

# On the selection of dose points for saturating exponential ESR/TL dose response curves

Rainer Grün and Edward J. Rhodes

Subdept. of Quaternary Research, Cambridge University, Free School Lane, Cambridge CB2 3RS, UK.

## Introduction

During the past few years, it has been suggested that linear extrapolation of the ESR/TL dose response may not be an appropriate procedure for determining the equivalent dose,  $D_E$ , when using the additional dose method (Apers et al., 1981; Berger et al., 1987; Poljakov and Hütt, 1990; Grün and Macdonald, 1989). In this paper we have performed numerical simulations of exponential dose response curves using random numbers which are more fashionably called Monte Carlo simulations. We wanted to investigate the influence of the applied dose distribution, the precision of the ESR/TL intensity measurement, and the number of data points on error in the  $D_E$  determination.

Franklin (1986) examined this question of optimization for linear fitting and concluded that, from a mathematical point of view, the minimum error in  $D_E$  determination might be attained from repeated measurements of the lowest and highest dose points. He suggested that the measurement of a dose point in the middle may be used to demonstrate linearity of the dose response. This may be directly applicable to single exponential fitting, when the dose response is converted into a straight line by fitting  $-\ln(1 - I/I_{\max})$  versus the artificial dose, where  $I$  is the measured ESR signal intensity and  $I_{\max}$  is the maximum ESR signal intensity when all traps are filled (Apers et al., 1981).

Barabas (1989) carried out some Monte-Carlo simulations on saturating exponential functions and came to the conclusion that at least 12-15 measurements with a maximum gamma irradiation dose,  $D_{\gamma(\max)}$ , of 1500 Gy ought to be carried out in order to achieve an error in  $D_E$  determination of about 10% for a  $D_E$  of 500 Gy and where the characteristic saturation dose,  $D_0$ , (see figure 1) is about 1500 Gy. However, as far as we are aware, no systematic numerical simulations have been carried out in order to determine the influence of the number of measurements and the placing of the radiation dose steps on the uncertainty in  $D_E$  determination. In many ESR or TL dating studies, the number of dose points and their placing seems very random.

## Experimental Procedures

The basic assumptions and procedures for our computational experiments are as follows: the dose response is described by a single saturation function of the type:  $I = I_{\max} \{1 - \exp(-(D_{\gamma} + D_E)/D_0)\}$  where  $D_{\gamma}$  is the additional artificial gamma dose and  $D_0$  is defined in

figure 1. In order to reduce computation time, the data points were converted into a straight line function by rearranging the above equation so that we could plot  $-\ln(1 - I/I_{\max})$  versus  $(D_{\gamma} + D_E)/D_0$ . The iteration process followed the following procedure: after inserting an initial value for  $I_{\max}$  into the above equation,  $D_0$  and  $D_E$  are readily calculated by applying the method of least squares (LSQ) to the straight line (see eg Berger and Huntley, 1986).  $I_{\max}$  was then iteratively optimized using the method of least squares to the original saturating exponential function and the whole process was repeated until changes in  $I_{\max}$  were smaller than 0.1%. All data points were given the same precision. Although this straight line conversion may not always give the best fit when compared to a three parameter optimizing procedure (see Grün and Macdonald, 1989), the differences of the calculated values compared to the latter approach (where  $I_{\max}$ ,  $D_0$ , and  $D_E$  are optimized at the same time) are usually smaller than 1% for all determined parameters. We used a random number generator to create a Gaussian distribution with a preset precision around a given mean value. The model assumes that the uncertainty in the dose can be neglected (see also Berger and Huntley, 1986; Berger et al., 1987).

Our calculations are based on a typical experimental ESR dose response curve of the paramagnetic centre at  $g=2.0018$  of tooth enamel with  $D_0 = 3333$  Gy. Since the results are generally valid for any  $I_{\max}$  value and  $D_E/D_0$  ratio, we also give the latter value.

Three basic different dose distributions were investigated:

- model 1: even dose spacing, (e.g. 0, 2, 4, 6, 8 ... );
- model 2: doubling of an initial dose step (e.g. 0, 1, 2, 4, 8.... );
- model 3: exponential dose distributions  
 $[D_{\gamma} = 10 \{ \exp(bN) - 1 \}]$ , where  $b$  is a constant (in the range 0.1 to 0.9) and  $N$  ranges from 0 to 9.

