

Bayesian analysis of Pleistocene Chronometric Methods

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ABSTRACT

Bayesian analysis of radiocarbon dates on Holocene archaeological sites has become well established. Application to Pleistocene sites dated by multiple techniques would be advantageous. This paper develops the necessary mathematical apparatus in the form of likelihoods for luminescence dating, and uranium series dating, and considers the possibility for amino-acid racemisation dating. Application of the new methods is illustrated using the stratigraphic sequences of dates from the sites of Saint Cesaire and La Chaise-de-Vouthon. For application to amino-acid racemisation dating fuller publication of data is found to be required.

Keywords: Bayesian statistics, luminescence dating, uranium-series dating amino-acid racemisation dating.

INTRODUCTION

Dating of Pleistocene archaeological and palaeoenvironmental sites is conducted using a wide range of chronometric techniques. In the last 30 years we have moved from a situation where the gap between the capabilities of radiocarbon dating and potassium-argon dating could be described as “the muddle in the middle” (Isaac 1975), to having the ability to apply a range of techniques (OSL, TL, ESR, Uranium-series, amino-acid racemisation, etc.). Often multiple techniques are applied at one site on samples with known stratigraphic relationships. It is also frequently the case that the object whose age is of interest (e.g. a hominid specimen) is not directly dated, but dated by stratigraphic association or ordering. In such cases we currently have no formal method to assess either the most likely age, or the uncertainty in that age, and as Ludwig (2003a) notes “for most studies *the uncertainty of the date is no less significant than the date itself*” (emphasis in original).

In contrast, in the Holocene radiocarbon is the dominant technique used (though far from the only one). Over the last 15 years Bayesian chronological modelling has been developed to the point of being a routine method for the analysis of stratigraphically related radiocarbon dates, allowing refinements in precision and the formal, numerical estimation of ages for objects related by stratigraphy to directly dated objects (Bayliss & Bronk Ramsey 2003, Buck 2003). The analysis of the ages of Pleistocene deposits and their contents could be greatly enhanced if we had similar mathematical methods which could be applied to stratigraphic sequences dated with multiple techniques (Millard 2003). This paper develops some of the necessary mathematical apparatus for such analyses and demonstrates its application to stratigraphic sequences of dates by single methods; extension to sequences dated by multiple methods will be treated in a future paper.

Each technique requires separate consideration of the mathematical structure of the calculations, and this paper tackles luminescence dating, uranium-series dating and amino-acid racemisation dating. ESR dating has been discussed elsewhere (Millard submitted).

The Structure of Bayesian Chronological Models

The basic idea of Bayesian chronological modelling is to combine dating information with the known ordering of the dates from stratigraphy:

refined dates = chronometry + stratigraphy,
 more formally this is expressed as a form of Bayes' theorem:

$p(\text{dates} | \text{chronometry}) \propto p(\text{chronometry} | \text{dates}) \times p(\text{dates})$,
 where $p(\text{dates})$ expresses our prior beliefs about the dates of events before obtaining
 chronometric measurements, $p(\text{chronometry} | \text{dates})$ is the likelihood which uses a
 mathematical model to express the probability of obtaining our observations if the
 dates were known and $p(\text{dates} | \text{chronometry})$ expresses our posterior beliefs
 incorporating our prior beliefs and the data. The prior beliefs can include statements
 about relative ordering of events, and thus incorporate stratigraphic information.

In addition, the dates of objects or strata not dated directly can be incorporated in the
 prior beliefs in order to obtain an estimate of their age given all the chronometric and
 stratigraphic data. Within the analysis of radiocarbon dates, the mathematical
 statement of the model is usually as a stratigraphic ordering in time of dates within
 broad limits, plus a mathematical statement of how a calendar date translates to a
 radiocarbon date via the calibration curve and laboratory measurement process (Buck
 2003). A similar structure is adopted here for other dating techniques, using a prior
 ordering statement combined with a likelihood describing the probability of observing
 the measured values given the age, chronometer mechanism and the measurement
 process. Thus the form of the likelihood needs to be derived separately for each
 dating technique, based on its chronometer mechanism and the required
 measurements. As in all previous models of prior ordering, the stratigraphic order is
 assumed known with certainty; although in principle uncertainty in stratigraphic
 ordering could be incorporated, the mathematical apparatus for doing this has not yet
 been developed.

A broad structure for constructing a chronology using multiple chronometric
 techniques applied to samples from a stratigraphic sequence may be developed as
 follows (Lanos 2003). The general equation governing the probability of a set of
 samples having a set of dates is:

$$p(\theta | \mathbf{M}, \mathbf{g}, S) \propto \prod_{i=1}^n p(\mathbf{M}_i | \mathbf{g}_i, \theta) \times \prod_{i=1}^n p(\theta_i | \theta),$$

where θ is the set of dates of interest, \mathbf{M} are the chronometric measurements, \mathbf{g} is any
 required set of calibration data or known parameters, and S is the stratigraphic
 information. The likelihoods of observations in two chronometric techniques are
 independent of one another if the techniques depend on different properties of the
 samples and their environments. In this case

$$p(\theta | \mathbf{M}, \mathbf{g}_M, S) \propto \prod_{j=1}^m \prod_{i=1}^{n_j} p(\mathbf{M}_{ij} | \mathbf{g}_{ij}, \theta_j) \times \prod_{j=1}^m p(\theta_j | \theta),$$

and we can use separate expressions for the likelihood, $p(\mathbf{M}_j | \mathbf{g}_j, S, \theta)$ of technique j .
 Further, some techniques date each sample separately and any parameters in common
 between samples (e.g. a half-life) are assumed to have negligible uncertainty. This is
 the usual assumption in the analysis of radiocarbon dates (Buck et al 1992) and is
 often applicable to closed-system uranium-series dates, but not to the other techniques
 considered here. In this case the joint likelihood over all samples for the method can
 be decomposed into the product of a series of likelihoods for the individual samples:

$$p(\mathbf{M}_j | \mathbf{g}_j, S, \theta_j) = \prod_{i=1}^{n_j} p(\mathbf{M}_{ij} | \mathbf{g}_{ij}, \theta_j),$$

and need only be derived for an individual sample.

In certain cases, the assumption of independence of techniques breaks down where they utilise a parameter in common, e.g. TL and ESR dating of a site may depend on one set of environmental γ -dose rate measurements. In these cases, a joint likelihood needs to be derived. This case is not tackled here. Falsely assuming independence of the techniques will lead to an underestimate of the uncertainty on dates compared to the dependent calculation. For sites where this occurs, the effort of deriving appropriate likelihoods should be weighed against the fact that commonality of one amongst many parameters may only lead to a small underestimate of uncertainty.

PRIOR PROBABILITIES FOR DATES

Early Bayesian modelling of radiocarbon dates assumed a uniform prior on all parameters, subject to the constraints of stratigraphic ordering and the limits of the radiocarbon calibration curve (e.g. Buck et al 1991, 1994a, 1994b, 1992). This very simple model attempted to allow the stratigraphy but no other aspects of the prior to influence the posterior probability distribution. Further research has shown that when there are large numbers of dates the choice of prior has little effect, but in cases with small numbers of dates (Nicholls & Jones 2001) or flat regions of the radiocarbon calibration curve (Steier & Rom 2000) the results can be significantly altered by choice of prior distribution. Steier and Rom (2000) note that stratigraphic ordering simply constrains the difference between dates to be positive and there is no obvious reason why the prior probability of difference should be uniform on the difference rather than on its logarithm, square root or some other transformation. Thus a uniform prior is mathematically convenient but has no strong modelling basis.

Bronk Ramsey (1998) observed that the usual method of conversion from $^{14}\text{C}/^{12}\text{C}$ ratio to conventional radiocarbon age is equivalent to an exponential prior on the radiocarbon age, but if this is replaced with a uniform prior normalisation becomes difficult for dates near the limit of the technique. He proposed the use of a prior proportional to the inverse square of age, as this is the “obvious choice mathematically”, being the lowest negative power which is integrable and scale invariant.

