

Assessing the error on equivalent dose estimates derived from single aliquot regenerative dose measurements

G.A.T. Duller

Institute of Geography and Earth Sciences, University of Wales Aberystwyth, Ceredigion SY23 3DB, United Kingdom

(Received 3 May 2007; in final form 6 June 2007)

Introduction

Measurement of the equivalent dose (D_e) is central to luminescence dating. Single aliquot methods have been developed in the last 10-15 years, first with the description of methods suitable for feldspars (Duller 1995) and subsequently those applicable to quartz (Murray and Wintle 2000, 2003). Such methods have the advantage that they are comparatively rapid, making it feasible to generate replicate determinations of the D_e , and generally yield D_e values of greater precision than multiple aliquot methods (e.g. Hilgers et al. 2001).

The ability to make replicate measurements of the D_e is one of the most significant advantages of single aliquot methods since this makes it possible to explicitly assess the distribution of apparent dose. This may be critical to demonstrate whether a sample was well bleached at deposition, and whether it has suffered from post-depositional mixing (e.g. Roberts et al. 1998; Jacobs et al. 2003). A number of approaches have been suggested for both displaying and analyzing dose distributions (Galbraith et al. 1999; Thomsen et al. 2003; Spencer et al. 2003; Galbraith 2003), but implicit to all these approaches is the assumption that the uncertainty in the individual D_e values is known.

Variations in D_e between different grains can be masked if many grains are measured simultaneously in a single aliquot (Wallinga 2002), and thus most analyses designed to study the dose distribution are undertaken on aliquots containing few grains (typically 20-50) or single grains. At this scale of analysis, not only do any variations in D_e become apparent, but so too do variations in the brightness of individual grains (McFee and Tite 1998; Duller et al. 2000; McCoy et al. 2000). One effect of such variations in brightness is that the precision with which D_e can be calculated varies from one aliquot to another. As shown by Bailey and Arnold (2006), accurately assessing the error on the D_e is vital in these situations if any method is used to combine these results that relies upon weighting the results

depending upon the accuracy of the individual results (e.g. the Central Age model or Minimum Age model, Galbraith et al. 1999).

Sources of uncertainty in the D_e can be subdivided into random and systematic sources. This paper only deals with random errors associated with the luminescence measurements and then their combination to determine D_e . Systematic sources of uncertainty, such as errors in the calibration of the beta or gamma source used to irradiate the sample in the laboratory need to be considered after the combination of individual D_e values. Similarly, there is an additional source of uncertainty in the suitability of the material for use with the SAR procedure; such uncertainty will be material dependent and has been the subject of much discussion by many authors (e.g. Bailey 2000; Murray et al. 2002; Jacobs et al. 2006a). Such issues are likely to become more significant as the luminescence signal gets close to saturation. The aim of this paper is to compare various approaches to estimating the error on individual D_e values, and provide some data sets that other workers may wish to analyse using their own methods. The paper will focus on examples where the growth is linear, or approximately linear.

Methods of D_e determination

The process of calculating D_e using the SAR procedure involves measurement of the natural luminescence signal (L_N) arising from irradiation in nature, assessing the sensitivity of the aliquot by measuring the luminescence signal (T_N) generated by a test dose (D_T), and then undertaking a number of cycles each of which involves irradiation (D_1, D_2, D_3 etc) to regenerate the luminescence signal (L_1, L_2, L_3 etc), followed by a test of the sensitivity (T_1, T_2, T_3 etc) using the test dose. The value of D_e is then found by comparing the ratio $R_N (= L_N/T_N)$ with the ratios R_1, R_2, R_3 etc (obtained from $L_1/T_1, L_2/T_2, L_3/T_3$ etc) to determine the laboratory dose that generates a signal equivalent to that obtained from the natural.

While all methods for determining D_e are very similar, for the purpose of this paper they can be grouped into three main categories. The simplest method is to compare the normalised signal from the natural ($R_N = L_N/T_N$) with that from the regeneration measurement which gives the closest ratio (e.g. $R_1 = L_1/T_1$) (Fig. 1a). The D_e is then simply given by the ratios of the signals and the dose (D_1) given in the laboratory to generate R_1 :

$$D_e = \frac{R_N}{R_1} D_1 \tag{Eqn. 1}$$

The second method is to interpolate between two regeneration points (R_1 and R_2), one of which is larger than R_N , and one of which is smaller (Fig. 1b). As with the first method, the mathematical calculation of the D_e is straightforward, and relies upon the three ratios of the luminescence signals, and the two known laboratory doses D_1 and D_2 given to the aliquot to generate R_1 and R_2 :

$$D_e = \frac{(R_N - R_1)(D_2 - D_1) + D_1}{(R_2 - R_1)} \tag{Eqn. 2}$$

The third method is to measure the response of the aliquot to a number of different regeneration doses (D_1, D_2, D_3 etc), and to fit an appropriate mathematical equation to the resulting data set R_1, R_2, R_3 etc (Fig. 1c). The equation fitted to this growth curve may be a straight line, or more commonly something involving a saturating exponential (e.g. Eqn. 3), reflecting the commonly accepted view that the luminescence signal saturates at high doses as the defect sites within the aliquot being measured become full.

$$R(D) = I_{Max} (1 - e^{-\frac{D}{D_0}}) + c \tag{Eqn. 3}$$

The ratio $R(D)$ (e.g. R_1, R_2, R_3 etc) measured following a laboratory dose, D , is dependent on the characteristic dose D_0 , which characterises the rate at which the defects in the aliquot become full, the maximum value obtainable, I_{Max} , and an offset, c . For the results shown in this paper the Levenberg-Marquardt algorithm has been used to fit both linear and saturating exponential functions. Hayes et al. (1998) have previously shown that such an algorithm can be used successfully for luminescence data, and the numerical routine from Press et al. (1986) is a convenient source of code.

