Optimising the reproducibility of measurements of the post-IR IRSL signal from single-grains of K-feldspar for dating

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Abstract

The reproducibility of the Risø single-grain measurement system has previously been quantified for the analysis of individual grains of quartz using the green laser and for single-grains of K-feldspar using the infrared (IR) laser at 50 °C. However, reproducibility estimates for a single-grain measurement system analysing K-feldspar grains using the post-IR IRSL (pIRIR) signal do not exist. This study provides the first measurement reproducibility estimates for both the $pIRIR_{225}$ and $pIRIR_{290}$ protocol using an IR laser. It is found that holding a sample at elevated temperatures (e.g. 225°C or 290°C) prior to measurement leads to loss of the pIRIR signal. The default single grain procedure implemented by the Risø reader may involve holding the sample at this elevated temperature for periods up to several hundreds of seconds, and that crucially this time may vary from one measurement to another, leading to poorer measurement reproducibility. The study demonstrates that the measurement procedure can be modified to standardise the time spent at high temperature (e.g. 290°C) and hence improve the reproducibility of the measurement system. The optimised procedure provides reproducibility estimates of 2.8 ± 0.3 % and 2.6 ± 0.3 % for the $pIRIR_{225}$ and $pIRIR_{290}$ signal, respectively, which are comparable to similar measurements performed with the green laser and the IR laser at 50 °C.

Introduction

Optically stimulated luminescence (OSL) dating with single-grains is a valuable approach in depositional environments where grains are likely to be incompletely-bleached. Single-grain dating involves analysing individual mineral grains (e.g. quartz or feldspar) to provide natural dose-distributions, which can then be statistically modelled to determine the true burial age. A major challenge for routine single-grain dating of sedimentary quartz from incompletely-bleached sediment is that typically only 5 % or fewer of the grains emit a detectable OSL signal, e.g. as few as 0.5 % of quartz grains could be detected from glaciofluvial sediments from Chile (Duller, 2006). In contrast to quartz, a larger proportion of K-feldspar grains emit a detectable infrared stimulated luminescence (IRSL) signal (e.g. Duller et al. 2003). However, the IRSL signal of K-feldspars measured at ~50 °C (IR_{50}) is reported to ubiquitously suffer from anomalous fading, and therefore may require fading-correction to provide an accurate depositional age (Huntley and Lamothe, 2001). Thomsen et al. (2008, 2011) extensively studied the fading rates of feldspars in response to different stimulation and detection conditions, and suggested that a more stable post-IR IRSL (pIRIR) signal can be accessed within feldspar grains using an initial IRSL stimulation at 50 °C followed by an elevated temperature IRSL stimulation, typically performed at 225 °C or 290 °C, hereafter termed the $pIRIR_{225}$ and $pIRIR_{290}$ signals. Given that large and variable fading rates are often reported for single-grain K-feldspars measured using the IR_{50} signal (e.g. Trauerstein et al. 2012), and also the difficulty in making accurate and precise fading measurements on individual grains, single-grain dating with K-feldspar would benefit from accessing the more stable pIRIR signal.

Measuring individual D_e values from single-grains normally results in more scatter in dose-distributions than is typical from multiple-grain analysis. Calculating the associated uncertainties in the D_e values for each grain requires knowledge of both the photon counting statistics and the reproducibility of the measurement system, though scatter may also arise from other factors that have not yet been identified. The reproducibility of the measurement system is dependent upon the thermal treatment, optical stimulation and material response for individual grains during measurement. Thus, measurement reproducibility is expected to vary between different readers used for single-grain measurements, the samples analysed, and between the IRSL and pIRIR signals used to measure K-feldspar grains. A challenge for IRSL analysis of K-

Step	**IR_{60}**	**$pIRIR_{225}$**	**$pIRIR_{290}$**
1	Dose (100 Gy)	Dose (100 Gy)	Dose (100 Gy)
2	Preheat 250°C for 60 s	Preheat 250°C for 60 s	Preheat 320ºC for 60 s
3	IR laser 2 s at 60°C	IR laser 2 s **or** LEDs 100 s at 60°C	IR laser 2 s **or** LEDs 100 s at 60°C
4	Test-dose (100 Gy)	IR laser 2 s at 225°C	IR laser 2 s at 290°C
5	Preheat 250°C for 60 s	Test-dose (100 Gy)	Test-dose (100 Gy)
6	IR laser 2 s at 60°C	Preheat 250°C for 60 s	Preheat 320ºC for 60 s
7	IR laser 2 s **or** LEDs 100 s at 290°C	IR laser 2 s **or** LEDs 100 s at 60°C	IR laser 2 s **or** LEDs 100 s at 60°C
8		IR laser 2 s at 225°C	IR laser 2 s at 290°C
9		IR laser 2 s **or** LEDs 100 s at 290°C	IR laser 2 s **or** LEDs 100 s at 330°C

Table 1: *Experimental details for the single-aliquot regenerative dose (SAR) pIRIR measurements performed throughout this study with the IR_{60}, $pIRIR_{225}$ and $pIRIR_{290}$ signals for single grains of K-feldspar. Note that the signal was measured for 0.15 s before and after the IR stimulation was performed so the IR laser was stimulating for a total duration of 1.7 s.*

feldspars is the thermal-dependence of the magnitude of the signal (Duller and Wintle, 1991; McKeever et al. 1997), which has the potential to make measurement of the pIRIR signal less reproducible than that of IRSL signal. Calculating the reproducibility of the measurement system is important so that the appropriate uncertainty is incorporated into D_e estimates. However, measurement reproducibility estimates are not currently available for the Risø single-grain K-feldspar system using the pIRIR signal. Estimates of the reproducibility of the single-grain measurement system only currently exist for sedimentary grains of naturally-occurring quartz analysed using the green laser (e.g. Truscott et al. 2000; Thomsen et al. 2005; Jacobs et al. 2006) and K-feldspar analysis using the IR laser at 50 °C (e.g. Trauerstein et al. 2012).

