



Reliability of single aliquot regenerative protocol (SAR) for dose estimation in quartz at different burial temperatures: A simulation study



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HIGHLIGHTS

- Ambient temperatures affect the reliability of SAR.
- It overestimates the dose with increase in burial temperature and burial time periods.
- Elevated temperature irradiation does not correct for these overestimations.
- Inaccuracies in dose estimation can be removed by incorporating pulsed irradiation procedures.

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ABSTRACT

The single aliquot regenerative protocol (SAR) is a well-established technique for estimating naturally acquired radiation doses in quartz. This simulation work examines the reliability of SAR protocol for samples which experienced different ambient temperatures in nature in the range of -10 to 40 °C. The contribution of various experimental variables used in SAR protocols to the accuracy and precision of the method is simulated for different ambient temperatures. Specifically the effects of paleo-dose, test dose, pre-heating temperature and cut-heat temperature on the accuracy of equivalent dose (ED) estimation are simulated by using random combinations of the concentrations of traps and centers using a previously published comprehensive quartz model. The findings suggest that the ambient temperature has a significant bearing on the reliability of natural dose estimation using SAR protocol, especially for ambient temperatures above 0 °C. The main source of these inaccuracies seems to be thermal sensitization of the quartz samples caused by the well-known thermal transfer of holes between luminescence centers in quartz. The simulations suggest that most of this inaccuracy in the dose estimation can be removed by delivering the laboratory doses in pulses (pulsed irradiation procedures).

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1. Introduction

The past four decades have seen the development of accurate and precise methods for estimating the equivalent dose (ED) in samples containing quartz. One of the most successful of these methods is the single aliquot regenerative (SAR) protocol, which is usually based on optically stimulated luminescence (OSL) signals (Aitken, 1998; Bailiff, 1994, 1997; Wintle and Murray, 2006; Wintle,

2008; Pagonis et al., 2011a, b, c; Chen and Pagonis, 2011; and references therein).

There have been several notable published experimental and simulation attempts to estimate the precision and accuracy of various TL/OSL dating techniques (McKeever et al., 1997; Bailey, 2001, 2004; Bailey et al., 2005; Adamiec, 2005; Pagonis et al., 2003, 2008b, 2011a). McKeever et al. (1997) carried out simulations using a simple OSL model, and concluded that sensitivity changes in their quartz samples depend only weakly on the added doses, and hence should not affect the SAR protocols significantly. Chen et al. (2001) by using a simple model of the SAR-OSL procedure were able to express mathematically the sensitivity-corrected

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dose–response curve (L/T) plotted against the regenerative dose. Bailey (2001, 2004) and Bailey et al., (2005) 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 a certain percentage of the original values. These authors studied the effect of several experimental factors on the accuracy and precision of the SAR-OSL technique, and found that the simulated SAR-OSL technique overestimated systematically the ED of the sample, especially at higher doses. They suggested the use of two possible methods of correcting for these SAR dose overestimations, either using an elevated irradiation temperature in the laboratory or using pulsed irradiation.

Pagonis and Carty (2004) and later on Pagonis et al. (2008a,b), used a modified version of the model by Chen and Leung (1998, 1999) to simulate the complete sequence of experimental steps taken during both the predose dating technique and the SAR-OSL protocol. They concluded that the SAR-OSL protocol is in general less sensitive to the specific experimental conditions than the various predose techniques. Duller (2007) discussed the nature of random and systematic sources of error in SAR-OSL measurements and identified several possible sources of systematic errors, and provided an estimate of the overall “instrumental error” in the SAR protocol at a value of ~1%. Thompson (2007) performed Monte Carlo simulations of SAR OSL and found that the mean ED value overestimated the palaeodose, particularly for larger luminescence measurement errors and larger paleodoses.

While the SAR protocol has been found to be quite reliable for dating of sediments using the OSL signal, it has been found to lead to unreliable results in certain situations (Murray and Wintle, 2000). The phenomena which might disturb the accuracy are the luminescence sensitization and thermal charge transfer, sometimes termed recuperation effect in the SAR protocol (Aitken and Smith, 1988; Murray and Wintle, 1999; Adamiec et al., 2008; Koul et al., 2014). The pre-heating involved in the SAR protocol might also induce these unwanted effects, due to charge transfer between different traps and centers. A typical SAR-OSL for dose estimation protocol involves preheating of the samples. The preheating treatment has been found essential in order to (i) evacuate charge from the traps responsible for the 110 °C TL peak, (ii) remove thermally unstable OSL components and (iii) generate parity in the sensitization of natural and laboratory irradiated samples (Murray and Roberts, 1998). It has been proposed that this disparity between natural and laboratory irradiations is due to in situ sensitization of the samples during their geological and burial history (Wintle and Murray, 1999). This in situ sensitization has been reported to be dependent on (i) the age of the specimen and (ii) the ambient temperature experienced by the sample during its lifetime.

