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Interpretation of single grain D_e distributions and calculation of D_e

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Abstract

Recent development of an instrument for measuring the optically stimulated luminescence signal from individual mineral grains has made it practicable to measure the equivalent dose (D_e) from many hundreds or thousands of single mineral grains from a sample. Such measurements can potentially be used to address issues such as sample integrity, and to make it possible to obtain ages from samples that consist of mixtures of grains, enlarging the range of materials to which luminescence dating can be applied. However, for reliable ages to be obtained, the characteristics of the equipment and the sample being analysed need to be understood.

Using sensitised sedimentary quartz grains, the instrumental uncertainty in repeated optically stimulated luminescence measurements made using a single grain laser luminescence unit attached to a conventional luminescence reader was evaluated; a value of 1.2% was obtained. Grains from this sample were then used to investigate the uncertainty in a measured dose distribution obtained using the single aliquot measurement protocol on each grain that had previously received a known laboratory dose; after systematic rejection of grains that did not pass defined acceptance criteria, overdispersion of 7% was found.

Additional spread in data was found when uniform aeolian sands were examined, resulting in overdispersion of $\sim 12\%$; this was attributed to a combination of factors relating to differences in field and laboratory conditions. A similar value was found for an archaeological horizon below this sand. For another sample from the same section, a significantly larger value was found, $\sim 29\%$; on this basis the finite mixture model was applied to obtain the likely dose components. The paper demonstrates the importance of correct assessment of error terms when analysing single grain D_e distributions and a number of rejection criteria that are vital to avoid the inclusion of data that could lead to misinterpretation of the degree of scatter present.

1. Introduction

Dating by means of the optically stimulated luminescence (OSL) signal from quartz has been developed over the last 20 years, since Huntley et al. (1985) presented measurements on a number of sedimentary samples. Recent technological advances have made it possible to measure OSL signals from individual sand-sized grains of quartz using a laser with its beam focussed down to 20 μm diameter at the surface of the grain (Duller et al., 1999; Bøtter-Jensen et al., 2000; Duller, 2004). This advance has occurred alongside the development of a measurement procedure that takes account of the sensitivity changes reported for individual grains (e.g. Adamiec, 2000). This measurement procedure, the single aliquot, regenerative-dose (SAR) protocol was developed by Murray and Wintle (2000) for single aliquots of quartz, each made up of a few thousand grains. Now it is possible to use SAR procedures to obtain the equivalent dose (D_e) for each of many thousands of grains.

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The large number of D_e values obtained for each sample need to be displayed, interpreted and combined in some way to obtain a single value of D_e that can be used in the final age calculation to obtain a true depositional age for the sediment. Grains within a single sample may have different apparent ages if some of them were not exposed to daylight for a sufficient period of time at deposition to reset their luminescence signal or if some younger or older grains intruded as a result of mixing between layers. Thus, a major advantage of dating single grains of quartz should be the ability to identify the presence of incompletely bleached grains or intrusive grains in a bulk sample.

However, before interpretations of D_e values can be made, it is essential to establish how much of the spread is caused by instrumental factors, counting statistics (e.g. Thomsen et al., 2005) and curve-fitting errors and how much is the result of using a SAR measurement procedure that is appropriate for the vast majority of grains, but not for all (Jacobs et al., Submitted). In theory, the instrumental factors and counting statistics can be improved through instrumental design. However, natural grain-to-grain variations will guarantee that SAR will be more suitable for some grains than others.

Once the instrumental uncertainty has been established and errors in calculating individual values of D_e quantified, the applicability of the SAR measurement procedure can be assessed on a grain-by-grain basis. Single grain analysis permits the rejection of data from individual grains that can adversely influence the dose distributions of a sample and the calculation of D_e used in the age equation. This is best demonstrated in a dose recovery experiment using laboratory-irradiated grains, where depositional and post-depositional processes cannot confuse the interpretation. In addition, study of a well-bleached sediment, (such as a dune sand), can be used to determine the percentage uncertainty caused by non-uniform irradiation in the natural environment. Only once all these factors have been quantified, is it possible to identify the presence of different populations of grains.

In this paper, several data sets for single grains are systematically evaluated for a number of sedimentary samples from two archaeological sites in South Africa, Blombos Cave (ZB4, ZB8, ZB13) and Sibudu Cave (SIB2). Although these sites contain an important archaeological record, the approaches that we have developed in order to provide reliable chronologies have a much wider applicability, i.e. to all sedimentary deposits.

2. Single grain D_e measurements

The first measurements of single grains were made by placing individual quartz grains on the type of discs used for measurement of single aliquots of quartz (e.g. Murray and Roberts, 1997; Roberts et al., 1997, 1998, 1999; Olley et al., 1999; Spooner et al., 2001); each grain was stimulated using either a tungsten halogen lamp giving $\sim 16\,\mathrm{mW/cm^2}$ of green plus blue (420–550 nm) light or by blue (470 ± 30 nm) light emitting diodes giving $\sim 40\,\mathrm{mW/cm^2}$. More recently, with the development of the single grain laser luminescence unit for the Risø TL/OSL reader (Duller et al., 1999), it has been possible to mount 100 grains in a 10×10 grid of holes drilled into the surface of a similar disc (Bøtter-Jensen et al., 2000). The OSL is stimulated from each grain in turn by the green (532 nm) light from a 10 mW $Nd:YVO_4$ diode pumped laser. This gives a power density at the grain of $\sim 50\,\mathrm{W/cm^2}$ which falls over a circular area 20 μm in diameter. This power level results in most of the quartz OSL signal being observed in the first 0.3 s of optical stimulation. Up to 48 such discs can be accommodated in the Risø OSL/TL reader.

Several recent papers have reported D_e values obtained following measurement of several thousand grains using the single grain laser luminescence system; however, only a small fraction of the measured grains resulted in usable D_e values. Jacobs et al. (2003b) reported the measurement of almost 9000 grains from three contemporaneous remnants of an aeolian sand unit that sealed Middle Stone Age deposits at Blombos Cave, South Africa. For these samples, no independent ages were available, but multiple grain SAR OSL dating gave ages of ~ 70 ka (Jacobs et al., 2003a) and the single grain ages were only 4% lower than these (Jacobs et al., 2003b). An age of ~ 70 ka for dune formation is compatible with the expected position of mean sea level at that time. Olley et al. (2004a) reported values for seven fine sand units from a marine core off the west coast of Australia. The single grain OSL ages were consistent with the chronology based on 15 AMS radiocarbon ages for the core, covering the period from ~ 43 to 1 ka. Feathers (2003) measured D_e values for between 1000 and 2000 grains for each of 19 samples from the Southern High Plains, USA. Most of the samples were of aeolian origin and each had associated radiocarbon ages, or archaeological ages based on point typology, over the age range from ~ 13 to 0.5 ka.

Although most single grain OSL ages in these studies were in agreement with the expected ages, there were instances of broader than expected distributions for some samples. For two samples from the marine core, Olley et al. (2004a) found a spread of as much as ~ 100 Gy; they explained this spread in D_e values in terms of the incomplete bleaching of some grains. Feathers (2003) proposed post-depositional mixing to be the cause of the wide distributions that he found in sand dunes. The samples studied by Jacobs et al. (2003b) did not show a spread in D_e and thus were considered to be undisturbed.

