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On the intrinsic accuracy and precision of luminescence dating techniques for fired ceramics

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ABSTRACT

Luminescence dating techniques have been used extensively for archaeological and geological samples. Such techniques are based on thermally or optically stimulated signals. This paper presents simulations of several luminescence techniques for equivalent dose (ED) estimation for ceramic materials containing quartz. The simulations are carried out using a recently published comprehensive kinetic model for quartz, consisting of 11 electron and hole traps and centers. The complete sequence of the experimental protocols for several thermoluminescence (TL) and optically stimulated luminescence (OSL) techniques are simulated using the same set of kinetic parameters. The specific simulated protocols are: additive dose TL protocol, predose technique (both additive and multiple activation versions), phototransfer protocol, single aliquot regenerative optically stimulated luminescence (SAR-OSL) protocol, and SAR thermoluminescence protocol (SAR-TL). One hundred random variants of the natural samples were generated by keeping the transition probabilities between energy levels fixed, while allowing simultaneous random variations of the concentrations of the 11 energy levels. The relative intrinsic *accuracy* and *precision* of the protocols are simulated by calculating the equivalent dose (ED) within the model, for a given natural burial dose of the sample. The intrinsic *accuracy* of these techniques is estimated by simulating natural irradiation of the samples with a known burial dose, followed by simulation of the luminescence method used to recover the estimated dose ED. The percent difference between the burial dose and the ED value represents the simulated accuracy of the luminescence technique. The relative intrinsic *precision* of these techniques is estimated by fitting Gaussian probability functions to the ED values obtained with the 100 model variants. It is found that the various techniques can reproduce natural paleodoses in the range 10 mGy–10 Gy with a typical intrinsic accuracy of +1 to 10%. Techniques based on single aliquot protocols were found in general to be more precise than techniques requiring the use of multiple aliquots. In addition, techniques based on interpolation of experimental data were found to be consistently both more precise and accurate than those based on extrapolation of experimental data.

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1. Introduction

Luminescence techniques are well-established experimental methods for determining the total cumulative dose from natural radiation sources for archaeological and geological dating, for accident dosimetry and for authenticity testing (Aitken, 1985; Wintle, 1996; Bailiff, 1994; Bailiff, 1997; Roberts, 1997; Bailiff et al., 2000; Wintle, 2008; and references therein). During the past four decades accurate and precise methods have been developed for

estimating the equivalent dose (ED) in fired or unfired samples containing quartz; such methods are based either on thermoluminescence (TL) signals, or more recently on optically stimulated luminescence signals (OSL). In a recent comprehensive review of luminescence techniques for ED estimation, Wintle (2008) summarized the historical and technological developments in luminescence dating techniques during the past 50 years, while in a previous paper in this journal Wintle (1996) discussed archaeologically-relevant luminescence dating techniques in a more general context.

Although TL and OSL techniques are well-established experimentally, further theoretical and modeling work is needed in order to obtain a better understanding of the various factors influencing

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both the precision and the accuracy of the various experimental protocols.

There have been several notable published experimental and simulation attempts to estimate the precision and accuracy of various TL/OSL dating techniques. Bailiff and Holland (2000) and Bailiff (1997) performed a comparative study of dating bricks using several different TL/OSL techniques, and by employing both multiple aliquot and single aliquot protocols. Bailiff and Petrov (1999) studied the possibility of using the 210 °C TL peak in quartz for retrospective dosimetry. Their study used ceramics and concluded that single aliquot protocols achieved precisions better than $\pm 5\%$, and that these precisions were much improved over multiple aliquot methods. Mejdahl and Bøtter-Jensen (1997) applied the SARA OSL method to archaeological and geological materials. Their results for 15 samples compared well with those from TL techniques. These authors also compared SARA results for 14 sediments with TL and IRSL estimates. Liritzis et al. (1997) investigated the use of green light stimulation in quartz dating, using ceramics removed from bore holes in areas of archaeological interest in Greece. They discussed various tests and single aliquot correction procedures when using the additive dose single aliquot method of ED determination. They concluded that this technique can be applied successfully when appropriate corrections are made for reusing quartz aliquots. Stokes et al. (2000) applied both SAR and SAAD luminescence techniques to 100 sedimentary samples of different origins. Murray and Olley (2002) examined the reliability of both published and unpublished SAR-OSL quartz ages, for which independent age controls are available. These authors concluded that OSL ages are accurate and that there was no evidence for systematic errors for ages extending at least to 350 ka. Thomas et al. (2008) used both SAR-OSL and TL techniques to study heated materials of archaeological importance from various parts of India. In general, they found reasonably good agreement between the SAR-OSL and TL ages and the corresponding archaeological ages.

There have also been several notable published simulation attempts to estimate the precision and accuracy of various TL/OSL techniques (McKeever et al., 1997; Bailey, 2001; Bailey, 2004; Adamiec et al., 2004, 2006; Kitis et al., 2006; Pagonis et al., 2003, 2006, 2008b). Duller (2007) discussed the nature of random and systematic sources of uncertainties in measurements of the equivalent dose ED during single aliquot regenerative dose measurements. He examined two different approaches to estimating the uncertainty in ED in the linear dose response region of the OSL signal, and found that both these approaches gave results very close and consistent with each other. Several authors have used a different approach, based on kinetic models for quartz. Pagonis and Carty (2004) used a modified version of the model by Chen and Leung (1998, 1999) to simulate the complete sequence of experimental steps taken during the additive dose version of the pre-dose technique. This simulation study was expanded by Pagonis et al. (2008b) using the comprehensive quartz model by Bailey (2001). These authors simulated both the additive dose and the multiple activation versions of the pre-dose technique, as well as the very successful single aliquot regenerative optically stimulated luminescence (SAR-OSL) protocol. Bailey (2004) used a Monte-Carlo approach in which a “standard” quartz model was used as a starting point and 300 versions of the parameters were generated by randomly selecting concentration values within $\pm 80\%$ of the original values, using uniformly distributed random numbers. For each of these variants the full sequence of irradiation and thermal history of the samples were simulated, and the SAR-OSL protocol was simulated in order to obtain an estimate of the precision of the SAR protocol. Thompson (2007) performed Monte-Carlo simulations of SAR-OSL dosimetry measurements to investigate the behavior of the measured equivalent dose (ED) as a function of

absorbed dose (palaeodose). It was found that the mean ED value overestimated the palaeodose, particularly for larger luminescence measurement uncertainties and for larger palaeodoses.

This paper describes an effort to simulate the complete experimental protocols for several luminescence techniques for fired ceramic materials, and to estimate their *relative intrinsic accuracy and precision*. To the best of our knowledge, there are no published simulation studies of these different TL and OSL techniques using the same kinetic model. The intrinsic accuracy and precision are estimated by simulating random variations of the concentrations of electrons and holes in natural quartz samples. The simulations are carried out using a recently published comprehensive model for quartz (Pagonis et al., 2008a), and by generating one hundred random variants of the concentrations of electron and hole traps in the model. The *intrinsic accuracy* of these techniques is estimated by simulating natural irradiation of the samples with a known burial dose, followed by simulation of the luminescence method used to recover the estimated dose ED. The percent difference between the burial dose and the ED value represents the accuracy of the luminescence technique. The *relative intrinsic precision* of these techniques is estimated by fitting Gaussian probability functions to the ED values obtained with the 100 model variants. These uncertainties in the ED values are clearly of a random rather than a systematic nature, due to the random distributions used for the concentrations in the model.

It is found that the various luminescence techniques can reproduce natural paleodoses in the range 10 mGy–10 Gy with an intrinsic accuracy and precision of ± 1 to 10%. The range of doses simulated in this paper was chosen as representative of typical natural doses for fired ceramics of archaeological interest. Techniques based on single aliquot protocols and on interpolation methods were found to be more accurate and precise than techniques requiring the use of multiple aliquots and extrapolation techniques. It is important to emphasize that this paper attempts to simulate the *intrinsic* accuracy and precision of the various luminescence techniques; the actual overall measured *experimental* precision of each technique will of course contain several additional sources of experimental uncertainties, which are not the subject of this paper. Furthermore, this paper addresses ED estimation for fired quartz samples only (such as ceramics), while samples containing unfired quartz or sedimentary quartz will be the subject of a separate study.

2. Description of the model

The simulations in this paper are carried out using the comprehensive quartz model developed by Pagonis et al. (2008a). This model is based on a previous model by Bailey (2001) that was developed on the basis of empirical data. Fig. 1 shows the energy level diagram in the model used in this paper. The computer code, the set of differential equations and the choice of parameters were presented recently by Pagonis et al. (2008a), and will not be repeated here. For easy reference a brief description of the various energy levels and their relevance in the various luminescence dating techniques is presented in this section. The values of the kinetic parameters used in these simulations are shown in Table 1.