Based on experimental observations and kinetics considerations Koul et al. (2009) postulated the participation of hole reservoir centers (termed R-centers) in the ambient sensitization process of the SAR-OSL signal. Specifically the thermally shallower center, R_1 , known to exist in quartz was observed by simulation to contribute significantly to this sensitization process. One possible mode of sensitization involves transfer of charge from a hole center (R-center) to luminescence center (L-center), and has been termed as the pre-dose sensitization effect (Zimmerman, 1971; Kitis et al., 2006; Pagonis et al., 2003; Oniya et al., 2012; Adamiec, 2005, 2008, 2010; Koul and Chougankar, 2007). In order to explain various quartz phenomena associated with the pre-dose process (thermal activation curves, isothermal sensitization, dose quenching), Bailey (2001) incorporated several R-centers (termed R_1 and R_2) in his quartz model. The main hypotheses governing the pre-dose phenomenon are that (i) a pre-dose (paleo-dose in the case

of natural samples) populates the reservoir R-center, (ii) thermal activation transfers holes from R to the luminescence center L and (iii) the sample sensitization results due to increase in the availability of the activated luminescence centers L. Unlike the transfer of the charge from R to L-center in one step in the laboratory by incorporating a high activation temperature, this charge transfer and associated sensitization might take place in nature as a trickle effect. However, considering the time scales involved, this trickle effect could lead to substantial sensitization over a period of time, especially for older samples.

Recently Koul and Patil (2015) carried out extensive simulation studies and examined the effect of geological and burial temperatures on the ambient sensitization of luminescence emission in quartz. The results of these simulations suggested that the temperatures prevailing during burial time have appreciable impact on the ambient sensitization of quartz, with higher ambient temperature leading to larger sensitization, and vice versa.

Any regenerative procedure for dating, like SAR-OSL, must ensure parity in the sensitization of the natural and artificially irradiated samples. Since the natural in situ sensitization has been reported to be dependent on ambient temperature, this raises the question: does SAR ensure the parity of the two sensitizations at different ambient temperatures prevailing on the globe? The simulations in this paper work were carried out to address this question. To the best of our knowledge, the effect of burial temperature on the SAR protocol has not been simulated before in the literature.

The specific goals of this paper are to:

- Investigate the validity of the SAR-OSL protocol for samples experiencing different burial temperatures in the range -10 °C to 40 °C.
- To study the effect of several variables involved in SAR experiments on the accuracy and precision of the protocol. Specifically the simulations were carried out for different burial doses and burial temperatures, test doses, pre-heating temperatures, cut-heat temperatures and laboratory irradiation temperatures.
- To examine by simulation possible methods of circumventing the inaccuracies introduced in the SAR-OSL protocol due to sample sensitization by exposure to different burial temperatures.

2. Simulation procedure

The simulations in this paper were carried out using the phenomenological general model of Bailey (2001). The model has been very effective to simulate various aspects of luminescence in quartz, and has been found to reproduce most of the phenomena observed in TL and OSL emissions of quartz (Pagonis et al., 2011a; Chen and Pagonis, 2011). The model comprises of five electron and four hole trapping centers. The electron trapping centers consist of the 110 °C trap, a 230 °C trap, fast and medium OSL traps, and a thermally disconnected trap. The hole traps consist of thermally unstable non-radiative recombination centers (R_1 and R_2), thermally stable radiative recombination center (L) and thermally stable non-radiative recombination center. It must be mentioned here that the model has been developed by considering only the 380 nm emission band of quartz. The transport equations describing the traffic of charge with time in various centers and traps of the model:

$$\frac{dn_i}{dt} = n_c(N_i - n_i)A_i - n_i\lambda_i(P, T) - n_i s_i e^{(-E_i/K_\beta T)} \quad (i = 1, \dots, 5) \quad (1)$$

$$\frac{dn_j}{dt} = n_v(N_j - n_j)A_j - n_j s_j e^{(-E_j/K_\beta T)} - n_c n_j B_j \quad (j = 6, \dots, 9) \quad (2)$$

$$\frac{dn_c}{dt} = R - \sum_{i=1}^q \left(\frac{dn_i}{dt} \right) - \sum_{j=q+1}^{q+r} (n_c n_j B_j) \quad (3)$$

$$\frac{dn_v}{dt} = R - \sum_{j=q+1}^{q+r} \left(\frac{dn_j}{dt} \right) \quad (4)$$

