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Exploratory Single Grain OSL Analysis of Sediments from Capu di Locu, Corsica

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A.J. Cresswell, T.C. Kinnaird, D.C.W. Sanderson

East Kilbride Glasgow G75 0QF Telephone: 01355 223332 Fax: 01355 229898

Summary

Single grain optically stimulated luminescence (OSL) measurements have been conducted on retained material from sediment samples collected during excavations of Neolithic monuments at Capu di Locu, Belvédère, SW Corsica. One sample was taken from a presumed burial chamber at Tola and two from the lower fill of the construction pit of a menhir at Stantare. Analysis was conducted on 150-250 µm quartz grains previously separated for single aliquot regeneration (SAR) measurements, and on 250-500 µm grains prepared for this analysis. Single grain measurements, including analysis of blank discs prior to dispensing mineral grains, were conducting on a Risø DA-20 automatic reader, with 800-1000 grains measured for each of two samples, and approximately 350 grains for the third sample.

These exploratory measurements have demonstrated that a relatively large proportion (>20%) of mineral grains produce significant OSL counts for both the natural stored dose and following a 25 Gy artificial dose. Previous analysis of these samples using multi-grain aliquots measured using a single aliquot regeneration (SAR) protocol had shown broad dose distributions, with mixtures of aliquots of different ages. The single grain measurements have confirmed that these are mixed age deposits. Two samples show stored dose distributions around mean values which correspond to archaeological ages, for the third the stored dose distribution is significantly broader with no apparent archaeological age component. Further statistical analysis of these data would be required to attempt to resolve archaeological age material, that could potentially be used to further constrain construction dates for these monuments. It is expected that similar luminescence properties would be exhibited by mineral grains from other samples collected during the excavations of these sites, and it is possible that further single grain OSL analysis of other parts of the site may produce dose distributions with less mixing and more readily resolved components with archaeological ages.

Meanwhile SG apparent ages were obtained from the Tola sample which are consistent with previous estimates, and two new estimates have been achieved from the Stantare site which indicate potential for recovering archaeological ages in the 3rd to 4th millennium BC from the Menhir site.

Contents

List of Figures

List of Tables

1. Introduction

Samples of sediments were collected from excavations of two prehistoric monuments, a menhir (Stantare) and chambered tomb (Tola) on the Capu di Locu plateau above Belvédère, SW Corsica and subjected to optically stimulated luminescence (OSL) analysis and dating (Sanderson *et.al.* 2014). Initial OSL profiling of 92 samples from 10 sedimentary sequences, was conducted during fieldwork using portable OSL equipment, and used to select samples for laboratory characterisation. Calibrated luminescence measurements were conducted on 33 selected samples, and single aliquot regenerative (SAR) OSL dating procedures applied to 7 individual samples, using multigrain aliquots. For the Tola site, it was possible to identify samples underneath the area of the principal slab and its chockstones which gave individual age determinations during the Corsican neolithic period. The combined results from the youngest of these gave a composite OSL age of 2870 ± 190BC. If this is representative of undisturbed sediment it puts an early 3rd millennium BC date on the original monument. Other samples in the vicinity returned slightly earlier dates. The equivalent dose distributions associated with the Stantare menhir indicated complex depositional histories with both young and old material intermingled in the sediments within root disturbed stone sockets. It was concluded that the Menhir could not be dated using multi-grain OSL methods.

In this study the single grain (SG) OSL properties from material from the Tola and Menhir samples have been examined, with the aim of assessing whether SG sensitivities and yields are potentially able to help to resolve the mixed age units encountered. Single grain analysis in principle might help to identify different equivalent dose populations within such sediments, providing signal strengths from single grains are adequate. However the precision of OSL measurements from individual grains is signal limited, and there are potentially microdosimetric variations which could also generate mixed dose distributions at SG scale. The study reported here therefore is an exploratory investigation aimed at assessing whether single grain dose distributions contain separable components, and if so whether this approach can be used to enhance the OSL dating of these monuments.

Two samples were initially selected. One sample was from the Tola site where the multigrain analysis had suggested a relatively simple dose distribution as a control, and one sample from the lower fill of the Stantare site. Single grain OSL analysis was conducted using approximately 800-1000 grains per sample, to assess signal sensitivity distribution, and the dose distributions from the measureable subset of grains to evaluate the potential for resolving the mixed age deposits. A further sample from the lower fill of the menhir, on the opposite side of the monument, was also analysed using a smaller number of grains. The data sets have been used for SG age estimation.