The original model by Bailey (2001) consists of 5 electron traps and 4 hole centers, and has been used successfully to simulate a wide variety of TL and OSL phenomena in quartz. This model was expanded by Pagonis et al. (2008a) to include two additional levels 10 and 11, as described below. Level 1 in the model represents a shallow electron trapping level, which gives rise to a TL peak at ~ 110 °C with a heating rate of 5 K/s. The TL and OSL signals from this trap play a major role in the pre-dose and phototransfer techniques simulated in Sections 3.2 and 3.3 of this paper. In addition,

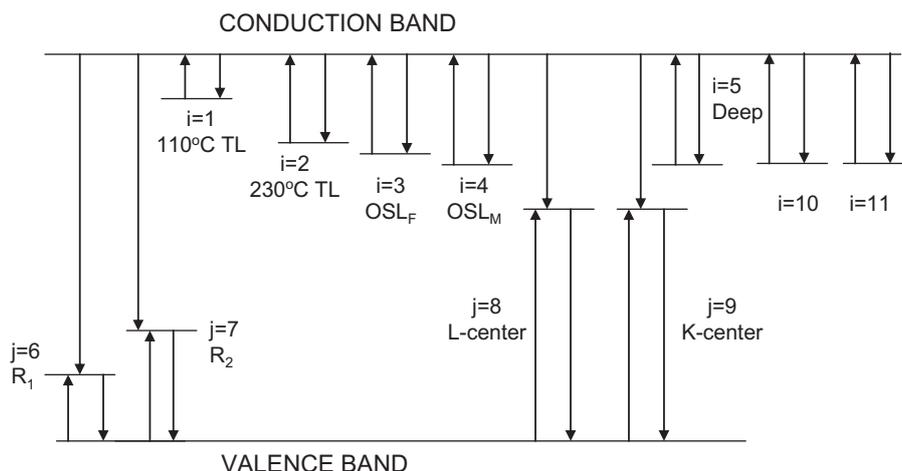


Fig. 1. Schematic diagram of the comprehensive quartz model of Pagonis et al. (2008a) used in this paper. The parameters in the model are given in Table 1.

the TL signal from this “110 °C TL peak” is used to correct for sensitivity changes occurring during simulations of the additive dose TL technique in Section 3.1.

Level 2 represents a generic “230 °C TL” trap, typically found in many quartz samples. The TL signal from this trap has been used successfully in several comprehensive studies (see for example Bailiff and Holland (2000); and references therein).

Levels 3 and 4 are usually termed the fast and medium OSL components and they yield TL peaks at ~330 °C as well as give rise to OSL signals. The OSL from levels 3 and 4 forms the basis of the very precise and accurate SAR-OSL protocols (Wintle and Murray, 2006). The TL and OSL signal from the fast OSL component (level 3) plays a major role in the additive TL, SAR-OSL and SAR-TL techniques simulated in Sections 3.1, 3.4 and 3.5 of this paper.

Level 5 is a deep electron center which is considered thermally disconnected. Levels 6 and 7 are thermally unstable, non-radiative recombination centers (“hole reservoirs”). These two levels play a crucial role in the predose sensitization mechanism which forms the basis of the predose dating technique. Level 8 is a thermally stable, radiative recombination center often termed the “luminescence center” (L). Level 9 is a thermally stable, non-radiative recombination center termed a “killer” center (K).

Levels 10 and 11 are the two new levels added to the original Bailey model by Pagonis et al. (2008a), and were introduced in order to simulate the experimentally observed thermally transferred OSL (TT-OSL) signals and basic transferred OSL (BT-OSL)

signals. Level 10 in the model represents the source trap for the TT-OSL signal and is a slightly less thermally stable trap with high dose saturation. It is assumed that electrons are thermally transferred from level 10 into the fast component trap (level 3). This trap (level 10) is assumed to be emptied optically in nature by long sunlight exposure. Although the model does not contain explicitly any of the slow OSL components which are known to be present in quartz, level 10 has very similar thermal and optical characteristics to such slow OSL components discussed in the literature (Bailey, 2004).

Level 11 is believed to contribute most of the BT-OSL signal in quartz; these traps are more thermally stable than either level 3 or level 10, and correspond to a TL peak at ~360 °C when the sample is heated with a heating rate of 5 K/s. We will refer to this TL peak as the “370 °C TL peak”, and the corresponding TL signal is simulated during simulations of the additive TL and SAR-TL methods described in Sections 3.1 and 3.5 in this paper. It is also noted that the original Bailey (2001) model does not contain an energy level corresponding to this “370 °C TL peak”.

In the rest of this paper it will be demonstrated that by using the model in Fig. 1 and the set of parameters in Table 1, it is possible to simulate the complete experimental protocols for several TL/OSL dating techniques. Table 2 shows the simulation steps for the additive dose TL technique. Tables 3 and 4 show in schematic form the steps simulated in the additive dose and in the multiple activation versions of the predose technique. The simulation steps in

Table 1

The kinetic parameters used in the comprehensive quartz model of Pagonis et al. (2008a) shown in Fig. 1. N_i are the concentrations of electron traps or hole, s_i are the frequency factors, E_i are the electron trap depths below the conduction band or hole trap depths above the valence band, A_i ($i = 1 \dots 5$, and $i = 10, 11$) are the conduction band to electron trap transition probability coefficients, A_j ($j = 6 \dots 9$) are the valence band to hole trap transition probability coefficient and B_j ($j = 6 \dots 9$) are the conduction band to hole center transition probability coefficients. Other parameters related to the photoionization cross-sections of the optically sensitive traps are the photo-eviction constant θ_{0i} , the thermal assistance energy E_{thi} .

Level #	Description	N_i (cm ⁻³)	E_i (eV)	s_i (s ⁻¹)	A_i B_i (cm ³ s ⁻¹)	B_j (cm ³ s ⁻¹)	θ_{0i} (s ⁻¹)	E_{thi} (eV)
1	110 °C TL peak	1.5e7	0.97	5e12	1e-8		0.75	0.1
2	230 °C TL peak	1e7	1.55	5e14	1e-8		0	0
3	Fast OSL (330 °C TL peak)	4e7	1.73	6.5e13	5e-9		6	0.1
4	Fast OSL (330 °C TL peak)	2.5e8	1.8	1.5e13	5e-10		4.5	0.13
5	Deep traps	5e10	2	1e10	1e-10		0	0
6	Hole reservoir	3e8	1.43	5e13	5e-7	5e-9	0	0
7	Hole reservoir	1e10	1.75	5e14	1e-9	5e-10	0	0
8	L-center	3e10	5	1e13	1e-10	1e-10	0	0
9	K-center	1.2e12	5	1e13	1e-14	3e-10	0	0
10	TT-OSL trap 275 °C TL peak	5e9	1.65	6.5e13	1e-11		0.01	0.2
11	370 °C TL peak	4e9	1.6	5e12	6e-12		0	0

Table 2

The simulation steps for the additive TL technique. Multiple aliquots are used.

1. Simulation of the quartz natural sample as discussed in the text.
 2. Irradiate sample in laboratory with dose D_i at a dose rate of 1 Gy/s.
 3. Heat sample to 500 °C – Record maximum of TL signal for the peaks at 300, 330 and 370 °C.
 4. Repeat steps 2–3 for the sequence of doses 0, 0.5, 1, 1.5, 2 and 2.5 Gy to reconstruct the dose–response curve TL vs. dose. Calculate the accrued dose ED by linear or non-linear extrapolation to the x-axis, as shown in Fig. 2b and c.
- The following additional steps to corrected for sensitivity changes on the quart sample were used in some of the simulations:
5. Irradiate sample in laboratory with test dose TD = 5 Gy at a dose rate of 1 Gy/s.
 6. Heat sample to 500 °C – Record maximum of TL signal for 110 °C peak. Use this signal S as a measure of the sensitivity of the sample.

Table 4

The simulation steps for the multiple activation version of the predose technique. A single aliquot is used for all measurements. The test dose is kept constant at 0.01 Gy in all steps of this table. The signals measured in all steps are those of the 110 °C TL peak.

1. Simulation of the quartz natural sample as discussed in the text.
2. Give test dose (TD). Measure S_0 .
3. Heat to 500 °C (first thermal activation).
4. Give test dose (TD). Measure S_N .
5. Give Calibration dose β .
6. Heat to 150 °C to empty the traps.
7. Give test dose (TD). Measure quenched sensitivity S_N .
8. Heat to 500 °C (second thermal activation).
9. Give Test Dose (TD). Measure $S_{N+\beta}$.
10. Calculate ED using S_0 , S_N , $S_{N+\beta}$ and S_N in equations (2) or (3).

the phototransfer protocol are given in Table 5. Finally Tables 6 and 7 outline the steps during the SAR-OSL and SAR-TL single aliquot protocols.

2.1. Simulation of the thermal and irradiation history of natural quartz samples

All simulations presented in this paper contain the following initial steps 1–6 to represent the thermal and irradiation history of the quartz samples. Steps 1–4 are adopted from the suggested simulated history of quartz samples in Bailey (2001). In order to simulate the “zero event” (such as the firing of ceramics in antiquity) as realistically as possible, an additional step 5 was added, which simulates heating the sample at 700 °C for 1 h. In step 6 the natural burial dose of 1 Gy is simulated by using a very low natural dose rate of 10^{-11} Gy/s at 20 °C. A similar simulation step was used in the simulation work by Pagonis et al. (2008b).

- 1 Natural quartz sample. Set all trap populations to zero.
- 2 Geological dose-1000 Gy at 1 Gy/s at 20 °C.
- 3 Geological time- heat to 350 °C.
- 4 Illuminate for 100 s at 200 °C, simulating repeated daylight exposures over long period of time.
- 5 Heat sample to 700 °C for 1 h, to simulate the high temperature firing of ceramics in antiquity.
- 6 Burial dose- 1 Gy at 20 °C at a very low natural dose rate of 10^{-11} Gy/s.

