1 **Uranium comminution age tested by the eolian deposits on the** 2 **Chinese Loess Plateau**

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10 **Abstract**

11 The ²³⁴U/²³⁸U ratio of fine particles can record the time since their separation
12 from bed rock because of the disruption of uranium series equilibrium
13 introduced by the recoil of daughter ²³⁴Th nuclei (precursor of ²³⁴U) out of
14 particle surfaces during the decay of ²³⁸U. Application of the uranium
15 comminution age method, which has great potential in tracing production and
16 transportation of sediments is however complicated by the weathering
17 dissolution of ²³⁴U depleted particle surfaces, the difficulty in determining the
18 fraction of recoiled nuclei, and the precipitation of exogenetic ²³⁴U. Here we
19 minimize these complications by using a newly developed precise size

separation using electroformed sieve, and a chemical protocol that involves reductive and oxidative leaching. Eolian deposits collected from the Chinese Loess Plateau (CLP) were used to test the validity of our method. Possible effects of weathering dissolution were also evaluated by comparing samples with different weathering intensities. The results show decreasing $^{234}\text{U}/^{238}\text{U}$ ratios in fine eolian particles with increasing sedimentation age, agreeing well with the theoretical prediction of the comminution age model. This successful application of the uranium comminution age approach to the eolian deposits on the CLP is also aided by a stable dust source, the low weathering intensity, the lack of consolidation, and the well-defined age model of the deposits. A transportation time of 242 ± 18 ka was calculated for the eolian deposits, which indicates a long residence time, and thus extensive mixing, of the dust particles in source regions, partly explaining the stable and homogeneous composition of the eolian dust over glacial-interglacial cycles.

Keywords: surface process; erosion; dust storm; paleoclimate; uranium isotope; dating

1. Introduction

The uranium comminution age of fine particles is the elapsed time since their separation from bedrocks (DePaolo et al., 2006). The principle is based on the recoiling effect during the α decay of ^{238}U . In large rock pieces older than

several half-lives of ^{234}U (245.6 ka, Cheng et al., 2013), the ^{234}U produced by the decay of ^{238}U is balanced by its decay to ^{230}Th :

$$\lambda_{238}^{238}\text{U} = \lambda_{234}^{234}\text{U} \quad (1)$$

where λ_{238} and λ_{234} are the decay constants of ^{238}U and ^{234}U , respectively. Thus, the activity ratio between ^{234}U and ^{238}U , i.e., $(\lambda_{234}^{234}\text{U})/(\lambda_{238}^{238}\text{U})$, equals 1. Such a secular equilibrium would be disturbed once small particles ($\leq 50 \mu\text{m}$) are generated because a significant portion of ^{234}Th , the precursor of ^{234}Pa and then ^{234}U , within the $\sim 30 \text{ nm}$ surface depth, will be ejected out of the particle due to the recoil effect during α decay of ^{238}U (Kigoshi, 1971). Consequently, the loss of ^{234}U in the particle exceeds its production. The activity ratio between ^{234}U and ^{238}U , which is normally expressed as $(^{234}\text{U}/^{238}\text{U})$, will thus decrease until a new equilibrium is established so that the decreased decay of ^{234}U associated with the decreasing amount of ^{234}U can compensate for the ejected ^{234}Th (DePaolo et al., 2006):

$$(^{234}\text{U}/^{238}\text{U}) = 1 - f \times (1 - e^{-\lambda_{234}t_c}) \quad (2)$$

where t_c is the comminution age, and f is the recoil fraction of the daughter ^{234}Th that is ejected out of the particle surface.

The uranium comminution age method can be tested using the relationship between $(^{234}\text{U}/^{238}\text{U})$ and t_c in Eq. (2). However, independent comminution age

determination is generally impossible. Alternatively, a test can be made by examining the correlation between ($^{234}\text{U}/^{238}\text{U}$) and deposition age in sedimentary sequences (DePaolo et al., 2012). Splitting the comminution age (t_c) into transportation time (t_T) and deposition age (t_D), Eq. (2) can be transformed to:

$$(^{234}\text{U}/^{238}\text{U}) = 1 - f \times [1 - e^{-\lambda_{234}(t_T+t_D)}] \quad (3)$$

Eq. (3) predicts a decreasing ($^{234}\text{U}/^{238}\text{U}$) with increasing deposition age if t_T and f can be assumed to be constant.

