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ON THE DETECTION OF OSL AGE OVERESTIMATION USING SINGLE-ALIQUOT TECHNIQUES

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Abstract: **Optically stimulated luminescence (OSL) dating is a quantitative dating method to determine the time of last exposure of sand and silt to (sun) light. However, insufficient resetting of the optically stimulated luminescence signal prior to sediment deposition can result in overestimation of the age of a sample. Therefore detection of so-called poor bleaching is of prime importance in OSL dating. Several methods proposed in the literature for detection of poor bleaching are based on the scatter in equivalent doses obtained by single-aliquot methods. In this paper numerical simulations are used to assess the validity of these methods. The simulations show that scatter in equivalent doses is largely dependent on the number of grains contributing to the luminescence signal of each aliquot, and proportion of poorly-bleached grains in the mixture under study. Thresholds for detection of poor bleaching based on inter-aliquot scatter in equivalent doses are therefore not valid. It is concluded that tight, symmetrical dose distributions obtained on small aliquots (ultimately consisting of a single grain) provide the best indication that the sample is homogeneously bleached.**

1. INTRODUCTION

The luminescence properties of quartz and feldspar grains can be used to determine the timing of the last exposure of sand and silt to (sun) light. Luminescence dating has seen major developments over the last decades, especially since the development of optically stimulated luminescence (OSL) dating (Huntley *et al*., 1985). This dating technique, also known as optical dating, can provide absolute ages in the range of decades to over a hundred thousand years, and is now widely applied in the fields of both geomorphology (reviewed in Aitken, 1998; Stokes, 1999) and archaeology (reviewed in Roberts, 1997). OSL dating is particularly applicable to deposits for which the light-sensitive OSL signal in all the grains has been zeroed by the exposure to sunlight during erosion, transportation, and deposition. If the OSL of some grains is not fully zeroed, an overestimation of the depositional age will occur. Thus, to have confidence in the accuracy of an optical age, it should be shown that the residual signal at the time of deposition was negligible for all the grains.

The OSL signal given by a grain is the result of the optical release of charge trapped in the crystal lattice. This charge is produced at an almost constant rate through time, as a result of exposure to ionising radiation from radioactive nuclides in the environment. The radiation dose is thus a measure of the burial time. For dating, its magnitude is determined by laboratory measurements of the OSL from the mineral grains. Thus derived, it is known as the equivalent dose. Put together with the measured environmental dose rate, an optical age can be calculated.

The OSL signal is very quickly zeroed by exposure to (sun) light, much faster than the thermoluminescence (TL) signal previously used for dating (Godfrey-Smith *et al*., 1988). This has opened the possibility of using OSL dating for materials that have only been exposed to light for a short period of time, or that have been exposed only to light of restricted intensity prior to deposition. Consequently, OSL dating has been used in a wide range of depositional environments, including fluvial, glaciofluvial, marine and colluvial. However, although the OSL signal is reset quickly, this does not imply that it is always completely reset at the time of deposition. Light exposure might have been too short in time, too dim, or too restricted in spectrum due to filtering in, for example, a turbid water column. Alternatively, the trapped charge in the grains prior to erosion might have been very large compared with that resulting from environmental sources since deposition. In this case, the OSL signal derived from

a small portion of the former, remaining after even quite extensive light exposure, can have a large influence on the value obtained for the equivalent dose (Aitken, 1998). Serious overestimation of age due to poor bleaching has been shown in several studies using OSL dating on feldspar separates (Duller *et al*., 1995; Lamothe, 1996; Lamothe and Auclair, 1997; Clarke *et al*., 1999). For quartz separates, significant residual OSL signals were shown to be present for samples from modern glaciofluvial sediments (Rhodes and Pownall, 1994), and to a lesser extent for modern fluvial sediments (Stokes *et al*., 2001).

The most rigorous test for poor bleaching is to compare the optical age with independent age control. However, these tests assume that: 1) the age control is without problems, and 2) there are no problems other than poor bleaching in luminescence dating. Especially for feldspar separates the latter assumption is questionable, because problems with anomalous fading (Wintle, 1973; Spooner, 1994) and sensitivity changes (Wallinga *et al*., 2000) can lead to an underestimation of age. Hence, an overestimation of age caused by poor bleaching can be masked by an underestimation of age caused by other mechanisms. To circumvent these problems, several studies have sampled modern deposits to investigate the remnant dose at the time of deposition (Stokes, 1992; Rhodes and Pownall, 1994; Murray *et al*., 1995; Olley *et al*., 1998; Bailey *et al*., 2001; Porat *et al*., 2001; Stokes *et al.,* 2001). However, luminescence dating is most useful when applied to deposits of unknown age. Therefore methods to detect poor bleaching are needed and these should be intrinsic to the luminescence dating measurements.

2. OBJECTIVES

Although differences in micro-dosimetry can cause different dose rates on a grain-to-grain scale (Murray and Roberts, 1997; Olley *et al*., 1997), it is normally assumed that ionising radiation produced within a sedimentary unit is uniform (McFee, 1998; Olley *et al*., 1999). If such is the case, grains of the same size and mineral type absorb the same post-depositional dose acquiring an OSL signal that represents the trapped charge this produces. The effective absorbed dose of each grain is then the sum of the grain-dependent remnant dose at deposition and the uniform post-depositional dose. Laboratory procedures for luminescence dating aim at determining this dose; the best estimate of this dose is normally referred to as the equivalent dose. Assuming that there is no intrusion of grains from other layers, the grain with the lowest equivalent dose is expected to have been a well-bleached grain at deposition and thus provide the post-depositional dose.

