Testing mathematical uranium migration models in combined ESR/U-series dating of fossil teeth from open-air sites

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Abstract

Several mathematical models have been suggested to describe uranium migration kinetics for ESR dating of fossil tooth. In the present study, different U-uptake models were used to calculate ages for fossil teeth from two open-air Paleolithic sites in Nihewan Basin – Donggutuo and Hougou and compared with independent age control of the sites. For Donggutuo site, the results show that the ages of three teeth calculated by EU and AU models were much younger than magnetostratigraphic age. At Hougou site, two teeth from the top of Nihewan lacustrine deposits give the consistent EU and AU ages within errors but younger than sediment OSL age. This study indicates that mathematical modeling could not always give convincing results for the open-air sites, which may experience dramatic hydrological variations and high erosion rates.

Keywords: ESR/U-series method, fossil teeth, uranium migration model, Donggutuo site, Hougou site

1. Introduction

Electron spin resonance (ESR) dating of fossil teeth is a valuable dating tool for archaeological sites, especially of Middle and Lower Paleolithic age. However, due to the open system behaviors of uranium in each dental tissue (enamel, dentine and/or cementum), the uranium migration history needs to be considered in determination of the dose rates of tooth samples. Several uranium uptake models have been proposed in ESR dating of fossil teeth: the early uranium uptake (EU) model (U-accumulation shortly after tooth burial) and linear uranium uptake (LU) model (continuous and constant U-accumulation) were the two U-uptake models commonly used in early ESR dating studies of teeth (Ikeya, 1982; Grün and Stringer, 1991). The EU model could be thought as a closed system of uranium in teeth, it always gives an age younger than LU

model, so the two model ages have been used to bracket the supposedly correct sample age (Wrinn and Rink, 2003). In the case of late uptake of uranium in tooth samples, a recent uptake model (RU) was proposed (Blackwell *et al.*, 1992). In order to apply US-ESR model for tooth dating, Grün *et al.* (1988) suggested combining uranium series analysis with ESR measurement in the US-ESR model by the determination a U-uptake parameter p ($p \geq -1$), which describes the uranium accumulation in dental tissues. As there is no pre-determined assumption for U-migration history, US-ESR could yield a more reliable ages than EU, LU and RU models, which corresponds to specific p -values: $p = -1$, $p = 0$ and $p = \infty$ respectively. The US-ESR model has already exhibited its potential in dating of early hominid sites, such as Swartkrans in South Africa (Curnoe *et al.*, 2001) and Atapuerca Gran Dolina in Spain (Falguères *et al.*, 1999), but limits were also demonstrated in Early Palaeolithic sites of Orce Basin, Spain (Duval *et al.*, 2012a, 2012b). The main limitation is that the model does not allow $^{230}\text{Th}/^{234}\text{U}$ activity ratios in dental tissues of more than 1.04, while this is often observed in natural samples. To solve this limitation, an Accelerating Uptake (AU) model (Shao *et al.*, 2012) was introduced for describing the U-uptake into dental tissue as an accelerating process. This model, with two parameters, initial uptake rate and acceleration of this uptake rate, is then able to reconstruct a process combining incorporation followed by leaching. An uptake parameter named n -value is introduced in AU model and corresponds to the ratio of these two parameters (Shao *et al.*, 2012). When $n = 0$, the U-uptake is equal to LU model and the U-system will evolve towards secular equilibrium state in the case of $n \geq 0$.

Grün (2000a) also proposed the CSUS model, assuming that the present day uranium in a sample was accumulated at the apparent U-series age within a short time span, and the measured uranium contents in dentine and enamel of the teeth are used to calculate the total dose during the apparent U-series ages. Since CSUS model describes an extreme case of U-migration, it gives the maximum possible age of the tooth for a given ESR and U-series data set. Although some other mathematical models of U-migration have been also suggested in ESR dating of fossil teeth (e.g. saturation uptake model (Ikeya *et al.*, 1997) or D-A model (Millard and Hedges, 1996)), they all have their own limitations, and are not widely applied in dating studies. The mentioned U-uptake and leaching models above in ESR tooth dating are summarized in Table 1.

