# INFLUENCE OF GROUND TEMPERATURE VARIATIONS AND OTHER FACTORS ON THE ACCURACY OF THERMOLUMINESCENCE DATING METHODS BASED ON THE SHAPE OF GLOW CURVES

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(Received 23 July 1986)

Abstract—The effect of ground temperature variations with time and depth as well as of other complicating factors are examined, with reference to the thermoluminescence dating methods which use the shape of glow curves as affected by ambient temperatures. It is shown that for accurate dating, detailed information is needed on the variation of temperature with time at the depth that the object being dated was found. Taking into account all the sources of error, a total uncertainty of around 20% in the ages is expected. This makes these methods of little use to archaeological dating but possibly of use in authenticity tests.

#### 1. INTRODUCTION

THE COMMONLY used methods of dating by thermoluminescence (TL) are based on thermally stable peaks in the glow curves. However, low-temperature peaks which are affected by ambient temperatures have been used in studies of the thermal or radiation histories of samples (Ronca, 1964; Johnson, 1965; Ronca and Zeller, 1965; Ronca, 1968; Christodoulides et al., 1971; Christodoulides, 1971; Durrani et al., 1972, 1976, 1977; Wintle and Aitken, 1977)

Peaks that had reached an equilibrium height, with the rate of trap filling by radiation being exactly offset by thermal draining at ambient temperature, have been used in determining this temperature when the dose rate was known or vice versa (Christodoulides, 1971; Durrani *et al.*, 1972).

Dating methods were also developed, which were based on low-temperature peaks for the determination of the dose rate or to avoid the need for having to determine the dose rate (Charalambous and Michael, 1976; Langouet *et al.*, 1976, 1979, 1985).

It is the purpose of this paper to consider the limitations of these methods, imposed by various factors, with special emphasis on the effects of the variation of ground temperatures.

#### 2. THEORY

In deriving the equations governing the growth of TL with dose when thermal drainage is taken into account, certain simplifying assumptions have to be made. The main assumptions are that traps are not destroyed or created by radiation, the rate of filling

the traps responsible for the TL peak under consideration is proportional to the number of empty traps and the dose rate, and that thermal drainage is governed by an Arrhenius expression as described below. Competition between traps is neglected and so is retrapping of liberated charges. With these assumptions, the mathematics is fairly simple (Christodoulides, 1971; Christodoulides *et al.*, 1971).

#### 2.1. Constant temperature and dose rate

If the phosphor is kept at a constant temperature T and irradiated at a constant dose rate r, and if N is the total number of traps per unit volume, of which n are filled at a given time, E is the energy depth of these traps and s their frequency factor, then

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \frac{1}{R_0} (N - n)r - sn \,\mathrm{e}^{-E/kT}. \tag{1}$$

The first term on the right-hand side gives the rate of filling the (N-n) empty traps.  $R_0$  is a constant of the peak and, as shown below, it is the saturation dose when thermal drainage is negligible. The second term gives the rate of draining the n filled traps by thermal excitation at temperature T.

Integrating (1) with all the traps empty at t = 0 and using the dose R = rt as variable, we obtain:

$$n = \frac{N}{\alpha} \left[ 1 - \exp\left(-\alpha R/R_0\right) \right] \tag{2}$$

with

$$\alpha = 1 + \frac{sR_0}{r} e^{-E/kT}.$$
 (3)

The parameter  $\alpha$  is large when thermal drainage is important and becomes equal to 1 when the

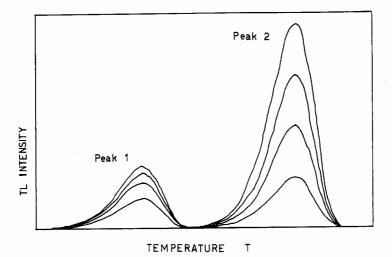


Fig. 1. Hypothetical glow curves for doses equal to 1, 2, 3 and 4 times a certain dose. The glow curve consists of two peaks, a low-temperature peak (1) and a high-temperature peak (2). Peak (2) increases in height linearly with dose, while peak (1), which is affected by thermal drainage, shows a sublinear variation of height with dose and signs of saturation.

irradiation temperature T is low enough for drainage to be negligible. When this happens, equation (1) becomes

$$n = N(1 - e^{-R/R_0}) (4$$

where the growth of n is given by a saturating exponential with saturation dose  $R_0$ .