In most optical dating procedures, the OSL signal is derived from a number of grains that form the sub-sample. A typical sample disc holds ~5 mg of material; this amounts to ~1000 grains when a 150 μm grain size is used. If the sub-samples contain grains with different equivalent doses, scatter will be seen for the luminescence data derived from both single-aliquot dating procedures (Li, 1994) and multiple-aliquot procedures (Rhodes, 1990; Duller, 1994a; Huntley and Berger, 1995). In single-aliquot methods, all measurements required for equivalent dose determination are made on a single sub-sample, thereby allowing comparison of equivalent doses obtained on different sub-samples (Duller, 1991). Li (1994) suggested that, using single-aliquot procedures, poorly bleached samples can be identified because they show a wide range of equivalent-dose values and an increasing trend of equivalent dose with natural luminescence intensity. His method of assessment of poor bleaching has been adopted by Duller (1994a; 1995), Duller *et al*. (1995), and Wintle *et al*. (1995).

Clarke (1996) refined Li's approach by presenting threshold values for scatter in equivalent dose (both absolute and fractional), indicating different degrees of bleaching. Clarke *et al.* (1999) used the same parameters to determine whether an OSL age can be considered to be accurate. Based on empirical studies of aeolian and fluvial sediments, Clarke *et al*. (1999) concluded that any luminescence date not showing significant (relative) scatter in equivalent doses is accurate.

Colls *et al*. (2001) use the correlation between equivalent dose and natural OSL intensity of aliquots as indication for poor bleaching. They classify samples as poorly bleached when a statistically significant correlation between these attributes is found. The statistical significance of the correlation is tested by a one tailed student's t-test on the test statistic

$$t_{calc} = \sqrt{R(n-2)/(1-R^2)}, \qquad (2.1)$$

where R is the Pearson's correlation coefficient for the normalised equivalent dose and the natural OSL intensity of the aliquots, and n is the number of aliquots. Samples that do not exhibit an apparent relationship between signal intensity and equivalent dose are regarded to be well (or at least uniformly) bleached.

A third indication for poor bleaching based on scatter in single-aliquot equivalent doses is the asymmetry of dose distributions. This attribute was first suggested by Murray *et al*. (1995) as a way of detecting poorly bleached samples.

In this paper the validity is assessed of poor bleaching detection methods that are based on characteristics of single-aliquot equivalent-dose distributions. Three different methods, proposed by Clarke *et al*. (1999), Colls *et al*. (2001) and Murray *et al*. (1995), are tested using numerical simulations of equivalent-dose distributions of poorly bleached samples.

3. COMPUTER SIMULATIONS

A simple model was constructed to study the influence of grain-to-grain variations in equivalent dose on the scatter in equivalent doses obtained for aliquots containing multiple grains. In this model virtual samples with a characteristic dose distributions are created. Different sub-samples from this virtual sample are taken, and the model used to calculate what equivalent dose would be obtained on this sub-sample when a single-aliquot procedure had been applied. Three factors control the equivalent dose that is obtained for each grain. Firstly, its intrinsic sensi-

tivity with regard to luminescence emission per unit dose. Secondly, the remnant dose that it had at the time of deposition and, lastly, the post-depositional dose (i.e. the dose that one wishes to obtain as the equivalent dose). Incorporation of the three factors in the model is briefly discussed below.

Sensitivity. Feldspar and quartz grains have been found with a wide range of intensities (Lamothe *et al*., 1994; Murray and Roberts, 1997; McFee and Tite, 1998). Even for the most uniform sand-sized quartz separates studied so far, 80% of the signal comes from only 30% of the grains, thereby being far from the 'ideal' of all grains contributing equally (Duller and Murray, 2000; Duller *et al.*, 2000). In the current computer simulation, the brightness distribution of the test-dose response of 891 grains of quartz sample TNE9503 was used (sample description and methodology can be found in Duller *et al*., 2000). The intrinsic OSL response (taken as the initial signal minus the signal observed at the end of stimulation) of a grain to the test dose is referred to as its brightness index (B_i); the average response was 1089 counts. For 20% of the grains no OSL decay was observed during stimulation, and it is assumed that these grains do not have any luminescence sensitivity; their brightness index was set to zero. Two grains where rejected because the OSL signal appeared to increase during measurement, which would result in a negative brightness. Sample TNE9503 was chosen for several reasons: Firstly, the test-dose luminescence response of a large number of grains was measured. Therefore, the brightness distribution (shown in **Fig. 1**) forms a true reflection of the variability of the sample. Secondly, the brightness distribution of this sample lies in the middle of those observed for quartz separates, and is therefore most likely to be representative for quartz in general (**Fig. 1**). Finally, the distribution is similar to that obtained on feldspar sample GDNZ1 (**Fig. 1**, previously described by Duller (1994b)), which makes the simulations described in this paper relevant to both quartz and feldspar samples. It should be noted that the brightness distribution for feldspar separate GDNZ1 was obtained with blue stimulation, but it is assumed that a similar distribution would be obtained with infrared stimulation.

Remnant dose. The equivalent dose that would be obtained if grains had been measured at the time of deposition (remnant dose, $D_{r,i}$) is most likely to be highly variable, depending upon the original nature of the deposit from which the grain was derived and the type and duration of transport. For the computer simulations, four types of original deposit were postulated, having received doses of 5, 50, 100 and 1000 Gy respectively. To allow for variations in light exposure during transport, it was assumed that the grains would have had their OSL bleached to some percentage of the original value. A top hat probability distribution was used, with equal probability of reducing the OSL signal by any fraction between 0 and 100%.

Post-depositional dose. All grains were specified to have received a post-depositional dose of 50 Gy (D_t).

