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

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ABSTRACT

The ^{234}U/^{238}U ratio of fine particles can record the time since their separation from bed rock because of the disruption of uranium series equilibrium introduced by the recoil of daughter ^{234}Th nuclei (precursor of ^{234}U) out of particle surfaces during the decay of ^{238}U. Application of the uranium comminution age method, which has great potential in tracing production and transportation of sediments is however complicated by the weathering dissolution of ^{234}U depleted particle surfaces, the difficulty in determining the fraction of recoiled nuclei, and the precipitation of exogenetic ^{234}U. Here we 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}U/^{238}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.

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:

$$\lambda_{238}{}^{238}\text{U} = \lambda_{234}{}^{234}\text{U} \tag{1}$$

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 μm) are generated because a significant portion of ^{234}Th, the precursor of ^{234}Pa and then ^{234}U, within the ∼30 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}U/^{238}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):

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

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}U/^{238}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}U/^{238}U) and deposition age in sedimentary sequences (DePaolo et al., 2012). Splitting the comminution age (t_c) into

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transportation time (t_T) and deposition age (t_D), Eq. (2) can be transformed to:

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

Eq. (3) predicts a decreasing $({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ 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 $({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ 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 $({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ 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 $({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ 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., $f = 1 - ({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ (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 ${}^{234}\mathrm{Th}$ 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 mechanism (Bandstra and Brantley, 2008).

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

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

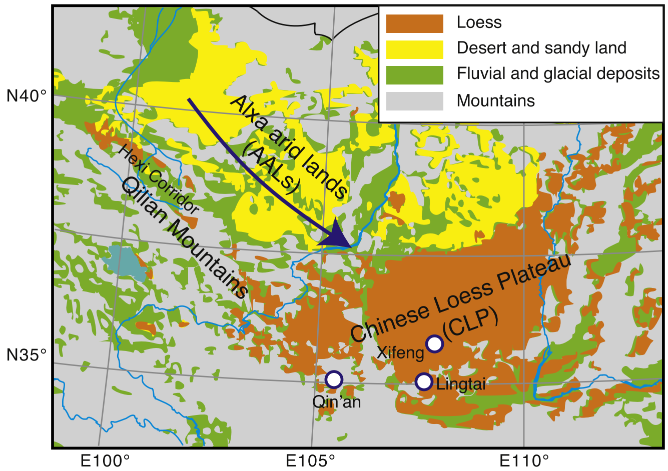

Fig. 1. Map showing geographic setting and locations of the sampling sites. Arrow illustrates the transport passway for eolian dust by the prevailing surface wind.

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

2. Samples and methods

2.1. Samples

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 $({}^{234}\mathrm{U}/{}^{238}\mathrm{U})$ 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