Truscott et al. (2000) used an early prototype of the single-grain system equipped with a green laser to perform repeated L_x measurements on sensitised quartz grains to calculate a measurement reproducibility of 3.5 % per stimulation. Jacobs et al. (2006) subsequently repeated the measurements of Truscott et al. (2000) on sensitised grains of quartz to calculate the reproducibility of an improved single-grain laser system. The calculated mean measurement reproducibility estimate was 2.6 % (range ~1 – 8 %) and 1.3 % (range ~ 0.5 – 5 %) for optical stimulation times of 0.04 s and 0.3 s, respectively. The reproducibility improved when a longer summation interval was used as the decay rate is controlled by the power of the laser and this may vary slightly for individual stimulations; summing a larger part of the decay curve reduces the impact of the variable laser power during individual stimulations. Thomsen et al. (2005) also measured the reproducibility of a similar single-grain system with repeated L_x/T_x sensitivity-corrected measurements and calculated a mean (± standard error) of 2.5 ± 0.3 % and 1.5 % per OSL measurement for 0.03 s and 0.57 s of optical stimulation, respectively, which is comparable to Jacobs et al. (2006). Finally, Trauerstein et al. (2012) adopted the approach of Thomsen et al. (2005) for single-grain analysis of K-feldspars using the IR_{50} signal and calculated an estimate of reproducibility of 2.4 % (1 s of optical stimulation).

The principle aim of this study is to calculate the reproducibility of single-grain measurements of K-feldspar using the pIRIR signal. In addition, this study also aims to assess whether the reproducibility of the measurement system using the pIRIR signal can be optimised by (1) reducing the temperature at which the single-grain disc is held during disc location from the elevated temperature (i.e. 225 °C or 290 °C) to room temperature, and (2) using IR light emitting diodes (LEDs) instead of the IR laser to perform the low temperature measurements (here made at 60 ºC) prior to the pIRIR measurements, and for bleaching grains at an elevated temperature at the end of each SAR cycle.

Experimental details

All luminescence measurements were performed using a Risø DA-15 automated TL/OSL single-grain system equipped with an IR laser (150 mW; 830 nm) (Bøtter-Jensen et al. 2003, Duller et al. 2003) at the Aberystwyth Luminescence Research Laboratory. The IR laser beam line was fitted with an RG-780 filter to remove any shorter wavelengths and a blue filter pack (Schott BG-39, GG-400 and Corning 7-59) was placed in front of the photomultiplier tube. The inclusion of the GG-400 filter is used to ensure complete removal of the thermally unstable UV transmission centred on 290 nm emitted during IR stimulation of feldspars (e.g. Balescu and Lamothe, 1992; Clarke and Rendell, 1997). The system was equipped with a $^{90}Sr/^{90}Y$ beta source delivering ~0.04 Gy/s. Table 1 outlines the protocols used for the three signals; IR_{60}, $pIRIR_{225}$ and $pIRIR_{290}$. Ten repeated

