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Research paper

Implications of broad dose distributions obtained with the single-aliquot regenerative-dose method on quartz fine-grains from loess

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Abstract

Six samples were collected from a section of Peoria Loess in Eustis, North America, for optically stimulated luminescence (OSL) dating of quartz, and all except one (LV90) produced narrow dose distributions. A comparative study was conducted on this sample and on a 'well-behaved' sample (LV91), involving other dating methods and examination of the quartz OSL. These investigations revealed differences in the quartz OSL growth with dose, OSL response to thermal treatments and the range of components within the OSL signals. An ultra-fast component was found in LV90 that displayed a higher rate of sensitivity change than the fast component and this had a malign influence on the determination of the equivalent dose. The distinctive luminescence characteristics of LV90 imply either a change in wind dynamics and/or the source area for the silt.

Keywords: OSL dating; SAR; Quartz; Dose distribution; Ultra-fast component; Loess

1. Introduction

The single-aliquot regenerative-dose (SAR) method (Murray and Wintle, 2000) for optically stimulated luminescence (OSL) dating, was developed on quartz coarse-grains and has been shown to yield excellent precision on the equivalent dose (e.g. Murray and Olley, 2002). Applying SAR to silty sediments involves a far more difficult sample preparation to extract the quartz than for sand samples. Banerjee et al., 2001, circumvented this problem with the introduction of a double SAR protocol. Feldspar grains in a polymineral sample are likely to dominate OSL signals, but they are also more sensitive to infra-red (IR) stimulation than quartz (Spooner, 1994). IR was therefore employed to reduce the trapped charge in the feldspars so that in the following blue-LED stimulation the signal from quartz, referred to as post-IR OSL, would be dominant. Stokes et al., 2003a, successfully employed this method on Loess from China, providing concordant ages for IR and post-IR OSL signals, but it produced discordant ages for another Chinese site (Roberts and Wintle, 2001).

It is preferable to apply the standard SAR protocol to a quartz extract, and so Mauz and Lang (2004) investigated both the efficiency of acid digestion and the methods to detect the presence of feldspar after acid treatment. Following their procedures, we chemically isolated the quartz fraction in order to determine ages for a section of Peoria Loess at Eustis, Nebraska, USA (Rousseau and Kukla, 1994; Rossignol et al., 2004). In general the results were excellent, but one sample gave us cause for concern. Our attempts to understand this variant behaviour are presented in this paper, and the implications for optical dating and loess accumulation at the site are discussed.

2. Samples and initial SAR results

Six samples (LV86-91) were collected from the Peoria Loess at the Eustis section (Fig. S1) with due care to shield them from exposure to light. Risø DA-15 TL/OSL readers recorded the luminescence of 11–20 μm quartz grains,

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extracted via standard methods and a 20% hydrofluoric acid (HF) digest (see online supplementary file). We also prepared 4–11 μm and 11–20 μm polymineral aliquots (no HF digest) for comparative studies on two samples.

SAR parameters are given in the online supplementary file and the mean equivalent dose (D_e) and standard deviation are listed in Table S1. The relative standard deviation (RSD) is a guide to reproducibility, and it is clear that most samples show little inter-aliquot variation (RSD from 5–9%), but the RSD for LV90 is 18%. Is the higher amount of scatter in LV90 acceptable for an undisturbed, aeolian deposit? Watanuki et al., 2003 achieved RSDs of 2–7% when dating Chinese loess, and Stokes et al., 2003b reported RSDs of 5–13% for deep sea sediments (deposited from dust plumes originating in nearby landmasses).

We compared the growth curves of a "well-behaved" sample, LV91 and LV90 (Fig. 1). The latter has a more linear growth than the former, with a mean characteristic saturation dose (D_0) of 148 ± 68 Gy, compared with 110 ± 9 Gy. However, both samples responded well to a dose recovery procedure (Murray and Wintle, 2003), thus encouraging confidence in the chosen SAR protocol. A beta dose of 96 Gy was given to 12 bleached aliquots, and the SAR routine used previously for natural aliquots of LV91 was employed to recover the dose. The result for LV90 was more accurate than LV91 (ratio of recovered to given dose was 0.97 ± 0.08 and 0.95 ± 0.03, respectively), but it was less precise (RSD 8% compared to 3%). Both results are acceptable, and it is clear that reproducibility for LV90 is much improved when a lower preheat temperature is employed (compare with Table S1). We attempted to discover the causes for the broad dose distribution displayed by natural aliquots of LV90 by exploring the luminescence behaviour of LV90 and LV91.