In each of these papers, the D_e distributions were presented as radial plots (Galbraith, 1988; Galbraith et al., 1999). These display both the individual D_e values and the related precision on a plot that also enables visual evaluation of whether a sample is dominated by one population of D_e values, or is more complex. Provided there is a statistically meaningful number of D_e values, and that the individual uncertainties have been evaluated correctly, it is possible to see whether 95% of the D_e values fall within $\pm 2\sigma$ of the central value. When more than 5% of D_e values lie outside this range, the dose distribution is termed overdispersed. Overdispersion (OD) in a D_e distribution is a quantitative measure that refers to the relative standard deviation of the distribution of true single grain D_e values from a central D_e value, after having allowed for estimation of the statistical error (Galbraith et al., 1999). Zero OD will suggest that there is a single population; OD that is significantly above zero suggests that a range of true D_e values is present.

If the instrumental uncertainty associated with the single grain measurement system is not included in the overall error estimates for single grain D_e's, it can falsely indicate OD in a data set. Conversely, when the instrumental uncertainty is overestimated, it may mask any spread in the data set. A reliable and quantified estimate of instrumental uncertainty is therefore essential and instrument reproducibility tests should be performed for each individual reader before D_e distributions are interpreted.

Galbraith et al. (1999) presented statistical models for calculation of an appropriate D_e, i.e. the one to be used to calculate the age of deposition of the quartz grains. The choice of model, e.g. common age, central age, minimum age or finite mixture model, is based on the OD value. This choice assumes that the precision assigned to each D_e measurement is correctly calculated. It combines the error related to counting statistics for each L (regenerative dose) and T (test dose) measurement in the SAR procedure with that related to the instrumental precision. Armitage et al. (2000) reported an uncertainty of between 1% and 2% on every L_x and T_x measurement on large single aliquots of bright grains measured using a standard Risø reader equipped with blue LEDs. For the Risø single grain laser system, the value of 3.5% measured by Truscott et al. (2000) has usually been adopted, e.g. by Jacobs et al. (2003b), Olley et al. (2004a) and Feathers (2003). This value has been re-evaluated by Thomsen (2004) and Thomsen et al. (2005) and will be determined, using a different approach, in the next section.

3. Instrument reproducibility

Truscott et al. (2000) obtained a measure of instrumental uncertainty of 3.5% on each OSL measurement for the prototype Risø single grain system. Many improvements have been made to both the software and the equipment itself during the course of the last 3–4 years and a new measurement of the instrumental uncertainty has been made.

Using the basic approach of Truscott et al. (2000), instrument reproducibility was measured by repeatedly irradiating, preheating and stimulating a single grain disc loaded with 50 bright grains of a sedimentary sample from Sibudu (SIB2). These grains were selected because they had more than 1500 counts when 0.04 s of optical stimulation time (two data channels) was used to integrate the OSL signal, thus reducing the contribution from counting statistics to a maximum of 2.5%. The grains had previously been subjected to a full SAR measurement cycle and it was assumed that the grains were, as a result, sensitised to a large degree. Therefore, unlike Thomsen (2004), no sensitivity corrections were applied. The grains were each given 80 Gy in situ using the beta irradiator in the reader, preheated at 260 °C for 10 s and the OSL was measured at 125 °C for 1 s at 90% laser power to obtain a measurement of L_x. A typical OSL decay curve is shown in Fig. 1. The signal integration regions of 0–0.04 s (two data channels) and 0–0.3 s (15 data channels) are indicated. The inset shows the normalised OSL for five repeated measurements on one grain. After each optical stimulation, the grains were again optically stimulated, but at a raised temperature (280 °C as opposed to 125 °C) to ensure that all remaining charge was evicted. This irradiation–preheat–OSL measurement cycle was repeated 10 times. The error resulting from these measurements will therefore be a combined product of counting statistics and instrumental uncertainty.

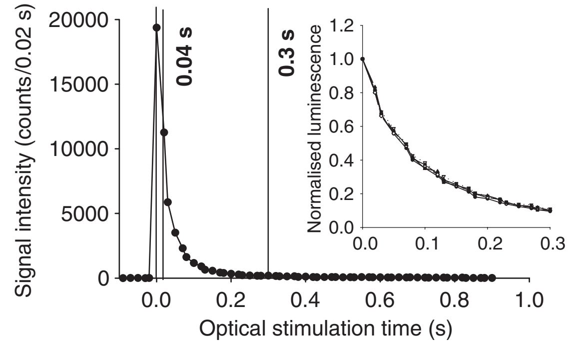

Fig. 1. Laser stimulated OSL decay curve for a grain of SIB2 given an 80 Gy dose. First five channel data points are dark count measurements. Inset shows five repeated OSL decay curves for one grain.

For each grain the 10 repeated L_x measurements $(L_1, \ldots, L_{10})$ were normalised relative to L_1. For some grains, however, sensitivity changes could still be observed as a function of cycle. Not all grains showed a linear trend in L_x with measurement cycle, and for these a second-order polynomial was fitted instead. The relative deviation about the fitted line, expressed as the residual sum of the squares, was taken as being the combined uncertainty arising from both instrumental uncertainty and counting statistics. The relative standard error (rse_{L_x}) of this combined uncertainty estimate was calculated as the square root of the residual mean square. It is therefore the standard deviation of the data about the trend line, rather than about the sample mean, and can be denoted as

$$\mathrm{rse}_{L_x} = 100 \times \sqrt{\frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{n-2}}, \quad (1)$$

where y is the normalised luminescence signal (L_x/L_1) for a specific grain, $\hat{y}$ is the normalised value predicted from the trend line for that grain, $\sum(y-\hat{y})^2$ is the residual sum of the squares and $n-2$ is the corresponding degrees of freedom associated with the sum of the squares.

As noted before, this estimate also includes the error arising from counting statistics; the contribution from counting statistics should therefore be estimated separately. The relative standard error (rse_{cs}) arising from counting statistics was estimated using the equation given in Galbraith (2002). The equation was

$$\text{rse}_{\text{cs}} = 100 \times \frac{\sqrt{\sum_i S_i + \bar{B}_n/k}}{\sum_i S_i - \bar{B}_n}, \quad (2)$$

where S_i is the signal from the ith channel integration and $\bar{B}$ is the average relative background corrected for the number of channels integrated for the OSL signal.

The relative standard error arising from instrumental uncertainty (rse_{iu}) is therefore the error arising from variability around the fitted line estimated from Eq. (1) minus the error that arises from counting statistics estimated from Eq. (2), where the subtraction was done in quadrature as

$$\text{rse}_{\text{iu}} = \sqrt{(\text{rse}^2_{L_x} - \text{rse}^2_{\text{cs}})}. \quad (3)$$

Two histograms showing the 50 estimates of instrumental uncertainty are given in Fig. 2a and b. The first represents the estimates when only the first 0.04 s (two data channels) of optical stimulation time are used for the OSL signal; the second histogram represents the estimates when most of the signal, or 0.3 s (15 data channels) of optical stimulation time, is integrated. The median value was used to quantify the instrumental uncertainty obtained from the 50 estimates at both signal integration levels. Values of 2.09% (mean = 2.57%) and 1.18% (mean = 1.28%) (indicated on the histograms as broken lines) were calculated for 0.04 and 0.3 s of optical stimulation time, respectively. As was observed by Thomsen et al. (2005), the instrumental uncertainty decreases as a longer integration period is used. Both values of instrumental uncertainty are notably lower than the 3.5% value obtained by Truscott et al. (2000), even though he used a longer integration time, and this reflects the technical improvements that have been made to the single grain system in this time.