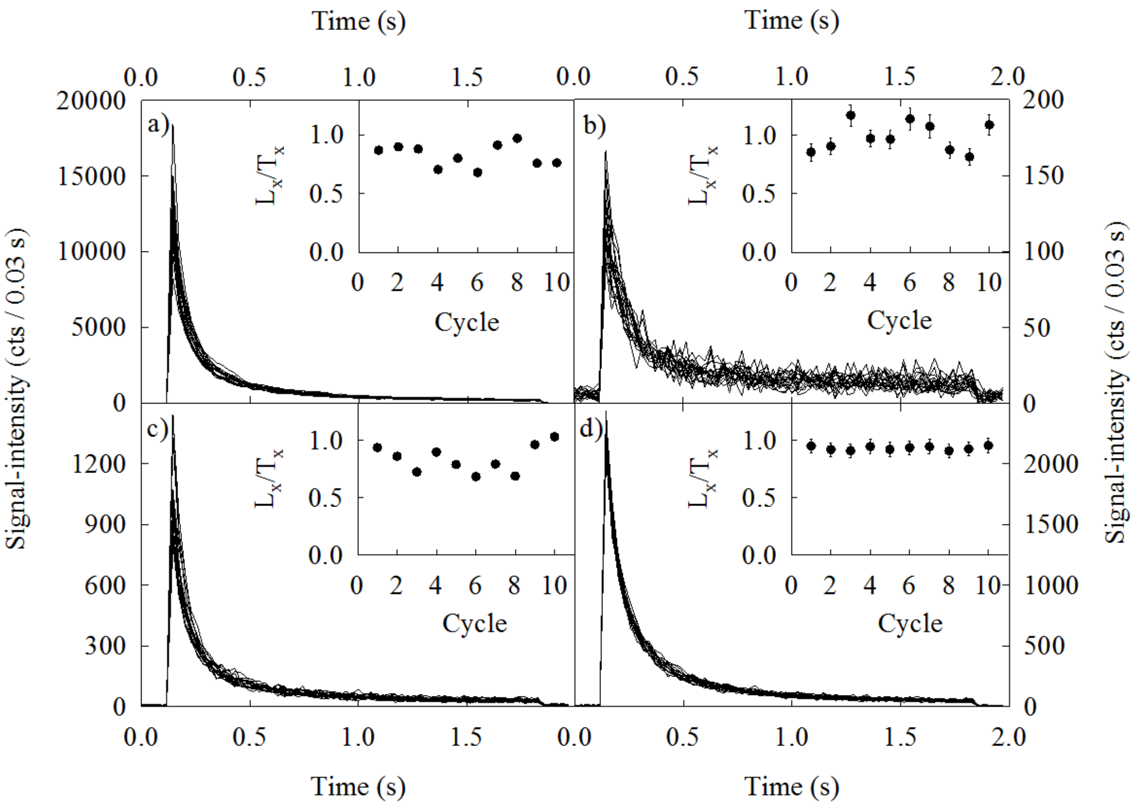

Figure 1: *Examples of the repeated L_x and T_x decay curves measured with the pIRIR$_{225}$ signal for individual grains of K-feldspar from GDNZ13 for a) the brightest grain (measurement reproducibility of 12.0 %), b) the dimmest grain (measurement reproducibility of 8.9 %), c) the grain with the largest measurement error estimate (measurement reproducibility of 18.7 %) and d) the grain with the smallest measurement error estimate (measurement reproducibility of 0.6 %). The inserts show the change in L_x/T_x for the repeated measurements. Note that the disc location was performed at the elevated temperature of 225 °C and the IR bleaching throughout the protocol was performed with the IR laser.*

L_x/T_x measurements were performed with the IR laser; 100 Gy doses were used to maintain an appropriate signal-intensity throughout analysis. The signal was recorded for a total of 2 s, which included the measurement of signal for 0.15 s before and after the IR stimulation was performed so the grains are stimulated using the IR laser for a duration of 1.7 s. The initial signal was summed over the first 0.3 s of stimulation and the background from the final 0.6 s. Grains were rejected if the test-dose uncertainty was greater than 10 %. The number of grains that fail this criterion varies for different measurement and analytical conditions, but this had no impact upon the patterns described in this study.

A sedimentary dune sand sample from New Zealand (GDNZ13) was used throughout this study. The sample was treated with a 10 % v.v. dilution of 37% HCl and 20 vols. of H_2O_2 to remove carbonates and organics, respectively. Dry sieving isolated the 180 – 212 µm diameter grains and density-separation provided the < 2.58 g cm^{-3} (K-feldspar) fraction. The K-feldspar grains were not etched in hydrofluoric acid. The grains were mounted in aluminium single-grain discs (a 10 x 10 grid of 300 µm holes). All the experiments were repeated on exactly the same suite of K-feldspar grains, which remained in the single-grain disc throughout the analysis. Prior to these measurements the grains had been heated up to 330 °C and thus were not expected to exhibit large changes in sensitivity during this sequence of measurements.

The uncertainty arising from the reproducibility of the measurement system was calculated by subtracting in quadrature the uncertainty arising from the counting statistics from the observed uncertainty in the repeated L_x/T_x measurements following Section 7.1 of Thomsen et al. (2005). The uncertainty arising from the counting statistics for each individual grain was calculated using the equations outlined in Section 3 of Thomsen et al. (2005). The reproducibility of a single L_x measurement was then calculated from this value by dividing by $\sqrt{2}$ (Section 7.1, Thomsen et al. 2005). Fig. 1 presents examples of the L_x and T_x decay curves produced from

Protocol	IR_{60} bleach	High temperature bleach	Disc location temperature	Mean ± Standard error (%)	Standard deviation (%)	Range (%)	'n'
IR_{60}	-	LEDs	60 °C	3.6 ± 0.3	2.4	0.3 – 10.0	49
$pIRIR_{225}$	Laser	Laser	225 °C	5.9 ± 0.5	3.7	0.6 – 18.7	49
$pIRIR_{225}$	Laser	Laser	60 °C	4.7 ± 0.4	3.1	0.5 – 14.6	48
$pIRIR_{225}$	**LEDs**	**LEDs**	**60 °C**	**2.8 ± 0.4**	**2.8**	**0.3 – 10.9**	**45**
$pIRIR_{290}$	Laser	Laser	290 °C	4.7 ± 0.6	3.6	0.5 – 16.2	38
$pIRIR_{290}$	Laser	Laser	60 °C	3.6 ± 0.4	2.4	1.0 – 11.3	43
$pIRIR_{290}$	**LEDs**	**LEDs**	**60 °C**	**2.6 ± 0.3**	**1.8**	**0.2 – 8.2**	**36**