We have performed our simulations for various preset  $D_E$  values of 10, 50, 100, 500, 1000 and 5000 Gy (which correspond to  $D_E/D_0$  ratios of 0.003, 0.015, 0.03, 0.15, 0.3 and 1.5, respectively). The basic computational procedure was as follows: first a saturating exponential function was defined by selecting preset values for  $D_0$  and  $I_{\max}$  followed by the

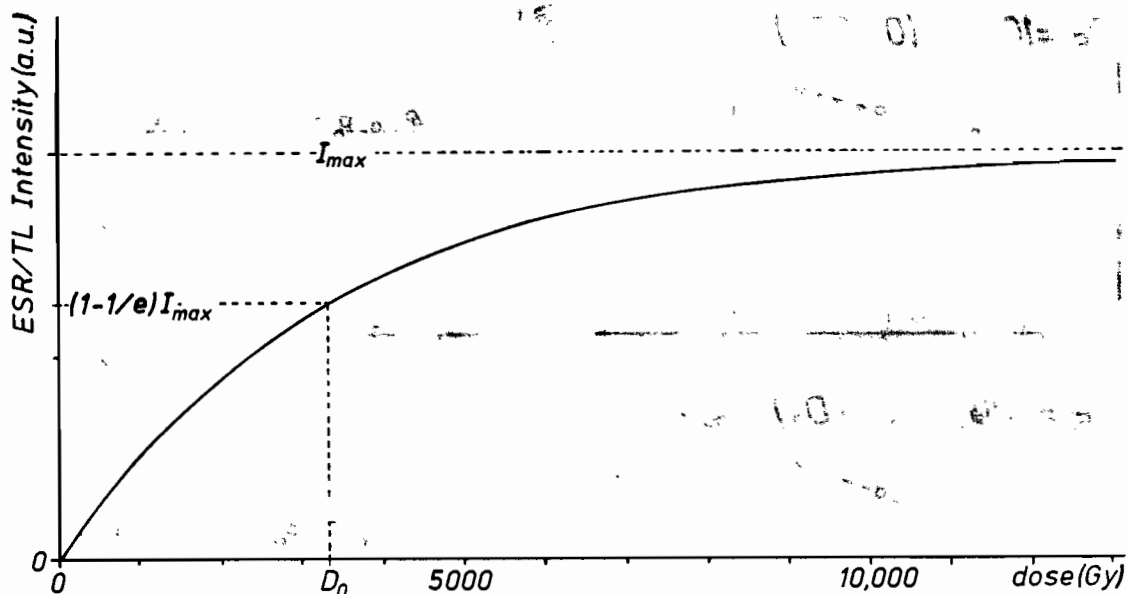


Figure 1. The dose response curve that was used in the computer simulation.  $I$ : measured ESR/TL intensity;  $I_{max}$ : maximum intensity when all traps are filled;  $D_0$ : characteristic saturation dose.

selection of the preset  $D_E$  and the number of different dose points. Then, the precision of the intensity measurements was defined and a dose distribution model with a particular  $D_{max}$  value was chosen. The random number generator then produced successively 1000 sets of ESR/TL intensity values within the preset precision. These sets were used to determine 1000  $D_E$  values.

We have checked the mean and the standard deviation of the randomly produced intensities and observed that the mean of the generated intensities differed less than 0.1% from the intensity values of the defined function, and the standard deviation showed variations in the range of 5% from the preset value. The computations were carried out on a computer with a 25 MHz 80486 CPU. The program was written in Microsoft Quick Basic.

## Results

### Effect of the maximum irradiation dose, $D_{\gamma(max)}$

In the first experiment, the precision ( $1\sigma$  - standard deviation) of each ESR/TL intensity is 2% (which is probably smaller than in most routine dating analyses) and 10 data points per dose response curve were used (most ESR dating results are based on fewer points).

Figures 2a and 2b show the standard deviation (s.d.) in the determination of  $D_E$ , and the systematic deviation of the computed mean  $D_E$  value from the preset  $D_E$ . The exponential dose step models 2 and 3 result in smaller errors than the even dose distribution (model 1). Additionally, the minimum error for models 2 and 3 is far less dependent on the applied maximum dose,  $D_{\gamma(max)}$ , than model 1.

In the  $D_E$  range from 10 to 1000 Gy (0.003 to 0.3  $D_0$ ), the optimum  $D_{\gamma(max)}$  value for model 1 is about 4 to 6 times  $D_E$ , whereas the optimum  $D_{\gamma(max)}$  value for models 2 and 3 is about 10 times  $D_E$ . For the optimum  $D_{\gamma(max)}$ , the s.d. of the  $D_E$  determination is in the 5-7% range. Above 1000 Gy (0.3 $D_0$ ), the calculated error becomes more critically dependent on the selection of  $D_{\gamma(max)}$  and an  $D_E$  determination of 1.5  $D_0$  has a s.d. in the range of at least 25%. This error rapidly increases with a change in  $D_{\gamma(max)}$ . Figure 2b seems to imply that  $D_{\gamma(max)}$  should not exceed about 3 times  $D_0$  when  $D_E \geq D_0$ .

