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Abstract

Potassium-rich feldspar (K-feldspar) grains were extracted from 28 sediment samples from sites in Asia, Europe and Africa and the dose response curves - or growth curves - were constructed from the post-infrared infrared stimulated luminescence (pIRIR) signals emitted by individual grains. The samples exhibit large between-grain variation in both pIRIR signal intensity and the shape of the corresponding growth curves; the latter can be largely explained as a result of measurement uncertainties, including counting statistics and instrumental irreproducibility. This between-grain variation can be reduced by applying a least-squares normalisation (LS-normalisation) procedure, which allows a common growth curve - or global standardised growth curve (gSGC) - to be established for individual K-feldspar grains from the range of samples investigated. Equivalent dose (D_e) values for single grains can be estimated using the fitting parameters of the gSGC, together with measurements of the natural signal (L_n), one regenerative-dose signal (L_x) and their corresponding test-dose signals (T_n , T_x). The D_e values obtained using the gSGC approach are consistent at 2σ with those obtained from the individual growth curves constructed for each of the grains. The establishment of a gSGC greatly reduces the time required to measure a large number of grains for old samples, and it also overcomes the problem of D_e underestimation that can occur using a full single-aliquot (or single-grain) regenerative-dose (SAR) procedure when a large number of grains are rejected due to issues of 'saturation'. The construction of a gSGC may, therefore, potentially extend the dating range of pIRIR signals beyond the reliable limit of D_e estimation using the standard SAR method.

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Single-grain dating of potassium-rich feldspar grains: towards a global standardised growth curve for the post-IR IRSL signal

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Abstract

Potassium-rich feldspar (K-feldspar) grains were extracted from 28 sediment samples from sites in Asia, Europe and Africa and the dose response curves — or growth curves — were constructed from the post-infrared infrared stimulated luminescence (pIRIR) signals emitted by individual grains. The samples exhibit large between-grain variation in both pIRIR signal intensity and the shape of the corresponding growth curves; the latter can be largely explained as a result of measurement uncertainties, including counting statistics and instrumental irreproducibility. This between-grain variation can be reduced by applying a least-squares normalisation (LS-normalisation) procedure, which allows a common growth curve — or global standardised growth curve (gSGC) — to be established for individual K-feldspar grains from the range of samples investigated. Equivalent dose (D_e) values for single grains can be estimated using the fitting parameters of the gSGC, together with measurements of the natural signal (L_n), one regenerative-dose signal (L_x) and their corresponding test-dose signals (T_n , T_x). The D_e values obtained using the gSGC approach are consistent at 2σ with those obtained from the individual growth curves constructed for each of the grains. The establishment of a gSGC greatly reduces the time required to measure a large number of grains for old samples, and it also overcomes the problem of D_e underestimation that can occur using a full single-aliquot (or single-grain) regenerative-dose (SAR) procedure when a large number of

grains are rejected due to issues of ‘saturation’. The construction of a gSGC may, therefore, potentially extend the dating range of pIRIR signals beyond the reliable limit of D_e estimation using the standard SAR method.

Keywords: K-feldspar, infrared stimulated luminescence, pIRIR, gSGC, D_e underestimation.

1. Introduction

The single-aliquot regenerative-dose (SAR) procedure (Galbraith et al., 1999; Murray and Wintle, 2000) has been widely used for optically stimulated luminescence (OSL) dating of quartz and infrared stimulated luminescence (IRSL) dating of feldspars. In this procedure, after measurement of the natural signal (L_n), a series of regenerative-dose signals (L_x), including a zero dose and one or more repeat doses, are measured for the same aliquot or grain to construct a growth curve of OSL or IRSL intensity as a function of dose. Following the measurement of L_n and L_x , a test dose is applied and measured (T_n , T_x) to monitor and correct for any sensitivity changes induced by different laboratory treatments, including irradiation, preheating and optical stimulation. The equivalent dose (D_e) of a grain or an aliquot is determined by interpolating its sensitivity-corrected natural signal (L_n/T_n) on to the growth curve of L_x/T_x intensity as a function of dose.

An alternative SAR-based method for D_e estimation involves establishing a common standardised growth curve (SGC) for separate aliquots of the same sample or of different samples (Roberts and Duller, 2004; Burbidge et al., 2006; Lai, 2006; Lai et al., 2007; Telfer et al., 2008; Long et al., 2010; Shen and Mauz, 2011). The construction of a SGC is based on the observation that T_x not only corrects for sensitivity change, but can also be used for between-aliquot normalisation. If a reliable SGC can be established, then it allows for D_e estimation based solely on measurements of L_n and T_n . A key benefit of a SGC is the considerable reduction in instrument time (by up to ~80% or more), especially when a large number of aliquots, or older samples, are measured. Li et al. (2015a) introduced the so-called ‘regenerative-dose normalisation’ — or ‘re-normalisation’ method — to

further reduce between-aliquot variations by normalising growth curves using one of the regenerative doses. They tested this method on samples with a range of depositional ages and collected from a variety of geological provenances and environmental settings around the world, and found that, for most samples, the OSL signal from single aliquots of quartz produced growth curves with similar shapes up to doses of ~250 Gy.

The previous studies are all based on large-aliquot measurements, however, where each aliquot consists of many hundreds to several thousands of grains, so any grain-to-grain differences are averaged. There are many instances where single-grain or small-aliquot analyses are preferable (Duller, 2008; Roberts et al., 2015), so large-aliquot SGC methods may be problematic due to grain-to-grain variations in growth curve shape and characteristic saturation dose (D_0) values (e.g., Roberts et al., 1999; Duller et al., 2000; Jacobs et al., 2003; 2013). Accordingly, Li et al. (2016) investigated the applicability of a SGC for single grains and small aliquots (~8–15 grains per aliquot) of sedimentary quartz from the site of Haua Fteah in Libya, which has a variety of independent age controls. They found that the samples exhibited large between-grain and -aliquot variation in the shape of their growth curves, which prevented the establishment of a universal SGC for all of the grains or aliquots. Instead, they showed that the between-grain variation in growth curve shape could be greatly reduced by applying an improved re-normalisation procedure — so-called least-squares normalisation (LS-normalisation) — and, as a result, the growth curves for their single grains of quartz could be divided into a minimum of three groups, each characterised by a different SGC.

