Concerning the normalization of additive dose optically stimulated luminescence data from quartz

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Abstract: *Since 'natural normalization' is not possible for low dose samples, several 'second dose' normalization procedures are compared. The use of light and of heat for bleaching before the second dose is considered. Attention is paid to any indication of second dose sensitivity being dependent on the magnitude of the first dose.*

Introduction

Since the introduction of optically stimulated luminescence dating by Huntley et al. (1985) some aspects of the technique have been developed analogously to established aspects of thermoluminescence dating procedures while others are unique to optical stimulation. The concern here is with one particular aspect of additive dose optically stimulated luminescence dating, namely sample to sample normalization when stimulating quartz of low natural dose with green light. 'Natural normalization', that is the use of the luminescence intensity resulting from a brief exposure of each sample to stimulating light prior to the addition of any laboratory dose, provides a very attractive approach to compensating for sample to sample variations (Stokes, 1992) and indeed natural normalisation has been much used with the infrared stimulation of feldspar (Wintle, 1993; Ollerhead et al., 1994). However the accuracy achievable from natural normalization must become less as the natural dose approaches zero and the method cannot be applied to investigations of the dose response properties of bleached material. In these circumstances normalization must be by an appropriate second measurement on the sample and Stokes (1992), using an argon ion laser for stimulation of young quartz, reduced the luminescence to about 1% of the initial value and then applied a dose of 5 Gy to each sample for normalisation. Variations on second dose normalization are compared below, involving the use of light or heat for bleaching before the second dose and looking for any indication of second dose sensitivity being dependent on the magnitude of the first dose.

The samples and measurement system

The quartz samples were prepared from BDH "acid washed sand" which was treated with concentrated HF for one hour. The purity of the quartz after this treatment was tested by verifying, for some randomly selected samples, that no luminescence was stimulated by exposure to infra red radiation. Any such signal would have indicated the presence of feldspar, the most probable contaminant. Some of the quartz was bleached by exposure to daylight for at least one week and some by heating for 5 minutes at 500°C before further use. Samples for measurement were deposited on silicone grease in the central portion of thin stainless steel discs 12 mm in diameter. Dosing was by exposure to a calibrated 90Sr beta source. A 16 green LED system with peak emission at 565 nm wavelength was used for stimulation (Galloway, 1992, 1993) while a carefully chosen filter combination of HA3, BG39, UG11, 7-59 and 7-60 filters, with a peak transmission at 365 nm wavelength, preceded the photomultiplier (EMI type 9635QA) which counted the luminescence photons. The multi-sample system described by Galloway (1991) was used for the measurements.

Method of measurement

First of all for each sample the background due to scattered light and photomultiplier noise was measured. A beta dose was then given, the sample preheated for 1 minute at 220°C to remove any unstable signal component and the luminescence measured. This was repeated for three samples for each dose and for eight doses ranging from 12.5 Gy to 175 Gy and the samples were then normalized to generate a growth curve showing the dependence of luminescence on dose. A separate set of 24 samples

was processed in this way for each of the normalization procedures considered.
Normalization required that the samples be bleached prior to application of the normalizing beta dose and bleaching was either by placing the samples in a Honle SOL-2 "solar simulator" for 1 hour or by heating the samples to 450°C for 1 minute. The sensitivity to the normalizing beta dose might depend on the dose to which the samples had been exposed before bleaching, as can occur in the case of thermoluminescence, Aitken (1985). To allow for this possibility the normalized dose response was compared for samples which had a second beta dose added to bring all to the same total dose before bleaching, with samples which had not; this was applied in both the SOL-2 and the heat bleaching procedures. In addition to these four normalization methods, one further procedure was investigated, based on a "single aliquot" approach developed for infra red stimulation of feldspar (Duller, 1991, 1992) and tested to a limited extent with green stimulation of quartz (Galloway, 1994). In this variant, each sample had a second beta dose added so that the sum of the two beta doses equalled the maximum beta dose applied to any of the set of samples to be normalized. The sample was then preheated, read and compensation applied in the single aliquot manner for loss of signal due to the first preheating and reading.

