



Title	Reconstruction of temporal variation of Asian dust provenances in silt and clay fractions in Japan Sea sediments since 10 Ma based on a quantification of each source contribution using a statistical end-member decomposition of powder X-ray diffraction profiles
Author(s)	李, 梅花 Alison
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Reconstruction of temporal variation of Asian dust provenances
in silt and clay fractions in Japan Sea sediments since 10 Ma
based on a quantification of each source contribution
using a statistical end-member decomposition of powder X-ray diffraction profiles
(日本海堆積物の粒度別粉末X線回折プロファイルの
統計的端成分分解に基づくアジア起源風成塵供給源別定量と
その時代変化の復元)

北海道大学大学院環境科学院
地球圏科学専攻

李 梅花

Abstract

Japan sea sediments consist of various detrital materials of eolian and riverine origin.

Understanding the provenance of eolian dust is vital for reconstructing the variability of wind patterns and monsoons in the past. A total of 180 samples were collected from the Hole 1425D, which had 370 m in length and 9.69 Ma geological age. Grain size separation was conducted to evaluate the mineral composition in silt ($>4 \mu\text{m}$) and clay ($<4 \mu\text{m}$) fractions by X-ray diffractometer (XRD). The mineral composition suggests the source variabilities in silt and clay fractions, which could be interpreted as provenance shifts occurring in 8 and 2.7 Ma.

Parallel factor analysis (PARAFAC) was applied to decompose X-ray diffractograms into individual subcomponents to identify the sources and quantify their contributions. A six-end-member PARAFAC model was established, and 3 Asian dust sources, Taklimakan (EM1), Gobi (EM2) and Ordos (EM5); a riverine source from the Japanese islands (EM3); and two biogenic sources, diatomaceous (EM4) and Opal-CT (EM6), were identified, and their contributions were quantified. Based on the provenance unmixing by PARAFAC, new and complete eolian accumulation rates from Taklimakan, Gobi and Ordos were reconstructed at Integrated Ocean Drilling Program (IODP) Site U1425 in the Japan Sea. In the late Miocene, the high mass accumulation rate (MAR) of riverine input from the Japan Islands reflected a strong summer monsoon during 9.7-8 Ma. Provenance shift occurred between 8 and 7 Ma from riverine source to eolian for a short period, which implied that Asian aridification was intensified at about 8 Ma, coinciding with the late Miocene cooling. Eolian flux

increased dramatically in Pleistocene to the maximum of 3.1 g/cm²/kyr, and the Gobi Desert, Taklimakan Desert and Ordos Plateau were the three main contributors of eolian flux to the Japan Sea.

The contribution of Taklimakan source to the Japan Sea ranged from 10 to 50% and the dust flux increased gradually in the Pleistocene to the highest of 1.67 g/cm²/kyr at 0.29 Ma. Lower flux occurred in the Late Miocene to Pliocene of <0.5 g/cm²/kyr. MAR of Taklimakan eolian in silt fraction was generally higher than clay fraction. The contribution of Gobi dust to the Japan Sea has ranged from 20-60% since 10 Ma. The highest MAR of 1.7 g/cm²/kyr occurred in 0.96 Ma and the lowest in Pliocene. MAR of Gobi dust in clay fraction was higher than silt fraction from 9.24 to 0.73 Ma. The results show that Taklimakan dust is dominated by the silt fraction, while Gobi dust is dominated by the clay fraction, and they are controlled by the relative contributions of different pathways of dust transport, such as the westerly winds and East Asian winter monsoon (EAWM).

Clay-size dust from the Gobi Desert increased during three periods, late Miocene global cooling (LMGC), intensification of Northern Hemisphere Glaciation (iNHG), and mid-Pleistocene Transition (MPT), which reflected increased EAWM winds associated with global cooling and glaciation. Taklimakan became the major dust contributor to the Japan Sea sediments during the warmer climate periods in the latest Miocene to early Pliocene and the Late Pleistocene, where westerly wind activity dominated eolian transport. Dust from the Ordos Plateau increased greatly from 0.95-0.85 Ma due to enhanced Asian aridification since 2 Ma in Northwest China.

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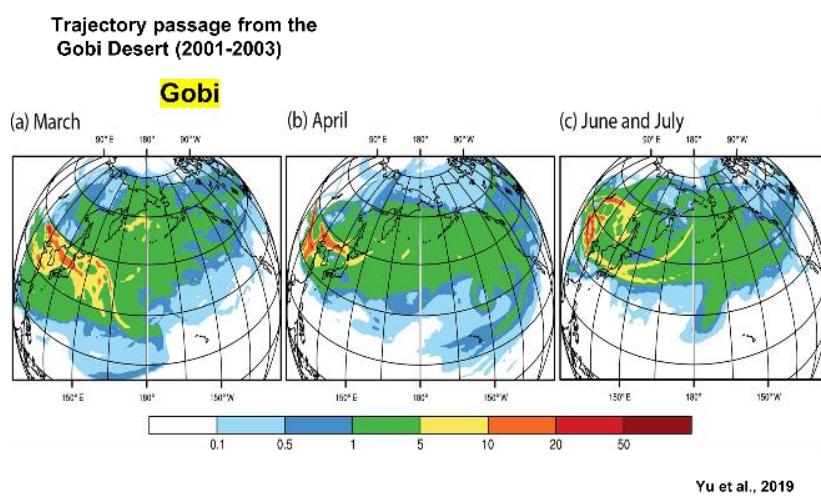
Chapter 1. Introduction

Dust is transported by wind as a large-scale movement of air which is sensitive to global climate changes. Eolian sedimentation in the North Pacific Ocean has been studied to evaluate the change in Earth's climate and wind circulation in the Cenozoic (e.g., LL44-GPC-3, Ocean Drilling Program (ODP) Sites 885/886 and 576/578; Leinen and Heath, 1981; Janecek, 1984). Westerlies and East Asian monsoons have prevailed in Asia since the early Miocene (Sun and Wang, 2005; Guo et al., 2008; Caves et al., 2015; Tada et al., 2016), in which East Asian summer monsoon (EASM) precipitation controls riverine detritus input to marine sediments and dust availability in desert areas, while the East Asian winter monsoon (EAWM) and westerly winds control dust transportation from Asian desert sources to marine sinks. Major climate changes in the late Cenozoic were accompanied by pronounced changes in the pattern and intensity of atmospheric circulation (Lamb and Woodroffe, 1970). An increase in ice sheets during the Northern Hemisphere Glaciation intensified the EAWM (Ding et al., 1995) in northern China. The latitudinal position of westerlies migrated equatorward by ~3-5° during the Last Glacial Maximum (Ono and Irino, 2004), while it was weakened and more poleward during the warm Pliocene (Abell et al., 2021). Currently, westerlies are migrating poleward in response to global warming (Yang et al., 2020). Accordingly, the interplay and response of atmospheric circulation to global climate changes are important factors for predicting future climate, and the reconstruction of past flux and grain size of dust accumulated in the downwind areas from inland deserts to the East

Asian region could allow us to understand the mechanisms modulating activities of westerlies and EAWM winds.

1.1 Eolian transportation paths at present

According to the 40 years of meteorological observation report in China, there are two dominant regions of dust storm events in the Central Asia deserts (Sun et al., 2001). One is the Taklimakan Desert in the Tarim Basin in western China and the other is Gobi deserts in southern Mongolia and the adjoining deserts in northern China. The two regions are under two different wind circulation systems prevailing in Eurasia (Sun 2008). In the northern Asia of Gobi and Mongolia areas, the surface wind system is mainly controlled by the pressure gradient between the Siberia-Mongolian High, which results in strong northwest winds (Winter Monsoon) in the spring season (Sun et al., 2001). Asian dust is carried by this low-level wind to a medium distance of 500-3,000 km of the Chinese Loess Plateau, Korea, Japan and SE China (Figure 1.11).



(The trajectory passage frequency: number of trajectories crossing over each 1° latitude × 1° longitude grid divided by the total number of trajectories from the sources in each month)

Multangle Imaging Spectro Radiometer (MISR) (>2km)

FIGURE 1.11 Trajectory passage of dust from the Gobi Desert (2001–2003) (Yu et al., 2019).

On the other hand, in the middle to high topography of Tarim and Qaidam basins, the atmospheric circulation is high-level Westerly Jet dominated (Sun et al., 2001). Westerly winds are responsible for the long-range transport dust (>5000 km) which crosses over the Japan Sea, North Pacific Ocean and America (Sun et al., 2001) (Figure 1.12). The peak of dust activation and transportation in spring over the Gobi Desert and in both spring and summer over the Taklamakan Desert.

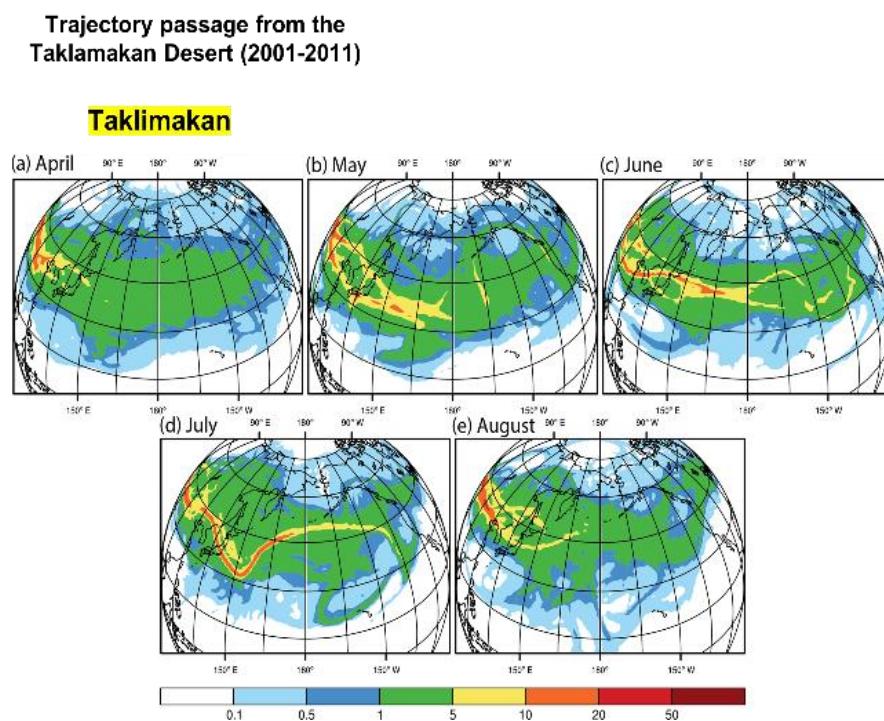


FIGURE 1.12 Trajectory passage of dust from the Taklimakan desert (2001–2003) (Yu et al., 2019).

1.2 Previous studies of eolian dust

Composition and flux of dust components could record the dust transportation mode and contribution from desert source materials, which could be well preserved in the sediments of the Japan Sea located at the eastern edge of East Asia on the main pathway of westerlies and under the influence of monsoon climate. Previous studies conducted at ODP Site 797 and IODP Site U1430 (Irino and Tada, 2000; Shen et al.,

2017; Anderson et al., 2020) in the Japan Sea presented the history of dust provenance change from the late Miocene to Quaternary. Although the relative contributions of dust to the Japan Sea sediments from different regions of Asian deserts have been intensely investigated, their association with the wind circulations of westerly, EAWM and the EASM are still poorly understood. It also remains unclear how the EASM and aridity have evolved over time in the inland deserts. The long-term aridification of inland Asia might have been associated with an uplift of the Himalayan-Tibetan complex in the Cenozoic (Guo et al., 2008; Zhang et al., 2022), while the aridity change and evolution of EASM and EAWM, as well as the latitudinal position of the westerly jet during the late Cenozoic are still controversial (Shen et al., 2017; Fan et al., 2006; Yang et al., 2017a; Yang Y. et al., 2021).

1.3 Objective

Examination of temporal and spatial variations of eolian deposits from different dust source regions could provide critical information on the response of westerlies and monsoonal fluctuations to the global climate change in late Cenozoic and the potential driving factors for these consequences, which are potentially relevant to our future climate change. Numerous Asian dust provenance studies are primarily based on the comparison of mineralogy (Leinen and Heath, 1981; Arnold et al., 1995; Biscaye, 1965) which can effectively reveal the mineral composition of parent rock in the dust sources region that controls the relative proportions of different minerals in the material carried by the wind. The mineralogical change in a specific eolian source region occurs over tens of millions of years time scale (Leinen and Heath, 1981; Prospero and Bonatti,

1969) providing a stable and significant feature to reconstruct the past atmospheric activity. Although the mineralogical composition is an effective proxy for eolian provenance study, the precise quantification of contribution from different sources is still challenging.

In this paper, we tried to solve the problems in quantification of dust contribution from different sources, their grain size dependences during the transport process, and climatic influences on dust sources and the transportation processes. For these purposes, parallel factor (PARAFAC) analysis was applied to mineral datasets in the silt and clay fractions of Japan Sea sediments to identify dust subcomponents from the complex mixture of materials, which enabled us to distinguish the variability of effects from the atmospheric circulation of westerlies, EAWM, and aridity changes in the dust source region for the last 10 million years.

Chapter 2. Site setting, stratigraphy, and sample selection

2.1 Integrated Ocean Drilling Program (IODP) Site U1425

A total of 180 samples were collected from IODP Site U1425 ($39^{\circ}29.44'N$, $134^{\circ}26.55'E$) (Figure 2.11), which is situated on the terrace of the Yamato Bank in the center of the Japan Sea. The higher topographic setting of the site minimizes the influence of turbidites, and slow sedimentation is ideal for detecting eolian dust (Tada et al., 2015).

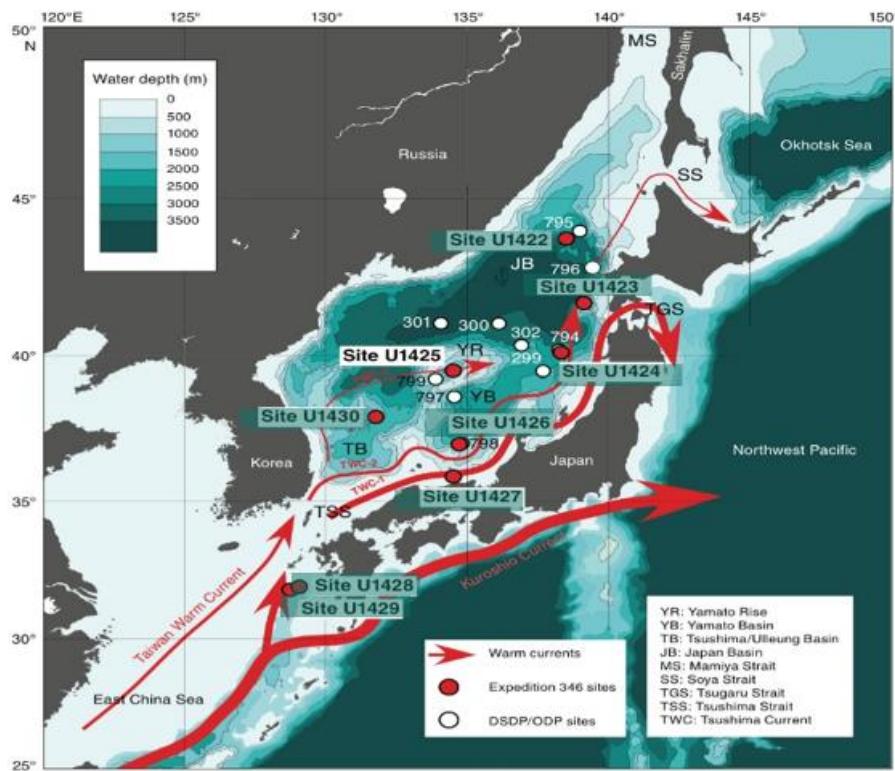


FIGURE 2.11 Location map of IODP Site U1425.

The sediment at Site U1425 is defined by three lithologic units (Tada et al., 2015). Unit I has an age from the Late Pliocene (2.7 Ma) to Holocene and consists of a higher proportion of terrigenous materials of clay and silty clay with small amounts of diatom-bearing clay. The most distinguishing sedimentary feature in this unit is the alternating bedding of dark and light intervals. Unit II has an age from the late Miocene

(7.4 Ma) to late Pliocene (2.7 Ma) and is mostly composed of a mixture of fine-grained clay minerals and biosiliceous components of diatom-rich clay, which indicates high biological productivity in this unit. Unit III, covering 7.4-9.7 Ma in this study, consists of clay, diatom-rich clay, diatom ooze, and siliceous claystone and has relatively higher terrigenous content in the sediment than Unit II. Volcaniclastic materials in this core represent a minor component of the sediments in general (Tada et al., 2015).

The age model for this site was established based on an integration of datums suggested by three age models by Tada et al. (2018), Kurokawa et al. (2019) and Kamikuri et al. (2017). From 0-1.45 Ma, we follow the age model of U1424, where the gamma-ray attenuation (GRA) density profile was tuned with LR04 (Lisiecki and Raymo, 2005), and the age was projected to U1425 using the correlation of dark-light sedimentary cycles by Tada et al. (2018). From 1.5-9.21 Ma, an age model orbitally tuned with long (405 ky) and short (100 ky) eccentricity cycles by Kurokawa et al. (2019) was used. From 9.22-9.69 Ma, biostratigraphic age controls by Kamikuri et al. (2017) were adopted (Figure 2.12).

Samples at Hole D of Site U1425 from the top to ~370 m CCSF-D_Patched_rev20170309 (revised core splice depth below sea floor; Irino et al., 2018), covering 9.6 Ma, where the average sample resolution was approximately 53 ky. Stratigraphic positions of samples collected from the interval out of the splice were projected to the corresponding stratigraphic position in Hole B by examining the core photograph. Details about the samples used, such as projected stratigraphic positions on the splice, revised core splice depth below the sea floor (m CCSF-

D_Patched_rev20170309), estimated ages, and the related sedimentation rates are shown in Table S1.

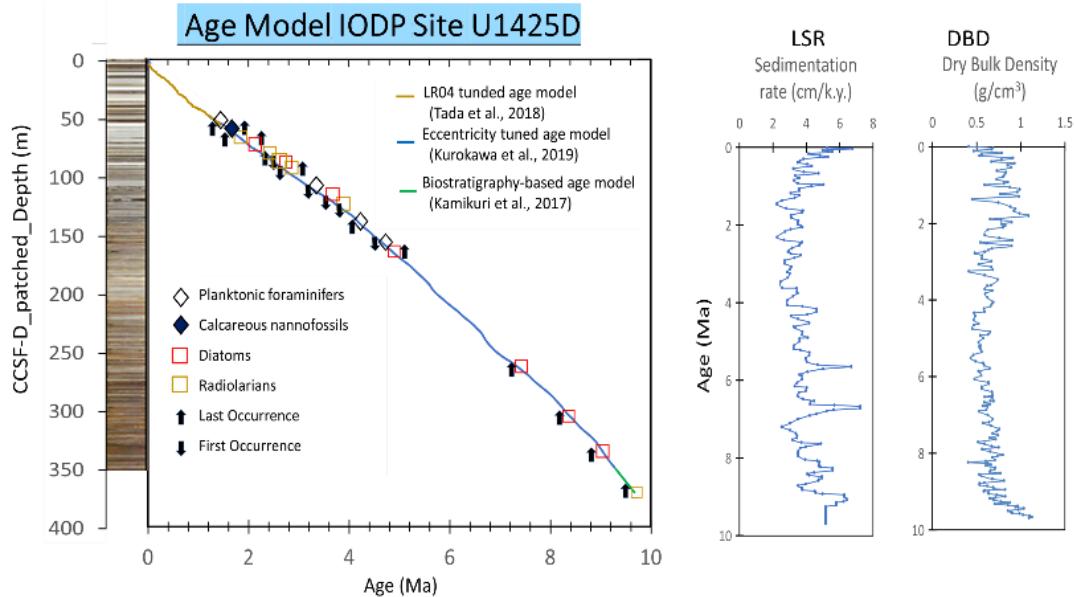


FIGURE 2.12 Age-depth relationship for the IODP Site U1425 sediment sequence. The age model for this site was established based on an integration of datums suggested by Tada et al. (2018), Kurokawa et al. (2019), and Kamikuri et al. (2017).

2.2 Potential source materials

The main sediment sources of the Japan Sea are the eolian dust from Central Asian deserts and riverine input from the nearby Japan islands (Shen et al., 2017). Asian dust is mainly emitted from the ten deserts and sandy lands in Central Asia, such as the Mongolian Gobi Desert, Gurbantuggut Desert, Taklimakan Desert, Qaidam Basin, Badain Juran Desert, Tengger Desert, Hobq Desert, and Mu Us sandy land, according to the meteorological observations (Sun et al., 2001; Wang et al., 2008). The detritus sources of these ten deserts are nearby mountains and underlying bedrock in the three major regions of the Northern Tibetan Plateau (NTP), the Central Asian Orogen (CAO), and the Ordos Plateau in North China which have been formed by tectonic activities in central to northern Eurasia since the Mesozoic (Chen et al., 2007). NTP contains Tarim

Craton and Qaidam Craton that provide clastic particles to the Taklimakan desert from the mountains of Kunlun Shan, Pamirs and Altun (Rittner et al., 2016). CAO contains the Junggar Craton and Siberian Plate, which supplied detritus to the Gobi and Mongolian deserts from the Gobi-Altai, Hangai Mountains, and Tianshan Mountains (north) (Chen et al., 2007; Sun et al., 2018). The Ordos Plateau is situated in the North China Craton delivering detrital materials to the eastern Mu Us Sandy land, Hobq Desert, and Ulan Buh Desert, although their contributions to the total Asian dust emissions are limited (Figure 2.21; Sun et al., 2018).