In a more recent, and differently executed, study, Thomsen et al. (2005) reported instrumental uncertainty on the same Risø single grain system as used in the present study. They calculated the relative standard deviation for 15 repeated L_X/T_X measurements for each of 92 grains and found it to be 3.6% when the luminescence signal is integrated over the first 0.03 s of optical stimulation time or 2.1% when the integration interval is increased to 0.3 s; thus, for a single measurement (e.g. L_X) this value is to be divided by $\sqrt{2}$, i.e. 2.5% or 1.5%. These values are very similar to the values obtained in this study (2.1% and 1.2%) for integration intervals of 0.04 and 0.3 s, respectively. The estimate by Thomsen et al. (2005), and those from this study, confirms the improvement in instrumental precision since the first measurements were made on the prototype system (Truscott et al., 2000). Thus, it is essential that the instrumental reproducibility is assessed for each Risø reader before D_e distributions can be interpreted. The values for the single grain laser luminescence unit are not dissimilar to the value of 1.9% ($2.7/\sqrt{2}$) obtained when single grains were measured using a conventional Risø TL/OSL reader equipped with blue LEDs (Thomsen et al., 2005).

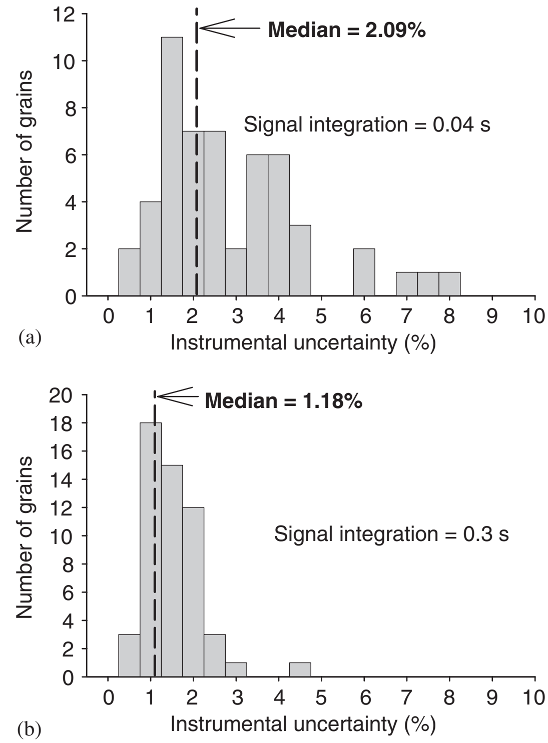

Fig. 2. Histograms showing the instrumental uncertainty (%) obtained for each of 50 grains given the same dose repeatedly. Results are shown using two different integrals of the optical stimulation time (a) 0.04 s and (b) 0.3 s (shown in Fig. 1).

4. Establishment of rejection criteria

Once the instrumental uncertainty has been established, measurements can be made to obtain D_e values for grains that have not been thermally treated or specially selected. Using the Risø single grain system, several hundred values of D_e can be measured; these can be analysed once any unreliable D_e values have been rejected. However, before any grounds for rejection of unreliable D_e values can be investigated, two types of grain behaviour need to be mentioned. There are some grains for which no photon counts can be observed for the test dose OSL signal, though a grain can be seen at the measurement position. For most samples the percentage of such grains is high (e.g. ∼ 70% was reported by Jacobs et al., 2003b for their three samples). For these grains, no D_e value can be obtained and these are not mentioned again. There are other grains for which the L_N/T_N ratio is well above the maximum L_x/T_x value that can be generated as the dose response curve is produced, even if the sample is taken to saturation; this characteristic results in an inability to obtain a D_e value because there is no intersection with the dose response curve.

For the remaining grains, the signal intensity of the test dose signal should be monitored, to ensure that the correction for sensitivity change is not hampered by a lack of recorded photon counts. Only then, for those grains for which it was possible to obtain a value of D_e can rejection criteria be proposed to eliminate those grains for which the SAR measurement procedure was not appropriate. Rejection can only be allowed when there is a valid reason for rejection. Inherent to the SAR measurement procedure are several tests that can indicate whether the procedure is appropriate or not. These are related to the recycling ratio tests, thermal transfer of charge and signal build-up and the OSL IR depletion ratio test.

In the next section, the rejection criteria are demonstrated being applied to artificially irradiated grains, since in this situation one can be confident of the dose received by the grains. Similar rejection criteria were used for the dating studies of Jacobs et al. (2003b), Feathers (2003) and Olley et al. (2004a) and have been reported by Olley et al. (2004b) for single grains from a wide range of Holocene sediments. However, these studies used naturally irradiated grains where the situation is more complex, so it is difficult to rigorously appraise the effectiveness of these rejection criteria. Such naturally irradiated grains were not necessarily exposed to sufficient sunlight at the time of deposition to reset the OSL signal, and so they may have retained some part of the geological dose. In addition, they may not have been exposed to an identical flux of ionising radiation during burial, with the result that grain-to-grain scatter would be expected due to beta micro-dosimetry. Also, the average dose received by the grains was not accurately known.

4.1. D_e distribution for a dose recovery experiment

In order to establish the rejection criteria proposed in this paper, a sample disc was held at a temperature of 125 °C and grains from SIB2 were bleached using the green laser for a sufficiently long time (2 s) to zero the OSL signal. The grains were then all given an identical beta dose (108 Gy) with the source mounted in the automated reader. The SAR procedure used in this dose recovery experiment employed a preheat of 260 °C for 10 s, a cutheat of 220 °C, and a stimulation temperature of 125 °C. A high test dose of 54 Gy was used to maximise the number of grains that could be observed. Previous experiments on multiple grain aliquots had shown that this high test dose (50% of the given dose) did not give rise to any problems (Jacobs, 2004). Also, after measurement of T_X (the signal resulting from the test dose), the grains were stimulated using the green laser for 2 s whilst held at 280 °C, a procedure based on the modification suggested by Murray and Wintle (2003) for single aliquots. Two doses were repeated as recycling points, 36 and 216 Gy. The signals used to construct the dose response curves were the first 0.3 s with the background being obtained from the average of the last 0.8 s of the 2 s stimulation time. For each grain this value was consistent with the photon count rate determined for an empty hole (∼ 3 cts/s).

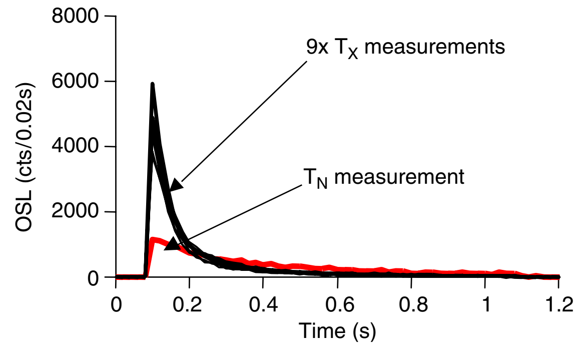

Fig. 3. Laser stimulated OSL decay curves for a grain of SIB2 given 10 repeated test doses of 54 Gy in the dose recovery experiment.

Of the 1000 grains measured, 749 grains gave measurable OSL signals and a dose response curve could be constructed (Table 1). Of these, 38 gave values of L_N/T_N that did not intersect the dose response curve, even though the dose had been delivered in the laboratory. This type of behaviour has been reported by Armitage et al. (2000), Yoshida et al. (2000), Jacobs et al. (2003b), Feathers (2003) and Olley et al. (2004a,b) for natural sedimentary samples. Bailey (2004) has suggested that this may be a result of charge competition caused by the non-identical field and laboratory conditions and that this may be alleviated using pulsed-irradiation and an elevated temperature. In the current study of SIB2, two other possible causes were identified. Firstly, the T_N OSL signal decays much slower than the decay curves for the nine other T_x decay curves. The shape of this T_N curve is also different from any of the L_X decay curves; this resulted in the L_N/T_N ratio being too great. The most probable reason is that the laser beam may have not hit the grain squarely, resulting in a lower optical stimulation power; this is demonstrated in Fig. 3. Such low values are also seen for L_x and T_x measurements for other grains, but are of particular concern in T_N (or L_N) measurements. Secondly, for some grains there appeared to be a continuous increase in background signal with each successive regenerative-dose cycle, rather than a dose dependent signal build-up.