Equation (2) shows that thermal drainage has the effect of decreasing the maximum number of traps that can be filled by a factor  $\alpha$ , and also decreasing  $R_0$  by the same factor. We may write

$$n = n_s (1 - e^{-R/R_s}) (5)$$

where

$$n_s = N/\alpha$$
 and  $R_s = R_0/\alpha$ .

Figure 1 shows a hypothetical glow curve consisting of a low-temperature peak which is affected by thermal drainage and a high-temperature peak which is not affected. Assuming that the TL intensity is proportional to n, which means that we again disregard retrapping and competition between traps during heating, we obtain the response curves of Fig. 2 for the two peaks, at three different temperatures. It is obvious that for a given dose rate, for higher temperature of irradiation we have a lower saturation dose  $R_s$  and saturation level  $L_s$ .

# 2.2. Periodically varying temperature

If the temperature of the sample changes during irradiation, we have variations in thermal drainage. For a complete solution, the exact value of tem-

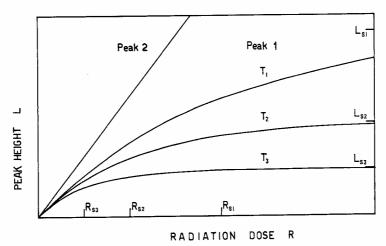


Fig. 2. The variation with dose of the heights of two peaks such as those shown in Fig. 1. Peak (2) which is not affected by thermal drainage is assumed to increase in height linearly with dose. Three different cases are shown for peak (1) which is affected by thermal drainage. The three cases correspond to three different temperatures  $T_1$ ,  $T_2$  and  $T_3$  in increasing order. To these temperatures there correspond saturation heights  $L_{s1}$ ,  $L_{s2}$  and  $L_{s3}$  with saturation doses  $R_{s1}$ ,  $R_{s2}$  and  $R_{s3}$  respectively.

perature as a function of time is needed. However, if the variation is periodic with a small period compared to the age of the sample, an approximate solution to the problem may be found. Over a time t, assuming that n does not change significantly, we have a change  $\delta n$  in n due to thermal drainage, where

$$\delta n \approx -sn \int_0^t e^{-E/kT} dt.$$
 (6)

We define a temperature  $T_B$  such that

$$e^{-E/kT_B} = \frac{1}{t} \int_0^t e^{-E/kT} dt$$
 (7)

which we call the Boltzmann-equivalent temperature. Assuming n changes by a small proportion over the time period t,  $T_{\rm B}$  if kept constant would have had the same draining effect on the peak as the variable temperature did. If T(t) has a period  $t_{\rm p}$  we take the average value of the Boltzmann exponential in equation (7) over a period  $t_{\rm p}$ .

In the case of periodically varying temperature, equation (1) may be rewritten as

$$\frac{\mathrm{d}n}{\mathrm{d}t} = \frac{1}{R_0} (N - n) r - sn e^{-E/kT_\mathrm{B}} \tag{8}$$

with the assumptions mentioned above. This equation describes the variation of n with time on a long time scale compared to  $t_p$  and neglecting the small fluctuations of n, having a period of  $t_p$ . The solution of equation (8) is identical to that of equation (2) with  $T_B$  replacing T.

#### 2.3. Periodically varying dose rate

If r varies periodically with time, with a period small enough so that n does not change significantly over a period, then we may use in equation (1) the average value of r. If both T and r vary with time, having the same period  $t_p$ , the average values of r and  $T_B$  used in equation (2) should be estimated over a period  $t_p$ . If their periods are different, the shortest interval of time being an integral multiple of both periods should be used as averaging time.

# 3. TL DATING METHODS BASED ON THE SHAPE OF GLOW CURVES

There are three methods of TL dating based on the theory developed above. All three make use of the changes in the shape of a glow curve caused by thermal drainage, to avoid the need for direct determination of the dose rate r. These methods will be briefly described below, and then there will follow a discussion of the characteristics of TL peaks which may be suitable for use in such methods of dating.

