THERMOLUMINESCENCE DATING AND ITS EXTENSION INTO THE PALAEOLITHIC

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Introduction

After demonstrations (Grogler et al., 1960; Kennedy and Knopff, 1960) of feasibility around 1960 thermoluminescence came to fruition as a dating technique (Aitken and Fleming, 1972) during the seventies. This was in respect of archaeological pottery, bricks and tiles and also the authenticity testing of art ceramics. The latter application had a particularly powerful impact, creating a 'TL revolution' in respect of some ranges of museum exhibit (see, for instance, Aitken, Moorey and Ucko, 1971; Fleming, Jucker and Riederer, 1971). In broad terms the accuracy attainable for archaeological dating is between 5 % and 10 % of the age when satisfactory assessment of the gamma dose-rate from the soil can be made, and between 20 % and 30 % of the age for authenticity testing; half-a-dozen sherds each 2 or 3 cm across are required for the former but only 50 milligram is required for the latter. Extension to burnt flint from palaeolithic sites was initiated (Göksu et al., 1974) in the mid seventies thereby giving the possibility of reaching beyond the ten thousand years during which pottery was available and eventually beyond the fifty thousand year limit of radiocarbon. This latter limit had meant that for important periods of man's development during the glacials and interglacials of the Middle and Lower Palaeolithic there was no direct dating available (later this gap was partly filled, in addition to TL, by uranium series dating of stalagmitic calcite where it was in close association with man's occupation, and by the somewhat problematical amino acid dating of bone). Hence the TL dating of burnt flint (e.g. Wintle and Aitken, 1977; Valladas, H., 1978; Huxtable and Jacobi, 1982; Bowman, 1982), and other extensions such as to speleothems (Debenham and Aitken, 1984) and volcanic lava (Guérin and Valladas, 1980) are of substantial importance in palaeolithic archaeology. Equally they are important in the whole multidisciplinary field of quaternary research and particularly relevant here is the revolutionary concept of using TL to date unburnt sediment. Stemming from work in the USSR it now appears feasible to date windblown sediment and some categories of waterborne sediment on the basis that the TL had been set to zero at deposition; by measuring the TL that has accumulated due to natural radiation dosage during burial the same age equation, viz:—

$$\text{Age} = \frac{\text{Accumulated TL}}{(\text{TL per unit dose}) \times (\text{dose per year})} \tag{1}$$

can be applied as for burnt materials where of course the $t = 0$ event is the firing by ancient man. The zeroing mechanism in the case of sediment is not fully established but certainly exposure to sunlight (e.g. while airborne, in the case of windblown sediment) is the most favoured candidate, with the possibility of weathering and glacial grinding being responsible in some circumstances.

In this paper we give a short summary of the TL technique with particular emphasis on its application to burnt flint and sediment from palaeolithic sites in western Europe. For fuller information concerning the technique and its applications the reader is referred elsewhere (see for instance: Aitken, 1974; Fleming, 1979; Mejdahl and Wintle, 1984; Wintle and Huntley, 1982; Aitken, 1985).

The TL technique

Thermoluminescence (TL) is a phenomenon exhibited to varying degrees by many minerals. Essentially it is the emission of light when a substance is heated, this light being additional to ordinary red-hot glow and usually occurring at a less elevated temperature. In so far as archaeological application is concerned the light is faint and a special high sensitivity photomultiplier is needed for measurement.

TL represents the release of energy that has been stored in the crystal lattice of the mineral, this energy being in the form of trapped electrons. These trapped electrons are the result of exposure to nuclear radiation and there is a weak flux of this from the radioelements (potassium-40, thorium and uranium) naturally present in minerals and soil. The basic notion of TL dating is that at the time of the event being dated the latent TL of the sample was effectively zero. Then during the millennia of burial the TL accumulated so that its intensity to-day is related to the age. The erasure of geologically-acquired TL at the time of the archaeological event is an obvious essential for dating. In the case of burned flint it is the action of heat that achieves this, a temperature of around 400°C being needed. With sediment it is the action of sunlight in 'bleaching' the TL before deposition; windblown sediment such as loess will have had a long exposure to sunlight while airborne and it appears that some types of waterborne sediment have been sufficiently exposed also. In stalagmitic calcite it is the formation of the calcite crystal that is being dated.

The basic equation is:—

$$\text{Age} = \frac{\text{Paleodose}}{\text{Annual radiation dose}}$$

The PALEODOSE is evaluated from laboratory measurements of the archaeological-accrued TL (the natural TL) and the sensitivity of the particular sample concerned, viz:—

$$\text{Paleodose} = \frac{\text{Natural TL}}{\text{TL per unit dose of nuclear radiation}}$$

Thus paleodose represents the total dose that the sample must have received since the event being dated. Alternative terminology is equivalent dose (ED) and archaeological dose (AD).