Table 2: *Measurement reproducibility estimates for the IR_{60}, $pIRIR_{225}$ and $pIRIR_{290}$ signals measured using the single grain system.*

repeated single-grain $pIRIR_{225}$ measurements. For these measurements the disc location was performed at the elevated temperature of 225 °C prior to the stimulation at 225 °C. The default setting of the Risø system is for the disc location to be performed at whatever temperature the optical stimulation will occur. All of the data shown in Fig. 1 was collected using a sequence in which the IR laser was used for the measurements at 60 °C prior to the pIRIR measurement, and the IR laser was used for the elevated temperature bleaching at the end of each cycle (step 9 in Table 1). The examples shown in Fig. 1 include a) the brightest grain (12.0 % measurement reproducibility) and b) the dimmest grain (8.9 % measurement reproducibility) from the single-grain population, in addition to the grains with c) the largest (18.7 %) and d) the smallest (0.6 %) measurement reproducibility estimates calculated from the single-grain population.

Table 2 presents the mean and standard error measurement reproducibility estimates for the different protocols employed, including the range of estimates for individual grains. The range of IRSL reproducibility estimates in this study (0.3 – 10.0 %) covers a similar range to those obtained by Jacobs et al. (2006) for quartz using the green laser and 0.04 s of optical stimulation (~1 – 8 %). However, the mean IR_{60} reproducibility estimate when the signal is summed over 0.3 s of optical stimulation (3.6 ± 0.3 %) is larger than the 2.4 % (1 s of optical stimulation) published by Trauerstein et al. (2012). When a longer summation interval is used (2 s of optical stimulation) then the IR_{60} reproducibility becomes 2.6 ± 0.4 %, almost identical to that of Trauerstein et al. (2012) because the effects from the reproducibility of the laser have been removed. Initial estimates of the measurement reproducibility for $pIRIR_{225}$ and $pIRIR_{290}$ single-grain measurements prior to making any alterations to the protocol to optimise the reproducibility were 5.9 ± 0.5 % and 4.7 ± 0.6 %, respectively (Table 2). Both estimates are larger than measurement reproducibility estimates presented here for the IRSL signal over comparable summation intervals (3.6 ± 0.3 %).

Optimising the reproducibility of the single-grain measurement system

Two aspects of the measurement procedure were modified in an attempt to optimise the reproducibility of the single-grain measurement system using the $pIRIR_{225}$ and $pIRIR_{290}$ signals; (1) reducing the temperature at which each disc is held during disc location prior to stimulation with the IR laser from the stimulation temperature (i.e. 225 °C or 290 °C) to room temperature, and (2) replacing the IR laser with the IR LEDs to perform the 60 ºC measurements and the bleaches at an elevated temperature.

Reducing the temperature during disc location

The three locating holes present on each single-grain disc allow the Risø single-grain system to locate the exact position of the single-grain disc throughout the analysis (Duller et al. 1999). For single-grain measurements the software as installed by Risø is currently configured to heat the single-grain disc to the elevated temperature required for optical stimulation (i.e. 225 °C or 290 °C for the pIRIR protocols) prior to disc location, e.g. the single-grain disc is held at the elevated temperature whilst the system locates the exact position of the disc; a process which can take up to ~200 s and may vary throughout the sequence.

The reason why the disc is heated to whatever temperature is going to be used for optical stimulation before disc location occurs is that it was feared that heating of the disc may cause the disc to rotate (Thomsen, Pers. Comm.). If this occurred after disc location then the disc co-ordinates may be

a) Default tlmsll.cmd script	b) Modified tlmsll.cmd script
[SGOSL]	[SGOSL]
; $1 Start Grain	; $1 Start Grain
; $2 Stop Grain	; $2 Stop Grain
; $3 Time	; $3 Time
; $4 Total Datapoints	; $4 Total Datapoints
; $5 Rate	; $5 Rate
; $6 PreHeat Temp	; $6 PreHeat Temp
; $7 Preheat Time	; $7 Preheat Time
; $8 Laser Power	; $8 Laser Power
; $9 Delay Before	; $9 Delay Before
; $10 Delay After	; $10 Delay After
; $11 Active Data points	; $11 Active Data points
; $12 LightSource	; $12 LightSource
5=PS $0	5=PS $0
10=#RS	10=#RS
15=#WLT	15=#WLT
20=LU	20=LU
25=#RS	25=#RS
30=LV OFF	30=LV OFF
35=ST $6 $5	40=#FD $12 $0
40=#RS	50=#RS
42=PA $7	55=LV ON
45=#RS	60=#RS
48=#FD $12 $0	62=ST $6 $5
50=#RS	64=#RS
55=LV ON	66=PA $7
60=#RS	68=#RS
75=LA SET $8	75=LA SET $8
80=LI SET $8	80=LI SET $8
85=#LOOP $1 $2	85=#LOOP $1 $2
90=#INITGRAPH $4	90=#INITGRAPH $4
95=#SG #LOOPCOUNT $12 $3 $4 $11 $10 $9	95=#SG #LOOPCOUNT $12 $3 $4 $11 $10 $9
100=#DATA	100=#DATA
105=#RS	105=#RS
110=#ENDGRAPH	110=#ENDGRAPH
115=#SAVE	115=#SAVE
120=#ENDLOOP	120=#ENDLOOP
125=#APPEND	125=#APPEND
130=LD	127=ST 0
135=#RS	130=LD
	135=#RS

Table 3: *The default (a) and modified (b) tlmsll.cmd scripts from the latest version of TL/OSL sequence editor.*

incorrect and this would affect the ability of the laser to accurately strike the grains during IRSL measurements.