$$L = n_c n_g B_g \eta(T) \quad (5)$$

Equations (1) and (2) represent the variation in the charge population with time, for electronic and hole trapping centers respectively. Similarly equations (3) and (4) represent the change in the charge population with time in the conduction and valence band respectively. The luminescence signal occurring due to recombination at the luminescence center L is described by equation (5). The various parameters described in the above equations are; N_i , the total concentration of electron traps (cm^{-3}); n_i , the instantaneous concentration of trapped electrons (cm^{-3}); s_i , the frequency factor (s^{-1}); E_i , the electron trap depth below the conduction band (eV); N_j the total concentration of hole traps (cm^{-3}); n_j , the instantaneous concentration of trapped holes (cm^{-3}); E_j , the hole depth above the valence band (eV); k_B , the Boltzmann's constant (eV K^{-1}); T , the absolute temperature (K); A_i , the conduction band to electron trap transition probability ($\text{cm}^3 \text{s}^{-1}$); A_j , the valence band to hole trap transition probability ($\text{cm}^3 \text{s}^{-1}$); B_j , the conduction band to hole trap transition probability ($\text{cm}^3 \text{s}^{-1}$); λ_i , the optical de-trapping rate (s^{-1}); t , the time (s); η , the luminescence efficiency factor which describes thermal quenching effects; β , the linear heating rate (K s^{-1}) and R , the ionization or pair production rate (s^{-1}). The optimum values assigned to these parameters for different centers and traps of the model are listed in Table 1 of Bailey (2001), as model variant Qtz-A1.

The computer code for numerically solving equations (1)–(5) was written in *Mathematica*, and typical running times for simulating a SAR-OSL procedure was ~30 s on a personal computer. It was found necessary to use a solver for “stiff” differential equations in some of the simulations, and the *Mathematica* software switched automatically between a stiff and non-stiff solver whenever necessary. The simulation procedure used in this paper for the natural quartz sample are identical to the ones used in Bailey (2001), and are listed as steps 1–4 in Table 1. The simulation of the natural irradiation during the geological and burial time

spans was performed by using an R value corresponding to a very slow dose rate of $6.34 \times 10^{-11} \text{ Gy s}^{-1}$, close to the dose rate one would expect in nature. These simulation procedures of the natural sample are of course a simplification, and the actual natural processes are much more complex. However, the simulations provide a useful insight into the various possible luminescence sensitization mechanisms taking place in quartz samples, and help explain some of the experimental inaccuracies previously reported for the SAR protocol. The environmental temperatures generally prevailing on the globe in the range -10 to $40 \text{ }^\circ\text{C}$ were used to represent the burial temperatures experienced by quartz grains in the simulation.

The SAR protocol simulation incorporated a pre-heat at $260 \text{ }^\circ\text{C}$ for 10 s and a cut-heat of 20 s at $220 \text{ }^\circ\text{C}$, as shown in Table 1. The dose recovery was done with different values of the paleodose, test dose, pre-heat and cut heat temperatures and for burial temperatures in the range -10 to $40 \text{ }^\circ\text{C}$. The degree of accuracy of the evaluated doses using SAR was studied with paleo doses of 2, 10, 50 and 100 Gy.

In order to simulate as accurately as possible the accuracy and precision of a typical experimental SAR protocol, the Monte Carlo simulation technique of Bailey (2004) and Pagonis et al., (2011a) was used as follows. Several random variants of the model parameters were generated; for each of these model variants the full sequence of irradiation and thermal history of the samples as well the SAR dating protocol were simulated, in order to obtain an estimate of the intrinsic accuracy and intrinsic precision of the SAR protocol. The accuracy of the simulated SAR protocol was then calculated by comparing the burial dose in the model with the evaluated SAR ED obtained from the simulated SAR protocol.

The precision of the simulated SAR protocol was estimated by using the following method. The experimentally observed variability in TL and OSL characteristics of quartz was simulated by assuming that all the fundamental transition probabilities in the model remained constant, while trap concentrations (parameters $N_1, N_2 \dots N_9$) were allowed to vary randomly within $\pm 80\%$ from the original values in Bailey (2001) by 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.

An initial set of simulations was carried out by using $N = 10$ – 100 random concentration variants and the results are shown in Fig. 1a. Comparison of the results with different N values showed a very small improvement of about 1% in the precision of the simulated results, while the corresponding accuracy of the simulations remained unaffected. On the basis of these initial results, it was decided to carry out the rest of the simulations using $N = 10$ natural variants of the sample, for the sake of saving computation time. Using $N = 10$ variants in the simulation is also closer to a typical experimental SAR protocol in which 5–10 aliquots of the same

Table 1
The simulation steps for a typical SAR-OSL technique. A single aliquot is used for all measurements. Steps 1–4 are a simulation of a “natural” quartz sample according to Bailey (2001), with a variable natural burial dose D.

- 1 . Geological dose irradiation of 1000 Gy at 1 Gy/s
- 2 . Geological time – heat to $350 \text{ }^\circ\text{C}$
- 3 . Illuminate for 100 s at $200 \text{ }^\circ\text{C}$
- 4 . Burial dose D at burial temperature $-10, 0, 10, 20, 30$ and $40 \text{ }^\circ\text{C}$ at a very low natural dose rate of $6.34 \times 10^{-11} \text{ Gy/s}$
- 5 . Irradiate sample with dose D_i at a laboratory dose rate of 1 Gy/s
- 6 . Preheat 10 s at $260 \text{ }^\circ\text{C}$
- 7 . Blue OSL for 100 s at $125 \text{ }^\circ\text{C}$ to record OSL (0.1 s) signal (L)
- 8 . Test dose TD = 5 Gy
- 9 . Cut-heat of 20 s at $220 \text{ }^\circ\text{C}$
- 10 . Blue OSL for 100 s at $125 \text{ }^\circ\text{C}$ to record OSL (0.1 s) signal (T)

Repeat steps 4–10 for the appropriate sequence of doses (0, 0.5D, D, 1.5D, 0 and 0.5D) to construct the L/T dose response and hence obtain the SAR estimated dose (SAR ED).

