How many grains are there on a single aliquot?

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Introduction

In our recent OSL studies on modelling equivalent doses (D_e) obtained using single aliquots the question arose how many grains are present on a single aliquot and consequently, how many grains on a single aliquot contribute to the measured signal?

To date, the usual practice reported in literature has been to calculate the maximum number of grains predicted on the basis of dense packing of circles on a plane assuming an average diameter of the hypothetically spherical grains (e.g. Rhodes 2007, Duller 2008, Arnold and Roberts 2009). When grains are attached to the disc surface through a medium like silicone oil sprayed on a disc through a circular mask, the number of grains is estimated from the size of the total area covered by grains and the surface area of the cross section of the spherical grain. So for example a 4 mm diameter aliquot of 90-125 µm diameter spherical grains would consist of a maximum number of grains (n):

$$n = \frac{\pi \cdot 2^2}{\pi \cdot 0.0538^2} \cdot \frac{\pi}{\sqrt{12}} \approx 1253 \text{ grains}$$

calculated for the middle of the diameter range, where $\frac{\pi}{\sqrt{12}}$ is the densest packing factor of a plane by circles of equal diameters in hexagonal arrangement, first given by Lagrange. A 6 mm diameter aliquot of 200-250 µm diameter grains would contain a maximum of :

$$n = \frac{\pi \cdot 3^2}{\pi \cdot 0.113^2} \cdot \frac{\pi}{\sqrt{12}} \approx 639 \text{ grains}$$

In general, grains are not spherical, their size varies, and the packing is unlikely to be perfect. These factors influence the number of grains present on a disc. In this communication we report our investigations of the number of grains present on a single aliquot of sedimentary quartz.

Experimental

The number of grains on an aliquot was determined for aliquots prepared using the standard method applied in the Bern OSL laboratory.

The discs are placed in a holder and after covering the holder with a mask with circular holes located at the centres of discs, the discs are sprayed with silicone oil. The oil circles have the desired diameter of the aliquot. Subsequently, the grains are placed on a sheet of paper and the discs held by tweezers are pressed on the grains with the side with the attached silicone oil. After that, the discs are tapped on their sides to remove the excess of grains to ensure that grains form a monolayer.

In the current study we used a mask with 6 mm diameter holes. In order to count the number of grains, we took photos of the aliquots using a Leica M 205C binocular microscope equipped with a digital camera Leica DFC 280. The photos were taken at a magnification of 12.5x, printed on A4 format and manually counted.

The quartz studied originated from NW alpine postglacial sediments from the profile Mattenhof located at Wauwilermoos on the Swiss Plateau. The 200-250 µm diameter grains were from the sample WAU-MH3 and grains 150-200 µm in diameter were from the sample WAU-MH2.

Results

The photographs (examples are shown in Figure 1) reveal that the quartz grains in these samples are only slightly rounded, mainly sharp-edged and the grains are of cubic, cuboid or elongated, thin and point-tapered shapes (Figure 1a). The heterogeneity of shapes may be the result of the rather short sedimentary history and limited mechanical abrasion by the glacier. It appears that within the 6 mm circle there is considerable space free of grains, contrary to the usual assumption of dense coverage by grains (Figure 1b).

Table 1: *Results of counting grains on single aliquot discs.*

Grain size	Aliquot diameter (mm)	Number of counted aliquots	Calculated[1)] maximum number of grains	Counted average number of grains	Standard deviation	Packing ratio (%)
150-200 µm	6	46	1073	677	48	63
200-250 µm		52	634	440	46	65

[1)] a monolayer of spherical grains with diameters of 175 and 225 µm is assumed

Table 2: *Estimation of the number of grains for 100-150 µm, 150-200 µm and 200-250 µm diameter grains.*

Grain size	Aliquot diameter (mm)	Estimated average number of grains	Assumed packing ratio (%)
100-150 µm	6	1406	
	2	156	63
150-200 µm	2	75	
200-250 µm	2	46	65

(a) (b)