The priors to be adopted before any stratigraphic constraints are imposed are thus not immediately obvious and deserve some consideration. In any dating technique based on the decay of one isotope in comparison to a stable isotope, we have a likelihood of the form

$$A \sim \text{Norm}(\exp(-\lambda\theta), s)$$

where A is the observed ratio, λ is the decay constant, θ is the age and s is the observed standard deviation, assumed to be known exactly. Where one isotope accumulates in relation to another, as in K-Ar or U-Pa dating then it is $1-A$ that is observed, but the principles remain the same. Similar but more complex considerations apply in multiple isotope systems such as U-Th dating. If a uniform, improper prior on positive values is adopted for θ then the posterior probability is

$$p(\theta) \propto \exp(-\lambda\theta) \exp(-\frac{1}{2s^2}(\ln A + \lambda\theta)^2)$$

This has a finite value when θ is infinite whatever the values of the other parameters, and is therefore always improper. Bronk Ramsey (1998) investigated this prior numerically and only found a problem for $A/s < 6$. This is probably due to numerical underflow in computation of the probabilities for large θ when A is far from zero. However, it seems unsatisfactory to use a prior which in principle gives an improper posterior distribution but in practice is often computable due to underflow. Likelihood based methods (e.g. Ludwig & Titterton 1994) do not have this problem; for Bayesian methods to produce the same distributions and confidence intervals as likelihood methods requires the assumption of a prior which is the differential of the dating equation,

$$p(\theta) \propto \frac{1}{\theta^2}$$

The posterior is then

$$p(\theta | \lambda) \propto \frac{1}{\theta^2} e^{-\lambda \theta} \quad (1)$$

and is proper. This might be an "obvious" choice of prior, but it has the disadvantage that the scale of the prior changes with the dating technique, so that when techniques are combined there is no "obvious" prior distribution. Ludwig (2003a) has proposed a system for stratigraphically constrained Bayesian analysis of U-Th dates, with this type of prior implied. If more than one technique were used (or even U-Th and U-Pa) there could be problems with the implied prior. Ludwig's implementation of the method in IsoPlot (Ludwig 2003b) allows either Gaussian errors or U-Th asymmetric errors, but not both in combination, and thus avoids the problem.

Inspection of equation (1) shows that the posterior will remain proper for any exponential prior, so the prior may have a different "decay constant" λ_0 from the radioactive decay constant λ , and thus the posterior is:

$$p(\theta | \lambda) \propto \frac{1}{\theta^2} e^{-(\lambda + \lambda_0)\theta}$$

λ_0 can be interpreted as the rate of a Poisson process which destroys potentially datable samples, and embodies an expectation that recent objects are more likely to be found than ancient ones (compare Joyce 1999). This is a usable prior but requires an arbitrary choice of the value of λ_0 . However, from a Bayesian perspective, λ_0 is simply another parameter and need not take a specific value, but can be uncertain with a probability distribution as a hyper-prior, and under appropriate circumstances this can be integrated out before calculating any dates. With an improper uniform hyper-prior on λ_0 , combined with the exponential prior proposed above, integrating λ_0 gives a prior on θ of $p(\theta) \propto \frac{1}{\theta^2}$, just as suggested by Bronk Ramsey (1998). This is tantamount to saying that we have no knowledge of the rate of loss of datable objects, and assuming that any rate of loss is equally likely. However the argument of Steier and Rom (2000) regarding choice of a vague prior on differences applies equally here. An alternative view might be that we are uncertain as to the mean lifetime of datable objects, and a uniform prior on $1/\lambda$ should be taken. Integrating out this hyperprior gives $p(\theta) \propto \frac{1}{\theta^2}$.

Although these hyperpriors have partial theoretical justification in terms of a loss process for datable samples, they both give a prior for θ with the problem of an infinite mass at $\theta = 0$ and thus an improper posterior for all values of A , s and λ_0 .

However the inverse square prior can be made proper with the small modification of setting an arbitrary small limit to the age (Bronk Ramsey 1998). This can be justified on the grounds that when we date an object we don't believe it was formed the day before yesterday. The inverse square prior is therefore a less arbitrary than might seem from Bronk Ramsey's (1998) paper, and requires fewer arbitrary choices than the $1/\theta$ prior, which can only be proper if both upper and lower limits to θ are set.

A LIKELIHOOD FOR TL DATES

Luminescence dating utilises a deceptively simple “dating equation”, which hides a series of complexities in the method. The account given here is abbreviated and biased towards thermoluminescence dating but should be applicable to other forms of luminescence dating. The dating principles of luminescence dating parallel those of the other trapped charge technique, ESR dating, and thus the statistical model here parallels that for ESR (Millard 2003, Millard submitted). Roberts (1997) gives a more detailed treatment of the measurements and procedures required. Rhodes et al. (2003) have applied Bayesian statistical techniques to OSL dates at Old Scatness Broch using OxCal (Bronk Ramsey 1995), but note that their method has the disadvantage that “there is no easy way to treat systematic errors in a rigorously Bayesian way.” Here the systematic and random uncertainties are examined to produce a more coherent approach.

The dating equation is simply:

$$\text{age} = \frac{\text{total exposure to radiation}}{\text{rate of exposure to radiation}} = \frac{D}{\dot{I}}$$

When appropriately stimulated (by heat for thermoluminescence (TL) or light for optically stimulated luminescence (OSL)), the sample emits light in proportion to the radiation dose it has received since last being stimulated. This palaeodose, D_E , is determined by measurement of the luminescence of the sample and the changes in intensity of luminescence after application of additional artificial doses of radiation.

The dose rate, \dot{I} , is the sum of the rates from a series of sources of radiation, which are measured in a variety of ways:

- the dose from sample itself, \dot{D}_{int} , determined by measuring the uranium, thorium and potassium content of the sample;
- the gamma radiation dose from the sediment, \dot{D}_{γ} , determined either by *in-situ* gamma-spectrometry measurements or from chemical analysis of the U, Th and K content of the sediment and an assessment of the water content of the sediment;
- the beta radiation dose from the sediment, \dot{D}_{β} , estimated by the same means as the gamma-dose, and adjusted for the geometry of the sample using an attenuation factor. In many cases using TL this dose is eliminated from consideration by removing the outer 2mm of the sample.

All of these are measured with an associated error term. \dot{D}_{int} is based on measurements with errors independent for each sample. The same is assumed here for D_E although there will be some correlated components of uncertainty error in this term, due to factors like calibration uncertainty of the artificial radiation sources; these constitute only a minor part of the overall uncertainty, most of which is due to scatter

in the measurements. \dot{D}_γ (and \dot{D}_β) determinations usually apply to groups of dates, so their errors are not independent between samples in a group. Such dependence needs to be taken into account in analysis of the dates.

The likelihood expresses the probability of the observed D_E values if we knew the true date and the true values of the components of the dose rate. Consequently the dating equation is re-expressed as $D_E = \dots$. Consideration of the components of the dose rate shows that where there are multiple samples they fall into a hierarchy of groups for these parameters (Figure 1), and therefore also for the associated uncertainties. The values for \dot{D}_{int} , α -efficiency (if measured), and the β -attenuation factor are unique to a measured sample, whilst one true (but unknown) date, θ , is shared by sub-samples (or aliquots) from the same object. The other parameters derive from measurements on the environment in common to different sets of samples. The sediment beta dose is common to a group of samples from the same sediment. The gamma dose, \dot{D}_γ , is homogenous on a larger spatial scale, often for all samples from a stratum, and all samples with the same beta dose will have the same gamma dose. Finally the cosmic rays dose, \dot{D}_{cosmic} is the same across a whole site, but may be attenuated by varying overburdens of sediment for different samples; thus it usually parallels \dot{D}_γ in grouping of samples, whilst the uncertainty is common to all samples.

