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Observations on palæodose determination with burnt flints

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Introduction

The thermoluminescence dating of prehistoric flints is now a solidly established technique (Aitken, 1985; Huxtable and Aitken, 1986; Valladas, 1992). However, in attempting to date Lower or Middle paleolithic hearths one is confronted by three problems:

1. The supralinear growth of the TL within a wide interval of radiation doses, up to 100 Gy in some instances (Mercier, 1991);
2. the onset of saturation at doses in the 150 to 700 Gy range;
3. the specific behaviour of the α radiation, which contributes significantly to the integrated radiation dose in the flint.

The first two are associated with the β and γ components of the radiation dose.

This paper deals with procedures for determining palaeodoses in archaeological flints when faced with obstacles of the type noted above and describes simulation experiments on raw unburnt flints collected near palaeolithic hearths. The experimental conditions are listed in table 1.

Table 1. Experimental conditions

The samples were annealed at 500 °C (except when indicated otherwise), crushed in an agate mortar and sieved to produce a size range of 100-160 μm, followed by washing with hydrochloric acid to remove calcium carbonate. TL emission was detected between 350 and 450 nm and glow curve measurements were made with six 2 mg aliquots for each sample.

The radiation sources were:

α	^{238}Pu	Flux (4MeV): $2.5 \cdot 10^6$ α/(cm^2·sec)
ß	^{90}Sr	Dose rate: ~8 Gy/min
γ	^{137}Cs	Dose rate: ~100 Gy/hr with 50 mg samples (Valladas, 1978).

In the discussion that follows the effects of β and γ radiation will be considered as equivalent.

The effects of α radiation.

It has been reported that the TL induced by α–particles is a linear function of the integrated α–flux (Aitken, 1985). However, it seemed advisable to check whether the linearity of the α–contribution (TLN_α) is maintained in flints heated 300 ka - 400 ka ago whose TLN ($TLN = TLN_\alpha + TLN_\beta$) indicates the onset of saturation. Thus two specimens (labelled T53 and T59) were subjected to the fine grain preparation (Zimmerman, 1971) and irradiated. As can be seen in fig.1 the growth curves remain linear up to integrated α fluxes of ~2.7 and ~$0.9 \cdot 10^{10}$ α/cm^2 for samples T53 and T59 respectively. Based on the uranium content of the two flints, the corresponding onset of saturation is expected after 1.14 and 1.67 million years respectively. Thus we will assume in this paper that the growth curve is a linear function of the integrated α–flux.

The effects of α and β (or γ) irradiation

To determine the relative efficiency of α and β (or γ) rays in the production of TL at different levels of saturation (Aitken, 1984), we worked on single layers of grains from prehistoric burnt flints T8 and T60 treated as described in table 1, except for the initial heating. The short path length of α rays compared with the grain size is unimportant since we are only interested in the relative effects of the two radiations. Four samples of each specimen were first exposed to γ radiation doses of 0, 150, 300 and 600 Gy respectively. A fraction of each sample received an additional $9 \cdot 10^9$ α/cm^2; another fraction received an additional β dose of 126 Gy.

The TL growth induced by the α and β radiation is shown in fig.2 (ΔTLα and ΔTLβ, respectively) as a function of each of the four initial γ doses. It can be seen that the higher the initial γ dose the smaller the subsequent ΔTLβ; such behaviour is indicative of saturation. In contrast, the ΔTLα is almost independent of the degree of saturation. Thus, the α/β efficiency ratio rises with increasing γ (or β) dose. This has to be taken into consideration when computing the β dose equivalent to a given α irradiation, which is:

$$S_\alpha = \frac{(TL_\alpha)D_\beta}{(TL_\beta)\,\varphi_\alpha} \quad Gy/(\alpha/cm^2) \qquad (1)$$

where,
$TL_\alpha = TL(N + \alpha) - TLN$; $TL_\beta = TL(N + \beta) - TLN$,
D_β is the β dose received by the sample and φ_α is the integrated α flux.

