ELECTRON SPIN RESONANCE DATING AND THE EVOLUTION OF MODERN HUMANS

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INTRODUCTION

Remarkable changes have been seen recently in our perceptions about the origin and early evolution of anatomically modern Homo sapiens (hereafter Homo sapiens or modern humans). Some of this change is the result of new approaches to the study of the palaeontological record, some to fossil discoveries, some to new genetic research on modern samples, and some to the development and application of dating techniques. These can for the first time cover the critical time zone of the later middle and early late Pleistocene where radiocarbon dating is inapplicable or of dubious value. This review examines the role of electron spin resonance (ESR) dating of tooth enamel, which, together with the related technique of thermoluminescence (TL) dating of burnt flint, has made the greatest single impact on chronologies. These techniques have demonstrated that the extensive early modern skeletal sample from Qafzeh Cave (Israel) is apparently, at about 90 to 120 ka, more than twice as old as many workers expected. The clear implication of these determinations is that Neanderthals and early modern humans were indeed separate and parallel evolutionary lineages, rather than ancestors and descendants. The repercussions of these results are still being felt. However, the age estimates are not yet universally accepted, with intimations that the dates cannot be verified independently and might be inaccurate (Trinkaus, in Bower 1988), are 'still experimental' (Trinkaus 1991) or might 'not stand the test of time' (Wolpoff 1989a, 104).

In this review of ESR dating and recent human evolution, we introduce the ESR technique and discuss its particular application to dating tooth enamel. We then review results obtained so far for fossil hominid sites relevant to the origin of modern humans, discussing the regions of Europe, the Middle East and Africa in turn. In conclusion we discuss the significance of the results so far and compare them with previous age estimates (see Figure 29). We also examine remaining problems, and possible future developments, including the further application of ESR dating to the chronology of recent human evolution.

First, however, it is necessary to provide a brief review of current ideas about the origins of Homo sapiens, in order that existing results from ESR dating can be put into context. Africa is now generally regarded as the original homeland of the hominids (the family containing
humans and our closest relatives), and that origin is believed to have taken place over 5 million years ago. However, it is still not certain whether Africa was also the homeland of our own species Homo sapiens. Humans of the earlier and much more robust species Homo erectus were widely distributed in Asia 700000 years ago, and according to different taxonomic views were also present as that species, or a closely related one, in Africa and Europe. Hence, potentially one, two or three of the inhabited continents could have been areas where our species began. Adherents of the multi-regional model of modern human origins argue that all three continents were involved, and propose Europe as the ancient homeland of the ‘Caucasoid’ peoples, and Asia as the homeland of at least two main groups of present-day humanity – the ‘Mongoloids’ (east Asians, Inuit and American Indians) and the ‘Australoids’ (Australian aborigines). From this model, there should have been evolutionary continuity in each inhabited continent, as semi-separate racial lines gradually developed into modern humans. Most ‘racial’ features would be of middle or even early Pleistocene antiquity, and the first appearance of modern humans should not have followed any particular geographic pattern, with rather indistinct taxonomic boundaries in the fossils caused by gradual evolutionary change and widespread gene flow (Wolpoff et al. 1984; Wolpoff 1989a and b; Smith et al. 1989).

Those who favour the ‘Out of Africa’ model argue that Africa was the sole location of modern human origins, and that all living people of whatever type or ‘race’ derive from an African source by evolution and dispersal over the last 100000 to 200000 years. The Neanderthal lineage in Europe and the Asian evolutionary lines of ‘Peking Man’ and ‘Java Man’ became extinct without giving rise to any living populations. ‘Racial’ features would only have developed in the late Pleistocene, after the African origin of Homo sapiens, as populations dispersed to their more recent homelands across the inhabited world. It would also be expected that there was a phased pattern of first appearances for the species:

(i) Africa – place of origin;
(ii) western Asia – secondary dispersal centre closest to Africa; and
(iii) finally, in this context, the peripheral areas: Europe, eastern Asia, Australasia and the Americas.

There could be coexistence of anatomically modern and non-modern hominin populations in areas where their geographic ranges overlapped, or during periods where modern humans were replacing archaic forms in the same areas (Stringer and Andrews 1988; Stringer 1989 and 1991; see also Bräuer 1991).

The importance of accurate dating to the resolution of the present dispute about the tempo and mode of origin of Homo sapiens is clear. Without an accurate chronological background, it is impossible to determine the location of the oldest Homo sapiens fossils, to document the geographic spread of the species, and to examine local lineages for signs of supposed evolutionary trends, the coexistence of distinct types of hominin, or actual replacement of one population by another (Smith and Spencer 1984; Stringer and Andrews 1988; Smith et al. 1989).

ESR DATING

ESR dating was introduced to earth sciences when Ikeya (1975) dated a speleothem from Akiyoshi Cave, Japan. Since then, ESR dating has been applied to many materials in
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geology, geography and archaeology. It permits dates on minerals which are precipitated, such as secondary carbonates (speleothems, mollusc shells, corals) or tooth enamel, as well as materials which were heated in the past such as volcanic minerals. Recent applications have tried to utilize the resetting of ESR signals during fault movement in order to establish the chronology of neotectonic processes. A summary of applications and a description of the dating procedures have been given by Grün (1989a and b). In the archaeological context, ESR dating of tooth enamel is of particular interest and this application will be described. Initial results on ESR dating of burnt flint seem promising (Porat and Schwarzc 1991), but this application is not yet far enough developed to be regarded as a dating technique.

Many important archaeological sites are not datable because they are beyond the radiocarbon dating range and lack material which is suitable for other dating techniques. Bones and teeth occur in most archaeological sites and are commonly coeval with other remains. As it turned out, bones are probably not datable with ESR (Grün and Schwarzc 1987; see also below), whereas ESR dating of tooth enamel can provide valuable chronological information for many sites.

Basic principles of ESR dating

Tooth enamel consists of more than 96% of the mineral hydroxyapatite (Driessens 1980), which is in contact with the more organically rich dentin and cement (see Figure 1 (A–C)). In ESR dating, the tooth is used as a dosimeter which records the natural radioactivity of the sample itself and its environment from the time the tooth was buried. An insulating mineral, such as hydroxyapatite, has two energy states at which electrons may occur: the valence band (ground state) and the conduction band (excited state; see Figure 2). When a tooth is formed, all electrons are in the ground state. Due to natural radioactive radiation, electrons are transferred to the conduction band. After a short time of diffusion, these electrons recombine with holes near the valence band. However, all natural minerals have impurities such as lattice defects or substituting atoms (traps) which can capture these electrons at various intermediate energy levels. These trapped electrons form paramagnetic centres and give rise to characteristic ESR signals (see Figure 3). The number of trapped electrons and, hence, the height of the ESR signal is directly proportional (i) to the number of traps in a mineral (and their cross-section for trapping = sensitivity), and (ii) to the strength of the radioactivity (dose rate), and (iii) to the time of irradiation (=age). Figure 2 also shows the trapping scheme of thermoluminescence on the right (as this technique is used to date burnt flint). The
Figure 2. Trapping scheme of electrons in insulating minerals. (left) Ionizing radiation transfers electrons to the conduction band. After a short time of diffusion most of the electrons recombine with holes near the valence band. Some electrons are trapped by impurities in the crystal lattice. These trapped electrons can be directly detected by electron spin resonance (ESR) (see Figure 3). $E_a =$ activation energy. (right) Subsequent heating releases the electrons and light emission can be observed (thermoluminescence).

sample is heated in the laboratory and at a temperature which corresponds to the activation energy, $E_a =$ trap depth, the trapped electrons are transferred back to the conduction band. Some holes near the conduction band are luminescence centres and when electrons recombine with these centres, light emission can be observed. The schemes shown in Figure 2 illustrate that in principle ESR gives a more direct view into the crystal and, additionally, measurements can be repeated on the same sample.

An ESR age is derived from the following relation:

$$\text{age} = \frac{\text{accumulated dose} (AD)}{\text{dose rate} (D)}$$

The accumulated dose, $AD$, is the radioactive dose the tooth has received since it was buried. This value is determined by the additive dose method (see Figure 4 or Aitken 1990). The dose rate, $D$, is derived from the chemical analysis of the radioactive elements (U, Th, and K; other elements are usually negligible) in the sample and its surroundings; direct on-site measurements may be employed in respect of the radioactivity of the latter. The isotopic concentrations are converted into dose rates by published tables (e.g. Nambi and Aitken 1986). The determination of the radioactivity that influences the sample is rather complex and has to be carefully evaluated.

As we can see from the equation above, two parameters have to be determined in order to estimate an ESR age, the accumulated dose (AD) and the dose rate (D).

**Determination of the accumulated dose (AD)**

The AD is the parameter which is actually determined by ESR spectrometry. Figure 5 shows
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Figure 3  (top) ESR signal of a relatively young enamel sample from Le Moustier and (bottom) ESR signal of an old enamel sample from Sterkfontein.

Figure 4  Determination of accumulated dose (AD) by the additive dose technique. Artificial gamma irradiation enhances the ESR intensity. The AD is obtained from the extrapolation of the data points. (left) Data points of sample 503a with linear fitting; (right) as above with exponential fitting. The SSQ (sum of square of the vertical distances of the data points from the calculated line) for the exponential fitting procedure is an order of magnitude smaller than for linear fitting. \( I_{\text{max}} = \) maximum intensity.
the block-diagram of an ESR spectrometer. The sample is brought into an external magnetic field and exposed to microwaves. The microwaves can change the energy level of the magnetic moment of a paramagnetic centre relative to the external magnetic field. Such atomic processes happen only at discrete energies, and for a given microwave frequency there are specific values of magnetic field strength at which these changes occur (resonance). In resonance position, microwave absorption is observed. In order to become independent of specific equipment configurations, the position of an ESR line in a spectrum is described by the g-value, which is proportional to the ratio of microwave frequency to magnetic field strength (see X-axis in Figure 3).

Figure 6 shows two ESR spectra of tooth enamel. The radiation sensitive signals at \( g = 2.0018 \) and the signal at \( g = 1.9976 \) are certainly due to hydroxyapatite (Houben 1971; Ostrowski et al. 1971 and 1974). In many cases the ESR signal is interfered with by organic radicals (Figure 6, lower spectrum). The marked quintet was attributed to alanine (Ikeya 1982). Newer investigations imply that this quintet is actually a septet with two very small lines further to the outside and this was related to a free dimethyl radical (Bouchez et al. 1988). Grün et al. (1987) showed that these organic interferences can be suppressed during the measurements by overmodulation (using a modulation amplitude of 0.5 mTpp).