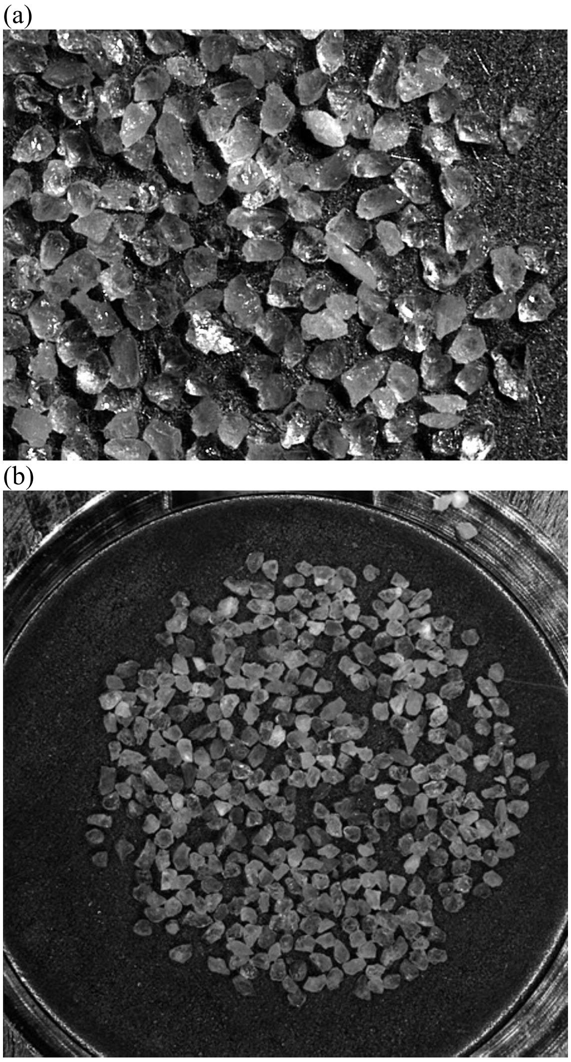

Figure 1: *(a) 200-250 µm grains of sample WAU-MH3 on an aliquot, (b) A view of the whole disc.*

Table 1 summarises the results of counting over 50 thousand grains attached to 6 mm aliquots. The table presents the average number of grains from diameter ranges of 150-200 µm and 200-250 µm calculated assuming densest packing, the average counted numbers of grains, their standard deviation and the ratio of those two values (called here the 'packing ratio').

We found that a 6 mm aliquot of 150-200 µm grains contains on average 677±48 grains and a 6 mm aliquot of 200-250 µm contains 440±46 grains. As shown in the last column of Table 1 the actual number of grains on an aliquot is lower than that predicted by dense packing by at least 35%.

Implications

Based on the results obtained from counting grains, we estimated the numbers of grains for the grain size 100-150 µm on 6 and 2 mm aliquots, and for 150-200 µm and 200-250 µm diameter grains on 2 mm aliquots. These estimates are listed in Table 2. In the case of the smaller grains, we assumed the same packing ratio as in the case of 150-200 µm, whereas for the two other ranges we used the packing ratio given in Table 1.

The experimentally determined packing ratios (Table 1) and the proportion of bright and dim grains (further jointly termed active grains) in the quartz extracts (for details and definition of "bright" and "dim" see Appendices A and B) makes it possible to estimate the probability of a given number of active grains being present on an aliquot.

The number of active grains present on an aliquot follows a binomial distribution. The probability of getting exactly k active grains on a disc (k successes) containing n grains (n trials) with a given proportion p of such grains in the total (probability p) equals:

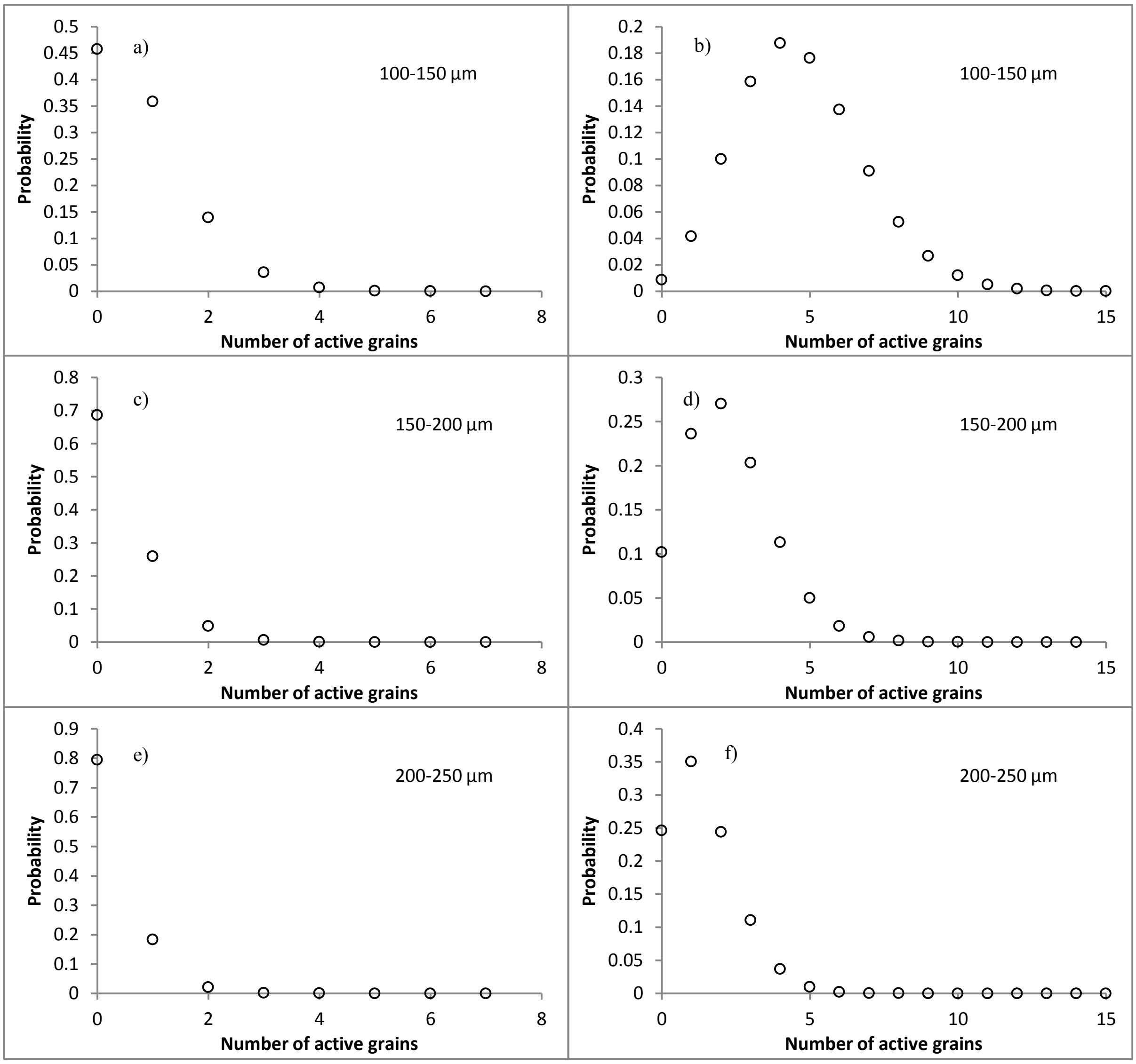

Figure 2: *The probability of a given number of active grains on 2 mm aliquots of quartz; a), c) and e) probabilities for bright grains; b), d) and f) for dim grains.*

$$P_k = \binom{n}{k} p^k (1-p)^{n-k} \quad (1)$$

Single grain measurements (see Appendix A for details) revealed that samples investigated in this study hardly contain any grains emitting light. For the calculations we assumed that only 0.5% of the grains are bright, and ~3% of the grains are dim, as defined in Appendix A. Figures 2 and 3 present the probabilities of encountering a given number of bright and dim grains on 2 mm and 6 mm aliquots, respectively. For example, 80% (probability of 0.8) of 2 mm aliquots containing 200-250 µm grains (Figure 2e) will not contain any bright grains and 25% of such aliquots will not contain any grains emitting light at all (Figure 2f). Consequently, well over 25% of such aliquots will not emit any measurable signal, however we do not have any measurements with 250 µm 2 mm aliquots.

In the case of 6 mm aliquots, 11% will not contain any bright grains, 24% one bright grain, 27% two bright grains, 38% more than two bright grains (Figure 3e). In addition, such aliquots will contain between 5 and 25 dim grains (Figure 3f).