Figure 1.
TL growth in flints T53 and T59 exposed to α radiation only. The solid lines represent polynomial computer fits through the experimental points.

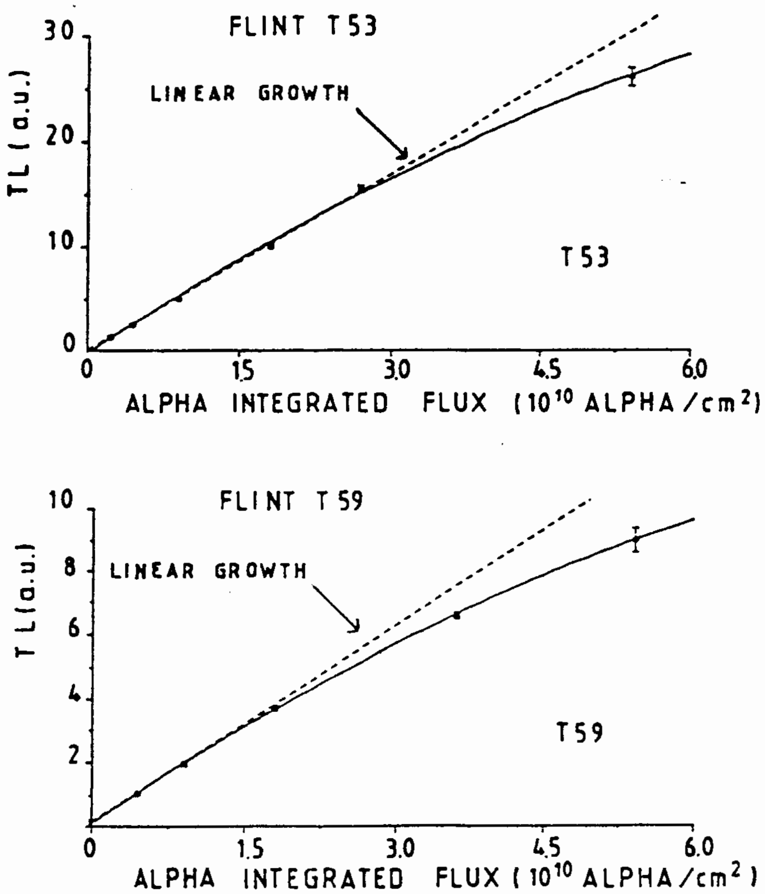
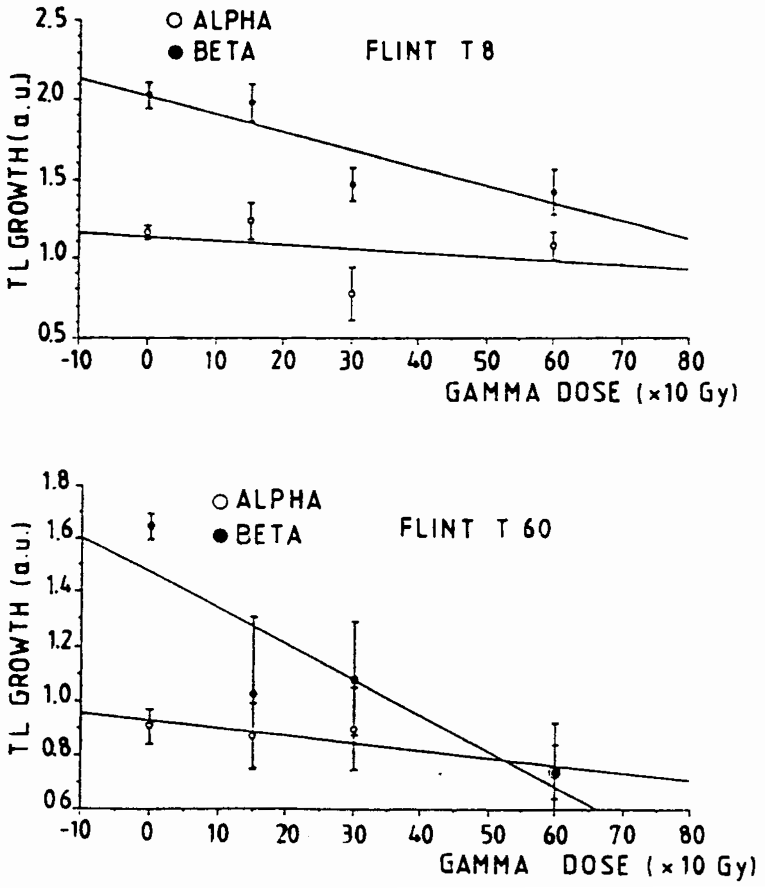