We have studied the effect of varying the parameters in steps 1–5 above during this simulation of the natural quartz sample. It was found that step 5 (heating of the ceramic sample for 1 h at 700 °C) is the critical step in the simulation sequence, while

changes in steps 1–4 above do not affect the results of the simulations significantly. The reason for step 5 being the most important step in the simulation of the natural quartz sample, is that during this high temperature annealing the concentrations of the traps and centers in the model are zeroed within the simulation; this is commonly referred to as the “zero event”.

We have investigated the effect of changing the parameters in steps 1–5 on the estimated ED values, by varying their numerical values over wide ranges. Specifically the geological dose in step 2 was increased up to 10^6 Gy, the geological time temperature in step 3 was changed up to 1000 °C and the illumination time and temperature in step 4 were similarly changed over several orders of magnitude. All of these drastic variations of the parameters resulted in a change of the estimated ED of less than 0.2%, which is well within the numerical accuracy of the results of the simulations. We conclude that the exact parameters used in steps 1–4 above are not critical for the results of the simulation.

The details of the high temperature annealing (step 5) are of paramount importance in the results of the simulations. We increased the annealing temperature in step 5 up to 1000 °C, with no appreciable changes in the results of the simulation. However, it was found that smaller annealing temperatures in the range 500–600 °C gave an overestimate of the ED values during the simulations. This overestimation is not surprising, since one would expect such lower annealing temperatures to result in an incomplete “zeroing event”, and hence a residual luminescence signal. Such residual signals would lead to overestimation of the ED values. The subject of incomplete firing of ceramics in antiquity is a rather important research topic, however, it falls outside the scope of this paper.

Table 5

The simulation steps for the phototransfer technique. A single aliquot is used for all measurements.

1. Simulation of the quartz natural sample as discussed in the text.
2. Heat to 160 °C to clear 110 °C traps.
3. Irradiate with beta test dose of 0.01 Gy.
4. Heat to 160 °C to measure the initial sensitivity S_0 – record height of 110 °C TL peak.
5. UV or optical stimulation for 1 min (phototransfer step).
6. Heat to 160 °C to measure the new sensitivity S_N – record height of 110 °C TL peak.
7. Prolonged UV irradiation to clear the deep donor traps.
8. Heat to 160 °C to clear 110 °C traps.
9. Irradiate with beta test dose of 0.01 Gy.
10. Heat to 160 °C to measure the sensitivity \bar{S}_0 – record 110 °C TL peak.
11. Give calibration beta dose $\beta \sim$ ED of 1 Gy.
12. Heat to 160 °C to clear 110 °C traps.
13. UV or optical stimulation for 1 min (phototransfer step).
14. Heat to 160 °C to measure the sensitivity \bar{S}_β – record 110 °C TL peak.
15. Calculate ED using equation (4).

Table 3

The simulation steps for the additive dose version of the predose technique. Multiple aliquots are used in this technique. The test dose is kept constant at 0.01 Gy in all steps of this table. The signals measured in all steps are those of the 110 °C TL peak.

1. Simulation of the quartz natural sample as discussed in the text.
2. Give test dose, usually 0.01 Gy. Measure initial sensitivity S_0 using the 110 °C TL peak.
3. Heat to 500 °C.
4. Give test dose. Measure thermally activated sensitivity S_N .
5. Use a new Aliquot; give test dose. Measure S_0 for 'normalization' purposes.
6. Give Calibration dose β (use $\beta \sim$ ED by using trial and error).
7. Heat to 500 °C (thermal activation).
8. Give test dose. Measure thermally activated sensitivity $S_{N+\beta}$.
9. Repeat steps 2–8 using added doses of 2β , 3β , etc. in step 6 to obtain the sensitivities $S_{N+2\beta}$, $S_{N+3\beta}$, etc. Obtain the graph of the sensitivity S vs. added dose and estimate the value of ED by linear extrapolation, as shown in Fig. 4a.

Table 6

The simulation steps for the SAR-OSL technique. A single aliquot is used for all measurements.

1. Simulation of the quartz natural sample as discussed in the text.
2. Irradiate sample with dose D_i .
3. Preheat 10 s at 260 °C.
4. Blue OSL for 100 s at 125 °C – record OSL (0.1 s) signal (L).
5. Test dose $TD = 0.1$ Gy.
6. Cutheat 20 s at 220 °C.
7. Blue OSL for 100 s at 125 °C – record OSL (0.1 s) signal (T).
8. Repeat steps 2–7 for the sequence of doses 0, 0.5, 1, 1.5, 0, 0.5 Gy to reconstruct the dose. Response curve L/T vs. dose. Estimate ED using interpolation as shown in Fig. 8a.

2.2. Simulation of random natural variations in quartz samples

Our simulation method is similar to the published work by Bailey (2004) who used a Monte-Carlo approach as follows; a “standard” quartz model was used as a starting point, and 300 versions of the parameters were created to form a statistical ensemble. The experimentally observed variability in TL and OSL characteristics of quartz was simulated by assuming that all the fundamental transition probabilities in the model remain constant, while trap concentrations are allowed to vary randomly from the values of a “standard quartz model”. The 300 variants of the model were generated in Bailey (2004) by randomly selecting concentration values within $\pm 80\%$ of the values in the standard model, using uniformly distributed random numbers. As discussed in Bailey (2004, p. 304) some variation of the transition probabilities may also be present in natural samples, but this variation is expected to be relatively insignificant. For each of these 300 variants Bailey simulated the full sequence of irradiation and thermal history of the samples, and the SAR-OSL protocol was simulated in order to obtain an estimate of the precision and accuracy of the SAR protocol.

Even though our modeling approach in this paper is similar to the approach of Bailey (2004), our goals are rather different. In this paper we are mostly interested in a comparative study of the *intrinsic accuracy and precision* of the simulated protocols. In a real experiment, one deals of course with several additional sources of experimental uncertainties, as described for example in some detail in the paper by Duller (2007).

An important question concerns the use of Gaussian curves to fit the simulated distribution of ED values. One could envision the use of a binomial, Poisson or another similar mathematical distribution instead. Our main reason for using a Gaussian curve is that there is a relevant precedent in simulation work using kinetic models in quartz; previous work by Bailey (2004) also used Gaussian fitting functions for their distribution of their ED values. We used the same fitting function in this paper as in Bailey (2004), in order to have a “reference comparison standard” for the magnitude of the

Table 7

The simulation steps for the SAR-TL technique. A single aliquot is used for all measurements.

1. Simulation of the quartz natural sample as discussed in the text.
2. Irradiate sample with dose D_i .
3. Preheat 10 s at 260 °C.
4. Measure TL by heating to 500 °C – record TL signal (L).
5. Test dose $TD = 0.1$ Gy.
6. Cutheat 20 s at 220 °C.
7. Measure test dose TL by heating to 500 °C – record TL signal (T).

Repeat steps 2–7 for the sequence of doses 0, 0.5, 1, 1.5, 0, 0.5 Gy to reconstruct the dose–response curve L/T vs. dose. Estimate ED using interpolation as shown in Fig. 9a.

accuracy and precision of the simulated luminescence dating techniques. In addition, Gaussian curves were found to fit the data reasonably well in most cases, and the R^2 value of the fits given in the relevant figures were found to be between 0.80 and 0.90. In principle, however, there is no absolute compelling reason for using Gaussian fitting curves.