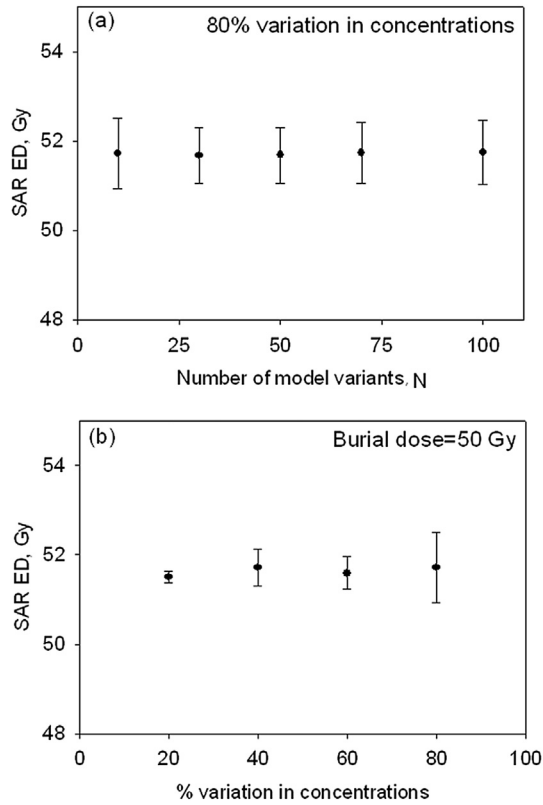


Fig. 1. (a) Simulations of the SAR protocol using $N = 10$ – 100 random concentration variants in the model. The variants are obtained by using a uniform distribution of random concentrations within $\pm 80\%$ of the original values in the model by Bailey (2001). The error bars represent the standard deviations of repeating the simulation N times. As N increases, both the accuracy and the precision (error bars) of the SAR ED value remains the same. (b) Simulations of the SAR protocol using random variants of the concentrations in the model, in the range ± 20 to $\pm 80\%$. The error bars represent the standard deviations of repeating the simulation with $N = 10$ random variants. As the percent variation of the concentrations increases from $\pm 20\%$ up to $\pm 80\%$, the accuracy of the recovered SAR ED remains the same, while the precision of the recovered SAR ED decreases, as shown by the gradually increasing error bars.

sample are commonly used.

A second initial set of simulations was carried out by using $N = 10$ random concentration variants in the range ± 20 to $\pm 80\%$, and the results are shown in Fig. 1b. These results show that as the percent variation of the concentrations increases from $\pm 20\%$ up to $\pm 80\%$, the accuracy of the recovered dose remains the same, while the precision of the recovered SAR ED dose decreases, as shown by the gradually increasing error bars. The simulation results in the rest of this paper are presented for these two extreme situations, for a small variation of $\pm 20\%$ and for a larger variation of $\pm 80\%$ of the concentrations N_i .

It is emphasized that the intrinsic precision and accuracy simulated in this paper are only one part of the overall precision and accuracy encountered during experimental applications of the SAR protocol (see for example Pagonis et al., 2011a for a detailed discussion of sources of error in various luminescence dating techniques).

3. Results and discussion

Some of the important experimental parameters in the SAR protocol, which also have bearing on its reliability, are the burial dose, burial temperature, test dose, pre-heat and cut-heat temperatures. Simulations were carried out with different values of

these parameters and the salient features of the findings are described below. All simulations were carried out by using the protocol shown in Table 1.