The measured doses for the remaining 711 grains are presented in a radial plot (Fig. 4a) where the reference value

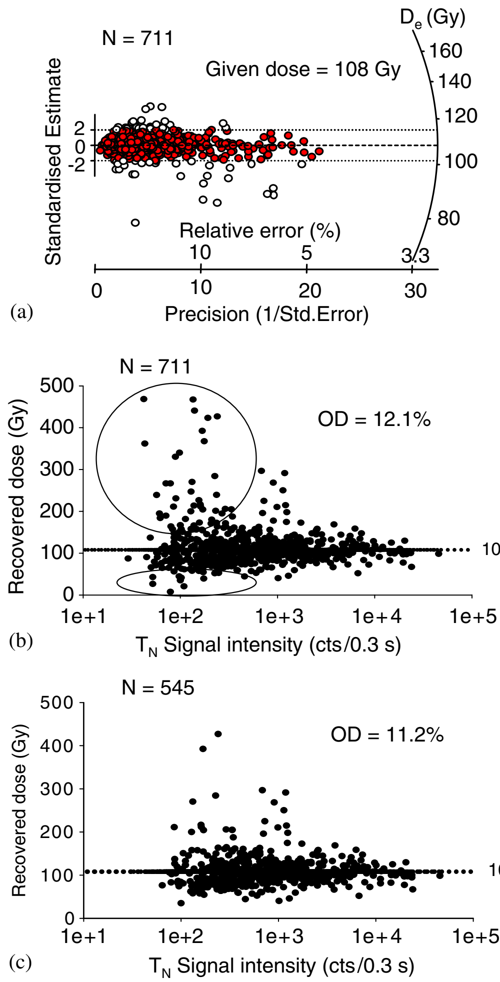

Fig. 4. Equivalent dose values for 711 grains of SIB2 that had been optically bleached and given a known beta dose of 108 Gy. Equivalent dose values obtained by SAR for grains for which a dose response curve could be constructed and an equivalent dose value obtained, displayed as (a) a radial plot; (b) and (c) as plots of equivalent dose (recovered dose) as a function of the first test dose measurement (test dose was 54 Gy); (b) shows all measured dose values, whereas (c) shows those left after rejection of grains for which the test dose signal was less than three times the background.

is the given dose of 108 Gy. The intrinsic brightness of the grains from SIB2 led to a large number of grains ($N = 711$) for which doses could be calculated. This can be compared to the dose recovery experiment of Roberts et al. (1999) for which only 76 grains gave measurable doses. The uncertainties in the measured dose for each grain were based on counting statistics, the instrumental uncertainty of 1.18% obtained in this paper and curve-fitting uncertainties using the Analyst software written by Duller that is widely available. A range of uncertainties are shown, with the smallest being about 5% (Fig. 4a). The dose value obtained with the central age model is 108 ± 1 Gy, which is consistent with the given dose. The open circles in the radial plot represent individual values that are not consistent with the given dose i.e. are not within 2σ; the OD value is calculated to be 12.1%.

In Fig. 4b the recovered dose is plotted as a function of the first test dose signal intensity (T_N) of each grain. The x-axis is plotted on a logarithmic scale to encompass the full range of signal intensities. The given dose (108 Gy) is also indicated on this plot. This can be compared with that of Olley et al. (2004b; Fig. 4b) for grains from a dune sand for which there was radiocarbon age control. For the current data set, there is considerably less scatter, as would be expected for recovery of a laboratory dose. However, the calculated OD is 12.1%, thus implying sources of error in making the measurement that are in addition to the instrumental uncertainty, counting statistics and curve-fitting uncertainty.

4.2. Signal intensity

The absolute intrinsic brightness of the individual grains presented in Fig. 4b varies by over two orders of magnitude and it can be seen that the range of D_e values obtained increases as the first test dose signal intensity (T_N) decreases. This phenomenon was also observed by Duller et al. (2000) for one of their Tasmanian dune samples. In this study of SIB2 some of the grains ($N = 166$) gave test dose signals barely above background; these grains were rejected using the criterion that the test dose signal following the first OSL measurement is less than three times the background ($T_N < 3 \times \mathrm{BG}$). This rejection did not make a significant difference to the shape of the distribution in the radial plot (not shown), since these are primarily low precision D_e values that may well be consistent with the given dose within two standard deviations. However, the OD value was decreased by approximately one percentage point to 11.2% after these grains had been rejected. The impact is seen in Fig. 4c for the D_e distribution for the remaining 545 grains. This results in a reduction in scatter in the low signal intensity part of the plot; in addition, it eliminated both the grains that overestimated, as well as the grains that underestimated, the given dose in those areas indicated by circles in Fig. 4b. However, there is still substantial spread in the data that was not dealt with using this rejection step.

4.3. Sensitivity correction—recycling ratios

Murray and Wintle (2000) recommended the use of a recycling ratio test to check on the sensitivity correction procedure when using the SAR measurement procedure for single aliquot measurements. In the current study two regeneration doses were repeated, one low (36 Gy) and one high (216 Gy) on the dose response curve; this approach was adopted because D_e was expected to be about 100 Gy, well up the dose response curve. A grain was first rejected when

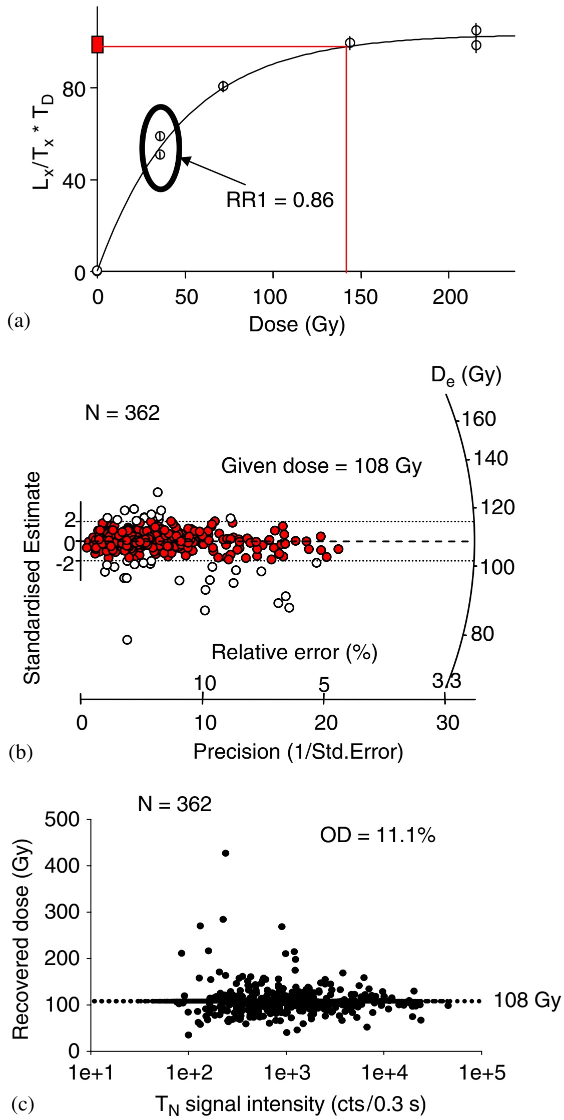

Fig. 5. (a) Dose response curve for one grain of SIB2 that fails the RR1 test with a recycling dose of 36 Gy, (b) radial plot following rejection of grains that failed the RR1 test and (c) recovered dose as a function of the first test dose measurement.

