

An alternative for model for open system U-series/ESR age calculations: (closed system U-series)-ESR, CSUS-ESR

Rainer Grün
Research School of Earth Sciences
The Australian National University
Canberra ACT 0200, Australia

(Received 18 January 2000, in final form 28 April 2000)

One of the major, well known problems in ESR dating of teeth is the modelling of the U-uptake history. Originally, Ikeya (1982) proposed the early (EU) and linear (LU) U-uptake models. Subsequently, Grün et al. (1988) developed combined U-series/ESR (US-ESR) dating where the U-uptake is modelled from the measured U-series disequilibrium values in the constituencies of a tooth (enamel and dentine). Some detailed U-series measurements on teeth from Pech de l'Aze (Grün et al. 1999) showed that the parametric U-series/ESR, US-ESR, results agree well with the predictive uranium diffusion of Millard (1993; also Millard and Hedges (1996)), see Pike and Hedges (in press). Interestingly, the model of Millard (1993) predicts that U-uptake of tooth enamel lies more or less in the middle between early and linear U-uptake, which agrees more or less with empirical observations (e.g., Grün and Stringer 1991).

In this paper, I present a new model for open system modelling which arose from the detailed analysis of a tooth from the Naracoorte Caves in South Australia (Grün et al., in press). Sample 1369 is a *Zygomaturus* (a large, wombat-like marsupial) tooth which was found within the sediment layers of the Fossil Chamber of Victoria Cave. The clastic, bone bearing sediments are capped by a small stalagmite with a TMS U-series age of 213 ± 7 ka (Moriarty et al. in press). Although the tooth was not analysed itself by U-series, a series of ten bone samples from various places within the sequence gave closely similar U-series results with average values for $^{234}\text{U}/^{238}\text{U}$ of 1.55 ± 0.16 and $^{230}\text{Th}/^{234}\text{U}$ of 0.65 ± 0.04 , corresponding to an average apparent age of 105 ± 11 ka. The U-series results had no relationship to the stratigraphic position in the sequence, implying that the overall U-accumulation was governed by processes that started after the

deposition of the whole sedimentary sequence. The average U-series ratios were used for open system U-series/ESR modelling.

Six enamel sub-samples were analyzed from the tooth. The enamel had a total thickness of about 2000 μm and samples were collected at different depths (see Figure 1). Figure 1 shows increasing U-concentrations from the outside (sediment side: base of Figure 1) towards the inside (dentine side: top of Figure 1), which implies that the main U-uptake of the enamel took place from the dentine side rather than from the outside of the tooth. ESR doses and other parameters were measured (all analytical details are given in Grün et al. (in press)) and age estimates were calculated according the EU, LU and US-ESR models (see Figure 2).

The average US-ESR uptake function is close to the LU model (Figure 2A) and consequently, the LU and US-ESR age estimates are close (Figure 2B). All age results have a clear trend in common: increasing apparent ages with decreasing U-concentration in enamel. This is caused by the fact that the D_e values do not show any such trend with depth or U-concentration. Thus, it appears that the dependence of the total dose rate on U concentration is less than would be expected from the applied open system models.

The data points can be used for extrapolation to zero U-concentration and the intercept with the Y-axis yields the ages that are independent of the internal U-concentration and should, in principle, be independent of the U-uptake model (see Blackwell and Schwarcz 1993). The Y-intercepts are 340 ± 35 ka (EU), 406 ± 38 ka (LU) and 454 ± 48 ka (US-ESR) using the York-fit option of the Isoplot program by K. Ludwig (see Ludwig and Titterton, 1994). The