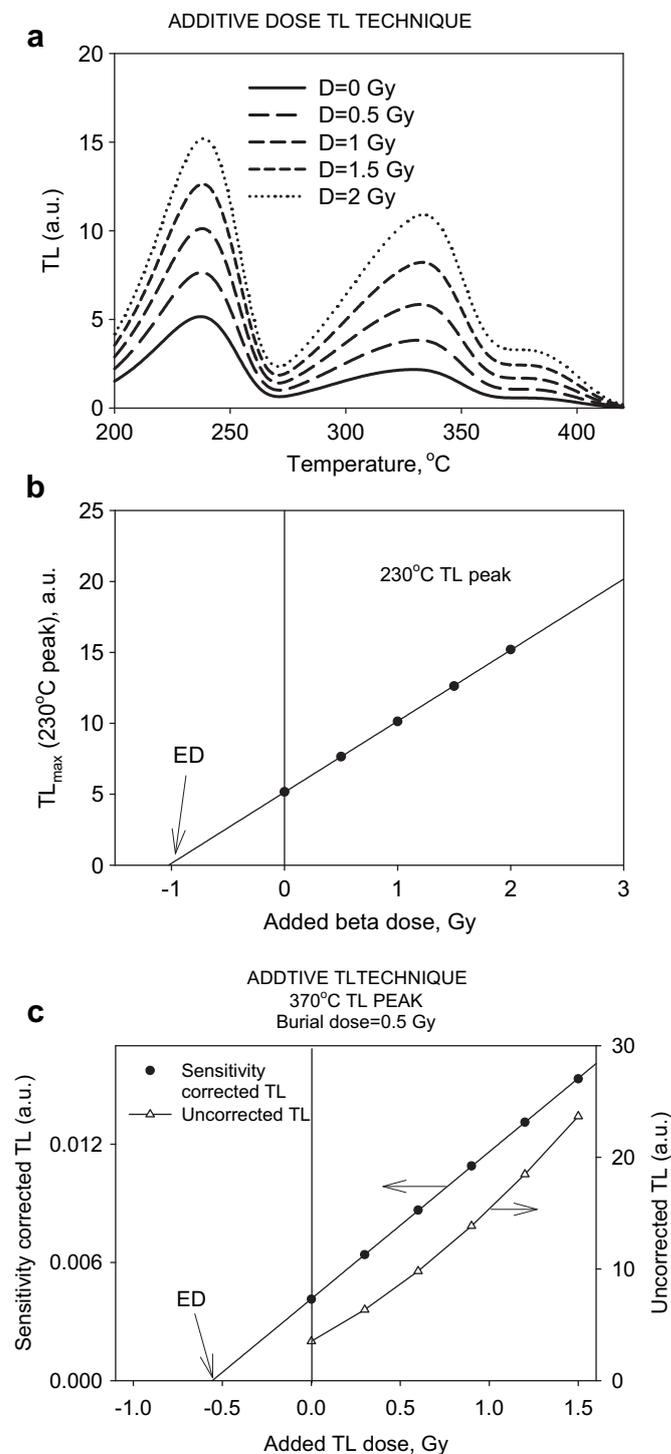


Fig. 2. (a) Simulated glow curves during the additive TL technique. (b) The dose response of the 230 °C TL peak is linear in the dose range simulated in this paper. (c) The dose–response of the 370 °C TL peak is superlinear in the same dose range (open triangles). After correcting for sensitivity changes using the height of the 110 °C TL peak, the dose–response of the corrected TL signal becomes linear (closed circles).

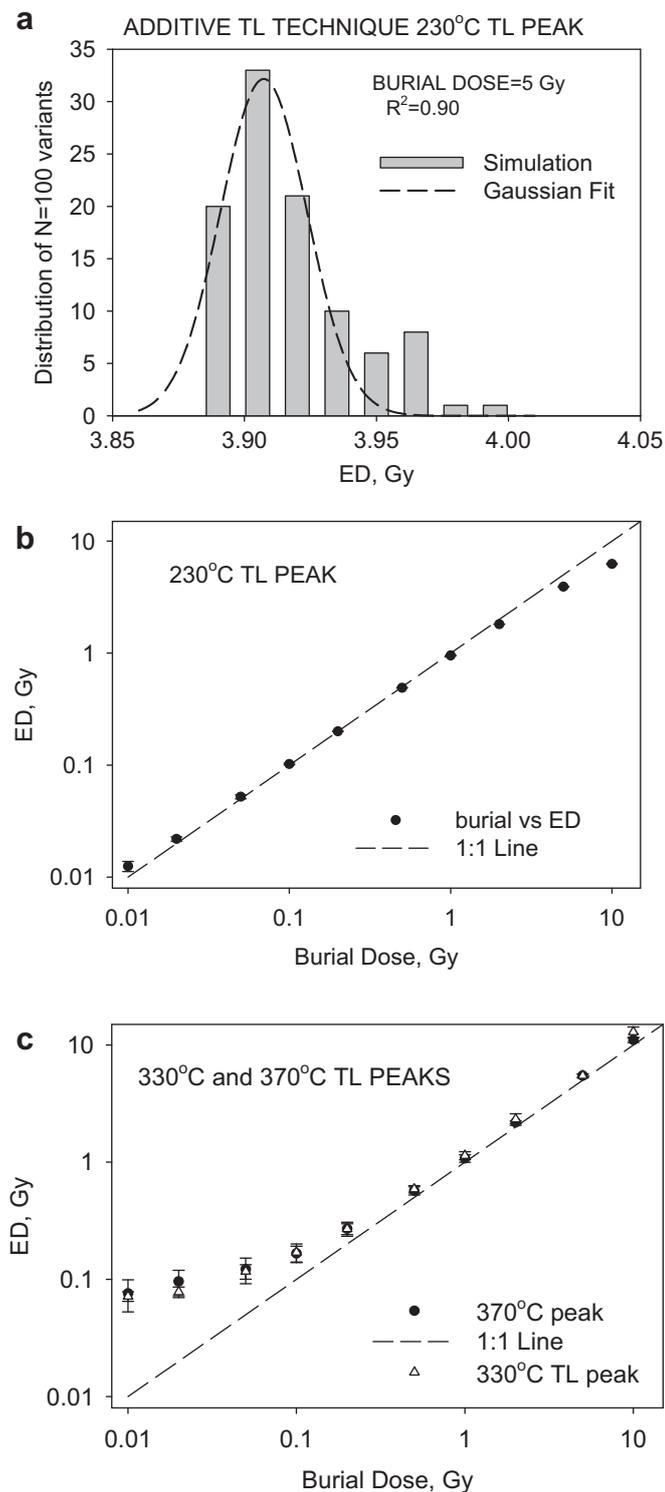


Fig. 3. (a) Simulated results for the intrinsic precision of the additive dose TL technique at a burial dose of 5 Gy, using 100 random variants of the natural samples. The histogram shown is for the 230 °C TL peaks, and is fitted to Gaussian distributions as discussed in the text. The simulated steps are given in Table 2. (b) The simulated equivalent doses (ED) obtained using the additive dose TL technique for the 230 °C TL peak, and for a range of burial doses. The error bars represent the standard deviation σ obtained from Gaussian fits similar to the one shown in (b). (c) Same as in (b), for the 330 °C and 370 °C TL peaks.

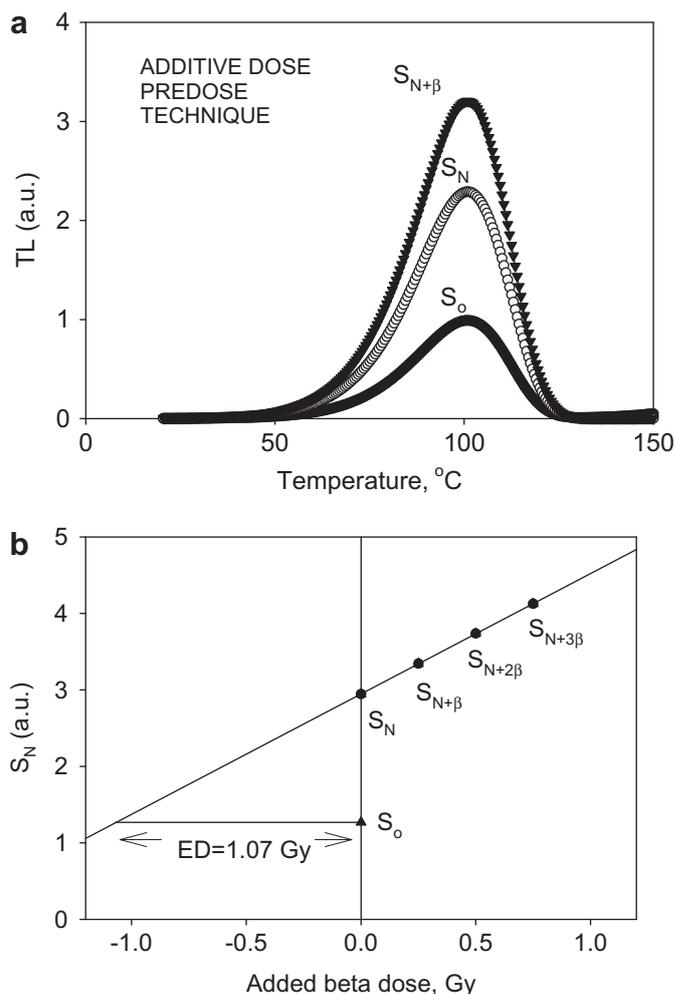


Fig. 4. (a) Simulated glow curves for the additive dose predose technique with the test dose TD=0.01 Gy, a paleodose PD=1 Gy, a calibration beta dose of 1 Gy and an activation temperature of 500 °C. The simulated steps are given in Table 3. (b) The simulated variation of the sensitivity S of the 110 °C TL peak with the added beta dose. Linear extrapolation gives the equivalent dose (ED) of the sample as shown.

3. Simulations of TL/OSL techniques for ED estimation

3.1. Simulation of the additive dose technique for TL

Historically the oldest method of TL is the additive dose technique, in which different aliquots of the quartz sample are given a sequence of added beta doses, and the TL glow curves are measured for each aliquot. Three TL peaks at 230 °C, 330 °C and 370 °C were used in the simulations for estimating the ED values of fired ceramic samples. Typical simulated steps in this method are shown in Table 2. The main assumption behind this technique is the existence of a linear relationship between the TL signals and the irradiation dose. A typical example of simulated glow curves at different beta doses is shown in Fig. 2a, in which the maximum intensity of all three TL peaks can be seen to increase with the added beta dose.

Fig. 2b shows the dependence of the TL signal from the 230 °C on the added beta dose; this dependence is linear within the range of the simulations, namely 10 mGy–10 Gy. The equivalent dose ED of the sample can be found by linear extrapolation to the dose axis, as shown in Fig. 2b.