The requirement of constant t_T , however, is not satisfied in many situations. Transportation time shows great variation in response to glacial-interglacial cycles, and thus rather scattered patterns of decreasing ($^{234}\text{U}/^{238}\text{U}$) with increasing deposition age have been observed (e.g., DePaolo et al., 2006; Dosseto et al., 2010). A case study on the fluvial sediment in the King's River Fan, which is assumed to have a constant zero transportation age, does show a monotonously decreasing ($^{234}\text{U}/^{238}\text{U}$) with sediment depth for particles finer than 6 μm (Lee et al., 2010). However, no reliable independent deposition age is available for this fluvial sequence (Lee et al., 2010). The 10–15 μm and 15–20 μm size fractions of the sediments even show several reversed trends in ($^{234}\text{U}/^{238}\text{U}$) with depth (Lee et al., 2010).

On the other hand, precise determination of f in Eq. (3) is also demanding (Bourdon et al., 2009; Maher et al., 2006). The recoil fraction depends on the size, geometry and surface roughness of the grain. While the size and geometry of grains can be quantified using traditional methods, such as laser based particle size analysis (Dosseto et al., 2010; Li et al., 2016), roughness shows considerable variation between samples (DePaolo et al., 2012; Lee et al., 2010). Measurement of surface area using BET gas adsorption has great potential for quantifying the recoil fraction (Aciego et al., 2011; Bourdon et al., 2009). However, this method requires fractal correction because recoil distance ($\sim 35 \times 10^{-9}$ m) is ~ 100 times the diameter of adsorbate molecule (e.g. 3.5×10^{-10} m for N_2). A practical technique to minimize the effect of changing recoil fraction in a sediment sequence is to sieve the sediment into limited size fractions (Lee et al., 2010). Assuming no change in geometry and roughness, the recoil fraction of the sieved grains can be treated as constant and its value can be estimated from old samples where equilibrium is established, i.e., $f = 1 - (^{234}\text{U}/^{238}\text{U})$ (Lee et al., 2010).

Other complexities associated with f include consolidation of sediment and influence of dissolution on specific area. In consolidated sediments, the ejected ^{234}Th nuclei may penetrate into nearby grains so that the apparent recoil fraction is reduced. Dissolution can both increase and decrease the specific surface area and thus the f value, depending on saturation state and reaction

99 mechanism (Bandstra and Brantley, 2008).

100 Testing the uranium comminution age in sedimentary sequences is also
101 complicated by the influence of dissolution and authigenic precipitation on the
102 ($^{234}\text{U}/^{238}\text{U}$) of particles (DePaolo et al., 2012; Dosseto and Schaller, 2016).
103 Using the low dissolution rate obtained from a 70 m deep vadose zone (Maher
104 et al., 2006), DePaolo et al. (2012) suggested that dissolution of the ^{234}U
105 depleted surface layer has negligible influence on the ($^{234}\text{U}/^{238}\text{U}$) of sediment.
106 However, dissolution rates of minerals vary significantly under different settings
107 (White and Brantley, 2003). Chemical weathering may also decrease the
108 $^{234}\text{U}/^{238}\text{U}$ of sediment by preferential leaching of ^{234}U that is mostly held in the
109 radioactively damaged crystal sites (Andersen et al., 2009; Chabaux et al.,
110 2003; Fleischer, 1980), the effect of which on uranium comminution age still
111 needs investigation (DePaolo et al., 2006; Dosseto and Schaller, 2016;
112 Handley et al., 2013).

113 Chemically leaching is generally applied to clean the authigenic phases
114 that contain exogenetic uranium (DePaolo et al., 2006). The best leaching
115 practice should obtain the lowest ($^{234}\text{U}/^{238}\text{U}$) values by removing all of the
116 authigenic phases that are likely to have ($^{234}\text{U}/^{238}\text{U}$) values > 1 , without
117 destroying the ^{234}U -depleted surface layer of the detrital minerals (Martin et al.,
118 2015). However, no general procedure has yet been established due to

119 variable natural sample matrixes.