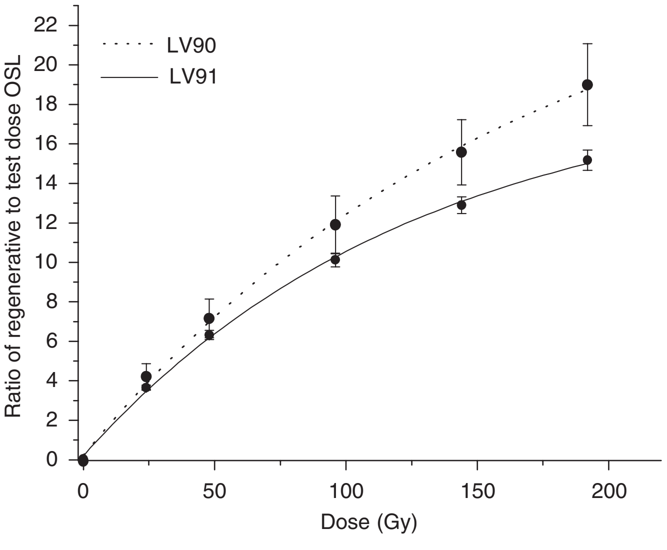

Fig. 1. Response to dose for LV90 and LV91 based on the mean ratios of regenerative to test dose signals in the SAR procedure on quartz aliquots (for LV90 $n = 15$; for LV91 $n = 35$). Error bars at one standard deviation.

3. Luminescence investigations on LV90 and LV91

A wide range of D_e may reflect sedimentary properties (such as poor bleaching during transport, or post-depositional mixing), or the luminescence properties of the quartz and thus the suitability of the SAR procedure. Another possible cause, feldspar contamination, was rejected as each aliquot was subjected to two tests to assess purity (see online supplementary file). Although poor bleaching and in situ mixing of grains is also thought unlikely for LV90, we investigated these possibilities by employing other luminescence dating methods to ascertain if this sample continued to present poor reproducibility. The study on quartz grains included decay curve shape, sensitivity changes and component-defined behaviour. For the latter we applied the linearly modulated (LM) technique (Bulur, 1996) for OSL measurement.

3.1. Other luminescence dating methods

We employed a double SAR procedure on polymineral aliquots (11–20 μm grains) to compare dispersion in both IR and post-IR D_e estimates, and to compare these with the results of the standard SAR protocol. The preheat was 200 °C/10 s, as previously used for quartz aliquots of LV91. We also used an IR-only stimulation in a SAR protocol (IRSL-SAR). This was done using the same preheat (temperature and duration) for regenerated and test-dose signals, as suggested by Blair et al., 2005, and a blue-transmission detection filter.

Additionally we employed the multiple aliquot additive dose method (MAAD) on polymineral 4–11 μm grains, detecting IRSL signals with the blue filter. This technique compared favourably with SAR procedures in studies by Banerjee et al. (2001) and Lepper et al. (2003), and has been successfully employed by Lang et al. (2003), for dating a loess section in Germany. Only one D_e is obtained with this method, and the error relates to scatter on the growth curve.

The D_e estimates (mean and standard deviation) for all the methods are listed in Table S2, and they are all lower than the quartz SAR estimates. The IRSL D_e from the double SAR method has the lowest value; this may be due to the influence of unstable UV emissions (Clarke and Rendell, 1997) and/or anomalous fading (Aitken, 1998). The post-IR data present an unexpected exchange in the level of reproducibility as LV91 has the highest RSD. We observed that the blue-stimulated signals for both samples were quite different to those of quartz aliquots. We suspect that our post-IR signals are dominated by feldspar emissions (a preponderance of traps that are stimulated only by low wavelengths; Blair et al., 2005; Duller and Bøtter-Jensen, 1993).

The results from this investigation indicate that precision on IRSL D_es is equally good for both samples, and suggests the problem with LV90 must relate to the quartz OSL. We also note that for every method the D_es for both samples are within errors of each other.