2. Methods

2.1 Sample Selection and Preparation

The control sample from the Tola site selected was SUTL2690B, taken from profile 7 from the thin humic horizon between the substrate and the principal slab. The multi-grain aliquot SAR analysis of this sample had shown a broad equivalent dose distribution, with the majority clustering around the robust mean (27.0 ± 5.3 Gy) with two aliquots tailing to a lower stored dose estimate of approximately 9-10 Gy. The individual multigrain OSL age from this sample was 2610 ± 930 BC, and its composite age when combined with adjacent sample SUTL2690A was 2870 ± 190 BC.

The sample initially selected from the Stantare site was SUTL2683, equivalent to SUTL2678H, from the bottom of profile 5 in the NW pit of the monument. The multi-grain SAR analysis of this sample had shown a bi-modal equivalent dose distribution with two distinct clusters around large equivalent doses of approximately 42 Gy, assumed to represent geological residuals, and low equivalent doses of 1-5 Gy that would be associated with modern material. The sample was selected to confirm the presence of the modern component, and to assess whether an archaeological age component could be identified using SG approaches from a substantial number of grains.

For both samples, a small quantity of prepared grains from the multi-grain SAR analysis had been retained. These had been sieved to select the 150-250 µm fraction prior to acid treatment. The >250 µm fraction from both samples was also processed for this analysis, with the 250-500 µm fraction selected after an HF acid treatment. Both size fractions were used for the Tola sample, with the results compared, but only the larger grains used for the Stantare samples.

A further sample, SUTL2680L, from the bottom of profile 2, the lower fill of the southern construction pit, was also selected for a small number of measurements with the larger grain size.

2.2 Measurements

A set of Risø single grain discs, which have an array of 100 sample holes, each 300 µm diameter and 300 µm deep, was cleaned prior to the start of this work, using ultrasonic treatment in deionised water, and acetone. A "hot tapping" clean, where SG discs are heated upside down in an anvil, and then tapped with a hammer (Davies & Sanderson 2009), was employed between measurement runs, in combination with additional ultrasonic washing. Before and after each set of sample measurements the cleaned SG discs were also put through explicit blank tests. These blank measurements were carried through to data analysis with the sample runs.

Measurements were conducted on a Risø DA-20 automatic reader in a similar manner to an earlier study of sediments from the Ring of Brodgar in Orkney (Sanderson et al 2010). The measurement cycle consisted of three stages as shown in Table 2.1. First, the check on the cleaned disks following 5 Gy irradiation took place. Second grains were dispensed onto each disc. Finally the measurement cycle summarised in table 2.1 was followed, which records the natural OSL signal, and regenerated responses to 25 Gy dose, interspersed with 5 Gy test doses. After measurement each disc was examined by optical microscopy and the number of occupied holes estimated visually.

2.3 Data Analysis

Each 2s measurement consists of 60 data channels. The first 5 and the last five channels were dark count, without the laser activated. The net signal in each channel was calculated as the difference between the channels 6-30 and channels 31-55, the first 1s of each measurement less a background from the second 1s. For each sample hole, the net counts for the natural signal, the signals following the two test doses, the signal following the regeneration dose and the final zero measurement were tabulated. Data which satisfied significance requirements for the signals obtained from either the first (natural) or third (following the regeneration dose) OSL measurements were selected for further analysis. Some data were observed to show slow decay curves, with counts significantly above background after 2 s, and were rejected. These were mostly associated with very large natural signals, assumed to be of geological origin. Approximately 0.25-0.50% (1-4 holes from the 700 holes in the 7 discs sets used) of sample holes showed a background following the blank check runs. These measurements were also rejected from further analysis.

For those measurements with significant signals in response to 5 Gy test doses the ratio of the two test doses was determined to assess sensitivity change trends across the analytical cycle. The mean values of the test does ratios were consistent with no overall change in sensitivity. This broadly consistent with the multi-grain results, where sensitivity changes of less than 10% had been observed across complete SAR sequences (Sanderson et al 2014). With no evidence of sensitivity change, the data for the natural signal and following the regeneration dose were used without test dose normalisation to minimise noise broadening of the estimated doses. The equivalent dose on each sample hole was calculated assuming a relationship between OSL net counts (N) and stored dose (D) of the form:

$$N = a(1 - e^{-bD})$$

Hence

$$D = \frac{\ln(1 - N/a)}{-b}$$

The relationship between 5 Gy response and 25 Gy response was examined to determine the dose response curvature in the vicinity of the expected dose for archaeologically significant grains. It was subsequently decided to use the parameter b values obtained from the multi-grain aliquot SAR analysis (0.0124 ± 0.011 for SUTL 2683, 0.0188 ± 0.025 for SUTL2690B) to correct the SG data for dose response curvature, on the basis that the dose response function is insensitive to b value at low doses, and that the non-linear corrections are small close to 25 Gy dose levels from archaeological aliquots. For one sample this parameter was not available due to insufficient SAR measurements, and a value for nearby samples was used.

