

# Estimation of equivalent dose in thermoluminescence dating - the *Australian slide* method

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## Introduction

An inherent difficulty in estimating equivalent doses for TL and ESR dating is that the shape of the dose growth curve can be directly measured only for doses greater than the natural dose,  $N$ . This can be done by adding doses to natural samples to give a series of  $(N+dose)$  points. What is required from the sample, however, is the amount of dose received since the TL clock was reset either by heat or by exposure to sunlight. This means finding the point where the dose curve would have been, had it been measured at the time it began to grow after resetting. This requires assumptions about the shape of the "missing" part of the glow curve at doses less than  $N$ .

Various approaches to this problem were discussed in the review paper of Wintle and Huntley (1982) and are summarised in Aitken (1985) and by Berger (1988). In one form or other they involve an extrapolation, which is direct in the case of the additive (including partial bleach) method, or implied in the regeneration method. In the first case it is assumed that the model used to fit the  $(N+dose)$  data correctly models the behaviour of the dose curve when extrapolated. In the case of regeneration, the natural dose point is made to fall on a dose curve "regenerated" from a sample which has had its TL reset in the laboratory. In this procedure, the regenerated curve indirectly models the missing part of the  $(N+dose)$  curve. It has usually been assumed that the shape of this curve is independent of (or at least is not strongly dependent on) the radiation and bleaching history of the sample. Although this is not necessarily true, the procedures described below go some way towards testing this assumption.

In the additive method, extrapolation gives satisfactory results provided that the dose response curve is not too different from linear. However, the uncertainties involved in the extrapolations make it unsuitable when

the dose curve is approaching saturation. Berger (1990), Berger and Huntley (1986), Berger et al (1987), Franklin (1986), Franklin and Hornyak (1989), Grün and MacDonald 1989, Grün and Rhodes (1991), Poljakov and Hütt (1990) and Scott and Sanderson (1988) have discussed statistical strategies for determining the uncertainty in extrapolation and measurement strategies for minimising this uncertainty.

An approach with a somewhat different philosophy was originally suggested by Valladas and Gillot (1978) for quartz that had been reset by heating. Faced with saturating dose curves, they used the shape of the second-glow dose curve to model the missing continuation of the first-glow dose curve for doses less than the natural dose  $N$ , making allowance for a change in sensitivity after heating.

Readhead (1982) and Smith (1983) made similar suggestions for describing the shape of the dose curve for sediments, where the resetting mechanism is exposure to sunlight. Readhead (op cit) added doses to samples that had been bleached in the laboratory, while Smith (op cit) suggested adding doses to sediments in the same sequence, but of younger age. By a variation of this argument, the growth curve of a zero-age sample (perhaps taken from the surface) may be used (Readhead, 1984). In these methods sufficient overlap is arranged between the  $(N+dose)$  curve and the regenerated curve so that the two can be made to coincide by shifting along the dose axis. It can then be determined whether the two sets of points are consistent with a single curve. If they are, then the two data sets jointly define the shape of the complete dose curve.

For some time the Adelaide TL laboratory has been using versions of these procedures which were first described by Readhead (1982; 1984) and Prescott (1983) and subsequently used by them for dating (see, for

example, Readhead 1988; Tejan-Kella et al, 1990). More recently, the method has been developed further in conjunction with a programme to test thermoluminescence dating against other geological evidence (Huntley et al, 1993). It is the purpose of the present note to set out the principles and practice of the method, together with an indication of modifications that may be suggested by differing circumstances.

#### Applying the method

At least twenty 5mg aliquots of prepared sample are given a variety of doses and their TL is used to define the (N+dose) growth curve. A further twenty are given a full sunlight bleach of 12 hours and then given a variety of doses (B+dose). These two curves define the (N+dose) and (B+dose) curves shown in fig 1. If all is well the two curves are the same except for a shift along the dose axis, in which case the (B+dose) curve is now used as a pattern for the missing part of the (N+dose) response. This represents the growth of the TL with radiation dose from the level to which the sample was originally reset at the time of deposition (point M in figure 1). If the sensitivity is not changed by the laboratory bleaching, and if the TL measured after such bleaching is the same as the TL that would have been obtained were it measured just after deposition, then displacement along the dose axis by an amount equal to the equivalent dose will bring the two curves into coincidence.

In practice, the possibility of a sensitivity change must be tested for and, also, the difference between the laboratory-bleached and depositional TL. These are further discussed below. We have used two somewhat different statistical techniques to obtain the dose shift from the data: weighted least squares and maximum likelihood. We describe the most recent versions.

To fit the (B+dose) data we use a single saturating exponential with the optional addition of a linear term which may be necessary in order to accommodate a steady increase at the largest doses. The saturating exponential is widely found to fit dose growth curves and is discussed in most of the references already quoted. Chen and Kirsh (1981), Huntley et al (1988) and Levy (1989) have shown that the function is an appropriate one, at least in some circumstances. Levy (op cit) also shows that an additive linear term can take account of the creation of new trapping centres. Berger (1990) has specifically discussed the use of an exponential-plus-linear model for TL and Grün (1990) has done so for

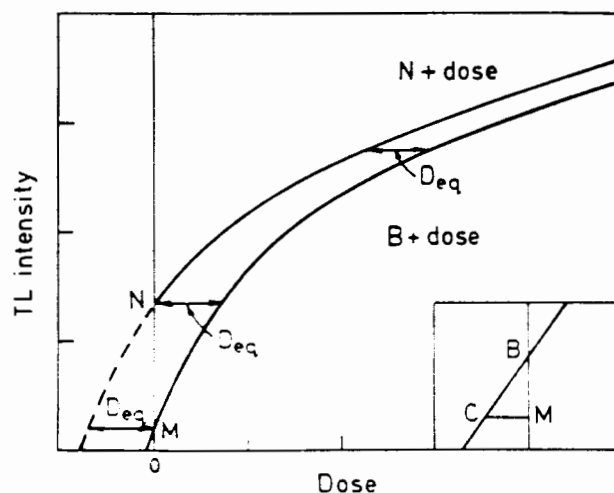


Figure 1. Schematic diagram illustrating the "slide" method used for finding equivalent dose,  $D_{eq}$ . The inset shows the method of correcting for a possible difference between laboratory (B) and natural bleaching (M).

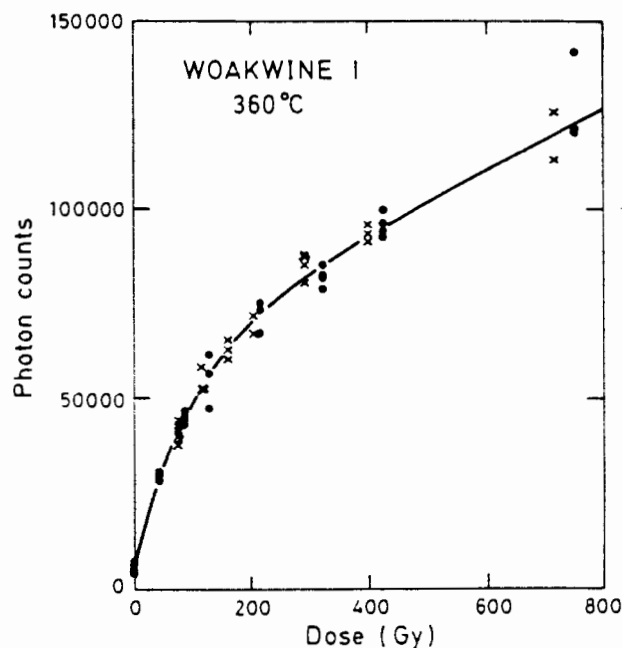


Figure 2. Dose curve for quartz sediment from the Woakwine I Range. Crosses define the (N+dose) data set of 30 points including 10 at zero dose; dots define the (B+dose) data set of 32 points. The two data sets have been made to coincide by sliding them one with respect to the other by an amount equal to the equivalent dose  $D_{eq}$ , which is estimated by least squares or maximum likelihood. The age is about 130 ka.

ESR. We have found that this model is satisfactory over a wide range of samples. It should be noted that both components of the model have an underlying physical basis.

It is worth remarking that the "slide" method which we are describing does not depend on the particular model chosen. The thing that matters is the shape of the curve and a polynomial fit could be used to represent it, as advocated by Readhead (1984).

Our fitting function can be written:

$$I = I_a \{ 1 - \exp(-(D - D_i)/D_o) \} + k(D - D_i)$$

where,

$I$  is the TL intensity,  $D$  is the laboratory dose,  $I_a$  is the high-dose asymptote of the saturating exponential,  $D_i$  the dose intercept;  $D_o$  and  $k$  are the scale parameters for the exponential and linear terms respectively.

The first step is to fit this function to the (B+dose) data and initial estimates of the parameters are obtained. This fit is with a least squares routine and a weighting chosen to reflect the assumption of a constant relative standard

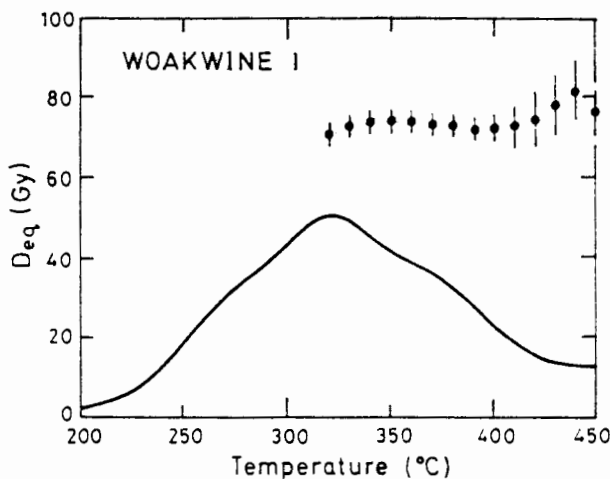


Figure 3.  
Equivalent dose plateau for the Woakwine 1 sample shown in figure 2. The natural glow curve is included to an arbitrary scale.

deviation at all doses. This function is then used as a first approximation to a least squares fit of both the (B+dose) and (N+dose) data sets taken together. In this fit there is an additional parameter, a shift along the dose axis for the (N+dose) data and, optionally, a TL intensity scaling factor.

In our experience (which may not be echoed in general) we have found that only rarely is the scale factor statistically different from unity. Bleached samples differ in this respect from heated samples of the same material. We note that Readhead (1988) reported sensitivity changes in some cases where artificial light sources were used for bleaching. When the intensity scaling factor is not statistically different from unity we set it at unity and re-run the programme. In this case we infer that the data points of the (N+dose) and (B+dose) belong to the same statistical population and can be jointly fitted to the same curve in order to estimate the parameters. This is statistically more efficient. The fitting routine gives the mean square deviation of the data points from the final fitted curve; and the uncertainty in the dose shift is found by a search in reduced chi-square space (Bevington 1969).

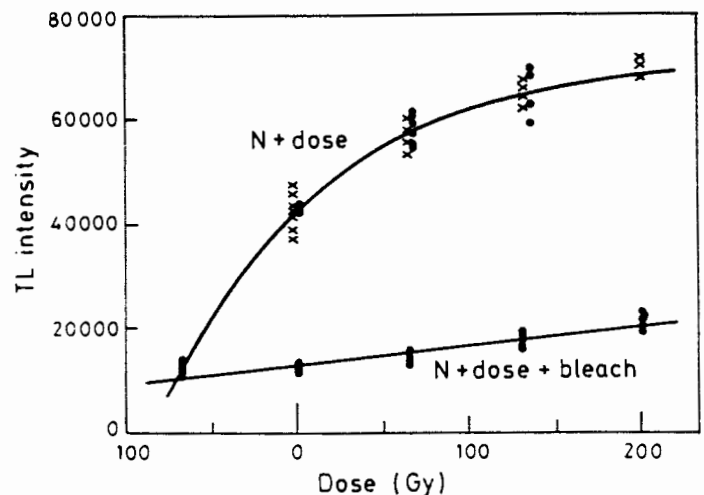


Figure 4.  
Modified Partial Bleach procedures applied to quartz sample TC2S1185 from aeolian infill of a fault scarp near Tennant Creek. The TL age is 50 ka. Crosses define the (N+dose) data set; dots define the (B+dose) and (N+dose+bleach) data sets. The two lowest plotted points each contain ten values, unresolved on the scale of the figure. As for figure 2, the first two of these data sets have been jointly fitted to a common curve by sliding. The lowest curve is a linear fit to the (N+dose+bleach) data.