These differing associations of parameter determinations with different subsets of the dated samples are expressed in a statistical model with a hierarchy of parameters. Thus the model may be expressed as:

$$D_E^{(ijkl)} | \theta \sim N(\mu_Z, s_Z^2)$$

$$\mu_Z = \theta + \dot{D}_\gamma + \dot{D}_\beta + \dot{D}_{\text{int}} + \dot{D}_{\text{cosmic}}$$

$$\dot{D}_\gamma \sim N(\mu_\gamma^{(l)}, s_\gamma^{(l)})$$

$$\dot{D}_\beta \sim N(\mu_\beta^{(kl)}, s_\beta^{(kl)})$$

$$\dot{D}_{\text{int}} \sim N(m_{\text{int}}^{(ijkl)}, s_{\text{int}}^{(ijkl)})$$

$$\dot{D}_{\text{cosmic}} \sim N(\mu_{\text{cosmic}}^{(l)}, s_{\text{cosmic}}^{(l)})$$

where i indexes over subsamples of sample j , from group k of samples with common sediment beta-doserate $\beta^{(kl)}$ and from group l of samples with common gamma dose-rate and with beta attenuation factor, $b^{(ijkl)}$, for each subsample. Depending on the site, the cosmic radiation dose may be common to all samples or particular samples. The equation as written assumes that it is common to the same groups as gamma dose. For any source of radiation, Z , μ_Z is the true underlying value associated with the observed rate \dot{D}_Z , and s_Z is its measured standard deviation. Following the methods used for radiocarbon dating it is assumed that each s_X is known, and the minor element of uncertainty in these values is ignored. The uncertainties are all assumed to be normally distributed.

This set of assumptions and relationships follows a simplified form of those normally used for luminescence dating, with the addition of recognising the hierarchically correlated uncertainties. As always, the results of the analysis cannot be better than its assumptions. Using these methods it is possible to reanalyse published data only where sufficient detail of the dose-rate components is given. For example, the sequences analysed by Rhodes et al (2003) would make a good test case for the new methodology, but they did not publish the breakdown of the dose-rate components.

CASE STUDY – TL DATING OF BURNT FLINTS AT ST CÉSAIRE

The site of the collapsed rock-shelter at Saint-Césaire, in the valley of a tributary of the Charente in western France, has yielded a stratigraphic sequence which covers the Middle to Upper Palaeolithic transition including Mousterian, Châtelperronian and Aurignacian stone-tool industries. Within one of the Châtelperronian levels was a well preserved Neanderthal skeleton (Lévêque et al 1993). The site is thus important for its lithic sequence and the fact that anatomically diagnostic human remains are associated with the “transitional” Châtelperronian industry.

The Châtelperronian is argued by some to represent acculturation of Neanderthals by incoming modern humans with Aurignacian technology (Mellars 2004), whilst others argue for its development by Neanderthals before contact with modern humans (d'Errico et al 2003). A key point of contention is whether the Châtelperronian clearly overlaps in time with the Aurignacian in the same region (Mellars 2004), or has clearly finished before the Aurignacian starts (d'Errico et al 2003). Refining the chronology of sites like St Césaire will help resolve these issues. The placing of the Neanderthal remains at 36 ± 3 ka (Mercier et al 1991) makes this one of the later dated specimens, and therefore important in understanding the wider picture of the tempo and mode of Neanderthal extinction.

The stratigraphic sequence at Saint-Césaire comprises a series of units which were clearly separated on the basis of colour and texture (Miskovsky & Lévêque 1993). There is an upper *ensemble jaune* (EJ) and a lower *ensemble gris* (EG) with layers labelled on the basis of colour. The primary dating of the site is by 20 TL dates on burnt flints (Mercier et al 1993, Mercier et al 1991). Figure 2 shows schematically the part of the stratigraphy relating to those dates. The luminescence measurements followed standard procedures. The internal dose rate was calculated using uranium, thorium and potassium contents measured by neutron activation analysis, after assessing the α -efficiency using the method of Valladas and Valladas (1982). The external γ -dose was measured by dosimeters placed in the site and a cosmic-ray dose was calculated following Prescott and Stephan (1982). Only the total environmental dose rate is reported by Mercier et al. (1993, 1991)

The stratigraphic information is transformed into prior information for a Bayesian analysis following the method of Zeidler et al. (1998). Within a layer no temporal ordering of flints is known, so they are all assumed *a priori* to be equally likely to fall anywhere between a start date α_n and an end date α_{n+1} for that stratum (n). There is no evidence for hiatus in deposition, so it is assumed continuous; so that the end date for one stratum is the start date for the next. Transition dates between strata are assumed to be equally likely to take any dates between 0 and 1000ka, as long as they have the correct ordering. Comparative calculations were also made using a prior

probability for dates proportional to the inverse square of age, subject to the ordering specified above.

The mathematical model above is modified in the following ways, and additional assumptions made, to compute the required values from the published data:

- uncertainty in the cosmic ray dose is ignored, as it is not given and Aitken (1985, Appendix B) argues that this uncertainty is negligible compared to the other uncertainties
- the external doses are assumed independently measured (there were 40 dosimeters for 20 flints), except where flints from the same layer are reported to have exactly the same external dose, where the external doses are assumed the same.
- the internal gamma dose is a fraction of the infinite matrix dose for the composition of the flint, depending on the geometry of the flint. These geometric factors were reconstructed from the difference between internal dose and the sum of internal alpha and beta doses reported in Mercier et al. (1991), scaled by the infinite matrix dose calculated from the composition using the tables of Bell (1979)
- uncertainty in the external dose is calculated by subtracting in quadrature the uncertainty in internal dose from the uncertainty in the total annual dose, and the 7% systematic uncertainty in the external dose.
- All dose-rates were then re-calculated using the dose-rate conversion factors of Adamiec & Aitken (1998) and the α -track lengths of Brennan and Lyons (1989). Revised figures are included in Table 1.

The model described above was implemented in WinBUGS 1.4 (Lunn et al 2000, Spiegelhalter et al 2004), after manipulation of the published data to the required form in a spreadsheet. For comparison, dates were calculated following the same methods as Mercier et al. (1993, 1991), using the original dose-rate factors and revised ones. Using the original factors and methods, no differences were observed in comparisons with the original calculations. Revised dose-rates and dates are shown in Table 1. The revised dose-rates increase the ages by 1-3% (0.4-1.3ka). The Bayesian stratigraphic analysis reflects this increase in age but also reduces the uncertainty for most of the dates, leading to higher chronological resolution.

The Neanderthal skeleton, placed at 36.3 ± 2.7 ka (95% CI 30.9-41.7ka) by Mercier et al. (1993, 1991), would be placed at 37.2 ± 2.6 ka (95% CI 32.0-42.4ka) using the revised dose-conversion factors now available and the same approach of considering only the mean of dates from stratum EJob sup (calculated using the methodology of Aitken (1985, Appendix B)). The stratigraphic model used here considers not only the uncertainties in the individual dates, but also the *additional* uncertainty that the skeleton could come from anywhere in the time period of the stratum, and the additional data of the stratigraphic relationships between dates. This now gives a mean for the dates of the stratum of 33.9-41.9 ka, and a date for the Neanderthal remains of 32.8-42.5 ka, at 95% credibility.

A LIKELIHOOD FOR URANIUM-SERIES DATES

Uranium-series dating is based upon the geochemical separation of uranium (U) and its decay products (progeny), deposition of that U in a new mineral and subsequent

build up of the progeny isotopes. An overview and summary is given by Latham (2001). Thorium (Th) is insoluble in water under most natural circumstances, whilst U is soluble in oxidizing conditions, so the separation is often and easily obtained. The essential assumptions of the method are

- no Th present at the time of formation of the mineral,
- no gain or loss of U or Th after formation, except by radioactive decay.