Taking into account the considerations presented in Appendix B it is possible to state that the signal of one 6 mm aliquot (of Alpine quartz) consists of the signal originating from a few tens of dim grains and some bright grains, however, there will be a few aliquots which will be dominated by the signal of single bright grains. Different conclusions may be

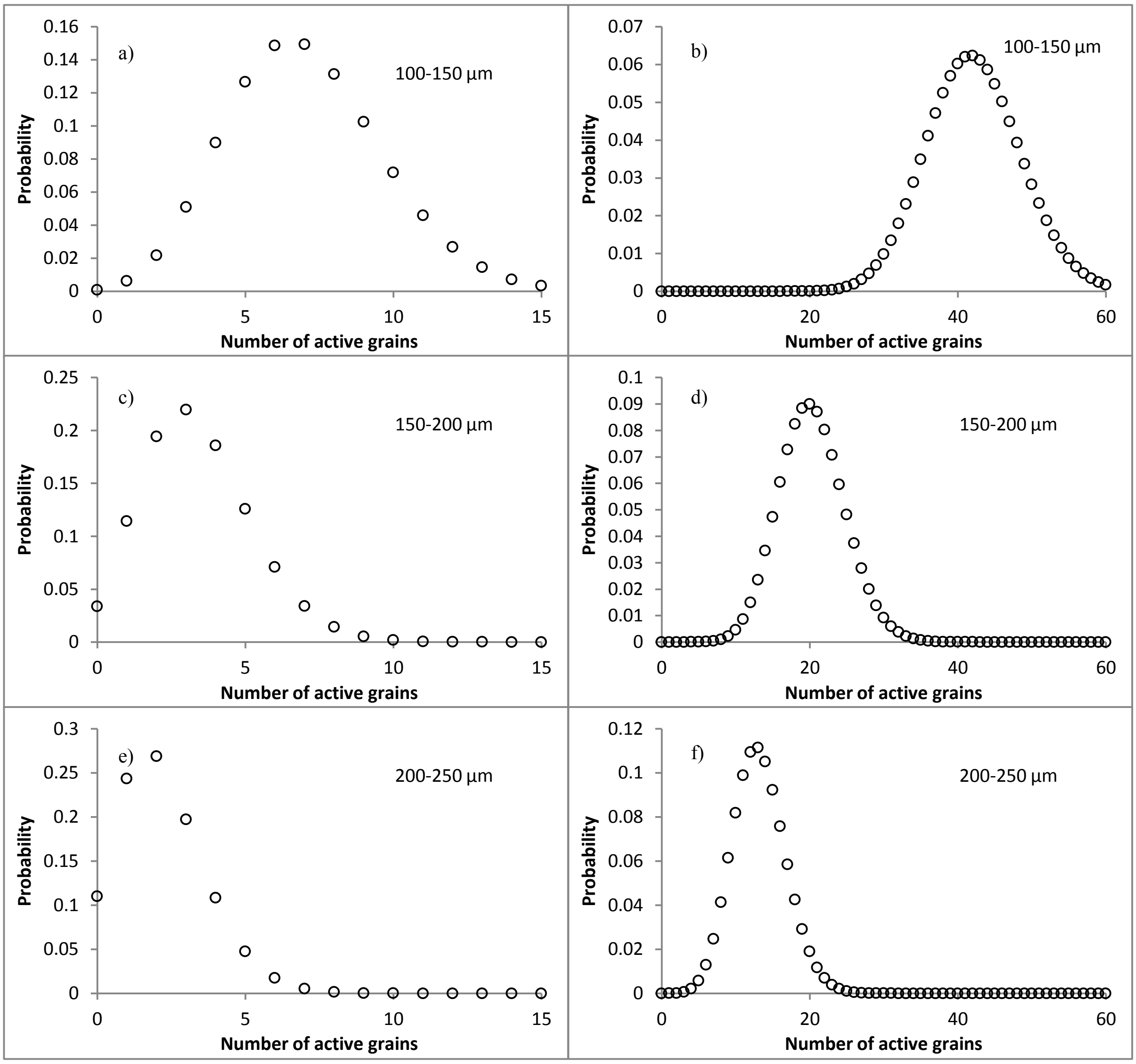

Figure 3: *The probability of a given number of active grains on 6 mm aliquots of quartz; a), c) and e) probabilities for bright grains; b), d) and f) for dim grains*

reached for another sample characterised by a different, experimentally determined, proportion of bright and dim quartz grains.

Conclusions

In this investigation we found that a significant discrepancy existed between the theoretically calculated number of grains and the actual number of grains. We suspect that there are several factors that may affect the magnitude of such discrepancy. These factors include the technique of aliquot preparation, the person who prepares the aliquots and the size and the shape of the grains, and hence the type of sediment, length of transport and duration of grain working.