In the  $D_E$  range 10 to 100 Gy (0.003 to 0.03  $D_0$ ), a low  $D_{\gamma(max)}$  value seems to lead to a systematic underestimation in  $D_E$ . However, if  $D_{\gamma(max)} > 5$  times  $D_E$ , this underestimation is less than 3%. In the  $D_E$  range of 500 to 1000 Gy (0.15 to 0.3  $D_0$ ), high  $D_{\gamma(max)}$  values lead to systematic overestimation of  $D_E$ . Again, if  $D_{\gamma(max)}$  is 10 times  $D_E$  or smaller, the overestimation is below 3%.  $D_E$  estimations in the 5000 Gy (1.5  $D_0$ ) range seem to be associated with severe overestimation (>15%).

The determination of  $I_{max}$  is also strongly dependent on the choice of  $D_{\gamma(max)}$ : a close approximation to the preset values of  $I_{max}$  and  $D_0$  is possible only when  $D_{\gamma(max)}$  is in the range of  $D_0$ . However, the correct estimation of  $I_{max}$  and  $D_0$  do not necessarily influence the precision and accuracy of the  $D_E$  determination. The  $D_E$  determinations with the smallest uncertainties in the low dose range (10 to 100 Gy or 0.003 to 0.03  $D_0$ ) may be associated with errors of many hundred percent in the determination of  $I_{max}$  and  $D_0$ .