A system satisfying these conditions is called a closed system. Equations for the activity ratios of various isotopes at time t from formation are readily derived (Ivanovich & Harmon 1992 Appendix A):

$$\frac{{}^{230}\text{Th}}{{}^{238}\text{U}} = \left(\frac{{}^{230}\text{Th}_0}{{}^{238}\text{U}_0} \right) e^{-\lambda_{230}t} + \frac{\lambda_{238}}{\lambda_{230} - \lambda_{238}} \left(1 - e^{-\lambda_{230}t} \right) - \frac{\lambda_{238}}{\lambda_{230} - \lambda_{238}} \left(1 - e^{-\lambda_{238}t} \right)$$

where each isotope symbol represents the activity of that isotope (i.e. the rate of radioactive disintegration per unit mass of sample) and λ_n is the decay constant for the isotope of mass $230+n$. Typically, corals and detritus-free speleothems can be dated using this equation. The isotope ratios are determined with uncertainties. Here I follow the common implicit assumption in the literature that they can be approximated as normally distributed errors (Ludwig (2003b, p. 25) gives a rare explicit statement of this approximation). Hence likelihoods suitable for incorporation in chronological models for U-Th dating are

$$r_{04} | \theta \sim \text{Normal} \left(\theta, s_{04} \right)$$

$$r_{48} | \theta \sim \text{Normal} \left(\theta, s_{48} \right)$$

$$\mu_{48} \sim \text{Normal} \left(\mu_{48}, s_{48} \right) + \text{Normal} \left(\mu_{48}, s_{48} \right)$$

where r_{nm} is the observed ratio of isotope $230+n$ to $230+m$, μ_{nm} is the true underlying value and s_{nm} is the observed standard deviation of the ratio. A prior on θ is specified from stratigraphic considerations, and a suitable prior is placed on μ_{48} , e.g. $\mu_{48} \sim \text{Unif}(0,20)$, would be a vague prior encompassing the entire range of values observed in natural systems.

Modern, precise, TIMS (thermal ionization mass spectrometry) measurements lead to a further complication, in that the uncertainty in the decay constants is no longer negligible and must be accounted for. Such uncertainties are, of course, common to all U-series dates in a chronological model, and the independence assumption, which simplifies the calculations in radiocarbon chronological models, cannot be maintained.

There are many attractive sample materials with high uranium contents and clear archaeological relevance that do not conform to the assumption of negligible initial Th. Detrital contamination in calcites is usually tackled using an isochron approach. Several sub-samples are taken, either mechanically or by sequential leaching with acid, and measurements made on them. Given that they are of the same age, but usually contain varying amounts of detritus, they yield differing isotope ratios which can then be used in an appropriate regression to determine the composition when there is zero detritus. Ludwig and Titterington (1994) discuss the various ways in which this can be approached, and maximum-likelihood estimation of the parameters of interest, together with error estimates. These methods can be adapted for Bayesian

stratigraphic analyses (Millard 2003). Alternatively, an initial $^{230}\text{Th}/^{232}\text{Th}$ ratio can be assumed, or measured on a suitable sample.

CASE STUDY –U-SERIES DATING OF SPELEOTHEMS AT LA CHAISE DE VOUTHON

La Chaise-de-Vouthon is a multi-chambered cave in the valley of the River Tardoir, in the Charente region of France. It has yielded a long sequence of stone tools (from the Acheulean to the Aurignacian) and numerous hominid remains, both Neanderthal and pre-Neanderthal. Samples from two of the chambers have been analysed in several chronometric studies. Blackwell et al. (1983) conducted the first full-scale study, dating various speleothems using uranium-series dating, and following up the initial study of Schwarcz and Debénath (1979). The relatively well defined dates from Blackwell et al (1983) for the site have subsequently been used to test ESR dating of tooth enamel (Blackwell et al 1992) and uranium-series dating of bone (Rae et al 1987). Both these later studies show underestimates of the age, which may be attributable to mobilisation of uranium in the deposits upsetting these techniques' assumptions about uranium uptake processes.

As an example of the application of the methods described above I present a re-analysis of uranium-series results for the Borgeois-Delauney chamber of the cave. Uranium-series measurements were obtained on two flowstones, forming Beds 7 and 11 of the stratigraphic sequence, plus stalagmites formed on top of Bed 7 before the deposition of Bed 6. Intercalated between Beds 7 and 11 are Beds 8, 9 and 10. Bed 8 was "very poor", and yielded little in the way of remains. Beds 9 and 10 yielded pollen indicating a cool wet climate, in contrast to the pollen from the flowstones which indicated a warm wet climate. Tools described as transitional Acheulean to Mousterian were found in Beds 9 and 10. Hominid remains were found at the very top of Bed 12, partly encrusted with the calcite of the Bed 11 flowstone.

Blackwell et al (1983) obtained uranium-series dates from multiple, stratigraphically related samples within individual speleothem deposits. The Bayesian analysis needs to respect these orderings and the ordering of the layers. Figure 3 shows the chronological model adopted. Stalagmite subsample LC11-2 of the original dataset has been omitted as it showed signs of recent recrystallisation, and flowstone sample LC13 has been omitted as there are some doubts about its stratigraphic integrity.

Blackwell et al (1983) made a correction for detrital contamination using the measured $^{230}\text{Th}/^{232}\text{Th}$ ratio and assuming that the detritus did not contribute to the extracted uranium, and had an initial $^{230}\text{Th}/^{232}\text{Th}$ ratio of 1.25. The same assumptions are followed here. In Blackwell et al.'s (1983) table of dates, there is one significant anomaly in that the date of sample LC47C-1 is reported as 112 ± 5 ka, but the reported isotope ratios correspond to an age of 143 ± 11 ka, and this date is mentioned in the text. For the purposes of calculations reported here, I have assumed the isotope ratios to be correct and the reported age to be a typographical error, as it duplicates the age of LC47C-3.

Of particular interest are the date of the hominid remains at the top of Bed 12, and the timing and duration of Beds 8-10, as well as the overall chronology. The analysis was conducted using Markov-chain Monte Carlo in WinBUGS (Lunn et al 2000,

Spiegelhalter et al 2004). Comparison was made of uniform priors and inverse-square priors with an upper bound on the layer boundaries of 1Ma. Use of a uniform rather than an inverse square prior made little difference to the results, so only results obtained using inverse-square priors are shown in Figure 4 and Table 2. Bed 10 is found to start at 92-116 ka and Bed 8 to end at 87-110 ka (both at 95% credibility). The overall duration of Beds 8-10 is 0.2-20 ka, implying a deposition rate of approximately 0.07 to 7 mm per year. The date of the hominid remains of Bed 12 is harder to evaluate, as there is no real constraint on their maximum age. A conservatively young estimate is that they are similar in age to the oldest dated sample from Bed 11. On this basis they are older than 151-188 ka.

These results place the hominid remains somewhat earlier than Blackwell et al.'s (1983) estimate of 151 ± 15 ka. They also demonstrate that although the accumulation of layers 8 to 10 must have been rapid, considerable uncertainty remains about their duration. The cool, wet climate found in layers 8-10 must correspond with some or all of oxygen isotope substages 5b-d.

AMINO-ACID RACEMIZATION

Amino-acid racemization (AAR) is a chemical dating technique reliant on the transformation of the L-amino-acids found in all living organisms to their mirror image molecules, D-amino-acids. After death the biochemical maintenance systems are no longer in place and L-amino-acids are gradually transformed to D-amino-acids until an equilibrium level is reached. AAR dating has been beset by problems caused by various processes which alter the D/L ratio, for example by leaching of amino-acids, and these have led to its general abandonment, unless a closed system can be demonstrated. Of the materials which maintain a closed system, ostrich egg-shell is the most commonly dated. For more details of the method see Aitken (1990) or Johnson & Miller (1997).

Amino-acid racemisation in ostrich eggshell has been used in the dating of a number of important archaeological and palaeoanthropological sites, e.g., Border Cave, Apollo 11 Cave (Miller et al 1999), Equus Cave (Johnson et al 1997), Bir Tarfawi (Miller 1993, Miller et al 1991), and Semliki (Brooks et al 1995).