For any particular grain (i), the total absorbed dose ($D_{e,i}$) will be the sum of the post-depositional dose (D_t) and the remnant dose ($D_{r,i}$). In order to use the same lingo in this paper as in applied studies, the total absorbed dose is referred to as equivalent dose here, although it is realised that the latter normally refers to the laboratory estimate of the former. For every grain in an aliquot, the natural luminescence intensity (I_i) is calculated by multiplying the equivalent dose by the brightness index ($I_i = D_{e,i} \cdot B_i$). A linear relationship between dose and luminescence signal is assumed in this calculation. From the attributes assigned to each grain, it is possible to calculate the equivalent dose that would be obtained for an aliquot containing a fixed number of grains; the simulation is carried out for aliquots consisting of 1, 5, 10, 100 and 1000 grains. The equivalent dose of an aliquot (a) holding n grains ($D_{e,a}$), is given by the summed natural luminescence intensity from all the grains on the aliquot, divided by the summed brightness index of all the grains on the aliquot ($D_{e,a} = \Sigma I_{1,n} / \Sigma B_{1,n}$). For each sample, 18 aliquots were simulated.

A second computer simulation was carried out, in which 90% of grains were constrained to have been well bleached at deposition. The remaining 10% of grains were assigned a remnant dose as in the previous simulation, and all grains were assigned a post-depositional dose of 50 Gy as in the previous simulation. Once again grains were picked at random to produce an aliquot with the desired number of grains (1 to 1000).

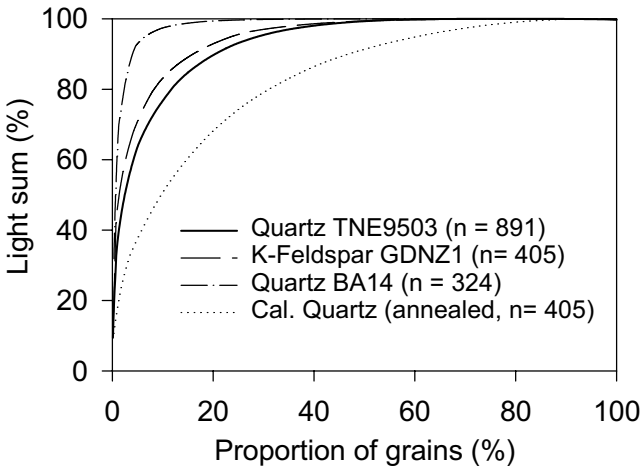

Fig. 1. *Brightness distribution of single grains of quartz and feldspar. The OSL response was measured in the Risø TL/OSL reader with single grain attachment (Bøtter-Jensen* et al.*, 2000), after administration of a uniform laboratory ionising dose and subsequent preheating. The data for this graph was kindly supplied by Geoff Duller of the University of Wales, Aberystwyth (UK). The brightness distribution of quartz sample TNE9503 is used in the computer simulations described in this paper. Its brightness distribution is similar to that obtained on feldspar from sample GDNZ1, and lies in the middle of the range of that obtained on other quartz samples (BA14 and calibration quartz). Sample descriptions can be found in Duller* et al. *(2000) and Duller (1994b), for the quartz and feldspar samples, respectively.*

4. SIMULATION RESULTS

For each scenario, the simulation described in the previous section was carried out ten times. Results are presented in **Table 1** for the simulations where all grains are poorly bleached, and in **Table 2** for the simulations where 10% of grains are poorly bleached. The values shown indicate the mean of the ten simulations, and the standard error on the mean. The percentage offset from the post-depositional dose (50 Gy) is indicated; samples for which the contamination with poorly bleached grains results in an offset in the equivalent dose of less than 7.5% are regarded as well bleached. Samples for which the equivalent dose is overestimated with 7.5% or more ($D_e \geq 53.75$) are interpreted as poorly bleached, and the percentage offset is printed in bold type-setting in **Tables 1** and **2**. Suggested indicators for poor-bleaching are also summarised in the Tables, where the values above suggested thresholds for poor bleaching are printed in bold type-setting. The indicators shown are: 1) the fractional error in the equivalent dose ($S_N = s/D_{e,av}$, threshold for poor bleaching: $S_N > 0.10$, Clarke *et al*., 1999); 2) the squared Pearson's correlation coefficient for the trend in the equivalent dose versus natural aliquot intensity (Colls *et al*., 2001, for 18 aliquots the threshold for poor bleaching is $R^2 > 0.16$); 3) the skewness of the equivalent dose distribution

$$\text{skewness} = \frac{n}{(n-1)(n-2)} \sum \left(\frac{D_{e,a} - D_{e,av}}{s} \right)^3 \qquad (4.1)$$

where n is the number of aliquots). No threshold for poor bleaching based on the skewness of the distribution has been suggested in the literature.

For graphical presentation, the equivalent dose of each simulated aliquot ($D_{e,a}$) and the natural luminescence intensity of each aliquot (I_a) are normalised to the sample average. The resulting normalised equivalent dose ($D_{e,a}$ (N)) and normalised natural OSL signal (I_a (N)) of a single simulation for each scenario, are presented in a scatter plots as suggested by Clarke (1996) and Clarke *et al*. (1999). Results of a single run of the first (all grains poorly bleached) and second (10% of grains poorly bleached) simulation experiment are shown in **Figures 2** and **3**, respectively. The group average equivalent dose ($D_{e,av}$) and standard deviation (s) are also shown on the plots. The influence of the number of grains per aliquot on the degree of scatter is shown in scenario I-V displayed down the page in increasing aliquot size, whereas the influence of the value of the previous dose is shown in scenario A-D. Dose distributions from selected scenarios are also presented as weighted histograms in **Fig. 4**.

The first simulation (**Table 1**, **Fig. 2**) shows that, for a sample where all the grains are poorly bleached, inaccuracy of the equivalent dose as a consequence of poor bleaching will only show as scatter if small aliquots are used, or if the previous dose is large compared to the post-depositional dose. When the aliquot consists of 100 or more grains (scenario IV and V), the equivalent dose is largely averaged within each aliquot. In the second simulation, the vast majority of the grains are well bleached, and only 10% of the grains contain a remnant dose. The simulation results (**Table 2**, **Fig. 3**) show that this type of contamination will result in larger scatter between aliquots for scenarios with a similar offset in equivalent dose.