The parameter a was determined from the net counts (N_R) measured following the regeneration dose (D_R).

$$a = N_R/(1 - e^{-bD_R})$$

The regeneration dose D_R dose was nominally 25 Gy. A zoned dose rate calibration for the single grain discs was used to correct for the spatially variable in dose rate across the discs. Values of net counts in excess of a indicate that the sample has received a dose in excess of that needed to saturate the trapping centres, and the stored dose cannot be determined. These doses are therefore not reported, although the number of grains is recorded.

Stage	Operation	Details
Check cleaned disks	Dose	50s irradiation with ^{90}Sr/^{90}Y source
	Pre-heat	160°C for 10s
	Read-out	2 s OSL with green laser, at 125°C
Dispensing grains		
Measurement	Pre-heat	230°C for 10s
	Read-out	2 s OSL with green laser, at 125°C
	Test dose	54s irradiation with ^{90}Sr/^{90}Y source
	Pre-heat	230°C for 10s
	Read-out	2 s OSL with green laser, at 125°C
	Regeneration dose	271s irradiation with ^{90}Sr/^{90}Y source
	Pre-heat	230°C for 10s
	Read-out	2 s OSL with green laser, at 125°C
	Test dose	54s irradiation with ^{90}Sr/^{90}Y source
	Pre-heat	230°C for 10s
	Read-out	2 s OSL with green laser, at 125°C
	Pre-heat	230°C for 10s
	Read-out	2 s OSL with green laser, at 125°C