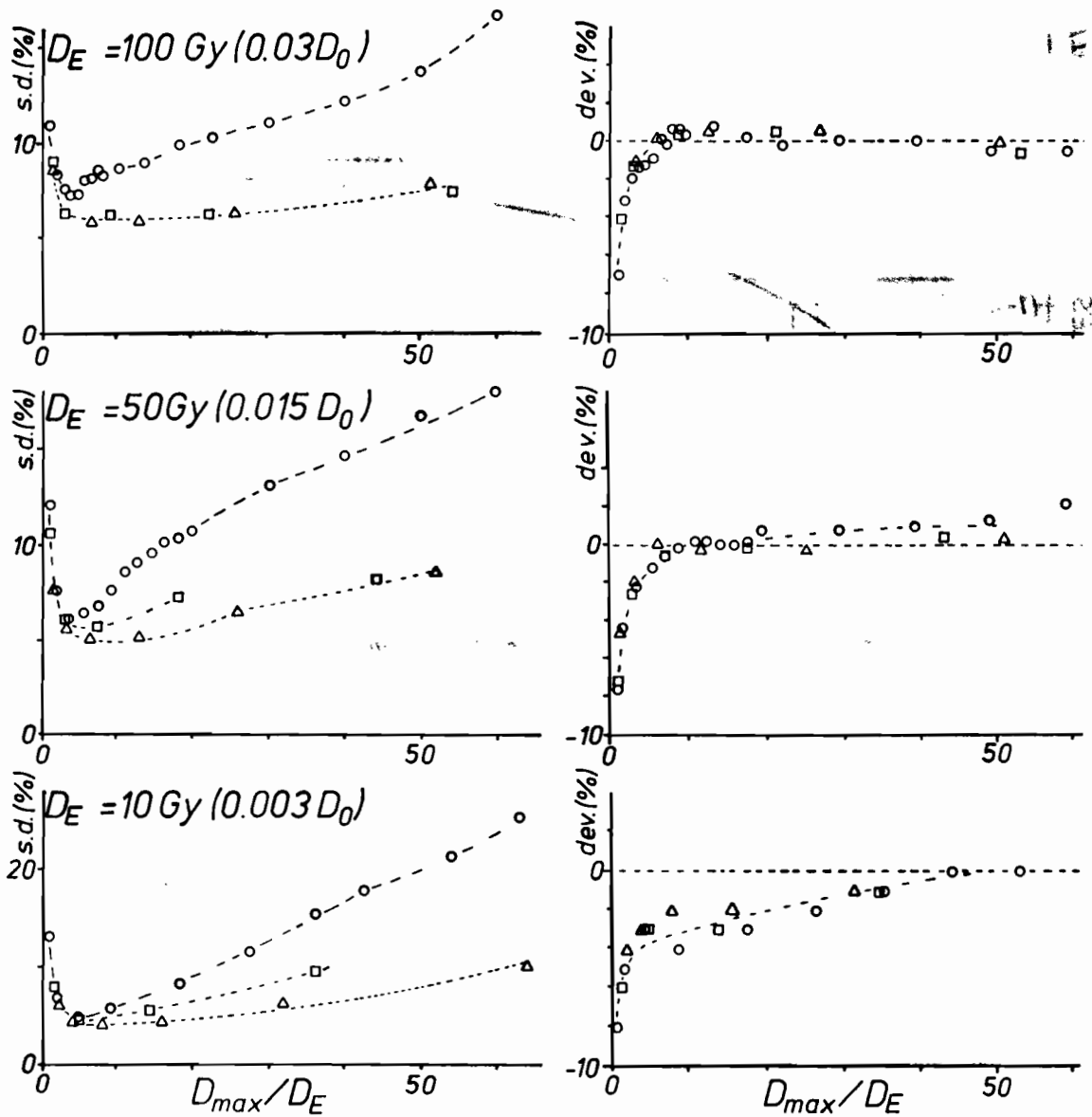


Figure 2a. Standard deviation, *s.d.*, (left) and systematic deviations (right) in  $D_E$  determination for preset  $D_E$  values of 10, 50 and 100 Gy (0.003, 0.015 and 0.03  $D_0$ ), and a precision in ESR/TL intensity of 2%. Dose spacing according model 1 (circles), model 2 (triangles) and model 3 (squares).

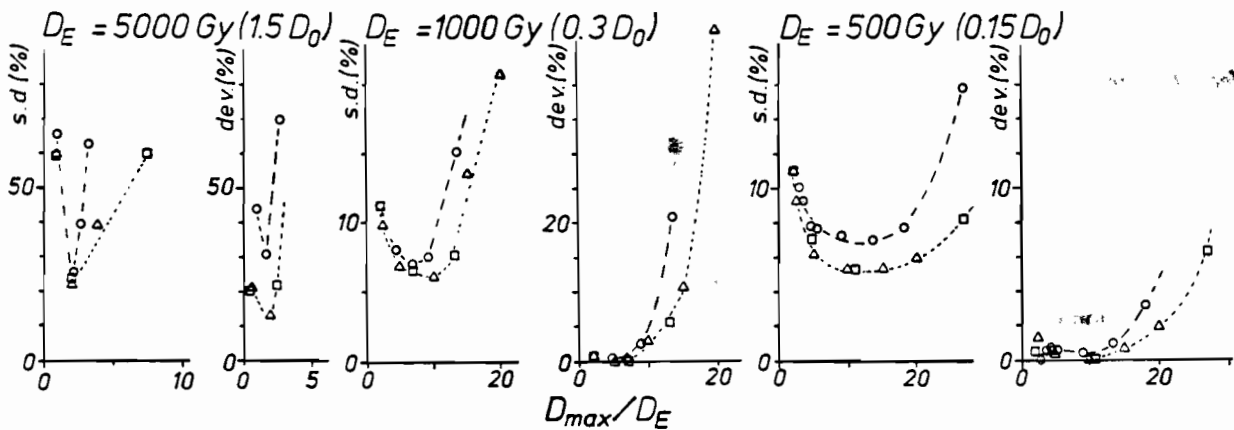


Figure 2b. Standard deviation, *s.d.*, (left) and systematic deviations (right) in  $D_E$  determination for preset  $D_E$  values of 500, 1000 and 5000 Gy (0.15, 0.3 and 1.5  $D_0$ ), and a precision in ESR/TL intensity of 2%. Dose spacing according model 1 (circles), model 2 (triangles) and model 3 (squares).

