Optically stimulated luminescence dating of heat retainer hearths from the Sahara: Insights into signal accumulation and measurement.

S.J. Armitage[1,2*], A. Krishna[1], L.E. Parker[1,3], G.E. King[4]

[1]Centre for Quaternary Research, Department of Geography, Royal Holloway, University of London, Egham, Surrey, TW20 0EX, United Kingdom.

[2]SFF Centre for Early Sapiens Behaviour (SapienCE), University of Bergen, Post Box 7805, 5020, Bergen, Norway.

[3]Archaeological Research Services Ltd, Angel House, Portland Square, Bakewell, Derbyshire, DE45 1HB, United Kingdom.

[4]Universität Bern, Institut für Geologie, Baltzerstrase 1+3, 3012 Bern, Switzerland.

Abstract

Heat retainer hearths are a prominent component of the Holocene archaeological record of a number of drylands. Rocks within these hearths were fired in antiquity, emptying the optically stimulated luminescence (OSL) source traps of mineral grains within the rock. Since partial bleaching and mixing of grains within a lithified heat retainer is impossible, these rocks offer the opportunity to test our understanding of OSL signal accumulation and measurement processes. First, we show that OSL ages calculated using grains size fractions from 4-11 µm up to 180-210 µm are indistinguishable for a single heat retainer, indicating that the environmental and instrumental dose rate correction factors routinely used in luminescence dating are accurate. Second, we used single-grain dose recovery and equivalent dose measurements to determine the overdispersion due to beta microdosimetry. For the heat retainers measured in this study, overdiseprsion due to beta microdosimetry ranges from 8.9 ± 1.8 to 20.3 ± 1.6 %. Third, we investigate the impact of mechanical crushing on the measured equivalent dose from quartz, to test the potential of using this technique to liberate dateable material from heat retainers which are not acid soluble. A small (<1 Gy) but significant increase in equivalent dose is observed in crushed zero-age samples, but the equivalent doses of crushed and uncrushed Holocene quartzes are indistinguishable. We conclude that crushing is a viable method for extracting dateable material from a heat retainer, but that some knowledge of the dosimeter's mean grain size is required for calculation of an accurate environmental dose rate.

* e-mail: simon.armitage@rhul.ac.uk

Keywords: optically stimulated luminescence; hearths; dose rate; microdosimetry; crushing.

1. Introduction

Heat retainer hearths are a prominent component of the Holocene archaeological record, and are particularly prevalent in the Sahara and Arabian Peninsula, where they are frequently associated with surface scatters which attest to the presence of ancient nomadic pastoralists. Rocks, which acted as heat retainers while the hearth was in use, are heated to sufficiently high temperatures to completely empty the OSL source traps. Consequently, mineral grains from within these heat retainer rocks are dateable using optically stimulated luminescence (OSL) techniques, providing an estimate of the time elapsed since last firing (Armitage and King, 2013; Rhodes et al., 2010; Rhodes et al., 2009). These fired heat retainers are often the only dateable and unequivocally anthropogenic component of the surface archaeological record. OSL dating of hearths is therefore potentially an important tool for understanding the nature of the surface archaeological record in drylands. Furthermore, fired hearth stones offer a number of advantages when studying the accumulation of the OSL signal. First, since the heat retainers are fired in a lithified state, and since post-firing mixing of mineral grains within a heat retainer is impossible, OSL dating of multiple grain-size fractions from a single rock allows the accuracy of grain-size specific corrections to environmental and instrumental dose rates to be tested. Second, since post-firing mixing of mineral grains is impossible, and since Armitage and King (2013, their Figure 5) have demonstrated that sunlight does not deplete the OSL signal in the inner portion of hearth rocks, sand sized quartz extracted from hearth rocks may be used to investigate single-grain equivalent dose (D_e) overdispersion due to beta microdosimetry. In the present study we: 1) Conduct paired single-grain dose recovery and D_e measurement experiments to investigate the degree of overdispersion caused by beta microdosimetry; 2) Calculate ages for a range of grain-size fractions extracted from a single heat retainer to test the internal consistency of the OSL method, and 3) Investigate the possibility of extending the OSL dating technique to non-carbonate lithologies where mineral grains cannot be extracted from a heat retainer by acid dissolution.