A different non-linear simulated dose response was obtained for the deeper TL traps at 330 °C and 370 °C, as shown in Fig. 2c for

the case of the TL peak at 370 °C. The response of this peak was found to vary almost quadratically with the irradiation dose, a phenomenon that is well known from a variety of TL studies (Wintle, 2008). A similar simulated non-linear behavior was found for the 330 °C TL peak (this data is not shown here). This quadratic dose dependence has been previously attributed to competition effects between different TL traps in quartz (Chen and McKeever, 1997).

The non-linear dose dependence of the TL signal is one of the major practical problems associated with TL dating. In some cases the non-linearity can be removed by making a sensitivity correction to the TL signal. We have simulated this sensitivity correction by adding a few steps to the simulation steps, as shown in Table 2. These steps consist of irradiating the sample with a test dose of 5 Gy, followed by a measurement of the TL signal from the 110 °C TL peak. The maximum height of this 110 °C TL signal is used as a measure of the sensitivity S of the sample; the corrected TL signal is obtained by dividing the TL signal from the 330 °C or 370 °C TL peaks by this sensitivity S . The result of applying is shown in Fig. 2c, where it can be seen that the sensitivity-corrected TL signal

becomes linear with the dose. The ED value can now be found by extrapolating the fitted linear curve to the dose axis.

The results of simulating 100 variants of the additive dose TL technique using the 230 °C TL peak are shown in Fig. 3a. The distribution of the ED values was fitted with a Gaussian distribution function shown as a dashed line. The equation for the fitted Gaussian distribution $N(D)$ is of the form:

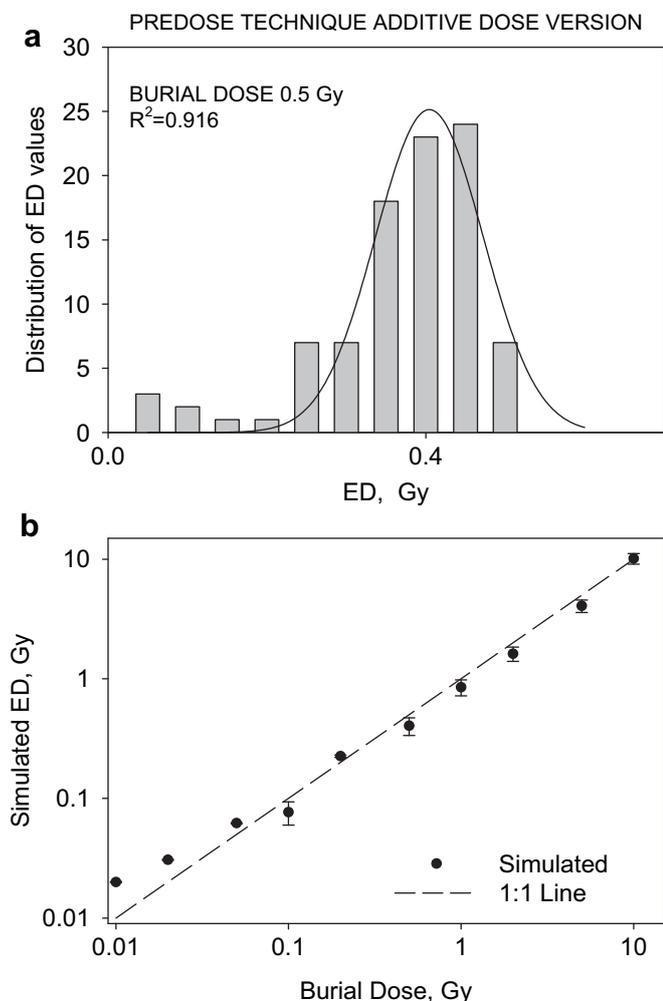


Fig. 5. (a) Simulated results for the intrinsic accuracy and precision of the additive dose predose technique at a burial dose of 0.5 Gy, using $N = 100$ random variants of the natural samples. The simulated steps are given in Table 3. The solid line represents a best-fit Gaussian curve to the simulated data. (b) The simulated equivalent doses (ED) obtained using the additive dose predose technique and for a range of burial doses. The error bars represent the standard deviation σ obtained from Gaussian fits similar to the one shown in (a).

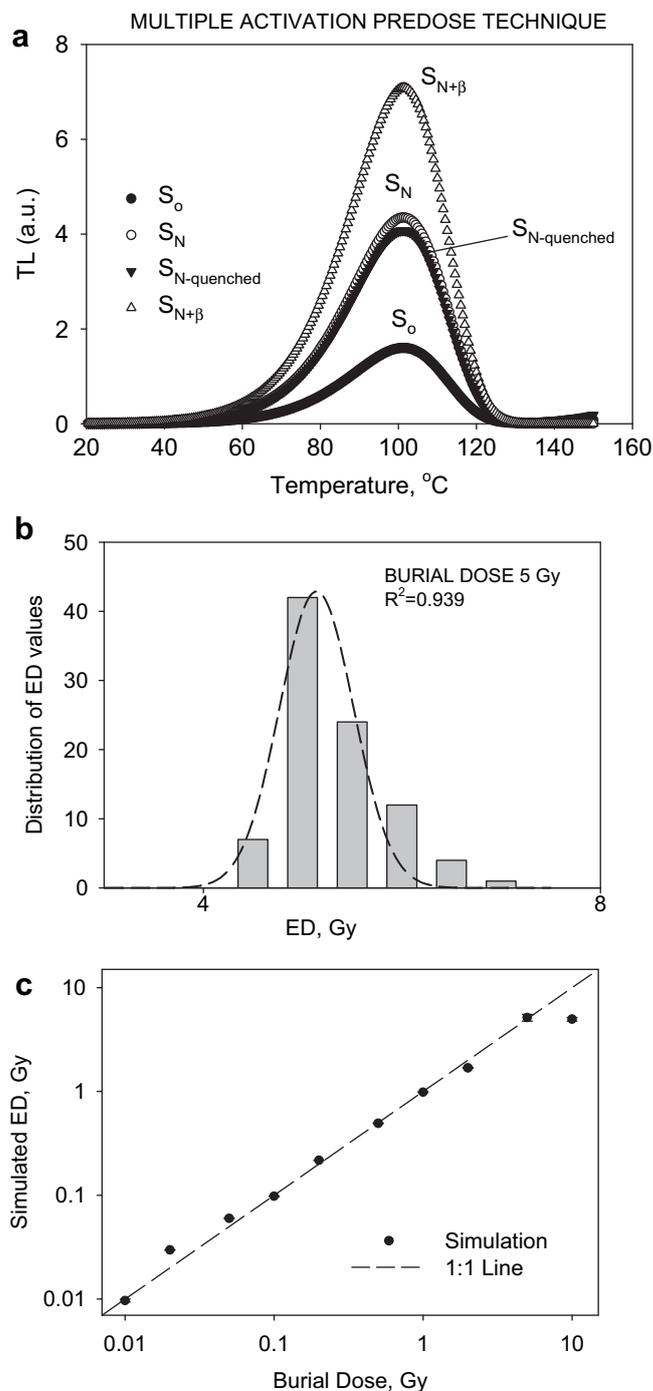


Fig. 6. (a) Simulated glow curves during the multiple activation predose technique. The simulated steps are given in Table 4. (b) Simulated results for the intrinsic precision of the multiple activation predose technique at a burial dose of 1 Gy, using $N = 100$ random variants of the natural samples. (c) The simulated equivalent doses (ED) obtained using the multiple activation predose technique and for a range of burial doses.

$$N(D) = A \exp\left(-\frac{(x - x_0)^2}{2\sigma^2}\right), \quad (1)$$

where the constant A represents the number of variants at the peak of the distribution which is centered at x_0 and has a standard