The ANNUAL DOSE, or DOSE-RATE, is determined by both laboratory and on-site measurements. It consists of two parts, the INTERNAL dose-rate from radioelements in the sample itself, and the EXTERNAL dose-rate from radioelements in the burial soil (up to a distance of about 0.3 metre from the sample). For flint and calcite the external component is often as much as 80 % of the annual dose and consequently the reliability of the age can be strongly dependent on an accurate evaluation of it. This component is alternatively called soil dose, environmental dose, gamma dose and gamma-plus-cosmic dose; the cosmic-ray contribution is typically about 20 % of the external component, less in deep caves.

For on-site measurements we use (i) TL capsules and (ii) a portable gamma spectrometer. A TL capsule, usually of copper, contains a highly-sensitive TL powder and is buried in the soil in a situation that represents that of the sample as closely as possible; it is about 8 mm diameter and 40 mm long. Minimum burial time is a few months, a year being preferred; it is placed at the end of a 0.3-metre long auger hole. The detector of the gamma spectrometer is 65 mm in diameter so that a correspondingly larger hole is required; however in this case the measurement time is only half-an-hour.

The annual dose is influenced by the water content of sample and soil during antiquity. An upper limit to the effect is obtained by measuring the saturation content. The as-dug content can also be measured (using a tightly-tied plastic bag to avoid evaporation during transportation to the laboratory); it is then a matter for discussion as to whether the average during burial was bigger or smaller than the as-dug value. Uncertainty about water content is one of the factors that limit the accuracy attainable and the collaboration of a sedimentologist is important.

The maximum age that can be reached is dependent on the TL characteristics of the sample, its radioactivity, and the radioactivity of the soil. In round terms both flint and calcite can reach about half-a-million years, perhaps more; the limit for sediment is not yet established. The accuracy obtainable varies with circumstances, often being limited by uncertainty in water content. It is realistic to expect ± 10 % of the age; somewhat better can sometimes be achieved but it is unlikely that the error limits will ever be reduced below ± 5 % of the age. Although the accuracy is not high it is usually adequate for deciding in which of several possible isotope stages a site should be placed.

The TL dating of burnt stones (other than flint) from Neolithic and later sites in Scandinavia has been undertaken extensively at the Nordic TL Dating Laboratory and V. Mejdahl gives an account of this in another paper at this meeting. It is also possible to use burnt stones in the Upper Palaeolithic and this has been done with success by Valladas (1978) on Magdalenian sites for instance. However the maximum age attainable with burned stones other than flint is rather limited. This is because of higher radioactivity and a lower level of TL saturation. Sediment dating of geological sites in Europe is being extensively developed by A. Wintle at Cambridge (see, for example, Wintle, Shackleton and Latridou, 1984). Dating of speleothems (stalagmitic calcite) in palaeolithic caves is also possible with TL (Debenham and Aitken, 1984); an allied method of dating for this material is by electron spin resonance (ESR) and this is being applied extensively to geological sites by Hennig and Grun (1984). Calcite can also be dated by the longer-established uranium-series techniques, though this technique does not extend much beyond 300 ka.

TL dating of flint samples

Pieces of flint which have been burned sufficiently to remove their geological TL by ancient man can be used for TL dating. There are two main problems to be considered; sample size and external environment.

The internal radioactive dose from most of the flints in this program has been very low so that the external dose contributes 80 % or more to the total dose which the flint receives. The importance of being able to monitor the exact environment of the flint being tested using TL dosimeters cannot be overemphasised. At the boundary between