The re-normalisation method was also tested on single aliquots of K-feldspar from different regions around the world (Li et al., 2015b). This study found that the post-infrared IRSL (pIRIR) signals from different samples produced similarly shaped growth curves, despite the variety of geological sources, depositional settings and burial ages. This finding indicates the potential to establish a global SGC (gSGC) for K-feldspar. However, the single-aliquot results of Li et al. (2015b) are still subject to the averaging effects of measuring multiple grains simultaneously. In the present

study, we take the gSGC approach a step further and examine the post-IR IRSL signals from single grains of K-feldspar extracted from sedimentary samples collected in Asia (Russia and China), Europe (France) and Africa (Kenya).

2. Sample descriptions

For this study, we selected 28 sediment samples from sites in Asia, Europe and Africa. Some of these samples have been dated previously and their D_e values determined using standard SAR methods; here we investigate their growth curves in detail for purposes of developing and testing a gSGC. Samples Chag12-1, -6, 9 and -10 were collected from sedimentary deposits at Chagyrskaya Cave, Russia (Derevianko et al., 2013, 2017; Slon et al., 2017), and samples DCE12-3, -4, -12 and -14, DCM12-3, -5, -26 and -27 and DCS12-5 were collected from sediments deposited in Denisova Cave, Russia (Krause et al., 2010; Reich et al., 2010; Slon et al., 2017). LC10-05 is a sample of colluvial–alluvial sediment collected from Early Aurignacian levels at Les Cottés, France (Talamo et al., 2012; Jacobs et al., 2015). Samples RUP-1, -2 and -3 were collected from Late Pleistocene sediments at Nyamita 2, and samples RUP-4 and RUP-5 are from Wakondo — both of these sites being located in Lake Victoria basin, Kenya (Blegen et al., 2015). An aeolian sand sample (Sm5) was collected from the Shimao section on the southeastern margin of the Mu Us Desert, central China (Sun et al., 1999; Li et al., 2011, 2013, 2014b). Samples Enderby-OSL1, -OSL2, -OSL3 and -OSL4 were collected from the Enderby Island, New Zealand. Samples BJZ-OSL1 and -OSL3 were collected from the Banjinzi site, Nihewan Basin, China (Guo et al., 2016). Sample HF-8 and HF-10 were collected from the site of Haua Fteah in Libya (Douka et al., 2014; Jacobs et al., 2017).

3. Experimental procedures and analytical facilities

All the samples were prepared using routine procedures (Aitken, 1998). The samples were first treated with HCl acid and H_2O_2 solution (to remove carbonates and organic matter, respectively), and then dried and sieved to obtain grains of 180–212 μm in diameter. The K-feldspar grains were

separated from quartz and heavy minerals using a sodium polytungstate solution with a density of 2.58 g/cm³. The separated K-feldspar grains were immersed in 10% HF acid for 40 min to etch the surfaces of the grains and remove the outer, alpha-irradiated portions, and then rinsed in HCl acid to remove any precipitated fluorides.

IRSL measurements were made on automated Risø TL/OSL-DA-20 readers equipped with infrared light-emitting diodes (LEDs, 870 nm) and a focussed infrared laser (830 nm) for stimulation (Bøtter-Jensen et al., 2003). The violet/blue emissions were detected with Electron Tubes Ltd 9235QA photomultiplier tubes fitted with Schott BG-39 and Corning 7-59 filters. The single-grain measurements were made using aluminium discs drilled with 100 holes, each 300 µm in diameter and depth (Bøtter-Jensen et al., 2003). Beta doses were administered using ⁹⁰Sr/⁹⁰Y sources mounted on the readers, with the dose rates calibrated for the individual holes.

For all samples, we applied a two-step single-grain pIRIR procedure (Blegen et al., 2015). The first infrared stimulation is made at 200 °C using infrared LEDs to stimulate all 100 grains on the disc simultaneously, and the second is made at 275 °C using the infrared laser to stimulate each grain individually (Table 1). A temperature of 275 °C was chosen for the second measurement, instead of 290 °C (e.g., Thiel et al., 2011), to avoid significant thermal erosion of the pIRIR signal due to each disc being held at an elevated temperature while the laser locates a disc and stimulates each of the 100 grains in turn. The suitability of this procedure for single-grain measurements of K-feldspars has been demonstrated previously using dose recovery, recycling ratio, recuperation and residual dose tests (Blegen et al., 2015; Guo et al., 2016).

4. Counting statistics and instrumental reproducibility

The experimentally observed grain-to-grain variability in the growth curves is governed by several sources, including counting statistics (σ_{cs}), instrumental irreproducibility (σ_{ins}) and the intrinsic differences among growth curves for individual grains. Based on numerical simulation, Li et

al. (2017) demonstrated that measurement uncertainties, including σ_{cs} and σ_{ins} , may result in a large apparent scatter of measured L_x/T_x ratios (and, hence, the growth curves), even for grains with the same true growth curve. To quantify the intrinsic differences in the growth curves for individual grains, the uncertainties arising from σ_{cs} and σ_{ins} must be allowed for. We used the same methods and equations provided in Adamiec et al. (2012), Bluszcz et al. (2015) and Li et al. (2017) to estimate the correction factors for the variance in the dark counts (k_{DC}^2) and photon counts (k_{ph}^2) for the two luminescence readers used in this study — Risø3 and Risø5. Risø3 has k_{DC}^2 and k_{ph}^2 values of 2.14 and 1.12, and Risø5 has k_{DC}^2 and k_{ph}^2 values of 3.18 and 1.86. We previously reported the k_{DC}^2 and k_{ph}^2 values for another of our readers (Risø2) as 3.69 and 1.88 (Li et al., 2017). These results indicate that all three readers have count data with greater variance than expected for a Poisson distribution; the correction factors are, therefore, incorporated into our error calculations, based on eqn (3) in Bluszcz et al. (2015).

By definition, σ_{ins} is an estimate of all systematic uncertainties arising only from the measurement instrument — for example, uncertainties in heating temperature, light stimulation power, and movement of discs between measurements. In this study, we estimated σ_{ins} for the single-grain pIRIR measurements by repeatedly irradiating, preheating and stimulating individual grains from several samples. This is similar to the method proposed for quartz grains (e.g., Truscott et al., 2000; Jacobs et al., 2006). In this test, four samples from Russia (Chag12-1, DCE12-4, DCE14-1 and DCS12-1) and one sample each from China (Sm5) and France (LC10-05) were measured on Risø3 or Risø5. The K-feldspar grains used for this test had previously been subjected to the full sequence of SAR measurement cycles to determine their D_e values. The SAR cycles were then repeated between 8 and 16 times, using the same regenerative dose each time; the size of this repeat dose ranged from 50 to 900 Gy (Table 2). A total of 100–300 grains was measured for each sample, but only those grains that yielded detectable signals (i.e., initial intensity of the test-dose signal at least 3σ above background) were accepted for analysis. Table 2 lists the repeat regenerative doses given to each

sample and the number of grains accepted for estimating the instrumental irreproducibility. Fig. 1a and b show the histogram distributions of the L_x/T_x signals from individual grains of samples Chag12-1 ($n = 576$) and Sm5 ($n = 672$) (measured in Risø3 and Risø5, respectively), normalised by the weighted mean L_x/T_x of each grain. Most of the normalised values are centred at unity, but some values deviate significantly from unity, by up to ~40%.