#### 3.1. The TL dating methods

Method A. The first method is based on a low temperature peak which has reached an equilibrium height as dictated by the dose rate and the rate of

thermal drainage, In this case, the dose rate may be found, (Christodoulides, 1971; Christodoulides *et al.*, 1971), using the value of  $n_s$  as determined by experiment and solving for r to obtain

$$r = \frac{R_0}{N - 1} s e^{-E/kT_B}.$$
 (9)

In addition to  $n_s$ , one must also determine N,  $R_0$ , s, E and  $T_B$ . The ratio  $N/n_s$  is the ratio of TL peak height when the sample is irradiated at a low temperature to saturation, to the equilibrium peak height as measured in the natural glow curve of the sample. Having determined r and the natural dose received by the sample using a high temperature peak and any of the usual TL dating techniques, the age is found.

Method B. After measuring the natural TL of a sample, a suitable pair of dose rate and temperature is found, such that by irradiating in the laboratory to the natural dose received by the sample, the exact shape of the glow curve is reproduced (Charalambous and Michael, 1976). The condition for this to be achieved is

$$t_{\rm A} \,\mathrm{e}^{-E/kT_{\rm B}} = t_{\rm L} \,\mathrm{e}^{-E/kT_{\rm L}} \tag{10}$$

where  $t_A$  is the archaeological age of the sample,  $T_B$ is the Boltzmann-equivalent temperature for the peak used over the time  $t_A$ ,  $t_L$  is the duration of irradiation of the sample in the laboratory to the natural dose at a constant dose rate and  $T_{\rm L}$  is the temperature of irradiation in the laboratory. This method is based on reproducing the TL vs dose growth curve in the laboratory at a much faster rate than that of the natural irradiation of the sample. This is achieved by imparting to the sample the same dose as its natural dose at a much higher dose rate than that of the natural irradiation and at the same time accelerating the rate of thermal drainage of the low-temperature peak by the same factor, by performing the irradiations in the laboratory at a suitably higher temperature. In this method E and  $T_B$  have to be known.

Method C. This is a method of relative dating by TL, known as DATE (Différence des Atténuations Temporelles des Emissions) (Langouet et al., 1976, 1979, 1985). It is based on determining a parameter g(t) which is proportional to the ratio of the height of the low-temperature peak affected by thermal drainage to that of a high-temperature peak which is not affected by ambient temperature. By laboratory irradiations the ratio of the rates of growth of the two peaks with dose is also found so that g(t) is normalized to unity for zero age (i.e. no thermal drainage).

By determining g(t) as a function of the age t for samples of known age, a calibration curve is obtained relating g(t) to age. A sample of unknown age is dated by comparison of its g(t) value with the calibration curve.

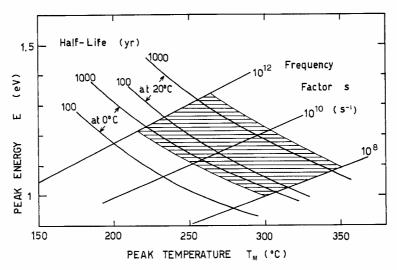


FIG. 3. The range of peak temperatures and peak energies (shaded region) which would exhibit thermal drainage effects at ambient temperatures between 0 and  $20^{\circ}$ C, and correspond to peaks which might be used for dating by the three methods described in the text. The region of interest is contained between the lines of  $s=10^8 \, {\rm s^{-1}}$  and  $10^{12} \, {\rm s^{-1}}$  which is the usual range for the frequency factor. It is also limited between the lines coresponding to half-lives of a few centuries at 0°C and a few thousand yr at 20°C, as these would be the ages of interest in archaeological dating. For geological dating the range of interest is shifted towards higher  $T_{\rm M}$  and E values.

#### 3.2. Choice of suitable TL peaks

Whether or not a TL peak would be suitable for use in the dating methods described above depends on its half-life at ambient temperatures. In dating of archaeological samples the ages range from a few centuries to a few thousand years. Each case must be considered independently, taking into account the method to be used, the expected age of the sample and the temperatures at the site and depth below ground surface at which the sample was found.