Each of these methods have implicit assumptions regarding the form of the dose response. For the first

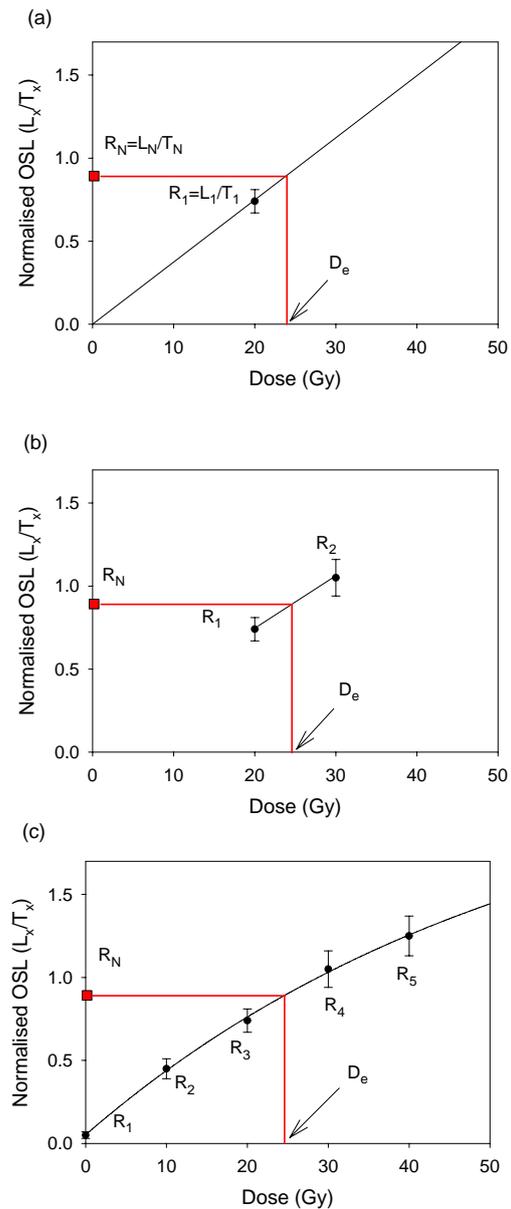


Figure 1: Diagram illustrating the three means of using SAR data to obtain a value of D_e . (a) By taking the ratio of R_N and a single value of R_X , (b) by interpolating between two values of R_X that straddle R_N , and (c) fitting a number of values of R_X measured at different regeneration doses to an equation (e.g. saturating exponential), and then interpolating the value of R_N onto that curve to determine D_e .

method it is assumed that the dose response (R_i) is proportional to dose (D_i) over the interval being analysed; for the second method that the dose response is linear between R_1 and R_2 ; and for the third method, that the chosen function adequately describes the dose response form. If these

Instrumental Error (%)	L _{Signal}	L _{BG}	T _{Signal}	T _{BG}	R _x	S _{Rx}	$\frac{S_{Rx} \times 100}{R_x}$
0	1,050	50	550	50	2.000	0.118	5.9%
1	1,050	50	550	50	2.000	0.122	6.1%
0	100,050	50	50,050	50	2.000	0.011	0.6%
1	100,050	50	50,050	50	2.000	0.030	1.5%

Table 1: Examples of the uncertainty (S_{Rx}) in the ratio of $L_x/T_x (=R_x)$ from counting statistics and from instrumental error.

assumptions are not met then this will introduce error into the value of D_e obtained, but such errors are unlikely to be estimated correctly by the methods described in this paper.

Sources of uncertainty in D_e

Assessing the error in the final value of D_e determined using these methods can be divided into two stages. The first involves calculating the uncertainty in each luminescence measurement and hence the ratios L_x/T_x , whether that is the ratio for the natural or a regeneration dose. The second stage is transforming the errors in those ratios, into an estimate of the uncertainty in the dose, D_e .