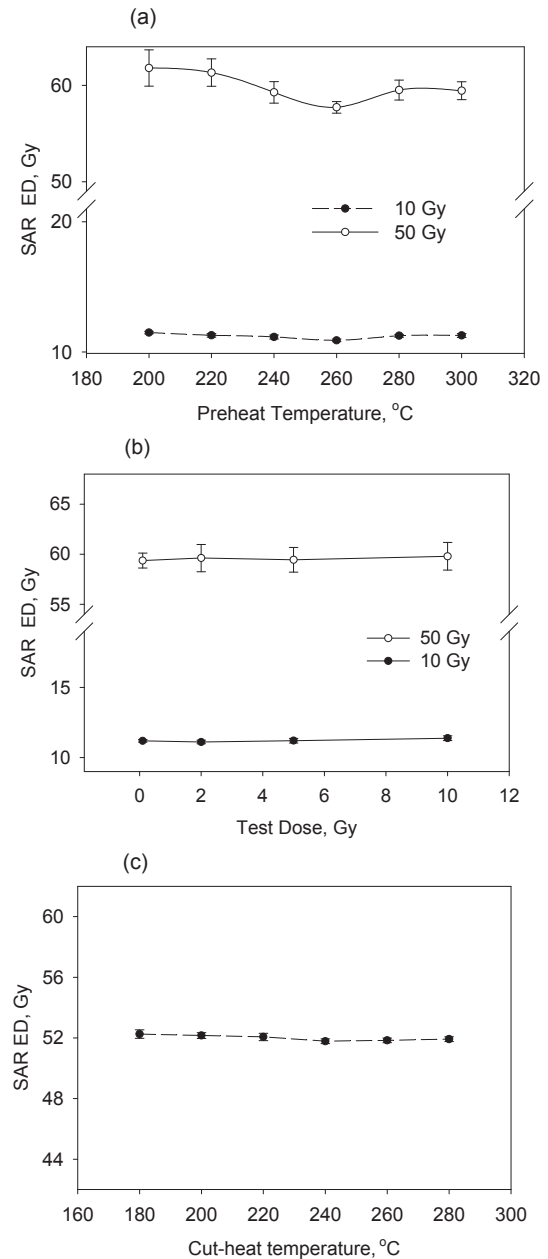


Fig. 2. (a) Simulations of the SAR protocol by varying the preheat temperature in the range 200 – 300 °C and for two burial doses of 10 , 50 Gy. As the preheat temperature is increased, the accuracy of the SAR protocol has a minimum at 260 °C. The error bars represent the standard deviation of $N = 10$ random variants in the model. The SAR protocol is both most precise and most accurate at a preheat temperature of 260 °C. (b) Simulations of the SAR protocol by varying the test dose used in the simulation in the range 0.1 – 10 Gy, and for two burial doses of 10 and 50 Gy. The accuracy of the SAR protocol is practically independent of the test dose, while it is most precise at the smallest test dose of 0.1 Gy (smallest standard deviation of $N = 10$ variants). (c) Simulations by varying the cut-heat temperature in the range 160 – 280 °C. Both the accuracy and the precision of the SAR protocol are practically independent of the cut-heat value.

3.1. Influence of test dose, preheat temperature and cut-heat temperature on the accuracy and precision of the SAR protocol

In order to simulate the precision and accuracy of SAR protocol, simulations were carried out with various preheat temperatures, test doses and cut-heat temperatures. Fig. 2 depicts the results of the simulations with different parameters employed here. Fig. 2a shows the recovered SAR ED as a function of the preheat temperature in the range 200–300 °C and for two burial doses of 10, 50 Gy. As the preheat temperature is increased, the error in the SAR ED has a minimum at 260 °C. The precision of the simulated SAR protocol is estimated by the size of the error bars in Fig. 2a; these error bars represent the standard deviation of $N = 10$ variants in the model. The simulated results in Fig. 2a show that the SAR protocol is both most precise and most accurate at a preheat temperature of 260 °C. It was therefore decided to use an optimal preheat temperature of 260 °C for the rest of the simulations in this paper.

Fig. 2b shows the recovered SAR ED by varying the test dose used in the simulation in the range 0.1–10 Gy, and for two burial doses of 10 and 50 Gy. As the test dose is increased, the accuracy of the SAR protocol stays about the same, practically independent of the test dose. The precision of the simulated SAR protocol is shown by the size of the error bars in Fig. 2b; it is found that the SAR protocol is most precise at the smallest test dose of 0.1 Gy. This result makes physical sense, since a small test dose is less likely to change significantly the luminescence properties of the sample during the SAR protocol, and therefore is also more likely to correct accurately for any sensitivity changes taking place in SAR. It was decided to use an optimal test dose of 5 Gy for the rest of the simulations, since this would correspond closely to SAR experimental protocols.

Fig. 2c shows the recovered SAR ED as a function of the cut-heat temperature in the range 160–280 °C. As the cut-heat is varied in the simulation, the accuracy and the precision of the SAR protocol stays about the same, practically independent of the cut-heat value. The small changes observed in Fig. 2c are of the order of 0.1–0.5%, which are well within the overall accuracy/precision of the simulations. It was decided to use an optimal cut-heat condition of 20 s at 220 °C for the rest of the simulations.

3.2. Influence of burial temperature on the accuracy and precision of the SAR protocol

Fig. 3a shows the recovered SAR ED as a function of the burial temperature in the range –10 to 40 °C and for four burial doses of 2, 10, 50, 100 Gy. These simulations use an optimal preheat of 10 s at 260 °C, a cut-heat of 20 s at 220 °C and a test dose of 5 Gy, based on the results of Fig. 2. The simulations in Fig. 3 were carried out by allowing the concentrations to vary by $\pm 20\%$ from their original values. Fig. 3b shows the same simulated results as Fig. 3a, by plotting the percent accuracy of the SAR protocol versus the paleodose, instead of using the actual SAR ED values as in Fig. 3a.