To estimate the instrumental irreproducibility, we adopted two methods. Both methods work with $y = \log(L_x/T_x)$ and its variance s ; the latter is equal to the relative standard variance of L_x/T_x . Formally, the j^{th} observed $\log(L_x/T_x)$ for the i^{th} grain can be written as

$$y_{ij} = \mu_i + \varepsilon_{ij} \quad (1)$$

where μ_i is the true $\log(L_x/T_x)$ value for the i^{th} grain and ε_{ij} is the deviation of y_{ij} from μ_i . We assume ε_{ij} to be a random quantity with mean (expected value) 0 and variance s_{ij}^2 ; the latter is the sum of the variances of counting uncertainty σ_{ij}^2 and instrumental uncertainty σ_{ins}^2 . It must be remembered that the σ_{ins}^2 value is associated with a single measurement (L_x or T_x), so the variance for L_x/T_x should be given as $s_{ij}^2 = \sigma_{ij}^2 + 2\sigma_{ins}^2$, since two measurements are involved.

The first method (Method 1) we used to estimate instrumental irreproducibility is similar to that used for quartz OSL by Jacobs et al. (2006), in which the relative standard error arising from instrumental uncertainty is calculated for individual grains. This was achieved by subtracting the error that arises from counting statistics from the corresponding error observed from repeated measurements, which can be expressed as follows

$$\hat{\sigma}_{ins}^2 = (s_{ij}^2 - \sigma_{ij}^2)/2 \quad (2)$$

Using this method, a range of $\hat{\sigma}_{ins}$ values were obtained for individual grains, and the median of these values was used to quantify the instrumental uncertainty. The estimates of σ_{ins} obtained using this

method on different samples are summarised in Table 2. A wide range of values from 0.6 to 3.4% were obtained. Mean values of 2.7 and 2.1% are obtained for Risø3 and Risø5, respectively.

The second method (Method 2) is based on a generalised method of moments estimator, which treats all the grains from the same sample simultaneously. We first estimate μ_i from the weighted mean of repeated measurements:

$$\hat{\mu}_i = \frac{\sum_{j=1}^n w_{ij} y_{ij}}{\sum_{j=1}^n w_{ij}} \quad (3)$$

where n is the number of SAR cycles measured for each grain, and w_{ij} is the weight and is calculated as

$$w_{ij} = \frac{1}{s_{ij}^2} = \frac{1}{\sigma_{ij}^2 + 2\sigma_{ins}^2} . \quad (4)$$

The best estimates of μ_i and σ_{ins} , denoted by $\hat{\mu}_i$ and $\hat{\sigma}_{ins}$, are the values of μ_i and σ_{ins} satisfying the following equation:

$$\sum_{i=1}^k \sum_{j=1}^n \frac{1}{\sigma_{ij}^2 + 2\hat{\sigma}_{ins}^2} (y_{ij} - \hat{\mu}_i)^2 = N - k \quad (5)$$

where N is the total number of observations (i.e., y_{ij}) and k is the number of grains analysed. The above equation can be solved numerically to obtain $\hat{\mu}_i$ and $\hat{\sigma}_{ins}$. The $\hat{\sigma}_{ins}$ values obtained using this method based on all the grains are summarised in Table 2 (column 6). The values range from 1.9 to 5.2 %, which are significantly higher than those obtained using Method 1. Since Method 2 deals with all the grains rather than individual grains as in Method 1, the overestimation of σ_{ins} value in Method 2 indicates that some of the grains may have significantly larger variance in repeated measurements. These ‘aberrant’ grains had a greater effect on results obtained for Method 2 compared to Method 1, because the median value would be less sensitive to extreme values. We offer two explanations for this observation. One is that these grains may have different measurement uncertainties from the

majority of grains; this can be ruled out, because instrumental irreproducibility should depend only on what instrument is used, rather than on the characteristics of the particular grain being analysed.

The other explanation is that these grains are poorly behaved, so that the SAR procedure does not sufficiently correct for sensitivity changes that occurred during the repeated measurement cycles. Fig. 2 shows examples of three ‘poorly-behaved’ grains (a–c) and one ‘well-behaved’ grain (d) from sample DCE14-1. When the moment method is applied to each of these grains, similar to Method 1, the σ_{ins} values are 8.6, 6.0, 5.2 and 0% (a–d, respectively). As expected, therefore, the observed variances of the repeat measurements for the ‘poorly-behaved’ grains are larger than those resulting solely from counting statistics and instrumental irreproducibility. We consider this to be a reasonable explanation for the observed difference between the two methods, because different grains may have different luminescence characteristics and not all grains need necessarily perform equally well using a SAR procedure. Our proposed explanation is supported by the corresponding growth curve data for these grains (insets in Fig. 2); that is, the poorly-behaved grains yielded poor growth curves, whereas the well-behaved grain produced a good growth curve. In practice, such poorly-behaved grains would be rejected from the data set prior to D_e determination, because of their poor recycling ratios (e.g., Fig. 2a and c) or the large scatter in their L_x/T_x values (e.g., Fig. 2b). For the same reason, these poorly-behaved grains should be discarded when estimating the instrumental irreproducibility. Including data for such poorly-behaved grains would likely result in overestimation of the instrumental irreproducibility error, especially when Method 2 is applied.