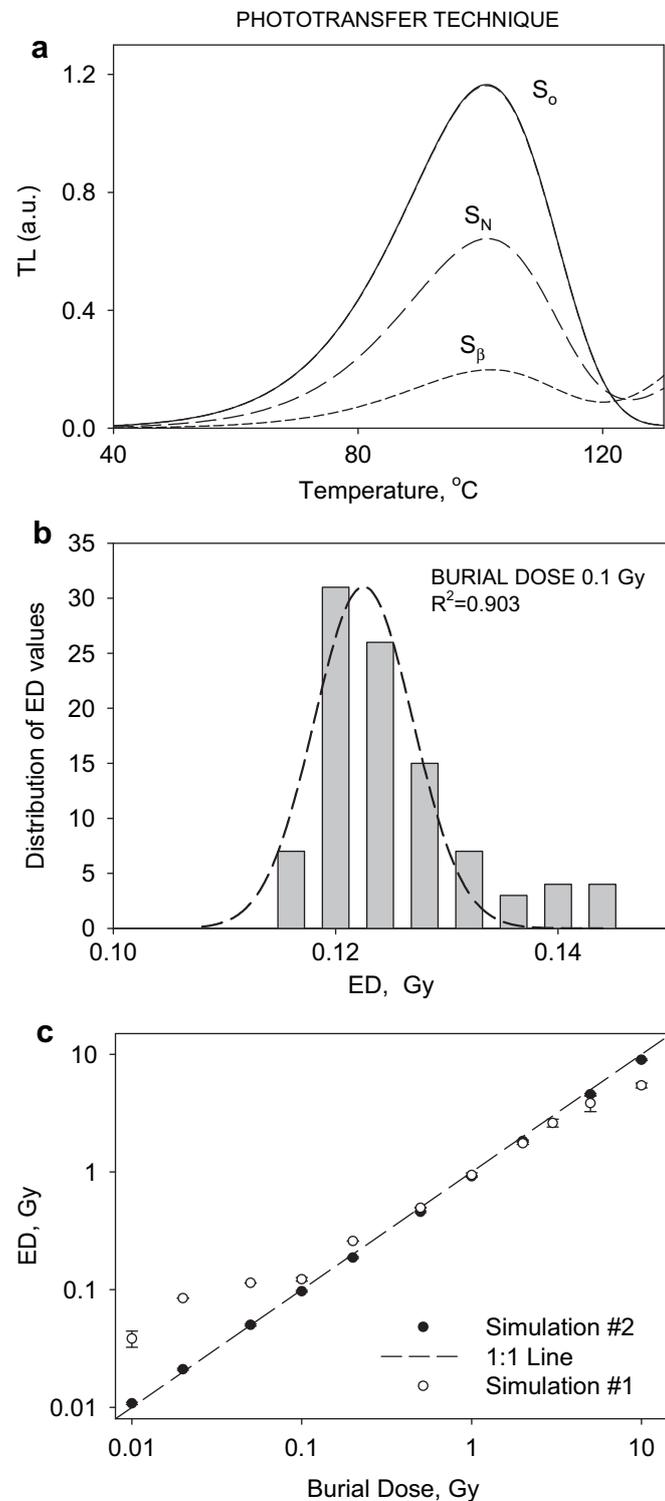


Fig. 7. (a) Simulated glow curves during the phototransfer technique. The simulated steps for this technique are given in Table 5. (b) Simulated results for intrinsic precision of the phototransfer technique at a burial dose of 0.1 Gy, using $N = 100$ random variants. (c) The simulated equivalent doses (ED) obtained using the phototransfer technique and for a range of burial doses.

deviation σ . The standard deviation σ obtained in this manner represents a measure of the *intrinsic precision* of the technique. In the example of Fig. 3a the accumulated natural dose was 5 Gy and the fitted Gaussian curve yields an average ED = 3.91 Gy, with a standard deviation σ of 0.02 Gy. We conclude that this dating protocol underestimates the burial dose by ~20%, and that the intrinsic precision of this technique is $\sigma/ED = 0.02/3.91 \text{ Gy} = 0.5\%$.

This procedure of simulating 100 variants and finding the standard deviation σ from the best-fitted Gaussian distribution was repeated for several burial doses between 10 mGy and 10 Gy, with the results shown in Fig. 3b. The 1:1 dashed line in this figure indicates the ideal case in which 100% of the burial dose is recovered using the simulated technique. The amount of deviation of the data points in Fig. 3b from the 1:1 line is a measure of the *intrinsic accuracy* of the technique. The small error bars in this figure are the standard deviation σ obtained from the Gaussian fits, and represent the *intrinsic precision* of this technique.

The results of Fig. 3b show that the additive TL method using the 230 °C peak can reproduce the ED values accurately in the range 10 mGy–2 Gy. The intrinsic precision of this technique is seen to be very good, in the range 2–5%, as indicated by the small error bars in Fig. 3b.

Fig. 3c shows the simulation results obtained for the additive TL method using the 330 °C and 370 °C TL peaks. It can be seen that using these two higher temperature TL peaks within this model leads to an overestimation of small doses between 10 mGy and 0.1 Gy. However, both methods seem to be both accurate and precise in the higher dose range 0.2 mGy–10 Gy. We attribute the systematic ED overestimation at lower doses to the following three factors. Firstly, the TL signal at 330 °C is a composite one, containing contributions from both the fast and the medium OSL/TL components (levels 3 and 4 in the model). Secondly, there is an additional TL peak at ~300 °C in the model, shown as a smaller hump in Fig. 2a. This peak overlaps with the TL peak at 330 °C, and represents level 10 in the model. Thirdly, using the signal from the 110 °C TL peak may not completely correct for sensitivity/superlinearity changes occurring in the sample. An important question about the histograms presented in this paper concerns the presence of several simulated data points which fall well outside the histogram, as seen for example in Fig. 3a. We have repeated the simulations in Fig. 3a using a larger number $N = 300$ of sample variants, in order to check whether the presence of these points is due to our use of $N = 100$ variants. The results of simulating $N = 300$ variants were found to be identical to those of Fig. 3a. This indicates that these outlying points are not an artifact of the simulation process, but rather they represent the inherent limitations in the accuracy and precision of the various dating techniques.

3.2. Simulations of the predose technique

Two main variations of the predose technique exist, known as the multiple thermal activation technique and the additive dose technique. In a typical experimental application of the predose procedure, a test dose $TD = 0.01 \text{ Gy}$ is commonly used, as well as a calibration dose β with a value close to the estimated paleodose of the sample. The exact value of the calibration dose to be used in the experiment is usually found by trial and error. The response to the test dose TD is measured by heating the sample to 150 °C, just above the “110 °C” TL peak. The effect of various experimental parameters on the accuracy of the predose technique was simulated in detail in Pagonis and Carty (2004) and Pagonis et al. (2008b). Our goal in this paper is to explore the relative accuracy and precision of the two predose techniques, rather than a detailed study of the effect of the various experimental parameters.

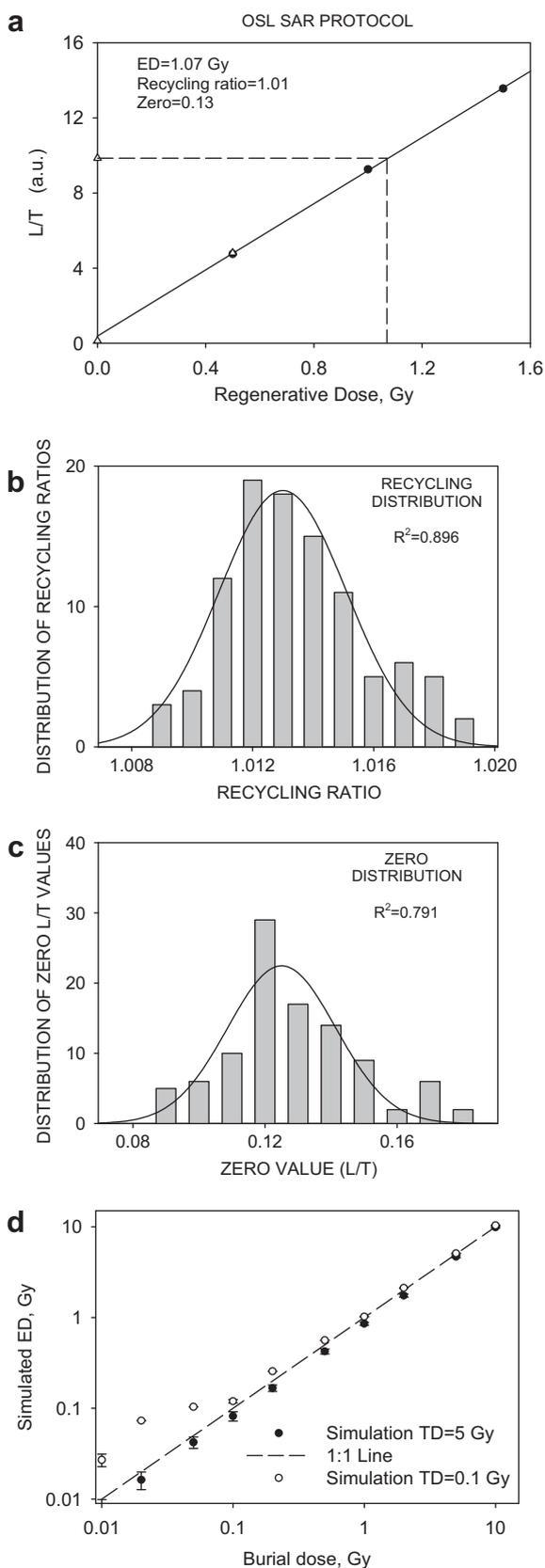


Fig. 8. (a) Simulated L/T signals during the SAR-OSL technique. The simulated steps are given in Table 6. (b), (c) Typical distributions of the simulated recycling ratios and zero intercepts for the SAR-OSL technique. (d) The simulated equivalent doses (ED) obtained using the SAR-OSL technique and for a wide range of burial doses.