On the basis of the published descriptions of the techniques it is in principle possible to develop a statistical model of the likelihood for AAR dating suitable for chronometric analyses similar to those described above for TL and uranium-series dating (Millard 2003). This would be useful in improving chronologies at the sites mentioned and in integrating AAR dates with other chronometric methods. Such an analysis would rely on having the basic chronometric data, in the form of D/L ratios and other measurements for stratigraphically related individual samples. However, this is where a major obstacle is encountered, as AAR data are rarely published for individual samples, but as averaged D/L ratios for layers. Moreover, as variation within a layer is often greater than the analytical uncertainties, the latter are not reported. I have found only two published AAR chronologies with individual sample measurements reported. At Equus Cave (Johnson et al 1997) the deposits appear to be mixed, and a stratigraphic analysis is pointless. At Bir Tarfawi, there are significant periods between most of the dated layers, (Miller 1993, Miller et al 1991) so that an analysis incorporating stratigraphy will produce little improvement in chronological

resolution. If full data on AAR dates were published, as is routine for luminescence, ESR and uranium-series dating, then the data would be amenable to analysis, and incorporation in more comprehensive attempts at chronology building.

CONCLUSIONS

This paper has shown that it is possible to develop a mathematical apparatus suitable for the incorporation of luminescence, uranium-series and other dating techniques into the Bayesian statistical framework which is currently available primarily for radiocarbon dates. This allows coherent analysis of stratigraphic and chronometric evidence, but reanalysis of published data relies on full publication of the parameters which go into calculation of a date. As expected, the incorporation of the additional data about stratigraphic relationships allows us to arrive at more precise estimates of the dates of events, at times even though additional stratigraphic uncertainty is allowed for as well. Although bespoke computer code is currently needed for each site, it should now be possible to start to integrate multiple techniques applied to a single site to obtain more coherent chronologies from Pleistocene archaeological and palaeoanthropological sites.

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NOTE:

The WinBUGS code use for the examples and instructions for using it are available from the author's website <http://www.dur.ac.uk/a.r.millard/>

REFERENCES

- Adamiec G, Aitken M. 1998. Dose-rate conversion factors: update. *Ancient TL* 16: 37-50
- Aitken MJ. 1985. *Thermoluminescence dating*. London: Academic Press
- Aitken MJ. 1990. *Science-based dating in archaeology*: Longman
- Bayliss A, Bronk Ramsey C. 2003. Pragmatic Bayesians: a decade of integrating radiocarbon dates into chronological models. In *Tools for constructing chronologies: crossing disciplinary boundaries*, ed. CE Buck, AR Millard, pp. 25-41. London: Springer-Verlag
- Bell WT. 1979. Thermoluminescence dating: radiation dose-rate data. *Archaeometry* 21: 243-5
- Blackwell B, Porat N, Schwarcz HP, Debénath A. 1992. ESR dating of tooth enamel: comparison with ²³⁰Th/²³⁴U speleothem dates at La Chaise-de-Vouthon (Charente), France. *Quaternary Science Reviews* 11: 231-44
- Blackwell B, Schwarcz HP, Debénath A. 1983. Absolute dating of hominids and Palaeolithic artifacts of La Chaise-de-Vouthon (Charente), France. *Journal of Archaeological Science* 10: 493-513
- Brennan BJ, Lyons RG. 1989. Ranges of alpha particles in various media. *Ancient TL* 7: 32-7

- Bronk Ramsey C. 1995. Radiocarbon Calibration and Analysis of Stratigraphy: The OxCal Program. *Radiocarbon* 37: 425-30
- Bronk Ramsey C. 1998. Probability and dating. *Radiocarbon* 40: 461-74
- Brooks AS, Helgren DM, Cramer JS, Franklin A, Hornyak W, et al. 1995. Dating and Context of Three Middle Stone Age Sites with Bone Points in the Upper Semliki Valley, Zaire. *Science* 268: 548-53
- Buck CE. 2003. Bayesian chronological data interpretation: where now? In *Tools for constructing chronologies: crossing disciplinary boundaries*, ed. CE Buck, AR Millard, pp. 1-24. London: Springer-Verlag
- Buck CE, Kenworthy JB, Litton CD, Smith AFM. 1991. Combining archaeological and radiocarbon information: a Bayesian approach to calibration. *Antiquity* 65: 808-21
- Buck CE, Litton CD, Scott EM. 1994a. Making the most of radiocarbon dating: some statistical considerations. *Antiquity* 68: 252-63
- Buck CE, Litton CD, Shennan SJ. 1994b. A case study in combining radiocarbon and archaeological information: the early Bronze Age settlement of St. Veit-Klingberg, Land Salzburg, Austria. *Germania*: 427-47
- Buck CE, Litton CD, Smith AFM. 1992. Calibration of radiocarbon results pertaining to related archaeological events. *Journal of Archaeological Science* 19: 497-512
- d'Errico F, Henshilwood C, Lawson G, Vanhaeren M, Tillier AM, et al. 2003. Archaeological evidence for the emergence of language, symbolism, and music - An alternative multidisciplinary perspective. *Journal of World Prehistory* 17: 1-70
- Isaac GL. 1975. Sorting out the muddle in the middle: an anthropologist's post-conference appraisal. In *After the Australopithecines*, ed. KW Butzer, GL Isaac. The Hague: Mouton
- Ivanovich M, Harmon RS. 1992. *Uranium-series disequilibrium : applications to earth, marine, and environmental sciences*. Oxford: Clarendon. xxxiv, 910 pp.
- Johnson BJ, Fogel ML, Miller GH, Beaumont PB. 1997. The determination of late Quaternary paleoenvironments at Equus Cave, South Africa, using stable isotopes and amino acid racemization in ostrich eggshell. *Palaeogeography, Palaeoclimatology, and Palaeoecology* 136: 121-37
- Johnson BJ, Miller GH. 1997. Archaeological application of amino acid racemization. *Archaeometry* 39: 265-88
- Joyce M. 1999. How fast do things disappear? *Genealogists' Magazine* 26: 173-4
- Lanos P. 2003. Bayesian estimation of archaeomagnetic reference curves: applications to dating and chronological interpretation. In *Tools for Constructing Chronologies: Crossing Disciplinary Boundaries*, ed. CE Buck, AR Millard. London: Springer Verlag
- Latham AG. 2001. Uranium-series dating. In *Handbook of Archaeological Sciences*, ed. DR Brothwell, AM Pollard, pp. 63-72. Chichester: Wiley
- Lévêque F, Backer AN, Guilband M. 1993. *Context of a Late Neandertal: Implications of multidisciplinary research for the transition to Upper Palaeolithic adaptations at Saint-Césaire, Charante-Maritime, France*. Madison, Wisconsin: Prehistory Press
- Ludwig K. 2003a. Mathematical-statistical treatment of data and errors for $^{230}\text{Th}/\text{U}$ geochronology. *Reviews in Mineralogy and Geochemistry* 52: 631-66
- Ludwig K. 2003b. *User's Manual for Isoplot 3.00: A geochronological toolkit for Microsoft Excel*. Berkeley: Berkeley Geochronology Center

