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Research paper

Monte Carlo approach to calculate US-ESR age and age uncertainty for tooth enamel

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ABSTRACT

The combination of electron spin resonance (ESR) and U-series dating approach is increasingly being used to fossil teeth from archaeological sites. A rigorous age uncertainty assessment is needed for this dating method. However, it is difficult to provide partial derivatives of the combined ESR/U-series (US) model, as required by the law of propagation of uncertainties. In this study, we developed a new age calculation MATLAB program, called USESR, using a Monte Carlo approach for estimating the age and the age uncertainty for tooth enamel. Tests have been performed with virtual samples ($n = 64$). The results suggest that this Monte Carlo approach can provide reliable US-ESR age and reduced age uncertainty in comparison with those obtained by the routinely used program, DATA. The results also show that the new program has higher tolerance limits of U-series disequilibrium than the DATA program for US-ESR age calculations.

1. Introduction

Electron spin resonance (ESR) dating has become an efficient tool in Quaternary geochronological studies. Although ESR dating can be applied to a variety of materials, tooth enamel is one of the most widely used materials for ESR dating studies owing to its ubiquitous nature. Given the open system behavior of fossil teeth, the ESR dating of tooth enamel needs to model the uranium uptake histories of the dental tissues for an accurate evaluation of the dose rate (Grün, 1989, 2006; Rink, 1997). To overcome this challenge, Grün et al. (1988) developed the combined ESR/U-series (US) model, assuming that the U-content of dental tissue at a past time (t) can be expressed as a function of the measured present-day U-content (U_m), the burial time (T), and the uptake parameter (p, $p \geq -1$) in an equation of the form $U(t) = U_m(t/T)^{p+1}$. On the basis of this function, Grün et al. (1988) then developed expressions for time-dependent $^{234}U(t)$, $^{230}Th(t)$, and the dose rates. Using this model, the dose rate evaluation includes the U-series disequilibria for each dental tissue; thus, this model may yield an age estimate (US-ESR age) compatible with both the measured ESR and U-series data (Grün et al., 1988).

The US model is now increasingly used in the ESR dating of tooth enamel, although many other U-uptake models have been developed over the past decades (e.g., Blackwell and Schwarcz, 1993; Ikeya et al., 1997; Grün, 2000; Hoffmann and Mangini, 2003; Shao et al., 2012). The US-ESR age is routinely calculated with a DOS based program, DATA, which can also be used for early uptake (EU) model and linear uptake (LU) model age calculations for tooth enamel (Grün, 2009a,b). So far, DATA has produced hundreds of US-ESR dates, and these dates are often compared with the results given by independent dating methods (e.g., Peresani et al., 2008; Valladas et al., 2008; de Torres et al., 2010; Porat et al., 2010; Wagner et al., 2010; Mercier et al., 2013; Michel et al., 2013; Rink et al., 2013; Falguères et al., 2013). However, few studies present the details of the US-ESR age calculation with the age uncertainty assessment. The present work attempts to evaluate the US-ESR age and the age uncertainty using the Monte Carlo approach, which is one of the easiest and most flexible methods for uncertainty estimation, regardless of the problem complexity (BIPM et al., 2008a). This paper first introduces the algorithm used for US-ESR age calculation, describes the Monte Carlo approach used for US-ESR age and age uncertainty evaluation, presents the results obtained on virtual samples ($n = 64$) using the new approach, and finally compares the results with those from the DATA program.

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2. Algorithm used for US-ESR age calculation

The basic principle of ESR dating of tooth enamel is that the accumulated dose in the enamel (D_e) is a function of the natural dose rate (R) and the time of radiation exposure (T):

$$D_e = \int_0^T R(t)\mathrm{d}t \tag{1}$$

Thus, D_e is the total dose that the enamel has received since the burial event. The dose rate primarily consists of an internal dose rate (α and β) generated by the U in the enamel, an external β dose rate due to the U in the dentine and/or cement, and an external dose rate (γ and sometimes β) related to the U, Th, and K in the surrounding sediment plus cosmic rays (Grün, 1989).

The open behaviors of dental tissues to uranium and the U-series disequilibrium, both in sample and its environment, cause R to vary with time. As a result, it is impossible to calculate T by direct division of D_e by R, as it is usually done for the ESR dating of quartz (e.g., Voinchet et al., 2010). Using the US model, the tooth enamel age can be calculated from the combination of the measured ESR and U-series data sets through the reconstruction of the U-uptake processes according to U-series disequilibrium states in each dental tissue. Fig. 1 shows a schematic of the algorithm used for US-ESR age calculation, which may be divided into five successive steps.

2.1. Calculating p–t relationship

The first step uses the measured ^{234}U/^{238}U and ^{230}Th/^{234}U activity ratios in each dental tissue to calculate the relationship between the uptake parameter (p) and time (t). Grün et al. (1988) provided expressions for time-dependent ^{238}U, ^{234}U and ^{230}Th as functions of p and t (U_8(p, t), U_4(p, t), and Th_0(p, t), respectively). Accordingly, the activity ratios of ^{234}U(t)/^{238}U(t) and ^{230}Th(t)/^{238}U(t) can be calculated for the US model. Then, a single model age can be derived from the activity ratio functions for a given p-value and pair of measured ^{234}U/^{238}U and ^{230}Th/^{238}U ratios. By repeating calculations of t for a series of p-values, a relationship between p and t can be established. Graphically, t increases with the increase of p ($p \geq -1$) (see Fig. 2 in Grün et al., 1988). The p–t relationship is crucial for calculating the internal dose from the enamel and the external β dose from dentine and/or cement (see step 2.2).