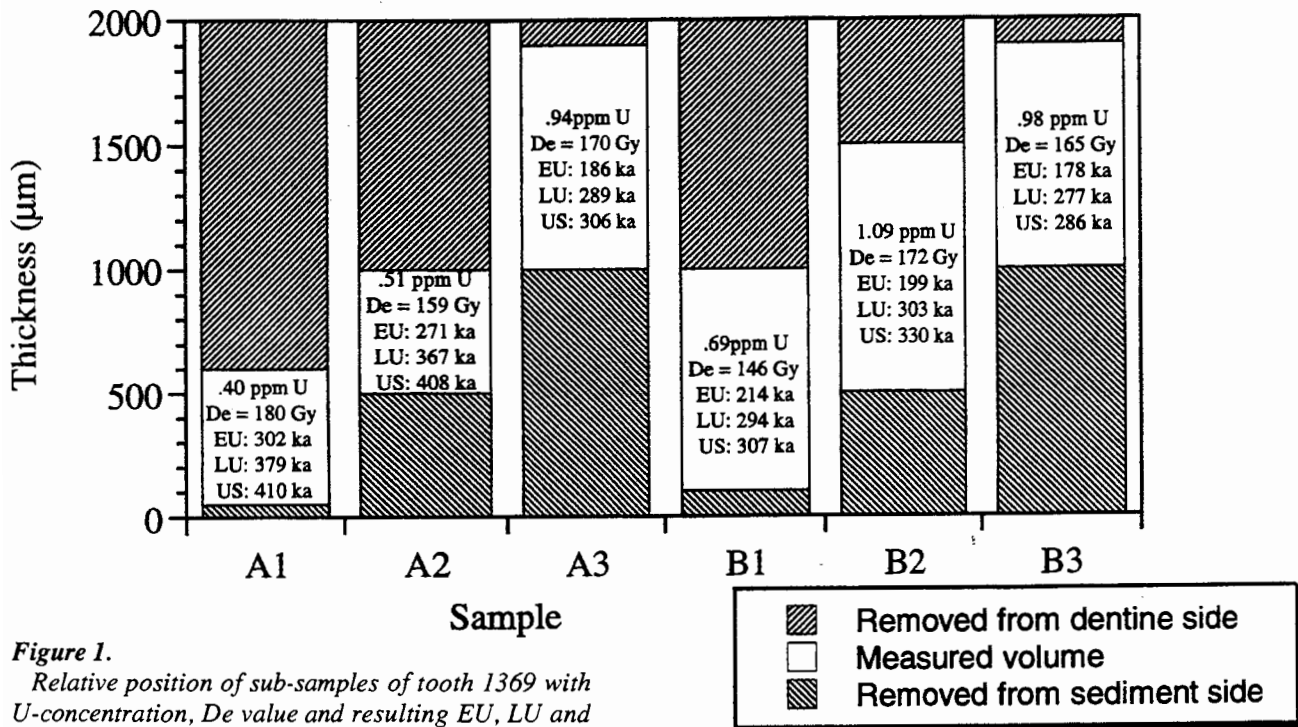


Figure 1.

Relative position of sub-samples of tooth 1369 with U-concentration, D_e value and resulting EU, LU and US-ESR model ages.

intercepts are all about 100 ka older than the weighted means (EU: 210 ± 7 ka; LU: 308 ± 10 ka, US-ESR: 342^{+13}_-6 ka). The EU model, which is clearly inappropriate for this sample (the sediment sequence ought to be older than the covering speleothem), yields an intercept which is considerably smaller than those of the LU and US-ESR model.

The question is, why are the D_e -values more or less independent of the U-concentrations? One reason may lie in the use of inappropriate parameters for dose rate calculation. For example, for the alpha efficiency, a value of 0.13 ± 0.02 (Grün and Katzenberger-Apel, 1994) was assumed and beta attenuation factors of Brennan et al. (1997) were used in all calculations. These factors could overestimate the U-dose rates. Another explanation lies in the appropriateness of the chosen U-uptake functions. If the uranium was acquired long after burial, the contribution of the U-dose rate to the total dose rate would be significantly smaller than calculated by the models.

The closely similar U-series ratios measured on the bone samples imply that U-accumulation took place after the deposition of the sedimentary sequence which ought to be older than about 213 ka. A possible model to explain the U-series results for the bones is that during a pluvial period around 105 ka, the faunal elements in the deposits accumulated their present uranium concentrations within a relatively short time span. Coincidentally, the analysis of speleothem frequency data (Ayliffe et al. 1998) established a wet period around this time (105-115 ka).

For modelling, I have used the U-series data of the bones. A simple delta function is assumed (similar to the EU model), i.e., all the uranium measured today in the sample was accumulated at the time of the apparent U-series date (Figure 3A). This simple model has several advantages: firstly, it gives a limiting, upper age for combined U-series/ESR modelling (unless U-loss is assumed and/or measured) and secondly, it is very simple to calculate. The enamel and dentine uranium dose for a given time and measured U-series isotopic data is calculated according to equation (A-4) in Grün (1989), considering appropriate attenuation factors, water concentrations and error calculations. The total closed system enamel and dentine U-dose is then subtracted from the D_e value. The resulting external dose is then divided by the external dose rate (sum of sediment beta, gamma and cosmic dose rates). Figure 3B shows the result of the CSUS-ESR age calculations. The Y-axis intercept, 440 ± 58 ka, is well within the error of the weighted mean of the individual CSUS-ESR results (417 ± 21 ka). The average CSUS age also agrees well with the extrapolated LU and US-ESR ages of 406 ± 38 ka and 454 ± 48 ka, respectively. It is clear that further work (measurement of all U-series ratios on all subsamples) ought to be carried out to check whether the CSUS-ESR model is correct for sample 1369. Interestingly, if linear uptake is assumed starting at about 213 ka (i.e. U-accumulation starts after the deposition of the capping speleothem), which corresponds roughly to the apparent closed system

age of 105 ka, the resulting ESR calculations show a negligible difference to the CSUS-ESR calculations.

