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Abstract

The establishment of standardised growth curve (SGC) for equivalent dose (D_e) determination can substantially reduce the amount of instrument time required for OSL measurements. In this study, we investigated the applicability of SGC for the optically stimulated luminescence (OSL) signal from single grains and small aliquots of sedimentary quartz from Haua Fteah cave, in Libya. The samples exhibit large inter-grain and inter-aliquot variation in the shape of their single grain and small aliquot dose response curves (DRCs) constructed from a range of sensitivity-corrected regenerative dose signals (L_x/T_x), which prevents the establishment of a single common SGC among different grains or small aliquots. Instead, the DRCs for the small aliquots can be divided into a minimum of three groups using the Finite Mixture Model, with the DRC saturating at a different dose level for each group. In order to establish a common DRC, or SGC, for each group, we propose a new normalisation method, the so-called least-squares normalisation procedure ('LS-normalisation'), which largely reduces the variation between aliquots within the same group and allows the establishment of a common DRC, or SGC, for each group. In order to apply the SGC method for these samples, two regenerative dose points are needed for each aliquot to attribute it to one of the groups based on the ratio of the L_x/T_x signals for two sensitivity-corrected regenerative dose points. Equivalent dose (D_e) values for each aliquot can be estimated using the fitting parameters calculated for the SGC of the relevant group to which it belongs, together with measurements of the natural signal (L_n), one regenerative dose signal (L_{x1}) and their corresponding test dose signals (T_n , T_{x1}). For the samples investigated from Haua Fteah, we found that D_e estimates obtained from the SGCs are consistent with those obtained using a full SAR procedure. Our results suggest that small single-aliquot and single-grain D_e values obtained from application of the SGC may be underestimated if there is a significant proportion of early-saturating grains present in the sample; such grains or aliquots are mostly rejected due to saturation when analysed using the full SAR procedure. In this case, it is necessary to calculate the D_e values based only on those grains or aliquots that have relatively high saturation levels.

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Investigation of the applicability of standardised growth curves for OSL dating of quartz from Haua Fteah cave, Libya

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Abstract

The establishment of standardised growth curve (SGC) for equivalent dose (D_e) determination can substantially reduce the amount of instrument time required for OSL measurements. In this study, we investigated the applicability of SGC for the optically stimulated luminescence (OSL) signal from single grains and small aliquots of sedimentary quartz from Haua Fteah cave, in Libya. The samples exhibit large inter-grain and inter-aliquot variation in the shape of their single grain and small aliquot dose response curves (DRCs) constructed from a range of sensitivity-corrected regenerative dose signals (L_x/T_x), which prevents the establishment of a single common SGC among different grains or small aliquots. Instead, the DRCs for the small aliquots can be divided into a minimum of three groups using the Finite Mixture Model, with the DRC saturating at a different dose level for each group. In order to establish a common DRC, or SGC, for each group, we propose a new normalisation method, the so-called least-squares normalisation procedure ('LS-normalisation'), which largely reduces the variation between aliquots within the same group and allows the establishment of a common DRC, or SGC, for each group. In order to apply the SGC method for these samples, two regenerative dose points are needed for each aliquot to attribute it to one of the groups based on the ratio of the L_x/T_x signals for two sensitivity-corrected regenerative dose points. Equivalent dose (D_e) values for each aliquot can be estimated using the fitting parameters calculated for the SGC of the relevant group to which it belongs, together with measurements of the natural signal (L_n), one regenerative dose signal (L_{x1}) and their corresponding test dose signals (T_n , T_{x1}). For the samples investigated from Haua Fteah, we found that D_e estimates obtained from the SGCs are consistent with those obtained using a full SAR procedure. Our results suggest that small single-aliquot and single-grain D_e values obtained from application of the SGC may be underestimated if there is a significant proportion of early-saturating grains present in the sample; such grains or aliquots are mostly rejected due to saturation when analysed using the full SAR procedure. In this case, it is necessary to calculate the D_e values based only on those grains or aliquots that have relatively high saturation levels.

Keywords: quartz, optically stimulated luminescence, small aliquots, standardised growth curves, least-squares normalisation

1. Introduction

Optical dating provides an estimate of the time since grains of quartz or feldspar were last exposed to sunlight (Huntley et al., 1985; Aitken, 1998; Jacobs and Roberts, 2007; Roberts et al., 2015). The burial age is estimated by dividing the equivalent dose (D_e , a measure of the radiation energy absorbed by grains during their period of burial) by the environmental dose rate (the rate of supply of ionising radiation to the grains over the same period). One of the most widely adopted procedures for measuring equivalent dose (D_e) values from quartz OSL signals is the single-aliquot regenerative dose (SAR) procedure (Galbraith et al., 1999; Murray and Wintle, 2000). In the SAR procedure, after the measurement of the natural signal, a series of regenerative doses, usually including a zero dose and one or more repeat doses, are given to the same aliquot or grain to construct a dose response curve (DRC). A test dose is applied and measured (T_n , T_x) following measurement of the natural (L_n) and each regenerative dose (L_x) signals to monitor sensitivity change induced by different laboratory treatments, including irradiation, preheating and optical stimulation. The D_e is determined from the sensitivity-corrected regenerative dose (L_x/T_x) and natural (L_n/T_n) signals. A range of regenerative doses bracketing the natural dose must be given so that the determination of D_e can be achieved by projecting of the natural signal onto the DRC and interpolating onto the dose axis.

Apart from monitoring and correcting for sensitivity changes that may have occurred during the SAR procedure, it has been suggested that the test dose response (T_x) can also be used as an inter-aliquot normalisation step to construct a standardised growth curve (SGC) that allows direct comparisons between different aliquots from the same or different samples (Roberts and Duller, 2004). The establishment of a reliable SGC allows D_e values to be obtained that are consistent with

those determined using the full SAR procedure, and based solely on measurements of L_n and T_n . The establishment of a SGC for D_e determination can substantially reduce the amount of instrument time required for OSL measurements and, hence, is especially useful for measuring a large number of aliquots for samples from a single site or region; the time saving is potentially greatest for older samples with high D_e values, because the SGC avoids the need to use large regenerative doses to reconstruct the DRC of every aliquot, as is required with the full SAR procedure. A number of studies have subsequently tested the SGC method and applied the procedure successfully to quartz OSL for samples from different regions (e.g., Burbidge et al., 2006, Lai, 2006; Lai et al., 2007; Telfer et al., 2008; Long et al., 2010; Shen and Mauz, 2011).

More recently, the SGC method was further improved by Li et al. (2015), who suggested that normalisation using one of the regenerative doses, the so-called regenerative normalisation (re-normalisation) method, can further reduce the inter-aliquot variation of DRCs for quartz OSL for the same or different samples. They tested this method using samples from around the world with a variety of geological provenances, environmental settings and depositional ages and found that single-aliquot quartz OSL from most samples have a similar DRC shape up to ~250 Gy.

All previous studies on SGC have been based on aliquots each consisting of a few hundred to a few thousand quartz grains. Any between-grain differences in luminescence behaviour, such as large variations in DRC shape and D_0 values (e.g., Roberts et al., 1999; Duller, 2000; Jacobs et al., 2003, 2013), are therefore expected to be averaged to some extent. In the present study, we examine whether the SGC method can be extended to small multi-grain aliquots (~8–15 grains per aliquot), which approximates one luminescent grain per aliquot using samples from Haua Fteah in Libya.