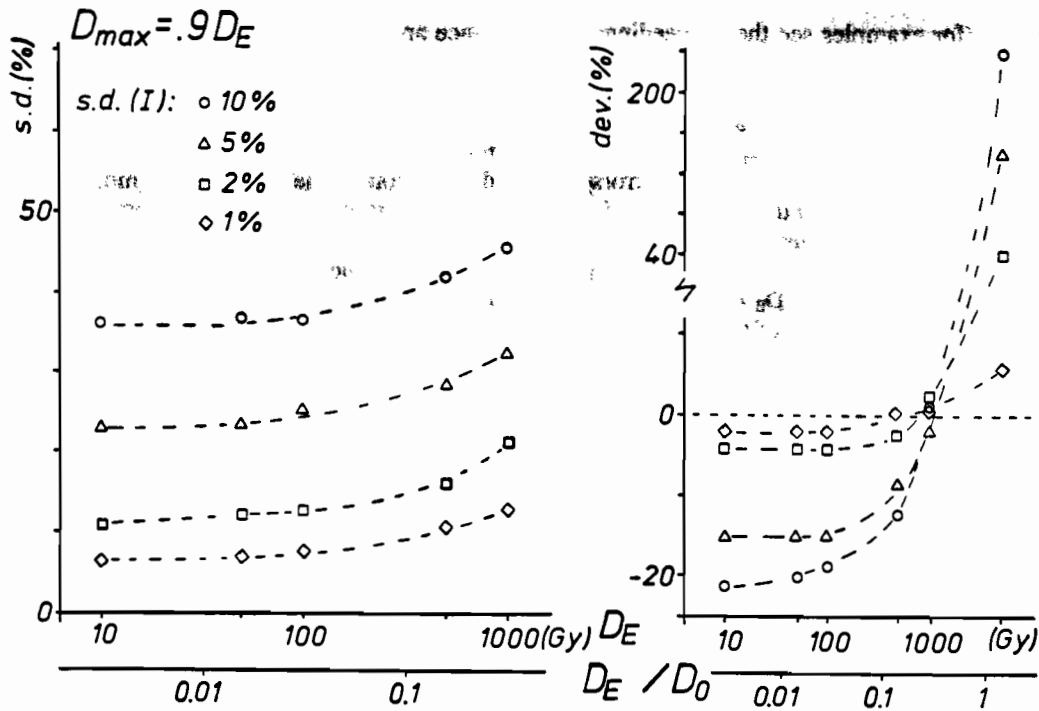


Figure 3. Standard deviation, s.d., (left) and systematic deviations (right) in  $D_E$  determination for  $D_{(max)} = 0.9 D_E$  using model 1. X-axis: preset  $D_E$ .

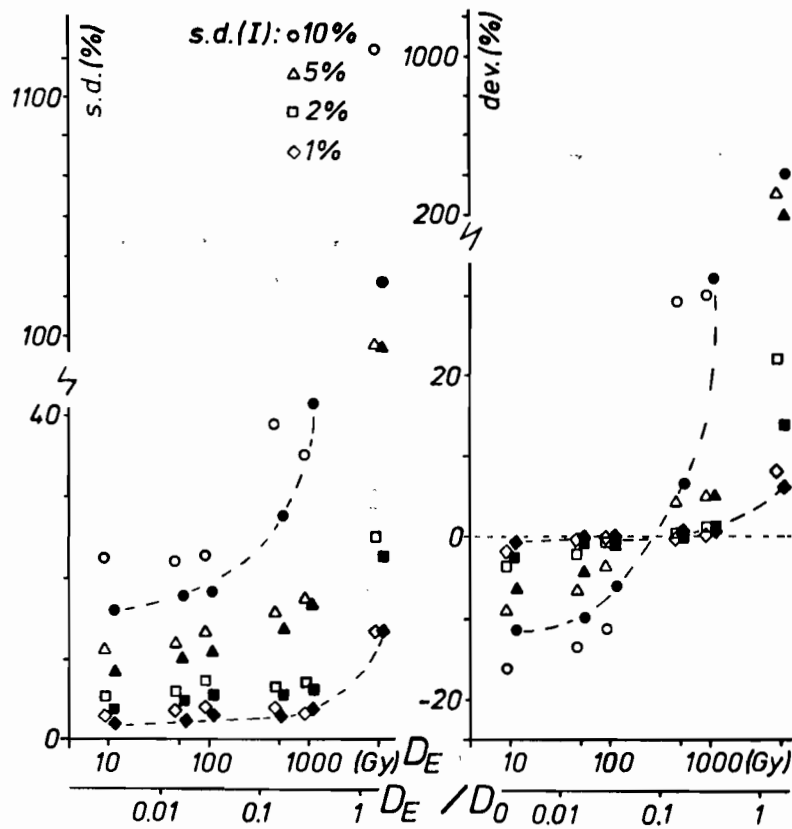


Figure 4. Standard deviation, s.d., (left) and systematic deviations (right) in  $D_E$  for different precisions in ESR/TL intensity and optimum  $D_{(max)}$  according Figure 2. Open symbols: dose spacing model 1; closed symbols: dose spacing model 2. X-axis: preset  $D_E$ .

Some publications (for examples see the Proceedings of the TL/ESR seminars in *Quaternary Science Reviews* vol. 7 (1988) or *Nuclear Tracks and Radiation Measurements* vols 10(1985), 14(1988) and 18(1991)) show dose response curves where  $D_{\gamma(\max)}$  is smaller than the determined  $D_E$  value. Figure 3 shows the errors that are involved in these cases (10 dose points). This selection causes a s.d. in  $D_E$  of at least 10% (with the same conditions as for the above simulation). Both the s.d. and the systematic deviation in  $D_E$  determination increase rapidly with lower precision of the data points (see also figures 4 and 5).