The assessment of the behaviour (i.e., reproducibility) of the grains in repeat measurements relies on a reliable estimate of instrumental uncertainty. The latter is, in turn, affected by the results obtained from the sample of measured grains. To address this issue, we applied the chi-squared statistical test iteratively to identify poorly-behaved grains and to estimate the instrumental uncertainty based only on the data collected from well-behaved grains. We first applied the moment method to all of the grains to obtain an initial estimate of σ_{ins} . Assuming that the individual

measurements are independent and standard normal random variables, the standardised residuals for individual observations R_{ij} , calculated as

$$R_{ij} = (y_{ij} - \hat{\mu}_i) / \sqrt{\sigma_{ij}^2 + 2\hat{\sigma}_{ins}^2} \quad (6)$$

are expected to follow the chi-squared distribution with $n-1$ degrees of freedom, and with mean R_{ij} equal to $n-1$ with a variance equal to $2(n-1)$. Fig. 3a shows the distribution of chi-squared values for all of the grains from sample Chag12-1, compared with the theoretical chi-squared distribution for $n-1$ degrees of freedom (red curve). Some grains have chi-squared values significantly higher than the 98th percentile of the chi-squared distribution (green line). We consider these grains to be ‘poorly behaved’ and discarded them before re-applying the moment method to the remaining grains again to obtain an improved estimate of the best-fit σ_{ins} and individual chi-squared values. This procedure is applied iteratively until all grains have chi-squared values smaller than the 98th percentile of the theoretical chi-squared distribution (Fig. 3b). In this particular example 17 (of 92) grains were identified as ‘poorly behaved’. The ‘well-behaved’ grains yielded an σ_{ins} estimate of 2.6%, which is substantially smaller than the value of 4.3% obtained using all grains. Furthermore, the distribution of chi-squared values for the ‘well-behaved’ grains, including the mean and variance, are broadly consistent with the theoretical distribution. We, therefore, consider that the value of 2.6% is the most reliable estimate of σ_{ins} based on the data for Chag12-1.

Using the above method, we calculated the σ_{ins} values for the different samples (column 8 in Table 2). A range of values was obtained and, similar to the results for Chag12-1, the corresponding estimates of σ_{ins} were reduced significantly following application of the chi-squared method to discard poorly-behaved grains. The extent of reduction is sample-dependent, as expected, because the proportion of poorly-behaved grains is also sample-dependent (column 7 in Table 2). The most reliable estimates of σ_{ins} for Risø3 range from 1.7 to 2.4%, with a mean of 2.2%, while the corresponding range of values for Risø5 is 1.5–2.8 %, with a mean of 2.2%. These values are similar

to, but less scattered than, those obtained from Method 1 based on the median of estimates for individual grains. These results also suggest that there is no systematic difference between the instrumental uncertainties for these two readers. Accordingly, we adopted the value of 2.2% for σ_{ins} obtained from Method 2 in our calculations of the L_x/T_x measurement uncertainties for the various samples analysed from the different sites in this study.

5. Comparing growth curves for different grains

The growth curve for each K-feldspar grain was constructed using the L_x/T_x ratios obtained from a series of regenerative- and test-dose signals, including a duplicate regenerative dose and a zero dose to determine the recycling ratio and extent of recuperation (Table 1). A key constraint on the precision with which L_x/T_x ratios can be measured is the absolute brightness of individual grains. We express absolute brightness as the net T_n signal for each grain, determined from the pIRIR counts for the first 0.1 s of infrared stimulation, minus the average background count over the final 0.2 s of stimulation. The test dose was fixed at ~50 Gy for all grains, enabling direct comparison of inherent brightness for grains from the same and different samples.

Table 3 lists the number of grains measured for each sample, and the number of grains rejected because of a weak T_n signal. The number of grains with detectable signals varies considerably between samples from different sites and also, in some cases, between samples from the same site. For example, only ~9% of the grains from sample Sm5 were rejected as having weak T_n signals, whereas some samples, such as LC10-05, RUP-1, and DCE12-14, had more than 90% of their grains rejected on this basis (Table 3). This result illustrating substantial within-site differences in intrinsic grain brightness.

Fig. 4a is a cumulative light sum plot for four samples, each from a different geographic region, showing considerable differences in their pIRIR sensitivities. There are two types of distribution: samples DCM12-26 and Sm5 have the most uniform distribution of grain sensitivities, with around

30–40% of the grains contributing 90% of the light sum, whereas only about 5–10% of the grains account for 90% of the light sum for samples LC10-05 and RUP-4. The distributions of individual grain sensitivities for the same samples are plotted in Fig. 4b. Sample Sm5 has the brightest grains (T_n intensities $>100,000$ cts/s) and the smallest relative difference in sensitivity (10×) among the brightest 20% of grains (i.e., $>100,000$ to $>10,000$ cts/s), while sample LC05-10 has the largest relative range in sensitivity (100×) for the brightest 20% of grains (i.e., $\sim 10,000$ to ~ 100 cts/s).

Fig. 5 displays example pIRIR decay curves and corresponding growth curves (left- and right-hand panels, respectively) for six individual grains from each of samples DCM12-26 (Fig. 5a and b), LC10-05 (Fig. 5c and d), RUP-4 (Fig. 5e and f) and Sm5 (Fig. 5g and h). The inset plots in Fig. 5a, c, e and g show the decay curves normalised to the first measurement channel, to account for the significant differences in signal brightness. All grains within and between samples have broadly similar decay curve shapes, and are depleted to instrumental background after ~ 0.5 s of laser stimulation.

The sensitivity-corrected growth curves were constructed from the L_x and T_x pIRIR signals using a general-order kinetic (GOK) function (Guralnik et al., 2015) of the form $f(x) = a[1 - (1 + bcx)^{(-1/c)}] + d$, where x is the dose and parameters a , b , c and d are constants. The growth curves appear to vary in shape — that is, a wide range of L_x/T_x ratios are observed at doses above ~ 100 Gy for three of the samples (Fig. 5b, f and h). The exception is sample LC10-05 (Fig. 5d), for which the highest regenerative dose used was only ~ 200 Gy, compared to doses of up to 800–1000 Gy for the other three samples. The saturation dose (and saturation level) also differ significantly from grain to grain for the latter samples (Fig. 5b, f and h) — for example, the D_0 values for sample DCM12-26 range from ~ 145 Gy (grain #4) to ~ 300 Gy (grain #1) using a single saturating exponential function to fit the growth curves (Fig. 5b). No correlation is evident between the shapes of the decay curves and the growth curves.

Differences in the shapes of the growth curves among grains from the same sample may result from several factors. First, individual grains may have intrinsic differences in luminescence behaviour that are governed by their physical properties, such as the type, relative concentration, ionisation and trapping cross-sections of defects involved in pIRIR production. Second, individual grains may have different post-crystallisation irradiation, bleaching and thermal histories, resulting in variable degrees of sensitivity change between measurement of L_x and T_x and, consequently, a range of L_x/T_x ratios at the same dose (Li et al., 2015a). The third factor relates to measurement uncertainties, including σ_{CS} and σ_{INS} , which can lead to significant variability in the observed L_x/T_x ratios and growth curves, even if the intrinsic luminescence behaviour of all grains is identical (Li et al., 2017).