120 In this study, we test the uranium comminution age method using the eolian
121 deposits on the Chinese Loess Plateau (CLP). The eolian sequences on the
122 CLP have a reliable age model that has been constrained by paleo-magnetic
123 reversals and orbital tuning (e.g., Sun et al., 2006). The low weathering
124 intensity of loess (Chen et al., 1998) may reduce the possible influences of
125 chemical weathering on ($^{234}\text{U}/^{238}\text{U}$). Nevertheless, effects of weathering can be
126 investigated by comparing samples with identified differences in weathering
127 intensity (Chen et al., 1999; Chen et al., 2001). The very old eolian deposits,
128 dating to the late Oligocene (Guo et al., 2002; Qiang et al., 2011) would help to
129 assess the effect of consolidation. For the purpose of this study, a chemical
130 procedure was designed to clean the U absorbed to the authigenic phases and
131 a precise size separation using electroformed sieve was applied to reduce
132 changes in recoil fraction. With all complexities minimized, we found that
133 ($^{234}\text{U}/^{238}\text{U}$) of the eolian deposits on the CLP decreases with increasing
134 deposition age as predicted by the uranium comminution age model.

135 **2. Samples and methods**

136 *2.1. Samples*

137 Samples of eolian deposits covering the past ~3 Ma were collected from

the Xifeng (107°47'E, 35°45'N) and the Lingtai (107°31'E, 35°01'N) sections on the CLP (Fig. 1). The stratigraphy of the two sections was determined by correlating the newly measured magnetic susceptibility data (Supplementary Fig. 1) to those previously published at a nearby ZJC site (Sun et al., 2006). The age model of the ZJC section has been established by tuning the grain size proxy, which mainly reflects the winter monsoon intensity, to orbital parameters with constraints from paleo-magnetic reversals (Sun et al., 2006). The age model has a sub-orbital resolution (~10 ka), which is accurate enough to assess the uranium comminution age.

Both glacial samples (loess layers) with low magnetic susceptibility and interglacial samples (paleosol layers) with high magnetic susceptibility were collected in the two sections (Supplementary Fig. 1). The resolution of the samples (Table S1) is designed to decrease with increasing age considering the decreasing rate of change in ($^{234}\text{U}/^{238}\text{U}$) through time (DePaolo et al., 2006).

Three old loess samples were collected from the Qin'an section (105°27'E, 35°02'N) to the west of Xifeng and Lingtai sections (Fig. 1). Deposition ages of the three samples were approximately determined to be 8.3 Ma, 9.4 Ma, and 21.5 Ma according to the previously published stratigraphy (Guo et al., 2002).

2.2. Sieving and chemical leaching

Grains of different sizes were separated by sieving the dispersed samples after removal of carbonate. Electroformed sieves were used instead of traditional nylon net sieves because the shape of the nylon net is readily deformed during sieving. More importantly, the accuracy of electroformed sieves can be as good as 1 μm (Irani and Callis, 1959). A diameter range between 20 μm and 25 μm was chosen because this is the main size range of the eolian deposits on the CLP (Sun et al., 2004). Two old loess samples (2.1 Ma and 21.5 Ma) were separated into five size ranges (2–10 μm , 10–15 μm , 15–20 μm , 20–25 μm , 25–64 μm) to investigate the dependence of f on grain size (Table 1). The precise size separation achieved by electroformed sieves is evident from microscopic observations (Supplementary Fig. 2).

A set of experiments has been conducted to establish the chemical procedure to clean the authigenic phases. The optimized procedure involves three steps. The first step is to remove carbonate using diluted acetic acid (0.5 mol/L) at room temperature for 2 hours (Chen et al., 2007). Similar ($^{234}\text{U}/^{238}\text{U}$) values of the residues have been observed for the experiments using 0.5 mol/L ($\text{pH} = 2.53$) and 0.1 mol/L ($\text{pH} = 2.88$) acetic acid solution, and the HAc–NaAc buffering solution ($\text{pH} = 4$) (Fig. 2), which indicates limited dissolution of the ^{234}U -depleted surface layer. Any ultrasonic shaking should be avoided because it generates higher ($^{234}\text{U}/^{238}\text{U}$) values for the residues (Fig. 2), possibly due to the breaking of the surface layer.