5. VALIDITY OF POOR-BLEACHING DETECTION METHODS

Relative scatter in equivalent dose

Using the relative-scatter criterion ($S_N > 0.10$; Clarke *et al*., 1999), samples from the first simulation (all grains poorly-bleached) are properly classified as poorly bleached when aliquots consist of 10 grains or less (**Table 1** and **Fig. 2**, scenario I-III). However, when aliquots are larger the method fails because grain-to-grain variations in equivalent dose are averaged within the aliquots. When the majority of the grains is well-bleached the method is slightly more successful, being able to detect severe offsets due to poor bleaching even in relatively large aliquots (**Table 2** and **Fig. 3**, scenario IV and V).

Some recent publications have suggested that the absence of scatter in equivalent doses obtained on single aliquots is a guarantee that the sample under study is well bleached (Clarke, 1996; Clarke *et al*., 1999). However, the simulations discussed in the present paper clearly show

Table 1. *Summary of results of poor-bleaching tests for the first simulation (all grains poorly bleached, scatter plots in* ***Fig. 2****). No.=number of grains per aliquot.*

No.	Scen.	Previous dose 5 Gy (Scenario A)			
	Fig.2	Offset D_e [%]	S_N	R^2	Skew
1	I	4.5 ± 0.1	0.03 ± 0.00	0.01 ± 0.01	0.25 ± 0.11
5	II	5.3 ± 0.3	0.02 ± 0.00	0.00 ± 0.01	-0.32 ± 0.12
10	III	4.8 ± 0.1	0.02 ± 0.00	0.02 ± 0.00	-0.05 ± 0.13
100	IV	5.0 ± 0.1	0.01 ± 0.00	0.00 ± 0.00	0.14 ± 0.13
1000	V	5.0 ± 0.0	0.00 ± 0.00	0.00 ± 0.00	-0.01 ± 0.21
		Previous dose 50 Gy (Scenario B)			
1	I	48 ± 2	0.20 ± 0.01	0.01 ± 0.01	0.10 ± 0.12
5	II	51 ± 2	0.15 ± 0.01	0.01 ± 0.02	0.03 ± 0.15
10	III	51 ± 2	0.12 ± 0.00	0.02 ± 0.01	0.00 ± 0.17
100	IV	49 ± 1	0.06 ± 0.00	0.05 ± 0.02	0.08 ± 0.26
1000	V	50 ± 1	0.02 ± 0.00	0.15 ± 0.04	0.22 ± 0.16
		Previous dose 100 Gy (Scenario C)			
1	I	96 ± 5	0.30 ± 0.02	0.01 ± 0.01	0.00 ± 0.19
5	I	103 ± 4	0.23 ± 0.02	0.04 ± 0.02	-0.12 ± 0.16
10	III	98 ± 3	0.19 ± 0.01	0.06 ± 0.03	-0.07 ± 0.09
100	IV	99 ± 2	0.11 ± 0.01	0.12 ± 0.03	-0.02 ± 0.19
1000	V	100 ± 1	0.04 ± 0.00	0.10 ± 0.04	0.14 ± 0.19
		Previous dose 1000 Gy (Scenario D)			
1	I	950 ± 56	0.58 ± 0.04	0.02 ± 0.02	0.14 ± 0.18
5	II	976 ± 25	0.39 ± 0.02	0.08 ± 0.03	0.04 ± 0.11
10	III	971 ± 22	0.36 ± 0.02	0.09 ± 0.03	-0.14 ± 0.14
100	IV	975 ± 13	0.20 ± 0.01	0.09 ± 0.03	-0.15 ± 0.13
1000	V	994 ± 9	0.06 ± 0.00	0.25 ± 0.04	-0.20 ± 0.16

that the scatter observed is largely dependent upon the number of grains that contribute to the luminescence signal of each aliquot. This result accords with that of previous studies. When first proposing the use of scatter for detection of poor-bleaching, Li (1994) already showed that the degree of scatter in equivalent doses obtained for single aliquots is dependent on aliquot size. Moreover, the simulations show that the type of mixture of well bleached and poorly bleached grains affects the scatter; a lesser abundance of contaminating grains results in greater scatter in equivalent doses obtained. Hence, paradoxically, poor-bleaching is more easily detected when it only affects a small percentage of the grains and thereby has a minor influence on the mean equivalent dose.