Table 2.1: Summary of measurement cycle

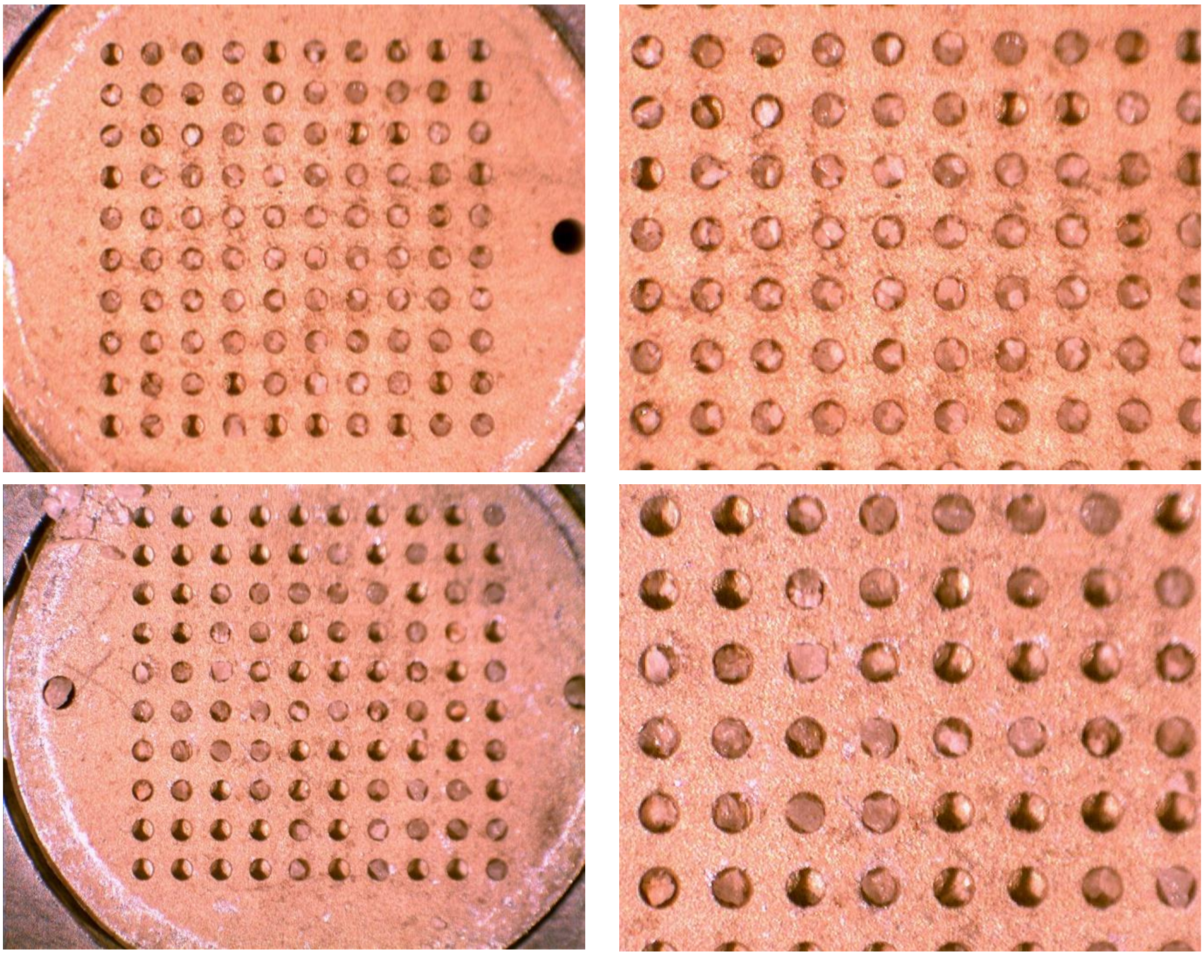

Figure 2.1: Examples of dispensed grains on disks, with 150-250 µm grains (top left) showing multiple hole occupancy (top right) and 250-500 µm grains (bottom left) with single grains in each hole (bottom right).

3. Results

Table 3.1 summarises the measurements conducted, indicating the numbers of occupied holes, based on visual microscopy, the number of holes from which significant signals were obtained on natural or regenerated cycles. Also tabulated are the number of holes selected after rejection of slow decays, an anomalous backgrounds. Further rejections were applied to discs whose natural signals were clearly at or above the saturation level of the dose response function, as tabulated here. The remaining numbers of accepted measurements from which dose estimates of varying precision could be obtained are also indicated.

Sample	Discs	Occupied holes	Significant holes	Selected holes	Saturated holes	Accepted measurements
SUTL2690B 150-250 µm	7	500	206	177	40	119
SUTL2690B 250-500 µm	7	350	129	149	27	88
SUTL2683 250-500 µm	18	900	776	379	70	270
SUTL2680L 250-500 µm	7	350	252	128	22	83

Table 3.1: Summary of measurements, the number of disks dispensed and the approximate number of sample holes occupied, the number of holes with significant signals (>3σ following the regeneration dose), the number of holes selected for further analysis, the number of these that had saturated signals, and the number of measurements where the uncertainty on the dose is less than 100%.

3.1 Tola Burial Chamber Sample

Figure 3.1 shows the relationship between the signals from the environmental dose and the nominal 25 Gy regeneration dose for the Tola sample, in both grain sizes.

Figure 3.2 shows a radial plot for the data from both grain sizes where the uncertainty on the calculated dose rate is less than 100%. Table 3.2 summarises the mean stored dose estimates for the two grain sizes, and the value determined previously by multi-grain SAR. It can be seen that there is no significant difference between the distributions for the two different grain sizes, and the distribution is consistent with that obtained using the multi-grain SAR method (Sanderson et.al. 2014).

Method	Number	Weighted Mean (Gy)	Robust Mean (Gy)
Multi-grain SAR	12		27.0 ± 5.3
150-250 µm single grain	119	24.7 ± 2.7	28.2 ± 2.2
250-500 µm single grain	88	25.2 ± 3.9	24.1 ± 3.2
All single grain	207	24.9 ± 2.2	26.4 ± 1.6

Table 3.2: Mean stored dose estimates for SUTL2690B (Tola)

In examining these dose distributions it is useful to consider the differences which grain size attenuation make to the dose rates. For the sample the dose rate evaluated for 150-250 µm grains is 5.6 ± 0.2 mGy a^{-1}. For 300 µm grains the corresponding value is 5.5 ± 0.2 mGy a^{-1}.

To first approximation a composite dose rate of 5.55 ± 0.20 mGy a^{-1} would apply to both grains sizes within uncertainties.