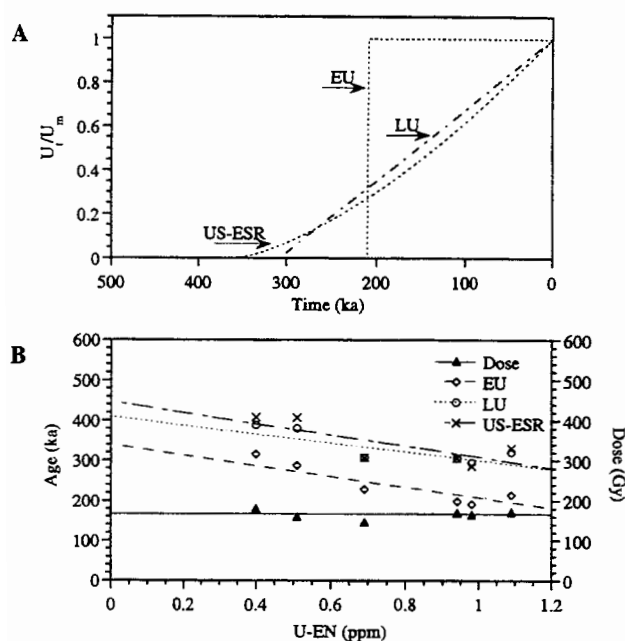


Figure 2.

(A) U -uptake functions for the average model ages of sample 1369.

(B) Calculated ages and dose values plotted versus internal U -concentration. All age calculations are dependent on internal U -concentration whereas the D_e value does not show any U dependency. This implies that the internal U dose rates are overestimations, either through incorrect choice of dose rate parameters (e.g., α -efficiency) or U -uptake model. Errors are omitted for clarity. The Y -intercepts (i.e. age estimation for zero internal U -concentration) calculated the York-fit option of Isoplot (see Ludwig and Titterton 1994) are: 340 ± 35 ka (EU); 406 ± 38 ka (LU) and 454 ± 48 ka (US-ESR).

The CSUS-ESR model can only be applied if U -series isotopic data are available. The calculation of age results is trivial. The CSUS-ESR model provides a *maximum possible age* for a sample because any delayed U -uptake will result in higher U -dose rates. As such, the model gives an indication of the robustness of the open system age calculations. The CSUS-ESR model seems appropriate in most cases where the apparent closed system U -series ages are significantly younger than the EU and LU ESR model ages (e.g., the results of Hoxne: Schwarcz and Grün (1993)). In most cases, the CSUS-ESR results differ little (within 10%) from US-ESR age calculations, particularly for relatively low U -concentrations (< 1 ppm U in enamel and 10 to 20 ppm in dentine). It seems therefore advisable to

calculate CSUS-ESR ages routinely in open system model calculations.

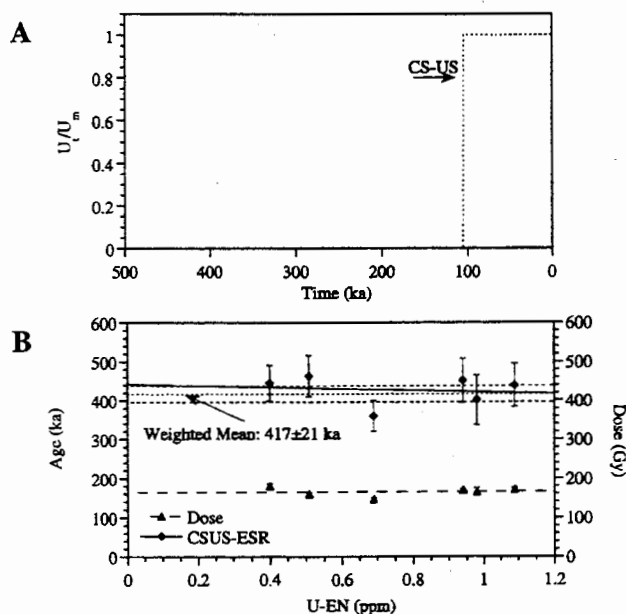


Figure 3.