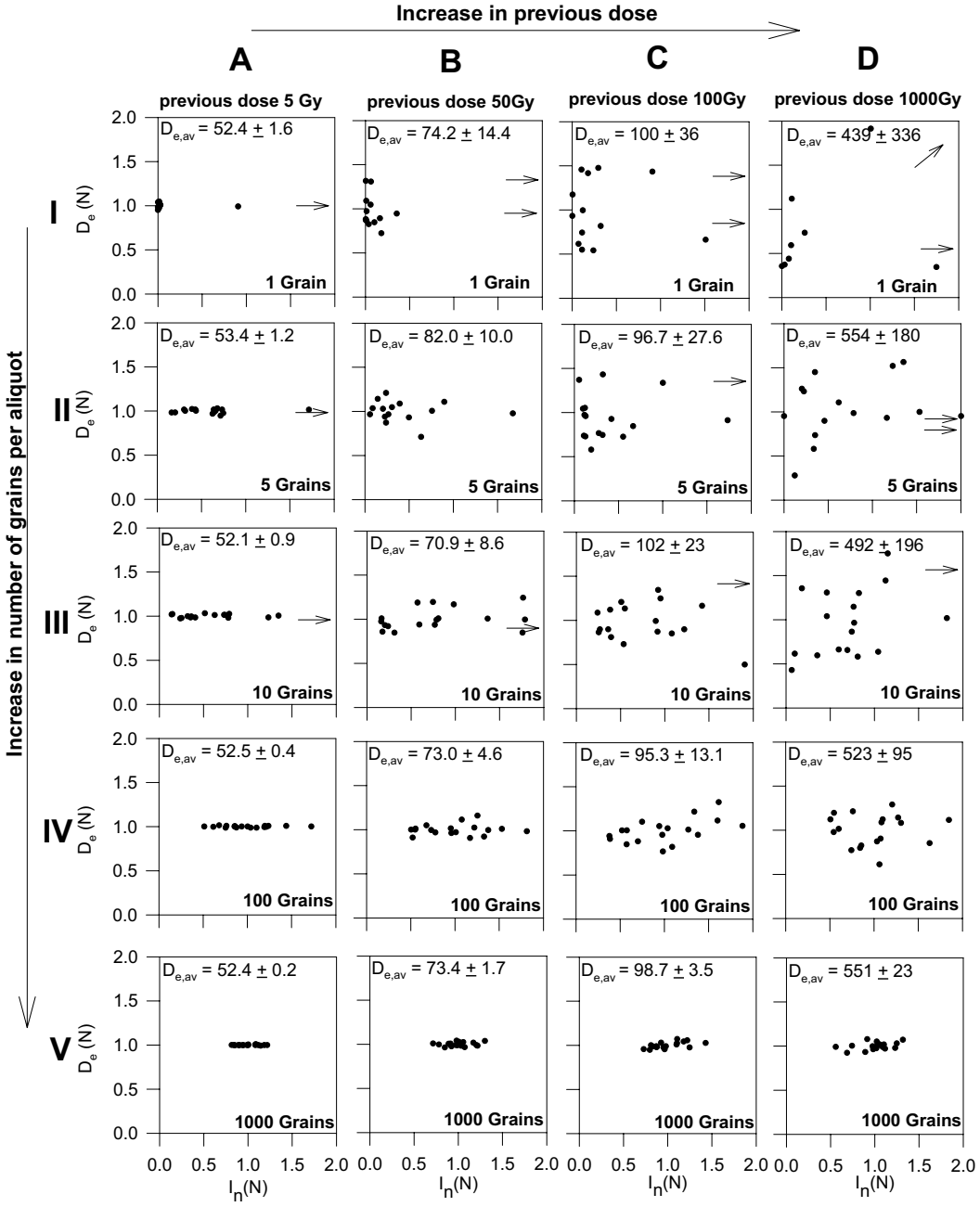

Fig. 2. *Using the model described in the text, equivalent doses and natural intensities are computed for aliquots consisting of grains picked at random from a poorly bleached virtual sample. Each grain is assigned the OSL sensitivity, as well as the equivalent dose, which consists of the post-depositional dose (=50 Gy) plus a random percentage of the previous dose. The previous doses are 5, 10, 100 and 1000 Gy for scenarios A, B, C and D respectively. Both the equivalent dose and the natural intensity obtained on each disc are normalised to the average from 18 discs, to construct normalised scatter plots (Clarke* et al.*, 1999). The mean equivalent dose ($D_{e,av}$) of the 18 discs is presented on each plot, as well as the standard deviation on the mean. Data are presented for aliquots consisting of 1, 5, 10, 100 and 1000 grains in scenarios I to V. For grains that have no luminescence response, an equivalent dose could not be calculated. Therefore, these grains are not plotted on the scatter plots with single-grain data (scenario I). Aliquots for which the normalised intensity or the normalised equivalent dose is beyond the limits of the graph, are indicated by small arrows. Scatter plots shown here represent one out of the ten model runs for each scenario. Average results of the ten runs are presented in* ***Table 1****.*

Correlation of OSL intensity versus equivalent dose

Statistically significant correlation between aliquot natural OSL intensity and equivalent dose ($R^2 > 0.16$, for n = 18; Colls *et al*., 2001) is only found for the simulations where 10% of the grains is very poorly bleached (**Table 2** and **Fig. 3**, scenario D) and for the simulation where all grains are very poorly bleached and aliquots contain 1000 grains (**Table 1** and **Fig. 2**, scenario D-V). The absence of this correlation in the other scenarios is a result of the wide spread in intrinsic OSL sensitivities of grains. A strong correlation is only to be expected when the OSL sensitivities of the grains are similar, or when large aliquots with a small percentage of poorly bleached grains are dated.

Asymmetry of dose distributions

The asymmetry of dose distributions as an indication for poor-bleaching of a sample (Murray *et al*., 1995) is the third method assessed. In this study, the skewness of the distribution is used to characterise the degree of asymme-