An ESR signal used for dating should have the following properties.

(i) A zeroing effect deletes all previously stored ESR intensity in the sample at the event which is to be dated.

(ii) The signal intensity grows in proportion to the dose received.

(iii) The signals must have a stability which is at least one order of magnitude higher than the age of the sample.

(iv) The number of traps is constant or changes in a predictable manner. Recrystallization, crystal growth or phase transitions must not have occurred.
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Figure 6 ESR spectra of tooth enamel (solid line: natural sample; dotted line: irradiated sample). The lower spectrum shows various interferences by organic signals. The marked quintet has been attributed to alanine or dimethyl radicals.

(v) The ESR signal is not influenced by sample preparation (grinding, exposure to laboratory light) or anomalous fading.

The ESR signal of recent teeth is below the sensitivity of an ESR spectrometer and in practical terms zero (except for an organism which was irradiated by an A-bomb explosion (Hoshi et al. 1985) or lived close to a leaking nuclear power plant).

For the determination of AD, the ESR response to radioactive irradiation must be known and mathematically expressed. For this purpose, aliquots of the sample are irradiated with a gamma source (e.g. $^{60}$Co). Many hospitals and chemistry departments have such gamma sources for various applications (e.g. sterilization or cancer treatment). During radiation an amount of energy is transferred to the sample and the unit for this energy dose is Gray (Gy). The old unit which is still often used is rad (100 rad = 1 Gy).

In the early days of ESR dating, many procedures were borrowed from TL dating, which mainly dealt with the measurement of relatively young archaeological samples such as pottery. Samples were usually irradiated with small doses (< 100 Gy) and the TL dose response was described as linear or within the linear part of the dose response curve. However, ESR dating of tooth enamel involves the determination of ADs in the range of 20 to 4000 Gy, which requires irradiation steps of up to 10 000 Gy. Since the number of traps in a mineral is limited, the probability of electrons being trapped decreases as more traps are already filled with electrons, and linear fitting cannot be applied. The dose response can be expressed by a simple exponential function:

$$I = I_{\text{max}} (1 - \exp(-a(D_{\text{irr}} + D_{AD})))$$

where $I$ = ESR intensity, $I_{\text{max}}$ = maximum ESR intensity when all traps are filled, $a$ = fraction of unfilled defects that trap electrons per dose unit, $D_{\text{irr}}$ = irradiation dose and $D_{AD}$ the dose
that corresponds to AD. Figure 4 shows the dose response of sample 503a from the site of Polledrara (Italy). The linear fit on the left side has a correlation coefficient of 0.9994 and this is often taken as proof for the validity of the fitting procedure. The right diagram shows the exponential extrapolation of the same data set. Although the curve looks more or less linear, the sum of squares of the deviation of the data points from the curve (SSQ) has improved by a factor of more than 10, but more importantly the AD is about 25% smaller than the one obtained by the linear fit.

There are several ways to determine the error in AD determination. In the case of repeated measurements at one dose and linear fitting by a conversion, the reader is referred to Berger and Huntley (1986), where the error is calculated from the scatter of the data points around the fitted function. In many cases this error seems rather small. Grün and Macdonald (1989) used a jackknifing procedure for error estimation in the case of single data points at a dose. This approach was attacked by Lyons and Brennan (1990) who were particularly unhappy that errors become very large when the data set contains an outlier. Another method was recently described by Poljakov and Hütt (1990) which was in turn critically evaluated by Berger (1990).

**Determination of the dose rate (D)**

The strength of the radioactive field which irradiates the sample is determined by the concentration of radioactive elements in the tooth and its surroundings plus a component of cosmic rays. In ESR studies, only the U and Th decay chains and the $^{40}\text{K}$-decay are of relevance (a minor contribution comes from $^{87}\text{Rb}$ in the sediment). There are three ionizing rays which are emitted from the radioactive elements (the ranges are given for a material with a density of about 2.5 g/cm$^3$).

(i) Alpha rays have only a very short range of about 20 μm because of the large size of the particles. Alpha particles are not as efficient in producing ESR intensity as beta and gamma rays, therefore an alpha efficiency has to be determined (see Aitken 1985 and 1990).

(ii) Beta rays (electrons) have a range of about 2 mm.

(iii) Gamma rays have a range of about 30 cm.

For tooth enamel, the following dose rate parameters have to be determined (depending on the configuration of the tooth enamel piece):

*Enamel* — U-concentration (Th and K are usually negligible)
- $^{234}\text{U} : ^{238}\text{U}$ ratio
- alpha efficiency
- thickness, for beta ray attenuation

*Dentin* — U-concentration (Th and K are usually negligible)
- $^{234}\text{U} : ^{238}\text{U}$ ratio
- water content

*Sediment* — U-, Th-, K-concentration and water content in immediate surroundings for beta dose rate
- gamma dose rate

**Cosmic dose rate**

The cosmic dose rate is dependent on the geographic latitude, the altitude and the thickness
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of the covering sediments. The cosmic dose rate is about 300 $\mu$Gy/a at sea level and decreases with depth below ground (see Prescott and Stephan 1982; Prescott and Hutton 1988).

Since the concentrations of radioactive elements in the sample and its surrounding are usually very different, it is necessary to determine the internal dose rate separately from the external dose rate.

**External dose rate** Material which is in contact with the piece of enamel (sediment or dentin) irradiates the sample with alpha, beta, and gamma rays, which are emitted from the containing radioactive elements (U, Th, and K). Figure 1 shows possible configurations for the enamel layer. The removal of the outer 20 $\mu$m of the sample eliminates the alpha irradiated volume (Figure 1(G)). Since the enamel layer is rather thin, it is normally not possible to remove the outer 2 mm which were beta irradiated. Since the beta dose rate decreases with depth, attenuation factors have to be considered. These have been calculated by Grün (1986).

The external beta dose rate has to be calculated separately from the external gamma dose rate because the beta dose rate is generated from the dentin or sediment immediately attached to the enamel layer, whereas the gamma dose rate originates from about 30 cm around the sample. The beta dose rate from the sediment is derived from the chemical analysis of U, Th, and K (in cases of Figure 1(D and E)). The beta dose rate of the dentin (Figure 1(E and F)) originates from its U-concentration. The dose rate for dentin is affected by U-disequilibria and U-accumulation (see below).

The gamma dose rate cannot normally be deduced from laboratory analyses but has to be measured *in situ* with a portable, calibrated gamma spectrometer or with TL dosimeters. This also has the advantage that the present-day water content is directly included; of course it is the average water content over the burial period that is relevant and the relationship of this to the present-day content has to be estimated from palaeoclimate information. However, this is not possible when working on museum collections. The gamma dose rate from dentin is negligible.

Water is a further attenuation factor and also has to be considered in the calculation of the beta and gamma dose rate (Bowman 1976; Aitken and Xie 1990). Dentin may contain up to 25% water.

**Internal dose rate** The internal dose rate is mainly generated by alpha and beta rays. Since teeth are virtually free of K and Th, only the U concentration has to be determined. An alpha efficiency of $0.15 \pm 0.02$ has been repeatedly measured (Grün 1985; DeCanniere *et al.* 1986; Katzenberger pers. comm.) and it is usual to assume this for all samples. Since the enamel pieces used are rather small, the internal beta dose rate is not 100% absorbed and a self-absorption factor has to be calculated. The internal gamma dose can normally be neglected. In many Quaternary samples, the U-decay chains display disequilibria, which are actually the basis for U-series dating. This is also the case for the uranium in enamel and dentin. These disequilibria affect the average dose rates and have to be taken into account mathematically.

**Early and linear U-uptake** One further effect complicates the dose rate determination of tooth enamel. Teeth and bones show a post-depositional U-uptake. This process of uranium uptake cannot normally be exactly determined. Dentin usually accumulates much more U than enamel (by a factor of 10 to 100). For teeth, two models have been suggested (Ikeya 1982; Grün *et al.* 1987):
Figure 7 Effect of U-accumulation on the calculation of an ESR age estimate. The dose rate of enamel and dentin is critically dependent on the U-uptake model. (left) With increasing U-concentrations in dentin and enamel, the contribution of the external sediment dose rate to the total dose rate becomes smaller and the dose rate calculation becomes more dependent on the U-uptake model. This causes an increasing difference between the early U-uptake (EU) and linear U-uptake ages (LU). At high U-concentrations the LU age is twice the EU age (right).

(i) a U-accumulation shortly after burial of the tooth (early U-uptake, EU); and
(ii) a continuous U-accumulation (linear U-uptake, LU).

Figure 7 shows the effect of U-accumulation on the calculated age. As long as the U-concentration in dentine and enamel is relatively low, the dose rate is mainly generated by the sediment (Figure 7 (left)) and there are only small differences in the EU and LU ages (Figure 7 (right)). However, as the tooth accumulates more uranium, the internal dose rate increases and there is more uncertainty. This effect becomes even more dominant when the external dose rate is lower. In the extreme case, the LU age is twice the EU age. The EU age is the minimum possible ESR age. In many cases we observe that the LU age agrees more closely with independent age estimates (e.g. Grün and Brunnacker 1987; Schwarcz et al. 1988a).

It has been suggested by Grün et al. (1988a) that U-series and ESR measurements should be made on the same sample. By comparing the results, it becomes possible to model the U-uptake. This approach requires very extensive analytical procedures, and U-series results with α-spectrometry have been found to be unsatisfactory because of the small size of the available sample and the usually low U-concentration in dentin. Additional problems occurred during the chemical preparation of the samples.

It has been shown by Grün and Invernati (1985) that uranium migrates into a tooth with a saturation front. One way to overcome this problem is to work on very big teeth (e.g. mammoth). The outside of the tooth acts as a buffer for the inside and samples cut from the middle have very low U-concentrations (see Figure 8). However, this is normally not feasible at archaeological sites.