- Ludwig KR, Titterington DM. 1994. Calculation of (230)Th/U Isochrons, Ages, and Errors. *Geochimica Et Cosmochimica Acta* 58: 5031-42
- Lunn DJ, Thomas A, Best N, Spiegelhalter D. 2000. WinBUGS - A Bayesian modelling framework: Concepts, structure, and extensibility. *Statistics and Computing* 10: 325-37
- Mellars P. 2004. Neanderthals and the modern human colonization of Europe. *Nature* 432: 461-5
- Mercier N, Valladas H, Joron J-L, Reyss J-L. 1993. Thermoluminescence dating of the prehistoric site of La Roche à Pierrot, Saint-Césaire. In *Context of a Late Neandertal: Implications of multidisciplinary research for the transition to Upper Palaeolithic adaptations at Saint-Cesaire, Charante-Maritime, France*, ed. F Lévêque, AM Backer, M Guilbaud, pp. 15-21. Wisconsin, Madison: Prehistory Press
- Mercier NM, Valladas H, Joron JL, Reeyss JL, Lévêque F, Vandermeersch B. 1991. Thermoluminescence dating of the late Neanderthal remains from Saint Césaire. *Nature* 351: 737-9
- Millard AR. 2003. Taking Bayes Beyond Radiocarbon: Bayesian Approaches to Some Other Chronometric Methods. In *Tools for Constructing Chronologies: Crossing Disciplinary Boundaries*, ed. CE Buck, AR Millard, pp. 231-48. London: Springer Verlag
- Millard AR. submitted. Bayesian analysis of ESR dates. *Quaternary Science Reviews*
- Miller GH. 1993. Chronology of hominid occupation at Bir Tarfawi and Bir Sahara East, based on the epimerization of isoleucine in ostrich eggshells. In *Egypt during the last interglacial: The Middle Paleolithic of Bir Tarfawi and Bir Sahara East*, ed. F Wendorf, R Schild, AE Close, associates, pp. 241-51. New York: Plenum
- Miller GH, Beaumont PB, Deacon HJ, Brooks AS, Hare PE, Jull AJT. 1999. Earliest modern humans in southern Africa dated by isoleucine epimerization in ostrich eggshell. *Quaternary Science Reviews* 18: 1537-48
- Miller GH, Wendorf F, Ernst R, Schild R, A.E. C, et al. 1991. Dating lacustrine episodes in the Eastern Sahara by the epimerization of isoleucine in ostrich eggshells. *Palaeogeography, Palaeoecology and Palaeoclimatology* 84: 175-89
- Miskovsky J-C, Lévêque F. 1993. The sediments and stratigraphy of Saint-Césaire: contributions to the paleoclimatology of the site. In *Context of a Late Neandertal: Implications of multidisciplinary research for the transition to Upper Palaeolithic adaptations at Saint-Cesaire, Charante-Maritime, France*, ed. M Guilbaud, pp. 7-14. Wisconsin, Madison: Prehistory Press
- Nicholls G, Jones M. 2001. Radiocarbon dating with temporal order constraints. *Journal of the Royal Statistical Society Series C-Applied Statistics* 50: 503-21
- Prescott JR, Stephan LG. 1982. The contribution of cosmic radiation to the environmental dose for thermoluminescent dating: latitude, altitude and depth dependences. *PACT* 6: 17-25
- Rae AM, Ivanovich M, Schwarcz HP. 1987. Absolute dating by U-series disequilibrium of bones from the cave of La Chaise-de-Vouthon (Charente), France. *Earth Surface Processes and Landforms* 12: 543-50
- Rhodes EJ, Bronk Ramsey C, Outram Z, Batt C, Willis L, et al. 2003. Bayesian method applied to the interpretation of multiple OSL dates: high precision sediment ages from Old Scatness Broch excavations, Shetland Isles. *Quaternary Science Reviews* 22: 1231-44

- Roberts RG. 1997. Luminescence dating in archaeology: from origins to optical. *Radiation Measurements* 27: 819-92
- Schwarcz HP, Debénath A. 1979. Datation absolue des restes humains de la Chaise-de-Vouthon (Charente) au moyen du déséquilibre des séries d'Uranium. *Comptes rendus de l'Academie des Sciences de Paris* 288: 1155-7
- Spiegelhalter D, Thomas A, Best N, Lunn D. 2004. *WinBUGS Version 1.4 User Manual* (<http://www.mrc-bsu.cam.ac.uk/bugs>). Cambridge: MRC Biostatistics Unit
- Steier P, Rom W. 2000. The use of Bayesian statistics for C-14 dates of chronologically ordered samples: A critical analysis. *Radiocarbon* 42: 183-98
- Valladas H, Valladas G. 1982. Effet de l'irradiation α sur des grains de quartz. *PACT* 6: 171-8
- Zeidler JA, Buck CE, Litton CD. 1998. Integration of archaeological phase information and radiocarbon results from the Jama River Valley, Ecuador: A Bayesian approach. *Latin American Antiquity* 9: 160-79

Figure 1: The hierarchy of parameters in common between different dating samples by luminescence. Each inner box is repeated within the box surrounding it, with different values of the parameters for different samples.

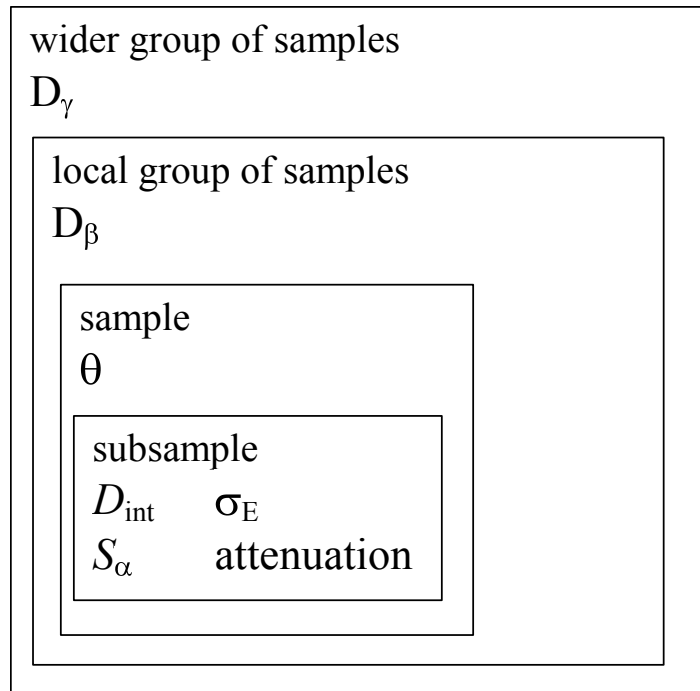


Figure 2: Dating results for Saint Cesaire. Thick grey bars represent two standard deviation results as reported by Mercier et al. (1991), thin black lines represent 95% posterior credible intervals for those dates using the model described in the text, thick black lines represent 95% posterior credible intervals of the boundaries between layers. Layer names and cultural associations are shown.