Figure 2.
TL growth in prehistoric burnt flints T8 and T60 receiving, respectively, an integrated flux of $9{\cdot}10^9$ α/cm² (ΔTLα) and a β dose of 126 Gy (ΔTLβ) as a function of the initial γ dose received (0, 150, 300 and 600 Gy).



It is apparent that for archaeological flints saturation will result in an overestimated S_α. One way of getting around this difficulty is to determine S_α on annealed samples. Tests on several flints which were not saturated showed that the value of S_α is unaffected by annealing at 500 °C. Hence, for samples at the onset of saturation, S_α should be determined in this manner.

Palaeodose determination in flints at the onset of saturation

The induced TL of an archaeological flint is normally plotted as a function of the added dose. Finding a function that passes through the experimental points and can be extrapolated to zero to give the palaeodose can present problems. The function currently used to deal with saturation has the form:

$$A\,(1 - e^{-d/M})$$

where, A is a proportionality factor, d represents the dose and M is a parameter giving the best fit to the experimental curve.

However, measurements on flints heated in the laboratory show that for d>0, the growth of TL is parabolic (i.e. $ad + bd^2$, where b>0) over a relatively wide dose range (0 to 100 Gy) which corresponds to supralinear behaviour (fig.3 in Mercier, 1991). It is noteworthy that expansion of the exponential formula gives $A(d - d^2/2M^2)$, a parabola with a curvature inverse of the experimental findings and clearly inappropriate. When extrapolated, such an exponential usually underestimates the palaeodose by about 50 Gy (fig.3 in Mercier, 1991).

We failed to find a simple function capable of fitting TL growth curves within the dose interval of interest for dating measurements. The only practical way to compute the palaeodose of flints exhibiting supralinearity and saturation is to use the glow curve recorded during second heating. Such an approach does not require any knowledge of the missing parts of the first TL glow curve, as has been shown in a previous study of quartz pebbles heated in a volcanic lava flow (Valladas and Gillot, 1978). However, in the case of flints, the difference between the plots of the TL growth induced by the α and β radiations must be taken into account. The procedure is applicable if the TL glow obtained after the second heating has values proportional to those obtained during the first heating. If the condition is fulfilled the first glow curve can be normalized to the second in the manner described below.

Let TL(D) represent the natural TL and TL'(D') the second TL glow, where D' is the applied laboratory dose under conditions where TL' (D') ≈ TL(D).

Let TL(D + d) and TL' (D' + d) represent the first and second glow after a dose, d, has been added.

We now define:

$$Y_o = \frac{TL(D)}{TL'(D')} \cdot (D') \qquad (2)$$

$$Y_1 = \frac{TL(D+d)}{TL'(D'+d)} \cdot (D'+d) \qquad (3)$$

$$Y_2 = \frac{TL(D+2d)}{TL'(D'+2d)} \cdot (D'+2d) \qquad (4)$$

To get the first estimate of the palaeodose, D_o, one extrapolates the points $(0,Y_o)$, (d,Y_1), .which usually yields a straight line, in the normal way (see fig. 7 in Valladas and Gillot, 1978). The second estimate, D_1, is obtained by replacing D' with D_o in equations (2), (3), etc. After 3 or 4 iterations the series D_o, D_1, D_2, ... converges to the correct palaeodose, D. The ratios

$$\frac{TL(D)}{TL'(D')},\ \frac{TL(D+d)}{TL'(D'+d)},\ \ldots$$

are then independent of glow curve temperature in the temperature range of the TL peak, because the doses D and D', (D+d) and (D'+d) are not significantly different and converge at the end of iteration.