Geological background of the 3 primary dust source areas

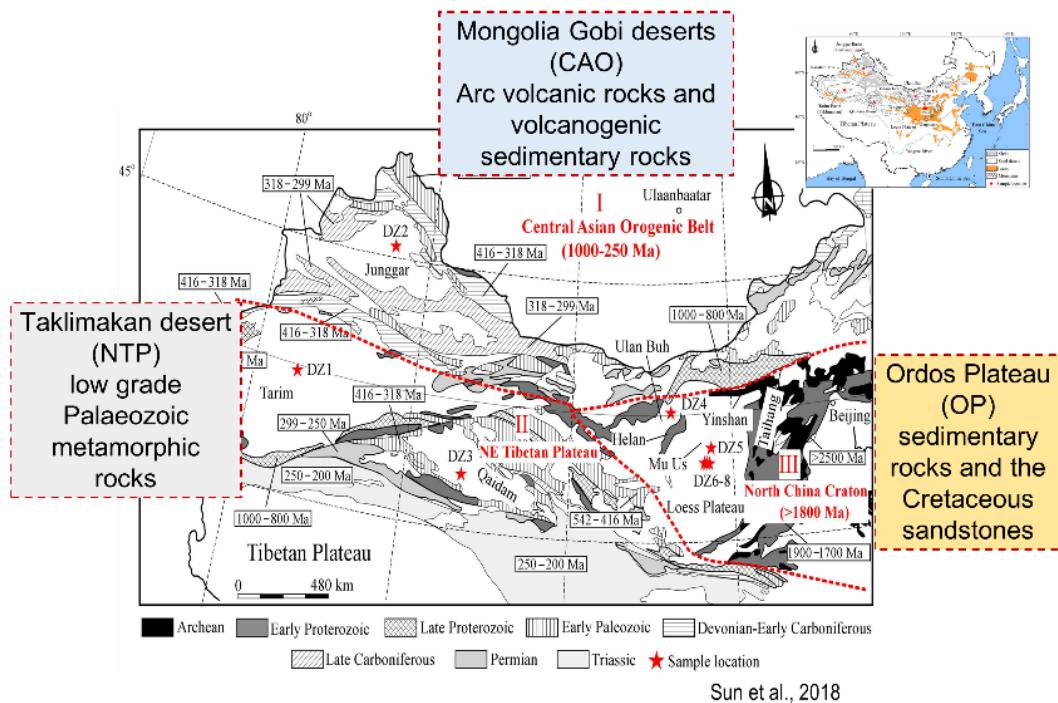


FIGURE 2.21 The three main dust source regions in the Northern Tibetan Plateau (NTP), Central Asian Orogen (CAO) and Ordos Plateau (Chen et al., 2007; Sun et al., 2018) in Central Asia and their geological background.

Despite that NTP and CAO are regional metamorphic mountain ranges, the combination of rock types and nature are different. Taklimakan desert is situated on the Tarim Craton in the NTP region, which consists of marine shale and sandstone in the

basin (Anonymous, 1991). The Tarim sandstone mainly derived from the interior of the Tarim Craton, which is dominated by monocrystalline quartz (Figure 2.22; Carroll et al., 1995). In contrast to the Tarim Basin, volcanic rocks and volcanogenic sedimentary rocks dominate in the Junggar Basin and these sandstones bear the compositional imprint of arc volcanics with felsic composition and plagioclase feldspar (Figure 2.22; Carroll et al., 1995). The East Gobi Basin was formed by extension and intracontinental rifting during the late Mesozoic, while the basement rock in southern Mongolia consists of volcanic islands with composition from lithic- and plagioclase feldspar-rich volcanic detritus to quartz and feldspar-rich granitic material (Watson et al., 1987; Johnson et al., 2001; Davidson, 2005). The distinctive features of the mineralogical composition in the two regions are inherited from the parent rocks which provide a stable and significant property for provenance study (Figure 2.22).

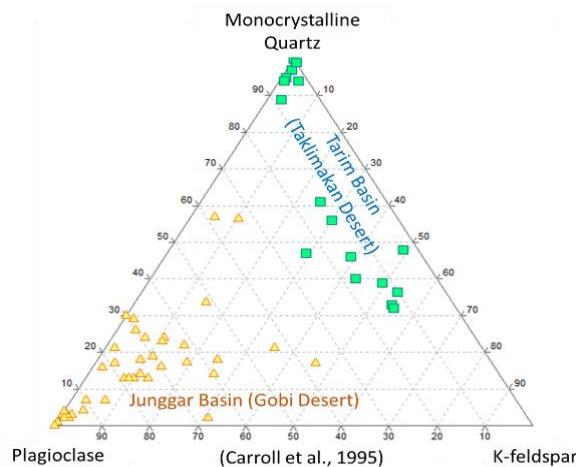


FIGURE 2.22 Ternary diagram of mineral composition of monocrystalline quartz, plagioclase and K-feldspar in the Tarim Basin and Junggar Basin (Gobi) according to Carroll et al. (1995).

Quartz, K-feldspar and muscovite are resistant to chemical weathering (Berner and Berner, 1996), while plagioclase will be partially weathered in the annual precipitation over 700 mm (Jeong et al., 2011). However, plagioclase (albite) is stable

and unchanged in the semi-arid and arid environments which rainfall is less than 500 mm. Because dust source regions in inland Asia such as Junggar Basin, Gobi, Mongolia and Tarim Basin underwent prolonged semi-arid to arid environments from the early Miocene to the present (Guo et al., 2008; Zhang et al., 2022), primary detrital minerals such as quartz, K-feldspar, and plagioclase (albite) could be altered insignificantly under such dry environment. The long-term resistivity of minerals under arid conditions well exemplified in L1 and L2 loess layers in the Chinese Loess Plateau (CLP) (Jeong et al., 2011).

To determine the provenance of the detrital end-members in U1425 sediments, we collected loess and desert materials from the three primary dust source areas of the Tarim Basin near the NTP, Tian Shan Mountains, Mongolian and Gobi Deserts in the CAO, Hobq, Inner Mongolia, and Mu Us Deserts on the Ordos Plateau (Isozaki et al., 2020; Nagashima et al., 2017; Sun et al. 2007) and neritic sediments close to the Japanese coast to serve as a riverine supply from the Japanese islands (Figure 2.23). The list of source samples from the Taklimakan Desert, Mongolian Gobi Desert, Ordos Plateau and Japanese islands is shown in Table S2.

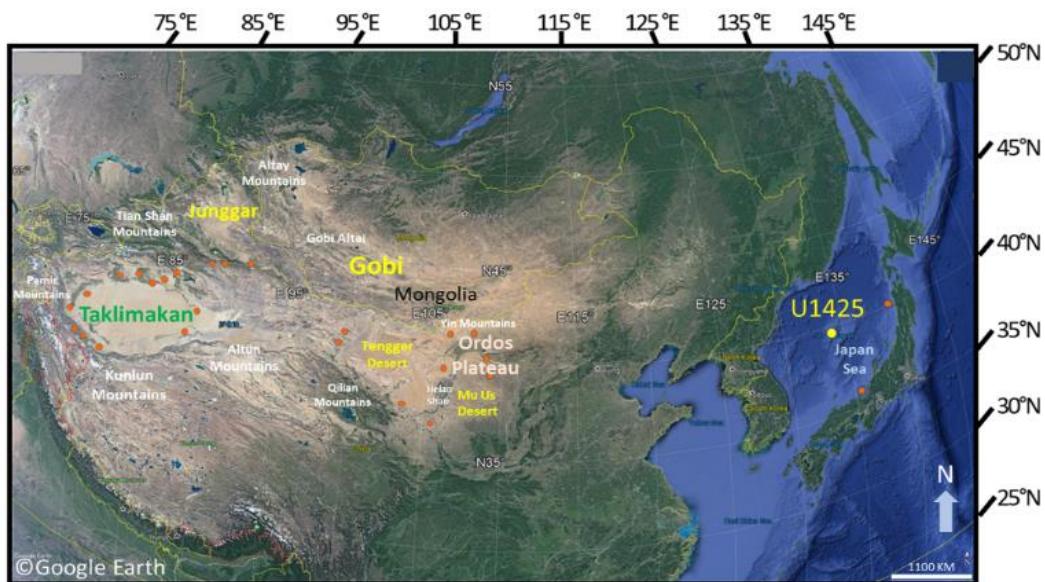


FIGURE 2.23 Sampling sites of the potential dust source materials from deserts in the three main dust source regions in the Northern Tibetan Plateau (NTP), Central Asian Orogen (CAO) and Ordos Plateau in Central Asia and neritic sediments from Japan Islands (orange dots).

Chapter 3. Analytical methods

3.1 Grain-size separation

To distinguish possible transport modes of detrital subcomponents such as westerlies, low-level wind during the winter monsoon, and river suspension and to eliminate mineralogical fractionation during sorting, it was necessary to measure compositional variability for various grain-size fractions within sediments. Nagashima et al. (2007) pointed out that eolian dust dominates the silt fraction of hemipelagic sediments in the Japan Sea, while riverine input is in the clay fraction. Therefore, we decided to separate sediment samples into different size fractions of silt ($>4 \mu\text{m}$) and clay ($<4 \mu\text{m}$). Grain-size separation was conducted by the repeated pipette method (Krumbein and Pettijohn, 1938).

3.2 Powder X-ray diffraction (XRD) Analysis

The mineral composition was analyzed for both silt ($>4 \mu\text{m}$) and clay ($<4 \mu\text{m}$) fractions, as well as potential source materials by using a powder XRD method. A powder sample mounted on a glass holder was scanned from 2° to 40° 2θ by 1° $2\theta/\text{min}$ at 40 kV and 20 mA using CuK α radiation, where the diffraction intensity (counts per second: cps) was recorded with a 0.02° 2θ step. Identification of major and clay minerals was performed according to the position of diagnostic peaks of these minerals on the XRD diffractograms (Figure 3.21; quartz 20.8° , illite 8.8° , chlorite 25.2° , kaolinite 24.2° , smectite maximum between 5 and 8° , K-feldspar 27.5° , anorthite 27.8° , and albite 28.0°). Since the 26.6° peak of quartz may overlap strong reflection of illite,

the 20.8° peak was used as the relative measure of quartz abundance (Biscaye, 1965).

The reproducibility of these measurements is better than 5%.

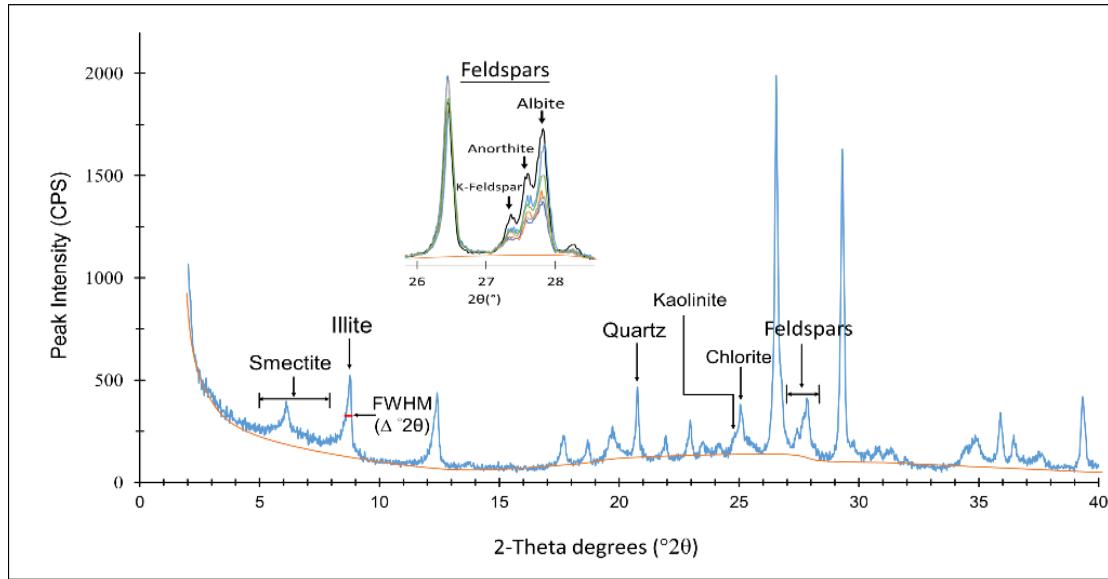


FIGURE 3.21 X-ray diffraction (XRD) spectra for a typical sample from IODP Site U1425. Identification of minerals was performed according to the position of their diagnostic peaks on the XRD diffractogram.

Illite crystallinity and illite chemical Index

Illite crystallinity and the chemical index of illite were also examined. The illite crystallinity was examined as a half-height width (FWHM) ($\Delta \text{ } ^\circ 2\theta$) of the 10 \AA (8.8°) peak on the XRD diffractograms (Figure 3.21). This is a measure of the crystallite size of illite and is often used to trace the possible source regions in marine sediments (Petschick et al., 1996). Higher values indicate poor crystallinities, and low values indicate well-developed crystallinities (Ehrmann, 1998). According to the Kübler index (1964), the illite crystallinity value is useful to determine what type of metamorphic conditions existed when a rock was formed. The illite chemistry was inferred from the ratio of $5 \text{ \AA}/10 \text{ \AA}$ peak areas on the XRD diagrams. Values above 0.4 are found in Al-rich illites (muscovite) formed by strong hydrolysis, while ratios below 0.4 represent

(Fe, Mg)-rich illites (biotite) characterizing for physically eroded/unweathered rocks (Ehrmann, 1998).

3.3 Measurement of diatomaceous content

Biogenic silica content (bioSiO_2) was estimated from the calibration curve with the height of diatomaceous hump between 16 and 32.5° on the XRD diagrams (Figure 3.31 (a)). A calibration curve was prepared from known mixtures of nearly pure diatom and diatom-free sediment in varying proportions to give a series of mixtures containing 0 to 90 percent diatomaceous and the samples were X-rayed. The height of the counting from XRD with the known amount of opal in the sediment was plotted as calibration curves (Figure 3.31(b)). The amounts of diatomaceous content in silt and clay fractions were calculated by this correlation:

$$\text{diatomaceous content (\%)} = \text{height of } 32.5^\circ \text{ hump (cps)} - 1.6831 / 3.9661.$$

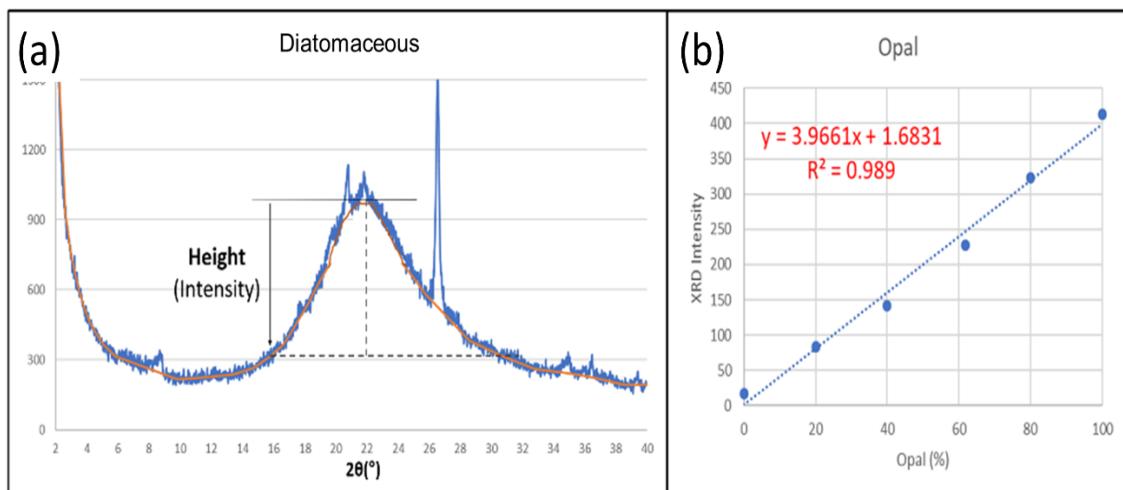


FIGURE 3.31 (a) Measurement of intensity (height) of diatomaceous content. (b) Calibration curve for determination of diatomaceous content.

3.4 Mass Accumulation Rate (MAR) Calculations

The MAR ($\text{g/cm}^2/\text{ky}$) for each detrital subcomponent was calculated using the dry bulk density (DBD) and linear sedimentation rate (LSR). DBD is defined as the mass (weight) of the dry solids divided by the total volume of the wet sample (Dadey et al., 1992). DBD (g/cm^3) was estimated from wet bulk density (WBD) assuming constant grain density (GD) and pore water density by Equation (1). As a measure of WBD for each sample, shipboard gamma-ray attenuation (GRA) density was used, which represented the WBD of the entire core and was measured for every 2.5 cm interval for all sediment core sequences (Tada et al., 2015). To maintain consistency between wet bulk density directly determined by shipboard “moisture and density (MAD)” measurements (Tada et al., 2015), we first calibrated GRA-based WBD (WBD_{GRA}) to MAD-based WBD (WBD_{MAD}) using a correlation between them, such as $\text{WBD}_{\text{MAD}} = 0.12901 + 0.89581 * \text{WBD}_{\text{GRA}}$ (S.E.= ± 0.10). A GD of 2.59 ± 0.15 (g/cm^3) averaged for all MAD measurements for the U1425 samples was assumed for all samples. The pore water density was assumed to be 1.024 g/cm^3 , as used in the MAD calculation (Tada et al., 2015).

$$\text{DBD} = \frac{\text{WBD} - 1.024}{1 - \frac{1.024}{\text{GD}}} \quad (1)$$

LSR (cm/ky) was determined from the revised core splice depth below the sea floor (m CCSF-D_Patched_rev20170309) and the integrated age model. MAR ($\text{g/cm}^2/\text{ky}$) of each end-member (EM) was calculated by the method of Rea and Janecek (1981) as $\text{MAR} = \text{fraction of EM} * \text{LSR} * \text{DBD}$.

Chapter 4. Application of parallel factor analysis (PARAFAC) to XRD diffractograms

4.1 Theoretical framework

The XRD diffractogram of each sample could be regarded as a linear combination of the diffractograms of subcomponents, such as dust from various sources, riverine input from surrounding lands, biogenic materials, and their diagenetic products. Each diffractogram of the subcomponent and its fraction in the sample should be nonnegative, and the total fraction of subcomponents should be (close to) unity. Parallel factor analysis was applied to decompose XRD diffractograms into those of subcomponents (end-members) and to quantify their contributions using drEEM toolboxes running on MATLAB® software (Murphy et al., 2013). The PARAFAC model was applied to data that were arranged in three-way arrays such as the excitation-emission matrix (EEM) of fluorescent organic matter (fluorescence intensity of sample \times excitation wavelength \times emission wavelength), where x_{ijk} is the intensity of fluorescence corresponding to the j -th emission wavelength and the k -th excitation wavelength for the i -th sample, and e_{ijk} is the residual representing the variability not accounted for by the model. Assuming the number of end-members is F , the decomposed matrices a_{if} , b_{jf} , and c_{kf} are the factor score (relative contribution of subcomponent in the i -th sample), factor loading spectrum along emission wavelength at a k -th excitation wavelength, and magnification factor at k -th excitation wavelength for f -th end-member, respectively, where a_{if} , b_{jf} , and c_{kf} are non-negative (Murphy et al., 2013).

$$x_{ijk} = \sum_{f=1}^F a_{if} b_{jf} c_{kf} + e_{ijk} \quad (2)$$

The calculation is conducted to obtain an end-member-mixing model that minimizes e_{ijk} . In the case of XRD in this study, the dataset can be regarded as a special case with $k = 1$, where x_{ijk} is the intensity of diffraction at the j -th diffraction angle ($2\text{-}40^\circ 2\theta$ with $0.02^\circ 2\theta$ interval; $1 \leq j \leq 1900$) for the i -th sample ($1 \leq i \leq 180$).

4.2 Model development

Steps included in PARAFAC analysis are to (1) import and assemble the dataset; (2) preprocess; (3) explore the data and develop preliminary models; (4) develop a final, validated model containing the correct number of components; and (5) export and interpret the results. These steps are well documented in Murphy et al. (2013). Here, the PARAFAC model development was initiated with a series of 2 to 7 components using the XRD diffractograms of the 180 samples and processing with the non-negativity constraint (which has positive values of scores and spectra) and default convergence criterion (0.01) for the outlier tests. Once the modelling constraints and criteria have been decided, the best way to obtain the best solution models for any given number of components is to repeat the modelling and ultimately adopt the model that represents the least-squares (minimum error) solution. When modelling does not produce a stable solution, indicating the model may have too many components or stricter (smaller) convergence criteria is required (Murphy et al., 2013). We applied a stricter convergence criterion of a minimum tolerance of 1×10^{-8} with nonnegativity constrained for the 4-7 components modelling (See Appendix 1). During the model development, no sample or data are removed.

4.3 Determination of the number of end-members

An appropriate model was found to minimize the sum of squares of the residuals (Bro, 1997). In this study, model development was initiated with a series of ($F =$) 2 to 7 end-members (EMs), which were applied to the 180 XRD profiles of silt and clay fractions with a nonnegativity constraint. Increasing the number of end-members from 2 to 7 reduced the score error and total residual sum of squares (RSS), and a significant decrease in error occurred between the 5- and 6-end-member models. As shown in Figure 4.31(a), for the silt datasets, the 4- and 5-end-member models showed higher RSS and larger score errors from 15-25%, while the 6- and 7-end-member models showed smaller score errors within 10%. On the other hand, some samples had a larger RSS in the 7-end-member model than in the 6-end-member model (Figure 4.31(a)). In the clay datasets, the 4-, 5- and 7-end-member models showed higher RSS and larger score errors than the 6-end-member model, which had the smallest RSS and score errors within 10% (Figure 4.31(b)). Therefore, 6-end-member models were adopted for both silt and clay datasets in this study.

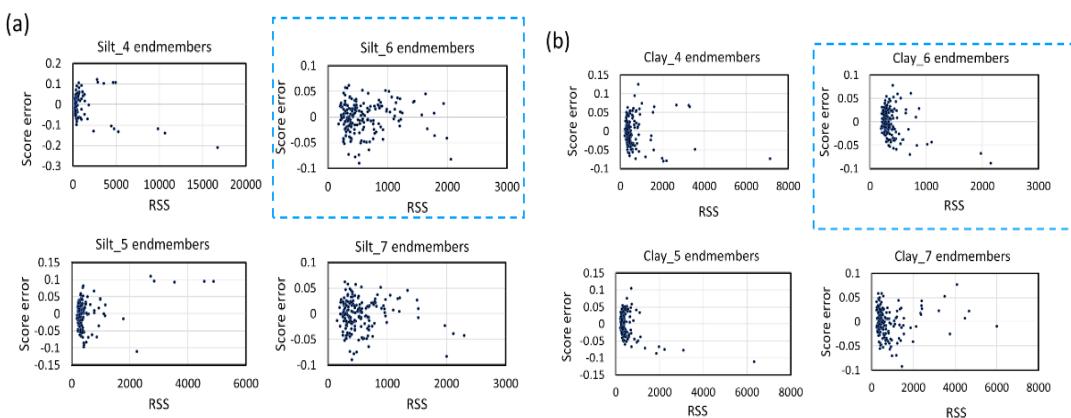


FIGURE 4.31 Residual sum of squares (RSS) and score errors of each sample in (a) silt and (b) clay datasets for the 4 to 7 end-member models calculated by PARAFAC.