Considering a sample of an age of 2000 years and ambient temperatures between 0 and 20°C, some arguments will be presented, which are relevant to the question of deciding the suitability of a TL peak. This will be done referring to Fig. 3, which shows the relation between peak half-life and peak temperature and energy.

If method A is to be used, a peak of half-life no higher than approx. 500 years at  $20^{\circ}$ C must be chosen, so that equilibrium height may be attained by the peak in about 2000 years. For *s* values between  $10^8 \text{s}^{-1}$  and  $10^{12} \text{s}^{-1}$  this means that the peak temperature must be between 250 and 360°C, corresponding to energies of 1.3 and 1.1 eV respectively.

Methods B and C have the same range of applicability. They may be used on peaks that have been affected by temperature irrespectively of whether they have reached equilibrium or not. It is preferable, however, to use peaks not at equilibrium as these are higher and can be measured with more accuracy. This means that their half-lives at  $20^{\circ}$ C may be up to about 5000 years for the case considered, extending the range of peak temperatures up to about  $350^{\circ}$ C for  $s = 10^8 \, \text{s}^{-1}$ .

The shaded region in Fig. 3 shows roughly the ranges of peak temperatures  $T_{\rm M}$  and energies E which are of interest in the three dating methods described. These may be roughly taken to be  $T_{\rm M}$  between 210 and 350°C and E between 1 and 1.4 eV, for archaeological dating. In all cases, however, an additional condition to be satisfied is that the low-temperature peak is high enough to be detectable and measurable with sufficient accuracy.

# 4. GROUND TEMPERATURE VARIATIONS

The effects of temperature variations were studied for various TL peaks and for the temperature at depths up to 1m below ground surface. The temperature data used were for the Athens region and in particular the Nea Philadelphia Meteorological Station.

#### 4.1. Temperature data

The monthly average surface temperature under shade for Athens is shown in Fig. 4(a) for each of the twelve months of the year. The curve is based on data collected between 1952 and 1971. The mean temperature over the year is 17.4°C, the minimum is about 9°C in January and the maximum about 27.5°C at the end of July. Superimposed on these there are the daily variations which on average have amplitudes of 3.5°C in January rising to 7.5°C in July, as found from data collected between 1946 and 1975, Fig. 4(b).

Ground temperatures were available for depths between 2 cm and 1 m, recorded at intervals of 6 h starting at noon every day. The data used in this

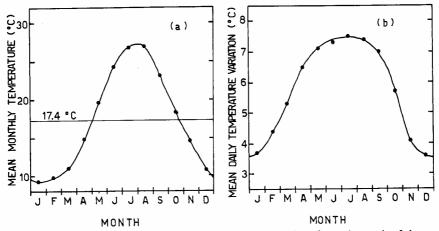


Fig. 4. Surface temperatures under shade, in the Athens region, given for each month of the year. (a) Monthly average temperatures over the period 1952–1971. The mean temperature for this period is 17.4°C. (b) Average values of the amplitude of temperature variation over the period of a day, given for each month, from data collected between 1946 and 1975.

study covered the period of 24 consecutive months between May 1982 and April 1984. The temperatures for the month of May 1982 are shown in Fig. 5 for the depths of 20 cm and 1 m. The picture is also representative of other months and depths. The general features are that for small depths the daily variation is clearly visible, superimposed on longer period fluctuations and the seasonal variation. At depths over 50 cm the daily variations (high-frequency) are not clearly visible, with only the seasonal variation remaining, together with some shorter period variations of small amplitudes.

# 4.2. The Boltzmann-equivalent temperatures

Using the definition of  $T_{\rm B}$  (equation (7)) and detailed temperature data such as shown in Fig. 5, for

depths of 2, 10, 20, 50 and 100 cm, the Boltzmann-equivalent temperatures were determined for each of the 24 consecutive months, as well as for each of the 2 years and for the whole 2-yr period. This was done for peaks of energies 0.8, 1, 1.2 and 1.4 eV. The exact number of days in each month was taken into account.