The second step is to remove Fe-Mn oxides through reductive leaching using the CBD method slightly modified from Mehra and Jackson (1958). The sieved samples after the removal of carbonate were bathed in a dithionite-citrate solution buffered with sodium bicarbonate at 85°C. Repeated CBD treatments and reductive cleaning using hydrazine (Boyle, 1981) resulted in higher ($^{234}\text{U}/^{238}\text{U}$) values of the residues possibly due to dissolution of surface layers (Fig. 2). Thus, only one CBD treatment is recommended.

The final step is to remove organic matter by bathing the sample in 5% hydrogen peroxide at 85°C for 1 hour. After removal of organic matter the samples show lower ($^{234}\text{U}/^{238}\text{U}$) values compared to the untreated ones (Fig. 2).

One loess sample (>1 Ma) is repeatedly processed in every batch to check the uncertainty of the whole procedure, which results in a ($^{234}\text{U}/^{238}\text{U}$) value of 0.912 ± 0.004 (mean $\pm 2\sigma$, the same hereafter; n=8; Fig. 3a).

3.3. BET measurement

BET surface area was measured for both the sieved and cleaned samples (with all authigenic phases removed) and the sieved but uncleaned samples (with carbonate minerals removed but not the Fe-Mn oxides and organic matter). Nitrogen adsorption-desorption at 77 K was applied to the samples,

with surface area evaluated by the single point BET method. The fractal dimension of the grain surface (D) was determined by the slope of the linear correlation between θ and the logarithm of P_0/P , where θ is the relative adsorption and P_0/P is the ratio between saturated absorption pressure and absolute pressure, respectively (Avnir and Jaroniec, 1989; Bourdon et al., 2009).

3.4. Isotope measurements

About 100 mg of cleaned sample was dissolved in 4 ml 1:1 mixture of HNO_3 and HF solution in tightly sealed teflon vials at 120°C for more than 48 hours. Uranium in the digested solution was separated using UTEVA resin following the method of Wang and You (2013). The $^{234}\text{U}/^{238}\text{U}$ ratios were measured using a newly launched MC-ICP-MS (Neptune plus, Thermo-Fisher Scientific) at the MOE Key Laboratory of Surficial Geochemistry, Department of Earth Sciences, Nanjing University. Signals for ^{235}U and ^{238}U were monitored using Faraday cups and that of ^{234}U was monitored using a secondary electron multiplier (SEM). The 2σ standard deviation of the ($^{234}\text{U}/^{238}\text{U}$) ratio is 0.001. Instrumental bias between ^{234}U and ^{238}U was corrected by normalizing the $^{238}\text{U}/^{235}\text{U}$ ratio to 137.84 (Andersen et al., 2017). Standard sample bracketing (SSB) is also applied to correct short-term variation of the yield factor between Faraday cups and SEM (Wang and You, 2013). An in-house U standard (NJU),

which gives a $^{238}\text{U}/^{235}\text{U}$ normalized ($^{234}\text{U}/^{238}\text{U}$) value of 0.950 ± 0.002 ($n = 42$; Fig. 3b) that is consistent with previous calibration to IRMM3184, is used as a standard for the bracketing. The ($^{234}\text{U}/^{238}\text{U}$) value was calculated using decay constants of $2.82206 \times 10^{-6} \text{ a}^{-1}$ and $1.55125 \times 10^{-10} \text{ a}^{-1}$ for ^{234}U and ^{238}U , respectively (Cheng et al., 2013). Repeated measurements of the USGS BCR-2 standard material give a mean SSB value of 1.001 ± 0.002 ($n = 21$; Fig. 3c).