the low dose recycling ratio (RR1) exceeded 1.1 or was less than 0.9, as suggested by Murray and Wintle (2000). No account was taken of the uncertainty on the recycling ratio. This is a more stringent procedure than using the uncertainty on individual grains and would have resulted in rejection of more grains. An example of a dose response curve for which the first recycling ratio is 0.86 ± 0.06 is shown in Fig. 5a. As for all grains, the two recycling points were used in the fitting of the growth curve, and all grains were either fitted with a saturating exponential function or the sum of a saturating exponential function and a linear term. One hundred and eighty-two grains were rejected on the basis of their recycling ratios and the dose distributions excluding these grains are presented in Fig. 5b and c. The shape of the radial plot has not changed much, since most of those grains had low precision dose values; it can be seen in Fig. 5c that the grains that were rejected included some of the grains for which the dose was overestimated. This rejection made very little difference to the OD value (11.1%). Feathers (2003) also used a single low dose recycle point, but increased the range for acceptance to between 0.8 and 1.2. Jacobs et al. (2003b) also used this wider range for their three sands and rejected 2% of their measurable grains on this criterion.

However, there are a number of grains ($N = 90$) for which the second higher repeated dose point (216 Gy) did not recycle well, but for which RR1 was acceptable. An example of a dose response curve for which RR2 was unacceptable is shown in Fig. 6a. The dose distribution following rejection of the grains that failed the RR2 test is shown in Fig. 6b and c. The radial plot has changed substantially, with the majority of the outliers that both overestimated and underestimated the given dose being eliminated. The OD has been reduced to a value of 9.0%.

4.4. OSL IR depletion ratio

The impact that the presence of grains that give an IR stimulable signal at room temperature (50 °C) can have on single grain D_e distributions has already been demonstrated for the three sand samples from Blombos Cave (Jacobs et al., 2003b). Although all grains have been density separated using a sodium polytungstate solution with a density of 2.62 and 2.70 g/cm^3 to remove potassium and sodium feldspar grains and heavy minerals respectively, and also have been etched using hydrofluoric (HF) acid for 40 min to remove any remaining feldspar, it is still possible that some feldspars or other mineral contaminants (possibly as inclusions) may remain in the quartz extracts (Baril, 2004). A procedure to identify the presence of feldspar has been developed by Duller (2003); it is based on the depletion of the OSL signal by a prior exposure to infrared. This test, the OSL IR depletion ratio test, is applied at the end of a SAR run.

When applied to the SIB2 grains, very few failed the OSL IR depletion ratio test. Only four grains out of 749 from which any OSL signal could be observed were rejected using this test. These grains had ratios of the sensitivity-corrected OSL signals (after/before exposure to infrared) that were significantly less than unity (at two standard deviations) and were regarded as grains containing feldspar. However, as for the data sets presented in Jacobs et al. (2003b), these few grains had a great impact on the OD of the distribution. These four grains are indicated as filled squares in Fig. 6b. All four underestimate the dose and fall below the 2σ limit. It should be noted that since this is a dose recovery experiment, anomalous fading cannot be invoked as an explanation, and it is more likely that SAR is inappropriate for such

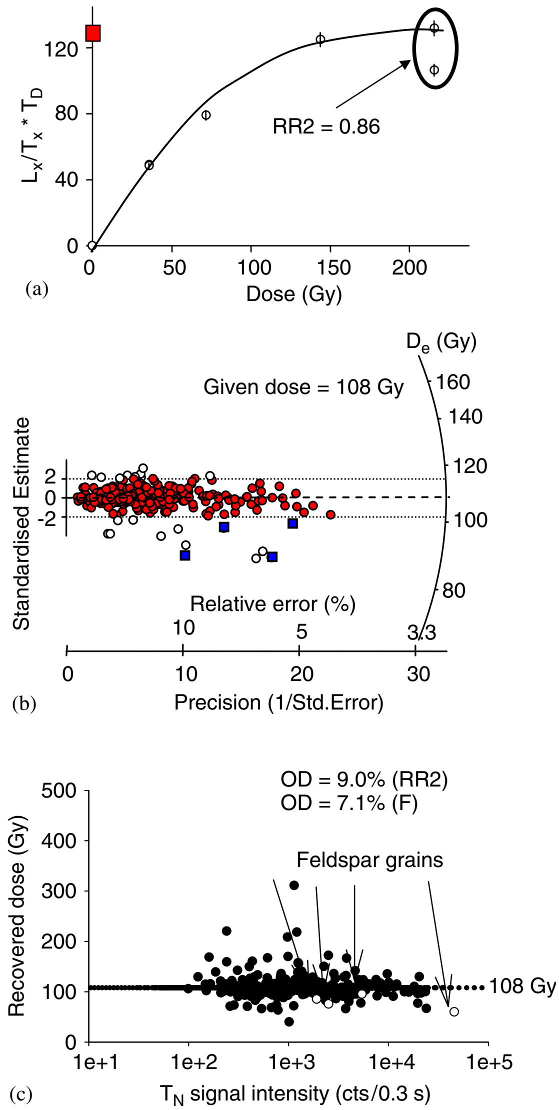

Fig. 6. (a) Dose response curve for one grain of SIB2 that fails the RR2 test with a recycling dose of 216 Gy, (b) radial plot following rejection of grains that failed the RR2 test and with feldspar grains identified as filled squares, and (c) recovered dose as a function of the first test dose measurement with the feldspars identified as open circles.

minerals (Wallinga et al., 2000). In Fig. 6c they are presented as open circles and indicated with arrows. Removing these four points reduced the OD from 9.0% to 7.1%. In this data set the brightest grain was a feldspar and it had the sixth smallest error. It is therefore very important to test for feldspar contamination and to reject these grains from the data set. Another characteristic of these grains is that their OSL signal did not reach background after 1 s of stimulation (Jacobs et al., 2003b, figure 4b).

4.5. Recuperation of the OSL signal

Recuperation can be assessed using the L_x/T_x ratio of the zero dose regeneration point and can be expressed numerically as a percentage of the L_N/T_N ratio. Murray and Olley (2002) suggested that a value of up to 5% is acceptable. Using this 5% criterion, 53 of the grains from SIB2 failed. It was found that the majority (45) of these grains were already rejected based on either the $T_N < 3 \times$ BG criterion or the RR1 and RR2 tests. The impact on the distribution was small (not shown) since it eliminated grains that both under- and overestimated the given dose; also these were low precision grains. The OD value was reduced from 7.1% to 7.0%.

5. Precision in D_e measurements

In Section 3, the instrumental uncertainty pertaining to a single OSL measurement was measured as 1.2%. For each D_e measurement in the dose recovery experiment of Section 4.1, individual measurements of precision were assigned, taking into account this value of instrumental uncertainty, counting statistics relating to each L and T measurement, and curve fitting. As mentioned in Section 2, the OD is the relative standard deviation of the values from the central value, after the above combined uncertainty has been allowed for. For the dose recovery experiment, OD was found to be 12.1%. Application of rejection criteria in Sections 4.2–4.5 resulted in this value being reduced to 7.0%; this suggests that there was a source of error that had not been accounted for.