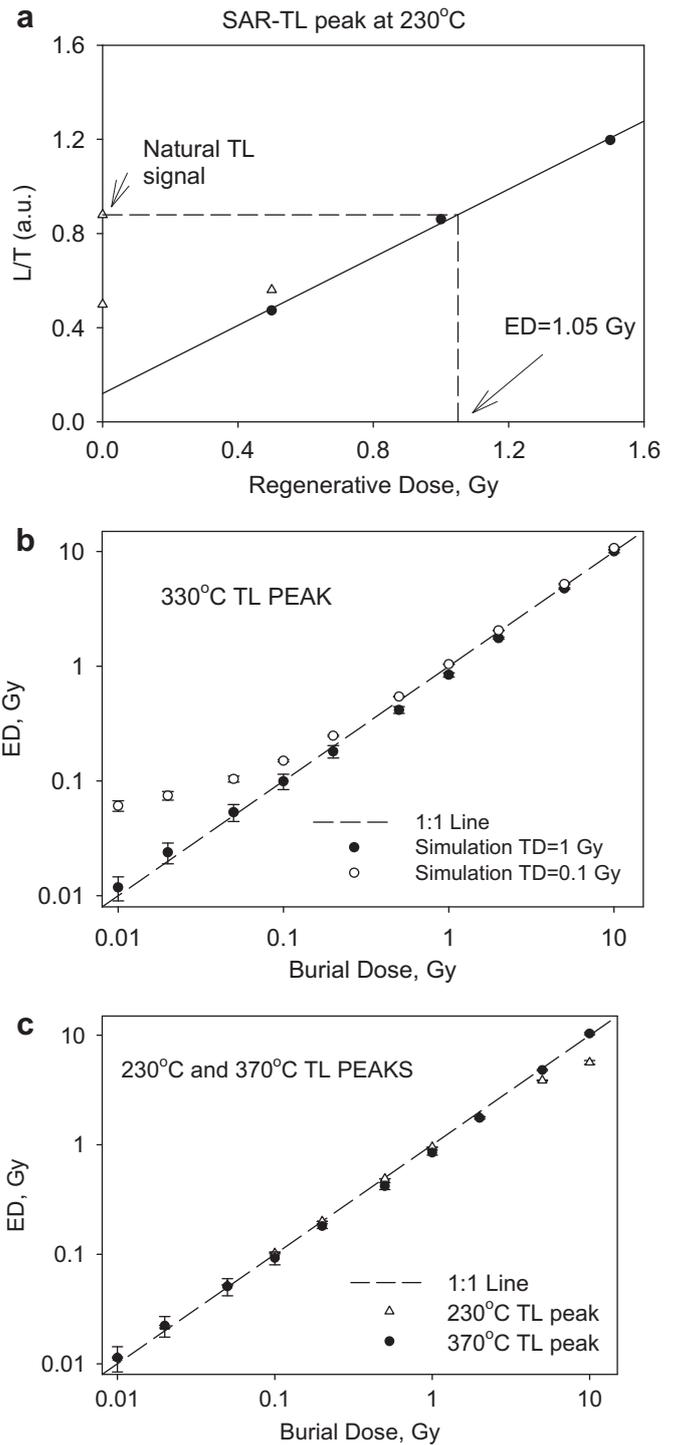


Fig. 9. (a) Simulated L/T signals during the SAR-TL technique, using the 230 °C TL peak in quartz. The simulated steps are given in Table 7. (b) The simulated equivalent doses (ED) obtained using the SAR-TL technique for the 330 °C TL peak, and for a range of burial doses. (c) Similar results for the TL peak at 230 °C and 370 °C.

3.2.1. The additive dose variation of the predose technique

The additive dose variation is a multiple aliquot technique. The basic sequence of measurements during the additive dose technique is shown in Table 3 (Aitken, 1985, p. 153–168). Using a first portion of the material the TL sensitivities S_0 and S_N to a small test dose (TD) are measured as shown in steps 2–4 of Table 3. Using the remaining portions of the material, the TL sensitivities of the irradiated samples $S_{N+\beta}$, $S_{N+2\beta}$, $S_{N+3\beta}$ etc. are measured as shown in

Table 8

Summary of all ED values and their uncertainties obtained from the simulations in this paper. All values are in Gy.

Burial dose, Gy	Additive TL 230 °C peak	Uncertainty ($\pm 1\sigma$)	Additive TL 330 °C peak	Uncertainty ($\pm 1\sigma$)	Additive TL 370 °C peak	Uncertainty ($\pm 1\sigma$)	Additive predose, 110 °C peak	Uncertainty ($\pm 1\sigma$)	Multiple aliquot predose, 110 °C peak
0.01	0.013	0.001	0.072	0.007	0.076	0.023	0.020	0.000	0.010
0.02	0.022	0.001	0.078	0.008	0.096	0.024	0.031	0.000	0.030
0.05	0.052	0.002	0.117	0.017	0.122	0.030	0.062	0.001	0.060
0.1	0.102	0.002	0.170	0.031	0.166	0.026	0.077	0.017	0.098
0.2	0.200	0.001	0.270	0.037	0.270	0.029	0.225	0.005	0.217
0.5	0.490	0.004	0.589	0.043	0.570	0.046	0.405	0.068	0.493
1	0.952	0.002	1.138	0.090	1.077	0.080	0.850	0.130	0.984
2	1.810	0.002	2.327	0.262	2.184	0.059	1.620	0.220	1.687
5	3.910	0.020	5.474	0.182	5.469	0.195	4.070	0.490	5.142
10	6.255	0.061	12.907	1.322	11.005	0.510	10.130	1.040	4.960

steps 5–9 of Table 3. Examples of simulated TL glow curves during this procedure are shown in Fig. 4a.

The additive dose variation of the predose technique avoids multiple thermal activation of the material and also is not affected by the phenomenon of radiation quenching which was simulated and discussed in some detail by Pagonis et al. (2008b). The obvious disadvantage of the additive dose technique is the need for inter-calibration between the different aliquots, and the necessity for samples exhibiting low S_0 -values (Aitken, 1985, p. 163; Pagonis et al., 2008b). The dose evaluation in the additive dose technique is based on the assumption of a linear variation of the sensitivity of the sample with the dose. By extrapolating the graph of the sensitivity S to the dose axis as shown in Fig. 4b, one obtains an estimate of the equivalent dose (ED) of the sample.

The results of simulating 100 variants of the additive dose version of predose technique are shown in Fig. 5a, and these were fitted with a Gaussian distribution as described previously. This procedure was repeated for several burial doses between 10 mGy and 10 Gy, with the results shown in Fig. 5b, together with the 1:1 line. The error bars in this figure represent once more the standard deviation σ obtained from the Gaussian fits, and are seen to be much larger than those in Fig. 3b. The results of Fig. 5b indicate that the additive dose variation of the predose technique systematically underestimates the ED values in the complete range 10 mGy–10 Gy, and that the intrinsic precision of this technique becomes progressively worse at smaller doses.

3.2.2. The multiple activation variation of the predose technique

The basic sequence of measurements during the multiple activation technique is shown in Table 4 and is based on the use of a single aliquot of the sample (Aitken, 1985, p. 153–168). The TL sensitivities S_0 and S_N of the material to a small test dose (TD) are measured as shown in steps 2–4 of Table 4. The thermally activated sensitivities $S_{N+\beta}$ and S_N are measured using the same aliquot, as shown in the rest of Table 4. Experiments using the multiple activation variation exhibit the phenomenon of radiation quenching consisting of the sensitivity S_N in step 7 being usually lower than the sensitivity S_N in step 4, due to the beta irradiation in step 5 of Table 4. In addition, in this technique the aliquot undergoes a multiple thermal activation, which can cause changes to its predose characteristics. A sequence of typical simulated TL glow curves obtained using the multiple activation predose technique is shown in Fig. 6a.

The equations used in the multiple activation technique are also based on the assumption of a linear response of the sensitivity of the sample. The equivalent dose (ED) can be calculated using the equation:

$$ED = \frac{S_N - S_0}{S_{N+\beta} - S_N} \cdot \beta. \quad (2)$$

When the effect of radiation quenching is taken into account, equation (2) is modified by using the quenched sensitivity $S_{N'}$, instead of the sensitivity S_N , to obtain a corrected estimate of the equivalent dose using the equation (Aitken, 1985):

$$ED = \frac{S_N - S_0}{S_{N+\beta} - S_{N'}} \cdot \beta. \quad (3)$$

The applicability of this corrected equation (3) is discussed in some detail in Pagonis et al. (2008b). The results of simulating 100 variants of the multiple activation predose technique are shown in Fig. 6b and c. The results of Fig. 6bc indicate that the multiple activation version of the predose technique reproduces both accurately and precisely the ED values in the complete range 10 mGy–10 Gy. By comparing Fig. 6c and Fig. 5c, it is clear that the multiple activation version is preferable to the additive dose version in the case of the predose technique. This technique becomes much less accurate at doses $ED > 10$ Gy.