In some cases it is possible to reconstruct the most likely U-uptake by systematic studies on long profiles. Grün et al. (forthcoming b) investigated the archaeological sequence of Pech de l’Azé II. The U-concentrations in the dentin of the larger teeth (horse, bovid) are relatively low in layers 2 to 4 (< 2 ppm) whereas the U-concentration in the dentin of equivalent teeth from layers 5 to 9 is much higher (10–20 ppm: see Figure 9). This implies that the major U-migration in the sediments took place before the deposition of layer 4, and that therefore the EU age results are more likely to be correct for the lower layers (5 to 9). Figure 10 shows the age estimates from the ESR analyses. There is clearly a hiatus between the deposition of layer 4 and layer 5 which corresponds to the last interglacial, with a warmer
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Figure 8  Cross-section through a mammoth tooth. The dark regions in dentin and cement are enriched in uranium. ESR samples are therefore cut from the interior of the tooth which is only little affected by U-accumulation.

and probably more pluvial climate. This may explain why during this time water (carrying uranium) was mobilized and the teeth of the lower levels accumulated uranium.

For the upper levels, it is not possible to postulate a general U-uptake model. Most larger teeth have low U-concentrations and therefore the EU and LU age estimates do not differ to a great extent. These teeth ought to give the most reliable results. For the teeth with high

Figure 9  U-concentrations in dentin of teeth from Pech de l’Azé II (see Figure 10).
Figure 10  ESR age results of teeth from Pech de l'Azé II. Triangles: mean EU ages with errors; closed circles: mean LU ages with errors; open circles: mean LU age estimates (oxygen isotope stage boundaries after Martinson et al. 1987).
U-concentrations, sometimes the EU uptake results seem to yield a better internal consistency (e.g. 633, 788), while in other cases the LU result seems more appropriate (636, 629). Without further evidence one can only assume that the ESR ages calculated with the EU and LU models more or less bracket the true age.

**WHY IS IT NOT POSSIBLE TO DATE BONES BY ESR?**

Although this application of ESR dating was extensively reviewed by Grün and Schwarcz (1987), their conclusion has not been widely recognized. Their views result from the following observations: (i) bones usually absorb more uranium than teeth and (ii) the mineral phase in bones is only somewhere in the region of 40–60% (enamel: ≈96%). Besides hydroxyapatite, bones contain a large proportion of an amorphous phase with a very similar chemical composition to hydroxyapatite. Along with the alteration of the organic constituents in bone during fossilization, the mineralogical compounds change. Besides the formation of new minerals (Piepenbrink 1989), disintegration of the mineral phase and conversion of the amorphous phase into hydroxyapatite is observed (Newesely 1989) as well as growth of the crystal size of hydroxyapatite (Hassan et al. 1977) even under sub-aerial conditions (Tuross et al. 1989). Formation of the new mineral phase (with new defects = traps) with time must generally lead to an age underestimation regardless of the U-uptake model applied. This was demonstrated by Grün and Schwarcz (1987) who compared results of tooth enamel and dentin (which is rather similar to bone).

Oduwole and Sales (1991) have suggested an alternative ESR procedure to date bones: they observe that the ESR signal begins to fade immediately after irradiation. In older bones this fading component becomes smaller relative to the total signal and therefore this effect was suggested as a dating technique. The fading has been attributed to collagen, proteins or surface interactions of small-sized hydroxyapatite crystals (Ostrowski et al. 1974). If the fading is controlled by the latter process, it actually shows the crystallinity of the hydroxyapatite component in the bone. The crystallinity is indeed age dependent, but also influenced by many other processes (see previous paragraph).

**Dating range**

The upper dating range of the ESR technique is mainly controlled by thermal stability of the paramagnetic centres. Trapped electrons only have a limited probability of staying in the traps. After some time, which is temperature dependent, the electrons leave the traps and recombine with holes. The thermal stability describes the mean life (τ) of a trapped electron at the defect site. The influence on the evaluated age of this recombination process is negligible only if the mean life is approximately 10 times higher than the age of the sample.

Preliminary annealing experiments by Schwarcz (1985) suggested a mean life in the range of 10 to 100 Ma. Apart from a very large scattering, an investigation of teeth from Sterkfontein (Grün and Schwarz, unpublished data) showed no particular trend of underestimation in an age range of 1.6 to 2.4 Ma, which corresponds more or less to the expected age (Grine 1988). Therefore, the effect of recombination seems negligible for dating of middle and late Pleistocene samples.

The other parameter which determines the upper dating range is complete saturation. When all traps are filled with electrons, the ESR signal cannot be enhanced by further irradiation. This effect has rarely been observed for tooth enamel.
The lower dating range is determined by the sensitivity of the ESR spectrometer. It is easily possible to detect a signal that is generated by 1 Gy. This may correspond to a few thousand years in a low dose rate environment. Appendix I describes sample collection, preparation and ESR measurement. This appendix also gives advice for the correct collection of ESR samples.

**Errors**

The major source of uncertainty in ESR dating of tooth enamel is usually introduced by the unknown U-uptake. Samples from Border Cave had individual errors in the range of 7 to 10%. However, in other cases, the laboratory error is much larger. A detailed discussion of the uncertainties involved in ESR dating of tooth enamel is given in Appendix II.

**SOME NOTES ON TL DATING OF BURNT FLINT**

The resetting mechanism for TL dating of burnt flint is the firing of the material by ancient humans. All the previously stored TL intensity is deleted and the signals grow again due to natural irradiation. The basic mechanism for TL is shown in Figure 2 (right). The determination of the AD is equivalent to that of ESR.

For flint, the following dose rate parameters have to be determined:

- **Flint**
  - U, Th, and K concentrations
  - alpha efficiency

- **Sediment**—gamma activity

- **Cosmic radiation**

The dosimetric situation for flint is simpler than for tooth enamel because of the greater size of the samples. Additionally, TL dating of flint is usually not influenced by disequilibria in the U-decay chain or by uncertainties introduced by U-uptake. Also, the mineralogical properties of hydroxyapatite (enamel) and microcrystalline quartz (flint) are considerably different. Therefore it is not very likely that ESR and TL dates from the same archaeological layer are influenced by the same systematic uncertainties to the same extent, and both techniques can be regarded as virtually independent. However, in the few cases where the proportion of the internal to external dose rate is similar for ESR and TL, uncertainties in the determination of the external dose rate (e.g. water contents or systematic errors in the sediment analysis) may influence both systems similarly.

**RECENT HUMAN EVOLUTION IN EUROPE**

Before 1970, it was believed by many workers that Europe or the Middle East were centres of origin for modern humans (Hrdlička 1930; Le Gros Clark 1964; Spencer 1984) and by some that a form of Neanderthal or ‘Neanderthaloid’ population was the most likely precursor (Hrdlička 1930; Weidenreich 1947; Brace 1964; Brose and Wolpoff 1971; Spencer 1984; see Figure 11). Moreover, sub-saharan Africa and the Far East were generally considered as insignificant or retarded backwaters in recent human evolution (Coon 1962; Howells 1967; but compare Weidenreich 1947). Such views were partly a reflection of the better (or better-known) fossil and archaeological materials from Europe and western Asia, but they were also a reflection of long traditions of Eurocentrism in palaeoanthropological thinking, no
doubt associated with the actual, or intellectual, origins of most of the active researchers. During most of the last century, European fossil hominids have generally been dated by their associated archaeology (if any), or by their associated fauna and/or flora in the context of the succession of recognized continental glacial and interglacial stages and sub-stages (Stringer et al. 1984). Problems have arisen recently with each of these approaches.

On the one hand, the idea of a unilinear cultural ‘progression’ in Europe has been challenged, so that the supposed ‘refinement’ or otherwise of artefacts is no longer automatically taken as a reflection of their relative antiquity. The significance of the Bordean classification of the Mousterian also continues to be debated. Most workers no longer consider that these typologically recognized variants could have been due to ethnic differences in Mousterian populations (Bordes 1961), but it is unclear whether they were instead primarily time-successive or could represent different facies of activity or adaptation by essentially the same populations (Laville et al. 1980; Ashton et al. 1986; Mellars 1986 and 1988). This latter argument has direct implications for the dating of a number of Neanderthal fossils which are associated with particular Mousterian variants, for example La Chapelle-aux-Saints (associated with La Quina/Charentian Mousterian) and Le Moustier (possibly associated with the Mousterian of Acheulian Tradition). A time-successive Mousterian scheme would place La Chapelle as considerably more ancient than the Le Moustier Neanderthal (Mellars 1988), whereas they might be penecontemporaneous under an alternative scheme.

On the other hand, there are also problems with using continental faunal and floral indicators for relative dating, now it is generally accepted from the marine oxygen isotope record that the global succession of warm and cold climatic fluctuations was considerably
more complex than indicated by terrestrial records. This means that terrestrial records are not only incomplete, but they may also fail to discriminate between distinct climatic stages because their faunas and floras were similar.

The site of Ehringsdorf (Germany) provides an excellent example of the potential pitfalls of both the 'cultural' and faunal/floral relative dating schemes discussed above (Cook et al. 1982). Until recently, it was believed that the Lower Travertine at Ehringsdorf contained early Neanderthal hominids associated with an 'advanced' Mousterian industry and a fauna and flora from the last interglacial ('Eemian' = oxygen isotope stage 5e, c. 120 ka: Steiner 1979). However, radiometric dating by uranium series, now supported by ESR, has shown that the Lower Travertine more likely belongs to the penultimate interglacial, at nearly twice the previously suspected age (230 ka = stage 7: Blackwell and Schwarcz 1986; Schwarcz et al. 1988a; Grün et al. 1988b; see Figure 12).

The effect of the various dating problems on the late Pleistocene hominid sequence has been to create an artificial clustering of fossils, with little potential to discriminate sites with morphologically comparable hominids, or similar archaeology or associated faunas/floras. This problem has been compounded by the few radiocarbon dates available for late Mousterian and early Upper Palaeolithic sites, many of which should only be regarded as minimum ages. This has led to a probable compression of the actual chronology for many of the sites into the period between 30 and 50 ka (see Figure 29), a compression which is beginning to become apparent with the application of accelerator dating, and comparisons with U-series dates (Straus 1989). In particular, it is in sites generally assigned to the critical
period of time immediately before 40 ka that we are likely to see the dating techniques of ESR, TL and U-series making their greatest impact as they 'stretch' a condensed chronology to a more realistic length. Having discussed the problems of existing dating schemes for European fossil hominids, we will now review the application of ESR dating to particular sites, beginning with those in France.