Inspection of Fig. 3b shows overestimation of the paleodose with increase in both paleodose, itself, and burial temperature. The maximum overestimation can be seen to occur with a dose value of 100 Gy simulated with a burial temperature of 30 °C. Even at a low paleodose value of 2 Gy the SAR protocol overestimates the paleodose by $\sim 2\%$ at an ambient temperature of –10 °C, while this overestimation gradually increases to $\sim 4\%$ at an ambient temperature of 30 °C and remains constant at 40 °C. Similar behavior is seen in Fig. 3b for all four paleodoses of 2, 10, 50 and 100 Gy up to a burial temperature of 30 °C. The inaccuracy in the dose estimation can be, again, seen to increase with dose and burial temperature. However, at the highest simulated burial temperature of 40 °C there is a decrease of the percent error in SAR ED. The inaccuracy of

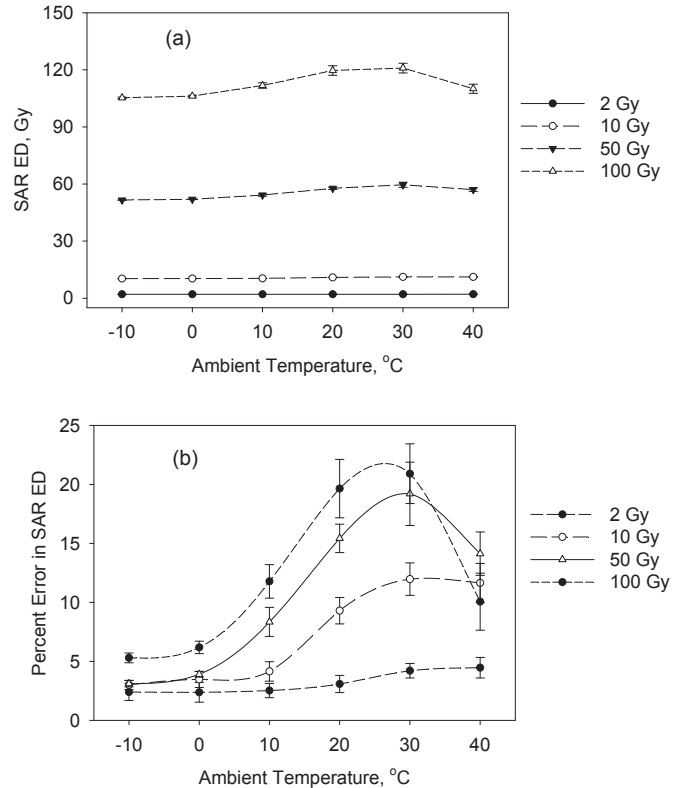


Fig. 3. (a) Simulations of the SAR protocol by varying the burial temperature in the range –10 to 40 °C and for four burial doses of 2, 10, 50 and 100 Gy. (b) The same simulated results as in (a) are shown by using the percent accuracy of the SAR protocol, instead of using the actual SAR ED values. The behavior of the curves in (b) is very similar to the previously simulated behavior of the TL sensitivity obtained by Koul et al. (2015, their Fig. 10a).

the SAR protocol is most pronounced for the highest paleodose of 100 Gy, and in this case the SAR protocol overestimates the paleodose by as much as $(21 \pm 3)\%$.

The behavior of the curves in Fig. 3b is exactly similar to the previously simulated behavior of the TL sensitivity in quartz found by Koul et al. (2015, their Fig. 10a). These authors found that the TL sensitivity remains constant for burial temperatures up to 0 °C, followed by drastic sensitization of the samples in the burial temperature range of 10–30 °C. The similarity between our Fig. 3b and Fig. 10a in Koul et al. (2015) indicates that these two simulated effects most likely have a common origin, namely the in situ sensitization of the quartz samples due to differences in burial temperatures.

The changes in the dose response of the samples were tested using a test dose of 10 Gy. When the burial temperature was changed from –10 to 40 °C, there was a significant change in the test dose response of the OSL signal, of the order of $\sim 10\%$. This effect was even stronger when the quartz samples were pre-annealed, before application of the SAR protocol in the laboratory. Samples annealed for 60 s at 400 °C showed an increased test dose response of $\sim 70\%$. These simulations show that irradiation temperature, annealing temperature and burial temperature affect significantly the response of the sample to a test dose. This is in agreement with the experimental work by Charitidis et al. (1996) and Kitis et al. (1990).

The simulation was repeated by changing the temperature in the geological time step of the Bailey model, within the range 300–350 °C. The results of the recovered dose in the SAR protocol changed very little, of the order of $\sim 0.5\%$. This is well within the

overall numerical limits of the accuracy of the model, and can be considered negligible. The simulation was also carried out by replacing the step of geological heating to 350 °C with a very slow irradiation for 1 million years at 20 °C at the natural irradiation rate of ~1 Gy/ka. The recovered dose from the SAR protocol was ~8% lower than the one using initial geological heating. So, it can be concluded that this step of the simulation is important for the overall accuracy of the simulations. Apparently slow irradiation in nature has a different effect than a short heating to high temperature, which is perhaps due to the fact that heating empties more effectively the traps and centers in quartz.