Errors in individual L/T ratios

The first constraint on the ability to measure a luminescence signal is its intensity. The uncertainty due to the counting statistics can be calculated using the method described in Galbraith (2002) based on a combination of both the number of counts in the signal and the magnitude of the background signal (L_{BG}) that has been subtracted. Li (2007) has shown that the situation becomes more complex at very low signal levels, but for this paper the approach of Galbraith (2002) has been used. The errors on the values of L_x and T_x can then be propagated in quadrature through Eqn. 4 to give the standard deviation S_{Rx} .

$$S_{R_x} = S_{\frac{L_x}{T_x}} = \frac{L_x}{T_x} \sqrt{\left[\frac{L_{Signal} + L_{BG}}{(L_{Signal} - L_{BG})^2} \right]^2 + \left[\frac{T_{Signal} + L_{BG}}{(T_{Signal} - L_{BG})^2} \right]^2}$$

Eqn. 4

A second source of uncertainty arises from the equipment used to undertake the irradiation, heating and optical stimulation of the aliquot. This uncertainty may be due to small variations in the intensity of the optical stimulation source, small variations in aliquot positioning under the radiation source or under the optical stimulation source, and small variations in the temperature of the aliquot both during preheating and during measurement. The magnitude of these effects is difficult to quantify

individually, but collectively they may be termed 'instrumental error'. These effects are likely to be small, but for bright samples where the uncertainty due to counting statistics is small, they may make a significant contribution to the total uncertainty. The size of this instrumental error can be assessed by making repeated measurements of the luminescence signal from a given irradiation. Such measurements have been undertaken for a standard Risø TL/OSL system by Armitage et al. (2000) giving an instrumental error of 1% on each measurement of L and T . A similar measurement by Rodnight (2006, p.167) yielded a larger value of 2.5%. Thomsen et al. (2005) and Jacobs et al. (2006b) made similar measurements for a single grain system, giving instrumental errors of ~1.5%. It is likely that this value may vary from one instrument to another, and possibly may change over long periods of time as instruments alter in their operation. For this paper a value of 1% has been used, and this contribution needs to be combined in quadrature with that from counting statistics for each L_x/T_x ratio.

A feeling for the magnitude of these effects, and their relative importance, can be gained from taking a number of examples. Table 1 shows data for a single aliquot measurement which gives a signal (L_{Signal}) of 1050 counts, a background (L_{BG}) of 50 counts, a test dose response (T_{Signal}) of 550 counts and a background (T_{BG}) of 50 counts, giving a ratio of L_x/T_x of 2.000. Based solely on the counting statistics, and using Eqn. 4, the standard deviation in that ratio is 0.118 (5.9%). Including the value of 1% for the instrumental error only marginally increases this to 6.1%, and the dominant source of uncertainty is the low count rate. In the second example in Table 1, the aliquot is approximately one hundred times brighter, and so the uncertainty due to counting statistics is very low (0.6%). In this case the instrumental error more than doubles this (1.5%).

Transforming the error in L/T ratios to an error in D_e

The quantity that we are ultimately interested in is the D_e and the standard deviation of this value (S_{De}). The approach to estimating S_{De} varies depending upon the method used to determine D_e . Using Eqn. 1, the

uncertainty can be simply obtained by combining the errors in the two ratios R_N and R_1 in quadrature, giving the following equation:

$$S_{D_e} = D_e \sqrt{\left(\frac{S_{R_N}}{R_N}\right)^2 + \left(\frac{S_{R_1}}{R_1}\right)^2} \quad \text{Eqn. 5}$$

For the situation where one is interpolating between two data points, as expressed in Eqn. 2, Thomsen et al. (2005, 2007) have shown that the standard deviation in the value of D_e is given by the expression in Eqn. 6.

$$S_{D_e} = \sqrt{\left(\frac{D_2 - D_1}{R_2 - R_1}\right)^2 \left\{ S_{R_N}^2 + \left(\frac{1}{R_2 - R_1}\right)^2 [(R_N - R_2)^2 S_{R_1}^2 + (R_N - R_1)^2 S_{R_2}^2] \right\}}$$

Eqn. 6

In the more complex situation where a large number of values of R_x are used to fit a mathematical equation to define the growth of the luminescence signal, then defining an analytical solution for the standard deviation in the D_e becomes more complex. For simple functions (e.g. linear) then it may be possible to do this, but in such methods it is normally necessary that the errors are relatively small. This is not always the case in luminescence data. The question then arises of how one can assess the value of S_{D_e} ?

Simple transformation of the S_{RN} to S_{De}

Frequently for luminescence data obtained using the SAR procedure, one of the dominant sources of uncertainty arises from measurement of the natural (S_{RN}) due to counting statistics and instrumental error. A straightforward means to estimate the uncertainty in the D_e that results from this is to interpolate the values $R_N + S_{RN}$ and $R_N - S_{RN}$ onto the growth curve and then see the variation in D_e that results. This approach makes it possible to deal with any form of equation that is fitted to the growth curve, and accommodates changes in curvature of the growth curve. However, it does not take into account the degree of certainty with which the growth curve is known, based upon the fit to the data R_1, R_2, R_3 etc. A first order approximation to incorporate the uncertainty in the growth curve that has been fitted can be obtained by calculating the deviation of the fitted growth curve from the n data points $R_{1..n}$ using Eqn. 7.

$$\text{Average Deviation} = \sqrt{\frac{\sum_{i=1}^n (\text{Predicted } R_i - \text{Actual } R_i)^2}{n}} \quad \text{Eqn. 7}$$

This figure for the typical deviation of the fitted growth curve from the measured data points is then combined in quadrature with the uncertainty in the ratio of R_N . This combined error is then transformed through interpolation of the upper and lower limits of R_N on to the growth curve, to calculate the limits on the estimate of D_e . Such an approach has the advantage of speed, but how accurate is such an approximation, and is a better approach available?