La Chapelle-aux-Saints

The partial skeleton of a Neanderthal male (see Figure 13) found in 1908 in the small cave of Bouffia Bonneval at La-Chapelle-aux-Saints, near Brive, almost certainly derived from a burial (Oakley et al. 1971). Although by no means the first Neanderthal skeleton found, it achieved particular significance because of the detailed and influential descriptions published by Boule (see e.g. Boule 1911 and 1921). The skeleton was excavated from a yellow clay with limestone fragments (bed 1), associated with a 'cold' mammalian fauna and artefacts now assigned to the La Quina variant of the Charentian Mousterian. Using the existing palaeoclimatic framework for south-west France, the hominid has, like most Neanderthals of this region, been assigned to 'Würm II (old usage)' (Oakley et al. 1971), which would indicate a relatively late Mousterian age, c. 50 ka. However, as already indicated, an alternative scheme proposed by Mellars (1988) would place the La Quina Mousterian variant rather earlier, at c. 60 to 68 ka.

Figure 14 shows the ESR age estimates (see also Raynal and Pautrat 1990). Since the
Figure 14  ESR age results on four mammalian teeth from the hominid level at La Chapelle-aux-Saints. Triangles: mean EU ages with errors; circles: mean LU ages with errors.

sediments in the cave are rather disturbed, the determination of the external gamma dose rate has to be treated with some caution. The EU age estimates suggest a rather young age for the La Chapelle-aux-Saints skeleton, 47 ± 3 ka, in line with conventional views, whereas the LU age estimates average 56 ± 4 ka, in between values previously estimated from the conventional dating scheme and that advocated by Mellars.

Le Moustier

Two hominid skeletons were recovered from this rock shelter, which gave its name to the term 'Mousterian' (Oakley et al. 1971). The first, of an adolescent male Neanderthal, was excavated under questionable circumstances in 1908, probably from bed J, while the second, of a young child, was excavated from a supposed grave cut through beds H–I in 1914. Both skeletons suffered post-extraction depredations, since Le Moustier 1 was partly destroyed by a fire in Berlin in 1945, while Le Moustier 2 was lost before it had even been described! Le Moustier 1 is conventionally dated to the later part of 'Würm II (old usage)' (Oakley et al. 1971) and was apparently associated with either a typical Mousterian, denticulate Mousterian or Mousterian of Acheulian Tradition. In each case, Mellars’s dating scheme would place this specimen rather late on archaeological grounds, at c. 40 to 50 ka.

Le Moustier was first dated by TL (Valladas et al. 1987a). Figure 15 shows a comparison of the TL and the preliminary ESR results (Grün et al. forthcoming a). The teeth were recovered from the same test excavation as the burnt flint for the TL study. The ESR determinations, whether EU or LU, agree with previous TL determinations in placing bed J as apparently very late in the French Mousterian sequence, since bed H is dated at c. 41 ka. This provides a maximum age for the overlying beds I–J. Le Moustier 1 would, therefore, appear to be one of the youngest Neanderthal fossils known.

Grotta Guattari

The Guattari cave, Italy, has produced three fossil hominids (Oakley et al. 1971). Two were
found on the cave surface (layer G0) in 1939: the famous 'Monte Circeo' Neanderthal skull (Guattari 1, see Figure 16) and an incomplete mandible of a second adult individual (Guattari 2). A better preserved mandible was excavated in 1950 from a breccia at the cave entrance (Guattari 3). All lie stratigraphically above the basal marine-influenced deposits (G7) which may relate to the final high sea level event of oxygen isotope stage 5 (5a = c. 74–84 ka, Martinson et al. 1987). The sediments of G0–G1 have been attributed to a cold phase, correlated to 'Würm II (old usage)', and there is now little evidence that layers G4–G5 contained a warmer fauna, including Hippopotamus, as previously claimed (Stiner 1991). There are few, if any, artefacts associated with layer G0, which is now interpreted as predominantly the result of occupation by the hyaena, Crocuta crocuta (Stiner 1991; White and Toth 1991). However, the lower deposits (G1–G2, G4–G5) contain two rather different artefact assemblages of the Pontinian Mousterian (Kuhn 1991).

The site was dated using U-series and ESR by Schwarcz et al. (1991). U-series dating on the innermost (earliest) calcite encrustations of surface bones give an age of 51 ± 3 ka, which provides an approximate minimum age for the hominids. Mammal teeth from the surface deposit have been dated by ESR to 44 ± 5 ka (EU) and 62 ± 6 ka (LU) and for the immediately underlying layer to 44 ± 6 (EU) and 54 ± 4 (LU). Hence Guattari 1 and 2 most probably date between c. 50 and 60 ka, with Guattari 3 bracketed between this last estimate and the end of oxygen isotope stage 5 (i.e. between 60 and 74 ka).
For the Middle East, the chronologies used by workers such as Howell (1951) in assessments of human evolution in the area were substantially modified during the following two decades (cf. Howell 1959). Howell had considered the Levant Middle Palaeolithic-associated samples from Skhul, Qafzeh and Tabun to represent a morphologically variable generalized or early Neanderthal population from the last interglacial, and one which lay close to the ancestry of both modern humans and 'classic' Neanderthals. However, by the early 1980s, further study, faunal analyses, limited radiocarbon dating, amino-acid dating using fossil bone, and lithic and morphological dating, produced a new chronology for the Levant hominids, where Neanderthals from sites such as Tabun, Shanidar and Amud dating from about 50 000 to 60 000 years were apparently succeeded by early moderns from the sites of Skhul and Qafzeh, dated to about 40 000 years ago (Jelinek 1982a and b; Trinkaus 1984). This inferred sequence paralleled that in Europe, but the Levantine Neanderthal/modern succession was apparently somewhat older, as it occurred during the Middle Palaeolithic rather than at its end. Some workers, therefore, proposed that modern humans evolved from Neanderthals in the Levant about 45 000 years ago, and then dispersed into Europe, where they either accelerated the evolution of the Neanderthals through gene flow, or directly founded the populations of the Upper Palaeolithic. It could be argued that our understanding of the chronology of Levantine Middle Palaeolithic sites has been hampered, rather than helped, by the application of radiocarbon dating (Weinstein 1984), since many determinations which should have been regarded as minimum values were taken as real ages. Equally, bone amino acid dating programmes were apparently never completed or published in detail (see e.g. Masters 1982), and provided no substantial clarification of the fossil hominid sequence in the Levant.
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Kebara

The Mount Carmel cave of Kebara (Israel) contains extensive archaeological deposits from the Holocene and Pleistocene, and produced a series of Upper Palaeolithic hominids in 1931, and a fragmentary infant skeleton from a Mousterian context in 1964 (Oakley et al. 1975). However, during the last decade further excavations in the Mousterian levels have yielded more hominids, including a very robust male Neanderthal skeleton which is virtually complete from the mandible down to the pelvis (Rak 1990). This skeleton was apparently a burial into unit XI1 (Bar-Yosef et al. 1988), associated with an industry which is considered to be late in the Levantine Mousterian sequence (Bar-Yosef 1989). ESR dating was conducted on seven mammal teeth, mostly gazelle, from the overlying unit X (see Figure 17).

The ESR age estimates average $60 \pm 6$ ka (EU) and $64 \pm 6$ ka (LU), and are close to the TL dates on burnt flints which average $60 \pm 4$ ka for units XI and XII (Valladas et al. 1987b). These suggest that the Kebara skeleton might be slightly older than the European Neanderthals dated so far by ESR, and in the case of Le Moustier considerably older.

Jebel Qafzeh

Remains of at least 20 hominids (infants, children and adults; see Figure 18) have been excavated from the Qafzeh cave near Nazareth, Israel, in two separate phases of excavation (1932–5 and 1965–79; Oakley et al. 1975; Vandermeersch 1981). The initial finds were from an Upper Palaeolithic context, but the majority were associated with the Middle Palaeolithic, and several appear to represent burials. The early work under Neuville concentrated on excavating the interior of the cave, while the later work, under Vandermeersch, concentrated on the terrace deposits. Because the early work was never completely published, there are some uncertainties about the exact stratigraphic positions of the hominids recovered, but the majority derived from layer L, which appears to correlate with layers XVII–XIX of the later excavations. Layer XVII produced the majority of the subsequent hominid sample.

Figure 17  ESR age results on teeth from layer X, Kebara (after Schwarcz et al. 1989). Open circles: mean EU ages with errors; closed circles: mean LU ages with errors; triangle: TL results from Valladas et al. (1987b).
The Qafzeh hominids have sometimes been regarded as Neanderthals, or transitional forms between Neanderthals and modern humans (Brace 1964; Brose and Wolpoff 1971; Wolpoff 1980), but most workers now regard them as closely related to, or actually representing, *Homo sapiens*, albeit with some archaic cranial (but apparently not postcranial) features (Stringer 1978; Stringer and Trinkaus 1981; Vandermeersch 1981; Trinkaus 1984). Assigning an age to the Qafzeh hominids has been difficult, with most workers until recently following either a morphological dating scheme, or an archaeological dating scheme, both of which placed the Middle Palaeolithic associated hominids of Qafzeh (and Skhul) as later than the Levantine Neanderthals, at c. 40 ka. Sedimentological work (Farrand 1979) and amino acid analyses on bone (Masters 1982) could be used to suggest a greater antiquity for Qafzeh, at least, but it was only the microfaunal analyses of Haas and Tchernov (Tchernov 1988; Bar-Yosef and Vandermeersch 1981; Vandermeersch 1981) which suggested that the Qafzeh Middle Palaeolithic levels might be earlier than those associated with the Neanderthals from Tabun and Kebara, with a possible antiquity of about 70 ka.

However, in 1988, TL dates on 20 burnt flints from Middle Palaeolithic layers XVII–XXIII were published, showing no systematic change with depth, and giving a weighted mean age of 92 ± 5 ka (Valladas *et al.* 1988). These results had already been circulated at scientific meetings, where they had provoked great controversy, so it was important to make further age estimates for the Qafzeh Middle Palaeolithic levels by other methods. Later in the same year, 14 ESR age determinations on six mammal teeth from layers XV–XXI were published (Schwarcz *et al.* 1988b).

As with the TL results, there was no systematic change with stratigraphic depth, and the average ages of 100 ± 10 (EU) and 120 ± 8 (LU) ka are comparable with, or slightly greater than, the TL determinations (see Figure 19; note that the results slightly differ from the ones published by Schwarcz *et al.* (1988b) due to recalculation of the AD with error (Grün and...
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Figure 19  ESR age results on teeth from Qafzeh. Open circles: mean EU ages with errors; closed circles: mean LU ages with errors; triangle: TL results from Valladas et al. (1988).