3.5 Statistics

The correlation between ($^{234}\text{U}/^{238}\text{U}$) and deposition age in form of Eq. (3) is regressed using Monte Carlo simulations to account for the uncertainties associated with ($^{234}\text{U}/^{238}\text{U}$). In each Monte Carlo simulation, a hypothetical dataset was generated from the observed ($^{234}\text{U}/^{238}\text{U}$) by adding random errors following a normal distribution defined by the observed external standard deviation of 0.002. The hypothetical dataset was then regressed to Eq. (3) using a least squares method. Regression coefficients (t_{T} and f), r^2 , and p values (p_{MC}) were recorded in every simulation. The simulation was repeated 100,000 times. The mean and standard deviation of the regression coefficients and r^2 from the 100,000 simulations were calculated as the final regression results. The significance of the final regression results was judged by the fraction (p) of insignificant correlations ($p_{\text{MC}} \geq 0.05$) among the 100000

simulations. A p value < 0.05 , i.e., when more than 95% of the simulated correlations were significant, means that the correlation between the observed x and y values is significant at the 95% confidence level considering the uncertainty in the data. The resultant values of t_r and f and their standard deviations from Monte Carlo simulations are the same as those derived from traditional regression using the least squares method.

3. Results

The BET surface area of the sieved and cleaned samples shows considerable variation from 1.3 to 4.9 m²/g, with a mean value of 2.3 ± 0.6 m²/g ($n = 12$, Table 2). In contrast, the BET surface area of the sieved but uncleaned samples (with carbonate minerals removed but not the Fe-Mn oxides and organic matter) shows smaller variability, with a mean value (21.7 ± 1.8 m²/g, $n = 5$, Table 2) of about 10 times the cleaned samples.

Decreasing ($^{234}\text{U}/^{238}\text{U}$) values with increasing depositional age has been observed for the 20–25 μm size fraction of the eolian deposits on the CLP (Fig. 4), which is consistent with the prediction of Eq. (3) ($p < 10^{-4}$, $r^2 = 0.89$). Fitting this relationship using Eq. (3) based on Monte Carlo simulations results in a transportation time of 242 ± 18 ka and an f value of 0.088 ± 0.002 .

The root mean square deviation (RMSE) of the ($^{234}\text{U}/^{238}\text{U}$) data from the

regressed correlation is 0.0045, which is about twice the standard deviation of the replicated sample (0.002). Thus, some unknown factors besides measuring uncertainty may contribute to the scatter of the correlation. Nevertheless, no clear differences can be found between interglacial and glacial samples, nor between the samples from Xifeng and Lingtai sections, along the fitting line within the $\pm 2\sigma$ analytical error (Fig. 4). The correlation between ($^{234}\text{U}/^{238}\text{U}$) and the deposition age is less scattered for young samples (< 200 ka, RMSE = 0.0037) compared to the older ones (> 400 ka, RMSE = 0.0045).

4. Discussion

4.1. Uncertainties related to pretreatment

The error in the measured ($^{234}\text{U}/^{238}\text{U}$) introduced by sample preparation might be related to the variation in grain size associated with sieving, or to chemical leaching of the surface layer. The limited variation in BET specific surface area of the sieved and uncleaned samples confirms the accuracy of size separation (Table 2). Leaching dissolution of the ^{234}U depleted surface layer would introduce smaller uncertainties for the samples with younger ages, because the depletion of ^{234}U is smaller in young surface layers (DePaolo et al., 2006). This might be supported by the less scattered correlation between ($^{234}\text{U}/^{238}\text{U}$) and deposition age for young samples compared to the older ones (Fig. 4). Chemical leaching may also cause preferential loss of ^{234}U relative to

^{238}U (Andersen et al., 2009; Fleischer, 1980), which might contribute to the decreasing ($^{234}\text{U}/^{238}\text{U}$) value after leaching. Such an influence of leaching can be traced by comparing the BET surface area-based and uranium isotopic equilibrium-based f values. The BET surface area-based f value (0.070 ± 0.004 , Table 2) is lower than that (0.088 ± 0.002) estimated by the ($^{234}\text{U}/^{238}\text{U}$) value (section 4.2). However, higher BET-based f values would be expected if there is preferential leaching of ^{234}U . Nevertheless, the small variation in ($^{234}\text{U}/^{238}\text{U}$) for the replicate pre-treated loess sample (± 0.004 ; Fig. 3a) indicates that the pretreatment involving size separation and chemical cleaning can give reproducible results.