This result differs from that obtained by Thomsen et al. (2005). In their work, when they gave a known beta dose to grains of a sample while they were within the single grain reader, they were able to entirely explain the observed dispersion in the values of D_e that they obtained, implying zero OD. However, there are a number of differences between their experiment and that described here. Firstly, in this paper the sample was bleached, but not heated, while Thomsen et al. (2005) annealed their sample prior to administering their known beta dose. The effect of this would be to largely equalise the sensitivity of all the grains. Secondly, Thomsen et al. (2005) used a much smaller beta dose (7 Gy) compared with that used here (108 Gy), and thus they would not have been affected by difficulties encountered when grains get close to their saturation level. These differences reflect the different purposes of the two studies. Thomsen et al. (2005) aimed to establish whether it was feasible to fully explain on the basis of statistics a suite of single grain D_e values when the sample and the measurement conditions were ideal. The aim of the current study was to assess the ability to recover a dose of a similar magnitude to that of the natural D_e of the samples (∼ 60–120 Gy) using grains in their natural condition.

Table 1
Summary of the number of grains measured and rejected using the various criteria presented in Section 4

	108 Gy	Natural dose	
	SIB2	ZB13	SIB2
Total number of grains measured	1000	4332	700
Contributing to total T_N light sum	749	826	446
Giving a dose response curve	749	228	446
Grains were then rejected for the following reasons:			
No L_N/T_N intersection	38	27	151
T_N signal $< 3 \times$ BG	166	0	25
Poor recycling ratio (RR1 and RR2)	272	23	138
Depletion by IR	4	8	6
Recuperation $> 5\%$	53	0	32
Sum of rejected grains	447	64[a]	322
Acceptable individual D_e values	302	164	124

Data are presented for the dose recovery experiment on SIB2 and the natural sediments ZB13 and SIB2. Note that some of the grains failed more than one rejection criterion; thus the individual numbers of rejected grains do not add up to the total.
[a]This number includes six grains that were identified as 'modern' (Jacobs et al., 2003b).

There are two ways to view the 7.0% OD that remains after application of the rejection criteria described in Section 4. The first is that there is a remaining source of random error that we have not taken into account, or that we have underestimated the magnitude of one or more components of the error. Alternatively, it is possible that we are correctly assessing the random error within our measurement procedure, but that in spite of our rejection criteria, there are still grains remaining which are unsuitable for the SAR protocol. This latter effect would be more likely to be seen in this experiment than that of Thomsen et al. (2005) since the combination of annealing the sample prior to dosing (causing the grains to behave more uniformly), and the use of a small known dose (well below signal saturation level), would minimise their impact.

Without the implementation of rejection criteria, the single grains measured in the dose recovery experiment (Fig. 4a) showed an OD of 12.1% in spite of allowing for measurement uncertainties. By removing 447 grains (Table 1, column 1) that were not suitable for SAR, i.e. ~60% of the grains with a measurable signal, this was reduced to 7.0%. It is likely that this value will be different for different samples, depending upon the range of behaviour found for individual grains. Further work is required to develop additional rejection criteria in order to reduce the OD value for a dose recovery experiment to near zero, but this is beyond the scope of this paper. Recent reviews of the SAR procedure suggest that an OSL signal dominated by the fast component is important for its success (Murray and Wintle, 2003), and criteria to reject grains that lack such a component may be essential.

In conclusion, it should be noted that for the dose recovery experiment the central D_e value and its calculated uncertainty was unchanged at 108 ± 1 Gy by application of the existing rejection criteria.

6. D_e distributions for naturally irradiated quartz

The same rejection criteria were applied to the D_e distributions from two naturally irradiated samples. D_e values for SIB2 (295 grains—excluding the grains with no L_N/T_N intersection) and ZB13 (201 grains—excluding the grains with no L_N/T_N intersection) are presented in Figs. 7a and 8a before rejection of any of the grains and in Figs 7b and 8b after those grains that failed the proposed rejection criteria were eliminated from the data sets. The numbers of grains that have failed each of the criteria are provided in Table 1 columns 2 and 3. (It should be noted that some of the grains failed more than one rejection criterion; as a result, the numbers of individually rejected grains do not add up to the total number of rejected grains.)

For SIB2 (Fig. 7a and b), the central D_e remained the same (125 ± 3 Gy) but the OD value was reduced from 41.9% to 21.8% as the number of grains for which D_e values can be determined were reduced from 295 to 124. In Fig. 7a there are a number of grains that appear to have a high D_e value and may have been taken as a second population, perhaps relating to in situ disintegration of roof spall or mixing in of grains from older units (upper ellipse in Fig. 7a). Most of these D_e values had relatively high uncertainties, with relative errors of greater than 10%. These grains were removed by application of the rejection criteria, primarily related to poor recycling ratios. This problem was also related to the proximity of L_N/T_N to the saturation level of the dose response curve for some grains, a condition that is influenced by the ability to recycle the high dose point. In addition, a number of grains that had similar uncertainties, but fell on the low side of the central D_e value, were also rejected (lower ellipse in Fig. 7a). However, there are still a relatively large number of outliers with relatively high precision (Fig. 7b). These results contrast with those in

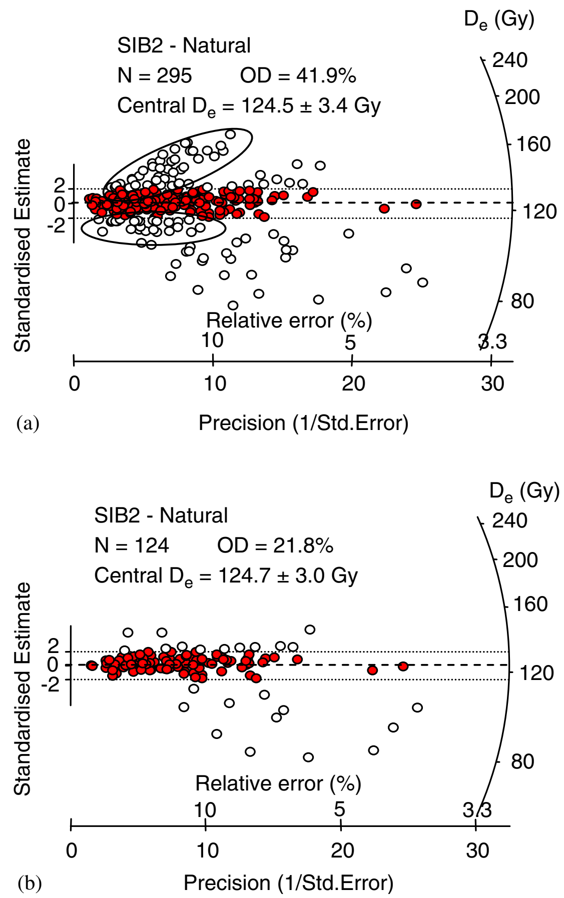

Fig. 7. Radial plot of D_e values for grains from a natural sedimentary sample from Sibudu Cave (SIB2) (a) before and (b) after rejection criteria were applied. The circled regions in (a) show the major groups of rejected grains.