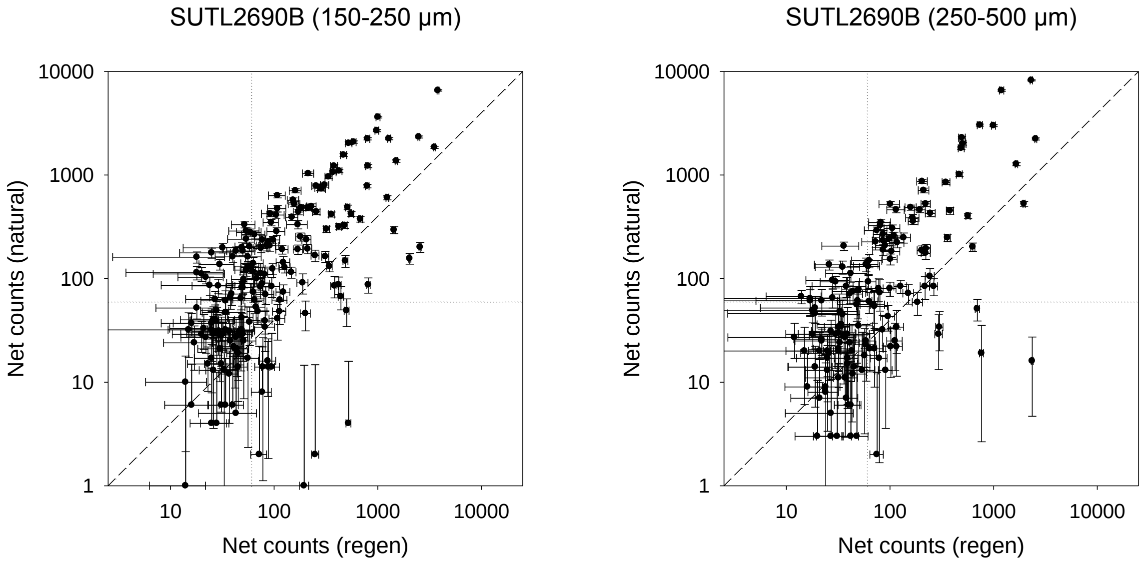

Figure 3.1: Relationship between net counts for the natural and regenerative dose measurements for the Tola sample. The dotted lines show the detection limits. The dashed line indicates the relationship expected for a 25 Gy natural dose, assuming a linear dose response curve.