2. Sample description

Six sediment samples (HF1, HF3, HF4, HF8, HF10 and HF11) collected from Haua Fteah were chosen for our study. These samples have been dated by Douka et al. (2014), with ages ranging from ~18 to ~70 ka and D_e values of between 25 and 130 Gy. Details about the samples, preparation

procedures and analytical facilities are given in Douka et al. (2014). Since very few sand-sized grains of quartz were present in these samples, Douka et al. (2014) used several different grain sizes for D_e determination. They conducted single-grain measurements on grains of 90–125 μm in diameter using customised single-grain discs, where each hole was 100 μm in diameter and 100 μm deep, and on grains of 125–212 μm in diameter using standard single-grain discs (each hole 300 μm in diameter and 300 μm deep). For measurements of multi-grain aliquots, they used the latter, where each hole contained about fifteen 60–90 μm -diameter grains or about eight 90–125 μm -diameter grains. They found that both multi-grain aliquots and single grains measured in this way yielded indistinguishable results (Table 2 in Douka et al., 2014).

In this study, we re-investigated the multi-grain aliquot OSL data obtained by Douka et al. (2014). We focused on the multi-grain aliquot data because a much larger number of aliquots were accepted compared to single grains (see Douka et al., 2014). Their single-grain measurements showed that detectable OSL signals (i.e., initial test dose signal intensities more than three times the background intensity) were emitted by only ~20% of grains in 9 of their 11 samples (the exceptions being HF1 and HF5), and that fewer than 5% of the measured single grains contributed to >80% of the total OSL signal (Fig. 1a). It is, therefore, reasonable to assume that, for the multi-grain aliquots that contain ~15 grains (60–90 μm) or ~8 grains (90–125 μm) in each hole, the measured OSL signal should arise from one or two bright grains only for most of the samples investigated from this site. The variability in luminescence behaviour among the multi-grain aliquots should, therefore, be comparable to that observed from single-grain measurements for these samples. This inference is supported by the similar spread in sensitivity-corrected (L_x/T_x) values observed for single grains and small aliquots of sample HF11 that had received regenerative doses of between 100 and 200 Gy (Fig. 1b). The L_x/T_x values for the small aliquots are typically more precise than those obtained for the single grains, as well as more numerous (4-fold in this example), because each aliquot will likely contain one or two bright grains and, thus, emit an intense OSL signal.

3. Variability in dose response curve shapes among individual aliquots

The variation in DRC among different grains and aliquots has been described in detail in Douka et al. (2014). They applied several standard criteria to select aliquots and grains that should produce reliable D_e values and reject those with unsuitable physical properties. For example, aliquots and grains were rejected if they exhibited one or more of the following properties: 1) weak test dose OSL signals (i.e., the initial intensity of the T_n signal was less than three times the background intensity); 2) high levels of recuperation (i.e., the sensitivity-corrected OSL intensity measured in the 0 Gy regenerative dose cycle was more than 5% of the sensitivity-corrected natural OSL intensity, L_n/T_n); 3) poor recycling ratios (i.e., the sensitivity-corrected OSL values for duplicate regenerative doses differed by more than 2σ); 4) natural OSL signals equal to or greater than the saturation limit of the DRC (i.e., the L_n/T_n signal exceeded that induced by the largest regenerative dose ('Class 3' grains of Yoshida et al., 2000) or lay in the saturated region of the DRC, so only a minimum estimate of D_e could be obtained); 5) significant loss of OSL signal after exposure to infrared stimulation (i.e., OSL IR depletion ratios (Duller, 2003) smaller than unity by more than 2σ); and 6) apparently 'modern' grains (i.e., grains with well-described DRCs but no T_n signals, resulting in zero or negative D_e values). Since the focus here is to examine the variability in DRCs, we applied the first three criteria and the fifth (but ignored criteria 4 and 6, which involve the L_n/T_n signals) to identify aliquots with reliable DRCs. We also rejected aliquots with DRCs that were hyperbolic in shape (see examples in Douka et al., 2014) and aliquots with L_x/T_x signals that were too scattered to be fitted reliably with any line or curve. A comparison between the rejection criteria used in this study and those used in Douka et al. (2014) is shown in Table S1.

Aliquots that passed the above criteria were considered to have yielded reliable DRCs. Among the accepted aliquots, we observed significant variation in DRC shape. Fig. 2 shows three typical DRCs of the test dose-corrected signals $[(L_x/T_x)*D_t]$ for three aliquots from sample HF11. The DRCs saturate at different levels and doses. For aliquot #1, a characteristic saturation dose (D_0) of 35 Gy was obtained when the data were fitted using a saturating exponential function. For aliquot #2, a larger D_0

value of 74 Gy was calculated, and for aliquot #3 showed no sign of saturation up to ~270 Gy; a single saturating exponential function plus an extra linear term provides the best fit to the DRC for aliquot #3. This observation suggests that a common DRC or SGC cannot be established for different aliquots or grains from the Haua Fteah samples.

To quantify the variability in the DRCs for a large number of grains or aliquots, so that the D_0 values for each DRC can be compared, would require that all of the DRCs are described with a single saturating exponential function. However, many of the aliquots required an extra linear term or a double saturating exponential function to fit their DRCs (e.g., aliquot #3 in Fig. 2), thereby prohibiting direct comparisons of D_0 values. Instead, we propose a simple method: to compare the ratio between the L_x/T_x values for two different sensitivity-corrected regenerative doses. For example, the ratios of the L_x/T_x values for two dose points (270 and 102 Gy) for the three aliquots shown in Fig. 2 are 1.03 (#1), 1.36 (#2) and 1.51 (#3). The variation in the shape of DRCs can be compared using this ratio, provided that the two regenerative doses chosen for comparison are not too similar in size and that one of the regenerative doses is beyond the linear range of the DRC. We calculated the ratios between the L_x/T_x values at 270 and 102 Gy for a total of 436 aliquots from HF11 that passed the criteria mentioned above. The ratios are shown as a radial plot in Fig. 3. The mean ratios range from 0.94 to 1.79. All aliquots with ratios smaller than unity have large uncertainties and are statistically consistent with unity at 2σ . These results indicate large variation in the shape of the DRCs for different aliquots. For aliquots with ratios consistent with 1, it is expected that their DRCs have saturated as early as ~100 Gy, similar to aliquot #1 in Fig. 2. For those with ratios equal to or greater than ~1.5, it is expected that they may have OSL signals that keep growing beyond 270 Gy, similar to aliquot #3 in Fig. 2. Many of the ratios lie between 1 and 1.5 and are representative of aliquots that may have DRCs similar to aliquot #2 in Fig. 2.