3.3. Compensating for SAR inaccuracies using pulsed irradiation procedures

Bailey (2004) and Bailey et al. (2005) suggested two possible experimental methods of overcoming inaccuracies in the SAR protocol due to differences in the natural and laboratory irradiation dose rates. Specifically they suggested either the use of elevated irradiation temperatures of ~200 °C during laboratory irradiation of the samples, or alternatively the use of a pulsed irradiation procedure in which the irradiation dose is delivered in several smaller doses, with a heating of the samples between irradiations.

We have explored the effect of these two procedures on the simulations, with the results shown in Fig. 4. In the simulations of

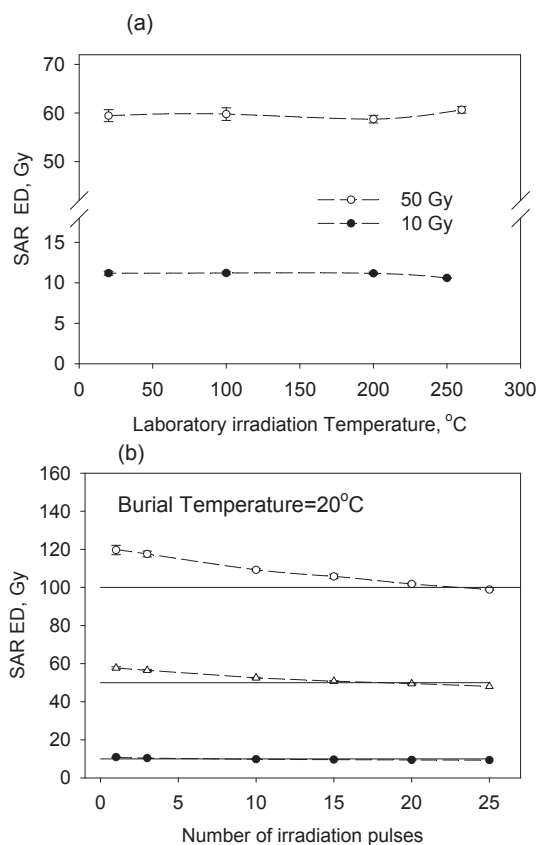


Fig. 4. (a) Simulations of the SAR protocol by varying the laboratory irradiation temperature in the range 20–260 °C, as suggested previously by Bailey (2004). The elevated irradiation temperature has practically no appreciable effect on either the accuracy or the precision of the SAR protocol. (b) The effect of using pulsed irradiation during the SAR protocol, the irradiation dose delivered with a variable number of irradiation pulses ($N_{\text{pulses}} = 1$ –20 pulses), to a sample with a burial temperature of 20 °C. As the number of pulses is increased, the accuracy of the SAR protocol is gradually improved at all three paleodoses (10, 50 and 100 Gy).

Fig. 4a the laboratory irradiation temperature is varied in the range 20–260 °C, and it is clear that using an elevated irradiation temperature has practically no effect on either the accuracy or the precision of the SAR protocol. This provides strong evidence that the SAR ED overestimations seen in our Fig. 3 most likely has a different physical origin than the dose rate effect studied by Bailey et al. (2005). This topic is considered further in the Discussion section of this paper.

Fig. 4b shows the effect of using pulsed irradiation during the SAR protocol, the irradiation dose delivered with a variable number of irradiation pulses ($N_{\text{pulses}} = 1$ –20 pulses), on the accuracy of the SAR protocol. Each pulse is followed by heating of the sample to 240 °C, as suggested by Bailey (2004). As the number of pulses, N_{pulses} , is increased, the accuracy of the SAR protocol is gradually improved at all three paleodoses (10, 50 and 100 Gy), and the correct paleodose is obtained when more than ~10–15 pulses are used. A typical case of sample experiencing a burial dose of 20 °C is depicted in Fig. 4b. The number of pulses required to attain the correct value is directly proportional to the magnitude of the paleodose.

Fig. 5 shows the result of repeating the SAR simulations of Fig. 3 by using pulsed irradiation in 10 smaller doses ($N_{\text{pulses}} = 10$). Fig. 5b shows that administering pulsed irradiation can significantly correct for the overestimation of the paleodose by the SAR protocol, in case of all irradiation doses and all burial temperatures. Fig. 6 shows the same results as in Fig. 5, by plotting the PULSED-SAR ED as a function of the burial dose. The results in Fig. 6 are close to the 1:1 line within the precision shown by the error bars, indicating that