Figure 6 shows the results for a peak of 1 eV and for the depth of 100 cm.  $T_{\rm B}$  is shown for each month, and varies between 10 and 28°C. For these same data the  $T_{\rm B}$  value for the first 12-month period is 21.0°C and for the second 20.6°C. For the whole 2-yr period it is 20.8°C.

The values of  $T_{\rm B}$  for the depths and peak energies mentioned above are plotted in Fig. 7, for the whole 2-yr period examined. The variation of  $T_{\rm B}$  with

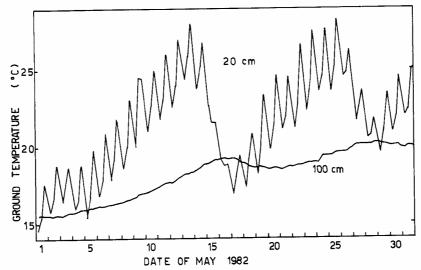


Fig. 5. Variation with time of the ground temperatures at two depths, 20 and 100 cm, for the month of May 1982. Readings are shown at intervals of 6 h. The location was the Nea Philadelphia Meteorological Station near Athens. For the depth of 20 cm the (short-period) daily variation is clearly visible, together with longer-period and seasonal changes. At a depth of 100 cm the daily variation is smoothed-out leaving only the longer-period variations superimposed on the slow seasonal increase of temperature.

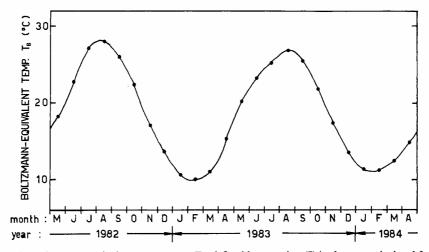


Fig. 6. The Boltzmann-equivalent temperature  $T_{\rm B}$ , defined by equation (7) in the text, calculated for each month between May 1982 and April 1984, for the depth of 1 m and for a peak of 1 eV energy depth. Detailed temperature data such as those shown in Fig. 5 were used in the calculations. The differences in  $T_{\rm B}$  for the same month of the two successive years are 1°C or less. The values given are of the temperatures  $T_{\rm B}$  which if kept constant over a month would have had the same thermal drainage effect on a 1 eV peak as the actual ground temperature did have over that period of time.

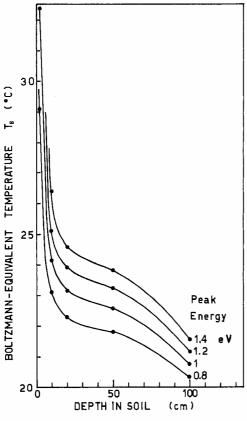


Fig. 7. The Boltzmann-equivalent temperature  $T_{\rm B}$  for the 2-yr period examined, as a function of depth, for peaks of energies 0.8, 1, 1.2 and 1.4 eV. The rate of change of  $T_{\rm B}$  with depth is very high at depths less than 20 cm. At greater depths the rate of change is about 2.5°C/m. These graphs may be used to find the errors in the values of  $T_{\rm B}$  that would be caused by a given error in the depth at which the object being dated was found.

energy should be noted, as well as the rapid changes at small depths. Due to the exponential dependence of thermal draining on 1/T, the few "heat-waves" of even short duration are felt at small depths with the result of increasing  $T_{\rm B}$ . It should be mentioned that for all depths examined, the average temperature over the 2-yr period was the same within  $0.05^{\circ}{\rm C}$  and equal to  $18.5^{\circ}{\rm C}$ .

Finally, Fig. 8 shows the variation with time of the factor  $\exp(-E/kT_{\rm B})$ , where  $T_{\rm B}$  is the Boltzmann-equivalent temperature for each month. This is plotted for traps of 1.2 eV energy and for two depths, 10 and 100 cm. The factor gives the fraction of filled traps which are drained over an interval of time equal to 1/s, where s is the frequency factor, with typical values between  $10^8$  and  $10^{12} {\rm s}^{-1}$ . The main feature of the graph is that it shows clearly that the main drainage of filled traps occurs during the 4 or 5 hottest months of the year.