#### *Effect of the precision of the ESR/TL intensity.*

Figure 4 shows the  $D_E$  estimations for different precisions of the generated ESR/TL intensities (10 dose points, model 1,  $D_{\gamma(\max)}$  selected to produce minimum errors in  $D_E$ ). For the s.d. in  $D_E$  to be smaller than 10%, the precision of the measured ESR/TL intensities must be better than 5% (in the  $D_E$  range of 10 to 1000 Gy [0.003 to 0.3  $D_0$ ]). A precision of 1% still causes a s.d. of > 10% for an  $D_E$  determination of 5000 Gy (1.5  $D_0$ ). Simulations with precisions of 5% and less are also associated with large systematic underestimations (10 to 100 Gy [0.003 to 0.03  $D_0$ ]) and overestimations (>500 Gy [0.15  $D_0$ ]).

#### *Effect of the number of dose points.*

Figure 5 shows that the error in the  $D_E$  determination cannot be significantly improved by additional measurements within the same dose range. In all cases, an  $D_E$  determination from 10 dose points with a 1% precision has a smaller s.d. and a smaller systematic deviation in  $D_E$  than from 40 dose points with a 2% precision. In the  $D_E$  range of 10 to 1000 Gy (0.003 to 0.3  $D_0$ ),  $D_E$  estimations based on 4 different dose points with a precision of 1% lead to better results than  $D_E$  estimations derived from 40 different dose points with a precision of 2%. This result parallels for exponential fitting the predictions of Franklin (1986) for linear fitting. It therefore seems advisable to put any spare energy into decreasing the uncertainty of the measured ESR/TL intensities (see also Berger and Huntley, 1989).

#### **Discussion**

Our simulation shows that significant deviations in  $D_E$  determination occur only at relatively small  $D_{\gamma(\max)}/D_E$  ratios (<3-5) where all dose points are relatively closely spaced or when the ESR/TL intensities have low precision and are dependent on the computational procedures that we have selected for our calculations: the largest possible  $D_E$  for a given data set is determined by a linear function (i.e.  $I_{\max}$  approaches infinity). While this determines the maximum possible  $D_E$ , there is basically no restriction for the determination of the minimum possible  $D_E$  and the resulting  $D_E$  values do not show a Gaussian distribution around the mean value. As soon as the curvature of the dose response curve is strong enough that no fitting effectively results in a straight line, the  $D_E$  distribution becomes symmetrical.

Since an exponential distribution of dose steps is less affected by the correct selection of  $D_{\gamma(\max)}$  and leads to slightly smaller errors in  $D_E$  than equally spaced dose distribution, it seems to be optimal to select a suitable maximum irradiation dose and to use an exponential distribution of the dose points (models 2 and 3). Our simulations show that any effort in minimizing the error in the  $D_E$  determination ought to go into minimizing the deviation of the ESR/TL measurement by repeated measurements at one dose, rather than measuring additional dose points. Figure 5 implies that 4 dose points with a 1% precision in the ESR/TL intensity lead to better results than 40 different dose points with 2% precision (the former can be achieved by measuring 4 different dose points with 4 aliquots with 2% precision resulting in a 1% standard error of the mean). However, we do not generally recommend the measurement of three or four additional dose steps only, because it will not allow the recognition of systematic deviations from the assumed mathematical model of the dose response curve. Additionally, see Berger's comments for restrictions in TL.

Our simulations suggest that in order to assess the errors that are involved in the  $D_E$  estimation, it is necessary to estimate the precision of a routine ESR/TL measurement (see also Berger and Huntley 1989). This precision may vary strongly from mineral to mineral and is generally larger in TL than in ESR (partly because the latter technique can measure much larger samples). Berger and Huntley (1986, 1989) and Berger et al (1987) point out that the intrinsic error of a TL measurement is not known. Little is known about the intrinsic error of an ESR measurement; repeated measurements of ESR intensities of speleothems varied between 2-5% (Grün, 1985), the reproducibility of the measurement of the OHC and E' centres in quartz are in the range of 1.5% (Rhodes, unpublished data) and the precision of ESR measurements in mollusc shells was assumed to be in the range of 3% (Barabas, 1989).

One way of estimating the precision of intensity measurements of a particular material is to calculate the scatter of the data points around the regression line for a large number of similar samples. ESR measurements of tooth enamel lie normally within less than 1% around the exponential regression line (using both equal weights and weights inverse proportional to the square of the intensity) and ESR measurements of corals lie in the range of 1 to 2% around the regression line (Grün, unpublished data).