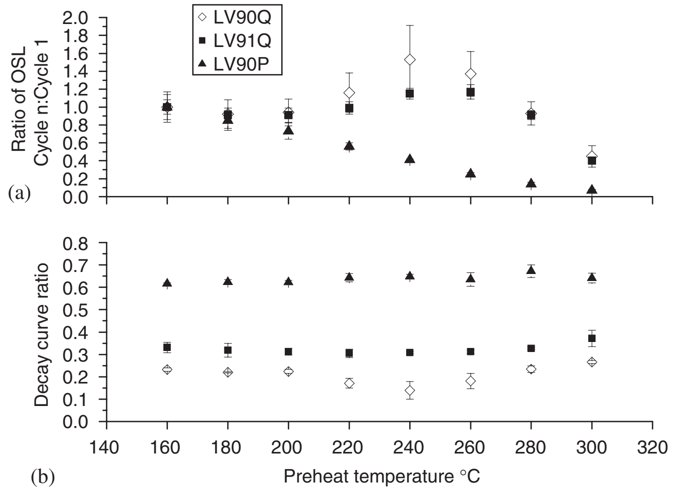

Fig. 2. Changes in (a) sensitivity and (b) decay curve ratios with increasing preheat temperatures for quartz (Q) aliquots of LV90 and LV91, and polymineral (P) aliquots of LV90. In Fig. 2a the mean net initial OSL signal (0–0.32 s minus the background equivalent from the last 3.2 s) of three aliquots for each sample is normalised to the first preheat cycle of 160 °C.

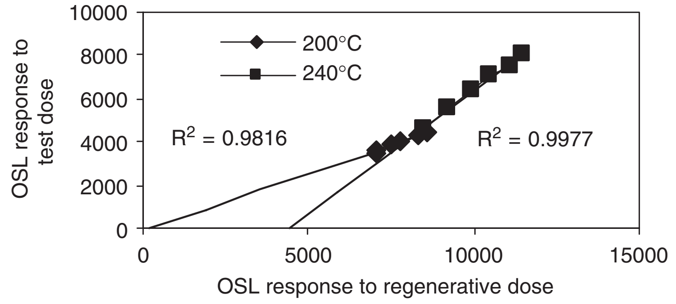

Fig. 3. Plot of the mean net initial signal from a repeated regenerative dose (L_i) against the test dose signal (T_i) for quartz aliquots of LV90 ($n = 3$). One set had a preheat of 200 °C/10 s, whilst the other had 240 °C/10 s for L_i. Both sets had a cut-heat of 200 °C for T_i.

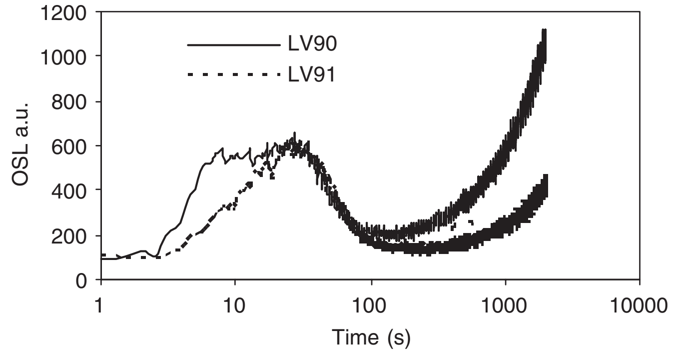

Fig. 4. LM-OSL curves (semi-log plot) for bleached and irradiated (96 Gy) quartz aliquots of LV90 and LV91 after a preheat of 200 °C/10 s. Stimulation was for 2000 s at 160 °C on Risø 2.

3.2. *Thermal effects and sensitivity changes*

3.2.1. *Experiment A*

Quartz and polymineral aliquots of LV90 and LV91 were subjected to repeated cycles of irradiation (12 Gy), preheat of 160 °C/10 s and blue-LED stimulation. On another set of aliquots the preheat was increased by 20 °C in each successive cycle.

There was a decrease in sensitivity with the fixed preheat for all aliquots, and for the polymineral aliquots when the temperature was increased. For quartz aliquots there was a trend of increasing sensitivity between 220–260 °C, and a decrease after 260 °C (Fig. 2a; also reported by Wintle and Murray, 1997). We determined decay curve ratios (net signal at 0.96 s stimulation time divided by the net signal at 0.16 s stimulation time) to ascertain if thermal treatment affected the shape of the OSL signal. The decay curve ratios for polymineral and LV91 quartz aliquots were unaffected by the increase in preheat temperature, but quartz aliquots of LV90 appeared to have a faster decay rate after a 240 °C preheat (Fig. 2b).

3.2.2. *Experiment B*

Quartz aliquots of LV90 were given repeated cycles of irradiation (12 Gy), preheat of 200 °C for 10 s or 240 °C for 10 s, and stimulation (for regenerative OSL, L_i). A 6 Gy test dose (for T_i) with a cut-heat of 200 °C was included after each cycle.