In this study, the default command script (tlmsll.cmd) originally installed with the software has been modified to undertake disc location at room temperature. Only once the position of the disc has been determined is the single-grain disc heated to the required temperature for the pIRIR measurement (i.e. 225 °C or 290 °C for the pIRIR protocols); this ensures that the period of time spent at elevated temperature is consistent from one set of single grain measurements to the next. The default tlmsll.cmd script as installed by Risø and the modified tlmsll.cmd script of this study are presented in Table 3a and b, respectively. The key difference in the modified script is that the Find Disc (FD) command is now undertaken first (line 40) before raising the hotplate temperature (ST) to the desired measurement temperature (line 62).

Experiments were performed using the $pIRIR_{225}$ and $pIRIR_{290}$ signal on exactly the same suite of grains to assess the effect of reducing the disc location temperature from the elevated temperature (i.e. 225 °C or 290 °C) to room temperature. Table 2 presents the mean and standard error, and range in measurement reproducibility estimates for the different single-grain populations. The $pIRIR_{225}$

measurement reproducibility fell from 5.9 ± 0.5 % to 4.7 ± 0.4 % when the disc location was performed at 225 ºC and room temperature, respectively, while the $pIRIR_{290}$ measurement reproducibility fell from 4.7 ± 0.6 % to 3.6 ± 0.4 %. Reducing the disc location temperature improved the mean measurement reproducibility by ~3 % for both signals (when subtracted in quadrature). Figure 2 presents the cumulative number of grains as a function of measurement reproducibility for the $pIRIR_{225}$ (circles) and $pIRIR_{290}$ (triangles) protocols using the disc location temperature of 225 °C or 290°C (closed, solid line) and room temperature (open, dashed line). The corresponding single-grain populations are shown in the histograms for the $pIRIR_{225}$ (top) and $pIRIR_{290}$ (bottom) signals. The number of grains with measurement reproducibility estimates ≤ 2 % increases from 10 % to 16 % for the $pIRIR_{225}$ protocol and from 20 % to 30 % for the $pIRIR_{290}$ protocol when the disc location temperature is reduced to room temperature. The data shown here demonstrate that the reproducibility of the single-grain measurement system using the $pIRIR_{225}$ and $pIRIR_{290}$ signals of K-feldspar grains improves by reducing the disc location temperature to room temperature.

Why does the reproducibility improve when the disc location temperature is reduced?

The mean signal-intensity of all the single-grain K-feldspars on the single-grain disc was calculated for the sequences when disc location was performed at the elevated temperature and at room temperature. When the disc location temperature was reduced from the elevated temperature to room temperature the mean measured signal-intensity increased by ~23 % and ~34 % for the $pIRIR_{225}$ and $pIRIR_{290}$ signals, respectively. The lower mean signal-intensity measured when using an elevated disc location temperature suggests that the pIRIR signal was thermally depleted throughout the period of time that it takes the single-grain measurement system to locate the disc. This thermal depletion would not occur if the disc was located at room temperature and could potentially explain the associated improvement in reproducibility. Additional experiments were performed to investigate whether thermal depletion of the pIRIR signal during disc location at elevated temperatures can explain the improvement in the reproducibility of the single-grain K-feldspar measurements.

Experimental details

To assess whether loss of the pIRIR signal could be observed due to holding the sample at elevated temperatures a multiple-grain aliquot of K-feldspar from sample GDNZ13 that had previously been

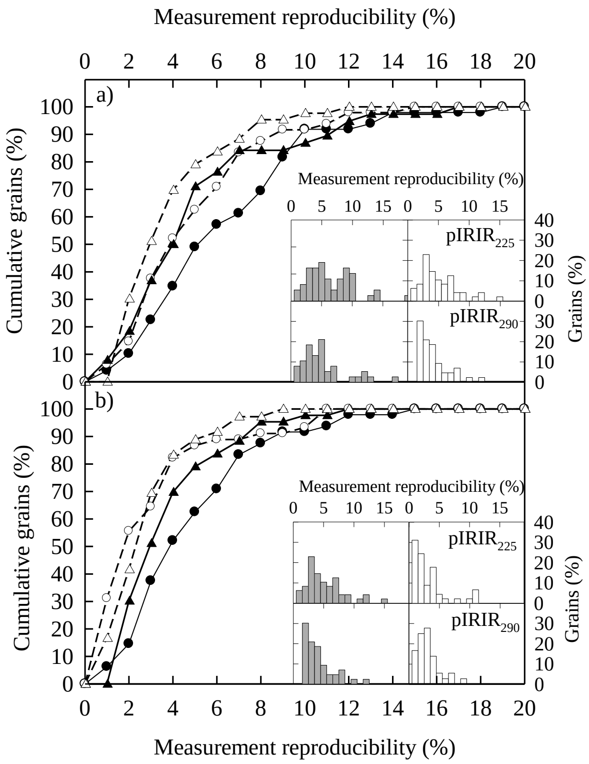

Figure 2: *Measurement reproducibility as a function of cumulative grains for the experiments performed to optimise the reproducibility of the $pIRIR_{225}$ (circles) and $pIRIR_{290}$ (triangles) single-grain K-feldspar measurements. The histograms show the corresponding $pIRIR_{225}$ (top) and $pIRIR_{290}$ (bottom) single-grain populations. Data are shown (a) comparing measurements made with disc location at an elevated temperature (solid line; filled histogram) with those made at room temperature (dashed line, open histogram), and (b) comparing measurements made undertaking the IR bleach in the pIRIR protocol with the focussed IR laser (solid line; filled histogram) to those undertaken with IR LEDs (dashed line; open histogram).*