Starting from the same premises and using the same model we have also used maximum likelihood to estimate the same parameters. The uncertainties in the parameters are found by inverting the Hessian matrix. A variety of comparisons show that there is very little difference between the least squares and maximum likelihood analyses. Which to use is, perhaps, a matter of personal preference.

### Total bleach

We first discuss the application of the method to a coarse-grain quartz sample to which the total bleach method can be applied. Partial bleach requires some modifications (see below). Selective bleach (Prescott and Purvinskis, 1991) which, like partial bleach, is needed when the degree of resetting is unknown or uncertain, calls for significantly different procedures which are the subject of a separate paper.

An example from the Woakwine Range in South Australia is shown in figure 2, adapted from Huntley et al (1993). The above procedure was applied at 10°C intervals to produce "plateau" graphs of  $D_{eq}$  vs T such as shown in figure 3 which is from the same sample.

As mentioned above, the dose shift is a close approximation to the equivalent dose  $D_{eq}$  and, under the usual assumptions of total bleach, the two would be identical. It may be necessary to make some adjustment to allow for the fact that the laboratory bleach may have brought the TL intensity to a larger or smaller value than the natural bleach.

Such an improvement in the estimate of  $D_{eq}$  can be obtained by using a modern sample from the same geological sequence, e.g., from the surface of an active dune, and assuming that its TL represents the intensity that would have been obtained from the sample being dated had it been measured immediately after its deposition. This measurement is used to make a correction, which is of the order of a few percent, by adding or subtracting a correction to the dose shift, as shown in the inset to figure 1. B represents the TL measured after a laboratory bleach, and M that from a modern dune. In the illustration the laboratory bleach was a little too short and the correction MC is added to the main shift.

### Partial Bleach

The above ideas can be used to improve the partial bleach method by reducing the uncertainty in the extrapolation. Fig 4 illustrates the steps. The data are from quartz sample TC2S/185 from the aeolian infill of an earthquake fault near Tennant Creek, Australia. The composite (N+dose) curve is obtained in the same way as the one we have been discussing. The curve marked (N+dose+bleach) has received a partial bleach of thirty minutes which has removed the easily-bleachable component of the TL. In the conventional application of the method, the equivalent dose is marked by the intersection of these two curves, which, it will be remarked, would ordinarily both have been extrapolated.

The fitting procedure already described defines the (N+dose) curve to an extent that negligible extrapolation is necessary for that curve. For the (N+dose+bleach) curve the same procedure could be followed viz, to construct a regenerated curve to use as a model for the partially bleached set. However our experience shows that, as in the present case, the partially bleached curve is often very nearly straight, at least for quartz--a result found by others, e.g. Franklin et al (1992). In these circumstances the practical extrapolation errors from this curve are relatively small and the extra work can probably be foregone. It is worth noting that an exponential fit to the (N+dose+bleach) curve gives no better fit to the data of figure 4 than does the linear.

Perhaps a fitting end-note to justify the present procedures is that, if the (N+dose) data are fitted in the "usual" (i.e. non-composite) way and extrapolated, the estimate of  $D_{eq}$  is 30% larger.

### Acknowledgements

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# Exponential regressions for TL/ESR using regenerated dose response curves

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## Introduction

For some years there has been much effort in improving exponential regression techniques for extrapolating the ESR/TL dose response curves (henceforth DRC) when using the additive technique, e.g., Mejdahl (1985), Debenham (1985), Berger et al. (1987), Poljakov and Hütt, (1990). However, at the same time it was recognized that in many cases the dose response curve were not strictly of the saturating exponential form neither in TL (e.g., comments of McKeever in Bulur and Özer, 1992) nor in ESR (Grün, 1991). As a result all errors evaluated from applying an exponential regression are underestimated (see Grün and Rhodes, 1991). Some specific solutions have been proposed, e.g., in the case where the dose response curve shows an exponential domain at low doses and a linear domain at higher doses (Berger, 1990; Grün, 1990). However, several mathematical functions would be necessary for extrapolating the various types of dose response curves that are experimentally encountered. For some years to come those functions will remain no more than acceptable approximations, because the basic phenomena are still partly unknown, are complex and the associated parameters are widely scattered - even for a given variety of minerals.

This paper deals with the case where the dose response curve of minerals which have been zeroed by heat in the past, such as volcanic materials, seem to be a single saturating exponential function at first glance. This kind of shape is very common, although the studied dose-range must be limited to low doses in many cases because changes in the shape occur when the dose is increased above a certain limit (e.g. a 'second rise').

The present work was intended to test a technique of regression in which account is taken of the regenerated dose response curve: the basic assumption is that the

dose response curve obtained with a laboratory reset sample (for brevity referred to here as the 2nd DRC) is often a close approximation to the additive dose response curve (1st DRC), with, in most cases, allowance for a scaling factor. This assumption of proportionality between the two growth curves had been already used by Valladas and Gillot (1978) in the normalization technique. Consequently, the 1st DRC will be fitted with a function in which some parameters derive from the fitting of the 2nd DRC. In the present work, a single saturating exponential function has been used in the fitting procedure, because it is commonly applied and has a realistic physical meaning but the principle can be extended to other, more developed functions. The technique (hereafter denoted 2+1) is illustrated by 2 simulations on test data from (i) a calculated curve, which seems to be exponential but is not strictly exponential and (ii) an experimental growth curve. An experimental application to dating will be presented in a forthcoming paper.

## The test data

*The simulated dose response curve.*

The calculated test curve was adopted from Li (1991); which consists of two overlying saturating exponential functions,

$$I_1 = 443 \{ 1 - \exp(-0.00268D) \}$$

$$I_2 = 443 \{ 1 - \exp(-0.0134D) \}$$

where,  $D(\text{Gy})$  is the radiation dose and  $I$  (a.u.) is the TL/ESR intensity.

A curve of this type may be proposed when two trapping processes occur simultaneously at one defect site or when two different minerals are mixed. In order to study various shapes of dose response curves in a variety of situations, three combinations of the two functions were used, namely  $0.25I_1+I_2$ ,  $I_1+I_2$  and  $I_1+0.25I_2$ , and referred to here as 0.25/1, 1/1 and 1/0.25

Figure 1.

Example of calculated test curves. The test function is a combination of  $I_1$  and  $I_2$  ( $I = I_1 + I_2$  in the present case) where,

$$I_1 = 443 \{1 - \exp(-0.00268D)\}$$

$$I_2 = 443 \{1 - \exp(-0.0134D)\}$$

The shift of the test curve along the dose axis allows to simulate different preset palaeodoses, i.e., 50, 100, 200 and 300 Gy (as shown).

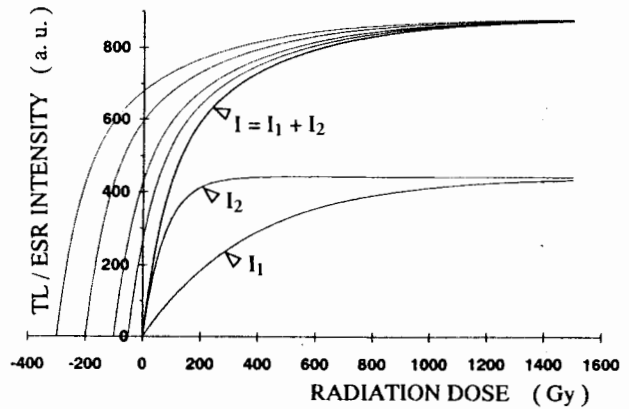


Figure 2.

Test curve  $I_1 + 0.25I_2$ ; preset palaeodose (PP): 200Gy. Black boxes: 2nd DRC test points; open boxes: 1st DRC test points; no errors on the data;  $P_{S1}$ : examples of extrapolations on 1st DRC only;  $P_{2+1}$ : examples of extrapolations with the 2+1 technique. Solid lines represents the theoretical growth-curve.

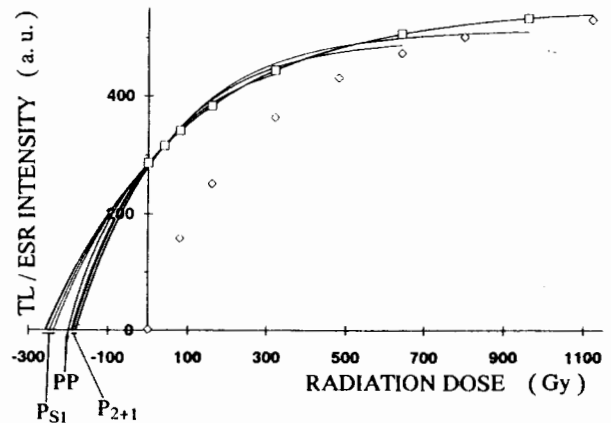
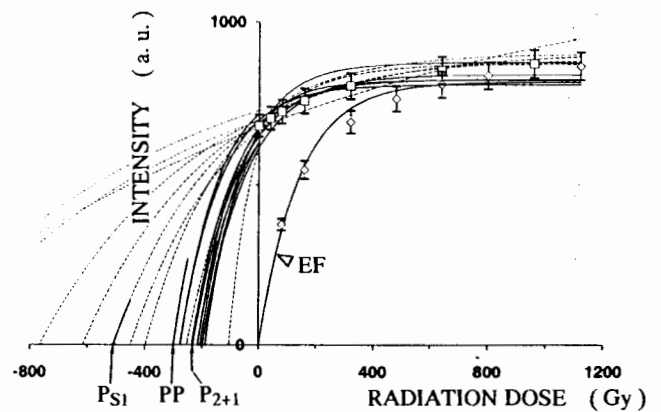


Figure 3.

Regression on a simulated test curve  $I/I$ , with 5% standard deviations on intensities; preset palaeodose (PP): 300 Gy; maximal dose taken into account on 1st DRC: 960 Gy.  $P_{S1}$ ,  $P_{2+1}$ : as in fig.2, the most probable value (the value obtained without errors). EF: exponential fit on 2nd DRC, using mean points. Dashed lines: ten first curves calculated with S1; solid lines: ten first calculated curves with 2+1.



(table 1); the preset palaeodose was varied from 50 to 300 Gy (fig. 1). Simulated experimental points were fixed at doses of 0, 40, 80, 160, 320, 640, 800, 960 and 1120 Gy. Several ranges of doses were used for curve fitting (see bottom of table 1). For every case, the range of doses for the second growth was taken in accordance with the usual choice, i.e., roughly equal to the total dose range of the first dose response curve (i.e. estimated palaeodose + maximum added dose).

#### *The experimental dose response curve*

The experimental set of data points was taken from the measured red TL peak (~620 nm, at 380 °C) of quartz grains annealed (at 400 °C for 15h) and afterwards irradiated in the laboratory. The standard deviation of the intensities was evaluated from repeated measurements (8-10 for each dose). The curve of peak height vs dose was used as a 2nd DRC (as in fact it is) and also as a 1st DRC by a shift along the dose axis of 145 or 290 Gy (see table 2). Thus, the palaeodose obtained by regression of the 1st DRC should be equal to the amount of dose shift, i.e. 145 or 290 Gy (fig.5).

In both cases, the two curves (1st and 2nd DRC) were adjusted to the same scale, so that the curves could be superimposed on each other by a shift equivalent to the palaeodose along the dose axis, i.e., no sensitization was simulated. The results given here would not be altered by a change in sensitivity because the regression technique presented below is able to take it into account.

#### **The 2+1 regression technique**

As a first step, the 2nd DRC is fitted using the function

$$I = I_{1\max} [1 - \exp(-D/D_0)] \quad (1)$$

where,

$I_{1\max}$  is the maximum intensity,  $D$  the dose and  $D_0$  the characteristic saturation dose. In the second step, the 1st growth is fitted using the function

$$I = I_{2\max} [1 - \exp\{- (D - D_e)/D_0\}] \quad (2)$$

where,  $D_e$  is the palaeodose to be evaluated;  $I_{2\max}$  is usually different from  $I_{1\max}$  because of sensitization after annealing.