4.4 Scaling factor calculations and reproducibility of end-member modelling

After validation of the 6-end-member PARAFAC model, scores (a_{if}) and spectral loadings (b_{jf}) of each end-member were obtained from the model, where c_{kf} was unity in this case ($k = 1$). Because the total sum of fractions of each end-member should be unity, scaling factors, α_f , were calculated to satisfy $\sum_{f=1}^6 \alpha_f a_{if} \approx 1$ for all i , where the square sum of residual errors $(1 - \sum_{f=1}^6 \alpha_f a_{if})$ was minimized. Using α_f , the fraction (absolute contribution) of the f -th end-member for the i -th sample was calculated as $\alpha_f a_{if}$. To obtain the XRD intensity profile of each end-member, outputs of drEEM toolboxes (Murphy et al., 2013) for spectrum loadings (b_{jf}) were first divided by the maximum of b_{jf} in terms of j for each f -th end-member, which was set as b'_{jf} . Then, b'_{jf} was divided by the scaling factor α_f to obtain a “realistic” diffractogram of each end-member.

The 6-end-member PARAFAC was modeled 20 times (Figure 4.41(a) and 4.41(b)) to ensure that the result was stabilized and not a local minimum (Stedmon and Bro, 2008). The 20 repetitions of score calculations of each end-member in the silt and clay datasets are stable and had no large variation (Figure 4.42(a) and 4.42(b)) except the Opal-CT (EM6) in the silt dataset. The content of diatomaceous (EM4) and Opal-CT (EM6) estimated from the PARAFAC Model is compared with the measured content of biogenic from XRD diffractograms at this site (Figure 4.43(a) and 4.43(b)). Both present similar content and temporal distribution.

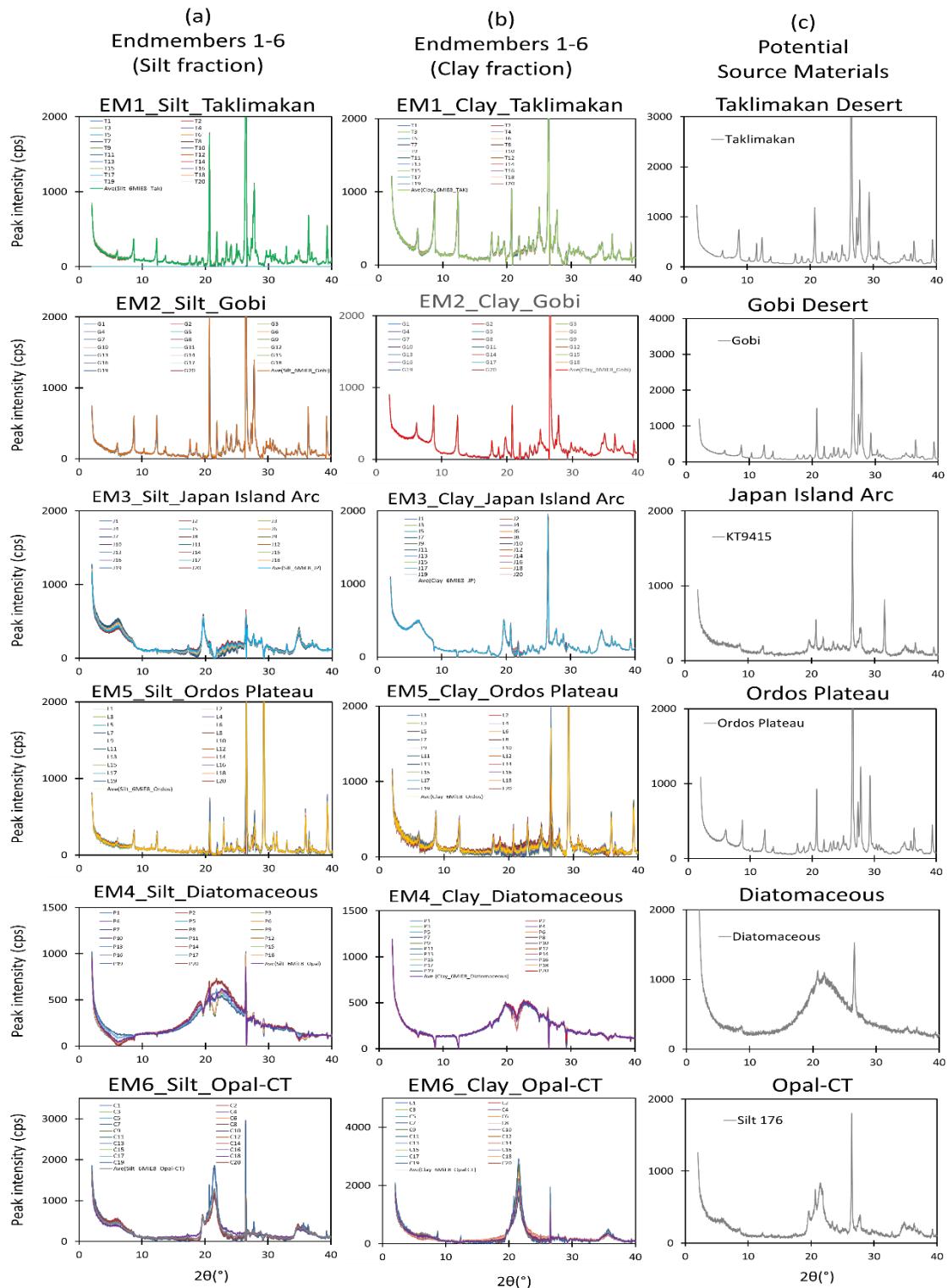


FIGURE 4.41 Summary of the 20 repetitions of spectral loading calculations and their average spectra for each end-member in the (a) silt and (b) clay datasets compared with the X-ray diffractograms of (c) potential source materials. Diffractograms for diatomite collected from the Monterey Formation in California and the sample from IODP U1425D 49H-2, 50 cm are shown as typical diatomaceous (opal-A) sediment and diagenetic form of sediment (opal-CT), respectively.

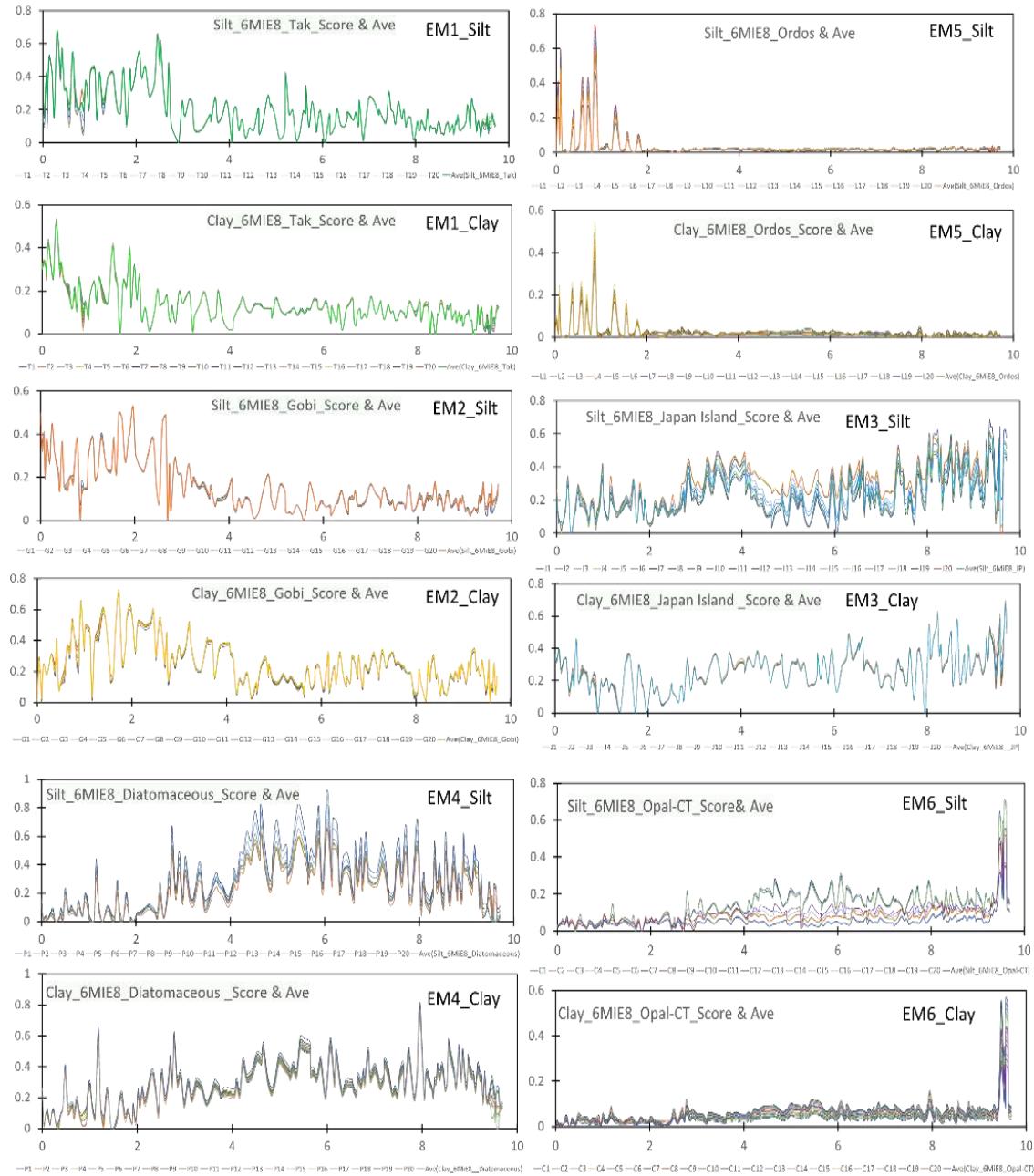


FIGURE 4.42 Summary of the 20 repetitions of score calculations and their average for each end-member in the silt and clay datasets.

Similar solutions with minor differences were reached, which implied that the obtained model was reproducible and reliable and could be used for a direct comparison between the mineral compositions of end-members and potential source materials. Therefore, the result was reported as the average of 20 models. The nonnegativity

constrained 6-end-member model explained 99.57% of the variation in silt datasets and 99.43% in clay datasets.

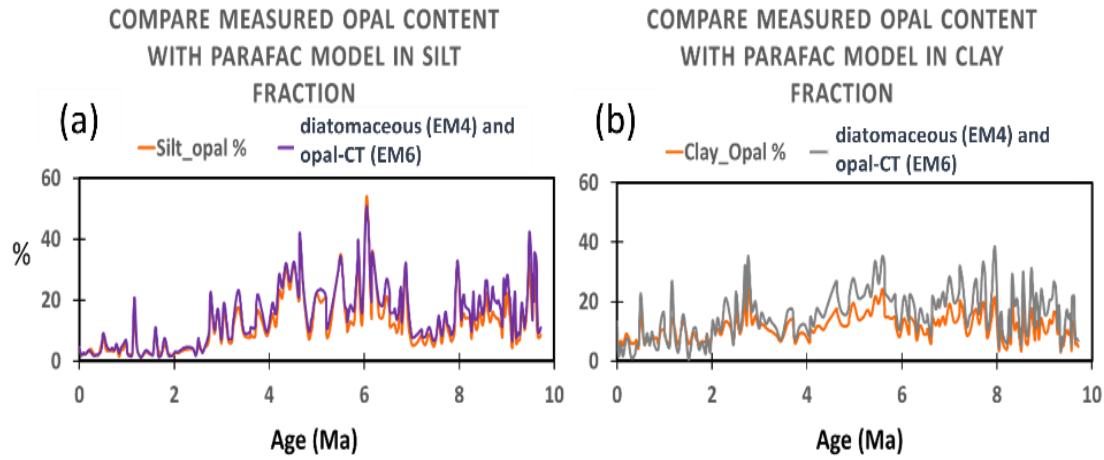


FIGURE 4.43 The total biogenic content of diatomaceous (EM4) and opal-CT (EM6) in silt and clay datasets (purple and grey line) estimated from the PARAFAC Model. They are compared with the measured diatomaceous content (orange line) in the (a) silt and (b) clay fractions.

Chapter 5. Results

5.1 Terrigenous components

The total terrigenous materials at IODP Site U1425 in Japan Sea sediments range from 25% to 82%. In general, the total terrigenous materials are low in Pliocene and higher between 8-9 Ma and after 2.7 Ma. The detrital silt fraction ranged from 3-63% and clay fraction ranged from 11 to 54%. The higher ratios of silt fraction occurred in the late Miocene during 9.6-8 Ma and lowered in 7.6-4 Ma. The higher ratios of clay fraction occurred in 7.6-6.3 Ma and after 2.5 Ma (Figure 5.11).

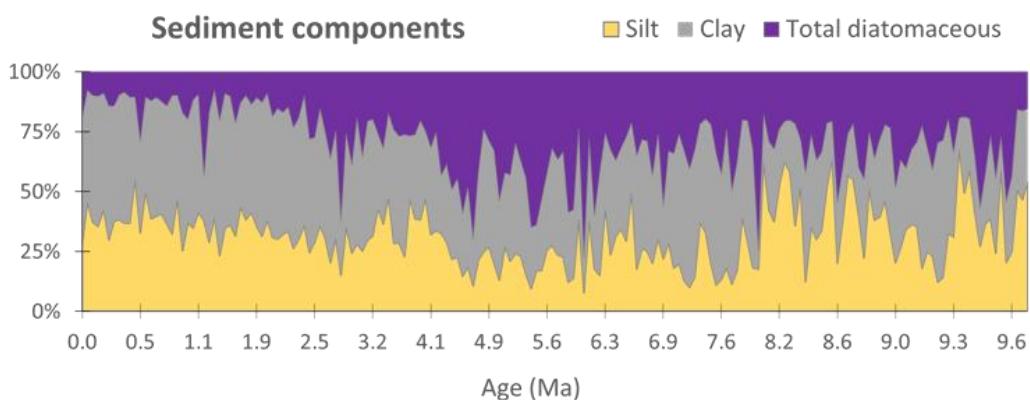


FIGURE 5.11 Temporal variation of sediment components at IODP Site U1425 in Japan Sea.

Lower contents of total terrigenous materials occurred in Pliocene which were associated with higher ratios of biogenic content. The biogenic content ranges from 6 to 68% (Figure 5.11). It is generally high in the late Miocene to Pliocene and especially high content between 5.5-4 Ma which corresponded to the higher abundance of diatoms and other siliceous components (Tada et al., 2015).

Terrigenous minerals from the IODP Site U1425 consists mainly of quartz, feldspar (including albite, anorthite and K-feldspar) and clay minerals (including illite, chlorite, kaolinite and smectite) as well as minor amounts of dolomite and pyrite.

Calcite is sparse throughout the upper 50 m of the core (mostly in the form of foraminifers and nannofossils) (Tada et al., 2015).

5.2 Mineral composition in silt and clay fractions in Japan Sea sediments

The relative content of minerals is inferred from changes in peak intensity (cps) of XRD analysis. Quartz is the most dominant mineral in sediments at this Site in Japan Sea. Quartz and feldspar are primary minerals that are related to parent rock composition.

Quartz has the highest peak intensity, ranging from 200 to 1,400 cps in silt fraction, and from 180 to 600 cps in the clay fraction. The highest quartz intensity occurred in the Pleistocene in silt fraction after 2.6 Ma to the present, and several high intensities were found during late Miocene (7Ma) to Pliocene (3Ma). The abruptly increased in quartz during the Pleistocene may indicate an abrupt increase in eolian dust as quartz is the major mineral component of Asian dust. Quartz intensity of clay fraction is stable from 200 to 400 cps and showed a gentle increase from late Miocene to the present (Figure 5.21).

Feldspars are the second most dominant mineral at this site. Albite and anorthite are the end-members of plagioclase feldspar. Albite is relatively common and important rock-forming mineral associated with the more acid rock type. Albite intensity varied between 100 and 1000 cps in the silt fraction, and below 400 cps in the clay fraction. The temporal distribution of intensity of albite in the silt and clay fractions is similar to quartz (Figure 5.21). Anorthite is a comparatively rare mineral but occurs in the basic

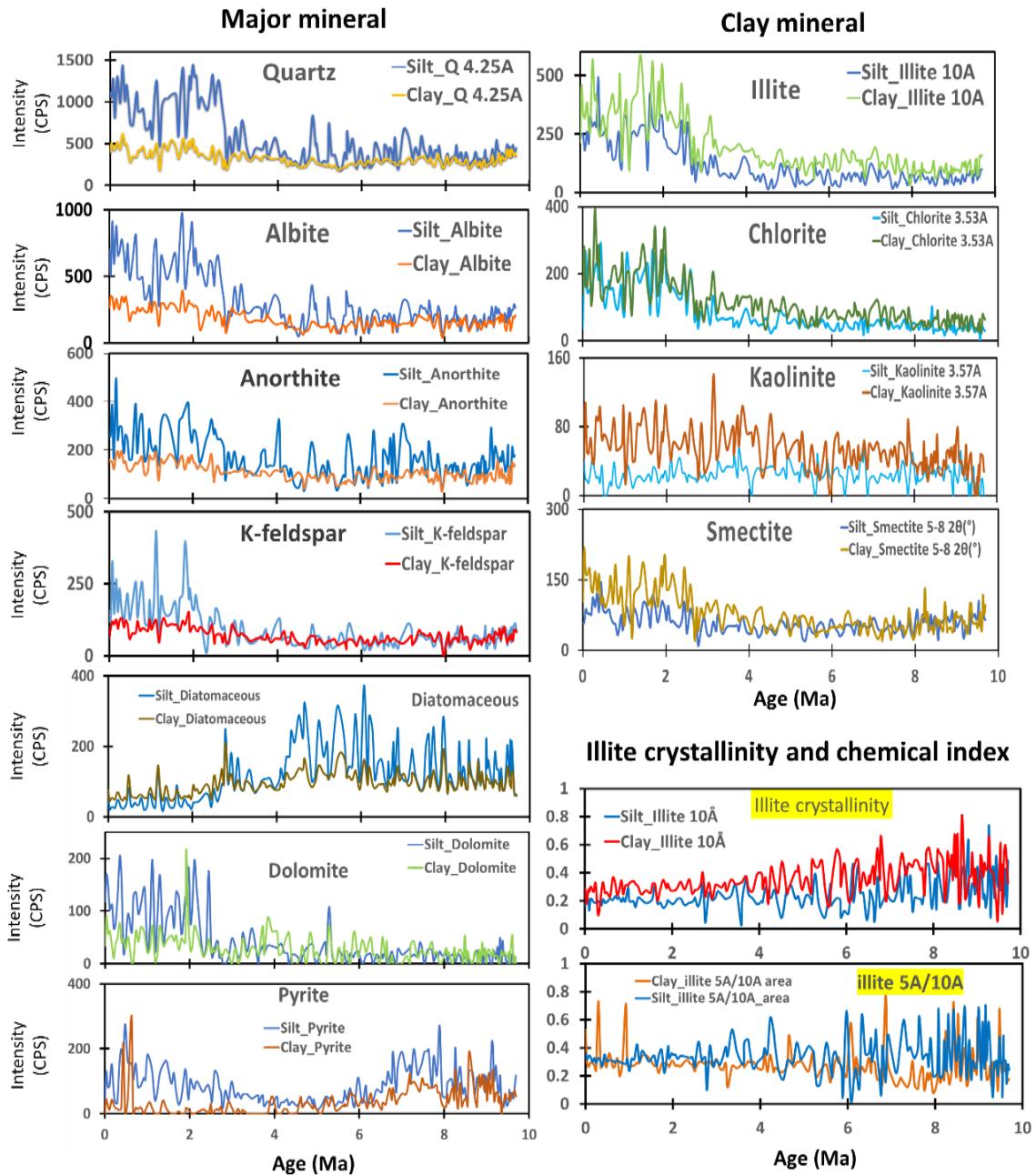


FIGURE 5.21 The temporal variability of mineral abundances of major and clay minerals in silt and clay sediments at IODP Site U1425 in the Japan Sea. Illite crystallinity and chemical index are also plotted with age in the silt and clay fractions.

plutonic rocks of some orogenic calcium-alkaline suites. Anorthite intensity varied between 50 and 500 cps in silt fraction and below 200 cps in clay fraction. In general, quartz and feldspars show higher intensity in the silt fraction than the clay fraction from the late Miocene to the Pleistocene (Figure 5.21).

Potassium-Feldspar (K-feldspar) is another end-member of alkali feldspar minerals which is a common constituent of most granites and other felsic igneous rocks. K-feldspar in silt fraction also showed an abrupt increase during Pleistocene after 2.1 Ma. Then intensity of K-feldspar in the silt and clay fractions is similar before 7.2 Ma, but slightly increased in the silt fraction since 7 Ma and reached the highest in the Pleistocene (Figure 5.21).

Illite is a ubiquitous terrigenous weathering product, generally associated with cool, dry environments and it is the dominant clay mineral in the Chinese Loess (Sun 2011). Illite is the third most dominant mineral at the site, with the intensity >500 cps during the Pleistocene in the clay fraction. Chlorite is usually indicating the mechanical weathering of terrigenous sediments. The peak intensity of chlorite in the silt and clay fractions are below 400 counts. The temporal distribution of illite and chlorite shows similar pattern in silt and clay fractions. In general, the peak intensity of clay fraction is higher than in the silt fraction and both increased abruptly at 3.1 Ma. Illite and chlorite show the highest intensity in the Pleistocene (Figure 5.21).

Kaolinite is a terrigenous weathering product usually associated with strong hydrolysis. The peak intensity of kaolinite in silt and clay fractions are generally low, below 140 cps. The downcore variation of kaolinite is different between silt and clay fractions. The peak intensity of kaolinite in silt fraction is around 50 cps between Miocene to the Pleistocene, while that in clay fraction increased gradually from Miocene to the Pleistocene with a sudden peak at 3.2 Ma with 141 cps (Figure 5.21).

Smectite is produced by continental weathering processes, weathering of volcanogenic material, and authigenic formation in sediments. The peak intensity of smectite in the silt and clay fractions are generally low from 10-100 before Pleistocene and increase to 150 cps after 2.7 Ma (Figure 5.21).