We wish to emphasize that the reported results are valid only if the ESR/TL dose response curve is precisely described by a single saturation function. In cases where this assumption is violated, for example by supralinearity, the occurrence of several overlaying saturation functions (Katzenberger and Willems, 1988), trap production (Grün, 1990; Berger, 1990), or occurrence of a "dose pit" (see Shlukov and Shakhovets, 1987; Grün, 1991), all errors and deviations that result from applying an incorrect mathematical function are much larger.

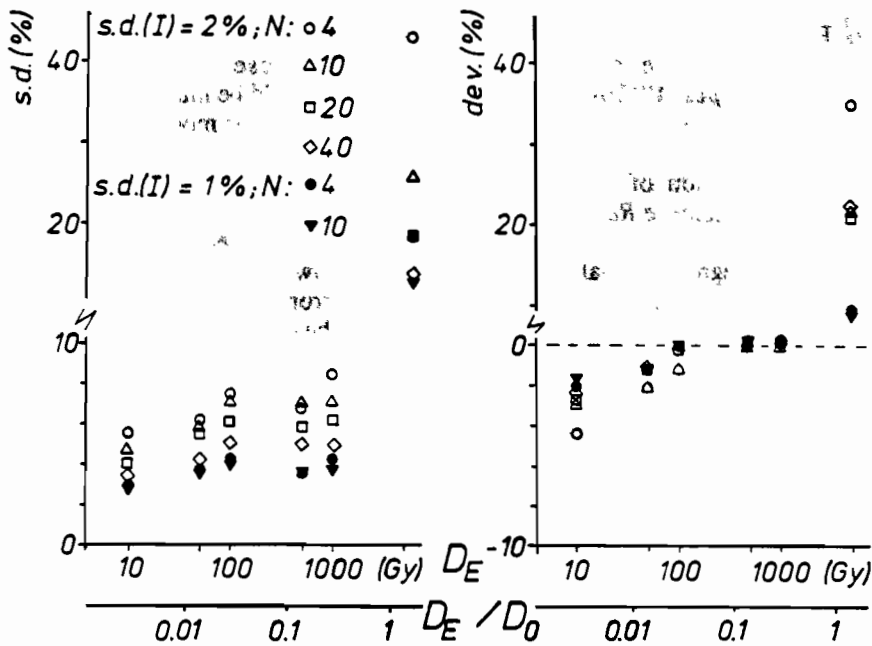


Figure 5. Standard deviation, *s.d.*, (left) and systematic deviations (right) in  $D_E$  determination for different number of data points,  $N$ , and optimum  $D_{E(max)}$  according Figure 2 using dose spacing model 1. X-axis: preset  $D_E$ .

Initial simulations using weights of the ESR/TL intensity that are proportional to the ESR/TL intensity (as suggested by Berger and Huntley, 1989) show that the results reported above are qualitatively correct, however, this approach leads to smaller overall errors. These results will be reported in a forthcoming paper.

### Conclusion

The size of the  $D_E$  error in our simulation is critically dependent on the precision of the measured ESR/TL intensity. Additionally, the uncertainty in the  $D_E$  estimation is dependent on the selected  $D_{E(max)}$  and the dose step distribution. This suggests that the samples should be monitored before irradiation in order to estimate the approximate  $D_E$ . Additional effort should go into increasing the precision of the data points. Measurements of less than 7 dose points can be recommended only if the dose response curve is known to follow a single saturation function.

The curve fitting procedure used in this paper is routinely applied in our laboratory; however, without further systematic simulations, the results cannot directly be generalized for other fitting procedures (e.g. using different weighting of the data points: see Berger and Huntley, 1986 and 1989; Berger et al., 1987) and methods of error assessment.

### Acknowledgements

This study was partly funded by a NERC research fellowship to E. Rhodes and a SERC grant to N. J. Shackleton. We thank the referee for many helpful suggestions.

### References

- Apers, D., Debuyst, R., DeCanniere, P., Dejehet, F. and Lombard, E. (1981) A criticism of the dating by electron paramagnetic resonance (ESR) of the stalagmitic floors of the Caune de l'Arago at Tautavel. In: DeLumley, H. and Labeyrie, J. (eds) *Absolute dating and isotope analysis in prehistory - methods and limits*. Preprint, S. 533-550.
- Barabas, M. (1989) ESR-Datierung von Karbonaten: Grundlagen, Systematic, Anwendungen. Inaugural Dissertation, Universität Heidelberg, 161 p.
- Berger, G.W. (1990) Regression and error analysis for a saturating-exponential-plus-linear model. *Ancient TL* 8, 23-25.
- Berger, G.W., and Huntley, D.J. (1986) Linear regression of TL data. *Ancient TL* 4, 26-29.
- Berger, G.W., and Huntley, D.J. (1989) Test data for exponential fits. *Ancient TL* 7, 43-46.
- Berger, G.W., Lockhart, R.A., and Kuo, J. (1987) Regression and error analysis applied to the dose-response curve in thermoluminescence dating. *Nuclear Tracks and Radiation Meas.* 13, 177-184.
- Franklin, A.D. (1986) Extrapolation errors in linear regression. *Ancient TL* 4, 31-35.
- Grün, R. (1985) Beiträge zur ESR-Datierung. Sonderveröffentlichungen des Geologischen Instituts der Universität zu Köln, 59, 1-157.
- Grün, R. (1990) Dose response of the paramagnetic centre at  $g = 2.0007$  in corals. *Ancient TL* 8(3), 20-22.
- Grün, R. (1991) Potential and problems of ESR dating. *Nuclear Tracks and Radiat. Meas.* 18, 143-153