Macdonald 1989) and by addition of one further specimen, 374). Taken together, these results strongly suggest a stage 5 age for the Qafzeh hominid-bearing Middle Palaeolithic deposits.

Skhul

Skeletal remains in varying degrees of completeness, representing at least 10 adults and children, were excavated from the small cave of Skhul, Mount Carmel (Israel), in 1931–2 (Garrod and Bate 1937; McCown and Keith 1939; Oakley et al. 1975; Trinkaus 1984). The fossils in some cases appeared to derive from burials in a deposit (layer B) which also contained nearly 10000 Middle Palaeolithic artefacts, and mammalian faunal remains. The hominids are morphologically similar to those from Qafzeh (Stringer 1978; Stringer and Trinkaus 1981; Vandermeersch 1981; Trinkaus 1984) in their relatively gracile post-crania,
and crania which combine modern and primitive characteristics (the latter including some strongly built brow-ridges and high total facial prognathism, see Figure 20). General assumptions about the age of the Skhul sample placed it as very late in the Middle Palaeolithic sequence of the area, as suggested by Jelinek’s (1982a and b) archaeological relative dating scheme. ESR dating was applied to seven samples of enamel from two bovid teeth (samples 521, 522) from layer B, curated at the Natural History Museum, London. Since the sediments of the cave were virtually entirely removed during the excavations, it was not possible to measure the gamma dose rate in situ. The gamma dose rate was therefore reconstructed from the analyses of the sediment attached to the teeth and of separate samples at the Museum. The mean ESR age estimates were 81 ± 15 ka (EU) and 101 ± 12 ka (LU) (Stringer et al. 1989).

Although one tooth (522) had an estimated age lower than the other (considerably so in the case of the EU estimate), the mean age determinations suggest a stage 4 or (more probably) stage 5 age for layer B, considerably older than the age expected for a late Middle Palaeolithic site. However, the Skhul age estimates are either slightly younger than, or in agreement with, the TL and ESR age estimates for Qafzeh discussed above. These determinations bring the morphologically comparable Skhul and Qafzeh samples close in time, and appear to establish an early modern presence in the Levant well before the demise of the Neanderthals, and penecontemporaneous with the earliest records of Homo sapiens in Africa.
Tabun

The large and deep cave of Tabun, Mount Carmel (Israel), was excavated between 1929 and 1934, and produced an extensive Middle and Lower Palaeolithic sequence and several hominid fossils (Garrod and Bate 1937; McCown and Keith 1939; Oakley et al. 1975; Trinkaus 1984), the most important being Tabun I (a partial adult female skeleton) and Tabun II (an adult, probably male, mandible). Both were recovered from layer C, associated with Middle Palaeolithic (Mousterian) artefacts, but it is possible that Tabun I (C1) was actually an intrusive burial from layer B (Garrod and Bate 1937). More fragmentary hominids were also recovered from stratigraphically higher deposits (the chimney, layer B and the terrace), layers C and E. The C1 skeleton is certainly that of a Neanderthal, albeit one with relatively small post-cranial bones, strong browridge and transversely flat upper face, while the Tabun II (C2) mandible appears to be that of a Neanderthal, but with some development of a chin (Trinkaus 1984).

Establishment of a reliable chronology for the Tabun archaeological sequence and hominids would be very significant in a regional context, because the site has been extensively used for late Pleistocene correlation within the Levant (Jelinek 1982a and b; Bar-Yosef 1989). Excavations under the direction of A. Jelinek (1967–72) lead to a revised chronology where Jelinek’s more detailed stratigraphic system of units can be correlated with the cruder scheme of Garrod (Jelinek et al. 1973; Jelinek 1982a and b). On this basis, layers B to C could date from c. 40 to 60 ka, D from c. 70 ka, and E from the later part of oxygen isotope stage 5 (c. 80 to 100 ka). Bar-Yosef’s (1989) alternative scheme is based partly on archaeology, partly on the microfaunal analyses of Tchernov (1988) and partly on absolute dates for other sites. He has proposed that there was a much longer hiatus between Garrod’s layers C and D, such that D might date from c. 100 ka and the lower part of E from stage 6, c. 150 ka. These schemes are summarized in Figure 21.

A series of radiocarbon determinations was made on charcoal and black soils from Tabun (Weinstein 1984), including samples from Garrod’s excavation of layers C (40 900 ± 1000 BP) and D (35 400 ± 900 BP), and Jelinek’s sequence (unit I ranging between 45 800 ± 2100 – 1600 and 51 000 ± 4800 – 3000). Although some workers seem to have accepted these determinations as real ages, it is clear that they should be viewed as minimum ages only.

Large faunal samples from Garrod’s excavation are curated at the Natural History Museum, London, and it was decided to sample 20 artiodactyl teeth from layers B to E for ESR dating, with separate labelled sediment samples used for the reconstruction of the external gamma dose rate (Grun et al. 1991). The results are shown in Figure 22.

The ESR age estimates suggest a greater age and much longer period of deposition for layers B to E. In fact, the age estimates for each layer are about twice those estimated from Jelinek’s chronology. Thus layers B to C are placed within stage 5, D represents stages 5 or 6, while E may cover much of stage 6 and even 7. If these age estimates are approximately correct, they would place layer C (and the associated Neanderthals) at approximately the same time period as, or slightly older than, the early modern hominids from Skhul and Qafzeh. However, there is insufficient precision to establish contemporaneity of Neanderthals and early moderns during stage 5 in the Levant. As well as suggesting that Tabun C1 may be an early Neanderthal, the Tabun age estimates have many further implications for existing hypotheses concerning the hominid sequence, archaeology, biostratigraphy, and palaeoenvironments of the Levant (see discussion in Grün et al. 1991) and it will be
important to compare the ESR chronology with that being developed using TL (Valladas pers. comm.).

Amud

A partial Neanderthal skeleton was discovered in the Amud Cave near Tiberias (Israel) in 1961 (Suzuki and Takai 1970; Oakley et al. 1975). The skeleton, which may have been from a contracted inhumation, had most body parts represented but many were poorly preserved. This discovery, Amud I, was followed in 1964 by Amud II (an adult maxilla), III (cranial and dental fragments of an infant), IV (an infant temporal fragment), and V (an isolated lower molar). All were recovered from 'formation B', associated with Middle Palaeolithic artefacts and intrusive later materials, with Amud I coming from the very top of this level. This skeleton (and as far as can be determined, the other hominids) represents a Neanderthal with numerous typical characteristics of the skeleton, but unusually tall and (for such a large male
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Mean ESR results for Tabun cave. Triangles: EU age estimates; circles: LU results. The averages for the sedimentological units are given with one standard deviation resulting from the scattering of the mean values.

individual) relatively small-browed and small toothed (Suzuki and Takai 1970; Trinkaus 1984).

Attempts at dating formation B met with only limited success (Suzuki and Takai 1970). A large series of radiocarbon determinations on collagen and carbonate fractions from mammalian fauna gave a range of terminal Pleistocene and Holocene ages (< 19 ka; see also Weinstein 1984) while 'uranium-fission track dating' (Suzuki and Takai 1970) gave an estimate of 28 ± 10 ka. These determinations are unrealistically young for material from a
Middle Palaeolithic context. It is hoped that the resumption of the excavations at Amud will provide a better chronology for past and future discoveries at the site. A preliminary ESR age estimate has been made on an artiodactyl tooth collected by the authors from near the top of formation B. Two sub-samples gave age estimates of $42 \pm 3, 41 \pm 3$ (EU) and $49 \pm 4, 50 \pm 4$ (LU).

This date is suggestive of a rather young age for the Amud Neanderthal compared with other Levantine Middle Palaeolithic hominids, and is of particular interest given some of the supposed 'progressive' evolutionary features of Amud I. If it is indeed late in time, it may document the existence of evolutionary trends in Levantine Neanderthals, or gene flow from contemporary *Homo sapiens*. Given the Amud age estimates, an ESR study of the Lebanese cave of Ksar Akil, which spans the Middle/Upper Palaeolithic transition (Bergman and Stringer 1989), would be useful.

**AFRICA**

As we have already indicated, prevailing views for much of this century were that Africa was an evolutionary and cultural backwater in the story of modern human origins, even if it merited an important place in early hominin evolution. As Coon (1962, 656) put it, 'If Africa was the cradle of mankind, it was only an indifferent kindergarten'! It was often regarded as an isolated continent, where relict archaic forms such as the 'Rhodesioids' (a term derived from the supposedly late Pleistocene Broken Hill skull of 'Rhodesian Man') persisted into the late Pleistocene, and the advanced behaviours associated with European Upper Palaeolithic peoples were slow to develop or penetrate (Coon 1962; Cole 1965; Howells 1967). However, reviewing the literature of the following decade from 1970 to 1980, it is clear that a growing number of fossil discoveries and re-evaluations, new radiocarbon dates and applications of other dating techniques were beginning to have an impact on such ideas about modern human origins.

In 1975, Clark reviewed the growing evidence for an important African role in modern human origins in his paper 'Africa in prehistory: peripheral or paramount?'. He made extensive use of an emerging revision of African archaeological chronologies, where it was becoming apparent that the African Middle Stone Age (MSA) was not time-equivalent to the Eurasian Upper Palaeolithic, but rather to the Middle Palaeolithic, and he made the following comment: 'far from Africa's having been the cultural backwater during the later Pleistocene that earlier notions has suggested, certain major technological advances appear to have been initiated there' (Clark 1975, 175). By 1980, it was realized that the archaic 'Rhodesioids' were more likely to date from the middle Pleistocene, in line with fossil evidence from Europe and Asia, and an increasing number of claims for an early appearance of anatomically modern humans in Africa were being made (Leakey et al. 1969; Beaumont et al. 1978; Howell 1978; see also Protsch 1975). However, because of the lack of suitable dating methods for the time immediately beyond the practical range of radiocarbon, such ideas were only slowly accepted, or not at all, so that Wolpoff in 1989 could still claim of the African fossil evidence 'not one specimen thought to be an 'early modern' has a defensible radiometric date' (Wolpoff 1989a, 64–5).