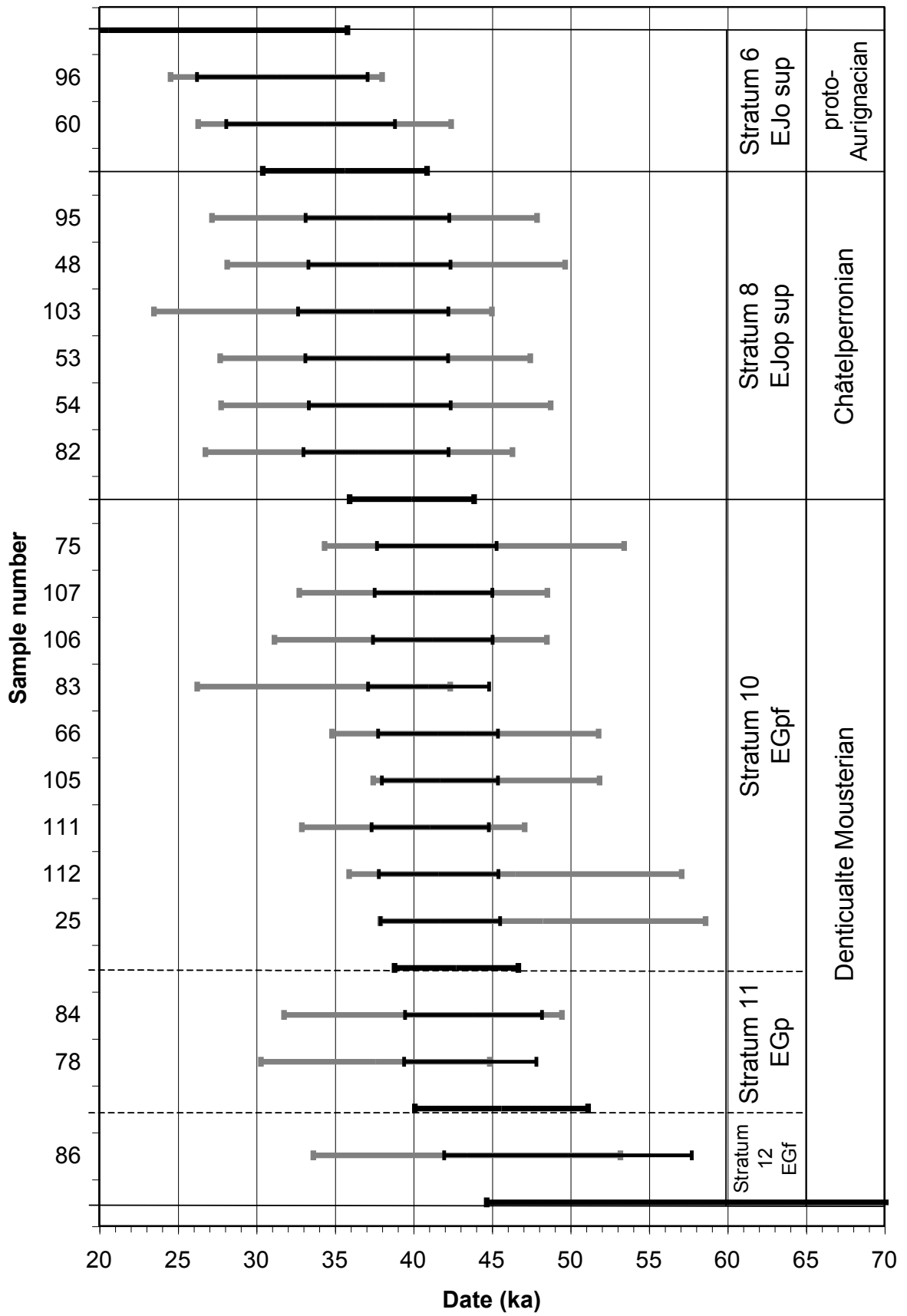


Figure 3: Harris matrix showing Stratigraphic model for Bourgeois-Delauney

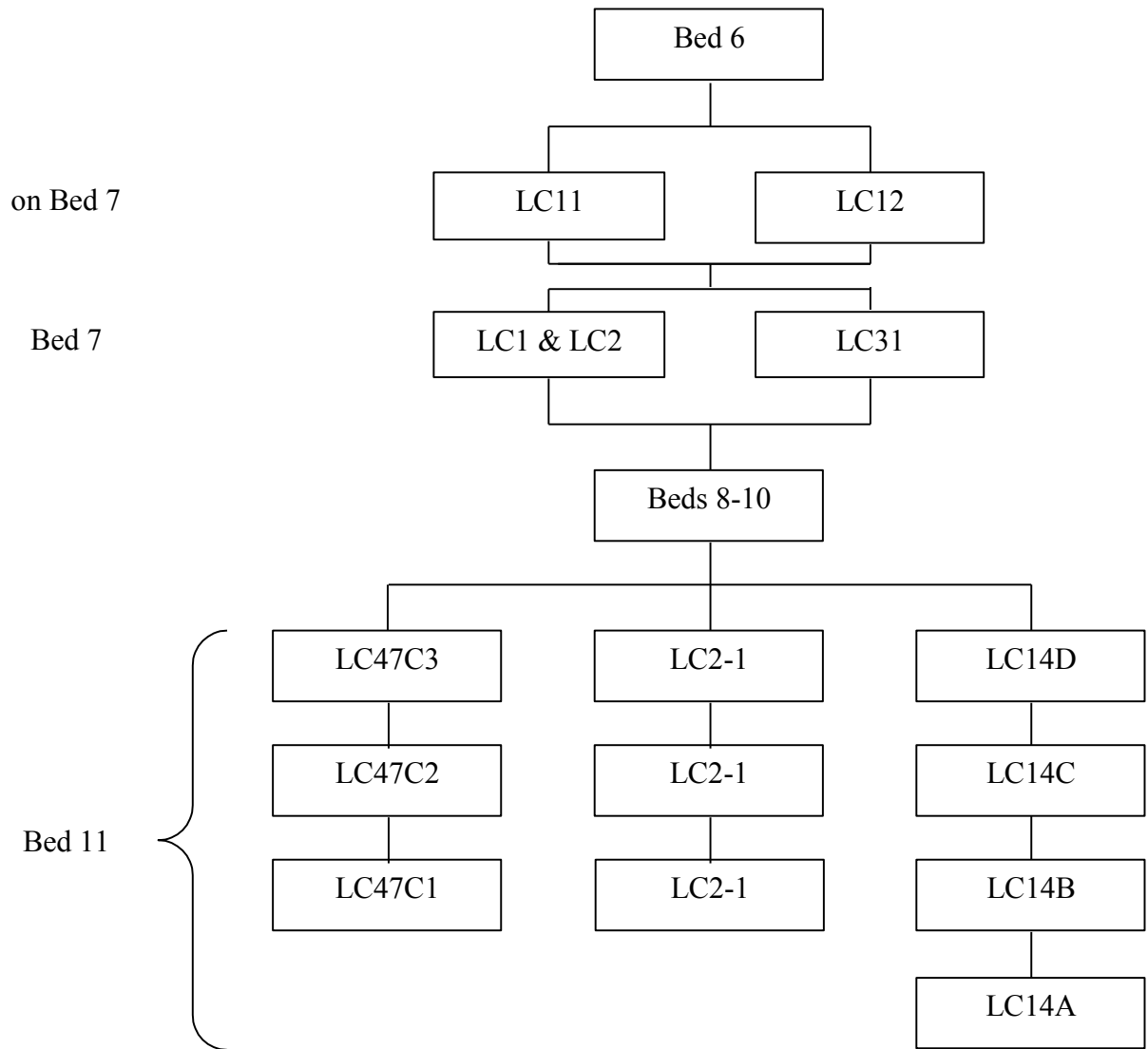


Figure 4: Dating results for Bourgeois Delauney. Thick grey bars represent two standard deviation results as reported by Blackwell et al. (1983), thin black lines represent 95% posterior credible intervals for those dates using the model described in the text, thick black lines represent 95% posterior credible intervals of the boundaries between layers.