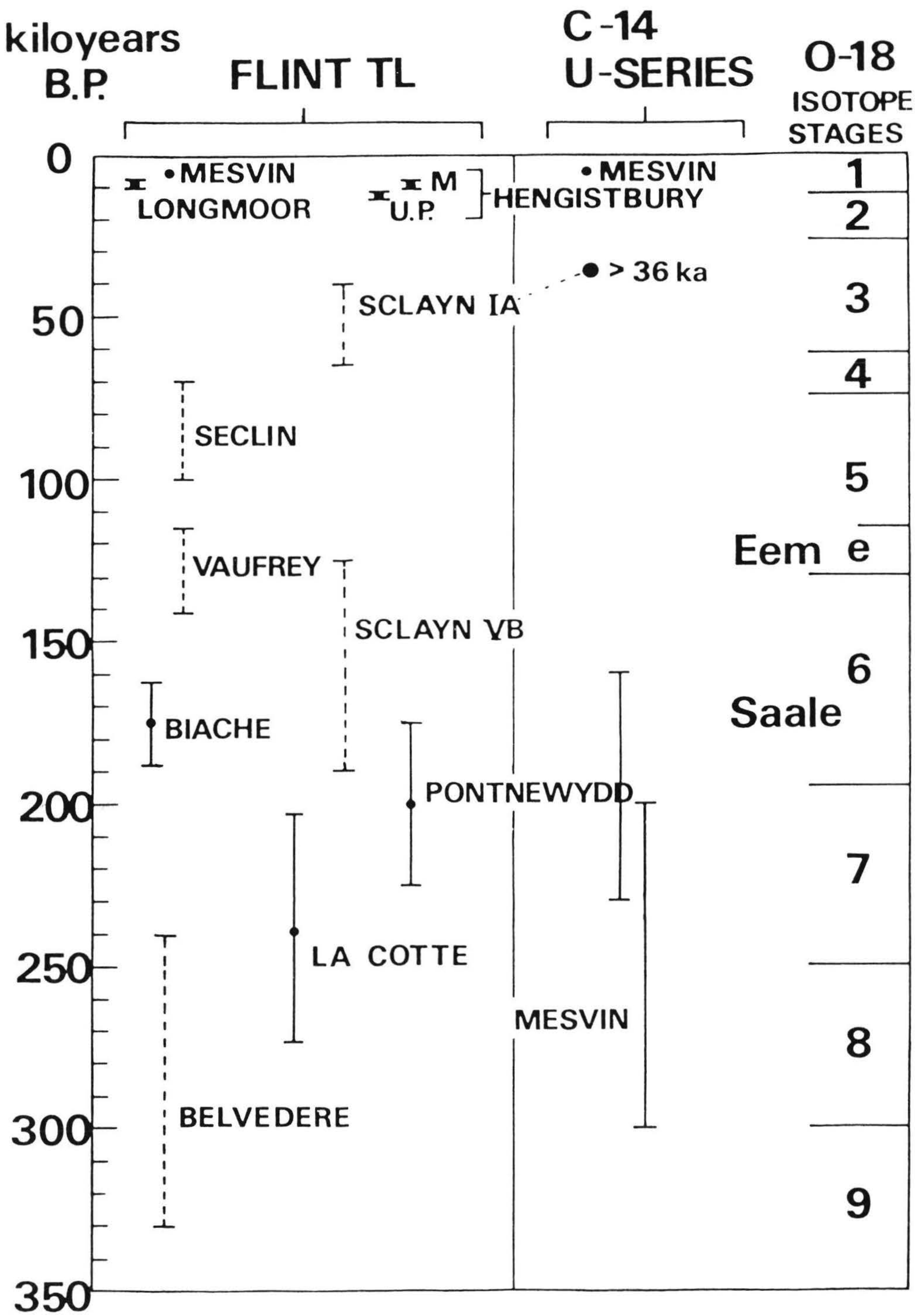

Fig. 1. TL results for various European sites displayed in conjunction with uranium series and carbon 14 dates where these are available.

the piece of flint of low radioactivity and the soil of usually much higher radioactivity, there is a layer of flint which has had an uncertain dose. This uncertainty extends about 2 mm into the flint, and so the routine practice is to remove and discard the outer 2 mm of the flint to be dated. The remainder of the flint is crushed, in the jaws of a vice, to produce the two sizes of grains used for dating, 90—150 microns, and less than 8 microns. The grains produced are treated with dilute acid before they are used. Because of the removal of the surface layer, and the method of sample preparation one needs a disc of flint roughly 3 cms × 1 cm thick to begin with, otherwise enough fine grains are not produced by the crushing process.

The results of several years application of TL dating to burned flints from European sites are displayed in Figure 1. It is to be emphasized that these TL dates are absolute in the sense of being based only on laboratory measurements and on-site radioactivity measurements. Hence they are essentially a 'physicist's view' of palaeolithic chronology and quite independent of other interpretations of the sites. Figure 1 also shows results obtained from uranium series and carbon-14 dating for the sites where these are available; in general there is satisfactory agreement, as also with the expectations from chronostratigraphic studies.

Sediments

Sediments contain, in varying proportions, a wide range of minerals. However, only two types contribute appreciably to the TL emissions, viz. feldspars and quartz; both these minerals have been used with success. Work at Oxford has been concentrated on one particular component of the feldspar TL which is advantageous for this method of dating because of its rapid and almost total erasure on exposure to sunlight. This property gives a high degree of assurance that, at the time of the sediment's deposition, this TL is effectively zero or at a manageably low level. The isolation of this signal is a simple matter because it is the only sediment emission found in the ultra-violet part of the spectrum. It can therefore be selected by placing a UV transmitting filter in front of the photomultiplier tube, avoiding the need for previous mineral separation.