We suggest that the knowledge of the packing ratio and the proportion of active grains in a given sample are very important in the choice of the used aliquot size to optimise the proportion of active grains on a single aliquot and the counting statistics.

For a sample the proportion of active grains and the packing ratio might be important for explaining the discrepancy between single grain and single aliquot results that is sometimes observed, especially when D_e modelling is used. It would be interesting for other laboratories to perform similar checks to explore the degree of variability of the packing ratio between laboratories and between quartz of varying provenance.

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Appendix A

Number of luminescent grains in quartz extract

In order to determine the number of luminescent quartz grains in the used extracts, the natural (L_N) and the regenerated (L_1) CW-OSL signals were measured for up to 500 single grains per sample.

The measurements were performed on automated TL/OSL-DA-20 Risø readers including a $^{90}Sr/^{90}Y$ beta source and an EMI 9235 PM tube. Optical stimulation was carried out using a 10mW Nd: YVO_4 solid state diode-pumped laser emitting at 532 nm (green light). The sample was preheated for 10 s at 220°C and read out at 125°C for 1s and recorded in 0.02 s time intervals. The OSL detection was performed through a Hoya U340 filter. Before measurement, each aliquot was tested for feldspar contamination by applying an IR-shine and eliminated if any response was observed.

Figure A1.a shows the net OSL signal curves, visualising the proportion of grains emitting more than 1ct/0.02s/Gy of OSL averaged over the first 0.06s of the decay, as described by Duller (2008). The signal level of 1ct/0.02s/Gy is thought to be acceptable when calculating single grain growth curves (Prof. G. Duller, personal communication). In the case of the investigated samples only 0.5-1% of grains gave a signal higher than this threshold (Figure A1.a). Figure A1.b, on the other hand, shows cumulative light sum curves as described by Duller et al. (2000). They indicate that apart from bright grains (ca. 60% of the luminescence), a significant part of the signal on a single aliquot originates from dim grains and some not further explored "noise" (ca. 40%) in the investigated samples. This observation differs from the frequently made assumption that mainly bright grains contribute to the measured signal. The data provided here indicate that often the signal gained from an aliquot is the sum of the signal of a few tens of grains, at least in the investigated Alpine samples. The situation will change in samples where some extremely bright grains occur. In such cases the total single aliquot signal may be dominated by the signal of such bright grains.

For the purpose of this study the threshold value dividing grains into bright and dim ones was chosen to be 80 cts in the first 0.02s channel of the OSL decay in response to a beta dose of 55 Gy – this value was selected on the basis of the considerations described in Appendix B. The brightest grains emitted a maximum of about 400 cts in the first 0.02 s channel of the OSL decay. Figures A1.c and A1.d show examples of OSL decay curves obtained from a dim and a bright grain, respectively.

Appendix B

Estimation of the magnitude of SG signal that can be distinguished from the background of a single multigrain aliquot

In order to estimate the magnitude of a single grain (SG) signal, measured under green light laser stimulation, necessary for the signal to be distinguishable from the background level in single aliquot (SA) measurement, using blue stimulation, we applied the following considerations.

For the ease of computation we assume that the SA signal (initial channels of the OSL decay curve) consists of the fast component of a SG and a background, which may consist of slow components and the PM tube background. The medium component is neglected. In addition, as this is a rough estimation, we ignore k_{corr}, described in Adamiec et al. (in press) and assume a Poisson distribution of the count numbers.

If the measured CW OSL of a single aliquot (SA) is S (understood as counts summed over time t_S) and the background to be subtracted is B (understood as background estimated on the basis of the last integral) then the OSL signal sum from the grain(s) is $I=S-B$. The uncertainty (u) of this signal is