The Monte Carlo method

A more robust approach to assessing the error on the value of D_e is to use a Monte Carlo method (Press et al. 1986). This approach has been used by some researchers for a number of years (Bailey Pers. Comm., e.g. Grine et al. 2007). Each value of R_x and its standard deviation S_{RX} , based on counting statistics and instrumental error, is represented by a Gaussian distribution of possible values. Repeated curve fitting and calculation of D_e are undertaken where the values of R_x that are used both for R_N , and for R_1, R_2, R_3 etc are drawn from Gaussian distributions whose widths are set by the calculated standard deviations. This approach explicitly assesses the nature of the distribution in the value of D_e , including its width and whether it is symmetric. An estimate of the standard deviation of D_e can then be explicitly calculated by analysis of the resulting distribution of D_e values. The central value of D_e is still obtained using the best fit through the original data.

Example data sets

To provide a comparison of these different approaches, a number of example data sets are shown (Fig. 2), and the results of D_e determination using the different methods and appropriate error calculations are shown in Table 2. The luminescence data used to generate the graphs in Fig. 2 are given in the Appendix. Although this is a limited data set with which to compare the methods, a number of points can be made.

Firstly, given the data sets used here, the results are consistent between the different methods. Given the variety of approaches both to determining the D_e and its uncertainty, this is reassuring. Moreover, the approximation described in the section headed “*Simple transformation of the S_{RN} to S_{De}* ” (shown under the heading “Curve Fitting” in Table 2) gives uncertainties that are very close to those derived using the more complex Monte Carlo approach. The Monte Carlo method is more time consuming than the approximation. The uncertainties shown in the final column of Table 2 are based on 1000 estimates of D_e using the Monte Carlo method (the distribution of D_e values are shown as histograms below the