Other minerals composition at Site U1425 in Japan Sea

Authigenic components are oceanic inorganic minerals that precipitate directly from the seawater, for example, dolomite, calcite and pyrite. Dolomite is a common rock-forming mineral. It is similar to the most common carbonate mineral calcite. These two share some similarities in structure and are very closely related chemically. Calcite is chemically CaCO_3 (calcium carbonate), dolomite is $\text{CaMg}(\text{CO}_3)_2$ (calcium-magnesium carbonate). Dolomite intensity is low in the late Miocene to the Pliocene and increases to 200 cps after 2.7 Ma. Pyrite is the most abundant and widespread sulfide mineral (FeS_2) and its relative content in the Japan Sea is high (200 cps) in the late Miocene and the Pleistocene, but low in the Pliocene (Figure 5.21).

Illite crystallinity and Chemistry Index

The illite chemistry index in the silt fraction varies between 0.02 and 0.7, with an average of 0.33. In general, the silt fraction showed a narrow range of the value of 0.3 during the Pleistocene and the very low value of <0.1 was found in some samples during 9.6-9.3 Ma, 6.7-5.9 Ma, and at 2.7 Ma. The illite chemistry index in the clay fraction varies between 0.08 and 0.78, with an average of 0.27, and showed a lower value (<0.3) in some samples during 8-7 Ma and 4.5-1 Ma (Figure 5.21). According to the illite chemistry index, values below 0.4 represent (Fe, Mg)-rich illites (biotite) characterized

for physically eroded and unweathered rocks. The highly crystallized illite and Fe-Mg rich illite in silt fraction at Site U1425 in Japan Sea may be eolian origin (Figure 5.21).

The crystallinity of illite in the silt and clay fraction varies between $0.05\text{--}0.74^\circ 2\theta$ and $0.1\text{--}0.8^\circ 2\theta$ with average values of about 0.25 and $0.36^\circ 2\theta$, respectively. The illite crystallinity of the silt fraction showed a lower and narrow range after the Pleistocene with the value of $\sim 0.2^\circ 2\theta$, while the clay fraction showed a higher value of $\sim 0.3^\circ 2\theta$ during the same period (Figure 5.21). High values indicate poor crystallinities and low values indicate well-developed crystallinities. The very low values of crystallinity were found in some silt fractions at 9.3 Ma and between 6.6 Ma and 2.7 Ma with the value of $<0.1^\circ 2\theta$. The highly crystallized nature indicates the illite was not formed at surface temperature and may have originated from a low-temperature metamorphic environment. According to the Kübler index, the illite crystallinity values of $<0.25^\circ 2\theta$ range correspond to the metamorphic greenschist facies that formed under the lowest temperatures and pressures usually produced by regional metamorphism (Figure 5.21).

Sources variability of silt and clay fractions

The illite/Quartz (Q), chlorite/Q and kaolinite/Q in silt and clay fractions show different fluctuation patterns. It is possible that the sources of these minerals are different between silt and clay fractions. The difference from 8 Ma where the two fractions show gradual change and the greatest difference occurred at 2.9 Ma (Figure 5.22(a)). The feldspars composition in the clay fraction shows more orthoclase, while the silt fraction contains more anorthite. Anorthite/Albite (An/Ab) range in the clay fraction is narrow from 0.4 to 0.6, while silt fraction shows a wider range from 0.2 to 1

which shows the highest value in the late Miocene and lower values in the Pleistocene and Pliocene. It indicates progressive changes in dust source areas or climatic (erosional) environments (Figure 5.22(b)).

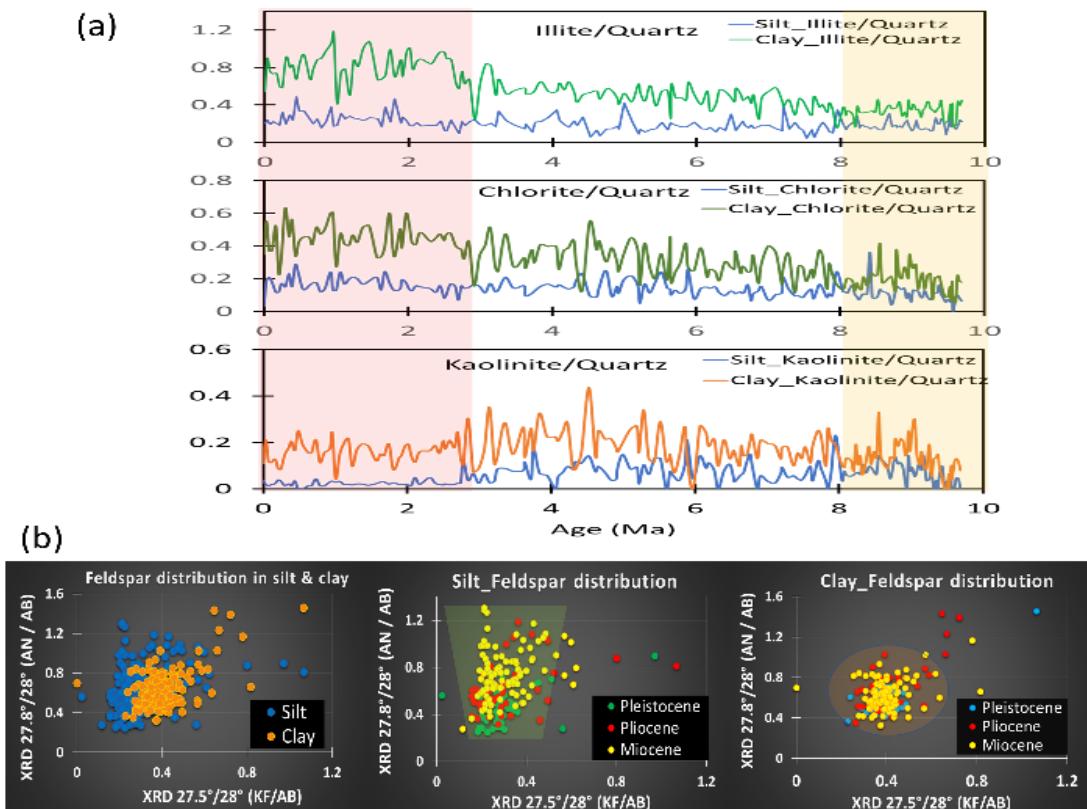


FIGURE 5.22(a) The illite/Quartz (Q), chlorite/Q and kaolinite/Q in silt and clay fractions. Shadings show the sudden increase of the difference between silt and clay fractions in 8Ma and 2.7 Ma. (b) The distribution of feldspar composition in silt and clay fractions. Silt fraction shows a progressive change of dust source areas or climatic (erosional) environments. AN/AB —Anorthite/Albite; KF/AB —K-feldspar/Albite.

Differentiation of Asian eolian and Japanese riverine sources

Quartz, plagioclase and illite were the most abundant minerals in the silt fraction during the Pleistocene, suggesting many dust-derived materials in this period (Table S3). The half-height width (FWHM) of illite in the silt fraction was low (high crystallinity) with a narrow range of approximately $0.2 \Delta^{\circ}2\theta$ in the Pleistocene. According to the Kubler Index (1964), highly crystallized illite (FWHM <0.25) was sourced from regional metamorphic rocks in mountain ranges, such as those on the

northern Tibetan Plateau (NTP) or in the Central Asian Orogen (CAO). Poor illite crystallinity ($\text{FWHM} > 0.4$) corresponded to diagenetic rock sources, such as those in the Japan Island arc.

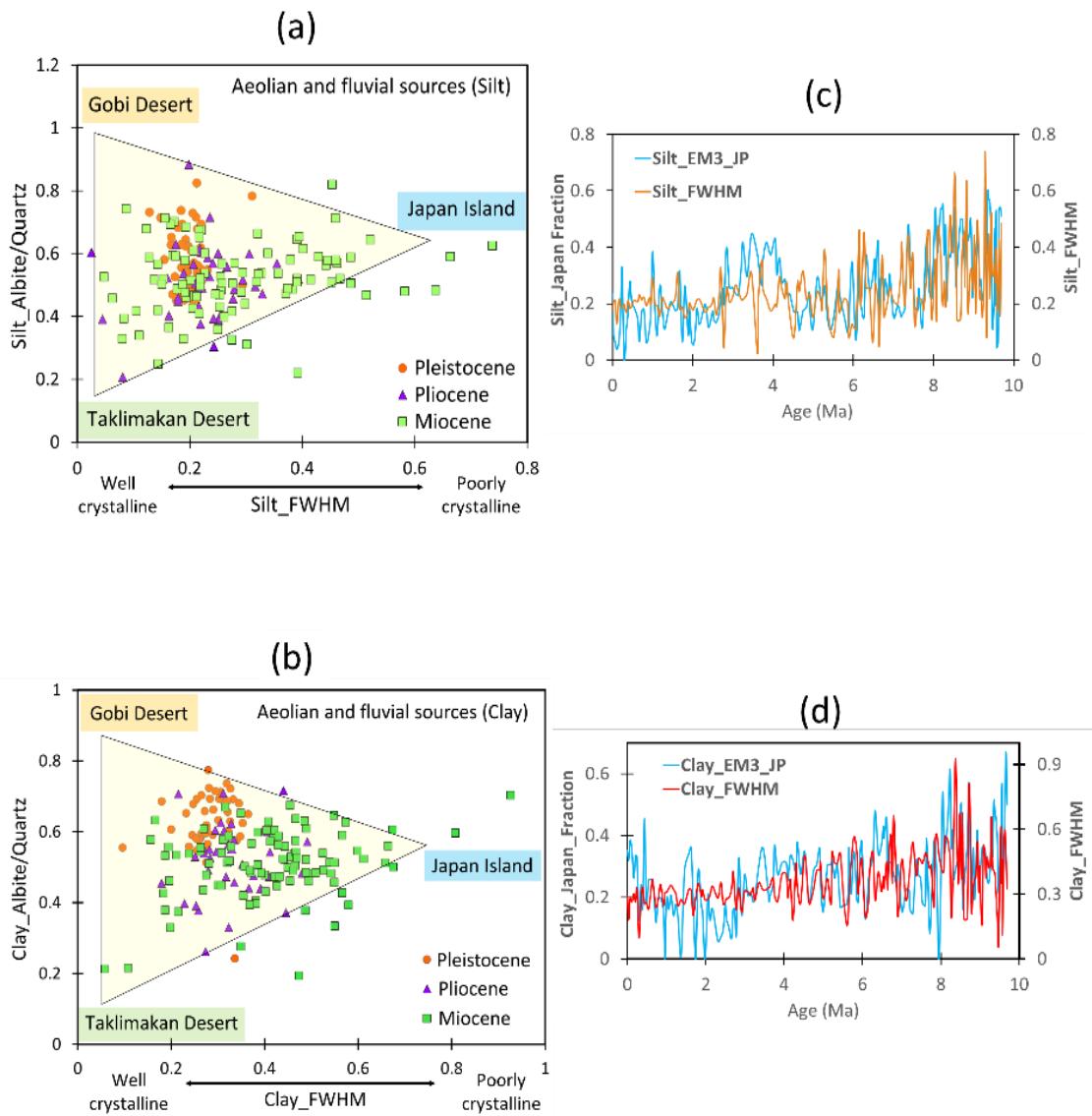


FIGURE 5.23 Differentiation of Asian eolian and Japanese riverine sources based on albite/quartz ratios and illite crystallinity (FWHM values) distributions in (a) silt and (b) clay fractions. Temporal variation of illite crystallinity compared with the contribution of EM3 (Japan Island arc origin) estimated by PARAFAC modelling in the (c) silt and (d) clay fractions.

Based on the mineralogical differences in the Tarim Basin (higher quartz content) and Junggar Basin in the Gobi Desert (higher plagioclase content) (Carroll et al., 1995 [33]) mentioned in above Section 2.2 (Figure 5.23), the distribution of albite (Ab) and

quartz (Q) ratios and illite crystallinity (FWHM) could effectively differentiate dust and riverine sources in silt and clay sediments in the Japan Sea. Taklimakan eolian sediment had the lowest FWHM values and the lowest Ab/Q ratios, and Gobi dust had a low FWHM but a higher Ab/Q ratio. Japanese riverine sediment had the highest FWHM values and higher Ab/Q ratios (Figure 5.23(a)). These interpretations are also applicable to the clay fraction (Figure 5.23(b)). This preliminary identification of the 3 sources is not sufficiently quantitative because of a lack of knowledge about exact mineral ratios for each detrital subcomponent and difficulty in eliminating contributions from biological components. Therefore, the results of more quantitative statistical end-member unmixing by parallel factor analysis (PARAFAC) are examined in the next section.

5.3 Provenance assessment of PARAFAC end-members

To determine the provenance of the 6 EMs in U1425 sediments calculated by PARAFAC, firstly we compared the diffractogram patterns of EMs with those of potential source materials. Figure 4.41 shows that desert source materials from the Taklimakan Desert, Gobi deserts and Ordos Plateau had high intensities of quartz, feldspar, and illite which match well with EM1, EM2, and EM5. Then, characteristic mineral ratios such as Ab/Q and K-felspar (K-fel) /Ab were used to distinguish the source materials (Figure 5.31(a) and 5.31(b)). Namely, Gobi and Ordos dust sources are characterized by higher Ab/Q, while Taklimakan shows lower Ab/Q (Figure 5.31(a)). On the other hand, Ordos materials are characterized by higher K-fel/Ab (Figure 5.31(b)).

Taklimakan materials and EM1 had lower Ab/Q ratios than Gobi materials and EM2 (Figure 5.31(a)). Sandstone in the Tarim Basin is dominated by monocrystalline quartz, which was a typical continental block-recycled orogen provenance of Paleozoic metamorphic rocks in the NTP (Carroll et al., 1995; Jia et al., 1997; Wang et al., 1986), while the Mongolian Gobi is characterized by back-arc basin deposits with plagioclase, feldspar-rich, volcanic detritus (Carroll et al., 1995, Davidson, 2005). Ordos materials and EM5 had high K-fel/Ab ratios (Figure 5.31(b)). The Ordos Plateau is situated in the northern part of the North China Craton, which contains largely undisturbed sedimentary rocks that are Carboniferous to Jurassic in age and Cretaceous sandstones (Kusky and Li, 2003). Therefore, we could attribute EM1, EM2, and EM5 to Taklimakan, Gobi, and Ordos, respectively. Again, it was also confirmed that EM1, EM2 and EM5 exhibited similar XRD diffractogram patterns to those of the desert materials from Taklimakan, Gobi, and Ordos, respectively (Figure 4.41).

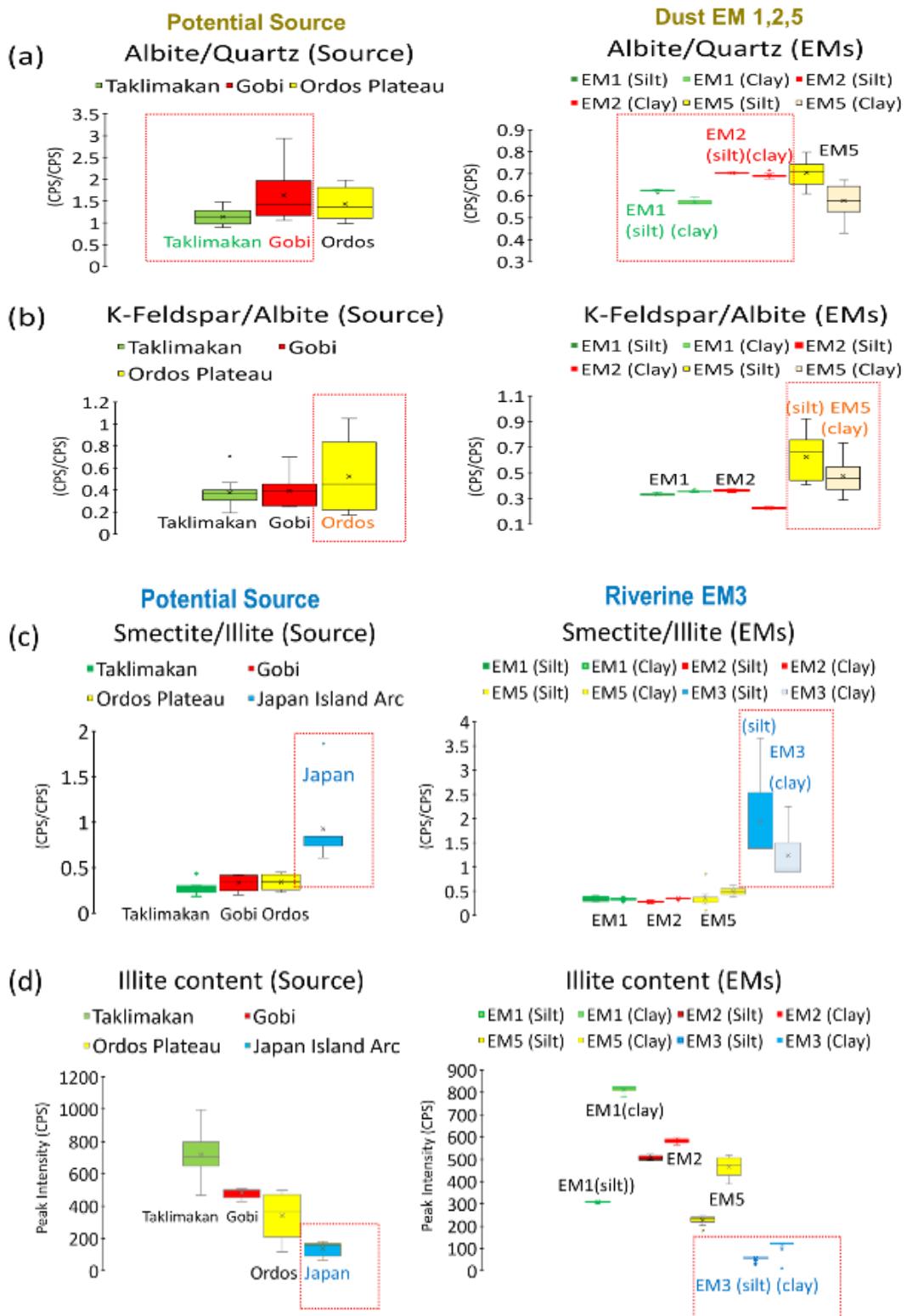


FIGURE 5.31 Summary of the mineral composition of source materials and end-members. Dust source materials from Taklimakan, Gobi and Ordos are compared with EM1, EM2 and EM5 for (a) albite/quartz and (b) K-feldspar/albite ratios. Japanese source materials are compared with EM3 for (c) smectite/illite and (d) illite contents.

Japanese island materials and EM3 had XRD diffractogram patterns different from those of the Asian dust sources and revealed significant peaks of smectite, low illite intensities (Figure 5.31(c) and 5.31(d)) and high anorthite/albite ratios. The Japanese islands are characterized by basaltic volcanic and sedimentary rocks (Nakada et al., 2016). Volcanic rocks often produce smectite by chemical weathering under wet and humid environments on Japanese islands (Liu et al., 2009). An attribution of EM3 to Japanese Island materials could be also justified by the similar temporal variations of illite FWHM and EM3 contribution (Figure 5.31(c) and 5.31(d)). Larger illite FWHM (low crystallinity) is generally associated with higher contribution of EM3 and both profiles show similar trend and critical changes at 8, 6, and 2.7 Ma both for silt and clay fractions. EM4 and EM6 were attributed to biogenic opal and opal-CT subcomponents, respectively (Figure 4.41).

5.4 Contributions and fluxes of end-members

A six-end-member PARAFAC model was established, and 3 Asian dust sources, Taklimakan (EM1), Gobi (EM2) and Ordos (EM5); a riverine source from the Japanese islands (EM3); and two biogenic sources, diatomaceous (EM4) and Opal-CT (EM6), were identified, and their contributions were quantified (Figure 5.41 and Table S4). Sediments at IODP Site U1425 in the Japan Sea indicate that biogenic blooms (EM4+EM6) occurred from the late Miocene to Pliocene. The contribution of Asian dust (EM1+EM2+EM5) to the Japan Sea was low in the late Miocene to Pliocene, at 20-40% (Figure 5.42(b)), and the mass accumulation rate (MAR) ranged from 0.2 to 1.5 g/cm²/ky but increased dramatically to 70% in the Pleistocene, with a maximum

MAR of $3.1 \text{ g/cm}^2/\text{ky}$ at 0.03 Ma (Figure 5.43(a)). The total Asian dust MAR in this study is similar to previous estimations of $1\text{-}3 \text{ g/cm}^2/\text{ky}$ at ODP Site 797 (Irino and Tada, 2000) and $2\text{-}3 \text{ g/cm}^2/\text{ky}$ at MD01-2407 (Nagashima et al., 2007) in the Japan Sea and is also consistent with the exponential decreasing trend along the eastward transportation of Asian dust (Zhang et al., 2016) during the late Quaternary.

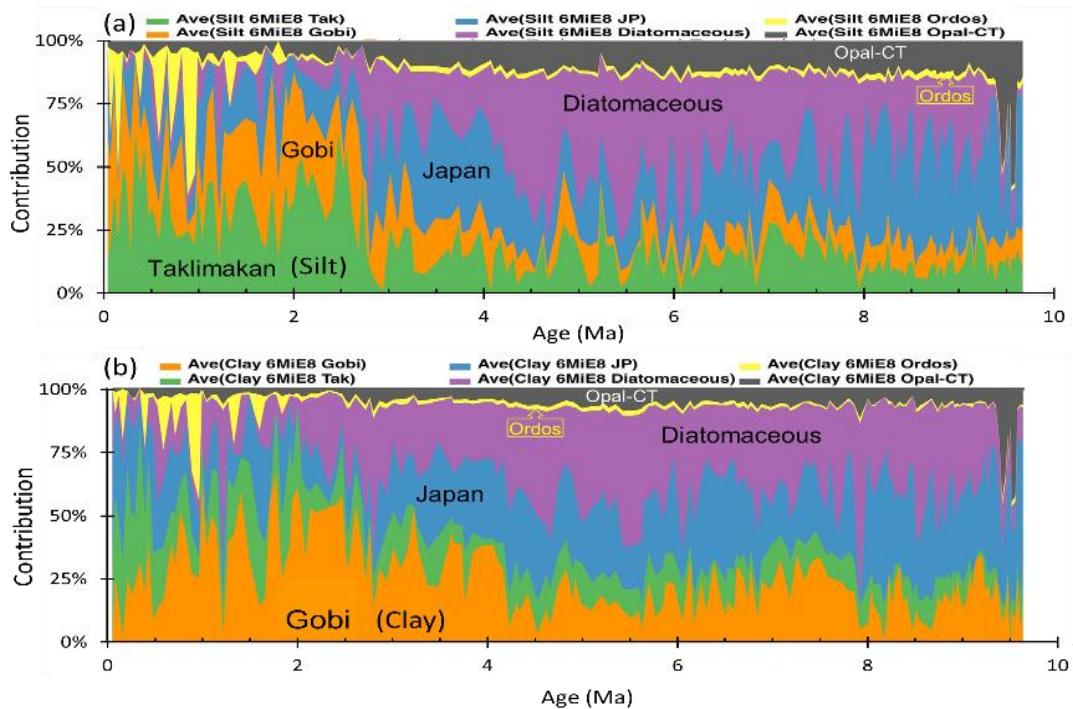


FIGURE 5.41 Temporal variation in the contribution of end-members 1-6 in (a) silt and (b) clay fractions at IODP Site U1425 in the Japan Sea. Green: Taklimakan (EM1), orange: Gobi (EM2), blue: Japanese islands (EM3), yellow: Ordos (EM5), purple: Diatomaceous (EM4), and gray: Opal-CT (EM6).