To visually compare the DRCs from different aliquots, we arbitrarily divided aliquots into three groups, based on their L_x/T_x ratios. Since different groups of ratios reflect DRCs with different

saturation characteristics, we termed these ‘early’, ‘medium’ and ‘later’ to reflect those DRCs with L_x/T_x ratios that are the smallest, intermediate and highest, respectively. To quantify the mean L_x/T_x ratio for each of the three groups, we used the Finite Mixture Model (FMM) of Roberts et al. (2000) to estimate the weighted mean ratios for each group and the probability of falling in each group for each aliquot. An overdispersion (OD) value of 3.5% was used when fitting the model (see next section for justification of this OD value) and a three-component mixture assumed. The FMM estimates the probability of an aliquot belonging to each of the fitted components (three in this case), and we assigned each aliquot to the component (i.e., ‘early’, ‘medium’ or ‘later’ group) with the highest probability. For sample HF11, the ratios for the ‘early’, ‘medium’ and ‘later’ groups are 1.103 ± 0.005 , 1.283 ± 0.009 and 1.539 ± 0.013 (Fig. 3), with 48, 38 and 14% of aliquots falling in each group, respectively. We also tested this approach on sample HF11 using the ratios of the L_x/T_x values for the dose points at 270 and 51 Gy (instead of 270 and 102 Gy) and identified the similar three groups with 48, 39 and 13% of aliquots falling in each group, respectively (see Fig. S1).

Douka et al. (2014) reported large variation in the OSL signal intensity and decay curve shapes for different grains and aliquots from the Haua Fteah samples. In this study, we investigated these properties for individual aliquots from each of the ‘early’, ‘medium’ and ‘later’ groups. Fig. 4a shows the natural test dose signal intensity (T_n) for all aliquots of HF11 as a function of the group to which the aliquots were assigned. Signal intensities ranged from a few hundred up to tens of thousands of counts per second, but there is no discernible difference between the ranges of sensitivities for aliquots from the three groups.

It has been shown previously that the DRC shapes of different components of quartz OSL may differ significantly; for example, some of the ‘slow’ components are thought to saturate at much higher doses compared to the ‘fast’ and ‘medium’ components (e.g., Singarayer et al., 2000; Singarayer and Bailey, 2003). To determine whether the decay curve shape of individual aliquots is correlated to the shape of its DRC, we calculated the ‘fast ratio’ of Durcan and Duller (2011) for each

aliquot, using the decay curve following a regenerative dose of 271 Gy. The ‘fast ratio’ for each aliquot is plotted in Fig. 4b as a function of the group to which it has been assigned. Apart from 2 aliquots from the ‘later’ group and 1 aliquot from the ‘medium’ group that have much faster decay rates (‘fast ratio’ >100) than the other aliquots, similar ranges of ‘fast ratios’ are observed for aliquots within each of the three groups. The results presented in Fig. 4 suggest that it is not possible to distinguish between the ‘early’, ‘medium’ and ‘later’ DRC groups on the basis of OSL decay curve shapes or OSL signal intensity.

4. A new method to establish standardised growth curves for different groups of aliquots

The DRCs of the test dose-corrected signals for individual aliquots of HF11, separated into the three different groups, are shown in Fig. 5a (‘early’), b (‘medium’) and c (‘later’). It should be noted that different aliquots in the same group do not necessarily have the same form of best-fit function—some DRCs, for example, may be best described by a single saturating exponential function, while others may be better fitted by a single saturating exponential plus linear function. Such variation is expected for single-grain and small single-aliquot measurements because of their generally poorer reproducibility compared to large single-aliquot measurements (e.g., Galbraith et al., 2005). As a result, the shape of the DRC (i.e., the form of best-fit function) for a particular aliquot or grain will be largely influenced by OSL measurement uncertainties, including those associated with counting statistics and instrumental irreproducibility. So, for example, the need to include a linear term in the best-fit function (e.g., a single saturating exponential plus linear DRC) will be influenced by the L_x/T_x signal(s) for the highest regenerative dose(s). Consequently, the allocation of an aliquot to a particular DRC group should not rely on the form of the best-fit function for its individual DRC. As a more reliable means of allocating aliquots to different groups, we suggest instead using the L_x/T_x ratio for two regenerative dose points, taking into consideration the associated measurement uncertainties.

Large between-aliquot scatter in the L_x/T_x values is observed for each group (Fig. 5a–c), even though they have similar L_x/T_x ratios. The differences within each group are masked by the large

scatter of data points. Li et al. (2015) suggested that such between-aliquot scatter could stem from different grains experiencing variable degrees of sensitivity change between measurement of the regenerative dose signal and its subsequent test dose signal, as a result of different dosing, bleaching and thermal histories. To overcome this problem of between-aliquot scatter caused by sensitivity change, Li et al. (2015) proposed an improved normalisation method: DRCs for different aliquots of the same or different samples should be normalised using one of the regenerative dose signals (L_{r1}), rather than the test dose as originally proposed by Roberts and Duller (2004). Li et al. (2015) found that this method reduced the between-aliquot variation significantly and allowed them to establish a SGC for aliquots and samples from many different regions.

Accordingly, to maximise the reduction in observed variation of the test dose-corrected signals in Fig. 5a, b and c, we propose the use of a similar, but improved, normalisation method: the ‘least-squares normalisation’ (LS-normalisation). In the re-normalisation method of Li et al. (2015), only one regenerative dose signal was used for normalisation, whereas all regenerative dose signals are considered in the LS-normalisation method. The re-normalisation method was developed specifically for large aliquots, whereas the LS-normalisation method is applicable to small aliquots and single grains, as well as to large aliquots.

The LS-normalisation method involves an iterative scaling and fitting procedure, which includes the following steps:

- 1) Choose a ‘starting curve’ for each group: the exact form of this curve is not critical to the method, as it is used only to provide an initial ‘target DRC’ for the subsequent iteration process and does not necessarily have to represent the true shape of the common DRC for the particular group. For all of the aliquots in each group, we chose to fit the sensitivity-corrected regenerative dose signals using a single saturating exponential function (dashed lines in Fig. 5a, b) and a single saturating exponential function plus an extra linear term (dashed line in Fig. 5c), as these broadly describe the overall trend of the data points in each group. Given the large inter-aliquot scatter observed in each group (Fig. 5a–c), it would

be equally plausible to fit other functions, but, as noted above, the choice of the fitting function for these ‘starting curves’ is not crucial and does not affect the final results after iteration; it only affects the number of iterations required to achieve convergence in step 4, below.

2) Re-scale the individual DRCs: the data set for each aliquot is then multiplied by a scaling factor, such that the sum of squared residuals is minimised—a residual being the difference between the observed value (i.e., the normalised experimental data) and the fitted value (i.e., the dashed line). Each aliquot is treated individually, so different scaling factors are determined for different aliquots to minimise the least-squared residuals. Importantly, this re-scaling process does not change the form of the DRCs for individual aliquots, because the same scaling factor is applied to all of the data points for any particular aliquot. That is, re-scaling shifts the data points up or down the y-axis by the same percentage of L_x/T_x at each dose, to minimise the difference between the DRC of each aliquot and the fitted ‘starting curve’ for the group to which the aliquot has been assigned. Thus, parameter A in the saturating exponential function (see Fig. 2 caption) is varied by the same percentage, but D_0 is not affected by re-scaling. Furthermore, as the same percentage shift is applied to the L_n/T_n ratio for each aliquot, re-scaling does not change the corresponding estimate of D_e .