2 Materials and methods

A total of thirteen heat retainers were collected from five hearths at the Al Wafa archaeological site, in the Libyan Sahara. A full site description is found in Armitage and King (2013). Briefly, Al Wafa is a gently sloping basin characterised by shallow, ephemeral drainage channels cut into an extensively deflated surface composed of unconsolidated sand protected by gravel-pebble sized carbonate clasts. The landscape is dominated by ~100 discrete hearths, which are usually seen as mounds rising a few tens of centimetres above the surrounding land surface (Figure 1), and date from ~7-9 ka (Armitage and King, 2013). These mounds are believed to result from topographic inversion,

with the hearths originally consisting of cobble lined pits dug into soil, which was subsequently deflated when the Sahara dried ~5 ka (Armitage et al., 2015). The cobbles, which served as heat retainers when the hearth was in use, are clasts of sandy limestone, which outcrops at the margin of the Al Wafa basin. Approximately cuboid heat retainers with no visible fracturing were sampled from discrete hearths.

The outer light exposed portions of the heat retainers was removed by repeated immersion in 1.16 M HCl, until the length of each axis of the clast had been reduced by a minimum of 25 mm. The dating sample was obtained by dissolution of the remaining material. Quartz separates were produced using the methods summarised in Supplemental Material Section A. For samples FZ39-48, only the 210-180 µm fraction was prepared for measurement. For samples FZ84, 85, 95 and 96, dating samples were prepared from the 4-11, 11-20, 20-40, 40-60, 60-90, 90-125, 125-150, 150-180 and 180-210 µm fractions. All size fractions were mounted on 10 mm aluminium discs. Additional single-grain analysis (212-180 µm grains) was performed using standard Risø single-grain sample holders.

All OSL measurements presented in this study were carried out using a Risø TL/OSL-DA-15 dating system (Bøtter-Jensen et al., 2003), fitted with a single-grain OSL attachment (Duller et al., 1999). Optical stimulation of single aliquots was carried out using a blue (470 ± 30 nm) light emitting diode (LED) array with a power density of 33 mW/cm^2, while single-grains were stimulated using a 10 mW Nd : YVO_4 solid-state diode-pumped green laser (532 nm) focussed to yield a nominal power density of 50 W/cm^2. Infra-red (IR) stimulation was carried out using an IR (870 nm) laser diode array. OSL was measured using an Electron Tubes Ltd 9235QB15 photomultiplier tube with 7.5 mm of Hoya U-340 filter interposed between the sample and photomultiplier. Irradiation was carried out using a 40 mCi ^{90}Sr/^{90}Y beta source, calibrated relative to the National Physical Laboratory, Teddington ^{60}Co γ-source (Hotspot 800) (Armitage and Bailey, 2005b). Due to the spatial heterogeneity of beta emitters across the active face of the ^{90}Sr/^{90}Y beta source, it was necessary to apply a grain position correction to single-grain D_e values, using the method of (Armitage et al., 2011).

Equivalent doses were determined using the single-aliquot regenerative-dose (SAR) method (Murray and Wintle, 2000). Following Armitage and King (2013), who worked on heat retainers from the Al Wafa site, we adopted a preheating regime of 260 °C, 10 s for PH1 (the pre-heat prior to measurement of natural or regenerated luminescence) and 220 °C, 10 s for PH2 (the pre-heat prior to measurement of the test dose luminescence response) for all measurements. OSL measurements were made at 125 °C, using blue diodes (40 s) for aliquot measurements or a green laser (2 s per grain) for single-grain measurements. For blue diode stimulation, the OSL signal was that recorded during the first 0.36 s of

stimulation, and a background signal from the last 4 s of stimulation was subtracted. For green laser stimulation, the OSL signal was that recorded during the first 0.3 s of stimulation, and a background signal from the last 0.3 s of stimulation was subtracted (Thomsen et al., 2005). Curve fitting, and D_e determination, were performed using version 3.24 of the Luminescence Analyst software (Duller, 2007). We adopted the single-grain rejection criteria of Armitage et al. (2011), Supplementary Material Section B.