The normalized dose response measurements

a) Quartz bleached by daylight

Dose response curves for quartz initially bleached by daylight and normalized by four of the procedures outlined above are shown in fig. 1. It is immediately clear that one of the methods of normalization, involving heating of the samples to 450°C before applying the normalizing dose, is not successful in reducing the scatter in the data. The two procedures which involved giving all samples an equal dose before normalization, one with bleaching by the SOL-2 and the other developed from the single aliquot approach, show an essentially identical dose response with little scatter in the normalised data. The procedure in which the samples were not given equal doses before bleaching and normalization gives a response which shows little scatter but falls below the others. This could be due to the sensitivity after bleaching increasing with previous dose. That preheating can cause thermal sensitization analogously to the thermoluminescence pre-dose effect has been observed by Godfrey-Smith (1994). The data in fig. 1 were measured with a 100 s exposure time to the green LEDs. The more encouraging normalization procedures, that is excluding the one involving 450°C heating as a bleach, were investigated further using green light measurement times of 50 s and 12.5 s with the results shown in figs. 2 and 3. These measurements confirm that the two procedures which involved giving all samples an equal dose before normalization show an essentially identical dose response with little scatter in the normalised data, while the procedure in which the samples were not given equal doses before bleaching and normalization gives a response which falls below the others.
However extending the investigation to higher doses at which saturation becomes more significant, fig. 4, shows that normalization based on measurements corrected in the single aliquot manner is no longer satisfactory; the simple correction procedure is strictly applicable only when the luminescence response to dose is linear (Duller, 1994).

b) Quartz bleached by heating at 500°C

The tests on quartz initially bleached by daylight should be relevant to sediment dating so for completeness similar tests were carried out on quartz which had all luminescence removed by heating which should be relevant to the dating of pottery. The response curves for five different normalization procedures are shown in fig. 5 and in terms of scatter in the normalized luminescence values there is nothing to choose between the procedures. Four out of the five procedures give consistent results and only the procedure in which the samples were not given equal doses before bleaching by the SOL-2 and normalization gives a response which differs by falling below the others. This procedure showed the same tendency in the tests on daylight bleached quartz in figs. 1, 2 and 3. The initially heated quartz behaved differently from the initially daylight bleached quartz discussed in section (a) above when heating to 450°C was used as a bleach prior to application of the normalizing beta dose, in that entirely acceptable normalization resulted (compare fig. 5 and fig. 1). Further, when 450°C heating was used as a bleach the normalization was not dependent on whether the samples had received the same dose before normalization, in contrast to the situation when the SOL-2 was used for bleaching.
It is interesting to note in passing that the dose response curves for the daylight bleached quartz show a different relationship between luminescence and dose from those for the 500°C heated quartz. Initially the response of the former is approximately linear, up to about 25 Gy say, whereas the latter is clearly non-linear in this region.

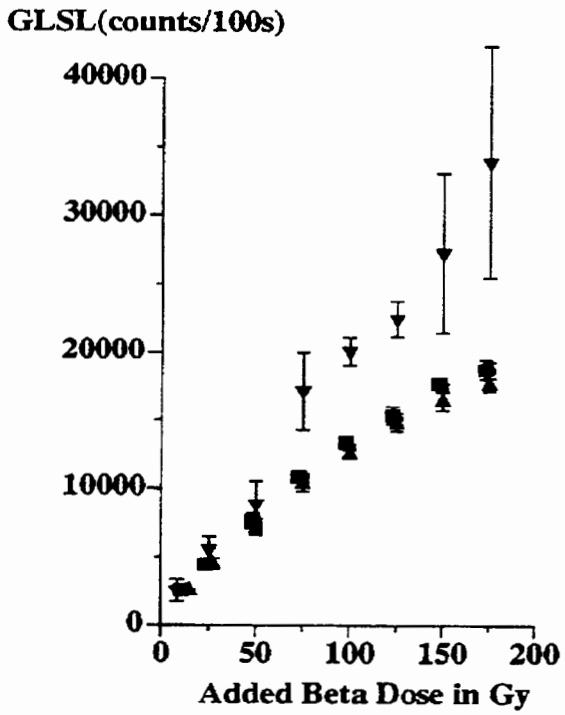

Figure 1
The dependence of the green light stimulated luminescence (GLSL) on beta dose for quartz previously bleached by daylight. The luminescence was measured for 100 s. Each point represents the mean of three measurements while the error bars indicate the spread in the three measurements. Four different normalization procedures are compared.

(1) samples were brought to the same total beta dose (175 Gy) before bleaching in the SOL-2 and application of the normalizing dose, indicated by squares.

(2) samples were brought to the same total beta dose (175 Gy), preheated and read for normalization with correction in the single aliquot manner, indicated by circles (which are indistinguishable from the squares on which they are superimposed).

(3) samples were bleached in the SOL-2 without bringing to the same total dose and the normalizing dose applied, indicated by upward pointing triangles.