4.2. Recoil fraction and roughness

The good correlation between ($^{234}\text{U}/^{238}\text{U}$) and deposition age as predicted by Eq. (3) requires a constant recoil fraction and transportation time. Otherwise, the effects of changing transport time and recoil fraction on ($^{234}\text{U}/^{238}\text{U}$) would have to exactly cancel out each other. To illustrate this, we calculate f value and transportation time of every sample assuming a constant transportation time of 242 ka and a constant f value of 0.088, respectively. The calculated f and t_{τ} show no variation, considering the propagated uncertainties (Fig. 5).

The constant f of the sieved size fraction is also confirmed by the rather constant BET specific surface area of the samples with carbonate minerals

removed, but not the Fe-Mn oxides and organic matter (Table 2). Thus, size separation using electroformed sieves can practically reduce the changes in f for the eolian deposits on the CLP. According to Eq. (2), the ($^{234}\text{U}/^{238}\text{U}$) value of samples older than several half-lives of ^{234}U equates $1 - f$. Thus, the value of f inferred from the replicated old loess sample is 0.088 ± 0.004 , which equals to the value (0.088 ± 0.002) derived from the correlation between ($^{234}\text{U}/^{238}\text{U}$) and deposition age (Fig. 4).

The determined value of f based on uranium isotopes can be compared to that based on BET surface area after correction of fractal dimension (Bourdon et al., 2009):

$$f = \frac{1}{4} \times \frac{2^{D-1}}{4-D} \times \left(\frac{r}{L}\right)^{D-2} \times L \times S \times \rho$$

(4)

where S is BET mass specific surface area, D is fractal dimension, r is the molecule diameter of N_2 (0.35 nm), L is recoil distance, and ρ is mineral density, about 2.7 g/cm^3 . The calculated f values using a recoil distance of $35 \times 10^{-9} \text{ m}$ (DePaolo et al., 2006; Maher et al., 2006) are about 0.011 ± 0.003 ($n = 12$) and 0.070 ± 0.004 ($n = 5$) for cleaned and uncleaned samples, respectively. The f values of cleaned samples based on BET surface area are nearly one order of magnitude lower than that determined from uranium isotopes. Instead, the f values of uncleaned sample based on BET surface area are close to that

determined from uranium isotopes. The lower and variable BET surface area based f values of cleaned samples may be related to the decreasing roughness of mineral surfaces during chemical leaching. Thus, BET surface measurements based on uncleaned samples are recommended in determining the value of f .

The slightly lower BET surface area based f values of the uncleaned samples compared to that derived from uranium isotopes could be related to the uncertainties in recoil distance (L) and/or difference in volume specific surface area ($S \times \rho$) between the main U holding heavy minerals and the bulk. Using a larger L value of 55 nm, as originally suggested by Turkowsky (1968), will result in f values (0.084 ± 0.005) much closer to that derived from uranium isotopes. Alternatively, planar grains of heavy minerals (such as apatite and zircon) may be preferentially transported compared to the bulk light minerals, and thus results in higher specific surface area for the main U-holding minerals.

Comparing the recoil fraction with grain size can infer roughness of the particles. Roughness accounts for both grain shape and surface structure relative to a spherical grain. The data observed from different grain size fractions of the eolian deposits on the CLP (Fig. 6) are consistent with previous suggestions that roughness tends to be higher for larger grains (DePaolo et al., 2012; Lee et al., 2010).

4.3. Influence of weathering

The influence of weathering is investigated by comparing samples with different weathering intensities. Paleosol layers deposited during interglacial stages, which are characterized by high magnetic susceptibility (An et al., 1991) and higher depletion of labile elements (Chen et al., 1999), have experienced much stronger post-depositional pedogenesis than the glacial loess due to the combination of a stronger summer monsoon, lower deposition rates, and higher temperatures (Chen et al., 1999; Chen et al., 2001). Weathering intensity is also different between the samples from Lingtai section and Xifeng section. The Lingtai site is located to the south of the Xifeng site, and thus receives higher precipitation (583 mm/yr vs. 528 mm/yr) due to the northward decreasing influence of the summer monsoon. The deposition rate at the Lingtai site is lower than that at the Xifeng site due to the southward transportation of eolian dust. Consequently, the eolian deposits at the Lingtai site experience much stronger post-depositional pedogenesis, which is evidenced by higher magnetic susceptibility and higher degree of chemical leaching compared to that at the Xifeng site (Sun et al., 2006). The consistency between glacial and interglacial samples, and between the Xifeng and Lingtai sections along the predicted ($^{234}\text{U}/^{238}\text{U}$)-deposition age correlation, implies that the influence of weathering on the ($^{234}\text{U}/^{238}\text{U}$) is very limited. The limited weathering influence may be related to the weak weathering of the eolian deposits on the CLP (Chen

et al., 1998).