The coefficients  $I_{1\max}$ ,  $I_{2\max}$  and  $D_0$  are obtained by minimization - by a simple and rapid method of successive approximations - of the sum of the squares of the relative differences between the experimental points

and the fitting curve. The use of relative differences is justified when the relative uncertainty  $\Delta I/I$  ( $I = \text{TL or ESR intensity}$ ) is constant, combined with a significant variation of amplitude of  $I$  in the given dose range (Nougier, 1985). Minimization of the sum of square absolute differences would give too much importance to the largest intensities. That is, the fitting curve would lie closer (relatively) to the high intensity points than to the lower ones; this effect is avoided by the use of the relative differences.

In our laboratory, the standard deviation of the calculated palaeodose (s.d.) is obtained by use of a Monte-Carlo technique (Pilleyre, 1991). Assuming that the error on the dose is negligible, the uncertainty of the intensity is estimated. The process of curve fitting is then repeated many times (e.g., 100 times) by randomly generating data sets that show a Gaussian distribution around the mean intensities of the experimental data set. The s.d. of the palaeodose is then derived from the distribution of the calculated palaeodoses. The palaeodose itself is calculated with the set of points corresponding to the mean intensities. This most probable palaeodose is generally not equal to the mean of the palaeodoses calculated with the Monte-Carlo technique. A Monte Carlo technique was also adopted by Grün and Rhodes (1991).

In the present work, no uncertainties were taken into account in the first (theoretical) simulation ( but in one case specified below), the points belonging to the calculated test curve; standard deviations (one sigma) were used in the second (experimental) simulation.

#### **Results**

Figure 2 illustrates the regressions and table 1 lists the results. As already specified, different regressions were calculated by varying the maximum dose (see table 1). It can be observed that very few extrapolations yield the correct palaeodose but, in nearly all cases, the exponential regressions on 1st DRC only (henceforth *S1*) gave worse results than the 2+1 technique. On the other hand, the lower the maximum dose taken into account, the better the results. This effect is more marked on *S1* extrapolations, where increasing the maximum dose has a strong influence on the calculated palaeodose. At the same time, the effect of the palaeodose value can be observed (table 1): when the test palaeodose for a given test curve is higher, the 1st DRC



Figure 4.

Distribution of the palaeodoses calculated with the Monte-Carlo technique (200 calculations); maximal dose taken into account: 960 Gy; error limits of 5% on intensities (table 1). Preset palaeodose (underlined): 300 Gy;  $P_{S1}$ ,  $P_{2+1}$ : most probable palaeodoses (= palaeodose obtained without errors on the data), with the two techniques of regression, i.e. 2+1 (upper figure) and S1 (lower figure).

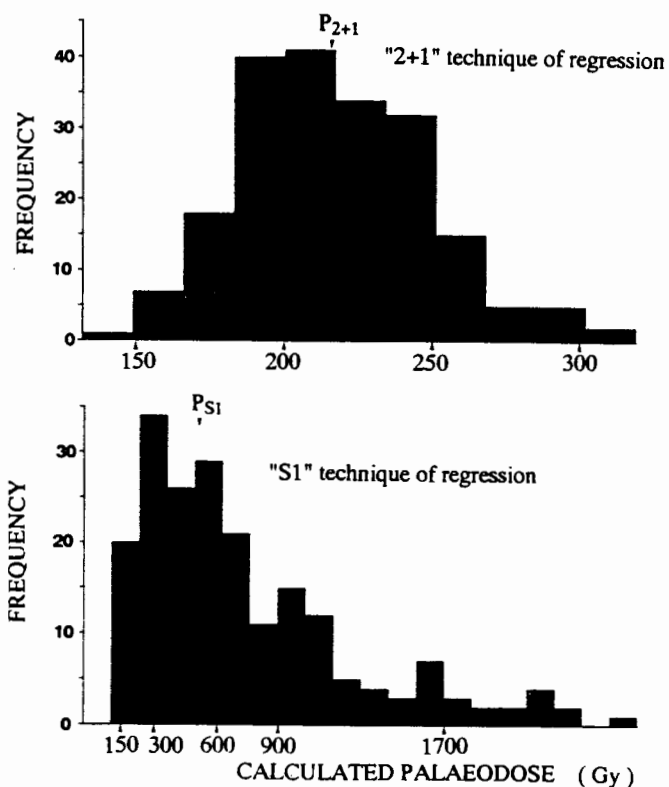
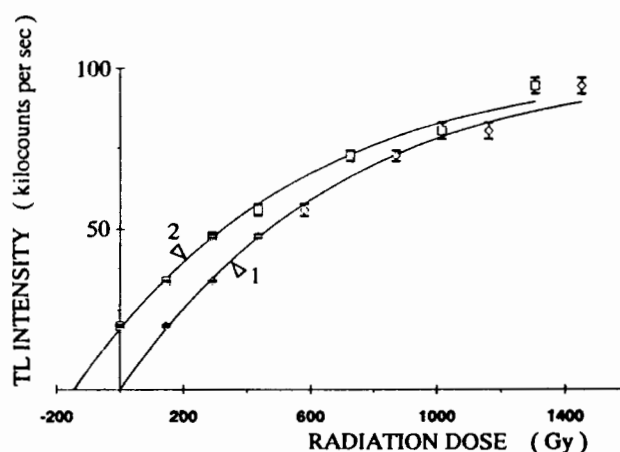


Figure 5.

Experimental test data: red TL of quartz grains annealed in the laboratory and irradiated with a  $^{137}\text{Cs}$  gamma source; red filter RG 610 Schott, 5 °C/s heating rate. Errors corresponding to one s.d. derive from several measurements for every dose. 1: 1st DRC, test palaeodose: 145 Gy; 2: 2nd DRC. Full lines: exponential fits (derived from 1st DRC).



lies closer to the saturating domain and the single fit  $S1$  is less relevant, although it might lie very near to the test points and seem, in this part of the curve, very good from a mathematical point of view.

It was verified that, when the test curve is purely exponential ( $I_1$  or  $I_2$  alone), both techniques give correct results.

The drawback with the above calculations is the lack of consideration of experimental errors, leading to situations far from actuality, especially for the highest palaeodoses. An attempt was made to simulate such

situations by use of Monte Carlo calculations, after an arbitrary standard deviation of 5% had been applied to the intensities of the test points. A particularly critical case was selected by taking the highest palaeodose (300Gy) and the 1/1 combination (fig.3).

Afterwards, the histograms of calculated palaeodoses with 200 draws were plotted (fig.4). It can be seen that the  $S1$  procedure gives an erroneous palaeodose with a very broad distribution, ranging from 90 to 2790 Gy. Moreover, the distribution is clearly non-Gaussian and asymmetric - as already mentioned by Grün and Rhodes (1991) - making the mean value and the s.d. given at the

bottom of table 1 almost meaningless. On the contrary, an approximately correct palaeodose value can be obtained with the 2+1 technique, in spite of the very unfavourable conditions. The corresponding probability distribution being not far from normal, the s.d. can be considered as realistic. Comparison of the mean results from the Monte Carlo calculations (in square brackets in the tables) and the most probable value (without brackets) gives a rough indication of the asymmetry of the palaeodose distribution.

Table 2 lists the palaeodose results obtained with the experimental test data, with the Monte Carlo calculation (100 draws). As above, the results obtained with the 2+1 technique are, in most cases, closer to the test palaeodose than with a single fit *S1*. The s.d. is larger with the latter fit but often not large enough to allow compatibility between the calculated and the preset palaeodoses. In this simulation, the mean calculated palaeodose is, in all cases, not significantly different from the most probable one.

Table 1.

Summary of the palaeodoses calculated with two different regression techniques; the ratio of the calculated and the preset palaeodose is given. *S1*: exponential fitting on 1st growth only, with minimization of the sum of the square relative differences between the fitting curve and the test points. 2+1: exponential fitting of 1st growth with use of the 2nd growth parameter ( see text). 3 different combinations of the functions  $I_1$  and  $I_2$  were used for the calculated test curve. The dose points taken into account are listed at the top of the table; for the 1st growth, those are the natural signal and the natural +6, +5, +4 or +3 added doses; for the 2nd growth, the zero point and 7, 6, 5 or 4 different doses.

The mean calculated palaeodoses obtained with the Monte-Carlo technique (100 draws) are given in square parentheses in the lower part of the table, where a s.d. of 5% has been taken into account for the intensities. The palaeodoses calculated with the mean points are given in the preceding row(s).

Test curve combination	Preset Palaeodose (Gy)	Calculated Palaeodose / Preset Palaeodose							
		<i>S1</i>		2+1		<i>S1</i>		2+1	
$I_1/I_2$	(Gy)	<i>S1</i>	2+1	<i>S1</i>	2+1	<i>S1</i>	2+1	<i>S1</i>	2+1
0.25/1	50	1.17	1.06	1.13	1.06	1.07	1.06	1.03	1.05
1/1		1.36	1.13	1.29	1.13	1.16	1.14	1.08	1.12
1/0.25		1.28	1.13	1.23	1.13	1.15	1.14	1.08	1.12
0.25/1	100	1.35	1.00	1.27	1.01	1.15	1.03	1.07	1.03
1/1		1.57	1.02	1.47	1.03	1.27	1.07	1.14	1.07
1/0.25		1.32	1.03	1.28	1.03	1.20	1.06	1.13	1.07
0.25/1	200	1.88	0.87	1.71	0.89	1.42	0.93	1.23	0.96
1/1		1.74	0.84	1.67	0.86	1.48	0.90	1.32	0.94
1/0.25		1.30	0.90	1.28	0.91	1.23	0.94	1.19	0.96
0.25/1	300	2.16	0.75	2.03	0.77	1.70	0.81	1.39	0.84
1/1		1.70	0.72	1.66	0.74	1.60	0.78	1.54	0.81
1/0.25		1.22	0.84	1.26	0.83	1.24	0.86	1.25	0.88
1/1	300	1.70	0.72			1.60	0.78		
with 5% s.d		±1.86 [2.55]	±0.11 [0.74]			±2.40 [2.40]	±0.22 [0.81]		
Radiation dose points (Gy)	1st growth	0, 40, 80, 160, 320, 640, 960		0, 40, 80, 160, 320, 640		0, 40, 80, 160, 320		0, 40, 80, 160	
	2nd growth	0, 80, 160, 320, 480, 640, 800, 1120		0, 80, 160, 320, 480, 640, 800		0, 80, 160, 320, 480, 640		0, 80, 160, 320, 480	

Another semi-experimental simulation was made identical to the second one but with another quartz

sample and a larger dose range and the results were qualitatively very similar to the preceding ones, with

more pronounced tendencies to give completely erroneous palaeodoses by fitting on the 1st DRC only. The shape of the growth curve appeared bi-exponential.

In the two simulations reported here, slightly better results were obtained with the simple regression (*SI*) in the special case of low palaeodoses and narrow dose range of extrapolation (bottom, right of the tables) but in these cases, the two palaeodoses calculated with the two regression techniques were close to each other.

### Discussion

The first example studied here can only give an indication of what can happen in reality because only a specific type of growth curve and fit was studied. However, it shows that, if the function chosen does not correspond exactly to the physical phenomenon to be fitted, large systematic errors can affect the calculated palaeodose. These errors can be reduced by using the shape of the 2nd DRC. Recognizing from the 1st DRC the appropriate function to be used, e.g., in the present case, a bi-exponential one, would evidently improve the results by application of an adapted treatment like the one proposed by Li (1991) or by Bulur and Özer (1992). In practice a major difficulty comes from experimental uncertainties, which, mostly at the onset of saturation, can accommodate a wide variety of fitting functions.

The use of the 2nd growth data will significantly reduce the freedom of movement, mainly in the dose range of interest, i.e., below the dose equivalent to the palaeodose and far from saturation. But a frequent drawback in the application of the  $2+1$  technique to experimental results is the effect of sensitization, which not only results in enhancement (generally) of the height of the signal for a given dose after annealing, but in frequent modifications of the shape of the dose response curve. Other reasons such as a differential behaviour under alpha and gamma irradiation can also modify the shape of the 2nd growth (see Aitken, 1985, p.139). In practice, the  $2+1$  technique can be used provided the whole fitting curve derived from the 2nd DRC lies within error limits of the experimental points of the 1st DRC. If this is not the case, no reliable palaeodose can be calculated by this technique and the situation reverts to that which obtains when only the 1st DRC is available.