In this study, we used combined ESR and U-series methods with different U-uptake models mentioned above (EU, LU, and ESR/U-series (US and AU) models) to the teeth from two open-air Paleolithic sites in

Nihewan Basin, China, Donggutuo and Hougou sites, which are well-constrained by independent dating methods, to test the reliability of these mathematical models in ESR dating.

2. Materials and method

Mammalian fossil teeth from two Paleolithic open-air sites in Nihewan Basin, Donggutuo and Hougou, were studied (Figure S1). The corresponding archaeological layers are well dated by magnetostratigraphy about 1.1 Ma (Wang *et al.*, 2005; Ao *et al.*, 2010) for Donggutuo site, and 363 ± 14 ka by OSL method for Hougou site (Nian *et al.*, 2013). In this work, combined ESR and U-series analyses were carried out with different U-uptake models for age calculation of the fossil tooth. The samples from Donggutuo and Hougou sites were all fragmental pieces, which may represent the distinguished taphonomic features of Nihewan Paleolithic sites (Wei Qi, personal communication). All samples present enamel and dentine tissues. The two dental tissues were separated mechanically with a dental drill, enamel surfaces was cleaned on both dentine and sediment sides to remove the effects of external alpha radiation, then ground, sieved (100–200 μ m) and split into 15 aliquots for gamma irradiation from 125 to 50000 Gy for Donggutuo samples and 10 aliquots from 80 to 5000 Gy for Hougou samples with a calibrated ^{60}Co GammaCell 220 research irradiator (LNHB, Saclay, France). U-series analyses were performed on each dental tissue of fine powder by α -spectrometry for uranium concentration and radioisotopic ratios ($^{234}\text{U}/^{238}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$) measurement, following the procedure of Bischoff *et al.* (1988). Radon loss of both enamel and dentine was assessed from combined γ and α -spectrometry measurements (Bahain *et al.*, 1992).

The ESR measurements were carried out on a Bruker EMX ESR spectrometer (X band, 9.82 GHz) at room temperature with the following acquisition parameters: 1 mW microwave power, 1024 points resolution, 100 kHz modulation frequency, 0.1 mT modulation amplitude, 20 ms conversion time and 5 ms time constant. Each sample was measured 4 times in order to check the reproducibility of the data.

The ESR intensity was extracted from peak-to-peak amplitudes (T1-B2) of the ESR signal of enamel (Grün, 2000b). In this study, the equivalent doses (D_E) were determined by fitting the experimental data points using an exponential plus linear (EPL) function with Origin 8.0 software and $1/I^2$ weighting (Figure S2). The single saturation exponential (SSE) function, used commonly in the previous ESR dating studies of fossil teeth, cause D_E overestimations for early Pleistocene samples (Duval *et al.*, 2009). The double saturation exponential (DSE) function may better describe the analytical data points than conventional single

saturation exponential (SSE) function (Duval *et al.*, 2009, Han *et al.*, 2011) but gives meaningless fitting parameters and much higher relative errors. As adjusted R-square (Adj. R^2) values obtained by using EPL function are very close to DSE function and considering the lower relative errors of D_E fitting results by EPL function (3% to 8% for Donggutuo samples, and 22% to 30% for Hougou samples) and meaningful fitting parameters compared with DSE, we chose to use D_E values of EPL fitting function for calculating the combined ESR/U-series age of fossil teeth.

The external γ -dose rate from the sediment was measured *in situ* by portable Canberra Inspector 1000 γ -spectrometer with a calibrated NaI detector. Since the fossil bearing lacustrine deposits are homogenous, there were no obvious dose rate changes in the culture layer of the same site. Sediments were also collected for U, Th and K measurement to evaluate β -dose rate contribution in the laboratory of Department of Prehistory, MNHN, Paris with HpGe γ -ray spectrometer at liquid nitrogen temperature. The sediment measured data were shown in the note of Table S1.