Taklimakan Desert (TAK: EMI)

The contribution of Taklimakan dust to the Japan Sea ranged from 10-50% (Figure 5.41(a) and 5.41(b)), and the flux increased gradually in the Pleistocene to a maximum of $1.67 \text{ g/cm}^2/\text{ky}$ at 0.29 Ma , in which both silt and clay fractions had the highest MAR at the same time (Figure 5.43(b) and 5.43(d)). TAK became the main dust contributor to the Japan Sea from 0.5 to 0.16 Ma and $5.6\text{-}4 \text{ Ma}$ (Figure 5.42(a)). Lower flux

occurred in the late Miocene to Pliocene at $<0.5 \text{ g/cm}^2/\text{ky}$. The temporal distribution of the Taklimakan dust flux in the silt and clay fractions had a similar trend and maintained a regular ratio between them from the late Miocene to the present, although the silt fraction dominated the total Taklimakan dust fractions (Figure 5.43(d)), which implies that both silt and clay dust were transported at the same time by the same transportation mode of wind.

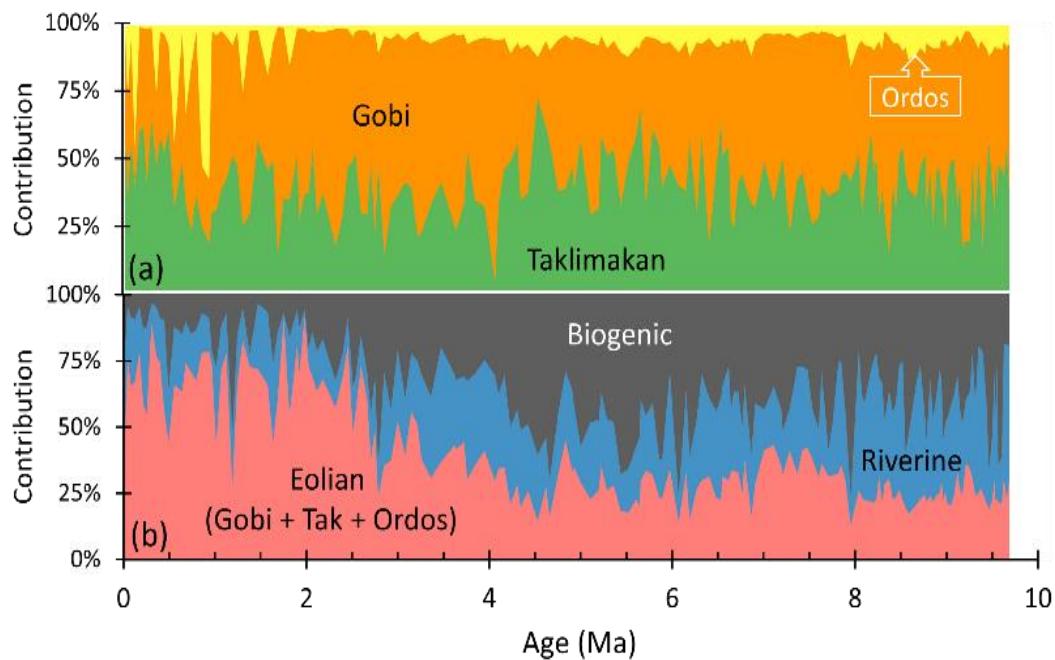


FIGURE 5.42 Relative contribution of end-members (EMs) in sediments at IDOP Site U1425 in the Japan Sea normalized for (a) total Asian dust, including Taklimakan (EM1), Gobi (EM2) and Ordos (EM5), and (b) total sediment subcomponents, including eolian (EM1, EM2, and EM5), riverine (EM3), and biogenic (EM4 and EM6) sediment subcomponents.

Gobi Desert (Gobi: EM2)

The contributions of Gobi dust to the Japan Sea have ranged from 20-50% since 10 Ma (Figure 5.41(a) and 5.41(b)). The highest MAR occurred 0.96 Ma at $1.7 \text{ g/cm}^2/\text{ky}$ and was the lowest in the Pliocene (Figure 5.43(b)). The clay-sized Gobi dust had a higher flux than the silt-sized fraction from 9.24 to 0.73 Ma. The highest MAR in the clay fraction was $1.56 \text{ g/cm}^2/\text{ky}$ in 0.96 Ma. The MAR of the silt fraction was

generally low at <0.8 g/cm²/ky. The temporal variation in Gobi fluxes in silt and clay fractions showed a great difference in pattern and trend, as well as dust quantity (Figure 5.43(c)), which implied that both were transported by different wind circulation from a dust source area to the Japan Sea.

Ordos Plateau (Ordos: EM5)

Ordos dust to the Japan Sea has been significant in the Pleistocene since 2 Ma with a 10-40% contribution (Figure 5.41(a) and 5.41(b)). The highest MAR occurred 0.92 Ma with 1.26 g/cm²/ky and decreased to 0.83 g/cm²/ky in the Holocene (Figure 5.43(b)). The silt fraction was dominant with Ordos dust since 1 Ma (Figure 5.43(e)).

Japan Island Arc (Japan: EM3)

The contribution of riverine input from Japanese islands to the Japan Sea was high during the late Miocene at 30-50% and decreased significantly in the Pleistocene (Figure 5.42(b)). The highest MAR was 2.98 g/cm²/ky, occurring 9.66 Ma but decreased to 0.2-1.2 g/cm²/ky from 8 Ma to the present, showing minima 8-7, 6-4.5, 2.7-2 and 1.5-0.6 Ma (Figure 5.43(a)).

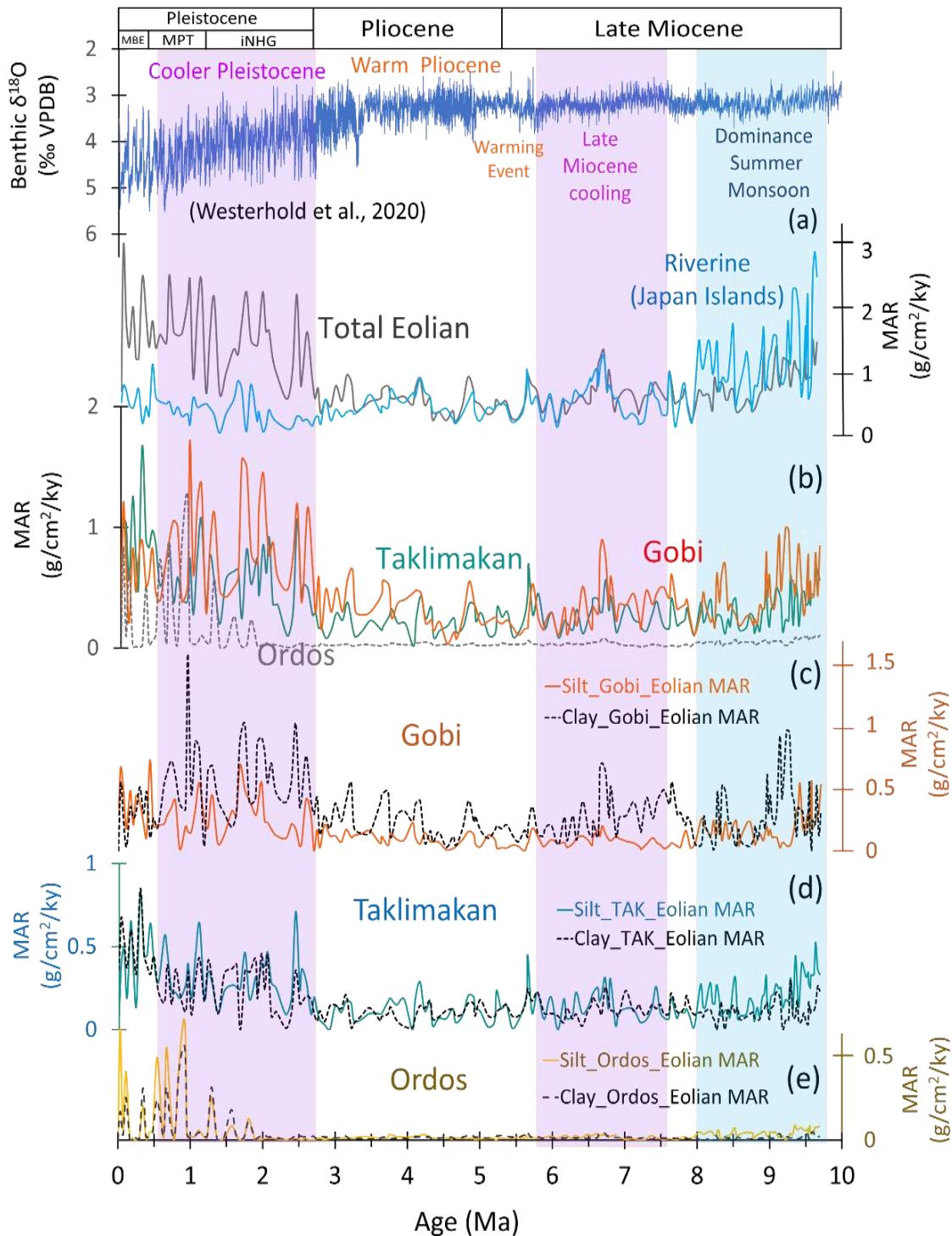


FIGURE 5.43 Temporal variations in eolian mass accumulation rates (MARs) at IODP Site U1425 in the Japan Sea since 10 Ma. (a) Total eolian and riverine materials and (b) total eolian material from Taklimakan, Gobi and Ordos; and dust fluxes in silt and clay fractions for (c) Gobi, (d) Taklimakan, and (e) Ordos. Benthic oxygen isotope data from Westerhold et al. (2020) are also shown at the top for comparison. The purple shading shows late Miocene Global cooling and Northern Hemisphere Glaciation. The blue shading indicates the dominance of the summer monsoon in the late Miocene.

Chapter 6. Discussion

In this research, we found Gobi silt and clay fluxes show significant differences in pattern, trend and quantity, while Taklimakan silt and clay fluxes show a similar trend and maintain a regular ratio from the late Miocene to the Pleistocene. These differences in flux variabilities might be controlled by the different modes of transportation paths from source regions to the Japan Sea during the past 10 Ma.

6.1 Grain-size variation of dust from Taklimakan and Gobi

Figure 5.43(d) shows that Taklimakan dust was generally dominated by the silt fraction (Silt_TAK) from the late Miocene to the Pleistocene, which may reveal a typical characteristic of a long-range transport of dust from Taklimakan by the westerly wind. The atmospheric circulation in the Tarim Basin is controlled by high-level westerly winds, which are responsible for long-range dust transport (>5000 km) to the Japan Sea and Pacific Ocean (Sun et al., 2001). Desert materials in Taklimakan were entrained to an elevation of >5 km in the troposphere and were transported by rapid and strong westerly winds to downwind areas (Figure 6.11). During transportation, no dust fall was observed in the proximal areas of Chinese loess until it traveled downwind to the Japan Sea (Tsai et al., 2008). Hence, all entrained materials in the deserts were transported directly downwind which is supported by the result of a climate model study in the Asian dust transportation by westerly (Yang et al., 2021). Typical 4 μm far-traveled Asian dust and larger particles (>10 μm) were transported to Canada in April 2001, indicating a strong and rapid westerly jet in the troposphere (Zdanowicz et al., 2007).

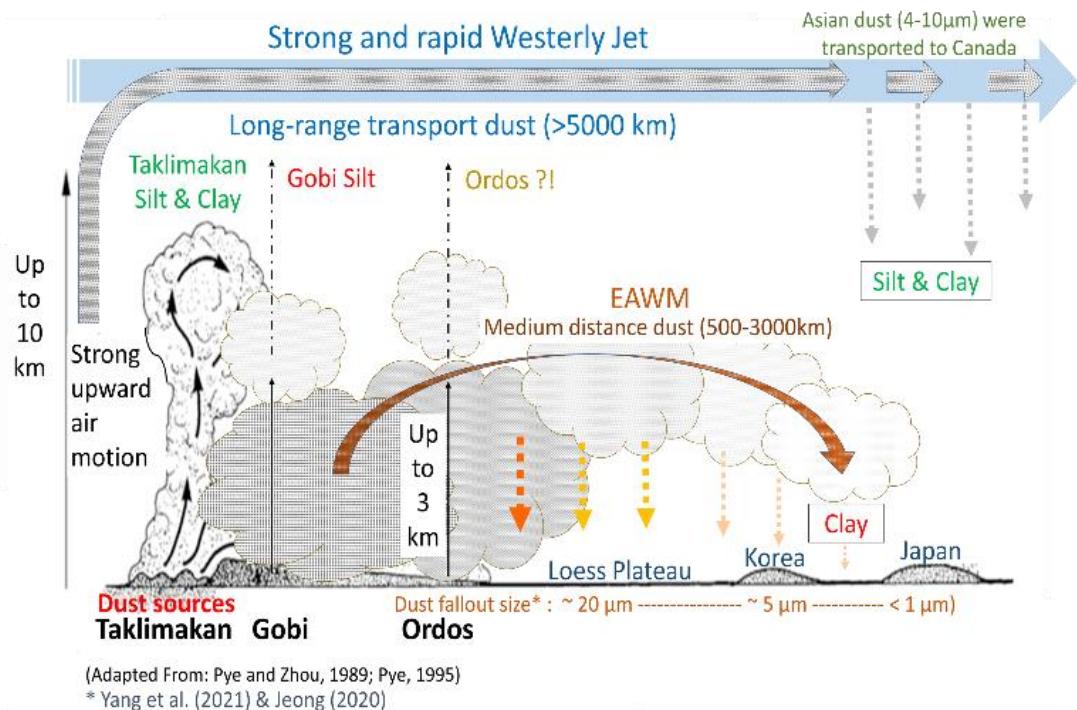


FIGURE 6.11 Schematic figure depicting the two dust transport modes, westerlies and low-level EAWM, as well as their depositional grain-size variations in the Japan Sea (modified from Pye and Zhou, 1989; Pye, 1995). Dust fallout sizes by the EAWM are according to Yang L. et al. (2021) and Jeong (2020).

In the case of Gobi materials, the clay-size fraction (Clay_Gobi) had a higher flux than the silt-size fraction (Silt_Gobi) from 9.24 to 0.73 Ma (Figure 5.43(c)). Because temporal flux variations of silt (Silt_Gobi) and clay (Clay_Gobi) fractions showed very different patterns and trends as well as their magnitude, silt and clay fractions were considered to be transported by different wind circulation modes from Gobi to the Japan Sea. Most desert materials in the Mongolian Gobi were entrained to an elevation of <3 km and carried by EAWM winds with a medium distance (500 to 3000 km) to the Chinese Loess, southeastern China, and the Japan Sea (Sun, 2002). When Gobi dust materials ascend to the highest elevation, the materials begin to descend and cause heavy dust to fall in the proximal region of CLP (Tsai et al., 2008; Sun, 2002). According to Liu (1988), CLP has been subdivided into three zones of sandy loess,

loess, and clayey loess zones from northwest to the south depending upon particle size which were sorted by wind blowing from the desert sources. Jeong et al. (2011) also found coarse silt was progressively replaced by the fine silt and clay from west to eastward on CLP. Coarse particle (5-20 μm) dust could be deposited on CLP (Yang L. et al., 2021), 2-5 μm dust has settled on the Korean Peninsula (Jeong, 2020), and only fine-grained dust (clay) could be suspended further to the Japan Sea and Southeast Asia (Yang et al., 2021; Figure 6.11). Although the effects of weathering should be considered, the gravitational settling during dust transportation by low-level winds of the EAWM is obvious. Therefore, it is reasonable to consider that clay-sized Gobi dust dominated from 9.24 to 0.73 Ma in the Japan Sea was mainly transported by low-level atmospheric circulation by EAWM winds (Figure 5.43 (c)). Figure 6.21 and 6.22 show that cooling events and trends found in the benthic $\delta^{18}\text{O}$ record (Westerhold et al., 2020) correspond well with the periods showing the higher flux of Gobi clay, which suggest that EAWM might be enhanced during cooling phase and glacial periods since 10 million years ago.

On the other hand, silt-sized Gobi dust in the Japan Sea revealed a similar MAR pattern and trend as those of Taklimakan dust, implying that coarser Gobi dust (silt) was mainly transported by westerlies to the Japan Sea. Sun (2002) and Tsai et al. (2008) also observed dust storm events in the Mongolian Gobi Deserts in which dust was entrained to the troposphere and transported by westerly winds to the Japan Sea and Pacific Ocean or America.

6.2 Detrital provenance variability in Japan Sea sediments

Late Miocene

High riverine flux from Japanese islands 9.6-8 Ma indicates a strong summer monsoon climate in East Asia, which is consistent with the clay mineral study at Linxia Basin in NE Tibet (Yang et al., 2021). A provenance shift occurred from 8-7 Ma; the Japanese riverine flux decreased to the lowest value, $0.2 \text{ g/cm}^2/\text{ky}$, and the eolian flux increased from 7.8-6.6 Ma in the Japan Sea (Figure 5.43(a)). Increase in eolian sedimentation at 8-7 Ma were also confirmed in the Loess Plateau, NE Tibetan Plateau, the Japan Sea and the North Pacific (Shen et al., 2017; Guo et al., 2002; Yang et al., 2017b; Rea et al., 1998).

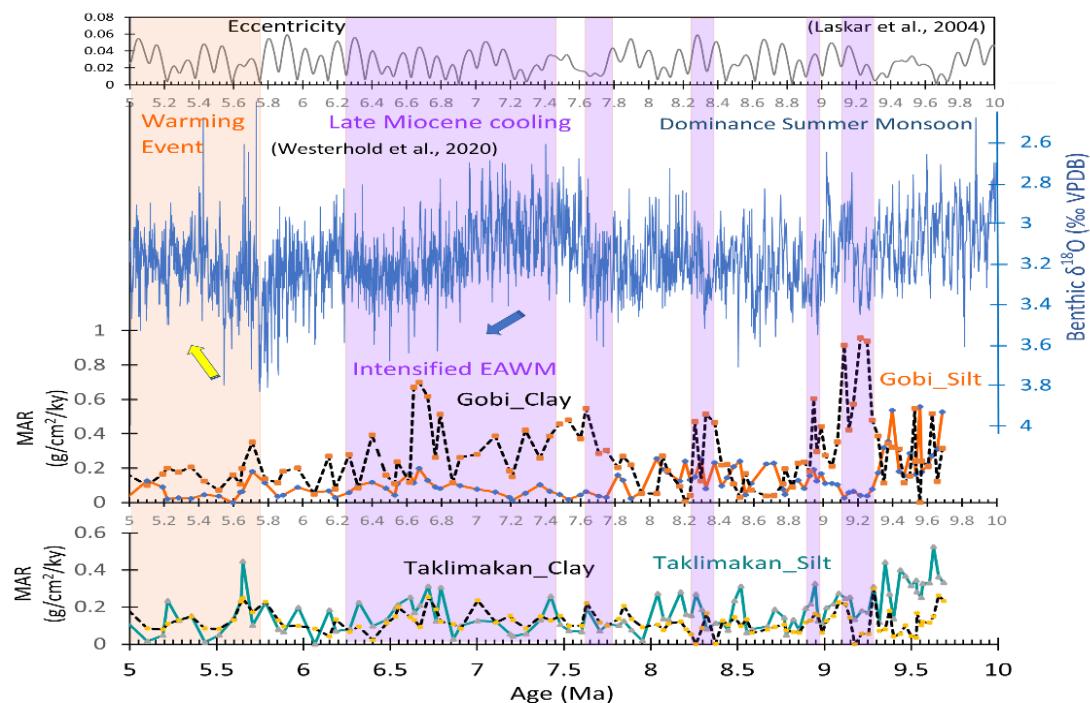


FIGURE 6.21 Dust fluxes in silt and clay fractions of Taklimakan and Gobi dust at IODP Site U1425 compared with benthic oxygen isotope data from Westerhold et al. (2020) and orbital eccentricity (Laskar et al., 2004) from 10 to 5 Ma. Gobi clay was associated with cooling (purple shading), and Taklimakan dust was associated with a warmer climate (orange shading) in the late Miocene to Pliocene. Arrows indicate the change in benthic oxygen isotope data during the late Miocene.

These data imply that Asian aridification approximately 8 Ma coincided with late Miocene global cooling (LMGC) between ~7 and ~5.5 Ma and with a short period of Northern Hemisphere glaciation between 6 and 5.5 Ma (Figure 6.21; Herbert et al., 2016; Holbourn et al., 2018). Eolian flux generally was low but occasionally increased to 1.39 and 1.02 g/cm²/ky in 6.72 and 5.65 Ma, respectively, suggesting the aridity of the Asian interior and the intensification of the winter monsoon in East Asia (Figure 5.43(a); Matsuzaki et al., 2020). This intensification of the EAWM was also confirmed by the increase of grain size in the eolian deposits on the CLP in 7.4 Ma and 5.3 Ma (Fan et al., 2006).