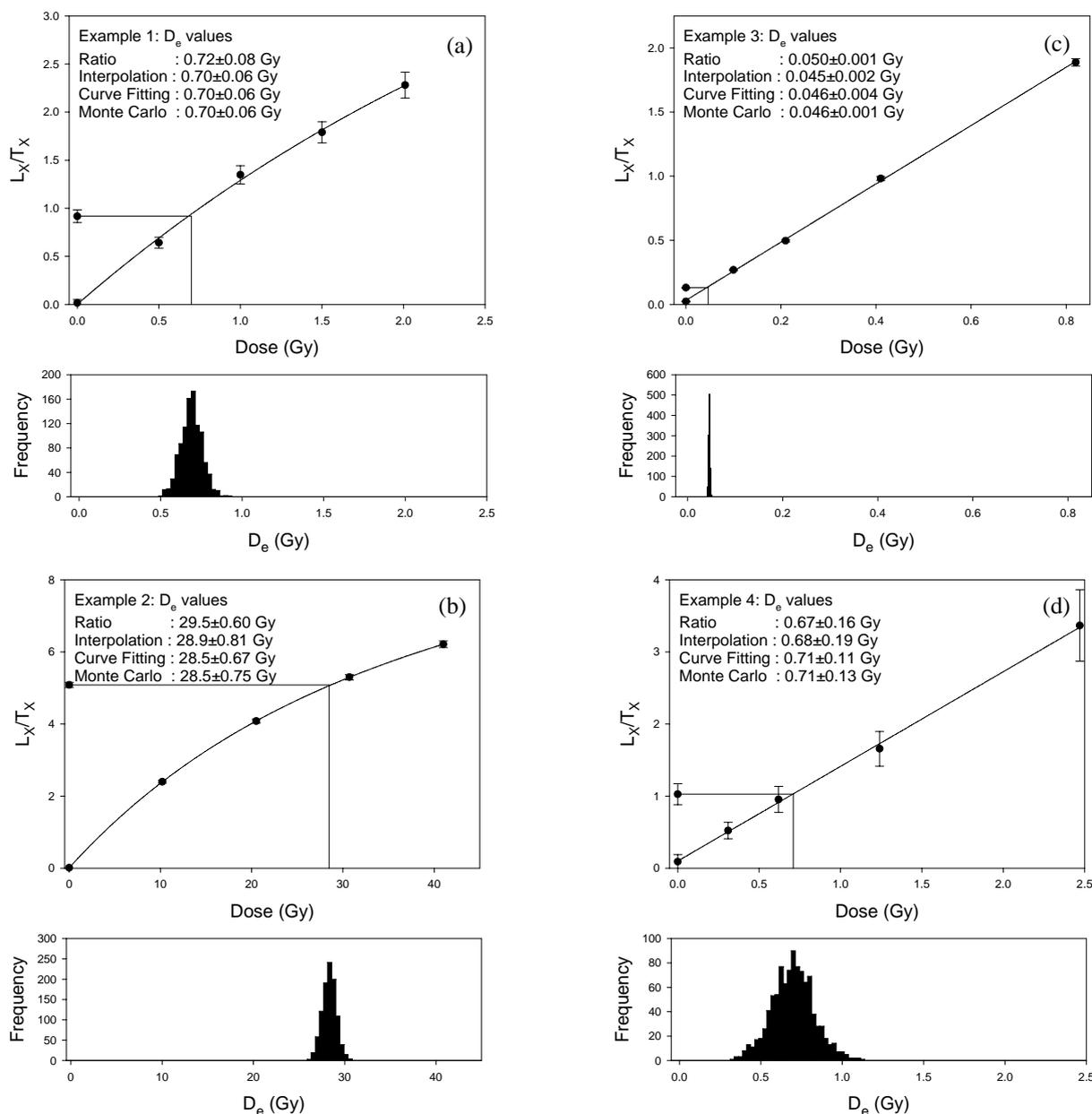


Figure 2: Growth curves for four example SAR data sets. In each case the values of L_x/T_x have been calculated based on the counting statistics and incorporating a 1% instrumental error. The D_e value indicated on each diagram is that based on curve fitting. The distribution of D_e values obtained using 1000 iterations of the Monte Carlo method are shown in the figures below each growth curve.

Example Number	Equivalent Dose (Gy)				
	Ratio	Interpolate	Equation fitted	Curve Fitting	Monte Carlo
1	0.72±0.08	0.70±0.06	Exponential	0.70±0.06	0.70±0.06
2	29.5±0.60	28.9±0.81	Exponential	28.5±0.67	28.5±0.75
3	0.050±0.001	0.045±0.002	Linear	0.046±0.004	0.046±0.001
4	0.67±0.16	0.68±0.19	Linear	0.71±0.11	0.71±0.13

Table 2: Comparison of equivalent dose calculated using the different analytical procedures described in the text.

growth curves in Fig. 2). However, the greater statistical robustness of the Monte Carlo method for determining the errors makes it preferable.

In general, measuring R_X for a range of regeneration doses provides information about the form of the growth of the luminescence signal. This is particularly important where the natural luminescence signal R_N may be close to saturation. Using a single data point and calculating the D_e by the ratio of R_N to R_X should provide an accurate value, especially where the D_e is low. However, the D_e may be inaccurate if the value of R_X does not closely match R_N . In the data sets shown in Fig. 2, this is not a major problem since R_X values at a range of doses have been measured, and the value of D_e shown in Table 2 is calculated by taking the ratio of R_N to R_X for that value of R_X that is closest to R_N . In spite of this, it can be observed that the D_e for Example 2 determined by this method (29.5 ± 0.60 Gy) is higher than that calculated using the other methods, because the value of R_X used (that relating to a regeneration dose of 30.7 Gy) is higher than R_N , and the method implicitly assumes that the growth of R_X is linear with dose. Using the value of R_X from the 41.0 Gy regeneration point gives an even higher D_e of 33.5 ± 0.68 Gy. Conversely, using values of R_X below R_N gives lower D_e values. Had the regeneration point at 20.5 Gy been used then the D_e would be 25.5 ± 0.52 Gy, and using the 10.2 Gy regeneration data would yield a D_e of 21.7 ± 0.44 Gy.

Interpolating between the two values of R_X that straddle R_N overcomes this fundamental problem, and approximates the slope of the growth curve at that dose. However, once again, the closer the two values of R_X are to R_N , the better the estimate of D_e . Precisely matching R_X and R_N is often difficult because of variability in the D_e from one aliquot to another. The best estimate of the form of the growth curve near R_N is achieved by fitting an equation to the entire data set.

Dose recovery experiment

One means of assessing whether the estimation of the errors on D_e values is appropriate is to undertake a dose recovery experiment on a sample that is thought to be well suited for the SAR procedure. Quartz extracted from a linear dune in north-eastern Tasmania (TNE9517, Duller and Augustinus 2006) was used for this experiment. Quartz grains were 180–211 μm in diameter, and these were mounted on aluminium discs using silicone oil. In order to create data with different OSL signal intensities, and thus with different uncertainties due to counting statistics, 22 aliquots were prepared so that the grains covered an area of ~ 5 mm diameter, while another 16 aliquots

were prepared where the grains covered an area of 2 mm diameter.

The OSL properties of quartz from this part of Tasmania have been described briefly in Duller and Augustinus (2006). The quartz has an intense OSL signal, and yields reproducible values of D_e using the SAR procedure. To undertake the dose recovery experiment, the 38 aliquots were bleached using the blue diodes in a Risø TL/DA-15 TL/OSL reader, given a dose of ~ 5 Gy using the $^{90}\text{Sr}/^{90}\text{Y}$ beta source mounted on the reader, preheated at 220°C for 10 seconds, and then had their OSL signal measured while holding them at 125°C . The aim of this test was not to assess the extent to which the sample is suitable for the SAR procedure, but to assess whether the calculation of the error in the calculation of D_e is appropriate. Thus, the bleaching and irradiation procedure was repeated at least 4 times in an attempt to 'condition' the aliquots and make them as reproducible as possible.