$$u(I) = \sqrt{u^2(S) + u^2(B)} \tag{1.1}$$

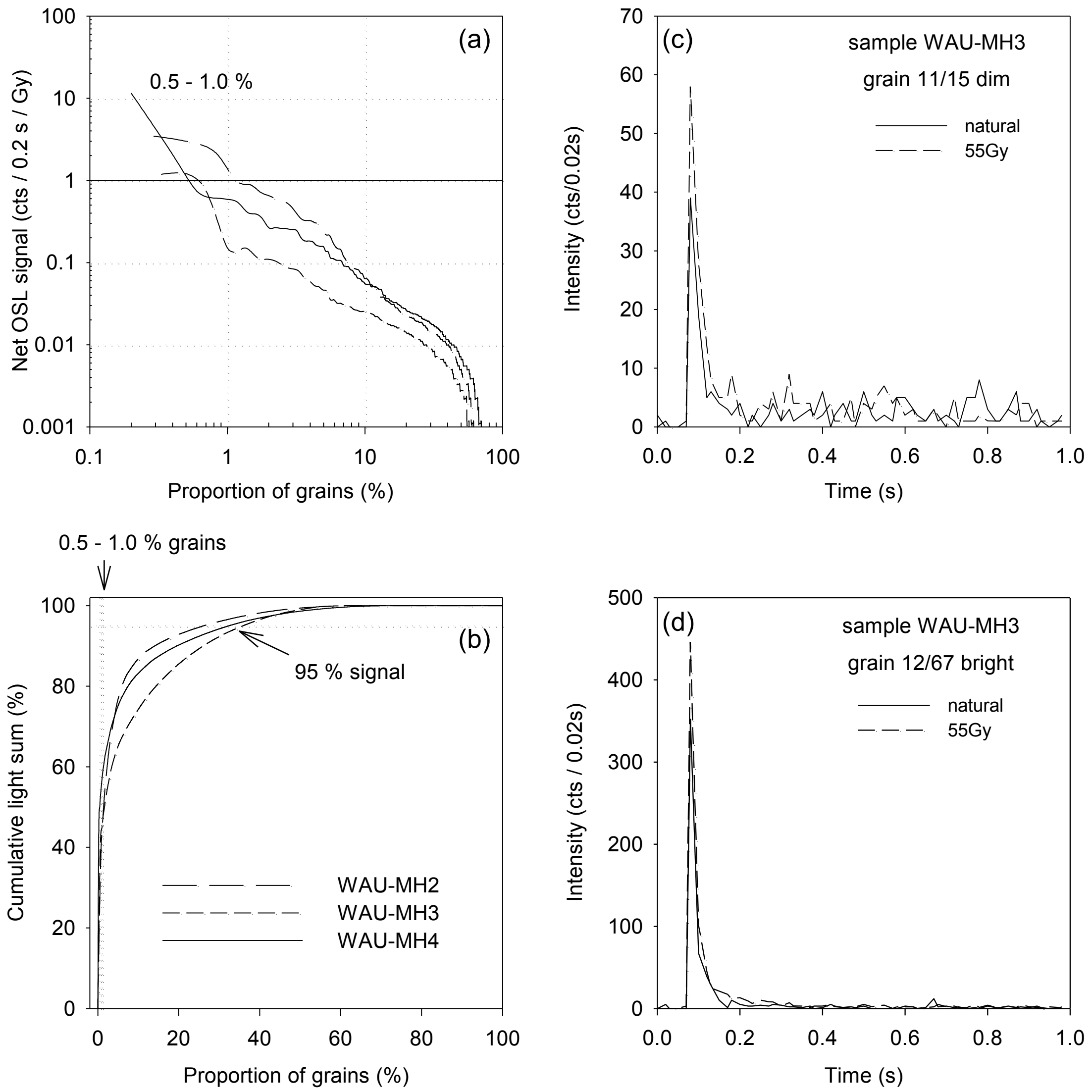

Figure A1: *Determination of the number and the quality of luminescent grains in the Wauwilermoos samples: a) net OSL curves (as defined in Duller 2008); b) cumulative light sum curves (as defined in Duller et al. (2000)) for samples WAU-MH2, WAU-MH3 and WAU-MH4; c and d) examples of OSL decay curves of the natural (solid line) and regenerated (dashed line) single grain OSL for a dim (c) and bright (d) quartz grains from sample WAU-MH3.*

If we assume that the count numbers are distributed according to the Poisson distribution this becomes

$$u(I) = \sqrt{S + B \cdot c} \quad (1.2)$$

where $c = \frac{t_S}{t_B}$ and t_S and t_B are the time of signal integration and background integration, respectively.

In order for the signal I to be distinguishable from the background the OSL signal should exceed its standard deviation and thus the following condition must be fulfilled

$$I > k \cdot u(I),$$

where k would be usually selected to be 2 or 3.