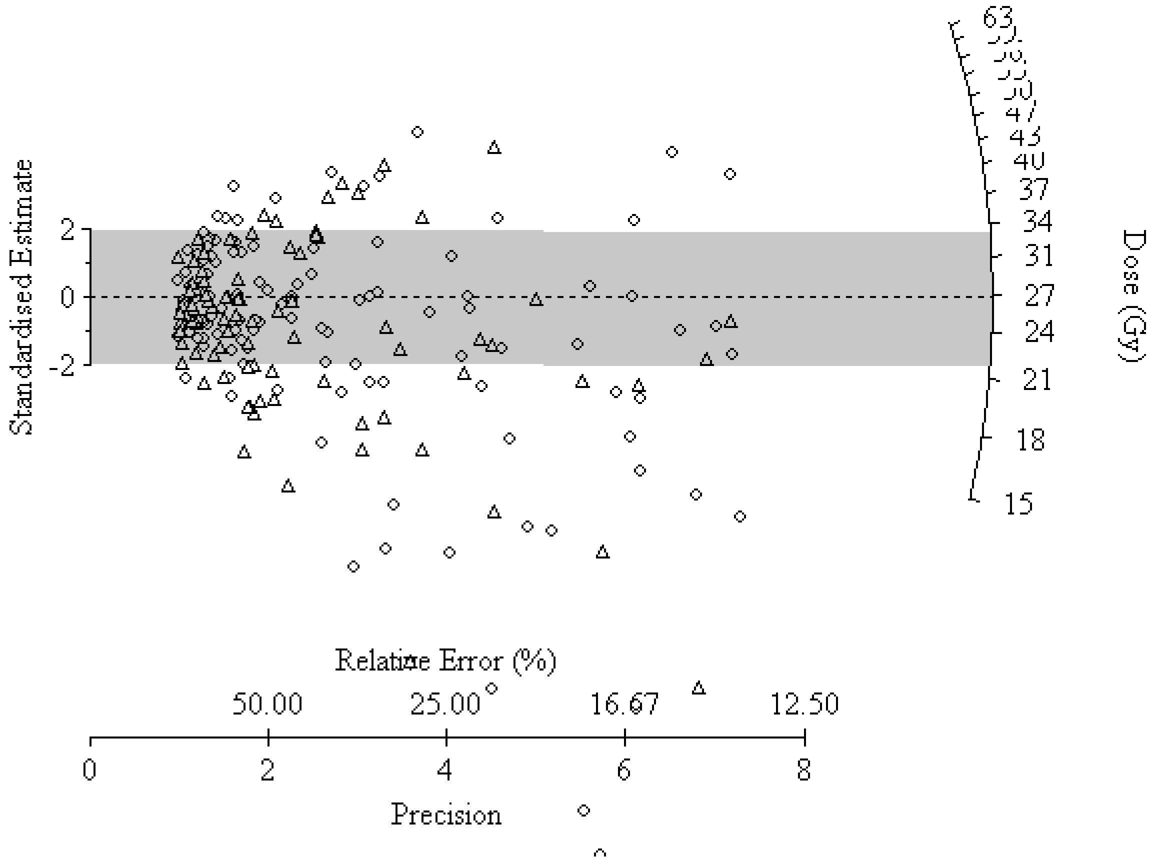

Figure 3.2: Radial plot for sample SUTL2690B (Tola) showing data for the 150-250 µm grains (circles) and 250-500 µm grains (triangles). The shaded area indicated the robust mean for the multi-grain SAR results (Sanderson *et.al.* 2014).

3.2 Stantare Menhir Samples

Figure 3.3 shows the relationship between the signals from the environmental dose and the nominal 25 Gy regeneration dose for the Stantare sample.

Figure 3.4 shows a radial plot for the data where the uncertainty on the calculated dose rate is less than 100%. Table 3.3 summarises the mean stored dose estimates, and the value determined previously by multi-grain SAR. For SUTL2683, the single grain data show a broad spread of stored doses, ranging from modern (1-5 Gy) to geological (>50 Gy), without any obvious clustering within that range that would correspond to an archaeological age. For SUTL2680L there is a much smaller spread in stored doses, with mean values that are consistent with an archaeological age.

Sample	Method	Number	Weighted Mean (Gy)	Robust Mean (Gy)
SUTL2683	Multi-grain SAR	24	42.3	
	250-500 µm single grain	270	29.4 ± 1.6	24.4 ± 1.6
SUTL2680L	250-500 µm single grain	83	18.8 ± 2.5	18.6 ± 2.0

Table 3.3: Mean stored dose estimates for SUTL2683 (Stantare, northwest construction pit) and SUTL2680L (Stantare, southern construction pit)

Taking account of grain size the estimated dose rates for these two samples are 3.85 ± 0.15 mGy a^{-1} (SUTL2683) and 4.08 ± 0.16 mGy a^{-1} (SUTL2680L).

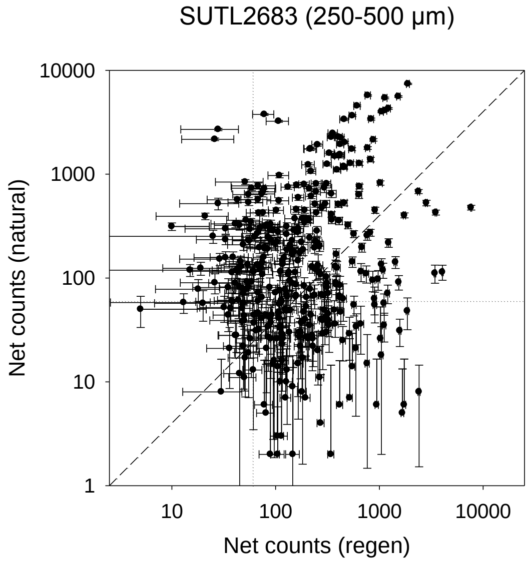

Figure 3.3: Relationship between net counts for the natural and regenerative dose measurements for the Stantare sample. The dotted lines show the detection limits. The dashed line indicates the relationship expected for a 25 Gy natural dose, assuming a linear dose response curve.