Figure 2 shows the method for determining the archaeological dose using this TL signal. The natural TL, as a function of temperature, is given by curve e, while the effect of a sunlight exposure is shown by curve a. Curves b, c and d result from the administering of different laboratory radiation doses to samples which had previously been bleached. It is generally found, by comparing the growth rates (versus radiation dose) of TL in the natural and bleached samples, that no change in TL sensitivity is brought about by the sunlight exposure. Then, the archaeological dose is simply the radiation dose which, given to the bleached sample, regenerates the same amount of TL as found in the natural sample. From Figure 2, it is seen that matching of the natural and regenerated glow curves occurs only above 270°C. This is because the natural sediment has, over the archaeological period, lost trapped electrons from the less stable traps which give rise to the lower temperature TL. Above 270°C, however, a unique value of the archaeological dose is obtained, which, in the example of Figure 2, is of the order of 180 Grays.

The time range over which the technique gives meaningful results is limited by (i) the saturation level of the TL, and (ii) the stability of the measured signal. While the growth of TL is non-linear over the dose range 0—1000 Grays, saturation is still not reached at the upper limit. This would, by itself, imply an age range back to at least about 300 ka, assuming typical dose-rates. However, the stability of the TL signal appears to set a lower bound. Although the effect of thermal drainage of the trapped

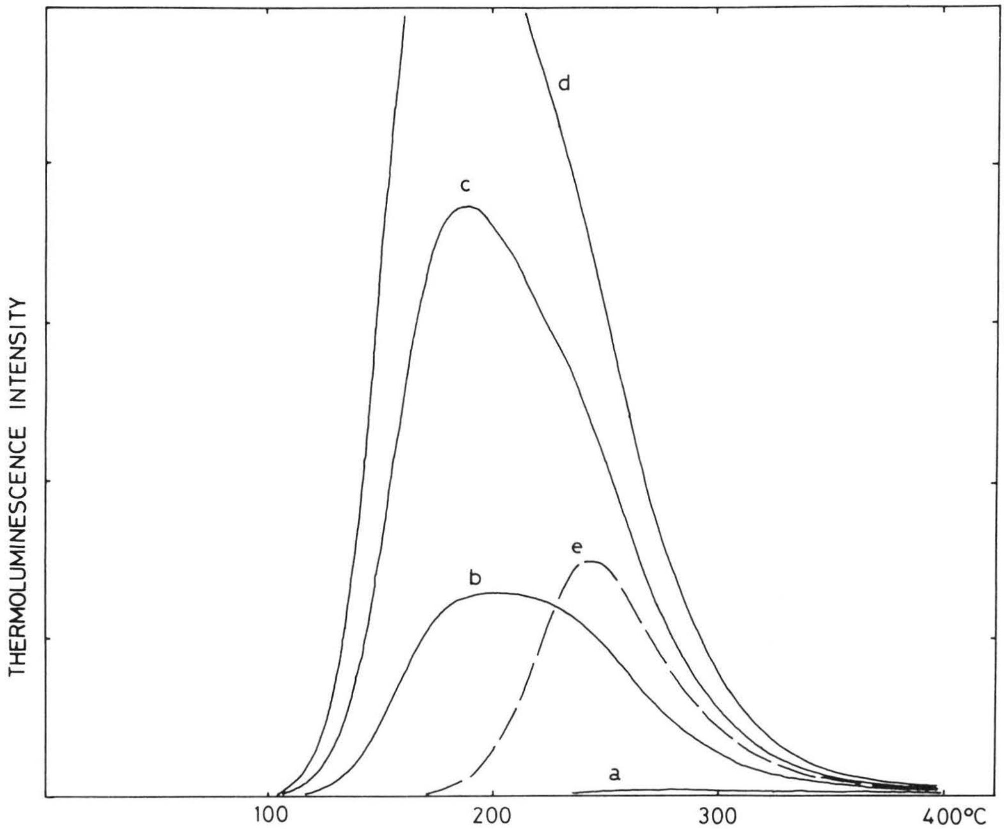

Fig. 2. Glow curves of a sediment sample, showing (a) the level of TL following exposure to sunlight; (b), (c) and (d) the regeneration of TL in samples exposed to sunlight by radiation doses of 85 Grays, 255 Grays and 510 Grays respectively. Glow curve (e) is that of the natural, as excavated, sediment.

electron populations from low temperature traps is clearly seen (Figure 2), there remains the possibility of a general fading of TL levels which occurs to equal degrees at all temperatures. By its nature, this latter type of fading is difficult to detect, but shows up strongly in old samples. From the results obtained from the older contexts so far investigated it appears that an instability of this type is present, at any rate with the fine grains (1—11 micron) presently used for dating (see Debenham 1985 for a full discussion). The upper limit that it imposes on the dating range probably lies in the region of 50—100 ka.

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