Sensitivity change is only correctly monitored by T_i when the preheat and cut-heat are at 200 °C (Fig. 3). The results from these experiments show that the two samples have different responses to thermal treatments. The behaviour of LV90 in Experiment B is similar to that reported by Bailey (2000), showing that, despite good recycling ratios, the sensitivity may not be monitored correctly by a SAR procedure.

3.3. *OSL components*

Smith and Rhodes (1994) proposed that the quartz OSL signal had fast, medium and slow components. Since the development of LM-OSL (Bulur, 1996) an ultra-fast component (UFC) has also been identified (Jain et al., 2003). Research by Bulur et al. (2000), Jain et al. (2003) and Singarayer and Bailey (2003) showed that the various components of quartz have different thermal stabilities, growth with dose and rates of sensitisation. We conjectured that LV90 might contain quartz grains that possessed a different range of components than LV91 so we applied LM-OSL to both samples. An extra signal was present in the quartz aliquot of LV90 at the beginning of the stimulation, which we deduce is an UFC (Fig. 4). It is present at 200 and 240 °C, and although Jain et al. (2003) reported that the UFC was removed completely by heating to 250 °C, further research has shown it to be stable to 300 °C in other samples (Jain, pers. comm.). The slow components were also much larger in LV90 than in LV91, being twice the intensity of the other peaks.

We examined sensitivity changes in the UFC and fast component (FC) with the following experiment.

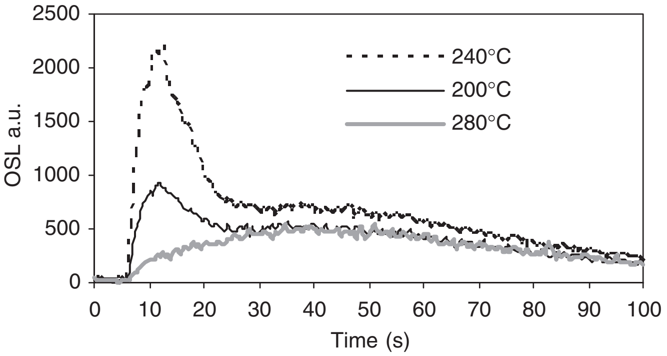

Fig. 5. Selected LM-OSL curves for the first 100 s for a quartz aliquot of LV90 after increasing preheat temperatures. OSL signals were recorded for three sets of cycles of a repeated regenerative dose for three preheat temperatures. The LM-OSL curves are from Cycle 3 (200 °C), Cycle 4 (240 °C) and Cycle 7 (280 °C). Stimulation was for 2000 s at 125 °C on Risø 1.

3.3.1. *Experiment C*

A bleached aliquot was subjected to a 47 Gy regenerative dose and a preheat of 200 °C/10 s for three cycles before increasing the preheat temperature to 240 °C for three cycles, and finally to 280 °C (Table S3). Sensitivity change was monitored by a 12 Gy test dose and a cut-heat of 200 °C. We used the integral photon count from 8–18 s to represent the OSL sensitivity of the UFC, and the integral 35–45 s for the FC.

For the low preheat cycles the UFC appears to show greater change in regenerative signals (L_i) than for test dose signals (T_i) as there is a 60% increase in the former but only a 35% increase in the latter from cycles 1 to 3. Sensitivity increases by a factor of two for both L_i and T_i when a 240 °C preheat is applied, but although T_i reflects the changes in L_i it is not at the same magnitude (Fig. S2). After a preheat of 280 °C the UFC is greatly reduced (Fig. 5), but T_i increases by a factor of two (Fig. 6a). The second peak, dominated by the FC, exhibits a much lower rate of sensitivity increase than the UFC (Fig. 6b). After a preheat of 240 °C there is only a 36% increase in sensitivity of L_i and a 24% increase in T_i.

These results indicate that a SAR procedure incorporating a 240 °C preheat would induce greater change in the sensitivity of the UFC than the FC, and affect the relative proportions of these two components. The UFC would then have more influence on rate of sensitivity change and response to dose of the net initial signal (0–0.32 s). This explains the poor reproducibility observed in the SAR data from natural aliquots, and the improvement witnessed in the dose recovery experiment.