Li et al. (2016) proposed that variation in growth curves due to intrinsic and measurement differences between grains can be overcome by appropriately normalising the growth curves using the least-squares normalisation (LS-normalisation) method, and they tested the procedure on small aliquots of quartz. To test its validity for individual grains of K-feldspar, we applied the LS-normalisation procedure to compare the single-grain growth curves constructed for the samples in this study. The procedure involves the following steps: (1) fit the L_x/T_x ratios for all grains using a best-fit model; (2) re-scale the L_x/T_x ratios for each grain using scaling factors determined in such a way that the difference between the re-scaled ratios and the best-fit growth curve is minimised through an optimisation procedure. Each grain is treated individually for re-scaling, so different scaling factors are determined for different grains; (3) repeat the fitting (step 1) and re-scaling (step 2) procedures iteratively until there is negligible change in the relative standard deviation of the re-normalised L_x/T_x ratios. The LS-normalisation procedure can be implemented using the built-in function `lsNORM()` provided in the R-package ‘numOSL’ (R Core Team, 2016; Peng et al., 2013; Peng and Li, 2017). A GOK function (Guralnik et al., 2015) is used to fit all growth curves during the LS-normalisation procedure.

Prior to application of the LS-normalisation procedure, grains with unsuitable pIRIR properties need to be rejected (Li et al., 2016). Here, we have used several criteria that are similar to, but slightly different from, those employed in previous studies (e.g., Jacobs et al., 2003, 2006; Blegen et al., 2015; Guo et al., 2016, 2017). Grains were rejected if: (1) the initial T_n signal is less than 3σ above of the corresponding background count, or the relative error on T_n is $>25\%$; (2) the ratio of the 0 Gy L_x/T_x value to the maximum regenerative-dose L_x/T_x value is $>5\%$; and (3) the L_x/T_x data points are too scattered to be fitted reliably with any line or curve, using two statistical criteria to determine whether the L_x/T_x ratios are too scattered. The first of these criteria is the figure-of-merit (FOM) value (e.g., Balian and Eddy, 1977; Peng et al., 2016) for the fitted growth curve, which is defined as

$$FOM(\%) = 100 \times \frac{\sum_{i=1}^n |y_i^o - y_i^f|}{\sum_{i=1}^n y_i^f} \quad (6)$$

where y_i^o and y_i^f denote the i^{th} observed and fitted values, respectively. The second criterion is the reduced-chi-square (RCS) value (e.g., Bevington and Robinson, 2002) for the fitted growth curve, which is defined as

$$RCS = \frac{1}{N-n} \times \sum_{i=1}^n \frac{(y_i^o - y_i^f)^2}{\sigma_i^2} \quad (7)$$

where N and n denote the number of observations and fitted model parameters, respectively, and σ_i is the standard error for the i^{th} observation. We set upper limits of 10% for the FOM and 5 for the RCS criteria, which appear to successfully select only grains with satisfactory growth curves. Fig. 6a–d display example growth curves for 4 grains from sample Sm5 with different FOM and RCS values. It can be seen that these criteria reflect the goodness-of-fit well, with a satisfactory fit requiring both values to fall below the set limits (e.g., Fig. 6c and d). We did not apply an explicit rejection criterion for the recycling ratio, as poor recycling is reflected in the FOM and RCS values. For example, the repeat dose points in Fig. 6c and d lie within 2σ of a common value, which is our standard recycling

ratio criterion of acceptance for single grains. The selection of grains based on the above criteria was achieved using the function `pickSARdata()` provided in the R-package ‘numOSL’. The numbers of measured and rejected grains for each of our samples are summarised in Table 3, along with the reasons for rejection.

The LS-normalisation procedure was applied to all accepted grains of sample Chag12-9 ($n = 280$) to test whether grains from the same sample share a common growth curve. Fig. 7a and b shows the test-dose standardised ratios ($L_x/T_x \times D_t$) before and after LS-normalisation, plotted as a function of regenerative dose. Large scatter is observed among the $L_x/T_x \times D_t$ ratios for the individual grains (Fig. 7a), but the scatter is reduced significantly after LS-normalisation (Fig. 7b). The values for the parameters a , b , c and d for the best-fit function in Fig. 7b (solid line) are 241, 0.00354, 0.838 and 2.52, respectively. The c value is significantly greater than zero, indicating a deviation from first-order kinetic behaviour (Guralnik et al., 2015). We calculated the ratios between the LS-normalised data (observed) and the best-fit SGC (expected) for all doses greater than zero, and these are plotted as a function of dose in Fig. 7c and as a radial plot in Fig. 7d. There is no observable systematic dose-dependency of the ratios (Fig. 7c) and, in line with statistical expectations, ~95% of the ratios are consistent with unity at 2σ (Fig. 7d). These results imply that the individual grains from Chag12-9 share a common growth curve, and that the observed scatter in their $L_x/T_x \times D_t$ ratios is mainly caused by measurement uncertainties and the differing extents of sensitivity change between measurement of any particular L_x signal and its subsequent T_x signal among the individual grains.

We then extended the LS-normalisation procedure to all accepted grains from 28 of the samples to test whether grains from different sites and geographic regions share a common growth curve. Fig. 8a shows the wide range of SGCs for the different samples, but these between-sample and -grain differences are significantly reduced after implementation of the LS-normalisation procedure. A common SGC can be fitted to these LS-normalised data (solid line in Fig. 8b), with values of 1.618, 0.00356, 0.871 and 0.0232 for the parameters a , b , c and d , respectively. Parameters b and c control

the shape of the growth curve and it is worth noting that their values differ slightly from those obtained for best-fit curve in Fig. 7b (0.00354 and 0.838). This is due mainly to the use of more data points at doses >1000 Gy to construct the growth curve in Fig. 8b, as parameter c is primarily controlled by the high-dose data points. This difference has a negligible effect on D_e estimation, however, when L_n lies within the range of given doses (<1200 Gy in Fig. 8b). The existence of a common SGC for the samples in this study is further supported by the distribution of ratios between the observed data and their corresponding expected values from the best-fit SGC for doses greater than zero (Fig. 8c and d) — that is, ~91% of the ratios are consistent with unity at 2σ , which closely matches statistical expectations. This result suggests that a gSGC may be established for a specific pIRIR signal for both single aliquots of K-feldspar (Li et al., 2015b) and individual K-feldspar grains.