The OSL measured is S and it is numerically equal to $B+I$, where I is the signal originating from a grain assuming that the SA signal consists of background and OSL emitted by one grain. If we combine the above equations we obtain

$$I > k\sqrt{B(1+c)+I}$$

This gives us the quadratic condition

$$I^2 - k^2 I - k^2(1+c)B > 0$$

Table B.1: *A summary of estimated I and signal necessary to distinguish a single grain's signal superimposed on the background of a multigrain aliquot for different values of parameters*

Background rate (s^{-1})	1500	1500	80	80	1500	1500	80	80
k	3	3	3	3	2	2	2	2
t_S	0.2	1	0.2	1	0.2	1	0.2	1
t_B	10	10	10	10	10	10	10	10
c	0.02	0.1	0.02	0.1	0.02	0.1	0.02	0.1
B	300	1500	16	80	300	1500	16	80
I	**57**	**126**	**17**	**33**	**37**	**83**	**10**	**21**
S = I+B	**357**	**1626**	**33**	**113**	**337**	**1583**	**26**	**101**

Solving this and rejecting the negative value finally gives the condition for the signal of a single grain to be distinguishable from the background of a SA

$$I > \frac{k^2 + \sqrt{k^4 + 4k^2(1+c)B}}{2} \quad (1.3)$$

Table B.1 summarises a few examples of calculated values of count numbers originating from grains in order to make them distinguishable from the background. For example, if we take into account the first second of the OSL decay curve, the background is estimated using the last 10 s of the decay curve we have c=1/10=0.1, *B* for 1 s equals 1500 (such a value was measured for sample WAU-MH3 due to a large slow component contribution). Choosing *k*=3 gives *I*=126 counts and *S*=1626 counts, while for *k*=2 *I*=83 and *S*=1583.

In order to compare this with the signal of a single grain under green laser stimulation we need to take into account the power of stimulation and the difference in the cross section for interaction of trapped electrons due to the different wavelengths. The typical power of blue light stimulating diodes (470 nm, photon energy 2.53 eV) is of the order of 50 mW/cm^2, while the power of the stimulating laser in single grain measurements (green light, 532 nm, photon energy 2.23 eV) is about 50 W/cm^2, giving a photon flux ca. 1000 times higher in the case of the laser stimulation.

The cross section for interaction with 470 nm photons is equal to $\sigma_{fast}=2.25 \times 10^{-17}$ cm^2 (calculated using the formulae given in Bailey et al., 2011; see Fig. 1 therein; more precise values of parameters needed to calculate the cross sections were supplied by Dr. R. Bailey, pers. comm.) and hence the decay time constant for blue light for a power of 50 mW/cm^{-2} is equal to 0.38 s which means that for a single aliquot measurement practically the whole fast component is emitted in the first second of decay. On the other hand, in the case of SG measurements using green light, $\sigma_{fast}=3.48 \times 10^{-18}$ cm^2 and the decay time constant is 2.2 ms. Therefore the whole of the fast component will be emitted during the first 0.007 s (the decay in case of SG green light measurements is ca. 170 times faster than in the case of SA blue light measurements). This means that the whole of the fast component is measured practically in the first channel only, as typically in single grain measurements channels of length 0.02 s are used. On this occasion it is worth noting that similar considerations made for the medium component return the decay time constant of 2.03 s for SA measurements and 0.025 s for SG measurements which means that the medium component will be practically emitted within the first 0.08 s in SG measurements.

All this information combined, under the assumption (for ease of computation) that the background in SG measurements gives negligible contribution, as it is often observed, leads to the conclusion that if one single grain of the dim Alpine quartz were to be seen above the high background of a single aliquot on a 2 σ level it would have to show more than about 80 counts (6^{th} column in Table B.1) in the first channel of the SG measurement. In the case of the 6 mm aliquots with a background equal to the machine background (let's assume 80 counts per second), this would amount to 21 counts for the single grain to be distinguishable (in Alpine samples there is always a high slow component contribution so this is a hypothetical situation).

These back-of-the-envelope calculations are a rough estimate and in order to check them one would have to select a bright grain and after irradiation place it on an aliquot to see whether this holds true.

We decided to classify the grains using the limit of 80 counts in the first channel of the SG OSL decay curve qualifying grains brighter than this limit as ‘bright’ grains while grains emitting a lower signal, though with a detectable OSL decay, as ‘dim’ grains.

Reviewer
K.J. Thomsen