Table 2.

Evaluation of the error associated with the determination of the palaeodose is not a simple task and, as outlined by Grün (1991) in the field of ESR dating, "little attention has been paid to the correct estimate of this parameter". It could be added, that most of the literature deals with standard deviations deriving from the regressions, but not with systematic errors generated by the use of improper functions. The present simulation suggests that those systematic errors can be substantially greater than the s.d.; it then appears illusory to make great efforts in improving the rapidity and performances of regression algorithms unless they are more relevant from a physical point of view. For the same reasons, it is not worthwhile to search for a perfect definition in the distribution of the palaeodoses calculated with the Monte Carlo technique by increasing the number of draws. A realistic estimation of the s.d. will not require more than 100 draws, and this is available with a very acceptable duration of calculation with a PC computer (see note at end of paper).

An additional improvement of the technique would be in taking into account the different precisions of the experimental points by weighting them by inverse variance (expressed in percentage of the intensity in this case), such as tested by Grün and Rhodes (1992). As concerns TL, no important modifications of the results can be expected because, as mentioned above, the relative uncertainty of the intensity is nearly constant for a given sample.

As illustrated on fig. 4 and in table 1, the use of the 1st DRC only results in very large uncertainties in unfavourable cases - where the maximum irradiation dose is small and the growth-curve is at the onset of saturation; because this situation is sometimes seen in the literature, this caution is emphasized.

The fact that reducing the dose range of the two growth-curves towards low doses in the regression improved the results can be qualitatively explained; as the dose range is reduced, the fit to the curvature improves, also leading to an improvement in the estimation of the palaeodose. However, as shown by Grün and Rhodes (1991), this is not always the case and reality is more complex. But we want to emphasize this principle for many cases and particularly when it seems more or less evident; e.g.,

Idem table 1, for the experimental test data (see fig.5), with a Monte Carlo technique (100 draws). Errors are quoted at one s.d..

Preset Palaeodose (Gy)	Calculated Palaeodose / Preset Palaeodose							
	S.1	2+1	S.1	2+1	S.1	2+1	S.1	2+1
145	1.18±0.06 [1.18]	1.05±0.04 [1.04]	1.10±0.07 [1.12]	1.03±0.04 [1.03]	1.15±0.07 [1.16]	1.03±0.04 [1.03]	1.11±0.09 [1.11]	1.01±0.05 [1.02]
*1st Growth	0, 145, 290, 435, 725, 1015, 1305		0, 145, 290, 435, 725, 1015		0, 145, 290, 435, 725		0, 145, 290, 435	
*2nd Growth	0, 145, 290, 435, 580, 870, 1160, 1450		0, 145, 290, 435, 580, 870, 1160		0, 145, 290, 435, 580, 870		0, 145, 290, 435, 580	

\* Radiation dose points (Gy)

Preset Palaeodose (Gy)	Calculated Palaeodose / Preset Palaeodose							
	S.1	2+1	S.1	2+1	S.1	2+1	S.1	2+1
290	-	-	1.10±0.11 [1.11]	0.91±0.04 [0.91]	0.95±0.12 [0.97]	0.95±0.05 [0.96]	0.98±0.14 [1.00]	0.89±0.06 [0.90]
*1st Growth			0, 145, 290, 580, 870, 1160		0, 145, 290, 580, 870		0, 145, 290, 580	
*2nd Growth			0, 145, 290, 435, 580, 870, 1160		0, 145, 290, 435, 580, 870		0, 145, 290, 435, 580	

when a 'second rise' occurs. Above a certain dose, the corresponding dose range can be discarded from regression by considering that the physical phenomenon which becomes dominant at high doses is unimportant in the low dose range. When high dose points have a negative influence on the extrapolation, this effect will be enhanced when the absolute differences between the points and the fitting curve (and not the relative ones) are used in the regression.

The second (experimental) simulation illustrated two aspects of this discussion. Firstly, although the dose response curve seemed to be nearly exponential, it was sufficiently different from an exponential to induce a significant error when the 1st DRC only was considered in the extrapolation and, secondly, the calculated error did not take into account this systematic error.

In order to get a crude estimation of the systematic error, we computed the mean value  $r$  of the ratios (calculated palaeodose/preset palaeodose) for the two techniques and for the two examples (i.e. table 1 and table 2). The results were:

Table 1:  $r_{S1} = 1.36$  ( $s = 0.26$ );  $r_{2+1} = 0.96$  ( $s = 0.12$ )

Table 2:  $r_{S1} = 1.08$  ( $s = 0.08$ );  $r_{2+1} = 0.98$  ( $s = 0.06$ )

It appears that an important systematic overestimation of the true palaeodose can be expected with an

extrapolation of the 1st DRC only (S1), whereas this systematic error remains within acceptable limits for the 2+1 technique. For the time being, it is necessary to assess a systematic error to be quadratically added to the random error in the quotation of the overall error using the 2+1 technique; the intermediate value of  $\pm 6\%$  can be used provisionally.

### Conclusion

It has been demonstrated, with two examples, that extrapolation of an additive growth curve can result in a completely erroneous palaeodose when the fitting function is taken to be exponential but does not in fact accurately represent the dose response of the sample. Generally, results are significantly improved when the shape of the fitting curve is derived from the shape of the second dose response curve, as in the 2+1 technique. Growth curves obtained with experimental data are rarely strictly saturating exponential but the width of experimental errors often hinders an appreciation of the exact mathematical description of the dose response curve. However, a necessary further step of improving the 2+1 technique will consist in developing more suitable fitting functions. We recommend a pragmatic approach such as the 2+1 technique of regression since it is more realistic than some sophisticated techniques applied to the 1st DRC only, even if the curves derived

from those techniques seem to provide better mathematical fits to the available data. At the same time, the simulations presented here should encourage one to be very generous with the size of the errors quoted on TL/ESR ages!

*Note on computing specification.* Calculations were made using a program written in *Turbo Pascal* and running on a PC with a 80386SX CPU and 80387 (16MHz) coprocessor. The overall duration for 100 draws ranged from a few seconds and a few tenth of minutes, depending on the shape of the curve.

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### PR Rainer Grün

The paper of Sanzelle et al. shows clearly the danger of estimating incorrect  $D_E$  values if an incorrect mathematical model is applied for fitting the data of the dose response curve using the additive dose method. The suggestion of applying curve parameters derived from a second dose response curve after the sample was reset is one way to overcome some of the problems to some extent. A similar technique was presented by Prescott et al. (1992) who combined the additive data set with a regenerated dose response curve ("Australian slide method"). This procedure has the general potential to become nearly independent of mathematical assumptions for curve fitting and, hence, the problems that have been outlined in this paper may be overcome.

Prescott, J.R., Huntley, D.J. & Hutton, J.T. (1993) Estimation of equivalent dose in thermoluminescence dating - *Australian slide method*. *Ancient TL* 11, 00-00.

# Uncertainties involved in the measurement of TL intensities

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Computer simulations (Grün & Rhodes 1991, 1992) have shown that the uncertainty in the estimation of dose,  $D_E$ , is critically dependent on the intrinsic uncertainty of the TL intensity measurement, which was also predicted by Berger et al. (1987). The fitting procedure of the dose response curve is also fundamentally dependent on the weight that is assigned to the data points. Usually, two weighting models are applied to the data sets: equal weights and weights proportional to variance (see e.g. Berger and Huntley 1986). If equal weighting is the correct procedure, the measured uncertainty for a sufficient number of repeated measurements should be constant, i.e. independent of the measured TL intensity. In case of weights inversely proportional to variance the relative uncertainty is constant for all values of TL.

We present the glow curves of three data sets of quartz, K-feldspar and Na-feldspar mineral separates from a Danish Cover Sand sample from Eggebaek and integral values of 23 other dose response data sets for each of the mineral separates. The samples have been the subject of an intercomparative dating study (Grün et al., 1989, Kolstrup et al., 1990). Quartz was obtained by heavy liquid separation and etching in conc. HF. The K-feldspar and Na-feldspar samples were separated by heavy liquids using 2.58 s.g. and 2.58-2.62 s.g., respectively. Samples were irradiated with 0, 8.8, 17.6, 44, 88 and 176 Gy. Each irradiation dose was applied to 12 aliquots of the respective mineral separate (and between 4 and 12 aliquots per dose for the integral values). Residual glow curves (6 for each mineral separate) were measured after exposure to light in a Hönlé sun light simulator for 160 min which corresponds to about 15 h of sun light exposure. All samples, N+dose and residual levels, were preheated in an oven for 140 h at 140 °C. TL glow curves were recorded with an automated Risø TL reader and a heating rate of 10 °C/s. The glow curves were digitised with a resolution of 50 channels in the

temperature range 200 - 500 °C (which is the resolution for the glow curves in all figures). The filters used for recording the glow curves were: quartz: 5-60 + HA3; K-feldspar: UG11; Na-feldspar: 7-59.

## Quartz

The left and right hand sides of figure 1 show the glow curves of the natural and irradiated samples and the residual level, respectively. The top figures (1A and 1G) show all recorded glow curves. The respective averaged glow curves are shown below (1B and 1H) with the standard deviation, henceforth s.d., (1C and 1I) and relative error (1D and 1J). The two lower most diagrams show the plots of the standard deviation (1E and 1K) and the relative error (1F and 1L) versus the measured TL intensity, respectively.

Figure 1C and 1E show increasing s.d. values with increasing TL intensity. The irradiated samples show a relatively narrow band of the relative error over most of the temperature range (1D), however, the natural sample shows significantly higher relative uncertainties at lower temperatures. Above about 1/5 of the maximum intensity the relative error remains more or less constant (1F). This implies that at least for this region weights inversely proportional to variance should be used in the fitting procedure. Figure 1E implies that even for the lower range, where a large scattering of the relative error can be observed, the standard deviation is not constant but increases with intensity. It follows that equal weighting is not the correct procedure for the lower intensity range.

Figures 1H to 1L show the same analyses as figures 1B to 1F for the measurements of the residual level. Again, above about 1/5 of the maximum intensity, the relative error remains more or less constant, but increases with