6. D_e estimation using a single-grain gSGC

6.1 Comparison of gSGC and SAR D_e estimates

The existence of a gSGC for individual K-feldspar grains allows D_e estimation without constructing a full SAR growth curve for each grain. Following Li et al. (2015a), SGC D_e estimation is achieved by re-scaling the L_n/T_n ratio using a scaling factor determined from the gSGC and an additional L_x/T_x ratio. The procedure can be described using the following equation:

$$f(D_e) = \frac{L_n}{T_n} \times \frac{f(D_r)}{L_r/T_r} \quad (8)$$

where $f(x)$ denotes the gSGC obtained by LS-normalisation, and D_r and L_r/T_r are the additional regenerative dose and the sensitivity-corrected ratio for the corresponding pIRIR signals, respectively.

To test the reliability of D_e estimates for different samples using the gSGC, we compared the D_e values obtained using the gSGC presented in Fig. 8b with those determined from grain-specific, full SAR growth curves. We used the same rejection criteria for calculating the SAR D_e values as those used above for selecting growth curves for LS-normalisation. We also rejected D_e values with relative

standard errors of >50%, although including these imprecise values has a negligible effect on the weighted mean D_e estimates. The results of the comparison are presented in Fig. 9 for all accepted grains ($n = 1953$) from the 28 samples in Fig. 8. The gSGC and SAR D_e values are clustered around the 1:1 line across two orders-of-magnitude of dose (Fig. 9a), with 99% of the grains having ratios of gSGC to SAR D_e values consistent with unity at 2σ . This outcome confirms that the gSGC developed for the samples in this study can produce reliable D_e estimates for individual grains — as accurate, at least, as those obtained using the full SAR procedure.

This comparison between SAR and gSGC D_e values is only possible for grains that yielded reliable SAR results. Several of the rejection criteria applied to the SAR data cannot be applied to the gSGC data, owing to the lack of information about recuperation (zero-dose response), recycling ratio and scatter of L_x/T_x data for each grain (i.e., FOM and RCS values). As a result, some of the grains rejected from the SAR data set are included in the gSGC data set, which could give rise to some inaccurate single-grain D_e estimates using a gSGC. Fig. 10 is a radial plot of all accepted SAR D_e values (filled circles) and gSGC D_e values (open triangles) for sample Chag12-1. Compared to the number of grains accepted for SGC analysis (210 out of 600 grains), 17 grains were further identified as ‘poorly behaved’ and rejected from the SAR data set. The SAR and gSGC D_e distributions are, nonetheless, indistinguishable and their weighted mean D_e values — calculated using the central age model (CAM: Galbraith et al., 1999; Galbraith and Roberts, 2012) — are statistically consistent (SAR: 132 ± 4 Gy; gSGC: 140 ± 6 Gy). The inclusion of ‘poorly behaved’ grains in the gSGC data set appears to have a negligible impact on the final D_e estimate for this sample. However, it is worth to mention here that the extent of the effect will depend on the fraction of ‘poorly behaved’ grains and the D_e from these grains in the samples studied. Hence, a comparison between the distributions of the SAR and SGC D_e values are necessary to validate the SGC results.

6.2 D_e estimation using gSGC and re-normalised L_n/T_n ratios

The SGC method can also overcome problems associated with D_e underestimation for aliquots and grains with natural doses close to saturation (Li et al., 2017). Previous studies (e.g., Duller, 2012; Gliganic et al., 2012; Li et al., 2016; Guo et al., 2017) have found that rejection of ‘early saturated’ grains may result in truncated D_e distributions and underestimation of the final D_e . Li et al. (2017) demonstrated, through numerical simulation, that uncertainties associated with σ_{cs} and σ_{ins} can give rise to L_n/T_n ratios that are consistent with, or lie above, the saturation level of the corresponding measured growth curve — even if all of the aliquots or grains share the same growth curve and D_0 value — resulting in truncated D_e distributions. They proposed a method for D_e determination based on the full (‘untruncated’) distribution of L_n/T_n ratios for all aliquots or grains, and showed that it can overcome the problem of D_e underestimation for old samples; it can also produce reliable D_e estimates up to 4–5 times the D_0 value, which is well beyond the $2D_0$ limit often applied to D_e values obtained using conventional SAR procedures. Accordingly, we tested the method of Li et al. (2017) on the samples investigated here, in order to maximise the benefits of the single-grain gSGC for K-feldspar.

Fig. 11a shows the SAR D_e values for the 72 accepted grains of Chag12-9. For this sample, 326 grains passed the ‘weak T_n ’ criterion (Table 3), but 143 of them (~44%) have L_n/T_n ratios consistent with, or higher than, the saturation level of the corresponding single-grain growth curves, so finite D_e values could not be determined. The CAM D_e value for these 72 grains is 569 ± 25 Gy, with the individual D_e values appearing to be randomly distributed around this central value (Fig. 11a). Given the large number of grains rejected due to ‘saturation’, however, we consider this D_e estimate to be an underestimate of the true sample D_e . As a test, we measured six single aliquots of Chag12-9 using the ‘pre-dose’ multiple elevated temperature pIRIR (pMET-pIRIR) procedure, which is capable of measuring higher D_e values than is feasible using conventional IRSL and pIRIR methods (Li et al., 2014b; Guo et al., 2015, 2016). Five successive infrared stimulations were made at 50, 100, 150, 200 and 275 °C. A preheat of 320 °C for 60 s was given before measuring the L_n , T_n , L_x and T_x signals. The weighted mean D_e values are plotted as a function of stimulation temperature in Fig. 11b.

In the absence of a D_e ‘plateau’ for this sample, we used the value obtained at the highest stimulation temperature (275 °C) as the final D_e (1116 ± 51 Gy). This estimate is twice that of the weighted mean D_e determined from the single-grain SAR D_e values, confirming our proposition that the latter D_e distribution is significantly truncated.

The method proposed by Li et al. (2017) involves re-normalising the individual L_n/T_n ratios — dividing them by the L_x/T_x ratios for one of the regenerative doses applied during the SAR procedure; we used the 537 Gy regenerative dose for re-normalisation. Fig. 11c shows the distribution of re-normalised L_n/T_n ratios for the 335 grains that passed the ‘weak T_n ’ criterion. The results for the 72 grains accepted for D_e determination using the full SAR procedure are shown as filled (black) circles, and the ratios for the 263 rejected grains are displayed as open (red) circles. The latter have re-normalised L_n/T_n ratios systematically greater than those of the accepted grains, which explains why the sample D_e based only on the accepted grains is significantly underestimated.