For the experiment itself, the 38 aliquots were exposed to the beta source for 15 seconds (~ 0.6 Gy). The magnitude of this dose was then determined using a SAR procedure, with 6 regeneration doses (0, 10, 20, 30, 40, 10 seconds beta dose), using a preheat of 220°C for 10 seconds, and a cutheat after the test dose (6 seconds beta dose) of 200°C . After measurement of the response to the test dose, the aliquots were exposed to the blue diodes for 40 s whilst holding them at 280°C in order to reduce any build up of slow components in the OSL signal (Murray and Wintle 2003).

The equivalent dose values and associated errors were calculated using the Curve Fitting method and the Monte Carlo method. Both sets of analysis were undertaken assuming an instrumental uncertainty of 1.0%, and the results are shown as radial plots in Fig. 3. There is a broad spread in the precision with which the values are known, as was hoped for by the use of two different aliquot sizes. In both analyses, the results are consistent with the given dose of 15 s (weighted mean is 14.6 ± 0.7 and 14.7 ± 0.7 s respectively for the two analyses). In detail the errors on the D_e values are subtly different. For the Curve Fitting method, 35 out of 38 of the aliquots (92%) are consistent with 15 s within two standard deviations, while for the Monte Carlo method 33 out of 38 (87%) are consistent. Both percentages are only slightly lower than would be expected from statistics (for a normal distribution, 95% of the data are within two standard deviations).

It is interesting to note that while the two radial plots are similar, there are differences. For instance the

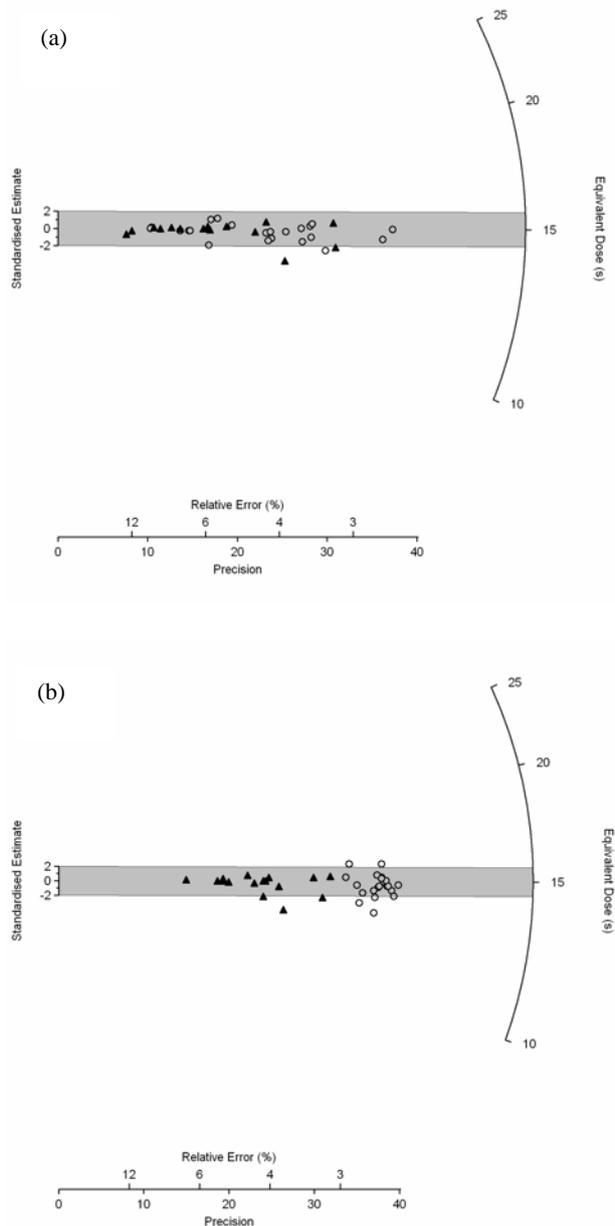


Figure 3: Radial plot of 38 D_e values obtained in a dose recovery experiment. The aliquots were irradiated for 15 s (~ 0.6 Gy), and then an SAR growth curve was constructed to measure this dose. The data were analysed (a) using the Curve Fitting procedure and (b) using the Monte Carlo method. Open points are those obtained using 5 mm diameter aliquots, filled points are those for 2 mm aliquots.

range in Relative Error observed is 2.7-13.1% for the Curve Fitting method, but rather narrower for the Monte Carlo method (2.5-6.7%). For a single population such as that shown in Fig. 3 this

difference is unimportant, but it may have a more serious impact if a data set containing a wide distribution of D_e values were to be analysed using some form of statistical model such as the Minimum Age model (Galbraith et al. 1999). Assessing the sensitivity of such statistical procedures to changes in the calculated uncertainty in D_e has been assessed by Bailey and Arnold (2006), who shows the importance of reliably estimating it.