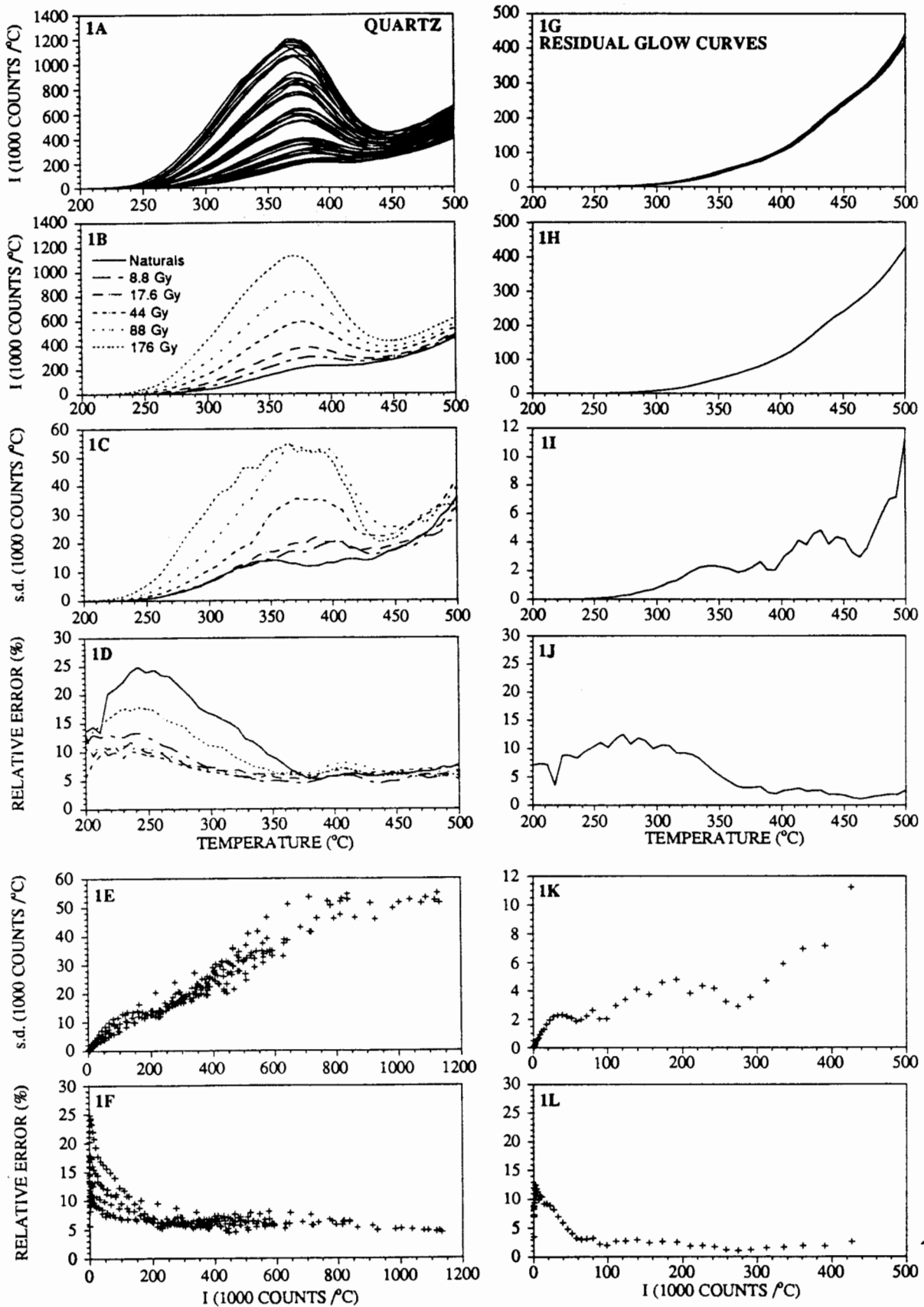
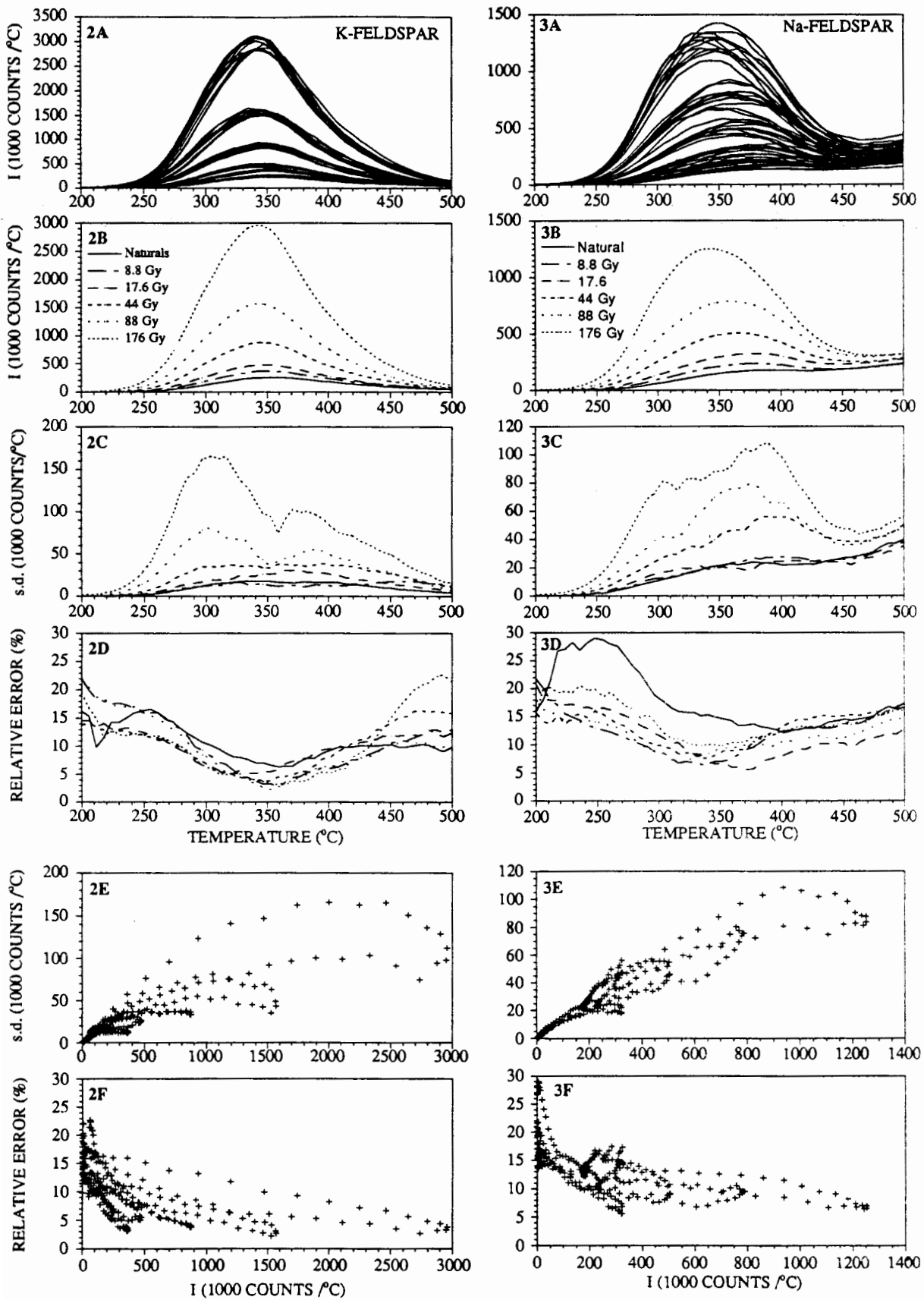


Figure 1.  
 Irradiated samples (left) and residual levels (right) for a quartz sample from Eggebaek, Denmark.  
 1A, 1G: all glow curves; 1B, 1H: averaged glow curves; 1C, 1I: plot of standard deviation, s.d., versus temperature;  
 1D, 1J: plot of relative error versus temperature; 1E, 1K: plot of s.d. value versus measured TL intensity;  
 1F, 1L: plot of relative error versus measured TL intensity.



Figures 2 (left) and 3 (right).

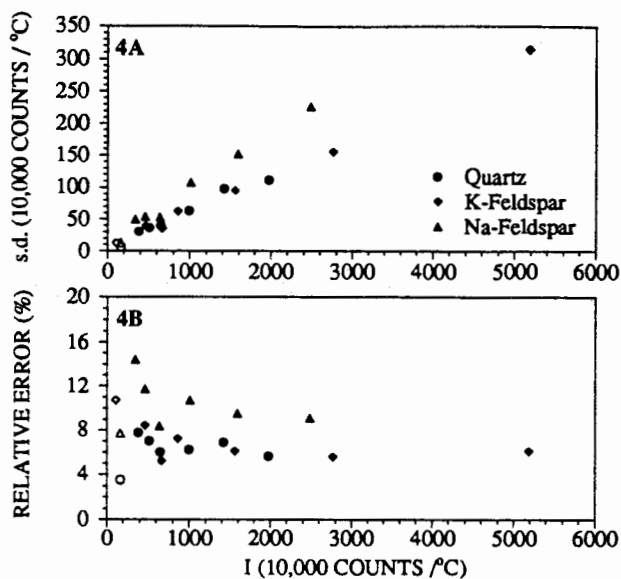
Irradiated samples for a K-feldspar (left) and a Na-feldspar (right) separate from Eggebaek, Denmark.

A: all glow curves; B: averaged glow curves; C: plot of standard deviation, s.d., vs temperature; D: plot of relative error vs s.d.; E: plot of s.d. value vs TL intensity; F: plot of relative error vs measured TL intensity.



Figure 4.

Plot of standard deviation (4A) and relative error (4B) of the integral 239 to 461 °C versus intensity of the glow curves shown in figures 1 to 3. Full symbols: natural and irradiated samples; open symbols: residual levels.



lower intensities. It is interesting to note that the measurement of the residual levels, which are much smaller than the levels of the irradiated samples, are associated with much smaller errors (2% compared to about 6%). This implies that for this sample the distribution of trapped electrons in non-light sensitive traps is more even than the distribution of trapped electrons that are light sensitive.

The integral values of the averaged glow curves for the range 261 to 439 °C are shown in figure 4. The relative errors of the irradiated glow curves show no particular trend and are in the range of about 5.5 to 7%, the natural glow-curve has a somewhat higher relative uncertainty of 8%. The residual level has an uncertainty of 3.5%.

Figures 5A to 5D show the analysis of the integral range of 261 to 439 °C for all 23 dose response data sets. It can be seen that there is a clear trend of increasing average intensity with increasing dose (5A) and increasing s.d. values with intensity (5C). There is no relationship, however, between the relative error and dose or measured TL intensity (5B and 5D).

#### K-feldspar

The display of figure 2 shows the analyses for the K-feldspar sample. The results differ to some extent from the ones for the quartz sample. There is not a trend towards a constant relative error (figure 2F) or constant absolute error (2E). The largest standard deviations are actually observed at the steepest slopes of the TL peak and coincide with the maxima of the first derivative of the glow curve (compare figure 2C with 2B; see also

Grün & Packman 1992). This effect may be attributed to small temperature shifts (temperature jitter). However, below 440 °C, the relative error at a particular temperature is more or less constant (2D), which means that for the dose versus temperature plot fitting with weights inversely proportional to variance seems appropriate. The natural glow curve is associated with the largest relative errors. The residual levels show a similar behaviour as the irradiated samples and do neither display a constant s.d. value nor a constant relative error versus measured TL intensity.

The integral values for the range 261 to 439 °C are shown in figure 4. The relative errors show no trend and are generally in the range of 5 to 8.5% which is very much the same range as for the quartz sample. The residual levels have an s.d. value of 10.7%, which seems rather large.

Figures 5E to 5H show the analysis of the integral range 261 to 439 °C of 23 dose response data sets. The average relative uncertainty of the natural sample is marginally larger than for the irradiated samples (5F), however, there is basically no correlation between relative error and dose or measured TL intensity, respectively (5F and 5H).

#### Na-feldspar:

The display of figure 3 shows the error analyses of the Na-feldspar sample. The irradiated samples show basically the same results as for the K-feldspar sample. The standard deviations of the averaged glow curves for one radiation dose step are largest at the steepest slopes

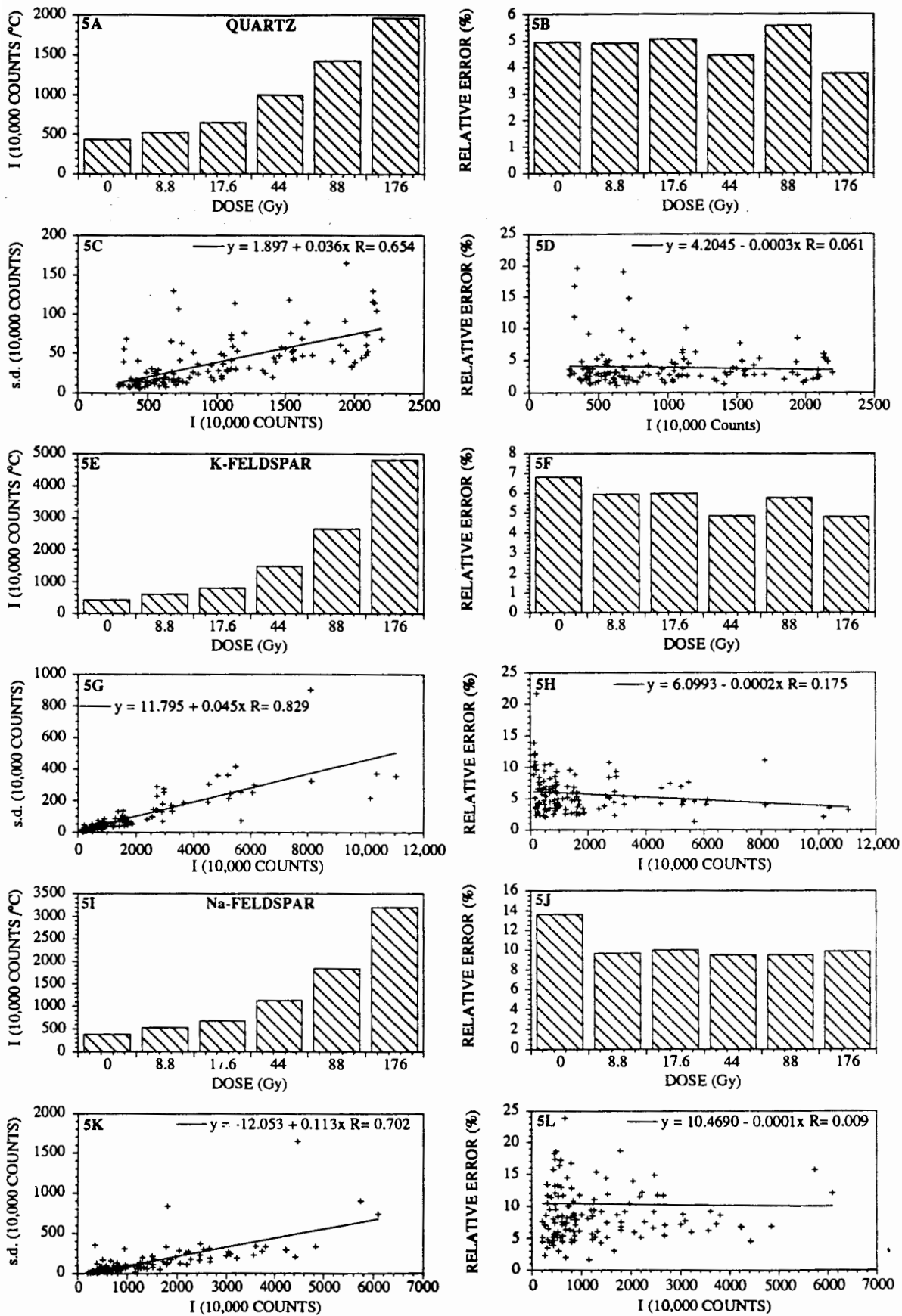


Figure 5.