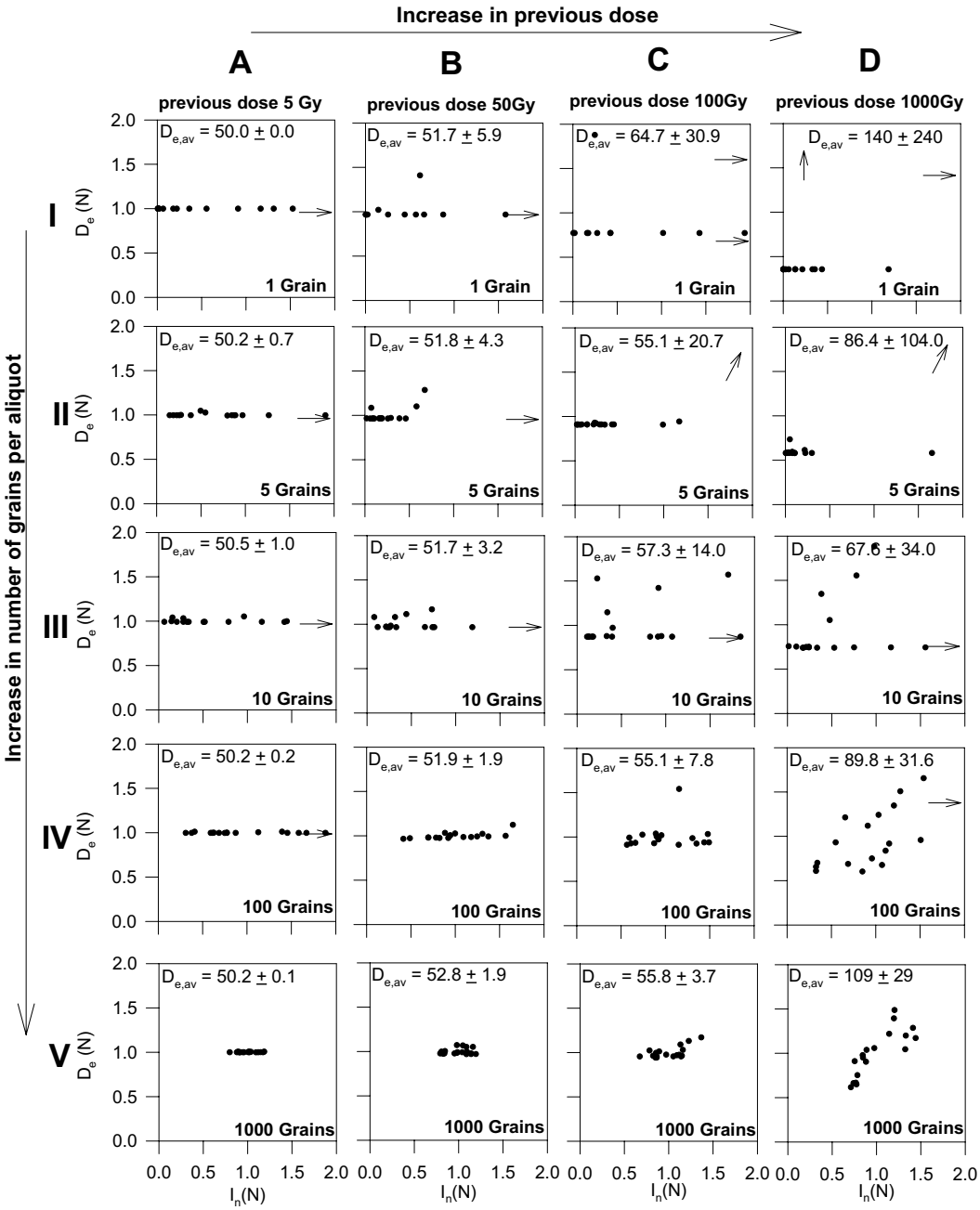

Fig. 3. *A simulation similar to Figure 2, with the difference that 90% of the grains in the virtual sample are well bleached, and thus only contain the OSL signal resulting from the post-depositional dose of 50 Gy. The remaining 10% of the grains is poorly bleached, and assigned a random fraction of the previous dose as in Figure 2. While the grains are picked randomly from the sample, not all aliquots contain exactly 10% of poorly bleached grains. Scatter plots shown here represent one out of the ten model runs for each scenario. Average results of the ten runs are presented in Table 2.*

try of the distribution around its mean; positive skewness indicates a distribution with an asymmetric tail extending towards more positive values. Lepper *et al*. (2000) proposed a different quantification for asymmetry of a dose-distribution

$$\text{Asymmetry} = (D_{e,\max} - D_{e,\text{mode}})/(D_{e,\text{mode}} - D_{e,\min}) \quad (5.1)$$

where $D_{e,\text{mode}}$ is the most frequently occurring equivalent dose). However, this quantification was not applicable for the simulated dataset as the values for $D_{e,\min}$ and $D_{e,\text{mode}}$ were often identical.

The simulations show that the dose distributions are skewed only when the majority of grains in the sample are well bleached (compare results in **Table 1** and **2**, as well as **Fig. 4B** and **4C**). Moreover, it was found that the skewness of the distribution decreases with aliquot size (see **Table 2**). Both findings accord with those made by Olley *et al*. (1999) in their investigation of the use of histograms for detection of poor bleaching. However, where Olley *et al*. (1999) indicate that the dose distribution will not be skewed when 10% or more of the grains is unbleached, such contamination is shown to produce skewed distributions in the present simulations. This difference is caused by the attribution of different OSL sensitivities to the grains in the present simulation, where Olley *et al*. (1999) assumed uniform sensitivity for all grains.

Table 2. *Summary of results of poor-bleaching tests for the second simulation (10% of grains poorly bleached, scatter plots in* ***Fig. 3****). No.=number of grains per aliquot.*

No.	Scen.	Previous dose 5 Gy (Scenario A)			
	Fig. 3	Offset D [%]	S_N	R^2	Skew
1	I	0.50 ± 0.22	0.01 ± 0.00	0.00 ± 0.01	1.9 ± 0.5
5	II	0.55 ± 0.13	0.01 ± 0.00	0.02 ± 0.02	2.6 ± 0.2
10	III	0.43 ± 0.10	0.01 ± 0.00	0.00 ± 0.00	2.3 ± 0.3
100	IV	0.47 ± 0.04	0.01 ± 0.00	0.00 ± 0.00	2.0 ± 0.3
1000	V	0.50 ± 0.03	0.00 ± 0.00	0.00 ± 0.00	0.77 ± 0.15
		Previous dose 50 Gy (Scenario B)			
1	I	4.8 ± 0.9	0.14 ± 0.02	0.00 ± 0.01	3.1 ± 0.4
5	II	5.2 ± 0.8	0.11 ± 0.02	0.00 ± 0.00	2.7 ± 0.2
10	III	4.3 ± 0.9	0.08 ± 0.02	0.01 ± 0.01	2.4 ± 0.3
100	IV	4.5 ± 0.3	0.04 ± 0.01	0.00 ± 0.00	1.5 ± 0.3
1000	V	4.9 ± 0.2	0.02 ± 0.00	0.05 ± 0.02	0.8 ± 0.2
		Previous dose 100 Gy (Scenario C)			
1	I	14.0 ± 3.0	0.34 ± 0.05	0.00 ± 0.00	2.8 ± 0.4
5	II	8.5 ± 1.6	0.21 ± 0.04	0.05 ± 0.04	2.9 ± 0.2
10	III	9.6 ± 2.1	0.18 ± 0.03	0.01 ± 0.02	2.9 ± 0.3
100	IV	10.7 ± 0.7	0.12 ± 0.01	0.09 ± 0.02	2.4 ± 0.2
1000	V	11.0 ± 0.4	0.05 ± 0.00	0.06 ± 0.03	1.03 ± 0.14
		Previous dose 1000 Gy (Scenario D)			
1	I	109 ± 24	1.5 ± 0.2	0.21 ± 0.08	3.1 ± 0.4
5	II	105 ± 18	1.2 ± 0.1	0.23 ± 0.07	2.9 ± 0.2
10	III	93 ± 16	0.87 ± 0.08	0.31 ± 0.09	2.4 ± 0.1
100	IV	107 ± 11	0.52 ± 0.06	0.43 ± 0.10	1.7 ± 0.3
1000	V	97 ± 5	0.23 ± 0.01	0.71 ± 0.03	0.7 ± 0.2