# 5. DISCUSSION

The effects of ground temperature variations and of other complicating factors on TL dating will now be discussed, with reference to the three dating methods mentioned.

### 5.1. Ground temperature variations

It should be mentioned at the start that the model used in the theoretical treatment is a very simplified one, assuming first-order kinetics both during irradiation and TL measurement, without taking into account competition between traps, retrapping and other effects. Nevertheless it is felt that even with these simplifications, a realistic qualitative exam-

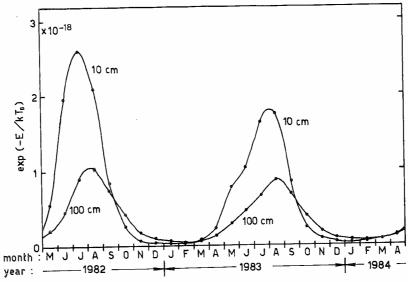


Fig. 8. The variation with time of the factor  $\exp(-E/kT_{\rm B})$  for  $E=1.2~{\rm eV}$  and  $T_{\rm B}$  estimated at two depths (10 and 100 cm) for 24 consecutive monthly periods. Multiplied by the frequency factor s, the ordinate axis gives the fraction of filled traps which are thermally drained per unit time. For the depth of 100 cm and for  $s=10^8 {\rm s}^{-1}$  the average fractional rate of drainage is about  $4\times 10^{-11}~{\rm s}^{-1}$  or about 0.126% per yr. For this rate the peak would be suitable for dating. The importance of the hottest 4 or 5 months of the year is obvious. The difference between the two successive years is also clear, especially at the smaller depths.

ination of the problem is possible and some quantitative estimates may be made.

It is evidently clear from the analysis made that when detailed knowledge of the temperatures at the depth at which the sample was found is not available, using an annual average temperature at the same site or at the region of the find, will result in large errors in the ages if corrections have to be applied due to thermal drainage. This was pointed out for the case of geological dating by Watanable and Pretti (1984), who used a peak at 367°C, of  $E=1.54\,\mathrm{eV}$  and  $s=8\,10^{11}\,\mathrm{s}^{-1}$  for the purpose. At the assumed annual average temperature of 24°C for the region, this peak has a half-life of  $3.6\times10^6\,\mathrm{yr}$  while at a temperature of (say) 2°C higher or lower the half-life would be different by a factor of 1.5.

Using the regional mean surface temperatures under shade would lead to errors also due to the fact that there may be substantial differences between neighbouring sites. As an example, it might be mentioned that in the Athens region, temperatures at the Athens Observatory (very near the Acropolis) are on average 1°C higher than those at the Nea Philadelphia Meteorological Station, 5 km away.

From the data used in this study it was also found that for a find at a depth of 50 cm, if the monthly average surface temperatures are used in estimating the Boltzmann-equivalent temperature instead of the detailed data for the depth of 50 cm,  $T_B$  will be underestimated by approx. 2.5°C and lead to errors in ages of between 30% for a 0.8 eV peak and 60% for a 1.4 eV peak.

For samples exposed to sunshine there exist great variations of temperature with time which cannot be

correlated with any accuracy to ground temperatures. The shape and surface texture of the sample play an important role in the temperature of such an object. For this reason,  $T_{\rm B}$  was not estimated for ground surface, although temperature readings were available.

When the exact depth is known at which the object being dated was found, it is necessary to have detailed data of temperature at that depth. There exist, up to a depth of 1 m, substantial temperature gradients which lead to significant variation of the Boltzmann-equivalent temperature  $T_{\rm B}$  with depth, as seen in Fig. 7. This variation is more pronounced at depths smaller than 20 cm. As seen in Fig. 8, the most important period for which temperatures are needed are the 4 or 5 hottest months of the year.

The thermal history of an object depends also on its immediate surroundings. The microclimate at the exact location of the object may be affected by ground morphology and inhomogeneities in the ground, caused by the presence of large rocks or other objects, things which are quite common at archaeological sites. The assumption is also made that the environment of the sample was not changed over a period of time equal to its age. Earthquakes and other disasters (natural or man-made) may alter this environment. This limitation is also a problem for other methods of TL dating.