Measurements were made to check if the flints satisfied the proportionality condition. Raw flints collected near prehistoric hearths were heated to 500 °C, within the temperature range estimated for prehistoric burnt flints (Valladas, 1983) and under the conditions given in table 1. Half of the grains prepared taken to represent flint freshly removed from the hearth, was set aside. The other half was annealed at 350 °C for 90 mins after receiving a dose of 300 Gy to simulate the palaeodose. The annealing was performed at a temperature slightly lower than that of the TL peak (380 °C) so as not to alter the TL properties of the second flint fraction in comparison with properties of the first. The TL of the first half was denoted by TL1, that of the second by TL2. Aliquots of both fractions were irradiated with doses of 50, 100, 250, 400, 600 and 800 Gy, respectively, and their TL measured. The corresponding TL1 and TL2 values were obtained by integration of the glow peak between 350 and 410 °C. For the different applied doses the TL1/TL2 ratios were computed from polynomials fitted to the experimental values for eight flints: A, B, C, D, E, H, I and J (see fig. 3). As can be seen, the ratios are relatively constant for flints B, C, D, E, I and J and vary by less than 5% in flints A and H.

Since the proportionality condition is fairly well satisfied, the method described above can be used to obtain a good estimate of the palaeodose in the following way: 1) determine the growth curve of one fraction of the grains of archaeological flint; 2) determine the growth curve of another fraction, annealed at 350 °C, at doses higher than the roughly estimated paleodose. Interpolation of a second order polynomial is generally sufficient. It should be pointed out that the

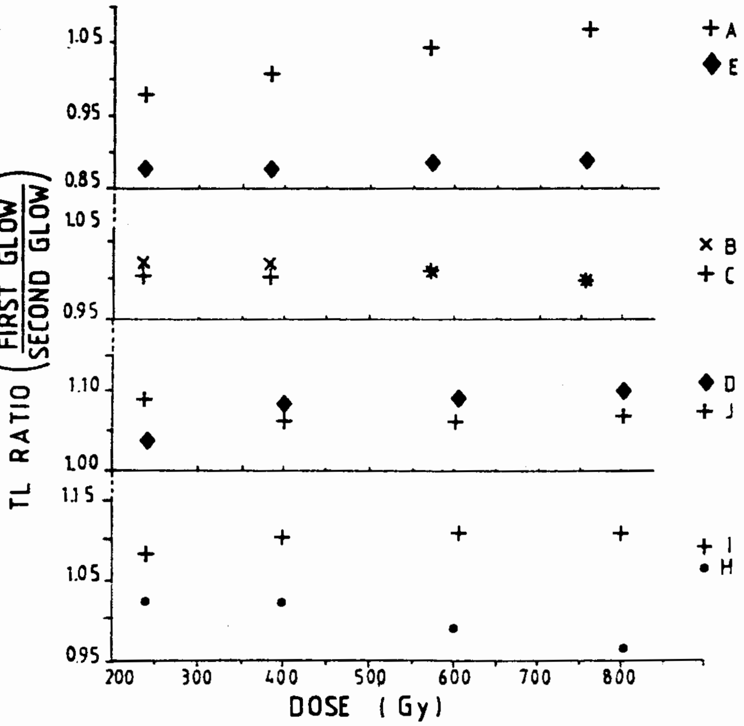


Figure 3.
Ratios of TL values from the first and second glows as a function of irradiation dose in experimental flints A, B, C, D, E, H, I and J.