3) Re-fit the re-scaled data points: the re-scaled data sets obtained for each aliquot in step 2 are then fitted again, using one of three commonly-used functions: a single saturating exponential, a double saturating exponential, and a single saturating exponential with an added linear term. As the amount of inter-aliquot scatter is greatly reduced after re-scaling in step 2, the best-fit function can be chosen on the basis of a chi-squared statistical test. For our particular data sets, we found that the re-scaled data for the ‘early’ group were best described using a single saturating exponential function, whereas those for the ‘medium’ and ‘later’ groups were best fitted using a double saturating exponential function. Due to the re-scaling of individual DRCs in step 2, the form of the best-fit functions for the group DRCs in this third step does not necessarily have to be the same as those used to establish the ‘starting curves’ in step 1.

4) Iterate the re-scaling (step 2) and re-fitting (step 3): the re-scaled data set for each aliquot is then multiplied by a new scaling factor to minimise the least-squared residuals with the newly fitted DRC for the group to which the aliquot has been assigned. These newly re-scaled data are then fitted again, with the best-fit functions determined from chi-squared tests. This iterative procedure is repeated until there is negligible change ($<1\%$) in the normalised data and best-fit DRC parameters. For the samples studied here, a single or double saturating exponential function provides the optimal fit to the data points for each of the 3 groups, but other functions may provide better fits for other samples.

This LS-normalisation procedure was applied to the data in Fig. 5a–c, resulting in the normalised data and best-fitting curves shown in Fig. 5d–f for the ‘early’, ‘medium’ and ‘later’ groups, respectively. For the Haua Fteah samples, we found that the LS-normalisation procedure required only 2–3 iterations before negligible changes in the final results were achieved. Importantly, inter-aliquot variation of the regenerative dose signals was reduced significantly after LS-normalisation (Fig. 5d–f). The normalised DRCs have clearly contrasting shapes for each group: the DRC of the ‘early’ group saturates by ~ 100 Gy and the DRC of the ‘medium’ group lies between that of the ‘early’ and the ‘later’ groups; the latter shows no sign of saturation up to a dose of ~ 270 Gy. The results in Fig. 5d–f suggest that the division of aliquots into the 3 groups is a reasonable approximation of the general pattern of dose response, but we acknowledge that some aliquots could be assigned to one of the other two groups. For example, we assigned each aliquot to one of the three groups by fitting the FMM to the distribution of L_x/T_x ratios and choosing the group with the highest probability (see section 3), but some aliquots could have belonged to one of the other groups with a lower, but not insignificant, probability. The uncertainty involved in assigning individual aliquots to just one group will contribute to the scatter of LS-normalised values about each of the DRCs.

5. Comparing DRCs for different samples

To test whether different samples have similar DRC characteristics to HF11, we investigated the DRCs from three additional samples—HF1, HF3 and HF10. We first applied the same criteria as we

did to HF11 to reject aberrant aliquots that may yield unreliable DRCs. We then divided the accepted DRCs from each sample into 3 groups, based on a FMM analysis of the ratios between the L_x/T_x values for two different regenerative doses. The number of measured and accepted aliquots for each sample, and the number assigned to each group, are summarised in Table 1. The numbers of accepted aliquots used to establish the SGC are slightly different from the numbers of aliquots used for D_e determination by Douka et al. (2014). This is because some of the aliquots with acceptable DRCs are unsuitable for D_e determination due to their sensitivity-corrected natural signals lying in or above the saturated region of the DRC; these aliquots are accepted in this study for purposes of establishing the SGCs. In addition, some aliquots may have had a sensitivity-corrected regenerative dose point that deviated significantly from the general DRC and was omitted to determine the D_e ; such DRCs have been excluded in this study. The majority of the accepted aliquots (between about 40 and 60%) fall in the ‘medium’ group, with 12–41% and 18–27%, in the ‘early’ and ‘later’ groups, respectively.

The sensitivity-corrected regenerative dose signals for individual aliquots from the different groups and for the various samples are summarised in Fig. 6a–c. All aliquots in each group were then normalised using the LS-normalisation method described above, with the results shown in Fig. 6d–f. The large between-aliquot scatter is reduced significantly, and, more importantly, the different samples appear to share a common DRC for each of the three groups. The parameters of the best-fitting function for each group are summarised in Table 2. The ‘early’ group can be adequately described by a single saturating exponential function, with a D_0 value of 36 Gy, which limits dose measurement to below ~100 Gy if the $3D_0$ value (i.e., the dose at 95% of the DRC saturation level) is considered as an upper limit for D_e estimation. For the ‘medium’ and ‘later’ groups, double saturating exponential functions best describe the shapes of their DRCs. The doses corresponding to 95% of the saturation level are ~295 and ~475 Gy, respectively. The high saturation dose of the ‘later’ group is similar to those grains described as ‘supergrains’ by Yoshida et al. (2000). However, since the maximum regenerative dose given to our samples was only 296 Gy, the D_0 values obtained for the

'later' group may not be reliable; further studies on the saturation characteristics of the 'later' group are required.

We used the central age model (CAM) of Galbraith et al. (1999) to estimate the weighted mean and standard error of the normalised signals at each regenerative dose point for all of the aliquots in each of the three groups. These estimates are plotted as a function of the corresponding regenerative doses in Fig. 7a and provide further confirmation that different samples in the same group have essentially the same DRCs once they have been normalised appropriately using the LS-normalisation procedure (bearing in mind that Fig.7a shows the DRCs for the 3 groups of aliquots, whereas Fig. 2 shows the DRCs for 3 individual aliquots, one from each group). Fig. 7a also shows that the DRCs for the three groups are indistinguishable in shape up to a dose of ~50 Gy, beyond which they start to diverge.

The OD value obtained from the CAM analysis allows us to check whether the spread of data around their corresponding trend lines (Fig. 6d–f) is caused solely by measurement uncertainty or also reflects minor differences in the shape of the DRCs among individual aliquots. For example, a zero OD value would imply that 95% of the aliquots in the same group DRCs that are indistinguishable at 2σ . The OD values obtained for each group at each regenerative dose are shown in Fig. 7b. We have excluded the OD results for the zero-dose point, because of the large relative standard error associated with the counting statistics of weak OSL signals (Li, 2007; Galbraith and Roberts, 2012), and because any minor thermal transfer or recuperation could result in a large relative change in the zero-dose signal. Fig. 7b shows that there is a strong dependence of OD value on the size of the regenerative dose: the OD values are largest (up to ~10%) for the signals measured at low doses (~25 Gy) and decrease as the dose increases. This trend is expected because the relative effect of thermal transfer or recuperation will decrease as the size of the regenerative dose increases. Similar trends are observed for the 'early', 'medium' and 'later' groups. It is noted that we have rejected all the aliquots with recuperation >5%, which is calculated as the ratio between the zero-dose signal and the natural signal.

However, a small recuperation or thermal transfer relative to natural signal does not necessarily mean a small ratio to the smaller regenerative dose signal, because many of the natural doses are larger than the smaller regenerative doses given. So the effect of thermal transfer or recuperation will be amplified in smaller doses, when compared to the larger doses (including natural doses). OD values are similar beyond ~100 Gy, with a median OD value of 2.5% obtained at doses >100 Gy. We consider this value (2.5%) to be a reliable estimate of the variation in the sensitivity-corrected signal (L_x/T_x) between aliquots in the same group, and used this value when fitting the FMM to distinguish between the different groups of aliquots on the basis of their L_x/T_x ratios; because two L_x/T_x values are involved in the latter comparison, the relevant OD is $\sqrt{2.5^2 + 2.5^2} = 3.5\%$ (see section 3).