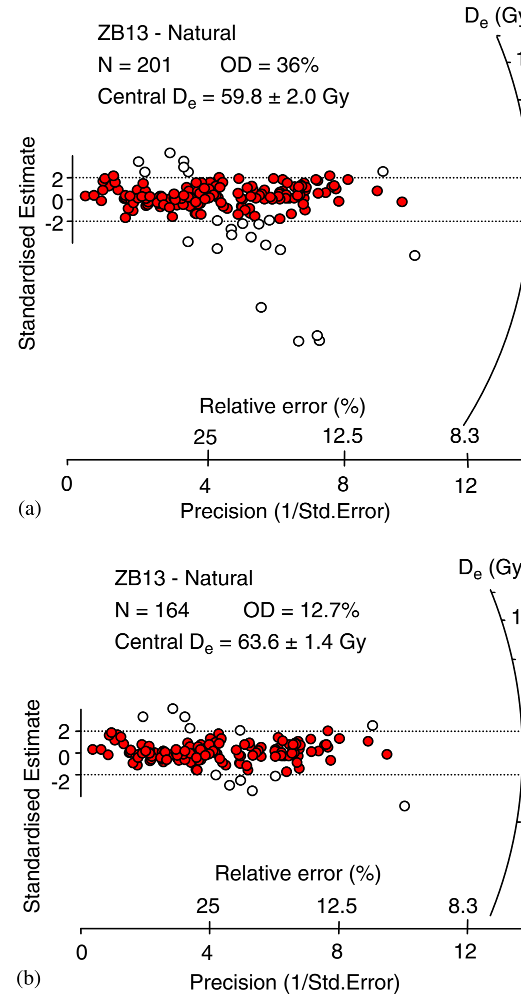

Fig. 8. Radial plot of D_e values for grains from a natural sedimentary sample from Blombos Cave (ZB13) (a) before and (b) after rejection criteria were applied.

Fig. 6b for the same sample given a laboratory dose and for which the same rejection criteria were applied.

For ZB13 the central D_e changed from 60 ± 2 to 64 ± 1 Gy and the OD value reduced from 36% to 12.7% as the number of grains were reduced from 201 to 164 (Fig. 8a and b). Thus, for ZB13, inclusion of those grains that were rejected may have led to the erroneous interpretation that the sample had been contaminated by younger grains. Now only one grain appears to be an outlier and is known with high precision. The value of 12.7% will contain the 7% OD from the measurement procedure (as discussed in Section 5), and will also contain the effects of non-uniformity in dose rate in the natural environment and incomplete zeroing. Olley et al. (2004a) reported that OD values of up to 20% could be found for uniformly bleached grains.

Two additional data sets for grains from Blombos Cave are presented in Fig. 9. Sample ZB4 (Fig. 9a) is taken from a level with shell beads (Henshilwood et al., 2004) and sample ZB8 (Fig. 9b) is taken from a hearth. For ZB4 and ZB8, 2200 and 2400 grains were mounted for single grain measurements, but dose response curves could only be obtained for 287 and 1321 grains, respectively, 190 and 1014 grains, respectively, were then rejected using the same rejection criteria as for SIB2 and ZB13. Applying the central age model to all the data points resulted in a D_e of 80 ± 2 Gy ($N = 97$) for ZB4 and 97 ± 2 Gy ($N = 307$) for ZB8. These two samples are representative of other samples found in Blombos Cave and will be used as the basis for the discussion of D_e selection in the next section. The OD values were 13.5% (ZB4) and 28.6% (ZB8) and these values encompassed the full range of values found for the five samples from the archaeological levels at Blombos Cave for which single grain measurements were made (Jacobs, 2004).

After rejection of quartz grains following the criteria outlined above, there is still considerable dispersion in the

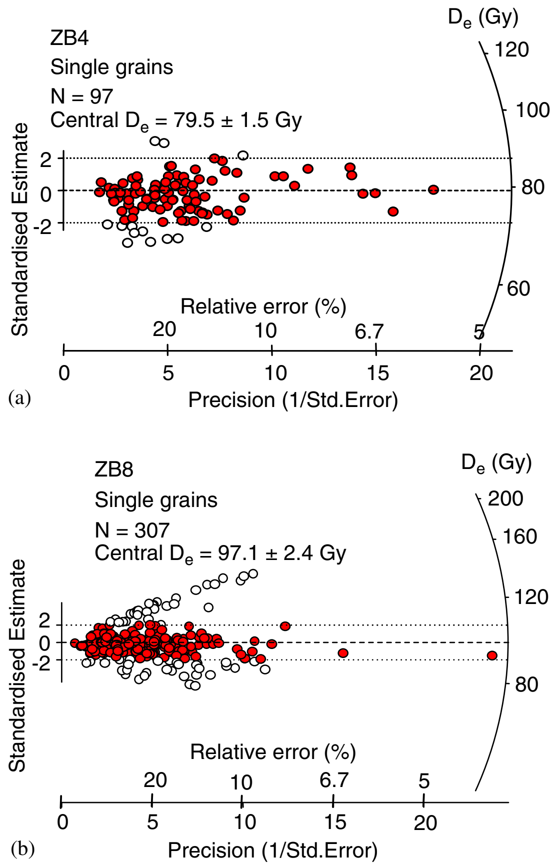

Fig. 9. Radial plots for natural samples from Blombos Cave (a) ZB4 typical of a sample with a single D_e population and (b) ZB8 typical of a sample with more than one population of D_e values.

single grain data sets for naturally irradiated grains, as compared to laboratory-irradiated grains. This OD will consist of two factors; the first being due to intrinsic factors (i.e. factors relating to the measurement procedure) as was demonstrated by our inability to account for all the OD in the laboratory-irradiated data set (Section 5), and the second being related to extrinsic factors that can include any factors related to depositional and post-depositional processes. Most published reports explained or attributed the observed D_e dispersions in the single grain data sets to (a) differences in the beta dose received by individual grains in their burial environment (Murray and Roberts, 1997; Olley et al., 1999; Roberts et al., 1999; Duller et al., 2000), (b) insufficient or heterogeneous exposure of some grains before burial, i.e. inadequate bleaching (Olley et al., 1999; Murray and Olley, 2002; Bøtter-Jensen et al., 2000; Duller and Murray, 2000; Duller et al., 2000), or (c) post-depositional intrusion of younger grains into older deposits or vice versa (Roberts et al., 1998). Galbraith et al. (2005) also suggest that some of the extra OD seen in natural samples may arise from the 'non-identical field and laboratory conditions'. This may include, among others (1) differences in the bleaching spectra of sunlight and the Risø illumination, (2) the different types of ionising radiation where natural samples are being exposed to a mixed radiation field as opposed to beta radiation in the laboratory, (3) the orders of magnitude difference between the laboratory and field dose rates and (4) the differential extent of charge redistribution resulting from natural and laboratory irradiation.

At archaeological sites such as Blombos Cave and Sibudu at least some, or maybe all, of these factors may have an influence on the D_e distributions and therefore on the estimation of the true burial dose calculated from them. In the previous sections, the central age model of Galbraith et al. (1999) was applied to all data sets so that they could be compared with one another. It is now appropriate to consider which statistical model should be applied to the data to obtain the best estimate of D_e.

7. Calculation of D_e

7.1. Single population D_e distributions

Galbraith et al. (1999) proposed two statistical models to calculate D_e for a single population of D_e values, namely the 'common age model' and the 'central age model'. An example of a single population D_e distribution is the laboratory data set from SIB2 obtained after rejection of those grains for which the SAR measurement procedure was not appropriate (Fig. 6b). The 'common age model' (Galbraith et al., 1999) assumes that the sample is an ideal one where all grains have received exactly the same dose and that any variation in the measured D_e values can be accounted for by the measurement uncertainties (these include the instrumental uncertainty and the uncertainties connected with counting statistics and curve fitting). The 'central age model' (Galbraith et al., 1999) assumes that the distribution of logarithmic D_e's is normal but that they are not consistent with a single value of D_e; instead they are randomly spread around a central D_e value with a standard deviation that is too wide to be accounted for by measurement uncertainties alone. The choice of which of the two models, 'common age' or 'central age', is most appropriate will therefore depend on whether any OD of the measured D_e's is observed; if there is no OD, then the 'common age' model reduces mathematically to the 'central age' model.