It is noted that if for any of the reasons mentioned above there is an overall uncertainty of  $1.5^{\circ}$ C in  $T_{\rm B}$ , there will be errors in the half-lives of peaks amounting to 17% for a peak of 0.8 eV, 20% for 1 eV and 32% for 1.4 eV. These will also be the errors in the ages of the objects being dated. For depths greater

than 20 cm, an error of  $1.5^{\circ}$ C in  $T_{\rm B}$  will be caused by an uncertainty of 50 cm in the exact depth at which the object was found (Fig. 7).

The storage temperature after excavation is also important. The height of a low-temperature peak may be drastically reduced by storage at a temperature which is only a few degrees higher than the Boltzmann-equivalent temperature during its history.

The temperature variations examined in this study were confined to detailed examination of a 2-yr period. There are however longer period variations in temperature. The mean air temperature in Athens for example has increased by 0.7°C during the last century, although there is no evidence that this rate of increase has prevailed for longer time periods. Fluctuations of temperature with approx. 1°C amplitude are present on the decade scale and of approx. 2°C amplitude for shorter periods. Finally, on the geological time-scale there are substantial fluctuations (5-10°C or more) as witnessed by the onset of ice ages. These have been well documented by studies of the isotopic abundance of oxygen in carbonate deposits or fossils, which is known to be temperaturedependent (Emiliani, 1955, 1958).

#### 5.2. Other factors affecting TL age

Other factors besides ground temperature may cause problems in TL dating based on the shape of glow curves.

The first important question that has to be decided is whether a low-temperature peak which is to be used has reached equilibrium height or not. To decide on this question one needs to have some idea of the expected age of the object and the parameters of the peak to be used so that an estimate of its half-life may be made. When dating burnt flint, for example, Wintle and Aitken (1977) had to consider the possibility of thermal drainage effects in some samples which gave unusually high natural doses for their expected archaeological ages but nevertheless low enough to exclude the possibility that they were the doses received by the flints since their geologic formation. Considerations similar to those of section 2.1 lead to the conclusion that the samples were not in fact adequately heated in antiquity to remove their geological thermoluminescence.

Dating by method A requires a peak at equilibrium height from which to determine the dose rate r. The parameters that need to be measured however are so many  $(E, s, N, n_s, R_0$  and  $T_{\rm B})$  that the accuracy of the method is very poor in age determination. It is however quite suitable for examining the thermal history of an object by determining  $T_{\rm B}$  if the dose rate r is known. The method has been used for example by Christodoulides (1971) and Durrani  $et\ al.$  (1972) in determining the maximum temperature reached on the surface of the Moon using material brought to Earth by the Apollo 12 mission.

For dating by method B, the low-temperature peak used must not be near equilibrium height although it

is necessary that it has been affected considerably by temperature to give accurate dates. It seems that natural quartz which is found in pottery has enough TL peaks to make possible the use of this method on objects with ages between 10<sup>3</sup> and 10<sup>4</sup> yr, a range which covers adequately the ages of archaeological interest.

A source of error is the fact that measurements performed in the laboratory for the determination of the half-lives of peaks have to be made at temperatures higher than those of the sample's environment, for practical reasons. One then either finds the E and s values of a peak or extrapolates to find the half-life of the peak at ambient temperatures. Apart from the errors caused by the extrapolation, the procedure is based on the assumption that the kinetics obeyed by the peak (or peaks) remain the same over a wide range of temperatures.