6. D_e estimation based on SGCs

In the previous sections we have argued that the quartz DRCs for samples from Haua Fteah can be divided into three groups. Aliquots assigned to the different groups have distinctive saturation characteristics, with those in the same group sharing similar DRCs (Figs 5 and 6). It is possible, therefore, to establish a SGC for each of the ‘early’, ‘medium’ and ‘later’ groups and estimate D_e values for individual aliquots based on the corresponding SGCs for the group to which it has been assigned. This should facilitate rapid and reliable estimation of D_e for a large number of single aliquots and single grains. In the original SGC method (e.g., Roberts and Duller, 2004; Lai, 2006; Lai et al., 2007), only the natural signal and its test dose signal (L_n/T_n) were required to determine D_e values. The re-normalisation method of Li et al. (2015) requires an additional regenerative dose OSL signal (and its corresponding test dose signal) to be measured. The original SGC and re-normalised SGC methods are applicable only if all aliquots can be described by a single SGC. As we have demonstrated above, this is not the case when single grains or smaller aliquots of quartz are measured, at least for these samples from Haua Fteah. Instead, measurement of two (or more) sensitivity-corrected regenerative dose signals is required to identify the DRC group to which the individual aliquot (or grain) belongs, and to assign the corresponding SGC to each aliquot (or grain) for D_e determination.

Two steps are involved in estimating D_e values using SGCs for samples that belong to different DRC groups. First, the L_x/T_x ratio for the two sensitivity-corrected regenerative dose signals needs to be determined to assign each aliquot to one of the three groups (see section 3). Second, the sensitivity-corrected natural signal (L_n/T_n) must then be multiplied by a scaling factor η (or re-normalisation factor), which is determined from one of the sensitivity-corrected regenerative dose signals (L_{r1}/T_{r1}) using the following equation:

$$\eta \frac{L_{r1}}{T_{r1}} = f(D_{r1}) \quad (1)$$

where $f(D)$ is the SGC function and D_{r1} is one of the two regenerative doses measured. For the D_{r1} , it is recommended that the regenerative dose close to the natural dose is chosen, in order to reduce the uncertainty resulting from the between-aliquot variation in individual DRCs (Li et al., 2015). The scaled or re-normalised natural signal ($\eta \frac{L_n}{T_n}$) can then be projected onto the corresponding SGC to estimate the D_e value for that aliquot; see Li et al. (2015) for a detailed description of this procedure and a worked example.

6.1. Separating aliquots into different groups

There is no need to discriminate between the different groups up to a dose of ~50 Gy, as they all share the same DRC, at least for these samples (Fig. 7a). The re-normalisation method of Li et al. (2015), which involves only one sensitivity-corrected regenerative dose, should therefore be applicable. For samples with D_e values in excess of 50 Gy, identifying the DRC group to which an aliquot belongs is crucial. In this case, two different sensitivity-corrected regenerative doses are required to assign aliquots to identified groups. Three factors should be considered when choosing the size of the two regenerative doses. First, they must differ sufficiently in size to maximise the difference between the two sensitivity-corrected regenerative dose signals. Second, at least one of the regenerative dose points must be beyond the linear region of the DRC (i.e., >50 Gy) because the re-normalised DRCs are identical in the linear dose region for the different groups (Fig. 7a). Third, the

two sensitivity-corrected regenerative dose points should bracket the sensitivity-corrected natural dose, to reduce uncertainties in D_e estimates resulting from extrapolating beyond the higher of the two chosen regenerative doses. Based on these considerations, we recommend that the lower regenerative dose falls within the range of 50 to 100 Gy, and the higher regenerative dose is ~200 Gy—or higher for samples with D_e values greater than 200 Gy.

6.2. *Identifying and rejecting poorly behaved aliquots*

One of the main drawbacks of estimating D_e values based on a SGC is that the performance of the SAR procedure cannot be fully assessed using the well-established criteria that form part of the full SAR procedure (i.e., recycling ratio test, and checks for the level of recuperation and unexpectedly large scatter in the regenerative dose signals around the DRC, the latter being required especially for single-grain measurements). Two sensitivity-corrected regenerative dose signals, however, may still provide useful information to enable rejection of some of the poorly behaved aliquots or grains.

Since the SGC for each group is established by selecting only those aliquots considered to be well-behaved and to produce reliable DRCs (see sections 3 and 4), the SGCs may provide some constraints on the range of reliable L_x/T_x ratios expected for the two sensitivity-corrected regenerative doses. For example, the SGCs of the ‘later’ and ‘early’ groups provide upper and lower limits of acceptable L_x/T_x ratios. Aliquots with L_x/T_x ratios lying outside this range can be considered as having unreliable L_x/T_x ratios and should be rejected prior to D_e determination. As another example, if the ratio of the L_x/T_x value at a dose of 200 Gy divided by the L_x/T_x value at a dose of 100 Gy is statistically smaller than unity, then at least one of these sensitivity-corrected values must be in error, because the former should be higher than, or equal to, the latter.

We have tested this approach on the 1000 aliquots measured for sample HF11. Two hundred and one of these aliquots were rejected because their initial test dose signal intensity was <3 times the background, or because the relative error on the test dose signal was >25%. For the remaining 799 aliquots, the L_x/T_x ratios for the two sensitivity-corrected regenerative doses (270 and 102 Gy) were

then calculated, and the upper (1.52) and lower (1.06) limits of the L_x/T_x ratios at 2σ calculated using the SGC functions for the ‘later’ and ‘early’ groups provided in Table 2. We also added, in quadrature, a relative error of 3.5% (for the two L_x/T_x values; Fig. 7b) to the error of each L_x/T_x ratio. Fig. 8a is a radial plot of the L_x/T_x ratios for the 799 aliquots. Any aliquot with a L_x/T_x ratio outside this range (1.06 to 1.52) was considered as ‘poorly’ behaved and was rejected from further analysis. Sixty-six aliquots were rejected on this basis, and these are shown as open triangles in Fig. 8a.

The DRCs from two aliquots that were rejected are shown in Fig. 8b and c; these DRCs are typical of rejected aliquots. Fig. 8b shows a DRC for an aliquot where one or more of the measured sensitivity-corrected regenerative dose signals deviated significantly from a monotonic DRC. The L_x/T_x ratio for the two sensitivity-corrected regenerative dose signals from such an aliquot can be significantly higher or lower, depending on whether the smaller dose point is underestimated significantly or the larger dose point overestimated significantly. In this example, the calculated L_x/T_x ratio was >1.52 (Fig. 8a). Fig. 8c shows a DRC for an aliquot that had a L_x/T_x ratio significantly smaller than the minimum accepted ratio of 1.06. This aliquot has a hyperbolic DRC and a recycling ratio that is not consistent with unity; both of these features would have led to a rejection of this aliquot during the full SAR procedure.

All accepted aliquots were then divided into three groups by fitting their L_x/T_x ratios using the FMM model, assuming a 3-component mixture and an OD value of 3.5%. The weighted mean ratios of each component, and the relative proportion of aliquots in each component, are shown in Fig. 8a. The D_e values for the aliquots in each group were then determined using their corresponding SGCs. For sample HF11, we have chosen the signal following the 102 Gy regenerative dose to determine the scaling factor η based on eqn. (1) for each of the accepted aliquots. The scaled natural signals ($\eta \frac{L_n}{T_n}$) were then projected onto the respective SGCs to determine the SGC-derived D_e values.