4.4. Influence of consolidation

The stable low ($^{234}\text{U}/^{238}\text{U}$) value of the very old loess samples (> 8 Ma), consistent with the prediction of Eq. (3) (Fig. 4), indicates that potential influence of consolidation, and thus the tendency for retuning the ($^{234}\text{U}/^{238}\text{U}$) back to one in the aged samples, is negligible. This is probably because the degree of consolidation is very low for the eolian deposits. In addition, the inter-granule spaces of the eolian deposits are mostly cemented by secondary carbonate (Li et al., 2013), which may prevent consolidation of detrital grains.

4.5. Transportation time

The transportation time of 242 ± 18 ka derived from the uranium comminution age indicates a very long residence time for the eolian dust in the source regions. Unlike the saltatorial transport of sand and gravel, fine eolian silt is mainly transported in the upper troposphere (Pye, 1995). Consequently, the time of eolian transportation is assumed to be from hours to days, as for modern dust storms (Uno et al., 2009). Thus, the transportation time derived from the uranium comminution age mainly reflects the residence time in source regions.

The long residence time in the source regions could be related to several

factors. Although the arid inland basins are generally regarded as source regions for the eolian deposits, due to the extensive eolian activities (Wang et al., 2004), it is argued that the silt dust particles are mainly generated from the surrounding high mountains (Sun, 2002). Thus, the long residence time may be related to storage of sediments in soils on the eroding mountain, and/or in river channels that transport the material to the arid lands, and/or in alluvial fans in the arid basins. Qilian Mountain has been identified to be the main debris source that feeds the most likely eolian source regions of the Alxa arid lands (Fig. 1) (Che and Li, 2013; Chen and Li, 2011; Hu and Yang, 2016; Li et al., 2009). The steep relief of Qilian Mountain (Pan et al., 2010) might not favor a long term storage in river channels. Instead, large volumes of Quaternary sediments has been found in the alluvial fans and in the arid basin (Li et al., 2014; Wang et al., 2015). Detailed research of the pathway from the source mountains to the arid lands may help to identify the places of silt storage.

The long residence time implies extensive fluvial and/or eolian mixing of the silt particles in the source regions (Gu et al., 1997; Gu et al., 1996), which may help to explain the arguably homogeneous composition of loess deposits. A residence time longer than one glacial-interglacial cycle also suggests that the potential influence of orbital-scale climate changes on the duration of silt storage could be largely buffered, and is thus barely reflected in the eolian deposits on the CLP (Chen and Li, 2011; Li et al., 2009).

5. Conclusion

The eolian deposits on the CLP show decreasing ($^{234}\text{U}/^{238}\text{U}$) value with increasing deposition age, as predicted by the uranium comminution age model. The successful application of the uranium comminution age method to the eolian deposits on the CLP benefits from several factors: (1) the specific surface area, and thus recoil fraction, of carefully separated size fractions show very limited variation in the loess-paleosol sequences, (2) the ($^{234}\text{U}/^{238}\text{U}$) values of the eolian deposits have not been influenced by post-depositional dissolution due to low weathering intensity, (3) carbonate cement may have largely prevented consolidation of the detrital minerals, and most importantly (4) the residence time of eolian dust in source regions is relatively constant due to the large buffering capacity.

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565 **Table**

566 Table 1. The f values constrained by uranium isotope analyses of different grain
567 size fractions of the eolian deposits.