(4) samples were brought to the same total dose (175 Gy) then bleached by heating to 450°C before application of the normalizing dose, indicated by downward pointing triangles and large error bars.

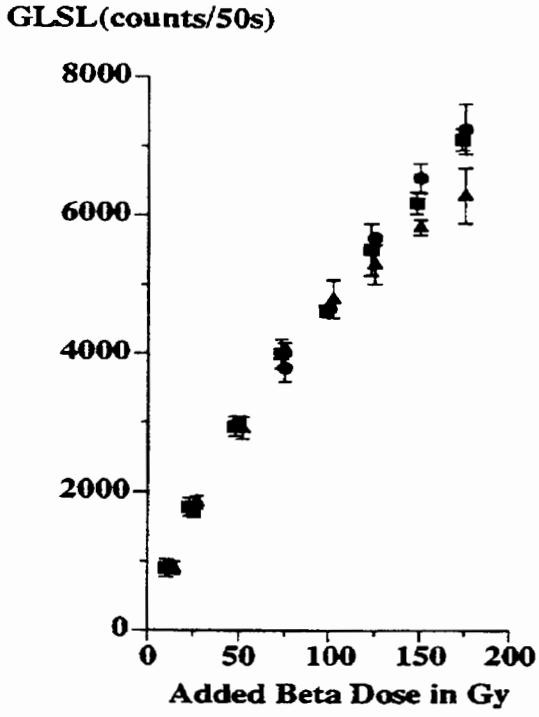

Figure 2
As fig. 1 but for 50 s green light stimulation, omitting the least successful normalization procedure involving heating to 450°C.

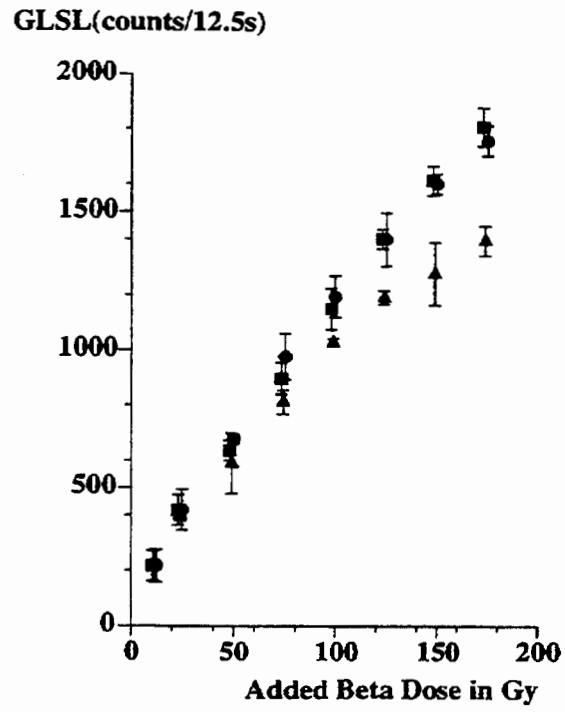

Figure 3
As fig. 2 but for 12.5 s green light stimulation.

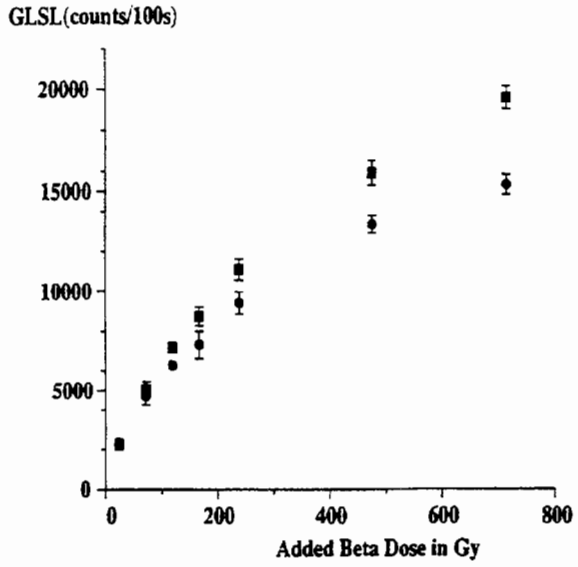

Figure 4
The dependence of the green light stimulated luminescence (GLSL) on beta dose for quartz previously bleached by daylight, as in fig. 1 but extending to higher doses at which saturation becomes more significant. Each point represents the mean of three measurements while the error bars indicate the spread in the three measurements. Two different normalization procedures are compared.
(1) samples were brought to the same total beta dose (700 Gy) before bleaching in the SOL-2 and application of the normalizing dose, indicated by squares.
(2) samples were brought to the same total beta dose (700 Gy), preheated and read for normalization with correction in the single aliquot manner, indicated by circles.