Conclusions

While there are differences between the D_e values obtained using the three methods of D_e determination illustrated in Fig. 1, these differences are small (Table 2). For these data sets, R_X has been measured for a range of different regeneration doses to ensure that the response of the luminescence signal in the dose range of interest is known. Using the ratio to a single data point may lead to larger errors than those seen here if the value of R_X is not close to R_N , or if the aliquot is close to saturation. Interpolating between two values of R_X decreases the magnitude of any such error, but the most accurate method will be to fit the entire data set, particularly if there is discernable deviation from linear growth. Two methods of estimating the standard deviation in the estimate of D_e have been described. Of the two, the Monte Carlo approach is more statistically robust, but both yield similar results, both for the detailed examples shown in Fig. 2, and the dose recovery data in Fig. 3, presumably because the dominant source of uncertainty is that which arises from the measurement of R_N and the instrumental error. One advantage of the Monte Carlo approach is that for samples approaching saturation it will correctly identify that the errors in the estimate of D_e are asymmetric. However, two issues arise in such situations. The first is that standard statistical methods for combining different D_e estimates which rely upon weighting of individual data points cannot easily be applied to data with asymmetric errors. The second is that in situations where the luminescence growth curve is sufficiently close to saturation for asymmetry in errors to become important, the reliability of the SAR method is unclear. Wintle and Murray (2006) recommend that the method only be used when R_N is less than 85% of I_{Max} (Eqn. 3).

While the procedures described here attempt to assess the random uncertainty in the estimates of D_e obtained using the SAR procedure, the error due to systemic failures of the SAR procedure, or inappropriate response of the dosimeter will not be captured. These could potentially be very large, and are likely to become more severe as the response of the aliquot to radiation decreases as it approaches saturation. Such effects are more difficult to assess,

and tests such as dose recovery experiments may be one of the few ways that it is possible to get some impression of their impact.

Acknowledgements

All analyses shown here, including the Monte Carlo calculations, have been performed using the software package Analyst (Version 3.24) which is now available. The author is very grateful to a large number of people who have provided encouragement and help in the development of this software over many years. Kristina Thomsen very kindly provided the opportunity to test a number of these routines and gave assistance in checking some of the calculations. Richard Bailey discussed Monte Carlo methods and provided extremely helpful suggestions that improved this paper. Ann Wintle and Helen Roberts also provided valuable comments on earlier versions of the paper.

References

- Armitage, S. J., Duller, G. A. T., Wintle, A. G. (2000). Quartz from southern Africa: sensitivity changes as a result of thermal pretreatment. *Radiation Measurements* **32**, 571-577.
- Bailey, R. M. (2000). Circumventing possible inaccuracies of the single aliquot regeneration method for the optical dating of quartz. *Radiation Measurements* **32**, 833-840.
- Bailey, R. M., Arnold, L. J. (2006). Statistical modelling of single grain quartz D_e distributions and an assessment of procedures for estimating burial dose. *Quaternary Science Reviews* **25**, 2475-2502.
- Duller, G. A. T. (1995). Luminescence dating using single aliquots: methods and applications. *Radiation Measurements* **24**, 217-226.
- Duller, G. A. T., Bøtter-Jensen, L., Murray, A. S. (2000). Optical dating of single sand-sized grains of quartz: sources of variability. *Radiation Measurements* **32**, 453-457.
- Duller, G. A. T., Augustinus, P. C. (2006). Reassessment of the record of linear dune activity in Tasmania using optical dating. *Quaternary Science Reviews* **25**, 2608-2618.
- Galbraith, R. F. (2002) A note on the variance of a background-corrected OSL count. *Ancient TL* **20**, 49-51.
- Galbraith, R. F. (2003). A simple homogeneity test for estimates of dose obtained using OSL. *Ancient TL* **21**, 75-77.
- Galbraith, R. F., Roberts, R. G., Laslett, G. M., Yoshida, H., Olley, J. M. (1999). Optical dating of single and multiple grains of quartz from Jinnium rock shelter, northern Australia: Part I, Experimental design and statistical models. *Archaeometry* **41**, 339-364.
- Grine, F. E., Bailey, R. M., Harvati, K., Nathan, R. P., Morris, A. G., Henderson, G. M., Ribot, I., Pike, A. W. G. (2007). Late Pleistocene human skull from Hofmeyr, South Africa, and modern human origins. *Science* **315**, 226-229.
- Hayes, R. B., Haskell, E. H., Kenner, G. H. (1998) An assessment of the Levenberg-Marquardt fitting algorithm on saturating exponential data sets. *Ancient TL* **16**, 57-62.
- Hilgers, A., Murray, A. S., Schlaak, N., Radtke, U. (2001). Comparison of quartz OSL protocols using lateglacial and Holocene dune sands from Brandenburg, Germany. *Quaternary Science Reviews* **20**, 731-736.
- Jacobs, Z., Duller, G. A. T., Wintle, A. G. (2003). Optical dating of dune sand from Blombos Cave, South Africa: II - single grain data. *Journal of Human Evolution* **44**, 613-625.
- Jacobs, Z., Duller, G. A. T., Wintle, A. G. (2006b). Interpretation of single grain D_e distributions and calculation of D_e . *Radiation Measurements* **41**, 264-277.
- Jacobs, Z., Wintle, A. G., Duller, G. A. T. (2006a). Evaluation of SAR procedures for D_e determination using single aliquots of quartz from two archaeological sites in South Africa. *Radiation Measurements* **41**, 520-533.
- Li, B. (2007). A note on estimating the error when subtracting background counts from weak OSL signals. *Ancient TL* **25**, 9-14.
- McCoy, D. G., Prescott, J. R., Nation, R. J. (2000). Some aspects of single-grain luminescence dating. *Radiation Measurements* **32**, 859-864.
- McFee, C. J., Tite, M. S. (1998). Luminescence dating of sediments - the detection of high equivalent dose grains using an imaging photon detector. *Archaeometry* **40**, 153-168.
- Murray, A. S., Wintle, A. G. (2000). Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. *Radiation Measurements* **32**, 57-73.
- Murray, A. S., Wintle, A. G. (2003). The single aliquot regenerative dose protocol: potential for improvements in reliability. *Radiation Measurements* **37**, 377-381.
- Murray, A. S., Wintle, A. G., Wallinga, J. (2002). Dose estimation using quartz OSL in the non-linear region of the growth curve. *Radiation Protection Dosimetry* **101**, 371-374.
- Press, W. H., Flannery, B. P., Teukolsky, S. A., Vetterling, W. T. (1986). *Numerical Recipes: The art of scientific computing*. Cambridge University Press.