Pliocene

The Warming Event appeared in the early Pliocene (Holbourn et al., 2018; Ao et al., 2021) after the late Miocene cooling was characterized by very low fluxes of eolian and riverine material, 0.2-1 g/cm²/ky (Figure 5.43(a)), but had the highest biogenic contributions to the Japan Sea (Figure 5.42(b)). Shifts in dust provenance sources occurred from Gobi to Taklimakan during the warming event in 5.6-4 Ma (Figure 5.42(a)). According to a study by Ao et al. (2021) for eolian records in CLP, this warming event only increased summer monsoon moisture in East Asia, but enhanced aridification by increasing evaporation than precipitation in most parts of Central Asia. The riverine input from Japan islands did not show a prominent increase during the warming period (Figure 5.43(a)), suggesting continuously invariable summer precipitation at the eastern margin of Asia at that time. A gradual increase of eolian

material 3.2 Ma with decrease in biogenic materials followed by sudden cooling 3.3 Ma (Figure 5.42(b) and 6.22).

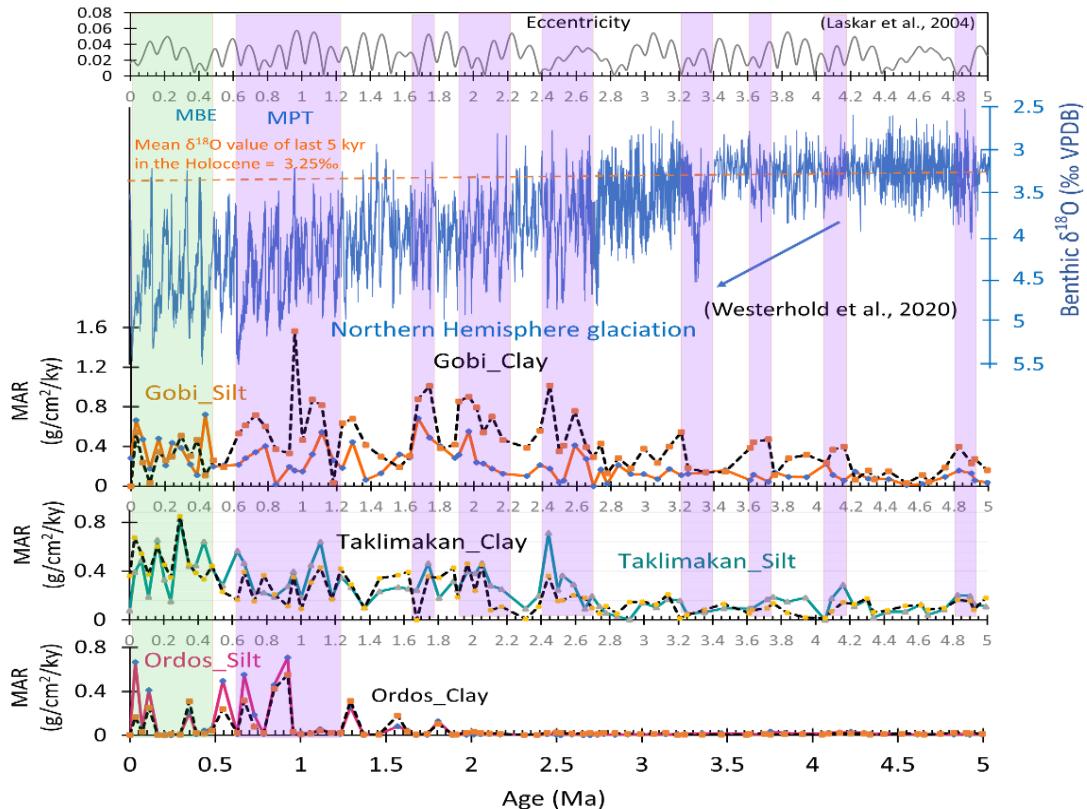


FIGURE 6.22 Dust fluxes in silt and clay fractions of Taklimakan, Gobi, and Ordos dust at IODP Site U1425 compared with benthic oxygen isotope data from Westerhold et al. (2020) and orbital eccentricity (Laskar et al., 2004) from 5 to 0 Ma. Gobi clay was associated with cooling/glaciation (purple shading), and Taklimakan dust was associated with a warmer/interglacial climate (green shading) in the Pliocene to Pleistocene. The orange dashed line is the mean $\delta^{18}\text{O}$ for the last 5 kyr in the Holocene (Raymo et al., 2018). The blue arrow indicates the substantial increase in benthic oxygen isotopes during the late Pliocene.

Pleistocene

A substantial increase in eolian MARs to $2.5 \text{ g/cm}^2/\text{ky}$ occurred after 2.7 Ma (Figure 5.43(a)) in the Japan Sea during the intensified Northern Hemisphere Glaciation (iNHG), which established a cold and dry environment, accelerating sediment erosion in the Central Asian Mountain ranges and strengthening the atmospheric circulation systems in the Northern Hemisphere. Low temperatures during iNHG caused a greater

equator-to-pole temperature gradient and stronger thermal gradients along polar frontal systems (CLIMAP, 1976), resulting in stronger westerlies (Gates, 1976) and intensification of winter monsoon winds.

Gobi was the most significant dust contributor to the Japan Sea in the Early Pleistocene but decreased sharply after 0.95 Ma (Figure 6.22). Ordos dust increased only 0.95-0.85 Ma during the mid-Pleistocene transition (MPT) due to enhanced aridification in Northwest China which is evidenced by Li et al. (2015) for the Ulan Buh Desert and southern Inner Mongolia. The formation of deserts in the Mu Us and southern Inner Mongolia could be as old as the MPT (1.1-0.9 Ma) (Li et al. (2015) resulting in increased dust materials from the Ordos Plateau (Figure 6.22).

Taklimakan became the main dust contributor to the Japan Sea after 0.5 to 0.16 Ma during the Mid-Brunhes Event (MBE) (Figure 5.43(b) and 6.22), which corresponded to a period when Earth eccentricity was close to 0 (Berger, 1978; Laskar et al., 2004), and the benthic $\delta^{18}\text{O}$ record shows the largest variation between 3.2 and 5.4‰ in the Pleistocene (Westerhold et al., 2020) (Figure 6.22).

6.3 Implications for atmospheric circulation variability during the past 10 million years

Based on our dust records, we reconstructed the variability of paleoatmospheric circulation of westerly winds by using fluxes from Taklimakan, as well as silt-sized dust from Gobi (Gobi silt). The EAWM was inferred from clay-sized dust from Gobi (Gobi clay). The EASM and aridity changes in East Asia were inferred from Japanese riverine sediments during the last 10 million years (Figure 6.31). Therefore, four paleo

climatic significances were ascertained for the last 10 Ma years in the Japan Sea sediments and described in the following subsections.

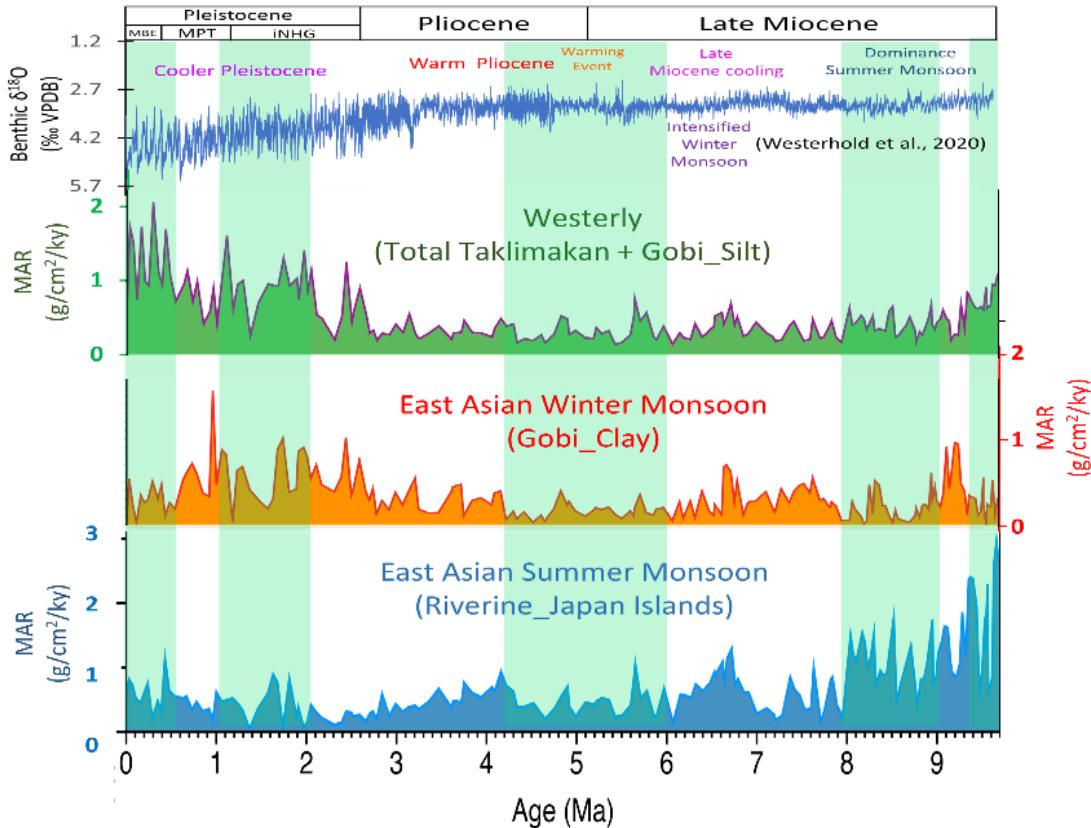


FIGURE 6.31 Variability of paleoatmospheric circulation of westerlies, East Asian winter monsoon (EAWM) and East Asian summer monsoon (EASM) reconstructed from the fluxes of eolian and riverine input to IODP Site U1425 in the Japan Sea since 10 Ma. The green shading shows the dominance of westerly winds. Benthic oxygen isotope data from Westerhold et al. (2020) are also shown at the top for comparison.

Dominance of the EASM 9.6-8 Ma

The dominance of the EASM in East Asia from 9.6-8 Ma suggested warm air and moisture from the southeast, which was inferred from the high Japanese riverine flux record (Figure 6.31). The eolian deposits accumulated on the CLP had a low deposition rate and strong pedogenesis, which revealed intensified precipitation during this period (Sun et al., 2015). In the same period, TAK and Gobi silt dust dominated 9.6-9.4 and 8.9-8 Ma, indicating that westerly winds were strong. In addition, high flux of Gobi

clay 9.3-8.9 Ma suggested that the EAWM winds were enhanced during a less pronounced, late Miocene cooling step ~9.0 Ma (Figure 6.21; Holbourn et al., 2013). The high-frequency fluctuations in dust grain size and dust source materials during this period suggested that unstable atmospheric circulation prevailed in East Asia during the dominance of the EASM in the late Miocene (Figure 6.21).

Increase aridification in inland Asia after 8 Ma

The EASM was strong 9.6-8 Ma, but relatively weak in 8-7, 6-4.5 and 3-2 Ma in the Japan Sea region, which might contradict some CLP records in China which proposed EASM increase in the Miocene–Pliocene climate transition (Ao et al., 2021). The evolution of EASM in East Asia during this period and its controlling mechanisms remain controversial (Wang et al., 2021) and complicated because EASM precipitation also depended on the variation of regional and local environments. The controls on aridification in inland Asia at ~8 Ma might involve the retreat of EASM, global cooling, and the uplift of the Tibetan Plateau. It seems widely accepted that the uplift of the Tibetan Plateau played a significant role in the long-term aridification of inland Central Asia during Cenozoic by modulating the atmospheric circulation and blocking moisture from southeast Asia (Tada et al., 2016; An et al., 2001). Based on a climate modelling study, Zhang et al. (2022) demonstrated the growth of the Tibetan Plateau and the uplift of the northern Tibetan Plateau expanded drylands in central Asia to the north of ~40°N resulting in the same dryland zone at higher latitudes as the present. In the latest Miocene or the Pliocene, the northern and eastern part of the Tibetan Plateau

and adjacent mountain ranges were still uplifted (Zheng et al., 2006) that may have intensified aridity in inland and Central Asia at 8 Ma and afterward.

Global cooling enhances EAWM

The dominance of the EAWM was linked to global cooling, such as LMGC (Figure 6.21), as well as the increase in ice sheets in the Northern Hemisphere, such as iNHG and MPT (Figure 6.22). This observation suggested that global cooling could have strengthened the low-level EAWM winds (Sun et al., 2020) and increased dust emissions from the Mongolian Gobi and northern China deserts to the Japan Sea.

During the late Miocene cooling period, Gobi clay significantly contributed to the total eolian flux in the Japan Sea (Figure 6.21) from 7.8 to 6 Ma, suggesting Asian aridification and intensification of the EAWM. Although this fast late Miocene cooling could be triggered by an abrupt decrease in CO₂ or the uplift of the Tibetan Plateau, the true mechanism is still unclear and under hot debate (Shen et al., 2017; Fan et al., 2006; Yang Y. et al., 2021; Lu et al., 2022). Slight increases in Gobi clay 4, 3.6 and 3.2 Ma (Figure 6.22) suggest progressive global cooling, which was associated with the occurrence of ice-rafted debris (IRD) at the north pole in the late Pliocene until the beginning of the Northern Hemisphere Glaciation (Raymo et al., 2018; Thiede et al., 1998).

During the iNHG, increasing Siberian High activity strengthened the EAWM winds (Ding et al., 1995) in northern China, which increased dust storm events as well as dust transportation from the Gobi and northern China deserts. Gobi clay increased substantially from the late Pliocene to the early Pleistocene until 0.6 Ma (Figure 6.22).

On the other hand, some studies suggested that the strong Siberian High and EAWM forced the westerly winds to migrate southward (Ono and Irino, 2004; Kapp et al., 2011; Pullen et al., 2011) during the strong glacial and cooling periods. Another study on the loess in the Tarim basin suggests that expansion of Northern Hemisphere ice sheets could intensify the strength of the westerly jet and push it southward, which enhanced aridity and intensified dust storm activity in the Tibetan Plateau (Fang et al., 2020). The increase in accumulation of Taklimakan dust and Gobi silt in the Japan Sea indicated a prolonged interval of westerly wind blowing on the inland desert area since the early Pleistocene (Figure 6.22).

Role of westerly for dust transport

During the warmer periods in the Warming Event in the late Miocene to early Pliocene and the Late Pleistocene, weakened cold air masses from the Siberian High could permit a northward migration of westerly winds in an earlier season (Abell et al., 2021; Yang et al., 2017a; Lu et al., 2020). Dust provenance changes from Gobi to TAK 5.8-4.2 Ma (Figure 5.42(a) and 6.31) during the Warming Event revealed the dominance of westerly winds but was characterized by low dust deposition in the Japan Sea.

The influence of westerlies increased gradually, reflecting the increasing fluxes of TAK and Gobi silt from 2.5 to 1.1 Ma and became dominant in the Late Pleistocene from 0.5 Ma to the present. The early northward migration of westerlies increased the duration of westerlies over the Taklimakan Desert and resulted in an increase in the length of season for dust storms and dust transportation from the Taklimakan Desert,

as well as deserts in northern China, to the Japan Sea. A similar situation can currently be observed with a longer dust season from April to August (Schiemann et al., 2009). During the dominance of westerlies, Gobi silt also increased accordingly and had a higher flux than Gobi clay from 0.5 Ma to the present (Figure 6.22).

Under recent global warming conditions, the dominance of westerly winds is obvious, which calls our attention to the influence on the regional and local climate. Extreme weather events such as heat waves and large storms are likely to become more frequent or more intense in the coming future. Although the behaviors of regional climate and weather are complicated to understand and forecast, long-term global climate changes in the past, such as the warming in the Pliocene or the dominance of the summer monsoon in the late Miocene, may provide the basis to solve the riddle and the problems that we are facing now or will face in the future.

Chapter 7. Conclusions

This work provides empirical evidence for the major changes in paleoatmospheric circulation of westerlies, the EAWM and aridity changes in the dust source region since 10 Ma using a sediment sequence from IODP Site U1425 in the Japan Sea. Mineralogical analyses of the sediment at IODP Site U1425 effectively established a proxy for the eolian provenance study. The mineral compositions could distinguish the detrital sources, such as dust from Taklimakan and Gobi and riverine input from the Japanese islands.

In order to solve the problems in eolian provenance, we applied PARAFAC end-member modelling, which is a powerful analytical tool to quantitatively differentiate eolian dust sources, such as those in Taklimakan, Gobi, and the Ordos Plateau, as well as the other subcomponents in the sediment. New dust records of Gobi, Taklimakan and Ordos source changes in the silt and clay fractions resolved the transport process and distinguished the effects of climatic intervention on dust transportation changes in the past.

There are four paleo climatic significances for the last 10 Ma years in the Japan Sea sediments. The dominance of the EAWM was linked to the LMGC, iNHG and MPT associated with global cooling and the increase in ice sheets in the Northern Hemisphere, which strengthened the low-level EAWM winds and increased the deposition of fine clay dust from Mongolian Gobi. In contrast, during the dominance of westerlies in the warmer periods during the Warming Event in the late Miocene to early Pliocene, as well as in the late Pleistocene, early seasonal northward migration of

westerlies prolonged the duration of westerlies over the inland desert area, which resulted in the deposition of coarser silt-sized eolian dust from the Taklimakan and Gobi Deserts.

The EASM was strong 9.6-8 Ma, while it showed fluctuation at moderate intensity after 8 Ma to the Pleistocene in the Japan Sea region. Stepwise Asian aridification occurred 8, 6, and 2 Ma and finally induced desertification in the Ordos Plateau area. A significant increase in Ordos dust contribution to the Japan Sea in the Late Pleistocene was related to the increase in aridity in Inner Mongolia and the Ordos Plateau since 2 Ma, and the enhanced aridification 1.5-0.6 Ma induced desertification in southern Inner Mongolia during the MPT.

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References

- Abell, J.T., Winckler, G., Anderson, R.F., and Herbert, T.D., 2021, Poleward and weakened westerlies during Pliocene warmth: *Nature*, v. 589, p.70-75.
- An, Z., Kutzbach, J., Prell, W., et al., 2001, Evolution of Asian monsoons and phased uplift of the Himalaya–Tibetan plateau since Late Miocene times: *Nature*, v. 411, p. 62–66, <https://doi.org/10.1038/35075035>.
- Anderson, C. H., Murray, R. W., Dunlea, A. G., Giosan, L., Kinsley, C. W., McGee, D., and Tada, R., 2020, Aeolian delivery to Ulleung Basin, Korea (Japan Sea), during development of the East Asian Monsoon through the last 12 Ma: *Geological Magazine*, v. 157, p. 806-817,
<https://doi.org/10.1017/S001675681900013X>
- Anonymous, 1991, Petroleum geology of the Tarim basin, Science Press (seven volumes), Beijing.
- Ao, H., Rohling, E. J., Zhang, R., et al., 2021, Global warming-induced Asian hydrological climate transition across the Miocene–Pliocene boundary: *Nature Communications*, v. 12.
- Arnold, E., Leinen, M., and King, J., 1995, Paleoenvironmental variation based on the mineralogy and rock-magnetic properties of sediment from Sites 885 and 886 in: *Proceedings of the Ocean Drilling Program, Scientific Results, Ocean Drilling Program*, p. 231–245.
- Berger, A., 1978, Long term variations of daily insolation and Quaternary climatic changes: *Journal of Atmospheric Science*, v. 35, p. 2362-2367.

- Berger, W.H., and Jansen, E., 1994, Mid-Pleistocene climate shift: the Nansen connection: in Johannessen OM, Muench RD, Overland JE (eds) *The polar oceans and their role in shaping the global environment*, p. 295–311, American Geophysical Union, Washington, <https://doi.org/10.1029/GM085p0295>.
- Berner, E. K., and Berner, R. A., 1996, *Global Environment—Water, Air, and Geochemical Cycles*, Prentice Hall, New Jersey.
- Biscaye, P. E., 1965, Mineralogy and Sedimentation of Recent Deep-Sea Clay in the Atlantic Ocean and Adjacent Seas and Oceans: *GSA Bulletin*, v. 76, p. 803–832.
- Bro, R., 1997, PARAFAC. Tutorial and applications: *Chemometrics and Intelligent Laboratory Systems*, v. 38, p. 149-171.
- Carroll, A.R., Graham, S.A., Hendrix, M.S., Ying, D., and Zhou, D., 1995, Late Paleozoic tectonic amalgamation of northwestern China: Sedimentary record of the northern Tarim, northwestern Turpan, and southern Junggar basins: *Geological Society of America Bulletin*, v. 107, p. 571–594.
- Caves, J.K., et al., 2015, Role of the westerlies in Central Asia climate over the Cenozoic: *Earth and Planetary Science Letters*, v. 428, p. 33–43.
- Chen, J., Li, G., Yang, J., et al., 2007, Nd and Sr isotopic characteristics of Chinese deserts: Implications for the provenances of Asian dust: *Geochimica et Cosmochimica Acta*, v. 71, p. 3904-3914,
<https://doi.org/10.1016/j.gca.2007.04.033>.

CLIMAP Project Members, 1976, The surface of the ice-age earth: Science, v. 191, p. 123-137.

Dadey, K. A., Janecek, T., and Klaus, A., 1992, Dry-bulk density: Its use and determination: Proceedings of the Ocean Drilling Program, Scientific Results, v. 126.

Davidson, S.C., 2005, Sedimentation History and Provenance Analysis of a Late Mesozoic Rifting Event at Tavan Har, East Gobi, Mongolia: Frontiers, The Interdisciplinary Journal of Study Abroad, p. 91-122.

Ding, Z., Liu T., Rutter N. W. et al., 1995, Ice-Volume Forcing of East Asian Winter Monsoon Variations in the Past 800,000 Years: Quaternary Research, v. 44, no. 2, p. 149-159.

Ehrmann, W., 1998, Implications of late Eocene to early Miocene clay mineral assemblages in McMurdo Sound (Ross Sea, Antarctica) on paleoclimate and ice dynamics: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 139, p. 213-231.

Fan, M., Song, C., Dettman, D.L., Fang X., and Xu, X., 2006, Intensification of the Asian winter monsoon after 7.4 Ma: Grain-size evidence from the Linxia Basin, northeastern Tibetan Plateau 13.1 Ma to 4.3 Ma: Earth and Planetary Science Letters, v. 248, p. 186-197, <https://doi.org/10.1016/j.epsl.2006.05.025>.

Fang, X., An, Z., Clemens, S. C., et al., 2020, The 3.6-Ma aridity and westerlies history over midlatitude Asia linked with global climatic cooling: Earth, Atmospheric, and Planetary Sciences, v. 117 (40),

<https://doi.org/10.1073/pnas.1922710117>.

Gates, W. L., 1976, Modelling the ice-age climate: *Science*, v. 191, p. 1138–1144.