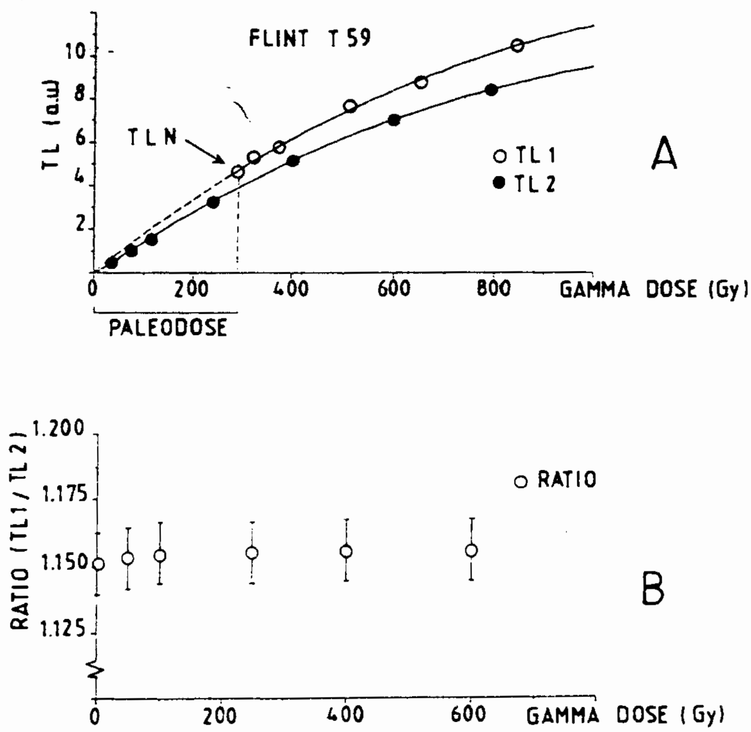


Figure 4.
Palaeodose determination in archaeological flint T59.

A. *TL growth for the first (TL1) and second (TL2) heatings, respectively, as a function of the added γ dose. Each point represents a value with an uncertainty of less than 3%. The solid lines represent polynomial computer fits through experimental points.*

B. *Ratios between corresponding points on TL growth curves from the first and second heatings as a function of γ radiation dose added.*

result is independent of the flint response at lower doses.

All the above concerns primarily flints that have received β (or γ) radiation. However, the TLN of prehistoric flints invariably contains some contribution from α radiation. As the proportionality condition has been found to be satisfactory with experimental β (or γ) irradiated flints, it is reasonable to work with prehistoric flints in the same way and subtract the α–contribution from the TLN. The palaeodose induced by β or γ rays from which the "β age" is computed by using the equation:

$$A = \frac{\text{Palaeodose}_{(\beta+\gamma)}}{\text{Annual dose}_{(\beta+\gamma)}}$$

The α–contribution to the TLN can be approximated by:

$$(\text{TLN})\alpha = \frac{(\text{TLN}) \cdot d_\alpha}{\text{Annual dose}}$$

where, d_α has the form: $d_\alpha = S_\alpha \cdot$ (annual α flux) and S_α is given by formula (1).

If the TLN is at the onset of saturation the age thus computed will be overestimated. In that case, a new $(\text{TLN})_\alpha$ can be calculated by using this approximated age and reiterating the calculations. In most instances the age thus recalculated falls within 2% of the true age.

A good example of this can be seen in fig.4, where the data from a prehistoric burnt flint are plotted. Part A shows the TL growth as a function of radiation dose during the first and second heating, respectively and the determination of the palaeodose. In part B of fig. 4, the relative constancy of ratios calculated by interpolating the two polynomials used to express the TL growth during the first and second heating respectively is evident. The radiation received by the flint ranged from the palaeodose (point 0 on the abscissa) to 600 Gy of additional laboratory dose.

Conclusion

We have shown that a hypothetical TL growth curve is not necessary to determine the palaeodose of burnt flints at the onset of saturation. After the α-contribution to the natural TL has been subtracted a β-palaeodose can be computed from glow curves recorded after second heating.

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