5A, 5E, 5I: Average integral (239 to 461 °C) intensities for 23 data sets. 5B, 5F, 5J: Average relative errors at each dose point. 5C, 5G, 5K: Plot of s.d. value versus measured TL intensity. 5D, 5H, 5L: Plot of relative error versus intensity.

of the TL peak (compare figures 3C and 3B). The relative error of the irradiated samples is not constant versus the TL intensity, however, the values are within a relative narrow band over the temperature. The natural sample shows a much larger relative error in the temperature range below 400 °C than the irradiated sample. The residual level shows a rather constant relative error over most of the temperature and intensity ranges.

Integral values for the range 261 to 439 °C are shown in figure 4. The relative errors of the irradiated samples show no trend and are generally in the range of 7.5 to 12%, whereas the natural sample has an uncertainty of about 14.5%. The residual level has a s.d. of 7.7%.

Figures 5I to 5L show the analysis of the integral range 261 to 439°C for 23 dose response data sets. The irradiated samples show the same trends as the quartz and K-feldspar samples. The relative error of the natural samples seems considerably higher than for the irradiated ones (5J). However, for the irradiated samples there is no relationship between relative error and dose or measured TL intensity, respectively (5K and 5L).

### Discussion

The size of the intrinsic uncertainty of a TL measurement has influence on the uncertainty in the  $D_E$  estimation (see Berger et al., 1987; Berger and Huntley, 1989; Grün and Rhodes, 1991). As shown above, this value may be intensity dependent and 12 aliquots are apparently not enough to get a better estimate of the intrinsic uncertainty. However, this value need not be known for the correct estimation of the  $D_E$  value, provided the dose steps have been selected appropriately (Grün and Rhodes, 1991 & 1992) and it is known whether TL measurements have constant standard deviations or constant relative errors. The form of the uncertainty has influence on the selection of the weighting model used and, hence, on the estimation of the  $D_E$  value, because the fitting procedure is very model dependent (Grün & Rhodes, 1992). Although one has only very little influence on the intrinsic error of a TL measurement, the error in the  $D_E$  estimation can be critically reduced by measuring repeatedly at each dose step (see Grün & Rhodes, 1992).

The quartz sample shows most clearly constant relative errors versus measured TL intensity (figure 1F). Although the other two mineral separates do not show

this general trend, the relative error is in a more or less narrow (but not constant) band over large ranges of the temperatures (figures 2D and 3D). All samples show a clear trend of increasing s.d. values with increasing TL intensities. The biggest absolute s.d. values, however, do not occur at the maximum TL intensity, but in regions where the slope of the TL peaks are the steepest (maxima in the first derivative of the TL glow curves (Grün & Packman 1992); see figures 2C and 3C). The steep slope regions are most affected by small temperature shifts (temperature jitter). All natural samples of the three data sets show larger relative uncertainties than the irradiated samples over most of the temperature range. All three sample sets show increasing s.d. values with increasing measured TL intensities and basically no relationship between relative uncertainty and measured TL intensity. This clearly implies that equal weighting is not the appropriate procedure for TL dose estimations.

The sets show that the relative error is within a more or less narrow band over most of the temperature range which indicates that weight inversely proportional to variance is the correct procedure for the fitting of the dose versus temperature plot. Whether this has any relevance for the correct estimation of the gamma equivalent dose that the sample has received in its geological past is another matter (see Grün & Packman, 1993). The relative error in the TL intensity for the integral irradiated samples of the three sets show also no particular trend (which basically follows from the above observation) and, hence, weighting inversely proportional to variance is appropriate.

The integral values and uncertainties of the 23 other sets reproduce the results of the three selected samples. There is basically no relationship between relative uncertainty and measured TL intensity and a stronger relationship between s.d. value and measured TL intensity. For quartz and K-feldspar, the relative uncertainty of the natural samples is within the range of the irradiated samples, however, the relative uncertainty of the naturals of the Na-feldspar separates seems somewhat larger than the relative uncertainty of the irradiated samples. Again these results imply that weighting inversely proportional to variance is the correct method. It is interesting to note that the average uncertainty of the quartz samples (in the 3-4% range) is smaller than of the K- and Na-feldspar samples with relative uncertainties in the 5 to 6% and 9% range, respectively,

although the quartz integrals have less counts.

For the estimation of the intrinsic precision of TL measurements the sample sets ought to contain more than 12 replicate TL measurements at each dose point. However, the results may only be valid for a particular sampling site and have actually little relevance for the correct estimation of the  $D_E$  value.

### Conclusion

The analyses of the three TL glow curve sets and integral values of 23 different data sets imply that fitting using weights inversely proportional to variance is the appropriate weighting method for determining dose values for these quartz, K-feldspar and Na-feldspar separates.

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PR John Prescott

# A model for mid-term fading in TL dating

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*Experimental evidence indicates that identical radiation doses acquired by archaeological materials during burial give smaller induced luminescent signals than those resulting from laboratory calibration dosing, thus leading to an underestimation of archaeological ages. A theoretical model is proposed that accounts for this behaviour based on the thermal release of electrons trapped at centres possessing a distribution of activation energies. The predicted degree of age underestimation is found to be sensitive to the ambient burial temperature. In the presence of several independent recombination centres this underestimation will depend on the centre selected by the employment of appropriate colour filters for detection.*

## Introduction

Suitable archaeological materials may often be used as dosimeters to reveal the palaeodose of radiation received during the specimen's burial. This information is generally obtained by the use of thermoluminescence (TL) techniques. It is essential in such determinations to use TL signals that are produced by stable charge trapping states capable of giving a response based on the total absorbed dose. However, when dating archaeological samples with ages in the range of many thousands of years, it has been found (Wintle 1973) in some cases that samples that are expected to be stable over this time range apparently lose part of their TL signal as anomalous fading. This means that the TL dependence on dose given in the laboratory is not the same as that imparted during burial. Clark and Templer (1988) explained this as being due to thermally assisted tunnelling, an effect previously discovered by Visocekas (1979) for calcite. In a series of papers, Mejdahl (1988a, 1988b; 1989) pointed out that when the ages to be determined are in the  $10^5$ - $10^6$  year range, the loss of TL that occurs may very well be due to a long-term thermal decay rather than the above mentioned anomalous fading. Thus, in a study of Tertiary sands, he found TL ages in the region of 300-500 ka rather than 700-2000 ka predicted from the stratigraphy. Recent measurements with feldspars have also indicated a systematic underestimation of TL ages when compared to expected geological ages, however, the extent of the underestimation appeared to be dependent on the wave lengths selected by the optical filter used to observe the luminescence (Balescu, et al. 1991 and 1992).

In a recent paper, Xie and Aitken (1991) describe a model which explains this 'mid-term' thermal fading during burial periods the order of  $10^5$ - $10^6$  years. They assume that the dating signal consists of a stable component and an unstable one, and that while the lifetime of the stable component is long compared to the oldest sample to be dated, that of the other is 'mid-term', namely, long compared to laboratory times but short compared to the ancient ages. They assumed that the unstable component has the same growth characteristics as the stable one, both exhibiting saturating exponential functions of the dose, and that the fraction of the signal which is not stable is constant for all samples. Using these assumptions they showed that for relatively old samples, ages may be appreciably under-estimated. In an example a sample known to be 730 ka old appeared to be only 127 ka old based on TL evidence, presumably as a result of the mid-term fading effect.

## Model

Xie and Aitken do not specify the nature of the fast and slow fading components. A plausible explanation is that a distribution of trapping states exists in which the deeper bound states are more stable than the shallower bound ones. The idea of a distribution of activation energies of trapping levels is as old as the theory of TL itself as first suggested by Randall and Wilkins (1945) who also gave evidence that such distributions exist in calcites and dolomites. Kikuchi (1958) showed that distributions of trapping level energies play an important role in the TL of glasses. Medlin (1961) suggests that discrete trapping levels may be broadened

into a continuous band of levels as the result of local distortions in the crystal fields due to dislocations, vacancies and impurities. Since both the local distortions and the trapping centres may be randomly distributed throughout the crystal, a logical form for the distribution of activation energies would be a Gaussian, namely, in obvious notation,

$$\rho(E) = N\sqrt{a/\pi} \exp[-a(E-E_0)^2]$$

The actual filling of these trapping states may or may not result in Gaussian charge distribution, depending on the circumstances. The effect of such a distribution on TL characteristics has been studied in detail (Hornyak and Franklin 1988).

Very likely, a distribution of closely spaced discrete or sharp trapping states having an occupation capacity following a normal distribution about some preferred activation energy would produce a very similar situation. In this connection it should be noted that a thermal or time sweep across an activation energy distribution possesses a substantial 'window' width thus making such detailed model distinctions difficult.

Hornyak et al. (1992) have recently shown that the TL characteristics of the 375 °C glow peak in quartz can be successfully explained using a model with such a Gaussian distribution for trapping states and two recombination centres yielding green and uv emissions. In the present study, the difference between dosing a sample in the laboratory times and in burial times is investigated when such a Gaussian distribution for traps is present. The differential equations governing the filling of traps and centres during long burial times are numerically solved and the results are compared to those found when the same dose is imparted during laboratory times (say of the order of minutes). The distribution of trap activation energies given in equation (1) is simulated with a 96 segment histogram following the procedure used in the above cited work. To avoid convergence problems and to achieve reasonably short computer running times the earlier VAX program had to be altered from a continuous dosing rate to that of doses administered in steps. For dosing during archaeological burial the probability of thermal release per unit time for a given trapped electron given by:

$$\alpha = se^{-E/kT}$$

is used while for laboratory dosing is set equal to zero. The results for this latter calculation were confirmed by

using the standard computer program with a total dose achieved in a 10 second period and an appropriately much higher continuous dose rate.

In order to present the relevant behaviour in its simplest form a hypothetical model for charge trafficking during the dosing and subsequent glow periods is adopted that consists of a single type of active electron trap and a single type of recombination centre. The physical parameters employed are taken from the above cited study using only the so called green member of the pair of recombination centres. These parameters are:  $E = 1.450$  eV for the central activation energy,  $s = 5.1 \cdot 10^{11}/s$  for the frequency factor, a ratio of retrapping to radiative recombination cross sections of 2.7, and  $N = 10^6/\text{mg}$  for the total available trap states (this corresponds to  $N = 10^{14}/\text{cm}^3$  when corrected for counting efficiency). An incomplete sun bleach is assumed at the beginning of the palaeodose period with  $n_0/N = 0.135$  for the relative trapped electron population density. The presence of a deep saturated thermally disconnected electron trap with a relative population density of  $p/N = 1.197$  is also assumed.