3.3. Simulation of the phototransfer technique

This technique has waned in popularity due to the development of the more precise/accurate SAR protocols. However, we include it here to demonstrate that the mechanism in the phototransfer technique is consistent with the model used in this paper. In quartz phototransfer, optical stimulation transfers trapped charge from optically sensitive deep donor levels like the 330 °C TL trap, into shallower acceptor levels like the 110 °C TL trap (McKeever, 1985). The main assumption behind this technique is that a fraction of the initially trapped charge in the deep donor levels is transferred into the 110 °C TL trap, and therefore, the TL signal from this trap will likely be proportional to the radiation dose received by the sample. The equivalent dose ED of the sample can be found by using a calibration dose of about the same magnitude as the natural dose. The simulated steps in this method are shown in Table 5. In steps 1–4 the initial sensitivity S_0 of the aliquot to the test dose of 0.01 Gy is measured, followed by measurement of the activated sensitivity S_N in steps 5–6. Step 5 is the main phototransfer event, in which UV or optical stimulation transfers charge from deep optically sensitive traps at 330 °C (the “fast OSL” trap of quartz), into the 110 °C TL trap. This charge transfer is assumed to take place through the conduction band. The deep traps are cleared by prolonged UV irradiation in step 7, and the sensitivity to the test dose is measured again in steps 8–10. Steps 11–14 are the calibration sequence for the phototransfer process, where the sensitivity of the aliquot to a known calibration beta dose β is measured (S_β). By assuming that the sensitivity signal from the 110 °C TL trap is proportional to the phototransferred charge, the estimated dose ED of the aliquot is given by the equation (McKeever, 1985, p. 268):

Uncertainty ($\pm 1\sigma$)	Phototransfer, 110 °C peak	Uncertainty ($\pm 1\sigma$)	SAR-OSL, 330 °C peak	Uncertainty ($\pm 1\sigma$)	SAR-TL, 230 °C peak	Uncertainty ($\pm 1\sigma$)	SAR-TL, 330 °C peak	Uncertainty ($\pm 1\sigma$)	SAR-TL, 370 °C peak	Uncertainty ($\pm 1\sigma$)
0.000	0.011	0.000	0.008	0.002	0.011	0.000	0.012	0.003	0.011	0.003
0.000	0.021	0.000	0.016	0.004	0.022	0.001	0.024	0.005	0.022	0.005
0.000	0.050	0.001	0.042	0.006	0.052	0.001	0.053	0.009	0.051	0.009
0.000	0.097	0.002	0.082	0.010	0.102	0.001	0.099	0.015	0.093	0.012
0.001	0.187	0.003	0.167	0.014	0.200	0.000	0.181	0.023	0.181	0.009
0.003	0.459	0.008	0.424	0.026	0.489	0.000	0.416	0.028	0.419	0.030
0.010	0.918	0.013	0.854	0.032	0.951	0.001	0.842	0.036	0.848	0.046
0.018	1.834	0.033	1.747	0.067	1.804	0.005	1.760	0.034	1.755	0.033
0.374	4.572	0.079	4.691	0.137	3.864	0.042	4.753	0.071	4.811	0.062
0.194	8.976	0.145	9.875	0.205	5.647	0.224	10.039	0.289	10.352	0.231

$$ED = \beta S_N \bar{S}_0 / S_\beta S_0. \tag{4}$$

Typical results obtained by simulating the steps in Table 5 are shown as “Simulation #1” in Fig. 7a, and the results of simulating 100 random sample variants are shown in Fig. 7b. In the example of Fig. 7a the two sensitivities S_0 and \bar{S}_0 are the same. In a second set of simulations we varied the calibration beta dose used in step 11 of Table 5; instead of using a fixed calibration beta dose of 1 Gy, we used a calibration dose equal to 0.1 times the natural dose of the sample. The results of using this optimized calibration dose are shown as “Simulation #2” in Fig. 7c. The results of Fig. 7c indicate that the phototransfer technique can be used for the whole range of doses 10 mGy–10 Gy, when the parameters are optimized.

3.4. Simulation of the single aliquot regenerative dose OSL technique (SAR-OSL)

Fig. 8 shows the results of simulating the popular and very successful single aliquot regenerative protocol (SAR) developed during the past 10 years (Wintle and Murray, 2006). The SAR technique has been developed for both OSL and TL signals, with the corresponding techniques referred to as SAR-OSL and SAR-TL. The most important part of the SAR technique is the correction of the measured OSL/TL signals by measuring the sensitivity of the sample due to a small test dose. The OSL/TL signal due to an irradiation dose is usually denoted by L , while the corresponding test dose signal is denoted by T . The ratio L/T represents the sensitivity-corrected OSL/TL signal.

During the SAR protocol the measured natural L/T signal is compared with additional L/T signals which are regenerated by irradiating the samples in the laboratory. A typical sequence of steps undertaken during the SAR-OSL protocol is shown in Table 6. In Step 2 the sample is given a laboratory dose D_i , known as the regenerative dose. In step 3 the sample is heated to a preheat temperature, typically for 10 s at 260 °C, in order to empty the shallow thermally unstable TL traps. In step 4 the sample is optically stimulated for 100 s using blue light (typically 470 nm), and the resulting OSL signal (L) is recorded. The optical stimulation at step 4 is carried out at an elevated temperature of 125 °C, in order to avoid complications due to the optically sensitive TL trap at 110 °C. In step 5 the sample is given a small test dose of 0.1 Gy, and in step 6 it is heated for 10 s at a lower temperature of 220 °C (known as the cutheat), to again remove electrons from shallow TL traps. Finally in step 7, the sample is again optically stimulated for 100 s, to measure the OSL signal (T), which is used to carry out the sensitivity correction for the OSL signal measured in step 4. Steps 2–7 in Table 6 are repeated for a sequence of doses D_i , with the first dose D_i taken to be zero in order to measure the OSL signal in the natural sample.

Fig. 8 shows the results of simulating the SAR-OSL protocol. The five regenerative doses used were 0.8β , β , 1.2β , 0 and 0.8β , and two

values of the test dose were used, namely 0.1 Gy and 5 Gy. The preheat temperature used in the SAR protocol simulation was 10 s at 260 °C for the regenerative dose measurements, and the cutheat used for the test dose measurements was 20 s at 220 °C. The sensitivity corrected signals L/T were used to reconstruct the dose–response curve as shown in Fig. 8a, and interpolation is used for estimating the accrued dose by the sample.

The simulation results of the SAR-OSL protocol showed good recycling ratios close to unity, and zero intercepts close to zero. Fig. 8b shows a typical distribution of recycling ratios and zero intercepts for the 100 variants in the model. Fig. 8c shows the results of two sets of simulations, which were carried out using test doses of 0.1 Gy and 5 Gy. It is concluded that the results of the SAR-OSL protocol can be optimized by choosing appropriate values of the test dose. The results of Fig. 8c indicate that the SAR-OSL technique is both accurate and precise in the range of doses 0.1 Gy–10 Gy. The intrinsic precision of this technique gets progressively worse at lower doses in this range.

It is noted that linear interpolation is used in all our SAR-OSL and SAR-TL simulations. In principle, one would expect that non-linear interpolation would give even better accuracy in the simulations.

3.5. Simulation of the TL single aliquot regenerative dose technique (SAR-TL)

Fig. 9a and b shows the results of simulating the SAR-TL protocol developed in analogy to the SAR-OSL protocol during the past 10 years (Wintle and Murray, 2006). The steps in this simulation are shown in Table 7, and are carried out using the TL signal from the 230, 330 and 370 °C TL peaks in the model. The five regenerative doses used were 0.8β , β , 1.2β , 0 and 0.8β , and two values of the test dose were used, namely 0.1 Gy and 1 Gy. The preheat temperature used in the SAR-TL protocol simulation was 10 s at 260 °C for the regenerative dose measurements, and the cutheat used for the test dose measurements was 20 s at 220 °C. The results of the SAR-TL protocol showed good recycling ratio close to unity, and intercepts close to zero.

The sensitivity corrected signals L/T were used to reconstruct the dose–response curve as shown in Fig. 9a, and interpolation was used for estimating the accrued dose by the sample. The results of Fig. 9b indicate that the SAR-TL technique is also sensitive to the test dose used, with more accurate results obtained using a test dose of 1 Gy. Fig. 9c shows the corresponding results using the TL peaks at 230 and 370 °C. The intrinsic precision of this technique can be seen to be good throughout the range of doses examined.

4. Conclusions

The comprehensive quartz model of Pagonis et al. (2008a) was used successfully in this paper to simulate the complete sequence of experimental steps taken during several TL/OSL experimental

protocols for ED estimation. The numerical results from the various simulations are summarized in Table 8 for easy reference.

By inspection of the compiled results in Table 8, we can draw some general comparisons for the relative accuracy and precision of the various luminescence dating methods. The precision of the 230 °C additive TL method is better than 1% for doses above 0.1 Gy, and becomes as poor as 5% at the lowest dose of 0.01 Gy. The additive TL method using the 230 °C appears to be more accurate and more precise than the corresponding additive TL methods using the 330 and 370 °C peaks. This is also obvious from a comparison of Fig. 3b and c, especially at low doses where the precision of the latter methods can be as poor as 30%. Several possible reasons for this difference between additive TL methods were given in the discussion in Section 2.1.

The additive predose technique in Fig. 5b and in Table 8 overestimates the low doses below 0.1 Gy, and overestimates doses higher than 0.1 Gy by as much as ~20%. The corresponding errors are rather large (up to ~22%) in most doses. The multiple activation predose techniques have similar accuracy to the additive dose version, while the precision is in general better with values smaller than 7% in all doses. The phototransfer technique in Fig. 7c can be seen to be both accurate and precise, with precision values less than 2% at all doses. The accuracy of the SAR-OSL technique in Fig. 8c becomes better for higher doses, while some underestimation can be seen for doses smaller than 2 Gy. The precision of this technique becomes worse at low doses (up to 20%), and better at higher doses (2–6%). Finally the SAR-TL techniques in Fig. 9b and c demonstrate overall reasonable accuracy over the whole range of doses, with the 230 °C SAR-TL technique being the most precise with precision values in the range 1–4%.

In general it is found that the various techniques can reproduce natural paleodoses in the range 10 mGy–10 Gy, with typical intrinsic precision of ± 1 –10%. Techniques based on single aliquot protocols were found in general to be more accurate than techniques requiring the use of multiple aliquots. In addition, interpolation techniques yielded better accuracy than extrapolation techniques.

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