For the data set in Fig. 6b, the 'common' and 'central' age D_e values have been calculated for these grains that had all received 108 Gy. This resulted in a 'common age' D_e of 107 ± 1 Gy and a 'central age' D_e of 108 ± 1 Gy. The values obtained using the 'common age model' and the 'central age model' are very close to each other, consistent with the knowledge that this data set represents a single dose population. As shown in Fig. 6c, the OD observed for this sample was calculated as 7.0%, and this must be included with the statistical error, curve-fitting error and instrumental error; since the OD is not zero, the central age model is more appropriate.

The data set for ZB13 given in Fig. 8b had an OD of 12.7%. This value is similar to that of another dune remnant outside the cave at Blombos, ZB20 for which OD = 11.4%, and also that for the dune layer inside the cave, ZB15 for which OD = 13.5% (Jacobs et al., 2003b). These values of OD contain the effects of micro-dosimetry and led the authors to select the central age model for D_e calculation.

7.2. *Multiple components D_e distribution*

One of two models may be appropriate if there are grains with different bleaching and/or irradiation histories, namely the minimum age model and the finite mixture model. Galbraith et al. (1999) suggested that the minimum age model should be used for samples that contained a mixture of grains that were well bleached, partially bleached and unbleached (e.g. some fluvial samples). The most appropriate D_e would therefore relate to the lowest range of D_e's, i.e. those that represent the bleached grains. Olley et al. (2004b) recommended use of the minimum age model for a range of Holocene sedimentary types that contained grains that had not been completely bleached. However, for cave sediments, such as the samples from Blombos Cave and Sibudu examined in this study, application of this age model is not always appropriate. Using the minimum age model may be erroneous because of the possibility of downward percolation of grains from a younger overlying deposit.

A number of methods of analysing data sets consisting of mixtures have been proposed (Roberts et al., 2000; Sivia et al., 2004). In this study the finite mixture model of Roberts et al. (2000) has been applied. The programme for the finite mixture model was written by Rex Galbraith; it uses the formulae and algorithm presented in Appendix A of Galbraith (1988), but is expressed in terms of the actual observations rather than the standardised observations as in the case of the publication. This finite mixture model enables one to estimate the number of dose components within a dose distribution, the corresponding D_e for each component, and the relative proportion of grains in each component.

In this model, the standard deviation (σ) of each component is treated as a known parameter that is the same for all the different components. This is unlike the calculation in the 'central age model' where σ is an unknown parameter. The applied standard deviation for the natural samples (in contrast to laboratory-irradiated samples) was based on the OD value obtained for the samples from the dune layer inside (13.5% for ZB15) and outside (12.7% for ZB13 and 11.4% for ZB20) Blombos Cave since this is believed to represent a single dose population (Jacobs et al., 2003b). The value of OD adopted was 12% and thus the standard deviation for each component was $\sigma = 0.12$. The number of dose components (k) was then determined using 1000 iterations.

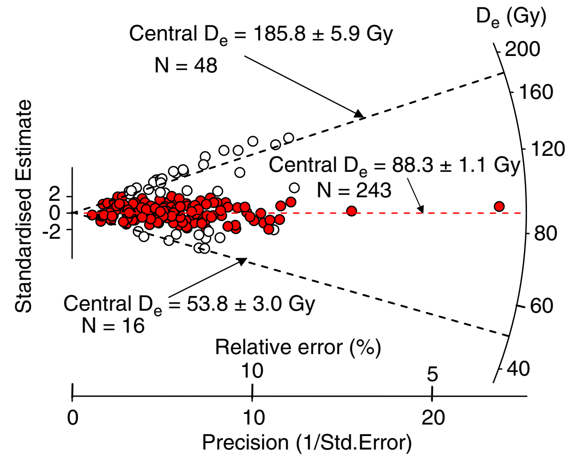

Fig. 10. Radial plot for ZB8 showing the three populations of grains identified by applying the finite mixture model. The central age model calculation was then applied to each population.

An example of a D_e distribution from Blombos Cave where it is likely that there are two or three discrete populations of grains with different doses is presented in Fig. 10. This sample (ZB8) is from a Middle Stone Age hearth where mixing between units is possible because of anthropogenic factors. A finite mixture model has been applied to this D_e distribution ($N = 307$) with $\sigma = 0.12$ and $k = 3$. This analysis resulted in three dose populations with central D_e values of 53.8 ± 3.0, 88.3 ± 1.1 and 185.8 ± 5.9 Gy with mixing proportions of 5.1%, 79.3% and 15.7%, respectively. The most appropriate D_e value for this sample is 88.3 ± 1.1 Gy because 79.3% of these grains are consistent with this D_e value. The higher dose population is similar to the D_e value (∼ 230 Gy, unpublished data) obtained for the older sterile sand that occurs a few centimetres below the sampling point for ZB8.

8. Conclusions

The instrumental reproducibility for the latest Risø TL/OSL single grain readers, obtained using the OSL signal from the first 0.3 s of optical stimulation time, was 1.2%. This is about 30% of the value for the prototype reader (Truscott et al., 2000) and similar to the value obtained by Thomsen et al. (2005)

A dose recovery test using optically zeroed grains given a 108 Gy applied dose gave a calculated OD value of 12.1%, even after incorporation of the 1.2% instrumental uncertainty and calculated statistical errors; the recovered dose value was 108 ± 1 Gy, as calculated using the central age model for a single population for the 711 grains for which values of D_e could be obtained. A series of rejection criteria (based on data available from the SAR runs on each grain) were applied to remove D_e values for which the SAR procedure was inappropriate and the OD was reduced to 7.0%. It is thought that this remaining OD arises because there are still grains present which are not suitable for the SAR procedure, and future work is required to determine additional rejection criteria to remove these.

The rejection criteria that were applied in the dose recovery experiment were applied to measurements made on naturally irradiated grains from two samples of archaeological interest from sites in South Africa, SIB2 from Sibudu and ZB13 from Blombos Cave. Out of the large number of grains measured for each sample, only those grains for which dose response curves could be constructed were selected for further evaluation. Grains were then rejected because the sensitivity-corrected natural measurement did not intersect the dose and response curve, gave a weak test dose signal, did not pass the recycling ratio test for an appropriate regeneration dose, and showed too high a level of recuperation or had their OSL signal depleted by exposure to IR (indicating the presence of feldspar).

For these two samples, the effect of rejecting these grains was assessed by calculating both the central D_e value and the associated OD. In the case of SIB2, the central D_e remained about the same, but the OD value was reduced from 41.9% to 21.8% when the number of grains for which values of D_e could be obtained was reduced from 295 grains to 124. For ZB13 the central D_e value increased by 6% and the OD value was reduced from 36% to 12.7% when the equivalent number of grains was reduced from 201 to 164. For SIB2 the OD value of 21.8% was 14.8% above the value of 7% found in the dose recovery experiment on the same sample. There are many possible reasons for this and they can be related to the non-identical nature of field and laboratory conditions as discussed by Galbraith et al. (2005).

Following application of the same rejection criteria, OD values of 28.6% and 13.5% were found for two samples from archaeological occupation levels at Blombos Cave (ZB8 and ZB4, respectively). For ZB8 a finite mixture model was required to select the most appropriate D_e value, rejecting outlying values on the basis of statistics.

This study lays the foundation for obtaining a rigorous chronology for the sandy archaeological levels at Blombos Cave and Sibudu Cave. Similar studies are recommended for other dating projects.

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