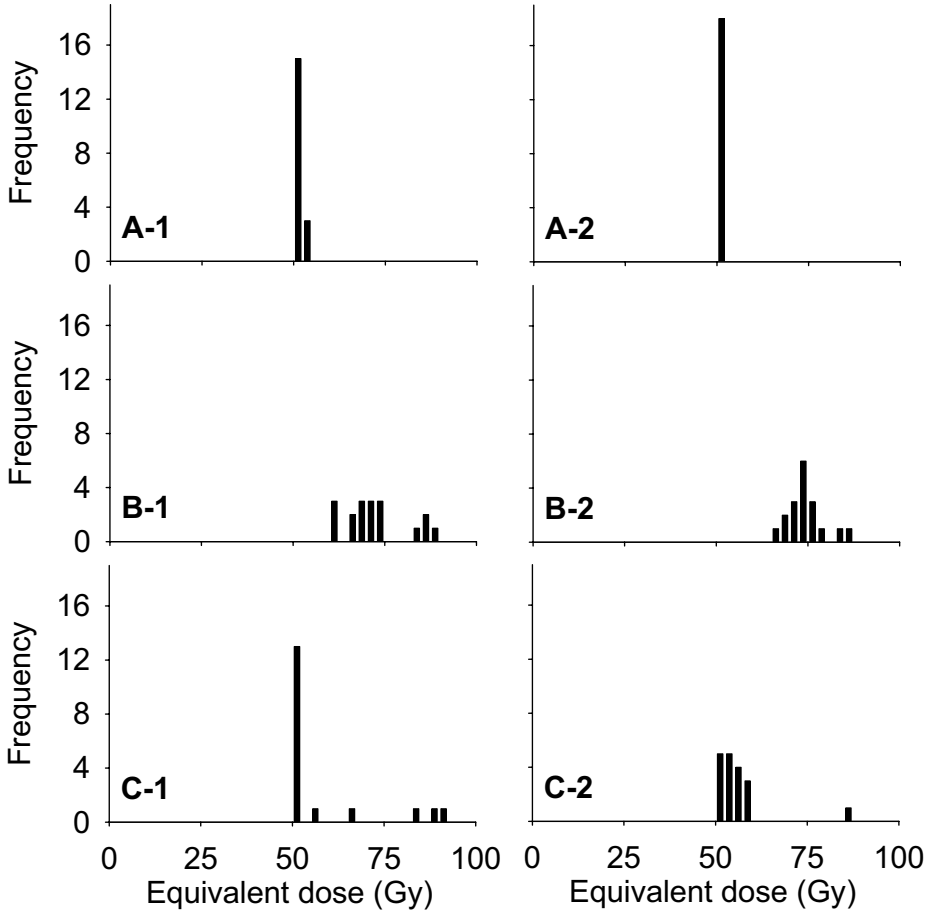

Fig. 4. *Typical equivalent-dose distributions for well bleached (A) and poorly bleached samples (B and C) presented in histogram plots. The dose distributions used are obtained from the different simulation scenarios: A) 10% of the grains poorly bleached with a previous dose of 5 Gy (scenario B in* ***Fig. 3****); B) All grains poorly bleached with a previous dose of 50 Gy (scenario B in* ***Fig. 2****); C) 10% of the grains poorly bleached with a previous dose of 100 Gy (scenario C in* ***Fig. 3****). Eighteen aliquots are used for each histogram; results are shown for aliquots containing 10 (left) and 100 (right) grains.*

6. IMPLICATIONS FOR DETECTION OF POOR BLEACHING

In interpreting the simulation results for suitability of methods for detection of poor bleaching in real samples, limitations of the model have to be kept in mind. In the simulations, the scatter in equivalent doses between aliquots arises purely from grain-to-grain differences in the remnant dose. In real samples there will be added sources for scatter: Firstly, grain-to-grain differences in micro-dosimetry can result in different post-depositional doses for the grains. Although these differences are generally thought to be small (Olley *et al*., 1999) they can be of importance for some samples (Murray and Roberts, 1997; Olley *et al*., 1997). Especially when small-aliquots or single-grains are used for equivalent-dose determination will differences in micro-dosimetry cause added scatter in equivalent doses. Secondly, in real samples there will be scatter arising from OSL-measurement imprecision resulting from photon-counting statistics. This again will be particularly the case when small aliquots or single grains are used. Thirdly, intrusion of grains from other layers can cause scatter in natural samples.

In ignoring the additive sources for scatter discussed above, the model provides a minimum estimate for the scatter in equivalent doses to be found in real samples. In isolating the scatter caused by poor-bleaching, it fits the assumptions made in the poor-bleaching detection methods based on the scatter of equivalent doses resulting from poor-bleaching. Thereby it creates the ideal circumstances for the detection of poor-bleaching by these tests. If the tests are not able to detect such simulated poorly-bleached samples, they cannot be regarded as reliable tests of poor bleaching in real samples.