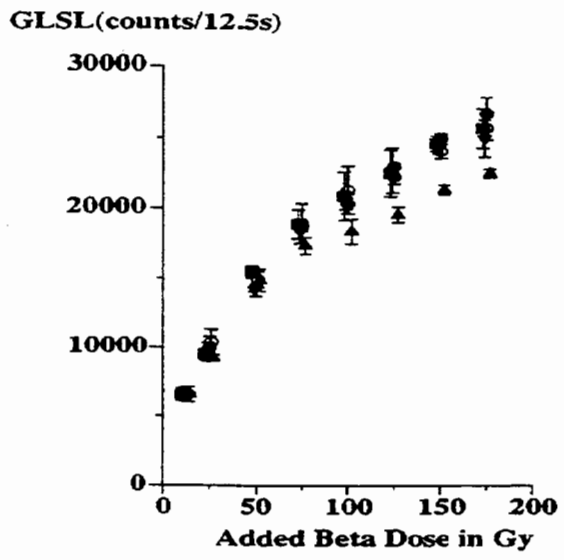

Figure 5
The dependence of the green light stimulated luminescence (GLSL) on beta dose for quartz previously bleached by heating to 500°C. The luminescence was measured for 12.5 s. Each point represents the mean of three measurements while the error bars indicate the spread in the three measurements. Five different normalization procedures are compared.
(1) samples were brought to the same total beta dose before bleaching in the SOL-2 and application of the normalizing dose, indicated by solid squares.
(2) samples were brought to the same total beta dose, preheated and read for normalization with correction in the single aliquot manner, indicated by solid circles.
(3) samples were bleached in the SOL-2 without bringing to the same total dose and the normalizing dose applied, indicated by upward pointing triangles.
(4) samples were brought to the same total dose then bleached by heating to 450°C before application of the normalizing dose, indicated by open circles.
(5) samples were bleached by heating to 450°C without being brought to the same total dose before normalization, indicated by downward pointing triangles.
Due to overlap not all symbols are inividually distinguishable.

Conclusions

Normalization involving sample bleaching by SOL-2 prior to application of the normalizing dose was successful provided all samples had been given the same total dose before bleaching.

Normalization involving bleaching the sample by heating prior to application of the normalizing dose was only successful for quartz which had been zeroed originally by heating.

A variant of the single aliquot method also provided a successful procedure for normalization within the dose range for which the luminescence response is approximately linearly related to dose.

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References

Aitken, M. J. (1985) Thermoluminescence Dating, Academic Press.

Duller, G. A. T. (1991) Equivalent dose determination using single aliquots. Nuclear Tracks and Radiation Measurements, 18, 371-378.

Duller, G. A. T. (1992) The use of a single aliquot method for intercalibration between radioactive sources. Ancient TL, 10, 8-11.

Duller, G. A. T. (1994) Luminescence dating of sediments using single aliquots: New procedures. Quaternary Geochronology (Quaternary Science Reviews), 13, 149-156.

Galloway, R. B. (1991) A versatile 40-sample system for TL and OSL investigations. Nuclear Tracks and Radiation Measurements 18, 265-271.

Galloway, R. B. (1992) Towards the use of green light emitting diodes for the optically stimulated luminescence dating of quartz and feldspar. Measurement Science and Technology 3, 330-335.

Galloway, R. B. (1993) Stimulation of luminescence using green light emitting diodes. Radiation Protection Dosimetry 47, 679-682.

Galloway, R. B. (1994) On the stimulation of luminescence with green light emitting diodes. Radiation Measurements 23, 547-550.

Godfrey-Smith, D. I. (1994) Thermal effects in the optically stimulated luminescence of quartz and mixed feldspars from sediments. Journal of Physics D: Applied Physics 27, 1737-1746.

Huntley, D. J., Godfrey-Smith, D. I. and Thewalt, M. L. W. (1985) Optical dating of sediments. Nature 313, 105-107.

Ollerhead, J., Huntley, D.J. and Berger, G.W. (1994) Luminescence dating of sediments from Buctouche Spit, New Brunswick. Canadian Journal of Earth Sciences 31, 523-531.

Stokes, S. (1992) Optical dating of young (modern) sediments using quartz: results from a selection of depositional environments. Quaternary Science Reviews 11, 153-159.

Wintle, A.G. (1993) Recent developments in optical dating of sediments. Radiation Protection Dosimetry 47, 627-635.

Reviewer
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