Guo, Z. T., Sun, B., Zhang, Z. S., Peng, S. Z., Xiao, G. Q. et al., 2008, A major reorganization of Asian climate by the early Miocene: *Climate of the Past*, v. 4, p. 153–174, <https://doi.org/10.5194/cp-4-153-2008>.

Guo, Z. T., Ruddiman, W. F., Hao, Q. Z., et al., 2002, Onset of Asian desertification by 22 Myr ago inferred from loess deposit in China: *Nature*, v. 416, p. 159–163.

Herbert, T.D., Lawrence, K.T., Tzanova, A., Peterson, L.C., Caballero-Gill, R., and Kelly, C.S., 2016, Late Miocene global cooling and the rise of modern ecosystems: *Nature Geoscience*, v. 9, p. 843–847, <https://doi.org/10.1038/NGEO2813>.

Holbourn, A., Kuhnt, W., Clemens, S., et al., 2013, Middle to Late Miocene stepwise climate cooling: evidence from a high-resolution deep water isotope curve spanning 8 million years: *Paleoceanography*, v. 28, p. 688–699.

Holbourn, A.E., Kuhnt, W., Clemens, S.C., Kochhann, K.G., Jöhnck, J., Lübbers, J., and Andersen, N., 2018, Late Miocene climate cooling and intensification of southeast Asian winter monsoon: *Nature Communications*, v. 9, 1584, <https://doi.org/10.1038/s41467-018-03950-1>.

Irino, T., and Tada, R., 2000, Quantification of aeolian dust (Kosa) contribution to the Japan Sea sediments and its variation during the last 200 ky: *Geochemical Journal*, v. 34, p. 59–93.

Irino, T., Tada, R., Ikebara, K., Sagawa, T., Karasuda, A., Kurokawa, S., Seki, A., and Lu, S., 2018, Construction of perfectly continuous records of physical properties for dark-light sediment sequences collected from the Japan Sea during Integrated Ocean Drilling Program Expedition 346 and their potential utilities as paleoceanographic studies: *Progress in Earth and Planetary Science*, v. 5, 23, <https://doi.org/10.1186/s40645-018-0176-7>.

Isozaki, Y., Tada, R., Sun, Y., et al., 2020, Origin of aeolian dust emitted from the Tarim Basin based on the ESR signal intensity and crystallinity index of quartz: the recycling system of fine detrital material within the basin: *Geological Magazine*, v. 157, p. 707-718.

Janecek, T. R., 1984, Eolian Sedimentation in the Northwest Pacific Ocean: A Preliminary Examination of the data from Deep Sea Drilling Project Sites 576 and 578,” in Initial Reports, G. R. Heath, L. H. Burckle et al., Eds., DSDP, v. 86, p. 589-603, Washington.

Jansen, J.H.F., Kuijpers, A., Troelstra, S.R., 1986, A mid-Brunhes climatic event: long term changes in global atmosphere and ocean circulation, *Science*, v. 232, p. 619–622, <https://doi.org/10.1126/science.232.4750.619>.

Jeong, G.Y., Hillier, S., and Kemp, R.A., 2011, Changes in mineralogy of loess-paleosol sections across the Chinese Loess Plateau: *Quaternary Research*, v. 75, p. 245–255.

Jeong, G.Y., 2020, Mineralogy and geochemistry of Asian dust: dependence on migration path, fractionation, and reactions with polluted air: *Atmospheric*

- Chemistry and Physics, v. 20, p. 7411–7428,
<https://doi.org/10.5194/acp-20-7411-2020>.
- Jia, C., Wei, G., Wang, L., Jia, D., and Guo, Z., 1997, Tectonic Characteristics and Petroleum, Tarim Basin, China: Petroleum Industry Press, Beijing (in Chinese with English abstract).
- Johnson, C. L., Webb, L. E., Graham, S. A., Hendrix M. S., and Badarch, G., 2001, Sedimentary and structural records of late Mesozoic high-strain extension and strain partitioning, East Gobi basin, southern Mongolia, in Hendrix.
- Kamikuri, S., Itaki, T., Motoyama, I., and Matsuzaki, M.K., 2017, Radiolarian biostratigraphy from middle Miocene to late Pleistocene in the Japan Sea: Paleontological Research, v. 21, p. 397–421,
<https://doi.org/10.2517/2017PR001>.
- Kapp, P., Pelletier, J. D., Rohrmann A., et al., 2011, Wind erosion in the Qaidam basin, Central Asia: Implications for tectonics, paleoclimate, and the source of the Loess Plateau: The Geological Society of America Today, v. 21, p. 4–10.
- Krumbein, W. C., and Pettijohn, F. J., 1938, Manual of sedimentary Petrography: Appleton-Century-Crofts, Inc., New York.
- Kübler, B., 1964, Les argiles, indicateurs de métamorphisme: Review Institute Francais du Pétrole, v. 19, p. 1093–1112.
- Kurokawa, S., Tada, R., Matsuzaki, K.M., Irino, T., and Johanna, L., 2019, Cyclostratigraphy of the late Miocene to Pliocene sediments at IODP sites U1425 and U1430 in the Japan Sea and paleoceanographic implications:

- Progress in Earth and Planetary Science, v. 6, 2,
<https://doi.org/10.1186/s40645-018-0250-1>.
- Kusky T. M., and Li, J., 2003, Paleoproterozoic tectonic evolution of the North China Craton: Journal of Asian Earth Sciences, v. 22, p. 383-397.
- Lamb, H. H., and Woodroffe A., 1970, Atmospheric circulation during the last ice age: Quaternary Research, v. 1, p. 29-58.
- Laskar, J., Robutel, P., Joutel, F., Gastineau, M., Correia, ACM., and Levrard, B., 2004, A long term numerical solution for the insolation quantities of the Earth: Astron Astrophys, v. 428, p. 261–285,
<https://doi.org/10.1051/0004-6361:20041335>.
- Leinen, M., and Heath, G.R., 1981, Sedimentary indicators of atmospheric activity in the northern hemisphere during the Cenozoic: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 36, p. 1-21.
- Li, G., Jin, M., Chen, X., et al., 2015, Environmental changes in the Ulan Buh Desert, southern Inner Mongolia, China since the middle Pleistocene based on sedimentology, chronology and proxy indexes: Quaternary Science Reviews, v. 128, p. 69-80, <http://dx.doi.org/10.1016/j.quascirev.2015.09.010>.
- Liu, T. S., 1988, Loess in China, China Ocean Press, Springer-Verlag, Berlin.
- Liu, Z., Zhao, Y., Colin, C., et al., 2009, Chemical weathering in Luzon, Philippines from clay mineralogy and major-element geochemistry of river sediments: Applied Geochemistry, v. 24, 11, p. 2195-2205.
- Lisiecki, L.E., and Raymo, M.E., 2005, A Pliocene-Pleistocene stack of 57 globally

distributed benthic $\delta^{18}\text{O}$ records: Paleoceanography, v. 20, PA1003,

<https://doi.org/10.1029/2004PA001071>.

Lu, H., Malusà, M.G., Zhang, Z., Guo, L., et al., 2022, Syntectonic sediment recycling controls eolian deposition in eastern Asia since ~ 8 Ma: Geophysical Research Letters, v. 49, <https://doi.org/10.1029/2021GL096789>.

Lu, Y., Dewald, N., Koutsodendris, A., Kaboth-Bahr, S., Rösler, W., Fang, X., et al., 2020, Sedimentological evidence for pronounced glacial-interglacial climate fluctuations in NE Tibet in the latest Pliocene to early Pleistocene: Paleoceanography and Paleoclimatology, v. 35,

<https://doi.org/10.1029/2020PA003864>

Matsuzaki, K.M., Suzuki, N., and Tada, R., 2020, An intensified East Asian winter monsoon in the Japan Sea between 7.9 and 6.6 Ma: Geology, v. 48, p. 919–923, <https://doi.org/10.1130/G47393.1>

Murphy, K.R., Stedmon, C.A., Graeber, D., and Bro, R., 2013, Fluorescence spectroscopy and multi-way techniques: PARAFAC, Analytical Methods, v. 5, p. 6557–6566.

Nakada, S., Yamamoto, T., and Maeno, F., 2016, The geology of Japan: The Geological Society, London.

Nagashima, K., Tada, R., Matsui, H., Irino, T., Tani, A. and Toyoda, S., 2007, Orbital- and millennial-scale variations in Asian dust transport path to the Japan Sea: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 247, p. 144–161, <https://doi.org/10.1016/j.palaeo.2006.11.027>.

Nagashima, K., Tada, R., Tani A., et al., 2007, Contribution of Aeolian dust in Japan

Sea sediments estimated from ESR signal intensity and crystallinity of quartz:
Geochemistry Geophysics Geosystem, v. 8, no. 2.

Nagashima, K., Nishido, H., Kayama M., et al., 2017, Composition of Asian dust
from cathodoluminescence spectral analysis of single quartz grains: Geology,
v. 45, p. 879-882.

Ono, Y., and Irino, T., 2004, Southern migration of westerlies in the Northern
Hemisphere PEP II transect during the Last Glacial Maximum: Quaternary
International, v. 118–119, p. 13–22.

Petschick, R., Kuhn G., and Gingele, F., 1996, Clay mineral distribution in surface
sediments of the South Atlantic: Marine Geology, v. 130, p. 203-229.

Prospero, J. M., and Bonatti, E., 1969, Continental dust in the atmosphere of the
Eastern Equatorial Pacific: Geophysical Research Letters, v. 74, p. 3362-3371,
doi:10.1029/JC074i013p03362.

Pullen, A., Kapp, P., McCallister, A. T., et al., 2011, Qaidam Basin and northern
Tibetan Plateau as dust sources for the Chinese Loess Plateau and
paleoclimatic implications: Geology, v. 39, p. 1031–1034.

Pye, K., and Zhou, L. P., 1989, Late Pleistocene and Holocene aeolian dust deposition
in North China and the Northwest Pacific Ocean: Palaeogeography
Palaeoclimatology and Palaeoecology, v. 73, p. 11-23.

Pye, K., 1995, The nature, origin and accumulation of loess: Quaternary Science
Reviews, v. 14, p. 653-667.

- Raymo, M. E., Kozdon, R., Evans, D., et al., 2018, The accuracy of mid-Pliocene $\delta^{18}\text{O}$ -based ice volume and sea level reconstructions: Earth-Science Reviews, v. 177, p. 291-302.
- Rea, D.K., and Janecek, T.R., 1981, Mass-accumulation rates of the non-authigenic inorganic crystalline (eolian) component of deep-sea sediments from the western mid-Pacific Mountains, Deep Sea Drilling Project Site 463, in Initial Reports, J. Thiede, T.L. Vallier et al., Eds, DSDP, v. 62, p. 653-659, Washington.
- Rea, D.K., Snoeckx, H., and Joseph, L.H., 1998, Late Cenozoic eolian deposition in the North Pacific: Asian drying, Tibetan uplift, and cooling of the Northern Hemisphere: Paleoceanography, v. 13, p. 215–224, <https://doi.org/10.1029/98PA00123>.
- Rittner, M., Vermeesch, P., Carter, A., Bird, A., et al., 2016, The provenance of Taklamakan desert sand: Earth and Planetary Science Letters, v. 437, p. 127-137, <https://doi.org/10.1016/j.epsl.2015.12.036>.
- Schiemann, R., Lüthi, D., and Schär, C., 2009, Seasonality and interannual variability of the Westerly Jet in the Tibetan Plateau region: Journal of Climate, v. 22, p. 2940–2957, DOI: 10.1175/2008JCLI2625.1.
- Shen, X., Wan, S., France-Lanord, C., Clift, P.D., Tada R., et al., 2017, History of Asian eolian input to the Sea of Japan since 15 Ma: Links to Tibetan uplift or global cooling?: Earth and Planetary Science Letters, v. 474, p. 296-308, <https://doi.org/10.1016/j.epsl.2017.06.053>.

- Stedmon, C. A., and Bro, R., 2008, Characterizing dissolved organic matter fluorescence with parallel factor analysis: a tutorial: Limnology and Oceanography, v. 6, p. 572-579.
- Sun, J., Zhang, M., and Liu, T., 2001, Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960-1999: Relations to sources area and climate: Journal of Geophysical Research, v. 106, p. 10,325-10,333.
- Sun, J., 2002, Provenance of loess material and formation of loess deposits on the Chinese Loess Plateau: Earth and Planetary Science Letters, v. 203, p. 845–859.
- Sun, J., Ding, Z., Xia, X., Sun, M., and Windley, B.F., 2018, Detrital zircon evidence for the ternary sources of the Chinese Loess Plateau: Journal of Asian Earth Sciences, v. 155, p. 21-34, <https://doi.org/10.1016/j.jseae.2017.10.012>.
- Sun X., and Wang P., 2005, How old is the Asian monsoon system?-Palaeobotanical records from China: Palaeogeography, Palaeoclimatology, Palaeoecology, v. 222, p. 181–222, doi:10.1016/j.palaeo.2005.03.005.
- Sun, Y., Tada, R., Chen J., et al., 2007, Distinguishing the sources of Asian dust based on electron spin resonance signal intensity and crystallinity of quartz: Atmospheric Environment, v. 41, p. 8537-8548.
- Sun, Y., Ma, L., Bloemendal, J., Clemens, S., Qiang, X., and An, Z., 2015, Miocene climate change on the Chinese Loess Plateau: Possible links to the growth of the northern Tibetan Plateau and global cooling: Geochemistry, Geophysics, Geosystems, v. 16, p. 2097–2108.

Sun, Y., Yan, Y., Nie, J., Li, G., Shia, Z., Qiang, X., Chang, H., and An, Z., 2020, Source-to-sink fluctuations of Asian aeolian deposits since the late Oligocene: Earth-Science Reviews, v. 200, 102963.

Tada, R., Murray, R.W., Alvarez-Zarikian, C., and the Expedition 346 Scientists, 2015, Proceedings of Integrated Ocean Drilling Program, volume 346, College 233 Station, TX (Integrated Ocean Drilling Program).

<https://doi.org/10.2204/iodp.proc.346.106.2015>

Tada, R., Zheng H., and Clift P. D., 2016, Evolution and variability of the Asian monsoon and its potential linkage with uplift of the Himalaya and Tibetan plateau: Progress in Earth and Planetary Science, v. 3, no. 4.

Tada, R., Irino, T., Ikehara, K., Karasuda, A., Sugisaki, S., Xuan, C., Sagawa, T., Itaki, T., Kubota, Y., Lu, S., Seki, A., Murray, R.W., Alvarez-Zarikian, C., Anderson, W.T., and Expedition 346 Scientists, 2018, High-resolution and high-precision correlation of dark and light layers in the Quaternary hemipelagic sediments of the Japan Sea recovered during IODP Expedition 346: Progress in Earth and Planetary Science, v. 5, 19,

<https://doi: 10.1186/s40645-018-0167-8>.

Thiede, J., Winkler, A., Wolf-Welling T., et al., 1998, Late Cenozoic history of the polar North Atlantic: Results from ocean drilling: Quaternary Science Reviews, v. 17, pp. 185-208.

Tsai, F., Chen, George T. J., Liu, T.H., Lin, W.D., and Tu, J.Y., 2008, Characterizing the transport pathways of Asian dust: Journal of Geophysical Research, v.

113, D17311, <https://doi:10.1029/2007JD009674>.

Wang, M. Y., et al., 2021, Late Miocene-Pliocene Asian summer monsoon variability linked to both tropical Pacific temperature and Walker Circulation: Earth and Planetary Science Letters, v. 561.

Wang, Y. Q., Zhang, X. Y., Gong, S.L., Zhou C. H., et al., 2008, Surface observation of sand and dust storm in East Asia and its application in CUACE/Dust: Atmospheric Chemistry and Physics, l. 8, p. 545–553,
<https://doi.org/10.5194/acp-8-545-2008>.

Wang, Z., Wu, J., Lu, X., Zhang J., and Liu, C., 1986, An outline on the tectonic evolution of the Tian Shan of China: Bulletin of the Institute of Geology, Chinese Academy of Geological Sciences, v, 15, p. 81-92 (in Chinese with English abstract).

Watson, M. P., Hayward, A. B., Parkinson, D.N., and Zhang, ZH. M., 1987, Plate tectonic history, basin development and petroleum source rock deposition onshore China: Marine and Petroleum Geology, v. 4, p. 205–225.

Westerhold et al., 2020, An astronomically dated record of Earth's climate and its predictability over the last 66 million years: Science, v. 369, p.1383–1387.

Yang, L., Shi, Z., Sun, H., et al., 2021, Distinct effects of winter monsoon and westerly circulation on dust aerosol transport over East Asia: Theoretical and Applied Climatology, v. 144, p. 1031–1042,
<https://doi.org/10.1007/s00704-021-03579-z>.

Yang, Y., Yang, R., Li, X., et al., 2017a, Glacial-interglacial climate change on the

northeastern Tibetan Plateau over the last 600kyr: Palaeogeography,

Palaeoclimatology, Palaeoecology, v. 476, p. 181-191,

<https://doi.org/10.1016/j.palaeo.2017.04.007>.

Yang, Y., Galy, A., Fang, X., et al., 2017b, Eolian dust forcing of river chemistry on

the northeastern Tibetan Plateau since 8 Ma: Earth and Planetary Science

Letters, v. 464, p. 200-210, <https://doi.org/10.1016/j.epsl.2017.02.009>.

Yang, Y., Ye, C., Galy, A., et al., 2021, Monsoon enhanced silicate weathering as a

new atmospheric CO₂ consumption mechanism contributing to fast late

Miocene global cooling: Paleoceanography and Paleoclimatology, v. 36,

<https://doi.org/10.1029/2020PA004008>.

Yu, Y., Kalashnikova, O. V., Garay, M.J., and Notaro, M., 2019, Climatology of

Asian dust activation and transport potential based on MISR satellite

observations and trajectory analysis: Atmospheric Chemistry Physic, v. 19, p.

363–378, <https://doi.org/10.5194/acp-19-363-2019>.

Zdanowicz, C., Hall, G., Vaive, J., Amelin, Y., Percival, J., Girard, I., Biscaye, P., and

Bory, A., 2007, Asian dustfall in the St. Elias Mountains, Yukon, Canada:

Geochimica et Cosmochimica Acta, v. 70, p. 3493–3507.

Zhang, R., Zhang, Z., Jiang, D., Ramstein, G., Dupont-Nivet, G., and Li, X., 2022,

Tibetan Plateau made central Asian drylands move northward, concentrate in

narrow latitudinal bands, and increase in intensity during the Cenozoic:

Geophysical Research Letters, v. 49, <https://doi.org/10.1029/2021GL093718>.

Zhang, W., Chen, J., Ji, J., and Li, G., 2016, Evolving flux of Asian dust in the North

Pacific Ocean since the late Oligocene: Aeolian Research, v. 23, p. 11-20,

<https://doi.org/10.1016/j.aeolia.2016.09.004>.

Zheng, D., Zhang, P.Z., Wan, J., Yuan, D., et al., 2006, Rapid exhumation at ~8 Ma

on the Liupan Shan thrust fault from apatite fission-track thermochronology:

Implications for growth of the northeastern Tibetan Plateau margin: Earth and

Planetary Science Letters, v. 248, p. 198-208,

<https://doi.org/10.1016/j.epsl.2006.05.023>.

Table caption

Table S1: List of samples used in this study. Sample numbers with stars (*) indicate that the sample was out of the splice. Since all samples were collected from IODP Site 1425 Hole D, the stratigraphic position of the sample out of the splice was projected to the corresponding horizon on the splice based on the core photograph, and the revised core splice depth below the sea floor (m CCSF-D_Patched_rev20170309) was calculated. Linear sedimentation rates (cm/ky) and dry bulk density (g/cm³) at each sample horizon are also shown.

Table S2: List of potential detrital source materials from the Taklimakan Desert, Mongolian Gobi Desert, Ordos Plateau, and Japanese islands.

Table S3: Peak intensities (cps) of minerals, illite FWHM ($\Delta \text{ }^{\circ}2\theta$) and illite chemical index ratio of 5 Å/10 Å peak areas identified by powder X-ray diffraction (XRD) in the silt and clay fractions.

Table S4: Contents (%) of silt (>4 µm) and clay (<4 µm) in dry sediment sample, and contribution (%) of each end-member in silt and clay fractions estimated by PARAFAC modelling. End-member contributions are expressed as the average and standard deviation of 20 times trial of PARAFAC calculation.

List of Appendix

Appendix 1: The PARAFAC modelling steps, matrix calculation and RSS comparison.

Table S2: List of potential detrital source materials from the Taklimakan Desert, Mongolian Gobi Desert, Ordos Plateau, and Japanese islands.

Region	Sample number	Latitude	Longitude
<u>Gobi Desert</u>			
	GB07-51	41°16'08.9"N	99°28'19.2"E
	GB07-52	40°53'20.2"N	99°23'15.5"E
	HT07-125	43°03'39.9"N	90°45'38.9"E
	HT07-124	42°55'03.5"N	89°46'08.5"E
	HT07-129	42°58'38.5"N	93°15'54.3"E
<u>TG-Tengger Desert</u>			
	TG07-36	39°08'17.1"N	105°39'17.6"E
<u>Ordos Plateau</u>			
	MU07-05	38°28'05.7"N	108°48'31.4"E
	MU07-10	39°24'44.9"N	108°57'30.9"E
	HB07-17	40°41'19.2"N	108°32'30.8"E
	HB07-18	40°47'46.5"N	108°15'18.8"E
	UB07-27	40°48'01.7"N	106°33'38.6"E
<u>Jingyuan (JY)</u>			
	JY1	36°21'00.0"N	104°36'00.0"E
	JY7	36°21'00.0"N	104°36'00.0"E
	JY13	36°21'00.0"N	104°36'00.0"E
<u>Qilian Mountains</u>			
	GL07-143	37°27'06.4"N	102°53'19.7"E
<u>Taklimakan</u>			
	TK07-116	38°45'21.9"N	87°30'21.1"E
	TK07-119	39°59'30.3"N	88°21'51.1"E
	TAK06-12	37°25'21.4"N	77°13'29.0"E
	TAK06-17	38°52'08.7"N	76°14'18.2"E
	TAK06-29	39°48'54.3"N	77°27'48.4"E
	TAK06-35	41°21'27.1"N	80°43'42.2"E
	KL10-03	37°16'07.8"N	77°51'41.4"E
	KKR10-02	37°00'57.8"N	79°43'46.3"E
<u>Tian Shan Mountains</u>			
	TS10-09	41°51'33.4"N	82°45'17.6"E
	TS10-11	41°29'18.5"N	84°05'32.1"E
	TS10-12	41°44'23.9"N	85°04'17.4"E
	TS10-13	42°10'04.7"N	86°11'56.7"E
<u>Japan Island</u>			
	MC02	36°12'15.82"N	135°29'16.69"E
	MC04	35°49'50.17"N	134°59'58.05"E
	MC05	36° 3'15.12"N	135°40'20.91"E
	MC07	35°46'3.18"N	135°24'33.08"E
	MC08	35°51'2.23"N	135°28'2.28"E
	KT94-15 PC9	39°34'21.60"N	139°26'24.69"E

Appendix 1:

The PARAFAC modelling steps, matrix calculation and RSS comparison.