**Klasies River Mouth Caves**

The complex of adjoining (and mainly continuous) cave deposits in a series of cave entrances at the mouth of the Klasies River, South Africa, were excavated in 1967–8 by Singer and
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Wymer (1982), and by Deacon in 1984–6 (Deacon et al. 1986; Deacon and Geleijnse 1988). These excavations produced a number of fragmentary, but apparently anatomically modern, hominid specimens in MSA levels (Rightmire and Deacon 1991). The age of these levels has been a source of controversy for many years (see e.g. Binford 1984; Deacon 1985), although an early late Pleistocene age for the lowest MSA deposits has been increasingly supported by recent studies (Deacon 1989). The early MSA1 and hominids occur in the light brown sands (LBS) and the lowest part of the sands-ash-shell (SAS) members, the MSA2 and associated hominids occur in the middle and upper SAS and (without hominids) in the rockfall (RF) members, while the Howiesons Poort (HP), MSA3 and a few associated hominids occur in the upper member (UM) (see Figure 23).

ESR dating was attempted using five mammalian tooth fragments from the basal (MSA1) SAS, the higher (MSA2) SAS and two parts (HP and MSA3) of the UM, producing 14 EU and 14 LU age estimates (Grun et al. 1990b, see Figure 24). In contrast to the specimens from Border Cave, all teeth contained considerable amounts of uranium and there is a large difference between the EU and LU results. The ESR age estimates clearly reflect their stratigraphic positions. The LU results of sample 543 (94 ± 10 ka and 88 ± 8 ka), which is associated with the lower SAS, show reasonable agreement with the U-series results that gave an upper limit of 110 ka for this layer. This strongly indicates that the LU results are more likely to be correct. The LU results for the Howiesons Poort layers and the layer immediately above range between 40 and 60 ka. This is in reasonable agreement with ESR dating of the comparable levels at Border Cave, where the dosimetry is completely different.

The ESR age estimates support an early late Pleistocene age for the earlier Klasies...
deposits, associated hominids and MSA artefacts. The oldest hominids, two maxillary fragments, are likely to be older than 90 ka, while specimens such as the mandible 41815 (see Figure 25 (right)) are probably about that age. The frontonasal fragment 16425, partial mandibles such as 13400 and 16424, and post-cranial fragments are likely to date to between 60 and 85 ka, while the more fragmentary specimens from the HP and MSA3 levels may be considerably younger, in the range 40–60 ka old. These ESR age estimates, like those from Border Cave, demonstrate the relatively great antiquity of the early MSA (and actual or putatively associated hominids) in South Africa. However, the age estimates also suggest a greater total time range for the MSA and a younger age for the HP facies than is sometimes considered likely. In addition, while some of the hominid specimens allied to modern Homo sapiens can be confirmed as close to 90 ka in age, others certainly appear to be younger than this.

Border Cave

This cave in north-east South Africa has produced four hominids believed to be of Pleistocene age (Oakley et al. 1977; Beaumont et al. 1978; Beaumont 1980). The main feature of the 4-m deep sequence of cave deposits is an alternating series of units termed ‘brown sands’ (BS) and ‘white ashes’ (WA). Border Cave 1 (BC1), an incomplete cranial vault and possibly associated post-cranial fragments, and BC2, a partial adult mandible, were recovered out of context in 1941–2. They must have been derived from a level above the 4WA if that unit was still intact in 1942, and it is claimed that matrix on BC1 most closely matches sediment at the base of unit 4BS (Beaumont 1980). BC3, the partial skeleton of an infant, was excavated from an apparent grave within the 4BS in 1941, BC5, a nearly complete adult mandible (see Figure 25 (left)) was collected from a shallow pit within the 3WA in 1974.
Most workers accept that the preserved parts of the Border Cave hominids are anatomically modern in morphology (but cf. Van Vark et al. 1989), but some have doubted whether the specimens (especially BC1 and BC2) are really contemporary with the Middle Stone Age (MSA) levels of the cave (Klein 1989). Radiocarbon has been used to date charcoal and bone collagen from the upper deposits, suggesting an age of > 41 to > 49 ka for levels below the late MSA of unit 2BS, and by inference for the hominids as well. The lower units have so far only been dated by extrapolation of dates obtained from the upper sequence (Butzer et al. 1978; Beaumont 1980). In order to develop an ESR chronology for the stratigraphy and the archaeological sequence, provenanced teeth of various large mammals were used to produce 66 age estimates (Grün et al. 1990a). Because, exceptionally, the teeth did not contain any significant amounts of uranium, the age results were not affected by uranium uptake. It is therefore possible to provide single age estimates for each specimen (see Figure 26).

The large series of ESR age estimates supports the early late Pleistocene date suggested for the lower MSA deposits of Border Cave, although the units appear younger than sometimes proposed, as does the MSA/Late Stone Age (LSA) transition (which occurs between 1WA and 2BS). Assuming that the stratigraphic positions of BC3 and BC5 are as already discussed, the former specimen should be about 70-80 ka, and the latter 50-65 ka in age. As BC1 and 2 derived from above the 4WA, they are probably less than 90 ka old, and may be of comparable age to BC3 (i.e. 70-80 ka). Unfortunately, attempts at accelerator dating the BC hominids in Oxford (see Grün et al. 1990a) failed due to insufficient collagen.

Although the Howiesons Poort layers in Border Cave and Klasies River Mouth Caves are not directly associated with most of the hominid remains, they are of particular archaeological importance and the ESR results need some further discussion. The Howiesons Poort (HP, also known as Epi-Pietersburg or MSA2) lithic industry shows several ‘advanced’ aspects that seem to resemble those of the subsequent LSA and Upper Palaeolithic elsewhere (Beaumont et al. 1978; Beaumont 1980; Sampson 1974; Singer and Wymer 1982; Mellars 1989; Mellars and Stringer 1989). Its chronological position is therefore of great interest.

Most previous arguments concerning the age of the Howiesons Poort are based on
sedimentation rates, faunal and floral investigation and oxygen isotope studies. Although many finite C-14 dates were derived from the HP levels, the oldest infinite dates have been emphasized, combined with suggestions of recent contamination for the finite determinations (see Beaumont et al. 1978). A widely accepted view seems to be that the HP layers represent a relatively short phase covering 85 000 to 95 000 years (Beaumont et al. 1978, fig. 6) or 65 000 to 75 000 years ago (Deacon 1989, fig. 28.1). However, oxygen isotope studies (Shackleton 1982) and sea level reconstruction (van Andel 1989) can provide a case for a younger age for these levels (around 40 000 years). Parkington (1991) summarizes the various views on the temporal position of the HP at Border Cave and elsewhere.

The Howiesons Poort dating range at Border Cave is estimated to be from about 45 ± 5 ka (HP/MSA3) to about 75 ± 5 ka (HP/MSA1). The LU-ESR age estimates for the HP layers at Klasies River Mouth are in the 40 to 60 ka range.

New amino acid racemization data on ostrich egg shells (Miller and Beaumont 1989) suggest that the sequence at Border Cave is about 30% older than estimated by ESR. The
amino acid dates are calibrated on an AMS radiocarbon date of 38,000 BP for a sample from 1BS (D/L = 0.262 ± 0.024). The D/L ratios for the lower layers then yield ages for the 1WA (D/L = 0.254 ± 0.022: 38 ka); 2BS (2BS.UP and 2BSLR.A: D/L = 0.338: 51 ka; 2BSLR.C: D/L = 0.385 ± 0.008: 59 ka); 2WA (D/L = 0.471 ± 0.008: 73 ka); and 5BS (D/L = 0.88: ≈ 120 ka). The respective ESR age estimates are: 1BS: 31 ± 2 ka, 1WA: 30 ± 3 ka; 2BS: 42 ± 7 ka; 2WA: 48 ± 2 ka; 4WA represents a time range of about 80 to 125 ka and 5BS: 128 ± 10 ka. Since the ESR set has a very high internal consistency, a random error can be excluded. Even accepting a systematic, non-identified 20–30% underestimation of the ESR dates (which gives them a surprising consistency for the lower layers), it is not possible to argue away a long duration for the HP deposits (then about 60 to 100 ka). Thermal annealing can be excluded, because its effect would be an increasing deviation with age (which is not the case for the lower layers). Anomalous fading can be excluded for hydroxyapatite in general, since at many other sites the ages were either in agreement with other evidence or found criticism because they seemed too old. On the other hand, a long duration of the HP cultural phase would resolve most of the contradictions that have been outlined by Parkington (1991). A resolution of the discrepancies in age estimates for the Border Cave deposits will clearly require much further work using different dating techniques.

**Jebel Irhoud**

The Jebel Irhoud cave is a solution cavity in a barytes quarry near Safi, Morocco (Ennouchi 1962; Oakley et al. 1977; Hublin and Tillier 1981; Hublin et al. 1987). Four hominids were recovered from the site between 1961 and 1969, but only the last of these has a precise provenance. All were found close to the cave floor, in association with Middle Palaeolithic (Mousterian) artefacts. Irhoud 1 is a fairly complete adult cranium, Irhoud 2 an adult calvaria (see Figure 27), Irhoud 3 a child’s mandible and Irhoud 4 a child’s humerus. The material has been termed Neanderthal (see e.g. Ennouchi 1962; Howell 1978), but it is evident that the cranial and facial material lack Neanderthal derived features (Howells 1975; Stringer 1978; Santa Luca 1978; Hublin 1989). However, the dentition of Irhoud 3 is very large, and the humerus, although immature, is strongly built (Hublin and Tillier 1981; Hublin et al. 1987). Overall, the specimens are certainly morphologically archaic, although they are seen as foreshadowing modern humans (Hublin and Tillier 1981; Stringer 1978 and 1989).

Dating the Irhoud site has been difficult, with only a minimum age radiocarbon determination on a mammal bone of greater than 30 ka as a guide (Oakley et al. 1977). The fauna has been correlated with the Soltanian or, more probably, pre-Soltanian stage, indicating an early late Pleistocene date (Hublin et al. 1987). However, the unsystematic collecting procedures used earlier at the site mean that a precise context cannot be provided for Irhoud 1–3. Nevertheless, ESR dating has been carried out on three mammal teeth, producing 5 EU and LU age estimates. These teeth derived from a level immediately overlying Irhoud 4.