3. Results and discussion

The U-series analyses of both Donggutuo and Hougou sites show relatively high uranium concentrations in the enamel (Table S2), which range from 1.17 to 16.33 ppm, and between 72.82 and 143.69 ppm in dentine. The $^{234}\text{U}/^{238}\text{U}$ ratios of enamel and dentine are similar, ranging from 1.75 to 2.10 and from 1.74 to 2.23, respectively. The $^{230}\text{Th}/^{234}\text{U}$ ratio ranges between 1.18 and 1.40 for enamel and from 1.35 to 1.75 for dentine, far beyond equilibrium, which indicate uranium leaching. In this case, US-ESR model cannot be applied for age calculation. Such limitations of US-ESR model were also observed during the study of Early Pleistocene sites in Orce Basin (Duval *et al.*, 2012a). Since EU and LU models are two special cases of US-ESR model (EU model: $p = -1$; LU model: $p = 0$), they also can only describe the uranium uptake instead of leaching, and are inappropriate for the age calculation of Donggutuo and Hougou teeth. The AU model can describe a process of uranium uptake followed by leaching, so in this study it is feasible for calculating sample ages. The dose rate and age results for the AU model calculations are shown in Table 2. The calculated n values from U-series and ESR data are all negative, as a result of uranium leaching. We place the EU and LU model ages in the Table 3 just for comparison.

The different components of dose rate contributed by dental tissues and surrounding environment are

shown in Figure 1. For the three teeth from Donggutuo, the internal alpha dose rates were the main components of the total dose rate, ranges from 42% to 79%; the beta dose rates from dentine are the second largest contributors to the total dose rate, between 13% and 33%. The environmental dose rates from the sediments and cosmic rays vary between 8% and 25%. For the two teeth from Hougou, both internal alpha dose rates and environmental dose rates made the largest contributions to the total dose rate, varying from 36% to 43% and 30% to 40% respectively. The beta dose rates from dentine contributed 24% to 27% to the total dose rate.

Comparing with the reference age of Donggutuo site (~1.1 Ma) constrained by palaeomagnetic study, both EU and AU model ages are much younger, and nearly 1/3 of the palaeomagnetic age for DGT0805 and DGT0807. This is probably attributed to the high uranium concentrations in the enamel of DGT0805 and DGT0807, both higher than 10 ppm. According to recent study by Joannes-Boyau and Grün (2011), the subsamples of tooth enamel with higher uranium concentration close to the dentine/enamel junction will yield smaller D_E values than those with lower U-concentration close to the surface because of their higher concentrations of non-oriented CO_2^- radicals generated by gamma irradiation. In this study, we did not observe this trend since we analyzed mixed enamel powder from both surface and dentine sides. In order to check whether the high uranium concentration caused alpha-efficiency variations, we introduced a k-value of zero into age calculation (Inverse correlation assumption, Bahain *et al.*, 1992). The age results of both EU and AU models become consistent with the reference age within error. In this case, we could consider the high U-concentrations in enamel as the main reason for age underestimation.

For the Hougou teeth, calculated EU and AU model ages are slightly younger than the reference age obtained by OSL (~ 360 ka, Nian *et al.*, 2013) using the same calculation parameters of sediment (water content, U, Th and K content etc.) (Figure 2). Although LU model ages obtained in this study and by some scholars (Schwarcz *et al.*, 1994) seems showing better agreement with other dating evidence, this simple U-uptake model does not have strong theoretical basis, and agreement with independent age control is purely incidental.

For Nihewan sites, the unexpected young ages seem related in part to the high U-content in enamel and to the high isotopic ratio, but we also could not exclude the effect of thermal instability of the ESR signal as well as a change of external dose rate related to the uplift and incision of the basin during the late

Pleistocene period. Chen *et al.* (2001) studied early Pleistocene teeth from Longgupo Cave with isothermal annealing, and observed mixed first and second-order kinetics for radicals involved in the ESR enamel signal which may lead an age underestimation. More detailed studies by Joannes-Boyau and Grün (2011) found that the unstable non-oriented CO_2^- radicals (NOCORs) in tooth enamel could not be removed by post-irradiation heating or long storage times. This may cause significant age underestimation. However, their experiments are based on fragment analysis, and annealing protocols of powder studies need to be developed in order to assess the amount of unstable NOCORs of irradiated signal.