Step 1: Arrange the data as EEMs

Worksheet for arrange XRD data to EEM formats for model processing

The 180 samples XRD data was arranged in a three-way structure as the excitation emission matrices (EEMs) for model processing

mod_silt0	200	300	400	500	600	700	800	900	1000	1100	1200	1300	1400	1500
2.02	932	0	0	0	0	0	0	0	0	0	0	0	0	0
2.04	965	0	0	0	0	0	0	0	0	0	0	0	0	0
2.06	889	0	0	0	0	0	0	0	0	0	0	0	0	0
2.08	865	0	0	0	0	0	0	0	0	0	0	0	0	0
2.1	803	0	0	0	0	0	0	0	0	0	0	0	0	0
2.12	722	0	0	0	0	0	0	0	0	0	0	0	0	0
2.14	680	0	0	0	0	0	0	0	0	0	0	0	0	0
2.16	648	0	0	0	0	0	0	0	0	0	0	0	0	0
2.18	655	0	0	0	0	0	0	0	0	0	0	0	0	0
2.2	585	0	0	0	0	0	0	0	0	0	0	0	0	0
2.22	570	0	0	0	0	0	0	0	0	0	0	0	0	0
2.24	548	0	0	0	0	0	0	0	0	0	0	0	0	0
2.26	545	0	0	0	0	0	0	0	0	0	0	0	0	0
2.28	510	0	0	0	0	0	0	0	0	0	0	0	0	0
2.3	499	0	0	0	0	0	0	0	0	0	0	0	0	0
2.32	474	0	0	0	0	0	0	0	0	0	0	0	0	0
2.34	443	0	0	0	0	0	0	0	0	0	0	0	0	0
2.36	502	0	0	0	0	0	0	0	0	0	0	0	0	0
2.38	446	0	0	0	0	0	0	0	0	0	0	0	0	0
2.4	466	0	0	0	0	0	0	0	0	0	0	0	0	0
2.42	420	0	0	0	0	0	0	0	0	0	0	0	0	0
2.44	407	0	0	0	0	0	0	0	0	0	0	0	0	0
2.46	402	0	0	0	0	0	0	0	0	0	0	0	0	0
2.48	411	0	0	0	0	0	0	0	0	0	0	0	0	0
2.5	406	0	0	0	0	0	0	0	0	0	0	0	0	0
2.52	415	0	0	0	0	0	0	0	0	0	0	0	0	0
2.54	409	0	0	0	0	0	0	0	0	0	0	0	0	0
2.56	370	0	0	0	0	0	0	0	0	0	0	0	0	0
2.58	372	0	0	0	0	0	0	0	0	0	0	0	0	0
2.6	365	0	0	0	0	0	0	0	0	0	0	0	0	0
2.62	412	0	0	0	0	0	0	0	0	0	0	0	0	0
2.64	409	0	0	0	0	0	0	0	0	0	0	0	0	0
2.66	379	0	0	0	0	0	0	0	0	0	0	0	0	0
2.68	393	0	0	0	0	0	0	0	0	0	0	0	0	0

Step 2: Input the data and start model developing

Model development by using MATLAB software and drEEM toolbox

MatLab for PARAFAC analysis commands for U1425 sediment unmixing analysis

(List of commands on MatLab)

Silt 6M_IE8_

```
>> filetype=1;ext='txt';range='A1..AU1901';headers=[1 1];display=0;outdata=0;
>> [X, Emmat, Exmat, filelist_U1425_Silt, outdata] = readineems(filetype, ext, range, headers, display,
outdata);
mod_silt001.txt
mod_silt002.txt
mod_silt003.txt
mod_silt004.txt
mod_silt005.txt
mod_silt006.txt
mod_silt007.txt
to
mod_silt180.txt

>> Ex = Exmat(1,:); Em = Emmat(:,1);
>> mydataU1425_Silt=assembledataset(X,Ex,Em,'QSE');
>> mydataU1425_Silt=subdataset(mydataU1425_Silt,[],[],mydataU1425_Silt.Ex>400);
>> TestU1425_Silt=outliertest(mydataU1425_Silt,[],6,'nonnegativity',[1e-8],'atonece');
```

Step 3: Develop preliminary models

PARAFAC Model report

Model development was initiated with a series of 2 to 7 components by using the excitation emission matrices (EEMs) dataset of the 180 samples with the non-negativity constraint and default convergence criterion (0.01) for the outlier tests. **A stricter convergence (10^{-8}) is applied in the 4-7 component.**

PARAFAC Model Report						
Info						
Toolbox	drEEM 0.3.0					
Date	31/07/2020 14:56					
Preprocessing						
nSample - full dataset	180					
nSample - modeled data	180					
No. excluded samples	0					
Excluded samples - indices						
Scatter Removal						
Zapped (Samples,EmRange,ExRange)						
Fluorescence unit	QSE					
Scaling						
PARAFAC model						
No. PARAFAC components	6					
No. Ex wavelengths	3					
OutlierTest_convgrcrit	0.00000001					
OutlierTest_constraints	nonnegativity					
Spectra						
mode	nm	Comp1	Comp2	Comp3	Comp4	Comp5
Ex	200	1	1	1	1	1
Ex	300	0	0	0	0	0
Ex	400	0	0	0	0	0
Em	2.02	0.039486	0.033532	0.070343	0.112118	0.112119
Em	2.04	0.038537	0.030702	0.067144	0.109392	0.109618
Em	2.06	0.034889	0.030136	0.063539	0.104493	0.105116
Em	2.08	0.033239	0.02814	0.060897	0.099175	0.099986
Em	2.1	0.032502	0.024106	0.05594	0.096556	0.099765
Em	2.12	0.030289	0.02574	0.055006	0.090509	0.094418
Em	2.14	0.028573	0.021403	0.051992	0.087068	0.093391
Em	2.16	0.027273	0.021776	0.048182	0.084001	0.087887
Em	2.18	0.02485	0.019969	0.046249	0.082464	0.082635

Step 4: Matrix calculation of actual intensity and score of each end-member

Scaling factors calculation

Fmax is calculated by multiplying the maximum excitation loading and maximum emission loading for each component by its score, producing intensities in the same measurement scale as the original EEMs (Murphy et al., 2013).

To calculate absolute contributions and intensities of spectral loadings for each end-member, (Fmax) and spectral loadings (Em) from model are adjusted by the scaling factors (α 1-6) with the definition, $S(\text{samples})=F(\text{spectral loadings, Em})*W(\text{Fmax})$.

$$\begin{array}{c}
 \text{IODP 142S} \\
 \text{samples} \\
 \begin{array}{cccccc}
 20 & S1 & S2 & S3 & \dots & S180 \\
 2.02 & S_{1(1)} & : & : & : & : \\
 2.04 & : & : & : & : & : \\
 2.06 & : & : & : & : & : \\
 2.08 & : & : & : & : & : \\
 2.10 & : & : & : & : & : \\
 : & : & : & : & : & : \\
 : & : & : & : & : & : \\
 : & : & : & : & : & : \\
 39.96 & : & : & : & : & : \\
 39.98 & : & : & : & : & : \\
 40.00 & S_{1(1901)} & S_{2(1901)} & S_{3(1901)} & \dots & S_{180(1901)}
 \end{array} \\
 = \\
 \begin{array}{cccccc}
 20 & Em1 & Em2 & Em3 & \dots & Em6 \\
 2.02 & f_{1(1)} & f_{2(1)} & f_{3(1)} & \dots & f_{4(1)} \\
 2.04 & : & : & : & : & : \\
 2.06 & : & : & : & : & : \\
 2.08 & : & : & : & : & : \\
 2.10 & : & : & : & : & : \\
 : & : & : & : & : & : \\
 : & : & : & : & : & : \\
 : & : & : & : & : & : \\
 39.96 & : & : & : & : & : \\
 39.98 & : & : & : & : & : \\
 40.00 & f_{1(1901)} & f_{2(1901)} & f_{3(1901)} & \dots & f_{4(1901)}
 \end{array} \\
 \times \\
 \begin{array}{cccccc}
 \text{Scores (Fmax)} \\
 S1 & S2 & S3 & \dots & S180 \\
 W1 & W_{1(1)} & W_{2(1)} & W_{3(1)} & \dots & W_{180(1)} * \alpha 1 \\
 W2 & W_{1(2)} & W_{2(2)} & W_{3(2)} & \dots & W_{180(2)} * \alpha 2 \\
 W3 & W_{1(3)} & W_{2(3)} & W_{3(3)} & \dots & W_{180(3)} * \alpha 3 \\
 : & : & : & : & : & : \\
 W6 & W_{1(4)} & W_{2(4)} & W_{3(4)} & \dots & W_{180(4)} * \alpha 6
 \end{array}
 \end{array}$$

$$\begin{array}{c}
 (1/\max Em1\dots6)*f1(1) / \alpha 1 \dots \alpha 6 \\
 \\
 \boxed{S = F \times W} \\
 \boxed{\frac{f_{1(1)} * \alpha 1}{\alpha 1} w_{1(1)}} \\
 \boxed{= F' * W'}
 \end{array}$$

Scaling factors

Scaling factors (α 1-6) for each component/end-member are calculated by the equation (2).

$$\left(\mathbf{W}^* \mathbf{W} \right)^{-1} * \left(\mathbf{W}^* \mathbf{W} \right) \begin{pmatrix} \alpha_1 \\ \alpha_2 \\ \alpha_3 \\ \alpha_4 \\ \alpha_5 \\ \alpha_6 \end{pmatrix} = \left(\mathbf{W}^* \mathbf{W} \right)^{-1} * \mathbf{W}^* \begin{pmatrix} 1 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ 1 \end{pmatrix} \quad (2)$$

Where $\mathbf{W} = (\mathbf{F}_{\max})$, ${}^t\mathbf{W}$ =transpose of \mathbf{W}

When the scaling factors of each component is calculated, the absolute contribution of each component is estimated by the equation: $({}^t\mathbf{W}) * (\alpha 1-6)$

Absolute intensity of spectral loading (Em) of each component is calculated by this equation: $[(1/\text{MaxEm}) * (\text{Em})]/(\alpha 1-6)$

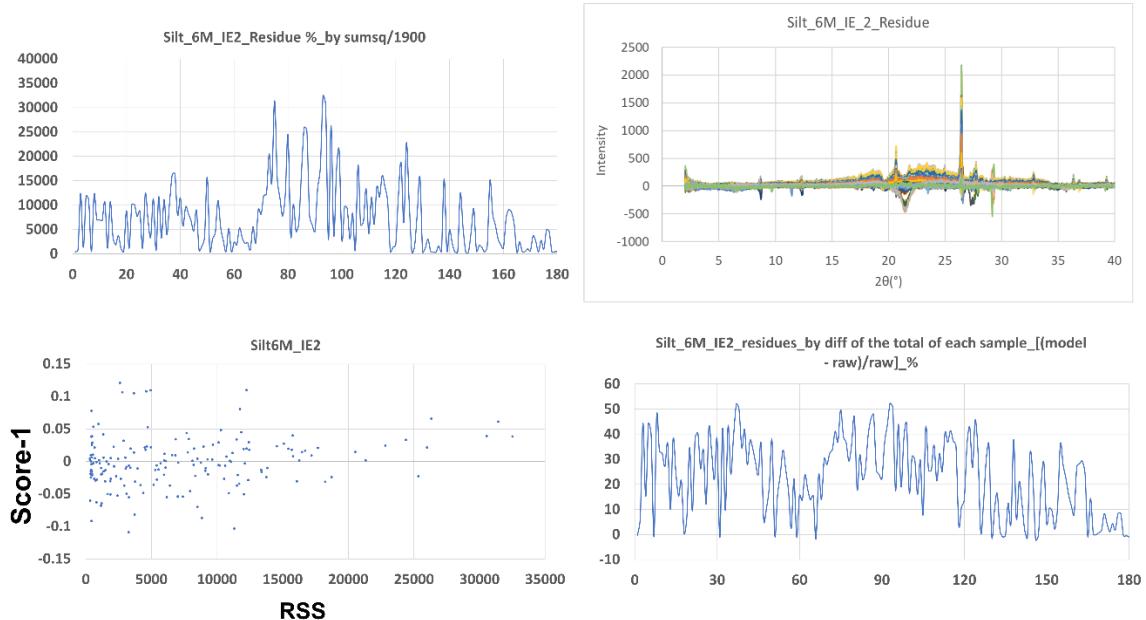
The absolute contribution of components multiplying the absolute intensity of spectral loading of the components to produce the same intensities as the original samples.

The worksheet presents the arrangement for calculating the scaling factors for each component

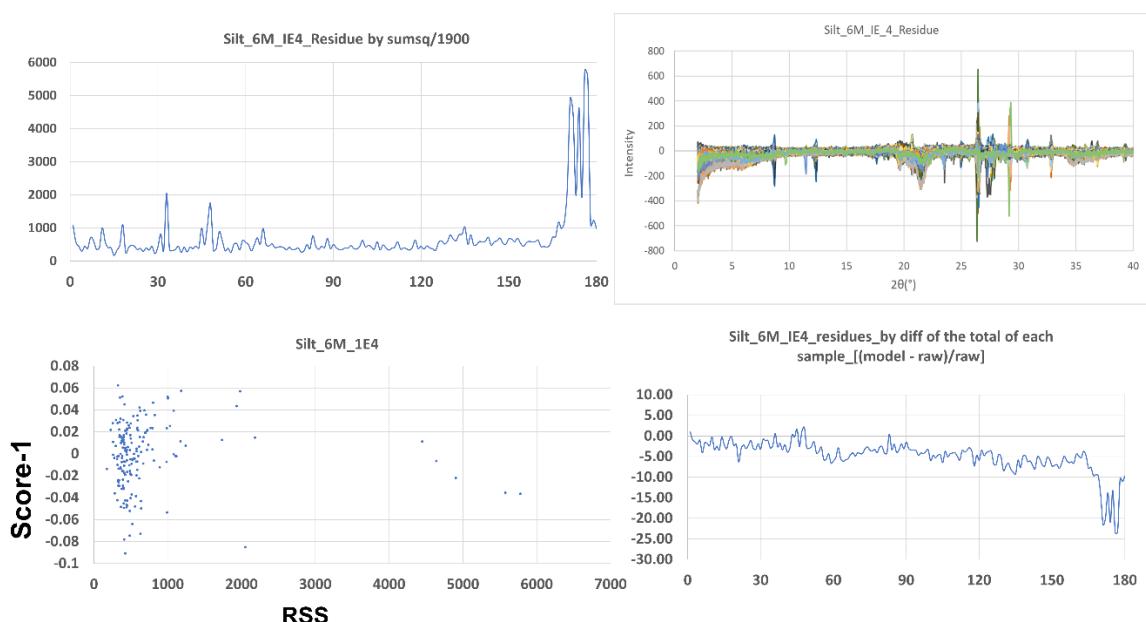
Silt_Scaling factor				\mathbf{W}_{Silt}	1	2	3	4	5	6	7
alfa1	0.0001514			$F_{\max1}$	1205.16	914.74	3472.32	807.83	4404.27	3641.92	1372.02
alfa2	0.000104058			$F_{\max2}$	4110.89	115.18	12.01	1811.65	3625.26	2575.42	4285.43
alfa3	0.0010452			$F_{\max3}$	110.89	10.01	24.12	34.58	10.86	38.31	26.63
alfa4	0.000880016			$F_{\max4}$	307.11	115.18	79.41	69.54	74.68	197.00	391.61
alfa5	0.000586707			$F_{\max5}$	37.83	64.20	81.94	93.66	86.44	132.06	74.32
alfa6	0.00025988			$F_{\max6}$	0.00	1588.77	283.22	2297.69	40.94	78.31	0.00
					$({}^t\mathbf{W})^{-1} \mathbf{W}^* \text{unity}$		$(\mathbf{W}^* \mathbf{W})^{-1} \mathbf{W}$	-1.01E-05	-5.33779E-06	2.1282E-06	-1.98382E-06
								1.98E-05	8.53313E-06	8.78791E-06	7.97179E-07
								1.25E-05	9.16251E-06	3.38691E-06	1.34753E-05
								-2.56E-06	-1.77973E-05	-2.99604E-05	-2.34503E-05
								-3.4E-05	-3.90798E-06	2.04679E-06	1.11985E-05
								-1.25E-05	4.96226E-05	8.68604E-07	7.8685E-05
${}^t\mathbf{W}_{\text{Silt}}$		$F_{\max1}$	$F_{\max2}$	$F_{\max3}$	$F_{\max4}$	$F_{\max5}$	$F_{\max6}$	unity	$\mathbf{W}^* \mathbf{W}$		
1	1205.163185	4740.013	110.849	307.1144	37.83014	0	1		713892419.7	493999299.9	44061971.54
2	914.7307006	3057.115	16.07416	115.1822	64.20485	1588.774186	1		493999299.9	562524462.9	33448185.44
3	341.1109472	1811.652	34.57723	69.53612	81.94117	283.2234076	1		44061971.54	33448185.44	11687409.74
4	807.8299562	1811.652	34.57723	69.53612	93.65735	2297.690551	1		88298679.23	78243119.3	13906427.86
5	4404.271705	3625.263	10.86139	74.67692	86.44186	40.94335614	1		37181810.49	32879581.32	5487058.416
6	3641.923735	2575.422	38.331367	197.0005	132.0584	78.30943861	1		36738401.94	42690784.82	2573509.754
7	1372.021892	4285.435	26.63047	391.6114	74.3196	0	1				
8	5731.484467	2987.354	0	0	104.4797	57.47476393	1				
9	3198.885232	1945.303	13.65458	140.5766	77.09193	947.2573495	1				
10	4868.640817	1321.966	32.92367	211.3096	33.80236	67.30722812	1				
11	2716.59588	3454.016	3.981817	329.6007	94.69674	111.6826366	1				
12	2739.639436	1465.293	159.9813	290.3891	92.7091	221.0411948	1	$(\mathbf{W} {}^t\mathbf{W})^{-1}$			
13	930.6987194	1295.019	50.37027	246.9537	70.66414	1677.133468	1		3.93879E-09	-2.90576E-09	-3.5755E-09
14	4679.050968	1993.589	55.47653	193.0523	41.27851	68.60210857	1		-2.90576E-09	5.21191E-09	4.93417E-09
15	1717.217094	1577.37	55.10948	104.6025	76.52925	1654.538078	1		-3.5755E-09	4.93417E-09	2.05997E-07
16	1418.382616	3000.686	35.12403	216.6351	90.92135	827.1703226	1		-1.87086E-09	-5.99946E-09	-9.2905E-08
17	1765.448891	3602.886	96.04193	239.7059	38.3382	52.69265648	1		-1.6625E-09	-2.55563E-09	-2.42776E-08
18	751.1669294	0	54.27734	114.7999	41.86524	2803.259113	1		2.21075E-10	-2.52937E-09	3.93259E-09
19	526.3910304	1138.635	41.3979	162.6342	83.11669	2360.921185	1				
20	3213.209343	1401.369	134.8958	241.2489	53.46601	82.16525978	1				
21	1650.909949	1474.59	68.89866	470.54	149.4413	117.688936	1				

The followings show the example of RSS result by applying a stricter convergence criterion of a minimum tolerance from 1×10^{-2} to 1×10^{-8} with nonnegativity constrained for the 6 endmembers model.

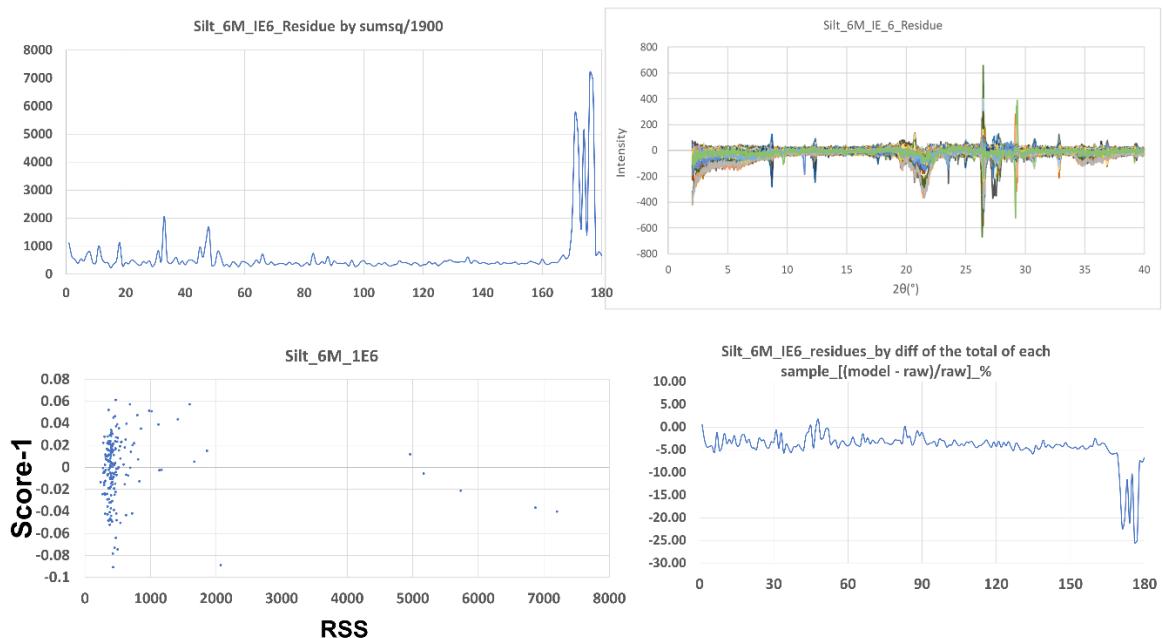
Silt_6M_IE2_Residue



Silt_6M_IE4_Residue



Silt_6M_IE6_Residue



Silt_6M_IE8_Residue