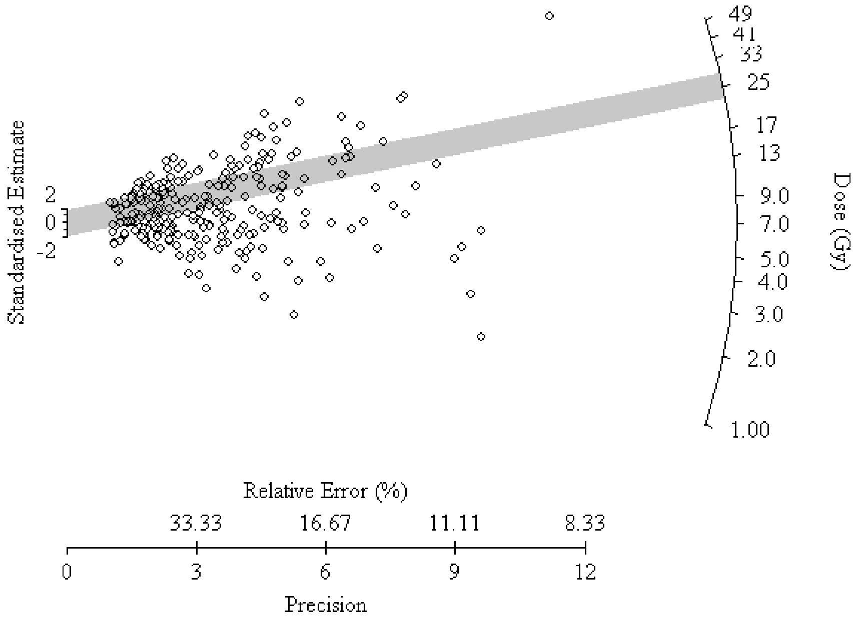

Figure 3.4: Radial plot for sample SUTL2683 (Stantare, northwest construction pit). The shaded area indicates the robust mean.

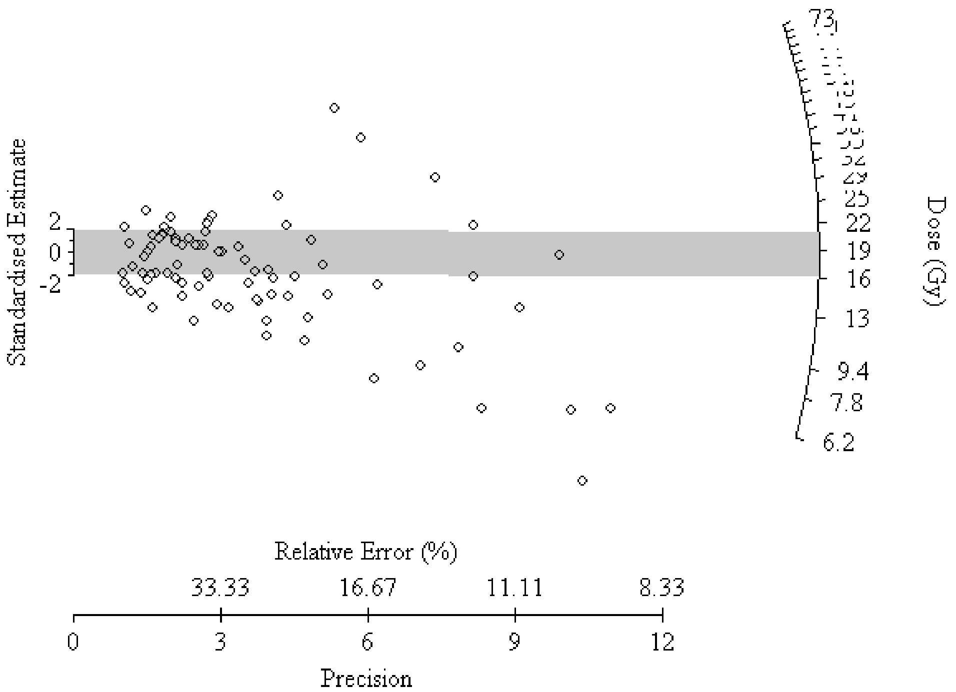

Figure 3.5: Radial plot for sample SUTL2680L (Stantare, southern construction pit), the shaded area indicates the weighted mean.

3.3 Apparent ages

Taking the mean effective dose rates as discussed above, and the dose estimates based on robust statistics, which as shown above are similar to the weighted values the apparent ages corresponding to the single grain data sets are given below in table 3.4.

For sample 2690B, from the Tola the outcome is within the analytical error of the multigrain result, and reinforces the early 3rd millennium BC age estimate from the multigrain analysis. The uncertainty of the SG results is considerable at 500 years, which reflects the combination of the uncertainties in individual SG dose estimation from these signal-limited individual grains and the overall spread in dose distribution.