The computer program assumes doses administered in steps of  $10^7$  s duration, delivering  $1.54 \times 10^{-4}$  Gy and generating the appearance of 0.96 electron-hole pairs in the conduction and valence bands. These values are again based on the above cited study; note that the actual e/h pairs created when corrected for counting efficiency yield  $10/\text{cm}^3$  per s). The calculation for a given total dose results in the number and distribution of trapped electrons remaining at the end of the dosing. This is then the starting point for calculation of TL glow curves or isothermal decay curves.

To generate a point in a plot of true age vs. apparent or laboratory age the dosing calculation is done twice, each time resulting in the same number of remaining trapped electrons and therefore (in the absence of thermal quenching) the same area under the glow curve assuming that this signal is used in the dating procedure. One calculation is done with  $\alpha = 0$ , representing laboratory dosing administered so rapidly that no thermal loss of trapped electrons occurs, and the time to reach the required number of trapped electrons is the apparent age. The other calculation is done with given by eqn (2) with  $T$  appropriate for the burial condition, thus turning on the thermal decay and allowing for the loss of trapped electrons. The time required to reach the same number of

trapped electrons in spite of this loss is then the true age to associate with the laboratory dosing signal or the resulting apparent age.

### Results

The quantity  $\Delta$  (= true age - apparent or laboratory age) is found to be very strongly dependent on the burial temperature in addition to the obvious dependence on the width parameter  $a = 2.77 (\text{FWHM})^{-2}$  appearing in Eq (1). Figures 1 and 2 show these results for as a function of apparent age for some typical cases of burial temperature and FWHM values.

Figure 3 displays these same results showing the TL signal (area under the glow curve) in units of  $n/N$  as a function of total dose both under burial and laboratory dosing conditions. As an example the indicated TL signal corresponding to  $n/N = 0.732$  would suggest a sample age of 300 ka based on laboratory dosing whereas the true burial age would be 90 ka older. Contrary to the model employed by Xie and Aitken the ratio of the TL signal generated by a laboratory dose to that developed under burial conditions by the same dose is not a constant, rather the ratio increases sharply with increasing dose. For the case illustrated, materials less than 10 ka in age would involve negligible values of  $\Delta$ .

Figure 4 shows the distributions of trapped electrons for the same value of  $n/N$  selected as the illustration in figure 3. The electron density is per mg-eV (uncorrected for detection efficiency). Under laboratory dosing the distribution has the same Gaussian shape as that for the trap activation energies. Under burial conditions for the same value of  $n/N$  the low energy electrons are severely reduced while the higher energy electrons are correspondingly increased as a consequence of retrapping. The areas under the two curves are equal. Note that the acquired dose is not the same in these two cases. For the same dose the TL signal for burial dosing would give  $n/N = 0.661$ , the loss of  $\Delta(n/N) = 0.071$  representing the thermal decay during burial.

Figure 5 shows the glow curves corresponding to the value of  $n/N$  previously selected as an illustration in figure 3 and displayed as electron distributions in figure 4. Again, as required, the area under the glow curves are equal. Although the peak position and FWHM of the two curves are almost the same, the effect of the reduced

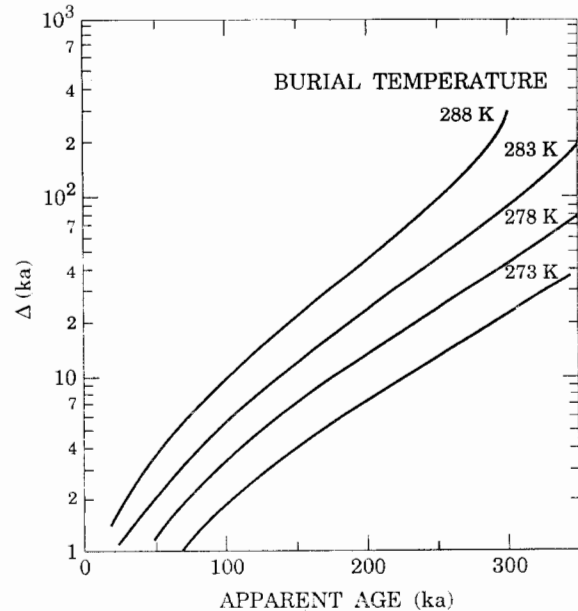


Figure 1.  
The quantity  $\Delta$  (= true age - apparent age) as a function of the burial temperature. The FWHM of the activation energy distribution is 0.186 eV and the central energy  $E_0 = 1.450$  eV.

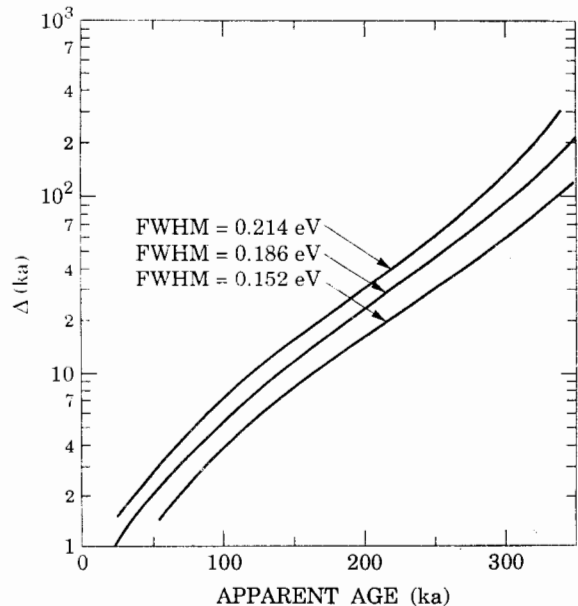
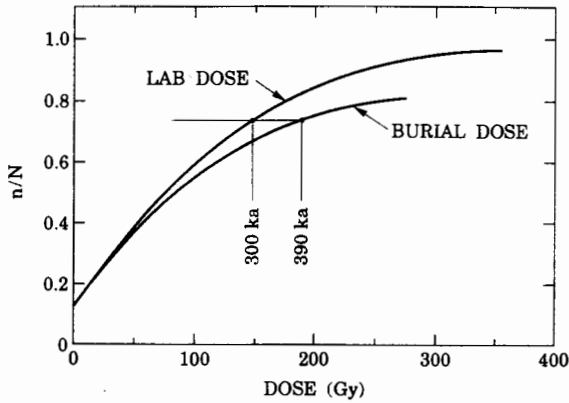
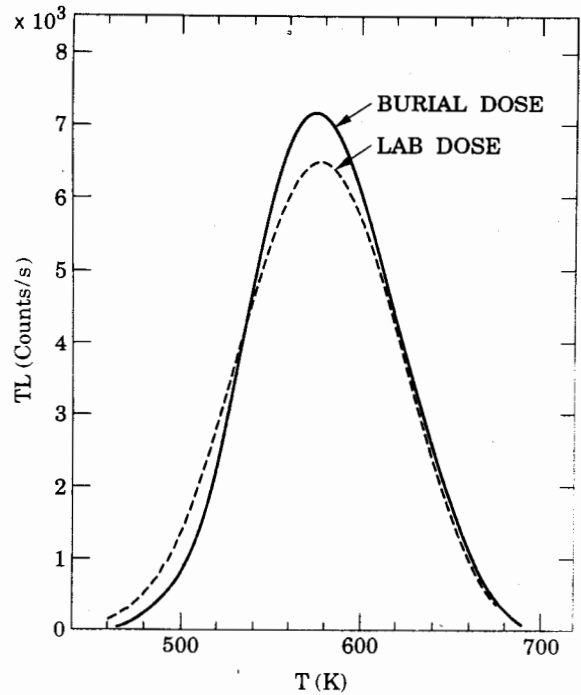


Figure 2.  
The quantity  $\Delta$  (= true age - apparent age) as a function of the width (FWHM) of the activation energy distribution. In all cases the central energy of the distribution  $E_0 = 1.450$  eV. The burial temperature is taken to be 283 K.

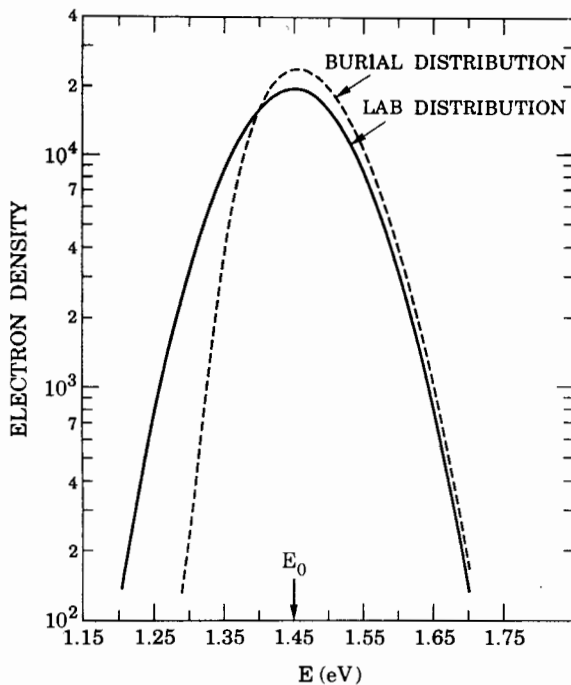




**Figure 3.**  
 The relative trapped electron population density,  $n/N$  as a function of applied dose for both burial and laboratory dosing situations. The burial temperature is 283 K and the FWHM = 0.186 eV with  $E_0 = 1.450$  eV. For the selected value  $n/N = 0.732$ , the corresponding TL signal (area under glow curve) would indicate an apparent age of 300 ka while the true burial age would be 390 ka for the same TL signal.



**Figure 5.**  
 The two glow curves corresponding to the electron distributions shown in fig. 4.



**Figure 4.**  
 The distribution of trapped electrons for the same value of  $n/N$  or TL signal selected as an illustration in fig. 3 contrasting the laboratory and burial dosing. The units for electron density refer to a total number of detectable electrons =  $7.32 \times 10^5/mg$ .

low energy electron population is clearly evident in the steeper initial rise for the burial situation. A standard plateau test comparing the two glow curves should also reveal the shift of the electron population to higher temperatures for the buried sample.

All the above described calculations were also repeated for the two recombination centre model for quartz reported in the earlier investigation (Hornyak 1992). This study demonstrated quite conclusively that the 375 °C peak in quartz involved a distribution of activation energies for the electron traps involved. Results very much like those shown in figures 1-5 followed. One novel feature worth reporting is the substantially larger mid-term effect noted for the uv emission compared to that in the green. This behaviour is attributed to the fact that the glow peak observed in the uv appears at a lower temperature (about 20 °C) than the glow peak observed in the green and hence is more sensitive to the loss of electrons with low activation energies under burial conditions.

**Conclusions**

The existence of a distribution of activation energies for an electron trap site may result in the appearance of a mid-term thermal fading phenomenon. It may readily



happen that supposedly long-lived states will possess a significant number of low activation energy components with intermediate or mid-term life times, long compared to laboratory dosing times but short compared to burial ages. In that event thermal losses that occur during the palaeodose period will not be evidenced in laboratory dosing experiments. Thus, the same absorbed dose will yield a smaller TL signal if acquired during archaeological burial than if administered under laboratory conditions. The usual techniques for age determination will yield palaeodose values that result in underestimating the archaeological age of these sample. To be precise even the various values of TL observed for different added laboratory doses will arise from electron distributions of mixed origins, those combining the original 'natural' population with the added laboratory population in some nonlinear superposition retrapping process.

A complicating phenomenon not considered in this investigation involves the possibility of defect migration. This behaviour can be simulated by allowing thermally driven transitions between the various portions of the activation energy distribution of N itself (Piters and Box, 1991). While this effect by itself would keep the glow area a constant, in connection with the presently described effect it might, if present, alter the escape rate of trapped charge under long burial conditions.

It would appear to be advisable to obtain glow curve data at various ramp heating rates and isothermal decay data on the natural sample to establish the activation energy distribution width and central energy. Such information would assist in sorting out the expected correction to the age underestimation. It would be ideal to use a glow peak occurring at a high enough temperature, of sufficiently narrow temperature width, and behaving with a more or less first order character in order to reduce the need for any important mid-term correction.

