changes (Cheng et al., 2016). Thus, the timing and duration of the MPT are similar between grain-size record of the loess deposits in NE China and benthic $δ^{18}$O records, same transitions in the frequency domain point to unique mechanisms governing the desert migration in NE China responses to the ice forcing in high latitude, rather than the summer monsoon influenced by the low-latitude hydrological changes.
Numerous studies (e.g., Clark et al., 2006; Sosdian and Hodell et al., 2008; Sosdian and Rosenthal, 2009) have shown that the MPT (1.25–0.7 Ma) experienced the most significant enhancement of the global ice volume, and the maximum volume of glaciations was established at the same time as the onset of the dominant 100-kyr cyclicity after the termination of the MPT at ~0.7 Ma. Moreover, the ice-rafted detritus (IRD) record (Fig. 9) of the North Atlantic indicates the volume of the Laurentide ice sheet exceeded a critical threshold with onset of the Heinrich events during MIS 16 (~0.64 Ma) (Hodell et al., 2008). Thus, the oldest loess deposits in NE China dated at ca.1.22 Ma based on magnetostratigraphy of the TDJZ section was roughly coincident with the beginning of the MPT which corresponds to the onset of aridification in this region (Zeng et al., 2016), and the substantial drying event at 0.7 Ma of the NYZG and SBH section was synchronous with end of the MPT (Fig. 5). In addition to the two major events of 1.25 Ma and 0.7 Ma, some paleoclimatic records suggest that 0.9 Ma within the MPT marked a major climatic event. The benthic δ¹⁸O records (e.g., ODP 1123) of the Pacific indicate that the maximum ice sheet extent was established during MIS 22 (~0.9 Ma) (Fig. 9), which was mainly related to an abrupt increase in the extent of the Antarctic ice sheet (Elderfield et al., 2012). In addition, MIS 24–22 are characterized by an extreme cooling event in most SST records (McClymont et al., 2013). Thus, the low amplitude drying event of the SBH section at 0.9 Ma was well coincident with the abrupt increase in the Antarctic ice sheet during MIS 22. Moreover, it is noteworthy that the grain-size record of the loess deposits in NE China generally exhibits a drying trend during MPT, which coincided with increased ice volume in the benthic δ¹⁸O record (Fig. 9). Thus, the abrupt drying events and drying
Fig. 7. Scanning Electron Microscope (SEM) images showing aggregations of clay-sized particles (a, b) and clay-sized particles adhering to the surface of quartz or feldspar grains (c, d) of the extracted 28–45 μm fraction. The sample names denote the sampling depth of the sections (in cm).

Fig. 8. Wavelet power spectra (Grinsted et al., 2004). (a) LR04 benthic δ¹⁸O stack (Lisiecki and Raymo, 2005); (b) stacked grain-size record of the loess deposits in NE China; (c) time series of the two signals (each normalized to the interval of [0 1]) over the past 1.2 Ma. The thick black contour designates the 5% significance level against red noise, and the cones of influence (COI) that are influenced by edge effects are shown as lighter shading at the beginning and end of the frequency-time plane. The grain-size records of the NYZG, SBH and TDJZ sections were stacked according to the procedures of Ding et al. (2002). The black dashed line indicates the onset of dominant 100-kyr cycles at 0.7 Ma. MPT: mid-Pleistocene transition (1.25–0.7 Ma). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
trend recorded in the loess deposits of NE China are both well correlated with the increase in high-latitude ice volume during the MPT (Fig. 9), which is consistent with the hypothesis that cooling and ice volume expansion at high latitudes was the driver of long-term stepwise aridification of the Asian interior during the late Cenozoic (Ding et al., 2005; Lu et al., 2010; Lu and Guo, 2014; Zeng et al., 2017). The numerical simulations have provided insight into the forcing mechanism of the enhanced aridity in Northern China linkage to the growth of high-latitude ice sheets. The latest experiments using the Community Atmosphere Model version (CAM4) have connoted that global cooling and ice sheet expansion can reduce annual precipitation in Northern China, resulting in enhanced aridity (Zhang et al., 2017). More specifically, the expansion of North American and European ice sheets can change the existing trough and ridge system in the Northern Hemisphere, leading to anomalous cyclone to the south of the European ice sheet in winter, which could result in strengthened westerly and northwesterly winds over inland China in winter (Zhang et al., 2017). As a result, the aridification and stronger winds could cause the large-scale expansion of desert and eventually entrain more and coarser dust particles which were deposited downwind (Ding et al., 2005; Lu et al., 2010; Zeng et al., 2017).

In summary, the large expansion of the Eurasian loess belt at ~1.2 Ma coincided with the beginning of the MPT (Zeng et al., 2017), during which global cooling and the growth of high-latitude ice sheets may have caused the aridification of the Asian interior to exceed a critical threshold, and promoted the extension of the desert belt to NE China. Thus, the Otindag sandy desert probably formed during this critical threshold, and promoted the extension of the desert belt to NE China during MPT (~1.25–0.7 Ma). The dashed lines in the records of LR04 and ODP1123 indicate the significant enhancement of the global ice volume at ~0.9 Ma and ~0.7 Ma, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

4. Conclusions

(1) The silicate Nd and Sr isotopic composition of the 28–45 μm grain-size fraction of the NYZG, TDJZ and SBH loess sections in NE China reveals that the sediments are derived directly from the Otindag sandy desert located a short distance upstream. In addition, the source of the loess deposits in NE China has remained constant since 1.2 Ma, and the prevailing wind direction and the tectonic-topographic configuration of the Otindag sandy desert during the past 1.2 Ma were unchanged.

(2) Unexpectedly, the 87Sr/86Sr ratios of the 28–45 μm fraction are influenced by a grain-size effect, and clay particles present as aggregates or adhering to the surface of larger particles are likely the main cause. This finding shows that caution is needed when using 87Sr/86Sr ratios of the 28–45 μm fraction to trace the source of loess deposits.

(3) The grain-size records of the three loess sections in NE China reveal the stepwise enhancement of aridification in the Otindag sandy desert at 1.2 Ma, 0.9 Ma, and 0.7 Ma. The drying trend and abrupt drying events recorded in the loess deposits of NE China are strikingly correlated to high-latitude ice volume increases during the MPT, which supports the hypothesis that cooling and ice volume expansion in high latitudes during the late Cenozoic were the major drivers of the long-term stepwise aridification of the Asian interior. The stepwise expansion of the Otindag sandy desert at ~1.2 Ma, 0.9 Ma and 0.7 Ma coincided with the beginning of the MPT, an extremely cold event at ~0.9 Ma within the MPT, and the establishment of a full glacial climate at the termination of the MPT.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.gloplacha.2019.103087.
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