Previous studies have shown it possible to identify poorly-bleached samples using the scatter in equivalent doses (Li, 1994; Duller, 1994a; 1995; Clarke, 1996; Clarke *et al*., 1999). In view of the results from computer simulations presented in this paper, it is concluded that the samples investigated in those studies must have fit one (or both) of the following two criteria: 1) the luminescence signal from each aliquot was dominated by a small number of bright grains with different equivalent doses; or 2) only a small proportion of the grains was poorly bleached. From the computer simulations it is evident that the absence of scatter in equivalent doses cannot be interpreted as an assurance that the sample was well bleached at the time of deposition.

In applying the correlation method for detecting poor-bleaching to fluvial quartz separates, Stokes *et al*. (2001) found that three out of four samples from the Colorado River (Texas, USA) showed evidence for partial bleaching, whereas for thirteen samples from the Loire River (France) only two were classified as poorly bleached. However, for the Loire River samples, mean equivalent doses of up to 14 Gy were reported for 'well-bleached' samples, whereas a sample from the Colorado River was classified as poorly bleached, in spite of having an equivalent dose of only 0.23 Gy. Colls *et al*. (2001) also worked on Loire samples and found that from 18 samples investigated, five should be classed as poorly bleached. Three of these were characterised by a single outlier (aliquot D_e > 3 SD from the mean) and the remaining two samples showed extraordinary wide scatter in equivalent doses. Folz *et al*. (2001) used a similar method to check for poor-bleaching in another suite of fluvial quartz from France. Although OSL ages overestimated the known age by 40%, no correlation was found between natural aliquot intensity and equivalent dose.

In expecting a correlation between equivalent dose and natural aliquot OSL signal for poorly bleached samples, this method assumes that the grains in the sample have similar OSL sensitivities. This assumption is clearly not valid (**Fig. 1**) and it is therefore not surprising that the method fails to identify poor-bleaching for simulated (**Table 1, 2**) and real samples (Stokes *et al*., 2001). For the natural samples where the method indicated poor-bleaching, results also showed a wide spread in equivalent doses or the existence of a single outlier (Colls *et al*., 2001; Stokes *et al*., 2001), which are probably better indications for poor-bleaching.

Histograms have successfully been used as a means to visualise the width and the skewness of dose distributions and to detect poorly bleached samples using small aliquots (Murray *et al*., 1995; Olley *et al*., 1998 and 1999; Lepper *et al*., 2000) and single grains (Roberts *et al*., 2000) of quartz. Primary limitation of this method is that the distribution will only be skewed if the majority of the grains in the sample are well bleached (compare **Tables 1** and **2**). Olley *et al*. (1999), have already pointed this out in their investigation of the use of this method. Murray and Roberts (1997) found wide, but symmetrical dose distributions for single-grains from supposedly well-bleached, aeolian quartz from Australia. The large standard deviation (23% of the mean) was attributed to differences in micro-dosimetry. Lepper *et al*. (2000) discuss ways to deconvolve the spread in equivalent doses arising from experimental errors and that from sedimentary processes. They propose that the correct equivalent dose can be obtained from an asymmetrical dose distribution after deconvolution.

The dependency of the degree of scatter on the aliquot size and on the percentage of poorly bleached grains, makes the use of thresholds for detecting poor-bleaching based on inter-aliquot scatter impossible. Although it is tempting to suggest such thresholds (Clarke, 1996; Clarke *et al*., 1999; Colls *et al*., 2001), they are not generally applicable, and hence do not provide an objective criterion for detecting poor bleaching. To improve the chances of detecting poor-bleaching using methods based on inter-aliquot scatter in equivalent doses, the aliquot size should be taken as small as possible, ultimately consisting of a single grain of sand. One should however keep in mind that scatter in equivalent doses can also results from other sources than poor-bleaching, for example differences in micro-dosimetry. Correlation of aliquot equivalent dose and natural OSL intensity is the only indicator that performs better for large aliquots; however, it only detects poor bleaching successfully when poor-bleaching results in a severe error in the equivalent dose.

A method for visualising scatter in equivalent doses in relation to its relative error is provided by radial plots (Galbraith, 1990). This type of plot has recently been used in studies using small aliquots (Olley *et al*., 1999) and single-grains (Roberts *et al*., 1998; 2000) of quartz. Although a promising method, the validity of radial plots for detecting poor bleaching is not assessed in this paper because the simulations do not allow calculation of an error on individual equivalent-dose determinations.

7. CONCLUSIONS

Scatter in equivalent doses obtained by single-aliquot OSL dating methods may indicate that the luminescence signal was not completely zeroed in all grains prior to deposition. However, the degree of scatter is largely dependent on the number of grains in each aliquot that contribute to the luminescence signal, the percentage of poorly bleached grains, and the remnant dose in the poorly bleached grains. For aliquots consisting of a number of grains, computer simulations indicate that:

1) The absence of scatter in equivalent doses obtained cannot be taken as assurance that all grains were well bleached prior to deposition.
2) A correlation of natural aliquot luminescence intensity versus equivalent dose should only be expected for poorly bleached samples when the luminescence sensitivity of the grains is relatively uniform. Therefore the absence of such a correlation is not an indication that the sample was well bleached.
3) Skewed dose distributions are an indication that the sample contains a small percentage of poorly bleached grains. A non-skewed distribution can indicate that the sample is well bleached or that a relatively large percentage of grains from the sample are poorly bleached.

The best method to check whether the equivalent dose of a sample might be overestimated as a consequence of poor-bleaching is to use small aliquots (ultimately consisting of a single grain), and to look at both the spread in the equivalent doses and the symmetry of the dose distribution. Tight, symmetrical dose distributions on very small aliquots indicate that the sample is homogeneously bleached, but even this does not necessarily mean that its OSL signal was completely reset during the last transportation event.

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