For the Stantare samples the NW section the use of robust statistics has apparently suppressed the anomalous modern components, which are attributed to root disturbance. However the remaining data sets fall into the late 5th millennium BC or early 4th millennium BC, and may potentially still contain residual age material. The analysis from the SE quadrant in sample 2680L has produced results which also come from the 3rd millennium BC, and are of similar or marginally younger age than that of the chambered tomb.

Sample	Stored Dose (Gy)	Dose Rate (mGy a^{-1})	Age BP (ka)	Date
SUTL2690B	26.4 ± 1.6	5.55 ± 0.22	4.75 ± 0.5	2735 ± 500 BC
SUTL2683	24.4 ± 1.6	3.85 ± 0.11	6.34 ± 0.7	4325 ± 700 BC
SUTL2680L	18.7 ± 2.5	4.08 ± 0.16	4.58 ± 0.6	2565 ± 600 BC

Table 3.4 Apparent ages for single grain analyses based on robust statistics

4. Discussion and Conclusions

Exploratory single grain OSL analysis has been conducted on retained material from samples collected for SAR analysis of multi-grain aliquots from two Neolithic monuments at Capu di Locu, SW Corsica.

A sample was used from the Tola site, where the SAR data suggested a relatively simple dose distribution, as a control on the method. Approximately 850 holes on the single grain discs were populated with two different grain sizes (150-250 µm grains, from material prepared for the SAR analysis, and 250-500 µm grains prepared for this work). There is no significant difference in the results from the two grain sizes. Approximately 40% of the sample holes produced a significant signal following a 25 Gy regeneration dose, with 207 (25%) producing a calculated stored dose used to produce the dose distribution. The sample produces a broad dose distribution within the same range of doses determined from the previous SAR analysis, with weighted and robust means consistent with the SAR value.

A second sample was selected from the Stantare site where the earlier SAR analysis indicated a bi-modal distribution of modern (1-5 Gy) and geological components. Approximately 900 holes on the single grain discs were populated with 250-500 µm grains. Over 80% of these sample holes produced a significant signal following a 25 Gy regeneration dose, with 270 (30%) producing a calculated stored dose used to produce the dose distribution. The sample produces a broad dose distribution, with modern (1-5 Gy), geological (>50 Gy) and intermediate doses without any obvious clustering. A smaller number of grains (approximately 350) were analysed from a second sample from Stantare where only two SAR analyses had been conducted, from the southern construction pit of the monument. This also shows a broad dose distribution, but with less apparent mixing compared to the first sample from Stantare, with mean stored doses consistent with an archaeological age. It is noted that the small number of SAR analyses means that the dose response of this sample is unknown, and it has been assumed to be similar to the other Stantare sample.

It is noted that both samples result in a broad distribution, with a mixture of grains with stored doses consistent with modern, geological and archaeological ages. It is noted that for both samples the mean stored doses are consistent with archaeological ages. For the Tola sample, with a dose rate of 5.55 ± 0.22 mGy a^{-1}, this would result in an age of 4750 ± 500 years BP (2735 ± 500 BC) which is consistent with the value determined by SAR analysis.

For the first Stantare sample, with a dose rate of 3.85 ± 0.11 mGy a^{-1}, this would result in an age of 6340 ± 700 years BP (4325 ± 700 BC), significantly younger than the age estimated from the weighted mean of the earlier SAR data (7100 ± 320 BC), but potentially still containing residual material, given the dose distribution. For the second Stantare sample, the mean dose rate from laboratory gamma spectrometry of other samples in the profile is 4.08 ± 0.16 mGy a^{-1}, comparable to the dose rate for the other Stantare sample, and the mean stored dose would result in an age of 4580 ± 600 years BP (2565 ± 600 BC).

This exploratory study has demonstrated the sensitivities and potential for single grain OSL methods with this material. While a reasonable large proportion of the grains (>20%) produce significant OSL counts from both the natural dose and a 25 Gy regeneration dose, the sensitivity and precision of individual dose estimates is limited. Nonetheless it has been possible to develop single grain dose distributions, which are broadly consistent with the previous SAR analysis from multigrain samples. For both samples studied there are broad dose distributions implying certain amount of mixed age content. It has nonetheless been possible to obtain broadly consistent age estimates from the Tola to those achieved with the original analysis. At the Stantare site the SG data produce improved outcomes, albeit with different working estimates from each side of the stone socket. This has shown signs of archaeological age estimates within the overall precision of the SG data sets.

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