- Grün, R., and Macdonald, P.D.M. (1989) Non-linear fitting of TL/ESR dose response curves. *Applied Radiation and Isotopes* **40**(10-12), 1077-1080.
- Katzenberger, O., and Willems, N. (1988) Interferences encountered in the determination of AD of mollusc samples. *Quaternary Science Reviews*, **7**(3/4), 485-489.
- Poljakov, V., and Hütt, G. (1990) Regression analysis of exponential palaeodose growth curves. *Ancient TL* **8**, 1-2.
- Shlukov, A.I., and Shakhovets, S.A. (1987) Kinetic studies of quartz thermoluminescence as applied to sediment dating. *Ancient TL* **5**(1), 11-15.

#### PR Reviewer's Comment (Glenn Berger)

This is a welcome addition to the published discussions on error analysis in TL/ESR dating studies. Monte Carlo simulations can provide powerful discrimination among the effects of the various inputs (assumptions and parameters) to regression and error models. Hopefully, such simulations will continue to be carried out in this area. However, as the authors point out, there are limitations to the conclusions one can draw from this first such detailed simulations in TL/ESR studies.

The assumption of equal weights to all data points (rather than weighting by inverse variance such as in Berger et al., 1987) may invalidate the author's conclusion for situations where the extrapolation from the range of artificial doses is relatively large (e.g.,  $D_e/D_0 > 0.1$  here, or relative extrapolations of  $>40\%$  in the terminology of Berger et al., 1987). It would be instructive to test this possible invalidation by simulations with models that use unequal weighting.

The authors' results also re-emphasize the importance of acquiring a firmer knowledge of the "intrinsic" variance or precision in each measurement of TL/ESR intensity. As discussed elsewhere (Berger et al., 1987; Berger and Huntley, 1989), there may be serious difficulties with attempting to measure reliably this variance for TL signals. However, it seems likely that the intrinsic variance of ESR signals can be measured reliably, though different values may be needed for each type of material. In this context ESR has an advantage over TL because signal intensities can be replicated on the same subsample, thus factoring out the disc-to-disc and related effects associated with replication of TL signals. Therefore, if reliable estimates of intrinsic variance in ESR signals can be obtained, then it would be straightforward to apply a Chi-squared goodness-of-fit test routinely to ESR growth curve data, and thereby provide an objective criterion for the recognition of spurious (outlier) data points (see e.g., Brooks et al., 1972). Of course, application of such a test depends for its validity on our confidence in the particular regression model that we choose (e.g., Berger 1990).

There is a further point to note. Although the authors qualify their results as applicable only to their curve-

fitting procedure, their discussion of the effects of the number of dose points (second paragraph in Discussion) could be misinterpreted as relevant to other "procedures" (error models), which it is not. That is, their model shows that increasing the number of data points at one dose point has a greater effect than if the same increase were distributed among several dose points. In contrast, equations 4 and 10 of Berger et al. (1987) show that with that error model (constant percent error, weighting inversely proportional to variance) the error in  $D_E$  can be reduced by any increase in the number of data points, without regard to how these data points are distributed among the chosen dose points, or whether new dose points are added. Of course, the low-dose (low TL intensity) data points have greater weight in this error reduction. The distinction of this behaviour from that of the authors' procedure may lie in their operational definition of precision. These comments are meant to draw further attention to the continued need for comparison of different error analysis models using common data sets (eg Berger and Huntley, 1989).

The above additional comments thus re-emphasize the importance of "model awareness" when interpreting computed regression errors, and the importance of acquiring a firmer knowledge of "intrinsic" TL/ESR errors, as discussed above.

- Brooks, C., Hart, S.R. and Wendt, I. (1972) Realistic use of two-error regression treatments as applied to rubidium-strontium data. *Rev. Geophys. Space Phys.* **10**, 5510577.