6.3. SGC D_e estimation and comparison with SAR D_e values

The procedure described above was applied to all six Haua Fteah samples investigated in this study. Aliquots were divided into three groups and their D_e values determined based on the SGCs. The number of rejected and accepted aliquots in each of the groups are summarised in Table 3. For these samples, an average of 41, 37 and 22% of accepted aliquots were assigned to the ‘early’, ‘medium’ and ‘later’ groups, respectively. We found that the scaled natural signals of many of the aliquots were at or above the saturation level of the SGCs, so finite D_e values could not be obtained for these ‘saturated’ aliquots. The number of such aliquots are summarised in Table 3 for each sample and each group. The number of rejected and accepted aliquots varies from sample to sample and group to group. For the youngest sample (HF1), all aliquots gave finite D_e values and contributed to the final D_e value for age determination. For the older samples, more aliquots were rejected due to saturation and, as expected, most of these fell into the ‘early’ group. For example, the percentage of aliquots from the ‘early’ group that could be considered as ‘saturated’ increased from zero for HF1 to ~70% for samples HF8, HF10 and HF11. In contrast, more than 80% of aliquots from the ‘medium’ group and nearly all (>94%) of the aliquots from the ‘later’ group in each sample produced finite D_e values. In Table 3, we have also summarised, for comparison, the number of aliquots in each group that were accepted for SAR D_e estimation by Douka et al. (2014). More aliquots were rejected during the SAR analysis because more rejection criteria were used and these criteria are not appropriate for SGC analysis (i.e., OSL IR-depletion ratio test; see section 6.2).

To test the reliability of D_e estimation using the SGCs for different groups, we compared the D_e values so obtained with those determined using the full SAR procedure (Fig. 9). The results show good consistency between the two methods, with D_e values scattered about the 1:1 dashed line. This concordance is further demonstrated in Fig. 10, where the ratios of the SGC and SAR D_e values for individual aliquots are shown as radial plots for each of the samples; the mean OD values for these distributions are 0% for HF1, HF4, HF8 and HF10, $1.3 \pm 2.7\%$ for HF3 and $6.4 \pm 1.2\%$ for HF11, with five of the six values statistically consistent with zero overdispersion. These results suggest that the

SGC method can produce reliable D_e estimates for individual aliquots from the three different DRC groups, and that no apparent biases have been introduced during the analytical process.

The weighted mean SAR and SGC D_e values for all accepted aliquots from each of the groups are summarised in Table 4 and shown in Figs 11a and b, respectively. The two youngest samples (HF1 and HF3) have SAR and SGC weighted mean D_e values for all three groups of aliquots that are statistically indistinguishable. This result is expected as nearly all the aliquots had natural signals below the saturation levels of the three SGCs and produced finite D_e values. For all older samples (HF4, HF8, HF10 and HF11), the SGC and SAR D_e values for the ‘early’ group are underestimated relative to the D_e values for the ‘medium’ and ‘later’ groups. The SGC ‘early’ group D_e values are underestimated by between 30% (HF4) and 50% (HF10) (Fig. 11a), whereas the SAR ‘early’ group D_e values are underestimated by ~20% relative to the corresponding D_e values for the ‘later’ group (Fig. 11b and Table 4). For these samples, many aliquots from the ‘early’ groups are saturated (Table 3), so finite D_e values could not be obtained and the D_e distribution is, therefore, truncated. The SGC and SAR D_e values for the ‘medium’ and ‘later’ groups are all statistically consistent at 1σ (Figs 11a and b and Table 4) and are not affected in the same way as the ‘early’ group.

It should be kept in mind that the comparisons between the SAR and SGC D_e values shown in Figs 9 and 10 are only available for those aliquots that produced reliable results using both approaches; a number of aliquots were rejected from the SAR D_e determinations (Douka et al., 2014) that were accepted for the SGC D_e determinations (Table 3). To investigate whether the ‘poorly behaved’ aliquots identified in the SAR procedure would result in erroneous results using the SGC approach, we compared the weighted mean SAR D_e values with the weighted mean SGC D_e values, including all accepted aliquots in each of the groups (Fig. 11c). The SGC and SAR D_e results for the ‘medium’ and ‘later’ groups are consistent but, for the ‘early’ group, the SGC produces smaller D_e values than the full SAR approach for the 4 older samples (HF4, HF8, HF10 and HF11). The underestimated weighted mean D_e values for the ‘early’ group contrasts with the overall consistency

between the SGC and SAR D_e values for individual aliquots (Figs 9 and 10). This suggests that some aliquots from the ‘early’ group that were considered ‘saturated’ on the basis of the SGC DRC may have intercepted their individual SAR DRCs and yielded finite D_e values, because the fitting function used for full SAR analysis is tailored to the individual DRCs, especially at doses close to saturation. Fig. 12 provides an example of such an aliquot from HF11, which explains why the SGC results for the ‘early’ group are underestimated relative to the SAR results. For the ‘medium’ and ‘later’ groups, no difference was observed because many fewer aliquots were rejected due to saturation (Table 3).

We also calculated the overall weighted mean D_e values for all accepted aliquots, regardless of the groups to which they had been assigned; these values are shown as black circles in Fig. 11. This procedure best approximates the conventional approach to obtaining D_e values from single aliquots and single grains. For the 4 youngest samples (HF1, 3, 4 and 8), the overall results are consistent with those obtained from the ‘medium’ and ‘later’ groups. For the 2 oldest samples (HF10 and HF11), however, the overall D_e values are underestimated compared to the D_e values obtained for the ‘medium’ and ‘later’ groups and the underestimation is greater for the SGC method (Fig. 11a) than when the full SAR procedure is used (Fig. 11b); the overall SAR D_e values are underestimated by about 5% and 6% for HF10 and HF11, respectively, while the corresponding overall SGC D_e values are underestimated by about 13% and 9%. This comparison implies that when a large number of aliquots are rejected due to the early saturation of some grains, the overall D_e could be underestimated because the full distribution of D_e values is truncated. In this case, it is necessary to divide the aliquots or grains into different groups, according to their saturation doses, and calculate the D_e values based only on those aliquots that have sufficiently high saturation levels (i.e., ‘middle’ and/or ‘later’ groups).

7. Discussion

The variability observed in DRCs for different aliquots is likely to be the result of several factors. Sources of variability include instrumental irreproducibility and experimental uncertainties, including counting statistics (Galbraith, 2002). In this study, a 2% uncertainty per OSL measurement

was included in the error calculations for individual D_e values to account for instrument irreproducibility (following the approach of Jacobs et al., 2006). We have shown that the wide range of DRCs observed for the Haua Fteah samples (Fig. 2) cannot be explained by this alone. Other sources of variability include, for example, differences in the characteristic saturation dose (D_0) level, allowing aliquots to be divided into three broad groups (Fig. 3), and the assignment of individual aliquots to only one group on the basis of the highest probability of fit as determined using the FMM. Variable degrees of change in luminescence sensitivity between the measurement of the regenerative and subsequent test dose signals can also cause differences in DRC shapes for different aliquots, but this effect can be reduced to a large extent by applying the LS-normalisation procedure (Figs 5 and 6).