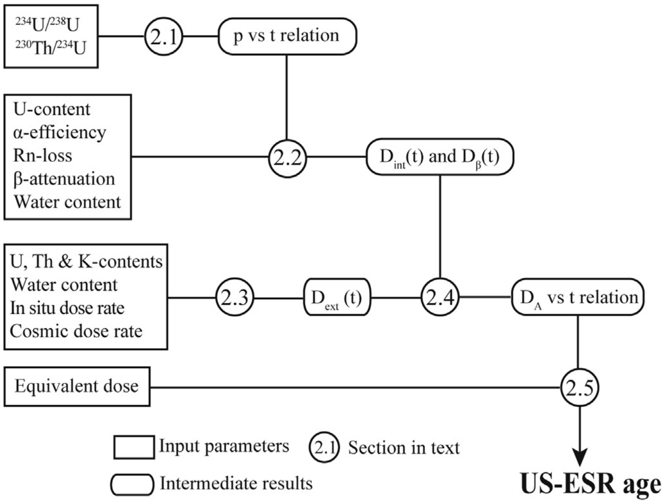

Fig. 1. Schematic illustration of the algorithm used for US-ESR age calculation.

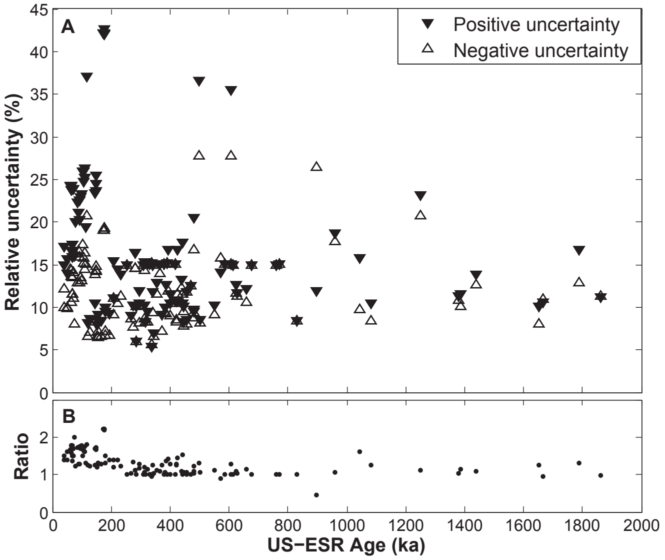

Fig. 2. Relative uncertainties of the US-ESR age estimates ($n = 125$) calculated by DATA program. Data in Fig. 2A are from La Micoque (Falguères et al., 1997), Hexian (Grün et al., 1998), Gran Dolina (Falguères et al., 1999), Panxian Dadong (Jones et al., 2004), Skhul (Grün et al., 2005), Ambrona (Falguères et al., 2006), Song Terus Cave (Hameau et al., 2007), Isernia (Falguères et al., 2007; Shao et al., 2011), Visogliano (Falguères et al., 2008), Liucheng (Rink et al., 2008), Arago (Han et al., 2010), Cuddie Springs (Grün et al., 2010), Mauer (Wagner et al., 2010), Venta Micena (Duval et al., 2011), Dali (Yin et al., 2011), Longgupo (Han et al., 2012). The ratios in Fig. 2B are the positive uncertainties in Fig. 2A divided by the corresponding negative ones.

2.2. Calculating internal dose (D_{int}) and external β dose ($D_β$)

The accumulated doses of D_{int} and $D_β$ are related to radioactive emissions produced by the U incorporated in the enamel and in the dentine and/or cement, respectively. The external α dose is not considered because the corresponding part of the sample is eliminated when ≥50 μm of the enamel surface layers are removed during sample preparation. Furthermore, the γ dose from the dental tissues (except for elephant teeth) is overlooked due to the long effective ranges of γ-rays in enamel (>20 cm). The D_{int} and $D_β$ doses generated in the time period from 0 to t is the sum of three decay components: the decay of ^{238}U to ^{234}U, the decay of ^{234}U to ^{230}Th, and the decay of ^{230}Th to ^{206}Pb (Grün et al., 1988). Using the p–t relationship and the expressions of $U_8(p, t)$, $U_4(p, t)$ and $Th_0(p, t)$, $D_{int}(t)$ and $D_β(t)$ can be efficiently evaluated for any t, including corrections for water content (Grün, 1994), α-efficiency (Grün and Katzenberger-Apel, 1994), β-attenuation (Marsh, 1999) and radon-loss (Bahain et al., 1992).

2.3. Calculating external dose (D_{ext})

In the third step, the external dose (D_{ext}) is quantified as the sum of the accumulated dose generated by cosmic rays and the radioactive elements in the surrounding sediment (mainly ^{238}U, ^{232}Th and their daughters, and ^{40}K). By assuming the U- and Th-series maintain a secular equilibrium, the dose calculation is simplified. Thus, D_{ext} can be assessed by in-situ γ dose rate measurements with thermoluminescent (TL) dosimeters (Valladas, 1982) or portable γ-spectrometry (Mercier and Falguères, 2007). On the other hand, D_{ext} can be calculated from the measurements of the U, Th, and K contents in the surrounding sediment (Adamiec and Aitken, 1998). Finally, the cosmic dose rate can be determined according to the latitude, altitude, and burial depth of the analyzed sample (Prescott and Hutton, 1994).