However, in general it is rather unfortunate that the amount of palaeodose underestimation due to mid-term thermal fading is very sensitive to the ambient burial temperature, particularly for samples of very old age. This effect will require the archaeologist to not only determine the present temperature of the burial site but to also involve some geological estimation of the temperature history of the site, a more daunting process.

A very favourable result of the present investigation is that for specimens not older than about 10,000 years no significant mid-term correction is called for if dating were based on using this glow peak. After ages over 20,000 years the ratio of observed TL under laboratory dosing to that under burial dosing rapidly increases with increasing dose. Underestimation of archaeological age will certainly occur.

Finally, the results obtained with the two recombination centre model suggests that when TL emission occurs at several wave lengths it may be advisable to examine each emission band separately for its dosing response. One may very well result in a smaller age underestimation, as apparently is the case for feldspar reported by Balescu and Lamothe. These investigators also report a much larger underestimation of age when observing the TL at shorter wavelengths.

#### Acknowledgements

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**PR Reviewer's Comments (S.W.S. McKeever)**

This is a very interesting paper and presents a feasible model for 'mid-term' fading. I think it should be stressed that a distribution of states is probably not necessary for the model to work; closely overlapping discrete states should produce the same effect. One may wonder what a plateau test looks like for the case described.

# Selective bleach: an improved partial bleach technique for finding equivalent doses for TL dating of quartz sediments

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*Improvements are suggested to the partial bleach method of finding equivalent doses for thermoluminescence dating of sedimentary quartz. They are based on the fact that the 325°C peak in the glow-curve can be completely removed by bleaching with light of relatively long wavelength, that it can be preferentially detected by the use of an optical filter transmitting at 380 nm, and by reduction of the glow-curve heating rate. Attention is drawn to a method of reducing extrapolation errors in finding the equivalent dose from dose growth-curves.*

## Introduction

The thermoluminescence (TL) dating of sediments is based on the assumption that the TL clock was reset by exposure to sunlight during the time that the sediment was being laid down. Doubts are likely to exist in the dating of sediments because the samples being dated may not have been exposed to sunlight for 'long enough' to reset the clock.

This has been recognised from the beginning (Wintle and Huntley, 1982; Huntley, 1985). They argued that since you do not know how much the sample has been bleached, then you should assume the worst and base your dating on a component in the TL that bleaches easily, assuming that there is one. They introduced the R- $\Gamma$  Partial Bleach method (R- $\beta$  if the radiation is from a beta source). The essential elements are shown in figure 1 which is adapted from Wintle and Huntley (*op cit*).

A range of doses is added to the natural samples being dated. Half of these are bleached by direct or artificial sunlight for a 'short' time, which is long enough to remove any rapidly-bleached TL component but not long enough to remove any significant amount of slowly-bleached TL. This bleaching may be with or without optical filters, depending on the circumstances. The samples are glowed out and the data are used to define a (natural+dose) growth curve and a (natural+dose+bleach) growth curve. The two curves are extrapolated to intersect at a point that defines the Equivalent Dose,  $D_{eq}$ .

## Optical Improvements

Franklin and Hornyak (1990) and Prescott and Fox (1990) have pointed out that, for quartz, there is an easily-identifiable, rapidly-bleached TL component, viz, the 325°C peak which emits at 380 nm. It is possible to bleach this component completely out selectively, without affecting other glow-curve peaks, with a short exposure to light of wavelength longer than about 475 nm (Spooner et al., 1988); and to enhance its detection in emission by the use of optical filters which are selective for ultra-violet, e.g. UG11, UG2, or either of these in conjunction with a 7-59.

These two practices can be profitably introduced to refine conventional R- $\Gamma$  (R- $\beta$ ) partial bleach:

- 1) By carrying out the partial bleach with full sun filtered through a long wavelength pass filter cutting at 475 nm or longer, e.g. GG475 or Chris James 101 (a theatrical filter). Bleaching times of the order of tens of minutes are sufficient to ensure that the 325°C peak is completely removed but that other components are unaffected. This component is thus effectively totally bleached.
- 2) By using an ultra-violet filter, as described above, during glow out.

We have tried out these modifications on a range of quartz sediments drawn from Australian sites. We have already reported (Prescott and Purvinskis 1991) that application of the procedures yields zero age for modern sites that formerly gave non-zero ages. Zero age

Figure 1.

Schematic, showing the application of conventional partial bleach to finding equivalent dose  $D_{eq}$  (after Wintle and Huntley, 1982)

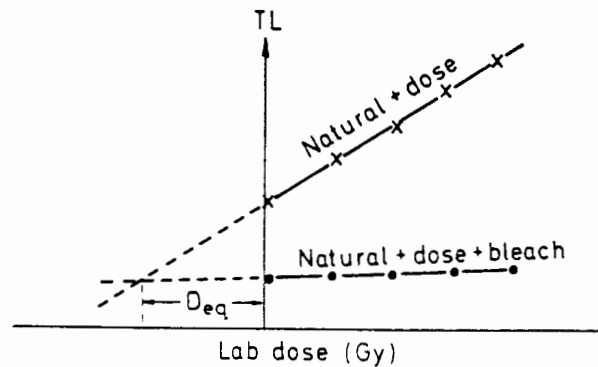
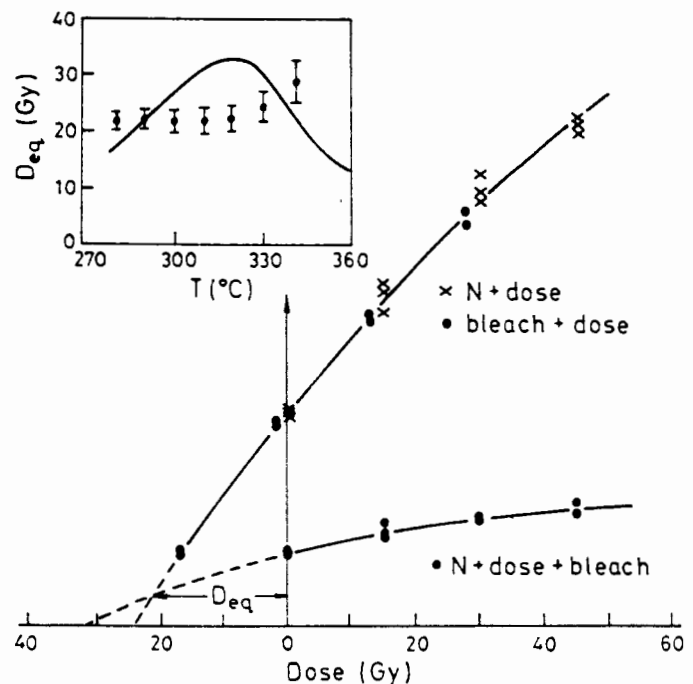


Figure 2.

Dose curves for sample TC2S/65 to which selective bleach procedures, as described in the text, have been applied. The inset shows a portion of the natural glow curve and the corresponding plateau. The rise on the upper side of the peak is attributed to the presence of the tail of the unbleached 375°C peak.



recovery was one of the tests proposed by Wintle and Huntley (1982) in their early review paper on TL dating of sediments.

So far as the use of u-v filters is concerned, we have found that the degree of selectivity of the 325°C peak differs from sample to sample. In many of them (such as PJ1ES/75 shown in figure 1 of Prescott and Fox, 1990) the background of unwanted peaks is reduced by a factor of five or more with respect to the 325°C peak. In samples where a long-lived peak of lower temperature is present, it interferes strongly since its emission lies within the pass band of the above filters; also it is not easily reduced by bleaching. In other samples, e.g., our Le Fevre Peninsula sequence, the 325°C peak is hardly present at all.

These latter difficulties would be present, of course, whether the selective bleach modifications were applied or not. The new procedures should result in at least some improvement in the estimates of equivalent dose in all cases.

The selectivity of the 325°C peak can be further improved by heating as slowly as practicable, so as to reduce thermal quenching (Wintle 1975) which affects this peak more than others (Spooner personal communication, 1992). We are grateful to A.D. Franklin for drawing our attention to this aspect.

#### Curve fitting

As indicated above, the partial bleach method requires the (natural + dose) growth curve and (natural + dose + bleach) growth curve to be extrapolated. The possibility

of consequent extrapolation errors is well-known and this has been extensively discussed in the literature on TL and ESR. This problem becomes the more challenging, the further the growth curves depart from linear. Procedures to minimise extrapolation errors have been widely discussed and are summarised elsewhere. (Berger 1988, Prescott et al., 1993).

The estimate of equivalent dose can be improved by reducing extrapolation errors using the fitting technique first suggested by Valladas and Gillot (1978). Faced with saturating dose curves for heated materials, they used the shape of the second-glow dose curve to model the missing continuation of the first-glow dose curve for doses less than the natural dose  $N$ , making allowance for a change in sensitivity after heating. Readhead (1982; 1984; 1988) independently applied and developed a similar idea for sediments; and further developments have been described by Huntley et al. (1993), or Prescott et al., (1993).

An example of the application of the foregoing techniques to a quartz sample, TC2S/65, from the aeolian infill of an earthquake scarp near Tennant Creek, Australia, is shown in figure 2. Bleaching was 30 minutes of natural sunlight through a Chris James 101 yellow filter with a cut-off to 1% at 475 nm. The optical filter was UG11+7-59, the photomultiplier 9635QA and the heating rate 5 K/s. The equivalent dose is  $21 \pm 1$  Gy and the TL age  $14 \pm 1$  ka.

#### Further comment

The foregoing applies to quartz. We have found no samples where the 325°C peak was not removed to a negligible level in bleaching times of the order of minutes of full sun. However, as was shown by Robertson et al. (1991) in a study of a representative collection of feldspars, there appears to be little evidence in the alkali feldspars for the existence of any readily bleachable peak with behaviour similar to that of the 325°C peak in quartz, although there was evidence for it in an oligoclase and a suggestion that easy bleaching may be associated with anomalous fading. The optical modifications will therefore not necessarily improve the estimates of equivalent dose in feldspars or undifferentiated mineral samples.

*A propos* of quartz, it should be noted that most ultraviolet filters also transmit in the red. Since many quartz samples emit in the red (Huntley et al., 1988),

unwanted light may be recorded unless this possibility is provided for.

One of the tests suggested by Wintle and Huntley (1982) for TL dating methods was that there should be an age plateau covering a range of temperatures, indicating that the TL originates from states of long lifetime. This test needs restating for selective bleach in quartz because we are most probably dealing with a single trap. We have found that there is a plateau covering the width of the 325°C peak and we should be concerned if there were not. The plateau for TC2S/65 is shown as an inset in figure 2.

Wintle (1975) shows that the lifetime of this level is about 30 Ma and Spooner (personal communication 1992) finds 20 Ma; this would appear to remove the uncertainty in the stability of the TL. We have found no evidence for anomalous fading on limited tests, although we note in passing that there is some evidence for long term fading in the 280°C peak.

#### Acknowledgements

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## Notices

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### Dr Jerzy Butrym

We report with sadness the sudden death a few weeks ago of Dr Jerzy Butrym of the Department of Physical Geography of the Marie Curie Sklodowska University of Lublion, Poland. Dr Butrym had been in the field of TL dating of sediments for about ten years.

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## Small Ads

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### Software

A number of visitors to Adelaide have expressed interest in obtaining our regeneration program. I have therefore tidied it up for general use and am offering it for sale.

The program takes two data sets, e.g.  $N+\gamma$  and  $N+\text{sun}+\gamma$ , and finds the dose shift of one from the other, with optionally an intensity scaling factor. This is achieved by jointly fitting the two data sets to a single function by a maximum likelihood method. Allowance for a modern analogue is provided.

The functions currently available are linear, cubic, saturating exponential, and saturating exponential plus linear.

Uncertainties in all parameters are provided. More details can be found in the article by Prescott, Hutton and Huntley in the last issue of *Ancient TL*, and in our forthcoming article in *Quaternary Science Reviews*.

The price is \$300 and the software is written for IBM PCs and compatibles. If you wish to obtain a copy please send a pre-payment or institutional purchase order to:

D.J. Huntley, Physics Department, Simon Fraser University, Burnaby, B.C., V5A 1S6. Canada.

If you wish a trial before purchasing please send a set of data on a disc and you should receive a sample output.