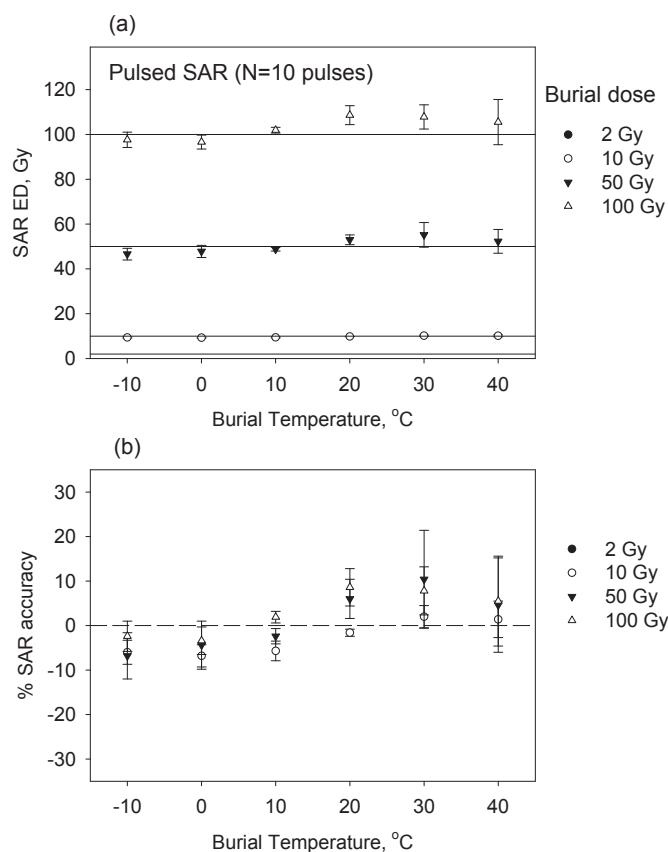


Fig. 5. (a) and (b): The simulations of Fig. 3 are repeated by using pulsed irradiation in 10 smaller doses ($N_{\text{pulses}} = 10$). Using pulsed irradiation can correct to a larger extent for the overestimation of the paleodose by the SAR protocol, at all irradiation doses and at all burial temperatures.

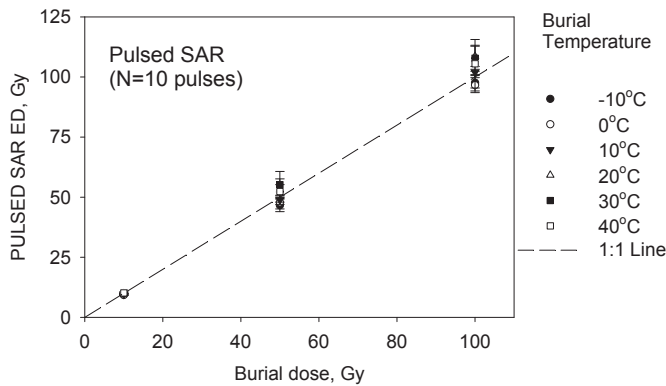


Fig. 6. The same results as in Fig. 5 are presented by plotting the PULSED-SAR ED as a function of the burial dose. These results are close to the 1:1 line within the precision shown by the error bars, indicating that the PULSED SAR protocol can adequately correct for the ED overestimations caused by sensitization of the samples during their burial.

the SAR protocol used with a pulsed irradiation can adequately correct for the sensitization of the sample taking place during their exposure to different burial temperatures.

4. Discussion and conclusion

Bailey (2004) and Bailey et al. (2005) attributed the SAR ED overestimation to the slow natural dose rate being many orders of magnitude smaller than the dose rates used in the laboratory. They concluded that this effect is due to different degree of competition for electrons from the conduction band encountered during natural and laboratory irradiation. Specifically, the difference between dose response in the laboratory and in nature was attributed to the thermally shallow hole center R_1 . They suggested that slow irradiation in nature leads to a relatively low equilibrium concentration of holes in this center. On the other hand, the high dose rates in laboratory irradiations lead to a considerably higher equilibrium concentration occurring in R_1 , which then leads to an overestimation of the SAR ED values.

The results of Fig. 4a show that using elevated irradiation temperatures does not correct for the SAR ED overestimations seen in Fig. 3; this is in contrast to the simulation and experimental results of Bailey (2004) who found that elevated irradiation temperatures can correct for SAR overestimations. Clearly, then the SAR overestimations due to elevated burial temperatures are related to, but do not seem to have the same mechanism as the dose rate effect studied by Bailey et al. (2005). By contrast, the results of Fig. 4b show that pulsed irradiation can correct for SAR overestimates caused by either differences in dose rate, or by elevated temperatures in nature. It looks that more experimental and simulation work is needed to clarify the exact differences in the mechanisms of these two phenomena. However, they both seem to have their ultimate result in the sensitization of luminescence signals of quartz samples.

It is also noted that in a recent experimental study Chapot et al., (2014) compared constant and pulsed-irradiation thermally transferred OSL protocols (TT-OSL) and investigated the possibility of thermal erosion of the TT-OSL dating signal. These authors concluded that partial annealing of the TT-OSL signal could indeed explain the difference between constant- and pulsed-irradiation protocols. It would then be interesting to extend the simulations in this paper to a study of TT-OSL signals from quartz, and examine in this manner the mechanism behind these two related phenomena, namely sensitization and thermal transfer of charge in quartz.

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