- Roberts, R. G., Bird, M., Olley, J., Galbraith, R., Lawson, E., Laslett, G., Yoshida, H., Jones, R., Fullagar, R., Jacobsen, G., Hua, Q. (1998). Optical and radiocarbon dating at Jinmium rock shelter in northern Australia. *Nature* **393**, 358-362.
- Rodnight H. (2006). *Developing a luminescence chronology for Late Quaternary fluvial change in South African floodplain wetlands*. Unpublished PhD thesis, University of Wales Aberystwyth, 304pp.
- Spencer, J. Q., Sanderson, D. C. W., Deckers, K., Sommerville, A. A. (2003). Assessing mixed dose distributions in young sediments identified using small aliquots and a simple two-step SAR procedure: the F-statistic as a diagnostic tool. *Radiation Measurements* **37**, 425-431.
- Thomsen, K. J., Jain, M., Bøtter-Jensen, L., Murray, A. S., Jungner, H. (2003). Variation with depth of dose distributions in single grains of quartz extracted from an irradiated concrete block. *Radiation Measurements* **37**, 315-321.
- Thomsen, K. J., Murray, A. S., Bøtter-Jensen, L. (2005). Sources of variability in OSL dose measurements using single grains of quartz. *Radiation Measurements* **39**, 47-61.
- Thomsen, K.J., Murray A.S., Bøtter-Jensen L., Kinahan J. (2007) Determination of burial dose in incompletely bleached fluvial samples using single grains of quartz. *Radiation Measurements* **42**, 370-379
- Wallinga, J. (2002). On the detection of OSL age overestimation using single-aliquot techniques. *Geochronometria* **21**, 17-26.
- Wintle, A. G., Murray, A. S. (2006). A review of quartz optically stimulated luminescence characteristics and their relevance in single-aliquot regeneration dating protocols. *Radiation Measurements* **41**, 369-391.

Appendix:

The luminescence data used to construct the growth curves in Fig. 2 are shown in the tables overleaf. In each case an instrumental error of 1% was used in the calculations. The luminescence signals were integrated from channels 1-10, and the background from channels 231-250. In the tables below, the value of the background has been adjusted to allow for the difference in the number of channels used for integration.

Reviewer

R.M. Bailey

Example 1

Dose (Gy)	L_x		T_x		L_x/T_x	$S(L_x/T_x)$
	Signal	BG	Signal	BG		
Natural	967	270	1064	304	0.917	0.066
2.01	2157	316	1219	412	2.281	0.134
1.50	1748	361	1139	364	1.790	0.110
1.00	1338	367	1122	401	1.347	0.095
0.50	870	364	1163	375	0.642	0.056
0.00	372	360	1159	383	0.015	0.035

Example 2

Dose (Gy)	L_x		T_x		L_x/T_x	$S(L_x/T_x)$
	Signal	BG	Signal	BG		
Natural	1961354	8978	398551	14323	5.081	0.072
0.0	13317	9690	353432	14664	0.011	0.000
10.2	680491	21238	294425	19112	2.395	0.034
20.5	983391	28554	256435	22292	4.078	0.059
30.7	1202981	36036	246823	26456	5.295	0.076
41.0	1445211	45549	257434	32002	6.209	0.089

Example 3

Dose (Gy)	L_x		T_x		L_x/T_x	$S(L_x/T_x)$
	Signal	BG	Signal	BG		
Natural	6566	968	44775	1798	0.130	0.003
0.82	83842	2268	46127	2918	1.888	0.029
0.41	44984	2683	46302	3234	0.982	0.016
0.21	24094	2682	46589	3390	0.496	0.008
0.10	14088	2788	45560	3486	0.269	0.005
0.00	3626	2676	46666	3458	0.022	0.002

Example 4

Dose (Gy)	L_x		T_x		L_x/T_x	$S(L_x/T_x)$
	Signal	BG	Signal	BG		
Natural	294	93	294	98	1.026	0.145
0.00	120	106	281	126	0.090	0.098
0.31	221	122	318	128	0.521	0.114
0.62	313	151	342	172	0.953	0.180
1.24	536	200	402	199	1.655	0.242
2.47	934	268	466	268	3.364	0.495