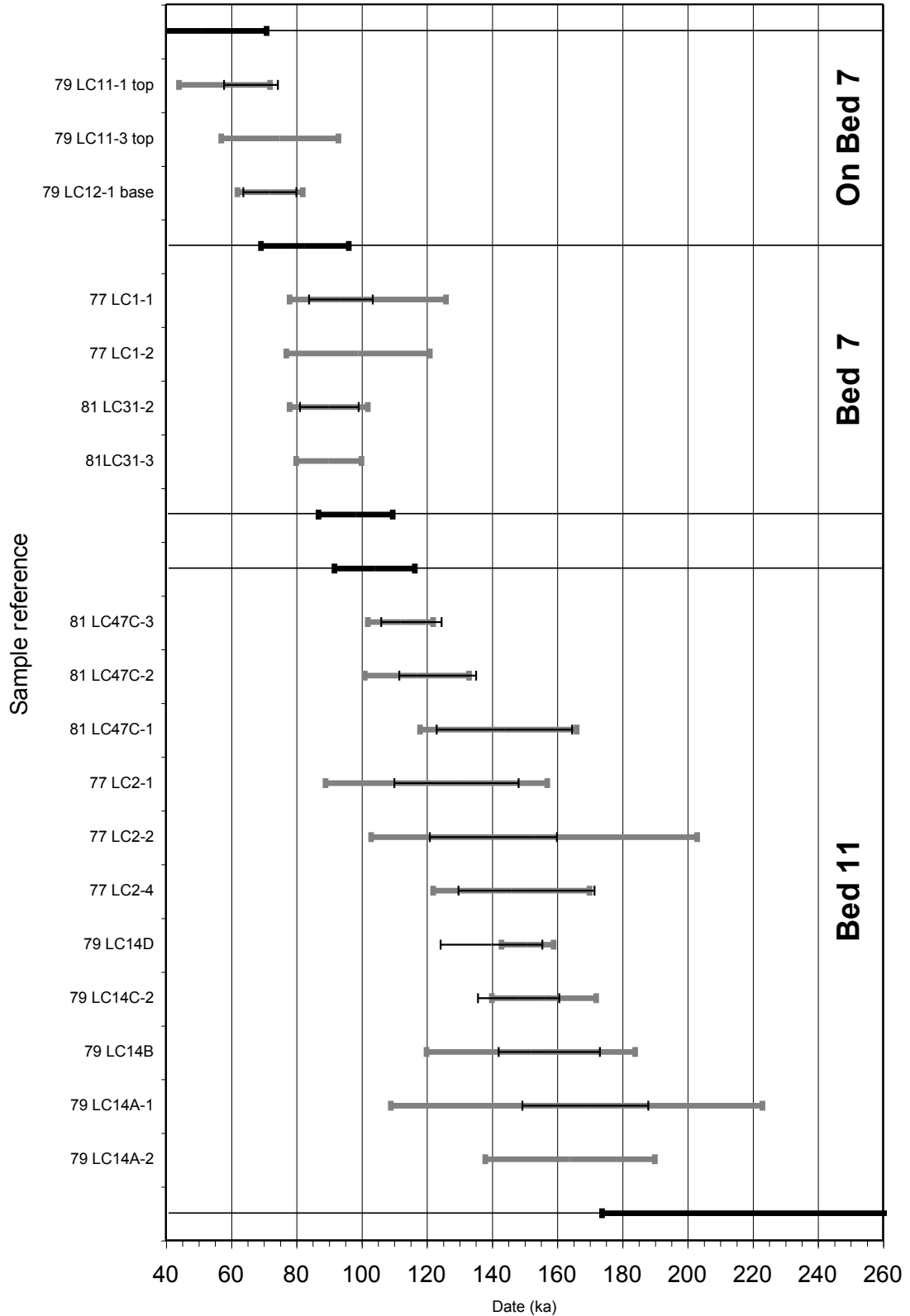


Table 1 Doserates and dates for St Cesaire

Lab. no.	Level	U†	Th†	K†	Sα‡	Dα	Dβ	Internal dose	External dose§	Annual dose	Paleodose	Age	Age (95% hpd)
		(ppm)	(ppm)	(ppm)	(μGy/(α cm ⁻²))	(μGy a ⁻¹)	(μGy a ⁻¹)	(μGy a ⁻¹)	(μGy a ⁻¹)	(μGy a ⁻¹)	(Gy)	(ka)	(ka)
96	6	0.53	0.491	666	1.99	240±7	143	388±27	610	998±104	31.23±1.20	31.3±3.4	26.2–37.1
60	6	0.541	0.237	430	1.45	159±9	119	283±22	610	893±102	30.70±1.03	34.4±4.0	28.1–38.8
95	8	0.687	0.466	1599	1.64	243±31	238	488±44	375	863±119	32.39±1.73	37.5±5.2	33.2–42.3
48	8	0.836	0.197	178	1.29	208±12	141	359±29	395	754±104	29.35±1.04	38.9±5.4	33.3–42.4
103	8	0.485	0.106	378	1.84	171±6	103	279±21	407	686±102	23.48±1.33	34.2±5.4	32.7–42.2
53	8	0.614	0.029	240	2.71	305±15	109	418±35	402	820±106	30.82±1.38	37.6±4.9	33.1–42.2
54	8	0.51	0.069	430	2.48	237±12	110	352±28	402	754±104	28.86±0.93	38.3±5.2	33.4–42.4
82	8	0.719	0.397	484	1.85	278±18	154	439±35	418	857±115	31.30±1.08	36.5±4.9	33.0–42.3
75	10	0.529	0.188	274	1.49	157±10	104	266±21	351	617±64	27.06±1.26	43.9±4.8	37.7–45.3
107	10	0.946	0.26	732	1.77	326±17	202	539±42	352	891±81	36.20±1.95	40.6±3.9	37.5–45.0
106	10	0.4	0.415	1206	1.69	158±3	164	326±21	359	685±63	27.29±1.75	39.8±4.3	37.4–45.1
83	10	0.616	0.27	292	1.74	218±7	120	343±25	281	624±65	21.40±1.38	34.3±4.0	37.1–44.8
66	10	0.461	0.12	440	1.79	160±11	105	270±22	315	585±55	25.35±1.18	43.3±4.2	37.8–45.4
105	10	1.051	0.212	864	1.48	297±14	227	531±40	318	849±72	37.92±1.25	44.7±3.6	38.0–45.4
111	10	0.652	0.289	780	1.93	256±18	164	426±34	315	741±69	29.64±1.02	40.0±3.5	37.3–44.8
112	10	0.603	0.342	842	1.3	164±13	163	335±26	336	671±75	31.19±1.39	46.5±5.3	37.8–45.4
25	10	0.472	0.389	610	1.84	193±10	127	329±24	310	639±65	30.82±1.56	48.3±5.2	37.9–45.5
84	11	0.746	0.384	472	1.14	176±12	156	339±26	257	596±66	24.22±0.91	40.6±4.4	39.5–48.2
78	11	0.452	0.337	364	1.67	165±8	104	274±20	284	558±54	20.96±0.74	37.6±3.6	39.4–47.8
86	12	0.629	0.272	320	1.49	190±12	124	326±26	251	577±65	25.06±1.00	43.4±4.9	42.0–57.7

Table 2 Isotope ratios and dates for La Chaise

	Sample	$^{230}\text{Th}/^{234}\text{U}$	$^{234}\text{U}/^{238}\text{U}$	$^{230}\text{Th}/^{232}\text{Th}$	Original age (ka)	95% posterior interval (ka)
Bed 7	77 LC1-1	0.646±0.032	1.018±0.038	9.6±2.2	102±12	84–104
	77 LC1-2	0.603±0.039	1.054±0.035	>1000	99±11	
	81 LC31-2	0.594±0.220	1.056±0.028	74.3±17.9	90±6	81–99
	81LC31-3	0.57±0.022	1.077±0.026	58.1±16.8	90±5	
on Bed 7	79 LC11-1 top	0.483±0.026	1.108±0.030	5.6±0.9	58±7	58–74
	79 LC11-3 top	0.526±0.031	1.117±0.057	15.6±4.9	75±9	
	79 LC12-1 base	0.511±0.021	1.076±0.033	15.1±2.7	72±5	64–80
Bed 11	81 LC47C-1	0.738±0.029	1.042±0.026	81.4±23.9	112*±5	123–165
	81 LC47C-2	0.666±0.027	1.073±0.030	>1000	117±8	112–135
	81 LC47C-3	0.647±0.018	1.050±0.019	>1000	112±5	106–125
	77 LC2-1	0.703±0.039	1.029±0.041	15.4±7.6	123±17	110–148
	77 LC2-2	0.775±0.048	1.056±0.048	20.4±14.2	153±25	121–160
	77 LC2-4	0.746±0.030	1.100±0.040	29.5±11.3	146±12	130–172
	79 LC14A-1	0.802±0.053	1.136±0.053	9.7±0.7	159 ₋	149–188
	79 LC14A-2	0.787±0.027	1.041±0.032	52.8±11.4	164±13	
	79 LC14B	0.772±0.035	1.032±0.045	20.1±2.8	152±16	142–173
	79 LC14C-1	0.631±0.023	1.037±0.029	30.0±8.0	107±6	136–161
	79 LC14C-2	0.748±0.019	1.054±0.022	>1000	156±8	
	79 LC14D	0.759±0.034	1.045±0.047	28.6±6.8	151±4	124–156