bleached using IR LEDs and heated up to 330 °C was subject to two experiments. Tables 4a and 4b describe the measurement sequences used for experiment 1 (pulsed stimulation using the IR LEDs) and experiment 2 (continuous stimulation using the IR LEDs), respectively. In both experiments any remnant charge was removed in step 1. The aliquot was then given a dose of 100 Gy (step 2) and preheated (step 3) using the same procedure (320 °C for 60 s) as that used normally for $pIRIR_{290}$ measurements, and used in Table 1. In experiment 2 the $pIRIR_{290}$ signal was measured continuously for 5 s, collecting data every 0.1 s, resulting in 50 data points. In experiment 1, 50 $pIRIR_{290}$ measurements were also performed, but they were carried out over a

Step	a) Pulsed IR LEDs	b) Continuous IR LEDs
1	IRSL at 330 °C for 100 s	IRSL at 330 °C for 100 s
2	100 Gy beta dose	100 Gy beta dose
3	TL 320 °C for 60 s	TL 320 °C for 60 s
4	IRSL at 60 °C for 100 s	IRSL at 60 °C for 100 s
5	Record TL signal as heating up to 290 °C	IRSL at 290 °C for 5 s
6	IRSL at 290 °C for 0.1 s every 10 s for 500 s	

Table 4: *Experimental details for the multiple-grain K-feldspar experiments performed in this study.*

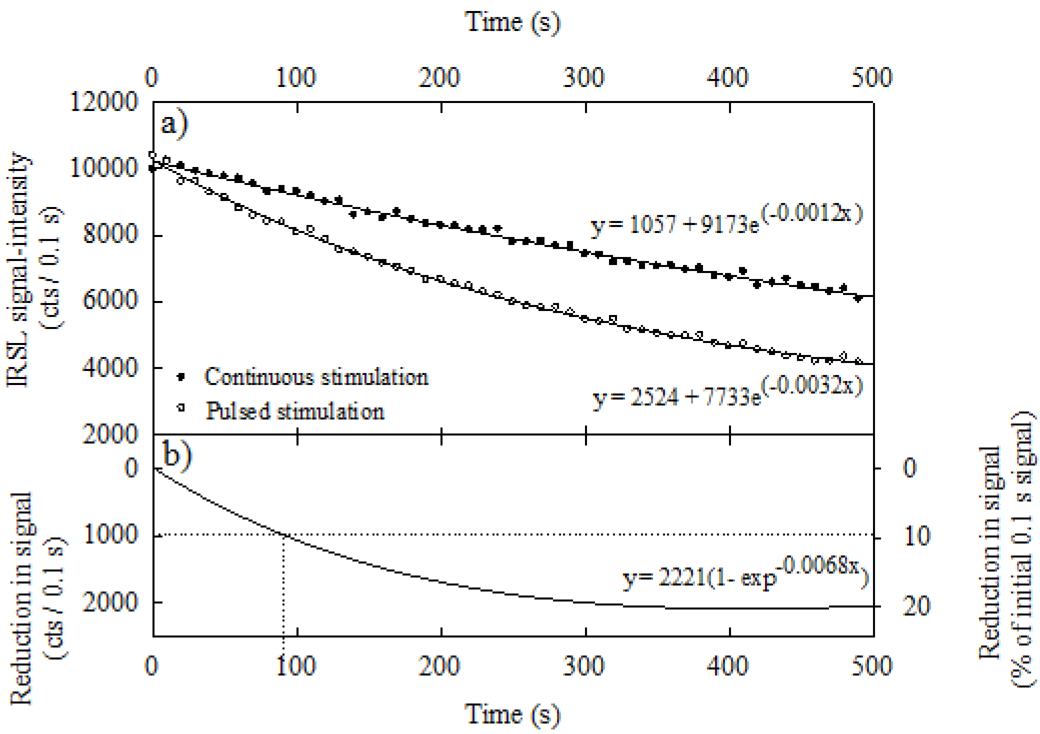

Figure 3: *a) The pIRIR signal recorded during the pulsed 0.1 s IR LED measurements every 10 s for 500 s (Table 4, step 6a) and the continuous IR LED measurement (Table 4, step 5b). Both datasets have been fitted with an exponential function. b) The exponential fit from the pulsed IR LED measurements as presented in Fig. 3a is subtracted from the continuous IR LED measurement to determine the reduction in signal that can be attributed to the effects of the prolonged heating at 290 °C. The reduction in signal is presented in absolute counts (left y-axis) and as a percentage of the signal in the first 0.1 s of IR stimulation (right y-axis). The dashed line marks the calculated reduction in signal (1,016 cts / 0.1 s, 10 %) after holding the disc for 90 s at 290 °C as an example of what is typical during routine dating measurements.*

period of 500 s. In both cases it is expected that the pIRIR signal will decrease due to optical eviction of charge, but if thermal depletion is also significant then the pulsed pIRIR data set collected over a longer period of time should show a greater decrease in intensity.