Closely related to this last problem is also the fact that irradiating at elevated temperatures may affect the kinetics quite drastically and yields a different TL vs dose growth curve than the one prevailing during the natural irradiation of the object being dated. That the sensitivity of a peak to dose depends in some cases on the temperature of irradiation has been demonstrated by Aitken et al. (1974), Khazal et al. (1975) and Durrani et al. (1977) for quartz and by Sunta et al. (1976), and Kathuria and Sunta (1982) for LiF (TLD-100). It may happen that at a temperature, a given peak has a very short half-life so that it does not take part in the competition processes of free charge carrier trapping and recombination. At a lower temperature, the traps producing this TL peak may retain charges for much longer periods, thus altering the kinetics of trap filling during irradiation. This would have an effect on the sensitivity of peaks at higher temperatures which would otherwise remain unaffected by thermal effects. For this reason, irradiations in the laboratory should be performed at the same temperature as that of the sample during its history, and this is more or less the procedure followed in the "classical" TL dating methods. It is impossible however to follow this rule when applying method B. The very basis of the method is the assumption that one can accelerate the trap-filling and trap-emptying mechanisms by the same factor while not disturbing the kinetics of these processes. This is a dubious assumption, and there does not seem to be a way of either by-passing it or of checking its validity, except perhaps by dating objects of known ages.

The assumption of constant dose rate is also not entirely correct. Variations of the dose rate with depth, weather conditions and month of the year were found at many different sites by Mejdahl (1970, 1978). Seasonal variations of the dose rate and correlation with weather conditions were also found by Liritzis and Galloway (1981), and Liritzis (1985). The only way of solving this problem would be by direct measurement of the dose rate as a function of

time at the exact spot the object being dated was found. This however, would invalidate the only advantage these dating methods offer, namely of not having to know the dose rate. Neither does there seem to be any regularity in the dose rate variations which might be used to overcome this problem.

In samples having more than one low-temperature peak affected by thermal drainage, problems arise if these are not well separated in the glow curve. For method B, if two such peaks exist, one may argue that since there are two adjustable experimental conditions, temperature and dose rate, glow curve matching may be achieved by finding the unique pair of temperature value T and duration of irradiation t which satisfy the conditions

$$t_{\rm A} \exp(-E_1/kT_{\rm B1}) = t \exp(-E_1/kT)$$

and

$$t_{\rm A} \exp(-E_2/kT_{\rm B2}) = t \exp(-E_2/kT)$$

where the subscripts 1 and 2 correspond to the two peaks. This however would make the procedure of finding t and T much more time-consuming and the errors in the age larger.

In method c, low-temperature peaks may be present after laboratory irradiations which do not exist in the natural glow curve. These must be removed by annealing before measurement. In the DATE method as applied, such a peak appears and contributes to the 325°C peak used for dating. Also, the peak at 375°C has some contribution at 325°C but this is not a serious problem since the parameter g(t) can still vary substantially (from 1 down to 0.4 for ages between 0 and 7000 yr). The range of ages covered may reach  $10^4$  yr and is due to the fact that the peak used has a mean life at 20°C of  $4250 \pm 350$  yr (Langouet et al., 1985).

Of the three methods mentioned, the DATE method seems to be the more successful, being a method of comparison not involving the determination of any peak parameters. If care is taken for the laboratory irradiations to be performed at temperatures close to those of the sample's environment during its history, the only serious source of error remaining is that of the uncertainty in the depth at which the sample was buried during its history. If objects of known age found at the same depth are used in establishing the g(t) calibration curve then the uncertainties in age may be as low as about 10% as deduced from published results obtained by this method.

#### 6. CONCLUSIONS

The variation of ground temperature with depth and with time was found to have significant influence on low-temperature peaks used in TL dating methods based on the shape of glow curves. A detailed knowledge of these temperatures at the exact spot where the object being dated was found must be available for

reasonable accuracy in the results. This is not always possible and in most cases is a more difficult problem than measuring the dose rate directly.

Other problems such as the variation of dose rate with time, weather conditions and nature of the environment, complicate matters further. In methods where the peak parameters have to be determined by measurements in the laboratory the uncertainties in age are further increased.

The DATE method, which is a method based on glow curve comparison, seems to be the most accurate of the methods examined, being able in favourable cases to give ages with errors of the order of 10%. All methods could be usefully employed in authenticity tests where a rough estimate of the age is considered satisfactory. There does not seem, however, to exist a way of overcoming the problems mentioned so as to improve the accuracy of the methods and make them useful for archaeological or geological dating.

Acknowledgements—The author wishes to thank the National Meteorological Service of Greece for providing the ground temperature data on which this work was based.

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