The two sites studied in this work are both open air sites. In the basin, a thick lacustrine layer was exposed to high erosion rates which were accompanied by variations of hydrological environments. The migration of uranium between ground water and dental tissues of is complex and may occur in multiple phases (Grün *et al.*, 2014). Grün (2009b) showed that the p-values of teeth from open-air sites vary greatly. Therefore, for the fossil teeth from open air sites, it is not possible to make simple U-uptake assumptions for age calculation. Developed U-uptake models may work on some Late or Middle Pleistocene sites, however, they do not always give the reliable results for old sites if complex erosion occurred with repetitive uplift and incision processes. If possible, detailed U-isotopes mapping is indeed a useful and essential technique to better understand the U-mobilization in the teeth samples and facilitate the sample selection (Grün *et al.*, 2014). Fossil teeth with high U-concentration in enamel and high $^{230}\text{Th}/^{234}\text{U}$ ratios in dental tissues should be avoided or abandoned since it is virtually impossible to obtain reliable ages regardless of the U-models used.

4. Conclusion

Combined ESR/U-series fossil dating has been applied on two open air sites Donggutuo and Hougou, in Nihewan Basin, China, and several commonly used U-migration models have been tested in this study. ESR/U-series models do have advantages on dating fossils from cave and fissure sites (Han *et al.*, 2012; Shao *et al.*, 2014), but this study also shows their limitation on dating of fossil teeth from open air sites. The ages calculated by EU and AU models are much younger than the references for Donggutuo site, and even if LU model ages agree with the reference occasionally for some samples, there is no theoretical basis of this parametric U-uptake model. This study indicates that the uranium migration models for combined ESR/U-series dating of fossil teeth from open sites are driven by the erosion and hydrological conditions of

burial environment. Both simple and developed mathematic U-uptake (leaching) models are difficult to apply to old (Early Pleistocene) fossil tooth samples when the complexity of U-migration during the burial history cannot be reconstructed. For these open air samples, U-uptake/leaching model ages should be used with caution. Information of dramatic hydrological variation and erosion processes at the site should be investigated. Systematic uranium series isotopes mapping of dental tissues before sample preparation is highly recommended in order to abandon the high U-content enamel samples and better understand the uranium distribution and migration in the fossil teeth (Duval *et al.*, 2011).

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Figure captions:

Figure 1 Percentage contribution by α , β , γ and cosmic dose rates to the total dose rate of Donggutuo and Hougou teeth samples

Figure 2 Comparison of three U-uptake (leaching) model ages with independent age control (the shade of blue indicates the reference age by paleomagnetic and astronomical dating methods (Wang *et al.*, 2005; Ao *et al.*, 2010); the red one brackets the range of reference age by OSL dating (Nian *et al.*, 2013))

Table captions:

Table 1 Different uranium migration models for ESR fossil dating

Table 2 Dose rate components calculated with AU model by combined ESR/U-series analysis

Table 3 Sample ages calculated with different U-uptake (leaching) models.

Figure 1

Figure 1 Percentage contribution by α , β , γ and cosmic dose rates to the total dose rate of Donggutuo and Hougou teeth samples