Reduction in signal-intensity when grains are held at an elevated temperature

The $pIRIR_{290}$ signals measured for the multiple-grain aliquot during the pulsed (experiment 1) and continuous (experiment 2) stimulation measurements are shown in Fig. 3a; each dataset is fitted with an exponential function. The reduction in signal attributed to holding the multiple-grain aliquot at an elevated temperature was determined by subtracting the reduction in the $pIRIR_{290}$ signal measured during the continuous stimulation (experiment 2) from the reduction in the $pIRIR_{290}$ signal measured during the pulsed stimulation measurements (experiment 1). The subtracted data are presented in Fig. 3b as absolute counts (left y-axis) and as a percentage of the signal in the first 0.1 s of IR stimulation (right y-axis). Fig. 3b demonstrates that there is an exponential reduction

pIRIR measurement	Disc location time (min:sec)
L_n	03:44
T_n	01:12
L_x (0 Gy)	01:14
T_x (0 Gy)	02:06
L_x (24 Gy)	01:13
T_x (24 Gy)	01:13
L_x (48 Gy)	01:19
T_x (48 Gy)	01:11
L_x (96 Gy)	01:13
T_x (96 Gy)	01:14
L_x (0 Gy)	01:12
T_x (0 Gy)	01:13
L_x (24 Gy)	01:11
T_x (24 Gy)	01:12
Mean ± st. dev.	**01:12 ± 00:36**

Table 5: *Periods of time it took the single-grain measurement system to locate the single-grain disc during a typical dating sequence using the pIRIR signal.*

in signal caused by thermal depletion of the pIRIR signal at 290 °C where after 300 s (5 minutes) the signal has depleted by ~2000 cts / 0.1 s (~20 % of the initial 0.1 s of signal). Beyond 300 s the signal does not appear to deplete any further. The typical time taken for locating a single grain disc (~ 90 s) is marked on Fig. 3b. The calculated reduction in signal after holding the disc for 90 s at 290 °C was ~1000 cts / 0.1 s (~10 % of the initial 0.1 s of signal). Thus, these experiments show that the pIRIR signal-intensity measured for K-feldspar grains was thermally depleted when the grains were held at an elevated temperature.

Implications for single-grain dating of K-feldspars

The thermal depletion of the pIRIR signal when K-feldspar grains are held at elevated temperatures has important implications for single-grain analysis as the time it takes for the single-grain system to locate each disc prior to each L_x and T_x measurement is not constant throughout the measurement sequence. Table 5 presents an example of the different disc location times recorded throughout a typical dating sequence using the pIRIR signal. If the single-grain disc is held at an elevated temperature during the disc location, the pIRIR signals are thermally depleted for different periods of time throughout the sequence. Thus, the pIRIR signals measured are not comparable for each L_n, T_n, L_x and T_x measurement. Moreover, the single-grain measurement system typically finds it most difficult to locate the disc during the L_n

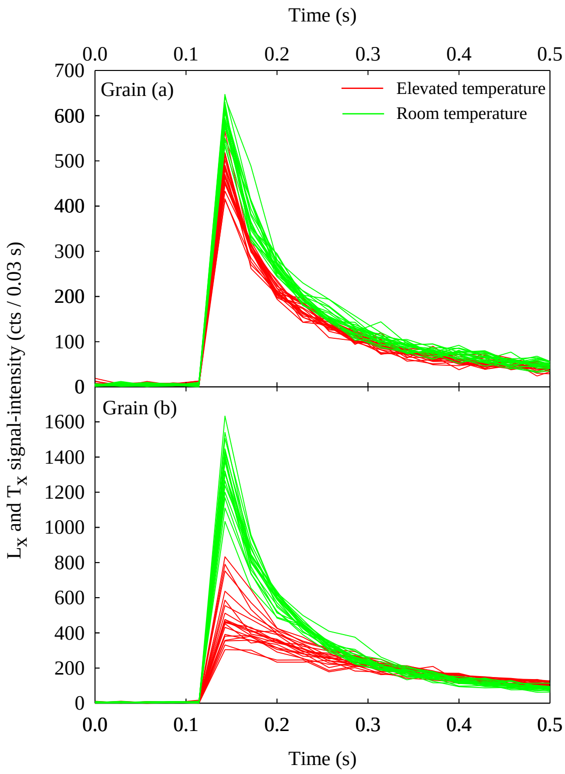

Figure 4: *Examples of the initial 0.5 s of L_x and T_x decay curves measured during the reproducibility experiments performed with the disc location temperature at 290 °C and room temperature. Grain (a) gives the best reproducibility estimate, and is consistent between the experiments using disc location temperatures of 290 °C (1.0 %) and room temperature (1.3 %). In contrast, the reproducibility of grain (b) improves from 11.5 % to 1.7 % when changing the temperature of disc location from 290 °C to room temperature.*

measurement (as shown in Table 5). Thus, it is likely that the L_n measurement is not comparable to the subsequent L_x and T_x measurements performed to construct the dose-response curve and provide sensitivity-correction.