(A) U -uptake function of the closed system U -series (CSUS) model.

(B) CSUS-ESR age calculations show little dependency on the internal U -concentration. The weighted average of 417 ± 21 ka agrees well with the extrapolated LU and US-ESR ages (see Figure 2B).

Acknowledgments

I thank Steve Robertson, Canberra, for corrections and Henry Schwarcz for a very lively discussion.

References

- Ayliffe, L.K., Marianelli, P.C., Moriarty, K.C., Wells, R.T., McCulloch, M.T., Mortimer, G.E. and Hellstrom, J.C. (1998) 500 ka precipitation record from southeastern Australia: Evidence for interglacial relative aridity. *Geology*, **26**: 147-150.
- Ayliffe, L.K. and Veeh, H.H. (1988) Uranium-series dating of speleothems and bones from Victoria Cave, Naracoorte, South Australia. *Chemical Geology*, **72**: 211-234.
- Blackwell, B.A. and Schwarcz, H.P. (1993) ESR isochron dating for teeth: a brief demonstration in solving the external dose calculation problem. *Applied Radiation and Isotopes* **44**: 243-252.
- Brennan, B.J., Rink, W.J., McGuirl, E.L., Schwarcz, H.P. and Prestwich, W.V. (1997). Beta doses in tooth enamel by "One Group" theory and

- the Rosy ESR dating software. *Radiation Measurements*, 27: 307-314.
- Grün, R. (1989) Electron spin resonance (ESR) dating. *Quaternary International*, 1: 65-109.
- Grün, R. and Katzenberger-Apel, O. (1994). An alpha irradiator for ESR dating. *Ancient TL*, 12, 5-38.
- Grün, R., Moriarty, K. and Wells, R. (in press) ESR dating of the fossil deposits in the Naracoorte Caves, South Australia. *Journal of Quaternary Science*.
- Grün, R., Schwarcz, H.P. and Chadam, J.M. (1988) ESR dating of tooth enamel: Coupled correction for U-uptake and U-series disequilibrium. *Nuclear Tracks*, 14: 237-241.
- Grün, R. & Stringer, C.B. (1991) ESR dating and the evolution of modern humans. *Archaeometry*, 33, 153-199.
- Grün, R., Yan, G., McCulloch, M. and Mortimer, G. (1999) Detailed mass spectrometric U-series analyses of two teeth from the archaeological site of Pech de l'Aze II: implications for uranium migration and dating. *Journal of Archaeological Science*, 26: 1301-1310.
- Ikeya, M. (1982) A model of linear uranium accumulation for ESR age of Heidelberg, Mauer, and Tautavel bones. *Japanese Journal of Applied Physics*, 21, L690-L692.
- Ludwig, K.R. and Titterton, D.M. (1994). Calculation of $^{230}\text{Th}/\text{U}$ isochrons, ages, and errors. *Geochimica Cosmochimica Acta*, 58, 555-564.
- Millard, A.R. (1993) *Diagenesis of archaeological bone: the case of uranium uptake*. DPhil. Thesis. University of Oxford.
- Millard, A.R. and Hedges, R.E.M. (1996) A diffusion-adsorption model of uranium uptake by archaeological bone. *Geochimica Cosmochimica Acta*, 60: 2139-2152.
- Moriarty, K.C., McCulloch, M.T., Wells, R.T. and McDowell, M.C. (in press) Mid-Pleistocene cave fills, megafaunal remains and climate change at Naracoorte, South Australia: towards a predictive model using U/Th dating of speleothems. *Palaeogeography, Palaeoclimatology, Palaeoecology*.
- Pike, A.W.G. and Hedges, R.E.M. (in press) Sample geometry and U-uptake in archaeological teeth: implications for U-series and ESR dating. *Quaternary Geochronology (Quaternary Science Reviews)*.
- Schwarcz, H.P. & Grün, R. (1993) ESR Dating of the Lower Industry. In (R. Singer, B.J. Gladfelder & J.J. Wymer. Eds) *The Paleolithic Archaeological Site at Hoxne, Britain*. Chicago: University of Chicago Press, pp. 210-212.

Rewiever

H. P. Schwarcz

Comments

Current research in ESR dating generally assumes uranium uptake by teeth either early in the burial history (EU) or by a process which has acted continuously up to the present. This paper introduces a new model: uptake as a discrete pulse, possibly controlled by some climatic event. Typically evidence for such an event-like uptake process would be seen in the uniformity of U-series dates throughout a deposit and, by implication, lack of stratigraphic order of these dates. It will be interesting to see if other examples of this phenomenon turn up.