4. Solutions and discussion

The malign influence of UFCs was encountered by Choi et al. (2003), when applying SAR to Korean sediments. They were able to employ a cut-heat of 200 °C to remove the UFC, but the UFC in LV90 is still present after a 240 °C preheat. Results from our preheat experiment using LM-OSL suggests that the optically insensitive 280 °C thermoluminescence (TL) peak may be responsible for charge transfer to the UFC-traps of the 325 °C TL/OSL trap system (Wintle and Murray, 1997). We would probably need a preheat of 300 °C to completely remove the UFC, and this would also cause thermal erosion of the FC.

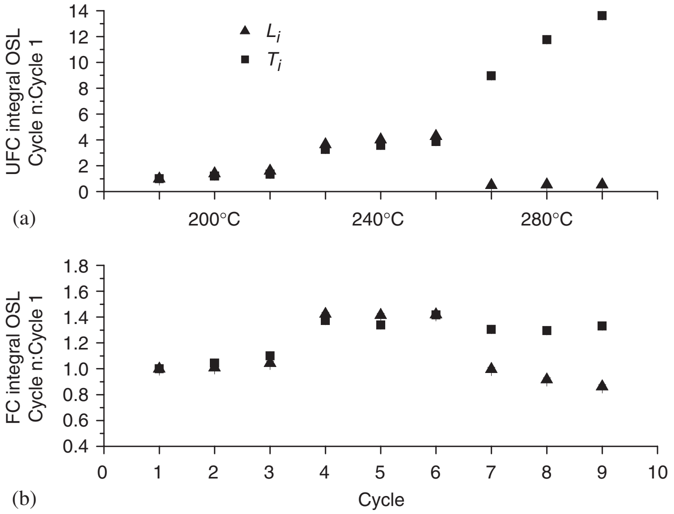

Fig. 6. Sensitivity change in (a) the UFC (integral 8–18 s) and (b) the FC (integral 35–45 s) for regenerative (L_i: triangles) and test dose (T_i: squares) signals for nine cycles of repeated doses at three different preheat temperatures. A background signal after a preheat of 350 °C was subtracted from all LM-OSL curves, and each integral was normalised to the first preheat cycle at 200 °C. Errors are based on counting statistics.

The results from Experiment B indicated that sensitivity change could be monitored successfully provided the preheat and cut-heat were at 200 °C. With this protocol we achieved a narrower range of D_e (RSD 7%) comparable with the other samples. It is interesting to note that the mean D_e has not changed (Table S4). However, the influence of the UFC is still present and likely to provide an erroneous D_e due to differences in charge transfer in the natural and laboratory-irradiated measurements (Wintle and Murray, 1997). We attempted to avoid the UFC by taking the net OSL signal from later in the stimulation (0.32–0.64 s instead of 0.0–0.32 s) as suggested by Jain (pers. comm.). This approach made no difference to the mean D_e for LV91, but increased the D_e for LV90 by 20% (Table S4). Both samples have similar D_es with this method, as was found for other luminescence dating techniques (Section 3.1 and Table S2). As reproducibility is improved with the 200 °C preheat, we might conclude that the “later-light” method is likely to provide greater accuracy in the $D_\mathrm{e.}$ However, the “later-light” method also produced a higher D_e for the dose recovery experiment on LV90 (recovered to given dose ratio of 1.17 ± 0.11). These conflicting results reduce confidence in the estimate for the palaeodose achieved with “later-light” method.

The different quartz luminescence signatures in these loess samples may have their origin in quartz from different source areas, and so the presence/absence of one of the characteristics may reflect changes in wind speed or direction during accumulation of the loess at Eustis. Such changes may be important for our understanding of atmospheric global circulation patterns during the Late Glacial (Roberts et al., 2003). We are investigating the composition of the loess to obtain corroborating evidence for our hypothesis of fluctuations in winds.

5. Conclusions

Our investigations confirmed our hypothesis that the broad dose distribution for LV90 was caused by quartz grains that had a different OSL signature to the other samples. This sample was more sensitive to thermal treatments in the SAR procedure because of temperature-induced sensitivity change in the UFC, possibly related to charge transfer from the 280 °C TL trap. It is possible that the relative proportions of this UF-quartz and "normal"-quartz varied between aliquots thus resulting in poor reproducibility.

As research progresses, it is becoming more evident that 'well-behaved' quartz, i.e. with a dominant fast component, is not always present. Our experience shows that a range in D_e may simply reflect an unsuitable SAR protocol or "abnormal" quartz signals. The variation in quartz signatures at Eustis may indicate a change in sediment derivation for the loess, providing a useful environmental indicator and highlighting the potential of quartz luminescence as a tracer for source rocks.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.quageo.2006.05.022.

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