Figure 4 shows two examples of the first 0.5 s of the L_x and T_x $pIRIR_{290}$ decay curves measured for two grains (denoted grains a and b) during the reproducibility experiments performed using disc location temperatures of 290 °C and room temperature. Grain (a) gave the lowest reproducibility estimate, and this did not improve when the disc location temperature was reduced to room temperature. Grain (b) gave a reproducibility estimate of 11.5 % when the disc location temperature was performed at 290 °C, but this fell to 1.7 % when disc location was performed at room temperature. The decay curves measured for grain (a) using disc location temperatures of 290 °C were generally slightly lower intensity than the decay curves measured when the disc was repeatedly located at room temperature, but both sets of decay curves are very similar in shape. In comparison, the decay curves measured for grain (b) using disc location temperatures of 290 °C were more varied and dimmer than the decay curves measured when the disc was located at room temperature.

The decay curves shown for both grains (a) and (b) support the hypothesis that the pIRIR signals of the grains were thermally depleted for different durations of time for the L_x and T_x measurements when the disc location was performed at 290 °C. Comparing the different behaviours between grain (a) and (b) suggests that this effect was more pronounced in some grains in comparison to others; thus, the pIRIR signals of some of the K-feldspar grains were more thermally-dependent than other grains. Locating the discs at room temperature during single-grain dating can circumvent the issues associated with grain-to-grain variability in the thermal erosion of the pIRIR signal during disc location. Performing the disc location at room temperature should therefore be a preferred approach for all routine single-grain dating of sedimentary coarse-grained K-feldspar samples.

Replacing the IR laser with IR LEDs for bleaching

The pIRIR protocol typically incorporates two steps where IR stimulation is used to bleach the sample; one at 60 °C (step 3, Table 1) to remove the influence of any unstable IR_{60} signal prior to the elevated temperature stimulation (i.e. 225 °C or 290 °C), and a second at an elevated temperature (i.e. 290 °C or 330 °C, step 9, Table 1) to prevent charge transfer between SAR cycles. The experiments performed using the pIRIR protocol in this study compared the difference between using the focussed IR laser and the IR LEDs for these two bleaching steps. Table 2 presents the mean, standard error, and range in measurement reproducibility estimates for the individual grains analysed. The mean measurement reproducibility reduced from 4.7 ± 0.4 % to 2.8 ± 0.4 % for the $pIRIR_{225}$ signal, and from 3.6 ± 0.4 % to 2.6 ± 0.3 % for the $pIRIR_{290}$ signal when using the IR LEDs to bleach the grains instead of the IR laser. The measurement reproducibility measured after modifying the $pIRIR_{225}$ and $pIRIR_{290}$ protocols are now comparable with that of the IRSL signal in this study and those published for the green laser (2.5 ± 0.3 %, Thomsen et al. 2005) and the IR laser at 50 °C (2.4 %, Trauerstein et al. 2012).

Figure 2b presents the grains as a function of measurement reproducibility estimates calculated for the $pIRIR_{225}$ (circles) and $pIRIR_{290}$ (triangles) signals using the IR laser (closed, solid line) and IR LEDs (open, dashed line) for bleaching during the measurement protocol. Histograms of the single-grain populations for the $pIRIR_{225}$ (top) and $pIRIR_{290}$ (bottom) signals plot the percentage of grains as a function of the measurement reproducibility and are also shown as inserts in Fig. 2b. Fig. 2b demonstrates the large improvement in the individual estimates of measurement reproducibility for the single-grain population when the IR LEDs are used for bleaching in the pIRIR protocol instead of the IR laser. The range in individual grain measurement reproducibility for the $pIRIR_{225}$ (0.3 – 10.9 %) and $pIRIR_{290}$ (0.2 – 8.2 %) is now comparable to the IR_{60} measurements in this study (0.3 – 10.0 %).

Conclusion

The Risø single-grain measurement system can be optimised to improve the reproducibility of the single-grain measurement system using the pIRIR signal of K-feldspars, and now provides estimates comparable to published single-grain quartz (Jacobs et al. 2006; Thomsen et al. 2005) and IR_{50} K-feldspar measurements (Trauerstein et al. 2012). Reducing the disc location temperature during pIRIR measurements from an elevated temperature (225 °C or 290 °C) to room temperature improves the measurement reproducibility by an average of ~3 % for the single-grain population. In addition, the use of IR LEDs instead of the IR laser to perform the bleaching at 60 °C and at elevated temperatures at the end of each SAR cycle improved the measurement reproducibility further to the estimates of 2.8 ± 0.3 % and 2.6 ± 0.3 % for the $pIRIR_{225}$ and $pIRIR_{290}$ signal, respectively. Both adaptations to the measurement of single-grain K-feldspars using the pIRIR signal are demonstrated to optimise the reproducibility of the single-grain measurement system; thus, the authors recommend that similar experiments are performed for individual readers to quantify the reproducibility of the equipment and measurement protocol used for single-grain dating.

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Reviewer

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