Our results suggest that, after allowing for uncertainties associated with counting statistics and instrumental irreproducibility, there remains a small amount of OD (~2.5%) associated with the re-normalised L_x/T_x values for different aliquots from the same 'early', 'medium' or 'later' group. This OD is the result of inter-aliquot variation in the DRCs for aliquots within each of these groups. This is further reflected in the higher OD values associated the SGC D_e values compared to the SAR D_e values (Table 4). We expect that dividing the aliquots into more groups can reduce this uncertainty. To test this proposition, the data from HF11 were analysed again using the same method described in section 4, but fitting the L_x/T_x ratios to 4 components with the FMM to establish the corresponding SGCs for each component. For this sample, a slightly lower OD value of ~1.3% was obtained, compared to ~2.0% using 3 components. Dividing data into more components increases the complexity of data analysis and increases analytical time and effort, thereby diminishing one of the chief benefits of the SGC approach. Given that dividing aliquots into 3 groups has yielded results that are consistent with those obtained from the analysis of individual SAR growth curves (Figs 9 and 10) we consider that for practical reasons 3 groups are sufficient to produce reliable results based on SGCs for the Haua Fteah samples. Independent studies are required to determine the optimum number of DRC groups for samples from other regions and sites.

Although the large variability in DRC shape for different grains could be characterised by dividing them into several groups, it is still unclear why different quartz grains have different DRCs and why they saturate at different dose levels. One of the groups (i.e., the ‘later’ group) appears to have a much higher saturation dose than the other pairs, suggesting that the aliquots comprising this group have the potential to extend the age range of conventional quartz OSL dating, by using small single aliquots or choosing grains with high D_0 values (e.g., Yoshida et al., 2000). We have demonstrated that all three groups of aliquots yielded indistinguishable results for the younger samples (Fig. 11), but that samples with D_e values in excess of ~100 Gy (e.g., HF4, 8, 10 and 11) produced significant underestimates of D_e from the ‘early’ saturating group of aliquots. It is crucial, therefore, to investigate the variability in the DRCs of single-grain measurements if a significant proportion of measured and accepted grains are rejected due to saturation. Future studies are required to confirm the validity of D_e determination based on the ‘medium’ and ‘later’ groups for samples older than those investigated in this study (D_e values >140 Gy), given the problems of D_e estimation in the high-dose range of quartz OSL for samples from some other regions (e.g., Lai, 2010; Lowick and Preusser, 2011; Chapot et al., 2012; Duller et al., 2015).

8. Conclusions

Single grains and small aliquots of sedimentary quartz from Haua Fteah exhibit large variations in the shape of the DRCs constructed from the sensitivity-corrected regenerative dose signals. This variability can be quantified and characterised using the ratio of the L_x/T_x signals for two sensitivity-corrected regenerative dose points. At least 3 groups of DRCs can be identified for the Haua Fteah samples, with each group saturating at a different dose level (‘early’, ‘medium’ or ‘later’). Inter-aliquot variation among the aliquots within any particular group can be reduced significantly applying a least-squares normalisation procedure (LS-normalisation), which allows common DRC or SGC to be established for each group. D_e values can then be estimated for each aliquot using the

fitting parameters for the SGC developed for each group, together with measurements of the natural signal (L_n), one regenerative dose signal (L_{x1}) and their corresponding test dose signals (T_n , T_{x1}). Future research should focus on investigating the variability in DRC shape among samples from different regions and the feasibility of establishing common SGCs that are applicable worldwide (i.e., global SGCs).

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Figure captions:

Figure 1: (a) Distribution of OSL signal intensities from 1000 grains of quartz from sample HF11.

Data are plotted as the proportion of the total light sum that originates from the specified percentage of grains. (b) Ratio of L_x/T_x values between two sensitivity-corrected regenerative doses of 203 and 102 Gy, for 799 multi-grain aliquots and 199 single grains of sample HF11. The two dashed lines mark the highest and lowest limits of the ratios.

Figure 2: Typical DRCs from three aliquots of sample HF11. The data set for aliquots #1 and #2 were fitted using a single saturating exponential function, $Y = A[1 - \exp(-X/D_0)] + Y_0$, where Y is the test dose-corrected signal, X is the regenerative dose, D_0 is the characteristic saturation dose, and A and Y_0 are constants. The data for aliquot #3 was fitted using a single saturating exponential function plus an extra linear term, $Y = A[1 - \exp(-X/D_0)] + CX + Y_0$, where C is a constant.

Figure 3: Ratio of L_x/T_x values between two sensitivity-corrected regenerative doses of 270 and 102 Gy, for 436 aliquots of sample HF11. The data points from the ‘early’ and ‘later’ groups are shown as filled circles and those from the ‘medium’ group are shown as open triangles.

Figure 4: (a) Test dose signal intensity (T_n) for individual aliquots of sample HF11 for each of the 3 different groups. (b) The ‘fast ratio’ of the regenerative dose signal following a dose of 270 Gy for single aliquots from the different groups of sample HF11. Each aliquot was optically stimulated by an intense, green (532 nm) laser beam for 2 s (see Douka et al., 2014), with the ‘fast ratio’ calculated as the ratio of the background-subtracted counts in the first 2 channels of OSL decay (i.e., 0–0.04 s) divided by the background-subtracted counts in the 9th and 10th channels (i.e., 0.16–0.20 s).

Figure 5: Test dose-corrected signals $[(L_x/T_x) * D_t]$ for individual aliquots from the ‘early’ (a), ‘medium’ (b) and ‘later’ (c) groups of sample HF11, and their re-normalised counterparts (d, e and f). The latter were calculated using the LS-normalisation procedure described in the text.

677 Figure 6: Comparison of the test dose-corrected signals $[(L_x/T_x)*D_t]$ for individual aliquots from the
678 ‘early’ (a), ‘medium’ (b) and ‘later’ (c) groups from samples HF1, HF3, HF10 and HF11, and their re-
679 normalised counterparts (d, e and f). The symbols in b–f are the same as those used in (a).

680 Figure 7: (a) CAM estimates of the re-normalised signals for the 3 groups of aliquots from samples
681 HF1, HF3, HF10 and HF11. (b) The OD values for each group at each regenerative dose.

682 Figure 8: (a) Radial plot showing the L_x/T_x ratios for 800 aliquots of sample HF11. Accepted and
683 rejected aliquots are shown as filled circles and open triangles, respectively (see text for discussion).
684 Two rejected aliquots are marked as b and c, and their DRCs are shown in the right-hand panels (b and
685 c, respectively).

686 Figure 9: Comparison of individual D_e values estimated from SGCs (SGC D_e) and from construction
687 of full SAR growth curves for single aliquots (SAR D_e) for six samples from Haua Fteah. The errors
688 bars are shown at 1σ on both axes. The dashed line in each panel indicates the 1:1 relationship
689 between the SGC and SAR D_e values.

690 Figure 10: Radial plots of the ratios of the SGC and SAR D_e values for the same six samples as shown
691 in Fig. 9.

692 Figure 11: Weighted mean values of SGC D_e (a) and SAR D_e (b) for all accepted aliquots for each of
693 the three groups for samples HF1, HF3, HF4, HF8, HF10 and HF11. (c) Comparison of the weighted
694 mean SGC D_e and SAR D_e values for the six samples in (a). The filled black circles denote the overall
695 D_e values obtained as the weighted means of all ‘early’, ‘medium’ and ‘later’ individual D_e values.