Grain sizes	2.1 Ma		21.5 Ma	
	$(^{234}\text{U}/^{238}\text{U})$	f^a	$(^{234}\text{U}/^{238}\text{U})$	f^a
2–10 μm	0.900	0.100	0.897	0.103
10–15 μm	0.910	0.090	0.908	0.092
15–20 μm	0.911	0.089	0.914	0.086
20–25 μm	0.909	0.091	0.908	0.092
25–64 μm	0.919	0.081	0.956	0.044

568 ^a $f = 1 - (^{234}\text{U}/^{238}\text{U})$.

569 Table 2. BET surface area and the corresponding f value for the 20–25 μm size
570 fraction of the eolian deposits on the Chinese Loess Plateau.

Deposition Age (ka)	Chemically cleaned			Chemically uncleaned			
	S (m^2g^{-1})	D	f^a	S (m^2g^{-1})	D	f^a	f^b
10	1.8	2.629	0.005				
124	2.1	2.328	0.016				
169	1.7	2.571	0.006				
242	2.4	2.308	0.020	22.4	2.609	0.070	0.084
252	2.1	2.460	0.010				
267	2.3	2.587	0.008	20.5	2.603	0.066	0.079
343	1.6	2.274	0.015	19.9	2.586	0.067	0.081
517	1.3	2.473	0.006				
800	2.3	2.615	0.007	21.3	2.599	0.069	0.083
1905	2.8	2.551	0.011				
2085	4.9	2.587	0.016	24.4	2.606	0.077	0.092
2320	2.2	2.612	0.007				
Mean	2.3	2.500	0.011	21.7	2.601	0.070	0.084
2 σ	0.6	0.078	0.003	1.8	0.009	0.004	0.005

571 ^aBased on $L = 35 \text{ nm}$ and $\rho = 2.7 \text{ g/cm}^3$.

572 ^bBased on $L = 55 \text{ nm}$ and $\rho = 2.7 \text{ g/cm}^3$.

Figure captions

Fig. 1. Map showing geographic setting and locations of the sampling sites. Arrow illustrates the transport passway for eolian dust by the prevailing surface wind.

Fig. 2. Influence of chemical cleaning on the ($^{234}\text{U}/^{238}\text{U}$) ratio of loess. Color of the bars illustrates method of acid leaching. The times of CBD reductive leaching is indicated by the number on the top of the bars excepting the one with hydrazine reductive cleaning. Removal of organic matter is applied to all samples except for those labeled with red triangles.

Fig. 3. Analytical uncertainties. (a) Long-term reproducibility of the whole procedure based on one loess sample of >1 Ma (SSB method). (b) Long-term analytical results of the $^{238}\text{U}/^{235}\text{U}$ normalized ($^{234}\text{U}/^{238}\text{U}$) of the in-house NJU standard. (c) Long-term analytical reproducibility of ($^{234}\text{U}/^{238}\text{U}$) based on BCR-2. The results of the SSB method and $^{238}\text{U}/^{235}\text{U}$ normalization are shown for comparison.

Fig. 4. The ($^{234}\text{U}/^{238}\text{U}$) of the chemically cleaned 20–25 μm size fraction of the eolian deposits on the Chinese Loess Plateau, plotted against deposition age. The solid red line is the best fit to Eq. (3) with red dashed lines showing the 95% confidence level.

Fig. 5. Calculated f value (a) and transportation time (b) assuming constant transportation time of 242 ka and f value of 0.088, respectively. The high-resolution portion of a glacial-interglacial cycle in (a) is illustrated in (c). The error bar propagates from an external error of ± 0.004 for ($^{234}\text{U}/^{238}\text{U}$). Only samples over the past 400 thousand years are shown in (b) because calculation for old samples results in large propagated errors. The red lines with 95% confidence level (dash lines) are from regression of the whole time series in Fig. 4. Also compared is the LR04 stack of benthic oxygen isotope (Lisiecki and Raymo, 2005) showing glacial-interglacial cycles. The glacial-interglacial cycle is also monitored by grain size and magnetic susceptibility in (c).

Fig. 6. Dependence of recoil fraction (f) on grain-size. Data source: CLP (this study); Hanford (Maher et al., 2006); Site 984 (DePaolo et al., 2006); Dome C dust (Aciego et al., 2011); King's River Fan (KRF) (Lee et al., 2010); Okinawa Trough (OT) (Li et al., 2016).

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