EU age estimates range between about 90 and 125 ka and the LU estimates between 105 and 190 ka. The teeth show increasing U concentrations with the estimated ages, which makes it unlikely that the one large sediment sample from which the external dose rate was derived was representative for the environment of all three samples. The distinct stepwise uranium concentrations in the dentin, and the range of ages determined, suggest that the site has a long depositional history, covering at least oxygen isotope stages 5 and 6. If the
hominids were indeed low in the stratigraphic sequence, a stage 6 age (130–190 ka: Martinson et al. 1987) might well be appropriate, but more data are required.

Singa

The Singa calvaria (see Figure 28) was discovered in 1924 in a caliche deposit within the Gezira clay of the Blue Nile, Eastern Sudan. Fauna and artefacts were also collected at Singa and the related site of Abu Hugar, 15 km further south. The hominid was initially considered to be anatomically modern, and perhaps that of a ‘protobushman’ because of its unusual shape, but Tobias (1968) and Brothwell (1974) noted archaic features, with Brothwell considering that pathology might have affected the shape of the skull. However, workers such as Rightmire (1984) and Clark (1988) have continued to view this specimen as essentially modern in morphology. Stringer (1979) concluded that it was primarily archaic in morphology, and agreed with Brothwell that cranial shape was affected by a growth anomaly in the parietal region. Stringer et al. (1985) conducted further analyses of the specimen, and of a newly-obtained endocranial cast, and provided more data on the archaic nature of the calvaria, as well as its possible pathology.

Dating the Singa site and hominid has proved difficult. Bate (1951) considered that the associated fauna, which contained some extinct species, might be of earlier late Pleistocene age. The artefacts, while mainly nondescript, have been considered to be Middle Palaeolithic
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Figure 28  Right lateral view of the Singa calvaria.

(Clark 1988). However, a crocodile tooth from Abu Hugar produced a young radiocarbon age of $17300 \pm 200$ BP, although contamination by more recent carbonate may have occurred (Whiteman 1971).

Two mammal teeth from Singa in the Natural History Museum collection were analysed. The external gamma dose was reconstructed from matrix samples of the caliche deposit. The equid tooth was labelled as associated with the Singa skull, the bovid tooth as collected by A. J. Arkell. The teeth contained very high amounts of uranium which results in a very large difference between the EU ($97 \pm 15$ ka) and LU ($160 \pm 27$ ka) results.

The age estimates must be considered very provisional given the uncertainties in dose rate calculations, the differences between the two teeth sampled, the large difference between the EU and LU values and the complex geology of the site (Clark 1988). However, the determinations certainly indicate a stage 5 or 6 age for the teeth. If the assessments of the Singa calvaria as archaic are correct, a stage 6 age for the hominid is feasible. If, instead, the Singa specimen is regarded as anatomically modern, it could represent one of the oldest examples known.

SUMMARY AND FUTURE PROSPECTS

The last decade has been a very exciting one for studies of modern human origins, as new ideas and data have made their impacts. In contrast with the previous decade, it is now
generally accepted that modern humans appeared in the Middle East and Africa before they appeared in Europe, where they were contemporaneous with the last Neanderthals. These changes have come about through both morphological and chronostratigraphic approaches. There has been a demonstration from metrical and anatomical studies of the real distinctiveness of early modern specimens from the Neanderthals, and there has been clarification of the relative (and in some cases absolute) age of the Neanderthal-early modern interface in each region. As we have shown above, ESR dating has played its part, in indicating that some early modern fossils in the Levant and southern Africa are in the region of 90 to 100 ka old. However, Neanderthals persisted in Europe until less than 40 ka ago (see Figure 29).

The 'genetic revolution' in studies of modern human origins is mainly a phenomenon of the last five years, and has served to demonstrate (with some significant exceptions) the distinctiveness and (in some cases) the greater diversity of sub-Saharan Africans compared with other modern populations (Cann et al. 1987; Cavalli-Sforza et al. 1988; Vigilant et al. 1989; Long et al. 1990; but cf. Excoffier and Langaney 1989). Calibration of the genetic
origin of Homo sapiens and of the African/non-African split is still controversial, but a number of estimates place the origin between 100 and 200 ka age (Cann et al. 1987; Cavalli-Sforza et al. 1988; Vigilant et al. 1989; Nei and Livshits 1991). The genetic data served to focus attention on southern Africa as a possible place of origin for our species, especially when coupled with the apparent presence of modern human fossils in relatively early MSA contexts. Although claims for equally ancient Homo sapiens were being made for other sites such as Omo-Kibish in Ethiopia (Leakey et al. 1969; Day and Stringer 1982) and Qafzeh (Bar-Yosef and Vandermeersch 1981), these were not so readily accepted. However, ESR and TL dating have now served to make the Middle East an additional focus of interest, and to broaden the possible areas of origin to include northern and eastern Africa, as well as the Levant itself (Stringer 1988). This is not incompatible with present genetic data, but would imply that the original genetic pattern for Homo sapiens in northern Africa has been disrupted by subsequent migration or gene flow from the north, which is why North Africans are now more closely allied to Eurasians, rather than to sub-saharan Africans. Support for this view comes from indications that some North African late Pleistocene crania are phenetically similar to modern sub-saharan samples (Bräuer and Rimbach 1990), while studies of dental morphology suggest that late Pleistocene Nubians were closely related to modern sub-saharan samples, but may have been replaced by populations with western Eurasian dental features (Turner and Markowitz 1990; Irish and Turner 1990).

ESR results presented here reinforce the view that northern Africa may have been an important area in the evolution of Homo sapiens, since they indicate a greater antiquity than generally believed (possibly > 130 ka) for the hominids from Irhoud and Singa. If the age estimates are approximately correct, then these fossils (which appear to show predominantly primitive features) could after all represent ancestral populations for modern humans, rather than relict groups. Such a view would be further supported if indications of a greater age (> 70 ka) for North African Aterian sites can be confirmed (Wendorf et al. 1990), which could in turn affect age estimates for the Aterian-associated, but anatomically modern, Dar-es-Soltane material (Ferembach 1976; Bräuer and Rimbach 1990). What is also required, and may be achievable in the near future, is further dating work for other critical African hominin sites such as Omo-Kibish (Day and Stringer 1991; Day et al. 1991), Eliye Springs (Bräuer and Leakey 1986), East Turkana (Guomde)(Bräuer et al. 1991), Ngaloba (Day et al. 1980), Cave of Hearths (Tobias 1968), Broken Hill (Stringer 1986) and Florisbad (Clarke 1985).

Turning eastwards, ESR dating has as yet made little impact on our understanding of modern human origins in the Far East and Australasia. ESR dating has been applied to some Chinese hominid sites (see e.g. Brooks and Wood 1990) but, unfortunately, some age estimates were carried out on bones and teeth without determining many dose rate parameters (e.g. Ikeya 1985) or were obtained by U-series dating of bones (Chen 1990), a method that is even more susceptible to open system behaviour of uranium than ESR dating. ESR dating on bone has also been used to estimate the age of the controversial WLH-50 calvaria from the Willandra Lakes region of southern Australia (Caddie et al. 1987). TL dating of sediments (not burnt flint) has been used more extensively in both regions and has made an important impact through indications of an early (> 50 ka) initiation of the human colonization of Australia (Roberts et al. 1990). However, many sites, especially in China and Indonesia, lack any independent chronological control and the potential for ESR dating is enormous (Stringer 1990a and b).
On the methodological side, the ESR dating technique has to be further refined, especially in view of the occasional large uncertainty that is introduced by the unknown uranium uptake. We hope that the combined U-series/ESR approach (Grün et al. 1988a) can be successfully developed into a routine technique.

We look forward to the next decade which promises to be every bit as exciting for studies of recent human evolution as the last one.

ACKNOWLEDGEMENTS

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APPENDIX I: SAMPLE COLLECTION, PREPARATION AND ESR MEASUREMENT

As can be seen in the dose rate section, the sample is irradiated from the sediment up to 30 cm by gamma rays and up to 2 mm by beta rays. In order to eliminate some of the effect of U-uptake, enamel samples are often cut from the surface of the tooth so that half of the external beta dose rate comes from the sediment which is not affected by U-uptake (see Figure 1(E)). Therefore, the tooth must be collected with the sediment attached to it. Teeth with relatively thick enamel layers (e.g. hippopotamus, mammoth, rhino) are generally better suited for dating than those with thin layers (e.g. deer). In practice, many archaeological sites contain abundant bovid or horse teeth which are quite suitable for ESR analysis.

Since single radiometric dates are usually meaningless, several samples (5-30) should be taken with stratigraphic control. The external gamma dose rate ought to be measured in situ with thermoluminescence dosimeters or a portable multi-channel gamma spectrometer. This also has the advantage that measurements include the present-day water content (although of course it is the average value over the burial period that is required). Another way to reconstruct the gamma dose rate is to collect representative sediment samples around the site which are then measured with a high resolution Ge-detector. This allows the detection of disequilibria in the U-decay chain. However, when working on museum collections from sites which have been extensively excavated (e.g. Skhul and Tabun), the only way to reconstruct the gamma dose rate is to analyse several surviving sediment samples from one archaeological layer and average these results. In some cases the sediment may have been homogeneous, in other cases this procedure introduces large uncertainties.

In the laboratory the tooth is cut with a diamond drill, then enamel and dentin are separated. Since the dose rate calculation is rather complex, two to four different pieces are prepared separately. This allows a check on the reproducibility of the results. The outer 100 μm of the enamel is removed with a diamond drill to cut off the volume which received an external alpha dose. For the calculation of the beta ray attenuation, the thickness of the enamel layer must be measured before and after this process. The enamel is then ground and passed through a sieve (≈ 150 μm) for homogenization. It has been shown by Desrosiers et al. (1989) that only very extensive grinding leads to the generation of radicals. Therefore, the enamel can be ground in a mortar, but mechanical devices should not be used. Ten to 15 aliquots are weighed and irradiated. If the AD is approximately known (from other chronological evidence), the irradiation steps are chosen to cover a dose range of about five to 10 times the expected dose. If the AD is not known, the sample is irradiated with exponential steps up to about 10 kGy. Since the signal at g = 2.0018 may be interfered with by short-living organic signals (Houben 1971; Caddie et al. 1985) and crystal-surface interactions have been observed (Ostrowski et al. 1974), there should be at least a one-week delay between irradiation and ESR measurement. Other laboratory influences such as light exposure do not seem to influence ESR behaviour.