Figure 2

Table 1 Different uranium migration models for ESR fossil dating

Models	Description	Expression	Diagram	Limitation	References
Early uptake (EU-ESR) model	Uranium accumulated shortly after tooth burial	$U(t) = U_m$ U(t) - U concentration at any time t; U_m - maximum U concentration; T - the age of tooth sample		Artificial assumption, ideal case based on geochemical process of uranium	Rae and Ivanovich, 1986
Linear uptake (LU-ESR) model	Uranium accumulated continously after tooth burial	$U(t) = U_m (t/T)$ * same parameters as in EU		Artificial assumption, based on experiences	Ikeya, 1982
U-series ESR (US-ESR) model	Uranium accumulated as a function of time, depending on a parameter p	$U(t) = U_m (t/T)^{p+1}$ * same parameters as in EU; uptake parameter $p \geq -1$ ($p = -1 \rightarrow$ EU; $p = 0 \rightarrow$ LU)		No solution in the case of U-leaching	Grun <i>et al.</i> , 1988
Recent uptake (RU-ESR) model	Uranium accumulated shortly and recently	$U(t) = U_m (t/T)^2$ * same parameters as in EU		Artificial assumption, only yield maximum age limit	Blackwell <i>et al.</i> , 1992
Close-system U-series ESR (CSUS-ESR) model	Uranium accumulated instantly at the time of the apparent U-series date	$U(t_a) = U_m$ t_a - apparent U-series age of tooth sample		An open system model provide a maximum possible age	Grun <i>et al.</i> , 2000
Accelerating uptake (AU-ESR) model	Uranium accumulated as an accelerating process	$\frac{dU_g}{dt} = f_g/\lambda_g + a_g t/\lambda_g \rightarrow U_g(t) = f_g/\lambda_g (n/2 \cdot t^2 + t)$ U_g - ^{238}U content at any time t; λ_g - decay constant of ^{238}U ; f_g - initial uptake rate of ^{238}U ; a_g - accelerations of f_g ; $n = a_g/f_g$		Applied to moderate U-leaching case; discussed in this study	Shao <i>et al.</i> , 2012

Table 2 Dose rate components calculated with AU model by combined ESR/U-series analysis

Sample No.	γ sediment+ cosmic ($\mu\text{Gy/a}$)	Internal dose ($\alpha+\beta$) enamel ($\mu\text{Gy/a}$)	β dentine ($\mu\text{Gy/a}$)	β sediment ($\mu\text{Gy/a}$)	Total dose rate ($\mu\text{Gy/a}$)	N-enamel	N-dentine	AU-ESR age (ka)
DGT0805	1052 ± 105	7721 ± 988	2579 ± 330	183 ± 18	11535 ± 1047	-0.0031 ± 0.00026	-0.0032 ± 0.00027	445 ± 32
DGT0806	1052 ± 105	1949 ± 199	1491 ± 152	107 ± 11	4599 ± 270	-0.0021 ± 0.00012	-0.0022 ± 0.00013	643 ± 33
DGT0807	1052 ± 105	11694 ± 1849	1895 ± 300	141 ± 15	14782 ± 1876	-0.0037 ± 0.00043	-0.0039 ± 0.00044	378 ± 36
HG0801	925 ± 93	892 ± 636	615 ± 438	73 ± 8	2505 ± 779	-0.0058 ± 0.0016	-0.0061 ± 0.0016	265 ± 59
HG0802	925 ± 93	1440 ± 600	921 ± 383	100 ± 10	3386 ± 719	-0.0046 ± 0.00091	-0.0049 ± 0.00092	304 ± 47

***Note:** The AU-ESR ages of the samples were calculated using a program based on MATLAB software (Shao *et al.*, 2012) with the following parameters and assumptions: alpha efficiency of 0.13 ± 0.02 (Grün and Katzenberger-Apel, 1994); dose rate conversion factors of Adamiec and Aitken (1998); Monte-Carlo beta attenuation factors (Marsh *et al.*, 2002) based on the thickness of the enamel layer, before and after its preparation; water contents of 3 wt% in the enamel, 7 wt% in the dentine and 15 wt% in sediment were assumed.

Table 3 Sample ages calculated with different U-uptake (leaching) models

Sample No.	EU age (ka)	LU age (ka)	AU age (ka)	Reference age
DGT0805	451 ± 44	835 ± 79	445 ± 32	1.06 ~ 1.12 Ma
DGT0806	776 ± 54	1204 ± 79	643 ± 33	
DGT0807	405 ± 50	764 ± 92	378 ± 36	
HG0801	274 ± 62	396 ± 90	265 ± 59	363 ± 14 ka
HG0802	315 ± 95	486 ± 147	304 ± 47	

***Note:** EU, LU ages were calculated with DATA program ([Grün, 2009a](#)) with the same parameters as in the note of Table 2.