696 Figure 12: Dose response curve for one aliquot from the ‘early’ group of sample HF11 that was
697 considered ‘saturated’ when compared to the SGC (black dashed line), but that intercepted the full
698 SAR DRC (orange full curve) and yielded a finite D_e value. The experimental SAR data were fitted
699 using a single saturating exponential function. The SGC curve has been scaled so that the re-
700 normalised natural signal is the same intensity as the L_n/T_n value shown on the y-axis.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Table 1: Number of aliquots measured and accepted for establishing SGCs for each of the DRC groups.

Sample	Grain size (μm)	No. of aliquots measured	Accepted for D_e estimation ^a	Accepted for establishing SGC	DRC group ^b		
					Early	Medium	Later
HF1	60–90	1000	593	673	197 (29%)	338 (50%)	138 (21%)
HF3	60–90	1000	534	488	58 (12%)	296 (61%)	134 (27%)
HF10	60–90	900	373	293	120 (41%)	113 (39%)	60 (20%)
HF11	90–125	1000	405	433	208 (48%)	167 (38%)	61 (14%)

^a Number of aliquots accepted for D_e estimation from Douka et al. (2014).

^b Proportion (percentage) of aliquots in each group is shown in parentheses below the number of aliquots.

Table 2: Summary of parameters for best-fit SGC functions shown in Fig. 6d–f for the ‘early’, ‘medium’ and ‘later’ DRC groups.

DRC group	Parameters of the SGC function $f(D)$ ^a					$2.3D_0$ (Gy) ^b	$3D_0$ (Gy) ^b
	A_1	D_1 (Gy)	A_2	D_2 (Gy)	y_0		
Early	45.1 ± 0.3	36 ± 1	-	-	0.87 ± 0.26	83	108
Medium	28.2 ± 4.6	29 ± 3	36.7 ± 3.4	122 ± 21	0.53 ± 0.20	230	295
Later	18.3 ± 4.8	25 ± 6	80.1 ± 2.2	170 ± 27	0.58 ± 0.40	355	475

^a The LS-normalised dose response data were fitted using either a single saturating exponential function of the form $f(D) = A_1 \left(1 - e^{-\frac{D}{D_1}}\right) + y_0$, or a double saturating exponential function of the form $f(D) = A_1 \left(1 - e^{-\frac{D}{D_1}}\right) + A_2 \left(1 - e^{-\frac{D}{D_2}}\right) + y_0$, where D is dose and the best-fit parameters of A_1 , D_1 , A_2 , D_2 and y_0 are listed above.

^b The $2.3D_0$ and $3D_0$ values correspond to the doses at 90% and 95%, respectively, of the saturation level for a DRC fitted using a single saturating exponential function – that is, the doses at 10% and 5% below the saturation level, respectively. An arbitrary $2D_0$ value, which corresponds to the dose at 86% of the saturation level, has been suggested as a safe upper limit for reliable D_e estimation (Wintle and Murray, 2006), but if the shape of the DRC is characterised accurately then much higher D_e values can be estimated reliably (Galbraith and Roberts, 2012; Roberts et al., 2015). D_e estimates have increasingly large standard errors at values above $3D_0$, as the natural signals impinge on the saturated region of the DRC.

Table 3: Number of aliquots measured, rejected, and accepted for D_e determination, together with the reasons for their rejection for SGC analysis.

Sample	Grain size (μm)	No. of aliquots measured	T_n signal < $3 \times \text{BG}$ or $\text{SE} > 25\%$ ^a	Poor L_x/T_x ratio	No. of identified aliquots in each group			Method ^b	No. of aliquots saturated			No. of aliquots accepted			
					Early	Medium	Later		Early	Medium	Later	Early	Medium	Later	Sum
HF1	60–90	1000	6	157	298	324	215	SGC	0	0	0	298	324	215	837
								SAR				160	296	107	563
HF3	60–90	1000	6	179	288	290	237	SGC	16	0	0	272	290	237	799
								SAR				74	288	145	507
HF4	90–125	1000	185	155	287	288	85	SGC	171	34	2	116	254	83	453
								SAR				60	239	54	353
HF8	90–125	500	104	69	126	106	95	SGC	96	18	5	30	88	90	208
								SAR				22	80	69	171
HF10	60–90	900	83	140	310	225	142	SGC	206	46	8	104	179	134	417
								SAR				115	139	86	340
HF11	90–125	1000	201	65	344	285	105	SGC	221	10	0	123	275	105	503
								SAR				84	182	76	342

^a T_n is the OSL signal measured in response to the test dose given after measurement of the natural OSL signal. BG and SE represent background and standard error, respectively.

^b Number of rejected aliquots are for SGC analysis only; those for SAR analysis are not shown because different rejection criteria were used (see Douka et al., 2014).

Table 4: Summary of the SGC and SAR D_e values, and corresponding overdispersion values, for the ‘early’, ‘medium’ and ‘later’ DRC groups of all six Haua Fteah samples.

Sample	Method	D_e (Gy)			Overdispersion (%)		
		Early	Medium	Later	Early	Medium	Later
HF1	SGC	23.3 ± 0.4	25.0 ± 0.3	25.0 ± 0.5	25.4 ± 1.3	22.5 ± 0.9	25.3 ± 1.5
	SAR	25.8 ± 0.4	25.2 ± 0.2	24.0 ± 0.4	15.8 ± 1.3	13.6 ± 0.8	14.6 ± 1.4
HF3	SGC	49.6 ± 0.8	53.0 ± 0.9	54.0 ± 1.0	21.9 ± 1.2	25.6 ± 1.2	27.5 ± 1.4
	SAR	50.6 ± 1.2	52.4 ± 0.6	52.5 ± 0.9	15.0 ± 2.0	16.4 ± 0.9	19.2 ± 1.3
HF4	SGC	69.7 ± 2.4	105.6 ± 2.9	94.3 ± 4.0	23.0 ± 3.2	33.9 ± 1.8	28.6 ± 1.6
	SAR	78.8 ± 3.9	102.0 ± 2.0	95.0 ± 3.0	23.0 ± 5.1	23.5 ± 1.7	19.8 ± 2.5
HF8	SGC	63.4 ± 3.4	110.5 ± 3.5	110.4 ± 3.7	20.8 ± 5.4	32.4 ± 2.5	30.3 ± 2.5
	SAR	81.9 ± 7.3	104.8 ± 2.4	107.1 ± 3.7	17.0 ± 12.8	0	23.8 ± 2.7
HF10	SGC	69.3 ± 2.3	137.1 ± 3.4	140.6 ± 4.8	28.1 ± 2.8	29.7 ± 2	37.5 ± 2.6
	SAR	111.4 ± 4.2	133.6 ± 4.1	141.3 ± 4.6	21.3 ± 4.1	25.2 ± 2.8	25.0 ± 2.7
HF11	SGC	85.5 ± 2.9	141.6 ± 3.0	136.0 ± 3.9	28.4 ± 2.8	30.5 ± 1.4	25.6 ± 1.7
	SAR	108.1 ± 7.2	133.6 ± 3.1	134.3 ± 4.1	43.8 ± 5.2	29.0 ± 1.8	25.9 ± 2.3