The sample has to be positioned in the middle of the cavity. When measuring powders, the signal intensity is crucially dependent on the packing of the powder (see Wieser et al. 1985). It is advantageous to measure a sample
several times by repeating the compaction in the sample tube. Samples are measured at room temperature with a modulation amplitude of about 0.5 mTpp (in order to eliminate interferences from organic signals), at a microwave power of about 2 mW (the measurements of Hoshi et al. (1985) imply ESR saturation effects at higher microwave powers).

Each piece of enamel and the attached dentin layers are analysed for uranium. If the teeth were collected in the field, the water content of the dentin should be measured. If the enamel layer was in contact with the sediment, this is analysed for U, Th, K, and water. If the gamma dose rate was not measured in situ, a representative sediment sample must be analysed for U, Th, K (and Rb), and water. If possible, disequilibria in the U-decay chain are also measured. These analytical results are then used in the age calculation. It seems valid to assume an alpha efficiency of 0.15±0.02 since all measured values are very close to this value. An assumed $^{234}\text{U}/^{238}\text{U}$ ratio between 1.2 and 1.4 does not introduce a large source of error. Ages are then calculated for early and linear U-uptake.

The explicit formulae for age calculation, and graphs for attenuation of beta and cosmic rays, have been given by Grün (1989a and b).

APPENDIX II: ERRORS IN ESR DATING

The error in AD determination derives:

(i) from the calibration of the source;
(ii) from the scatter of the data points around the extrapolation curve; and
(iii) from the use of a mathematical function for extrapolation which is only approximately correct.

Errors in the calibration of the gamma source are routinely in the range of 2 to 5%. However, if basic gamma dosimetric rules are disregarded (such as the establishment of a charged particle equilibrium between sample and sample containment, see Attix 1968), this error may be much higher. The error in AD determination should be in the range of 5 to 10%. In some cases it may be better, in others worse. Old samples normally show larger scattering than young samples. The third form of error may represent a large source of uncertainty. For example, one can observe systematic overestimations in the 25% range when linear fitting is applied instead of exponential fitting in the so-called 'linear part of the growth curve' (see Figure 4).

In the best case, one may get a systematic error for source calibration in the 2% range and a fitting error in the 3% range, resulting in an AD-uncertainty of about 4%, disregarding mathematical problems. In routine measurements, a minimum error of about 5 to 7% seems far more realistic.

The uncertainty in dose rate estimation derives from:

(i) laboratory uncertainties for measurements of radioactive isotopes;
(ii) reproducibility of these measurements;
(iii) assumption or variation of the k-value (alpha efficiency);
(iv) unknown mode of U-uptake;
(v) calibration of the gamma spectrometer;
(vi) assumption or variation of the water contents;
(vii) U-series disequilibria in sample and sediment;
(viii) variation of cosmic dose rate; and
(ix) mobilization of Rn and Ra.

Since the total dose rate is normally composed of a complex arrangement of radioactive sources, the influence of a given individual uncertainty on the error in age may vary to a large extent. For example, an assumed water content of 10±10% in a sediment may cause an error of about 10% in age when the external gamma and beta dose rate make up more than 90% of the total dose rate. On the other hand, when the external gamma and beta dose rates contribute only 10% to the total dose rate, the same uncertainty causes only a negligible 1% error.

Although the given laboratory uncertainties for the determination of U, Th, and K are usually very low (<1%), the reproducibility may be far worse. Since it is difficult to determine the alpha efficiency by ESR, this parameter is often assumed. All measured results for enamel were very close to 0.15±0.02. The calibration error for a gamma spectrometer is in the 5% range. The water content is a critical value and may have changed in the past. Aitken (1985) gives a typical variation of 7% for archaeological sites. Other effects such as Ra and Rn mobilization and Ra excess in dentin and enamel are rarely measured and may be a considerable source of uncertainty in some samples (see Papastefanou and Charalambous 1978). The cosmic dose rate may change by a variation of the sediment thickness above the sample (erosion or sedimentation).
Table 1 Influence of typical uncertainties ($\pm \sigma$) on age calculation

<table>
<thead>
<tr>
<th>Tooth with low U-concentrations</th>
<th>Early U-accumulation</th>
<th>Linear U-accumulation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Calculated age (a)</td>
<td>% deviation from mean</td>
</tr>
<tr>
<td></td>
<td></td>
<td>+</td>
</tr>
<tr>
<td>AD (Gy)</td>
<td>33.0 ± 1.5</td>
<td>103000</td>
</tr>
<tr>
<td>U (ppm)</td>
<td>0.05 ± 0.01</td>
<td>98600</td>
</tr>
<tr>
<td>$^{234}$U $^{238}$U</td>
<td>1.4 ± 0.4</td>
<td>99300</td>
</tr>
<tr>
<td>Alpha efficiency</td>
<td>0.15 ± 0.02</td>
<td>98200</td>
</tr>
<tr>
<td>U(SED) (ppm)</td>
<td>1.17 ± 0.01</td>
<td>97800</td>
</tr>
<tr>
<td>Th(SED) (ppm)</td>
<td>0.05 ± 0.05</td>
<td>97700</td>
</tr>
<tr>
<td>K(SED) (%)</td>
<td>0.318 ± 0.001</td>
<td>97700</td>
</tr>
<tr>
<td>Water (SED) (%)</td>
<td>10 ± 10</td>
<td>99600</td>
</tr>
<tr>
<td>Cosmic dose rate (μGy a)</td>
<td>243 ± 15</td>
<td>102000</td>
</tr>
<tr>
<td>U(DE) (ppm)</td>
<td>1.77 ± 0.01</td>
<td>97800</td>
</tr>
<tr>
<td>Water (DE) (%)</td>
<td>5 ± 5</td>
<td>98100</td>
</tr>
<tr>
<td>Thickness (μm)</td>
<td>1700 ± 200</td>
<td>99600</td>
</tr>
<tr>
<td>S1, S2 (μm)</td>
<td>100 ± 60</td>
<td>98100</td>
</tr>
<tr>
<td>Mean age</td>
<td>97700</td>
<td></td>
</tr>
<tr>
<td>Total error</td>
<td></td>
<td>105000</td>
</tr>
<tr>
<td>50% Rn-loss (tooth)</td>
<td></td>
<td>99900</td>
</tr>
</tbody>
</table>

Tooth with high U-concentrations

| AD (Gy)                          | 490 ± 40              | 84200   | 73700   | 6.6      | 6.8      | 139000  | 121800  | 6.9      | 6.9      |
| U (ppm)                          | 5.21 ± 0.01           | 79000   | 79000   | 0        | 0        | 130000  | 130000  | 0        | 0        |
| $^{234}$U $^{238}$U              | 1.4 ± 0.4             | 85700   | 73700   | 1.5      | 6.8      | 140000  | 122000  | 7.7      | 6.2      |
| Alpha efficiency                 | 0.15 ± 0.02           | 80800   | 77400   | 1.8      | 2.0      | 133000  | 128000  | 2.3      | 1.5      |
| U(SED) (ppm)                     | 2.95 ± 0.01           | 79000   | 78800   | 0        | 0.2      | 130000  | 129000  | 0        | 0.8      |
| Th(SED) (ppm)                    | 3.0 ± 0.1             | 79100   | 79000   | 0.1      | 0        | 130000  | 130000  | 0        | 0        |
| K(SED) (%)                       | 1.87 ± 0.001          | 79000   | 79000   | 0        | 0        | 130000  | 130000  | 0        | 0        |
| Water (SED) (%)                  | 10 ± 10               | 80600   | 77500   | 2.0      | 1.9      | 135000  | 126000  | 3.8      | 3.1      |
| Cosmic dose rate (μGy a)         | 150 ± 50              | 79500   | 78500   | 0.6      | 0.6      | 132000  | 129000  | 1.5      | 0.8      |
| U(DE) (ppm)                      | 184 ± 1               | 79200   | 78800   | 0.2      | 0.2      | 130000  | 130000  | 0        | 0        |
| Water (DE) (%)                   | 5 ± 5                 | 81400   | 76700   | 3.0      | 2.9      | 134000  | 127000  | 3.1      | 2.3      |
| Thickness (μm)                   | 1170 ± 150            | 81800   | 76500   | 3.5      | 3.8      | 135000  | 125000  | 3.8      | 3.8      |
| S1, S2 (μm)                      | 100 ± 60              | 79500   | 78600   | 0.6      | 0.5      | 131000  | 129000  | 0.8      | 0.8      |
| Mean age                         | 79000                 |          |          | 12.0     | 11.0     | 130000  |          | 12.4     | 10.9     |
| Total error                      |                       | 12.6     |          | 144000   |          | 10.8    |          |          |          |
| 50% Rn-loss (tooth)              |                       | 89000    | 12.6     |          |          | 140000  |          | 10.8     |          |
Table 1 shows the influence of the analytical results on age calculation. The first example shows a tooth with relatively little uranium. The total error is in the range of 8% and this can indeed be achieved (see example from Border Cave). In cases where the U-concentration is high, parameters which are connected with it (e.g. the $^{234}$U/$^{238}$U-ratio) and the unknown U-uptake cause the major source of uncertainty. An assumed Rn-loss of 50% would have only a minor influence on the first set of ages (<2.5%), but significantly changes the results of the second set, where the ages would be about 11% older.

For the ages shown in the diagrams above, the error in AD was determined by jackknifing (Grün and Macdonald 1989). Other assumed parameters were: alpha efficiency 0.15 ± 0.02; $^{234}$U/$^{238}$U = 1.2 ± 0.1, water in sediment: 20 ± 10% (in most cases). The analytical data of the results presented in this paper are published elsewhere and the reader is referred to the respective publications for more details.

It has to be emphasized that ESR age estimates based on assumed total dose rates or linear fitting (especially if the AD is greater than 100 Gy) are very likely to give extremely erroneous results, even when the site is of great importance and the ESR results are 'in good agreement with . . .' (see also Grün 1989c and 1991).

REFERENCES


Attix, F. H., 1968, Basic γ-ray dosimetry, Health Physics, 15, 49–56.


Clark, J. D., 1975, Africa in prehistory: peripheral or paramount?, Man, 10, 175–98.


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ESR dating and the evolution of modern humans


Steiner, W., 1979, *Der Travertin von Ehringsdorf und seine Fossilen*, A. Ziemsen, Wittenberg.
R. Grün and C. B. Stringer


ESR dating and the evolution of modern humans