To obtain a more accurate estimate of sample D_e , we re-scaled the L_n/T_n values for all 335 grains using the L_r/T_r ratios at $D_r = 537$ Gy for re-normalisation. Following Li et al. (2017), the re-scaling is based on the following equation:

$$\frac{L'_n}{T'_n} = \frac{L_n}{T_n} \times \frac{f(D_r)}{L_r/T_r} \quad (9)$$

where L'_n/T'_n denotes the re-scaled L_n/T_n ratio, and $f(x)$ denotes the single-grain gSGC. The individual L'_n/T'_n ratios for Chag12-9, together with the gSGC, are shown in Fig. 11d. The weighted mean L'_n/T'_n ratio was estimated using the CAM, and this was projected (blue horizontal line) on to the gSGC (red line). The resulting D_e estimate (1244 ± 86 Gy) is consistent with that obtained from the single-aliquot pMET-pIRIR signal stimulated at 275 °C (Fig. 11b). This finding confirms that the relatively large measurement uncertainties associated with single-grain measurements may give rise to D_e underestimates using conventional SAR procedures when large numbers of grains are rejected as

‘saturated’; the same caveat also applies to single aliquots that contain a high proportion of such grains.

7. Discussion

The K-feldspar data presented in Fig. 5 and 7 show that variation in the shape of growth curves for different grains from the same sample is much larger than that observed for single aliquots (Li et al., 2015b). This is to be expected, as single-grain signals are associated with larger measurement uncertainties, arising from σ_{cs} and σ_{ins} . The uncertainty from σ_{cs} depends on the particular sample measured and instrument used (see section 4). For a sample containing a large number of dim grains, σ_{cs} may play a significant role in the observed variability of L_n/T_n and L_x/T_x ratios; and the variability will be even larger for instruments with greater-than-expected variance in k_{DC}^2 and k_{ph}^2 . For samples with many bright grains (e.g., Sm5), the main source of variability is σ_{ins} . Based on numerical simulation, Li et al. (2017) found that, assuming a σ_{ins} value of 2–4% per measurement, the observed D_0 values for individual grains could vary by more than two orders-of-magnitude, even though they had been assigned the same true D_0 value. Our experimental results likewise show that individual grains appear to saturate at different doses (Fig. 5), but these differences are reduced significantly after LS-normalisation (Fig. 7b and 8b).

We have demonstrated that more than 90% of the K-feldspar grains measured in this study share a common growth curve (Fig. 8), despite originating from a range of geological and environmental contexts, and exhibit considerable differences in pIRIR sensitivities (Fig. 4) and scatter in L_x/T_x ratios (Fig. 5). Our single-grain results also confirm the results from Li et al. (2015b), who observed that a gSGC could be established for single aliquots of K-feldspar extracted from a variety of samples, although that study used a different stimulation source (infrared LEDs) and different preheat and stimulation temperatures. We expect, therefore, that the gSGC constructed in the present study will also be applicable to other single grains of K-feldspar measured using the same procedure. We endorse the recommendation of Li et al. (2015a, 2015b) that some of the grains from each sample

and site should be measured routinely using a full SAR procedure, to confirm that their growth curves have the same shape as the gSGC established here before applying the gSGC method to a large number of samples. It is worth noting that, as growth curve shapes can differ significantly for pIRIR signals measured at different stimulation temperatures (Li et al., 2014a), it is necessary to construct SGCs specific to each set of measurement conditions.

We have shown that single-grain D_e values based on the LS-normalised gSGC are consistent with those obtained from individual SAR growth curves (Fig. 9), thereby confirming the gSGC as a robust and rapid means of determining D_e values for single grains of K-feldspar. This method is particularly advantageous when a large number of grains need to be measured or when dating older samples that require large regenerative doses. A drawback of the method is the loss of information about recuperation and sensitivity correction (recycling), which may result in more dispersed D_e values due to the inclusion of grains that would ordinarily be rejected using the full SAR procedure. Therefore, we recommend that a comparison between the distributions of the SAR and SGC D_e values using some of the samples should be conducted to validate the SGC results before the application of the SGC method to the other samples. Finally, the existence of a SGC or gSGC does not automatically imply that reliable age estimates will be obtained. To validate the ages, dose recovery (Roberts et al., 1999), anomalous fading and residual dose tests should be performed and comparisons made, if possible, with independent chronological controls (Li et al., 2015b).

8. Conclusions

The pIRIR signals from single grains of K-feldspar exhibit large variations in the experimentally observed growth curves. Such variation results mainly from measurement uncertainties, which can be significantly reduced by applying a LS-normalisation procedure. We established a gSGC for individual K-feldspar grains from sedimentary deposits in a variety of geological regions and environmental settings. The gSGC D_e values are statistically consistent with those obtained from full SAR growth curves constructed for each grain. Use of the gSGC greatly

reduces the time required to measure a large number of grains, and is particularly useful for dating single grains of K-feldspar with large D_e values. The gSGC procedure also helps to overcome problems of D_e underestimation associated with truncated D_e distributions for samples with natural doses close to saturation, and has the potential to extend the time range of optical dating far beyond the limit of standard SAR procedures.

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Figure captions

Figure 1: Histograms of normalised L_x/T_x ratios for individual grains from samples Chag12-1 (a) and Sm5 (b), measured using Risø3 and Risø5, respectively. The L_x/T_x ratios of individual grains have been normalised using the weighted mean L_x/T_x ratio for each grain.

Figure 2: Examples of repeated measurements for ‘poorly-behaved’ (a, b and c) and ‘well-behaved’ (d) grains from sample DCE14-1. The inset in each panel shows the corresponding dose response data for the same grain.

Figure 3: The chi-squared values for all of the grains (a) and ‘well-behaved’ grains (b) from sample Chag12-1. The red curves show the theoretical distributions for $n-1$ (i.e., 8) degrees of freedom. The black, red and green vertical lines denote the values for the 68%, 95% and 98% percentiles of the theoretical distributions.

Figure 4: Cumulative light sum plots (a) and single-grain T_n intensities (b) for samples DCM12-26, LC10-05, RUP-4 and Sm5, plotted against the percentage of grains ranked according to brightness.

Figure 5: Representative pIRIR decay curves (left-hand column) and corresponding growth curves (right-hand column) for six individual grains from each of samples DCM12-26 (a and b), LC10-05 (c and d), RUP-4 (e and f) and Sm5 (g and h). The insets in (a), (c), (e) and (g) show the decay curves normalised to unity at the start of stimulation.

Figure 6: Example growth curves for four grains from sample Sm5 with different FOM and RCS values, showing the test-dose standardised ratio plotted as a function of dose.

Figure 7: Test-dose standardised ratios for all accepted grains from sample Chag12-9 (a), and the same data after LS-normalisation (b). The solid lines in (a) and (b) are the best-fit curves to all data points, shown in a variety of colours to distinguish different grains. (c) Ratio between the LS-normalised data and the best-fit gSGC in (b), plotted against regenerative dose. (d) Radial plot of the ratios shown in (c); the shaded band of ± 2 standardised estimates captures 94.3% of these ratios, which closely matches the proportion (95%) expected for ratios consistent with unity at 2σ .

Figure 8: Test-dose standardised ratios for all accepted grains from 28 samples (a), and the same data after LS-normalisation (b). The solid lines in (b) are the best-fit curves to all data points, shown in a variety of colours to distinguish different samples. (c) Ratio between the LS-normalised data and the best-fit gSGC in (b), plotted against regenerative dose. (d) Radial plot of ratios shown in (c); the shaded band of ± 2 standardised estimates captures 91% of these ratios.

Figure 9: (a) Comparison of single-grain D_e values estimated from the gSGC and full SAR growth curves for all grains accepted after application of both sets of rejection criteria. Errors bars are shown at 1σ on both axes, and the solid line indicates the 1:1 relationship between the gSGC and SAR D_e values. (b) Radial plot of the gSGC and SAR D_e values shown in (a), converted to ratios; the shaded band captures 99% of these ratios.

Figure 10: Radial plot of accepted single-grain SAR D_e values (filled circles) and SGC D_e values (open triangles) for sample Chag12-1.

Figure 11: (a) Radial plot of accepted single-grain SAR D_e values for sample Chag12-9. (b) Weighted mean D_e values obtained from pMET-pIRIR L_x/T_x ratios for six single aliquots of the same sample, plotted as a function of infrared stimulation temperature. (c) Re-normalised L_n/T_n ratios (L'_n/T'_n) for the 335 grains from sample Chag12-9 that passed the ‘weak T_n ’ criterion: the 72 accepted grains and 263 rejected grains are shown as filled (black) and open (red) circles, respectively. (d) L'_n/T'_n ratios for individual grains (blue squares on y-axis) and gSGC curve (red line) shown in Fig. 9b. The horizontal dashed (blue) line represents the weighted mean L'_n/T'_n ratio calculated using the CAM.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Table 1: Single-grain pIRIR measurement procedure.

Step	Treatment	Observed
1 ^a	Give regenerative dose, D_i	
2	Preheat at 320 °C for 60 s	
3	IR diodes stimulation at 200 °C for 200 s	$L_{x(200)}$
4	IR laser stimulation at 275 °C for 1.5 s	$L_{x(275)}$
5	Give test dose, D_t	
6	Preheat at 320 °C for 60 s	
7	IR diodes stimulation at 200 °C for 200 s	$T_{x(200)}$
8	IR laser stimulation at 275 °C for 1.5 s	$T_{x(275)}$
9	IRSL measurement at 325 °C for 100 s	
10	Return to step 1	

^a For the natural sample, $i = 0$ and $D_i = 0$ Gy, and the observed signals are denoted as L_n and T_n . The entire sequence is repeated for several regenerative doses, including a zero dose and a repeat dose.

Table 2: Summary of samples measured, repeat regenerative doses given, and the number of grains accepted for estimating the instrumental irreproducibility (σ_{ins}) for each of two Risø readers estimated using both Method 1 and Method 2 (see section 4).

Reader	Sample	Repeat dose (Gy)	Total number of grains	Method 1	Method 2		
					σ_{ins} based on all grains (%)	No. of well-behaved grains	σ_{ins} based on well-behaved grains
Risø3	Chag12-1	100	92	2.8	4.3	75	2.6
Risø3	DCE14-1	100	110	2.1	4.6	91	2.0
Risø3	DCS-1	100	54	2.5	3.9	45	1.7
Risø3	SM-5	100	83	3.4	5.2	54	2.4
Risø5	Chag12-1	200	94	2.9	4.0	81	2.8
Risø5	DCE-4	900	70	1.3	2.5	66	1.9
Risø5	DCE-4	300	70	1.6	2.2	66	1.6
Risø5	DCE-4	600	73	0.6	1.9	70	1.5
Risø5	LC10-05	100	40	3.1	4.2	37	3.1
Risø5	SM-5	50	99	2.5	3.1	94	2.8

Table 3: Number of individual K-feldspar grains measured, rejected and accepted for LS-normalisation, and the reasons for rejection: T_n intensity, extent of recuperation, figure-of-merit (FOM) and reduced-chi-square (RCS) values.

Sample name	No. of grains measured	Weak T_n signal ^a	Recuperation >10% ^b	FOM > 10% or RCS >5	No. of accepted grains
BJZ-OSL1	100	65	3	9	23
BJZ-OSL3	400	226	37	48	89
Chag12-1	600	307	10	54	229
Chag12-10	200	76	3	37	84
Chag12-6	100	65	1	11	23
Chag12-9	600	265	7	48	280
DCE12-12	600	515	0	17	68
DCE12-14	600	552	0	9	39
DCE12-3	200	56	1	49	94
DCE12-4	600	304	2	67	227
DCM12-26	500	421	0	28	51
DCM12-27	500	426	0	12	62
DCM12-3	500	315	1	31	153
DCM12-5	400	259	0	21	120
DCS12-5	400	252	0	38	110
Enderby-OSL1	600	584	1	6	9
Enderby-OSL2	600	560	2	20	18
Enderby-OSL3	300	290	2	6	2
Enderby-OSL4	500	440	2	36	22
HF-10	600	417	17	56	110
HF-8	600	412	19	79	90
LC10-05	400	344	42	4	10
RUP-1	500	447	10	8	35
RUP-2	500	419	14	26	41
RUP-3	500	417	12	21	50
RUP-4	500	426	6	27	41
RUP-5	500	458	5	15	22
Sm5	400	34	58	60	248

^a Initial T_n signal is within 3σ above the corresponding background count, or the relative error on T_n is >25%.

^b Recuperation is defined here as the ratio of the zero-dose to the maximum regenerative-dose pIRIR signals.