3 Age estimates generated using different grain-size fractions

In principle, any grain-size fraction from fine silt to fine sand can be used to measure the equivalent dose of a sample. For any given sample, the grain size used may be dictated by availability or the post-depositional processes which a sample has experienced e.g. silt may be translocated by percolating water in a sandy sediment, but be the only clast size available in a loess deposit. Consequently, sedimentary characteristics and geomorphic setting often necessitates the measurement of different grain size fractions for samples within the same dating study. However, numerous factors determining the environmental or laboratory dose rate experienced by the luminescence dosimeter vary with grain size. These variations may be progressive, e.g. the ~13% increase in beta dose rate from a Risø irradiation unit as grain size increases from 4-11 µm to >63 µm (Armitage and Bailey, 2005b) or abrupt e.g. at the transition between sand-sized material, which are routinely etched to remove the alpha dosed rind, and silt-sized material. If a single dating project uses more than one grain size fraction, then the minimum requirement for a successful study is that different grain sizes yield the same luminescence age for sediments which were deposited simultaneously. In practice, the types of sedimentary deposit which are normally targeted for OSL dating tend to be well sorted. Conversely, the heat retainers from Al Wafa contain quartz grains from <4 to >210 µm in diameter, all of which must have a common OSL age, making these samples ideal for testing the internal consistency of OSL ages produced using different grain size fractions.

Twelve aliquots of quartz from each size fraction from each of four samples were measured using a single luminescence reader. Across the range of grain sizes measured, progressive variations in environmental alpha (Brennan et al., 1991) and beta (Guérin and Mercier, 2012) attenuation factors, and laboratory beta dose rate (Armitage and Bailey, 2005b) occurred. An abrupt alteration in dose rate occurred between the 60-90 and 90-125 µm fractions due to HF etching of >90 µm grains, resulting in the loss of 100% of the environmental alpha dose (making the common assumption of isotropic etching), and 1% of the environmental beta dose (Brennan, 2003). An abrupt change in laboratory dose rate is also present between 20-40 and 40-60 µm, since the <40 µm grains were deposited from suspension, coating the entire face of the disc, whereas the >40 µm grains were

attached using Silkospray applied via a 5 mm diameter mask. However, since the instrument was calibrated using samples mounted by the same methods, no correction for the known variation in instrumental dose rate between the core and periphery of the disc was made (Ballarini et al., 2006).

Equivalent dose, dose rate and calculated age are presented for nine different grain size fractions in the range 4-210 μm (Tables S2-S4). For sample FZ84 (Figure 2), dose rate falls steadily as grain size increases, from a value of 0.98 ± 0.07 Gy/ka (4-11 μm) to 0.83 ± 0.05 Gy/ka (180-212 μm), a decrease of ~18 %. For all four of the samples for which data are available, equivalent dose also falls with increasing grain size, though for each sample there are outliers from this general trend (e.g. 11-20 μm in Figure 2). Nonetheless, ages calculated for each grain size fraction from a single sample are consistent with each other. This result indicates that for these samples at least, the dose rate correction factors used in this study produce consistent ages across the full range of grain sizes commonly adopted for luminescence dating work.

4 Single-grain overdispersion due to beta microdosimetry

Overdispersion (OD), the relative spread of equivalent doses after measurement uncertainties are excluded, is a key input for several of the statistical models used in single-grain dating e.g. the Minimum Age Model (MAM) (Galbraith et al., 1999) and the Finite Mixture Model (FMM) . However, successful application of these models requires an estimate of the OD of a well-bleached sample, which is otherwise identical to the sample being measured. Incorrect estimation of the well-bleached OD may lead to the adoption of the incorrect statistical model, and the production of an incorrect age. For example, an erroneously young age would be calculated if an underestimate of the well-bleached OD caused the MAM to be applied to a sample that was not partially bleached. The heterogeneous distribution of beta activity within a sediment, e.g. where occasional potassium feldspars are present within a quartz rich sand (Mayya et al., 2006), has the potential to cause such an underestimation of the well-bleached OD. Although some success has been achieved in modelling OD due to beta microdosimetry (Cunningham et al., 2012; Guérin et al., 2015; Martin et al., 2015), few studies have measured OD in ancient sediments where other causes may be excluded or isolated. Heat retainers which were fired in antiquity offer the opportunity to isolate OD due to beta microdosimetry, since lithified samples cannot experience post-depositional mixing, and firing completely empties the OSL source traps (Armitage and King, 2013, section 8), eliminating the possibility of heterogeneous bleaching. Consequently, OD observed in a D_e dataset (OD_{De}) from a heat retainer consists of contributions from the measurement process (e.g. instrumental variability and intra-sample variation in luminescence characteristics) and from beta microdosimetry. Since the OD observed in a dose recovery dataset (OD_{Rec}) is caused by the measurement process alone, it is

possible to isolate OD due to beta microdosimetry (OD_{β}) by subtracting quadratically OD_{Rec} from OD_{De}.

Dose recovery measurements were made on six samples from Al Wafa. Following Armitage and King (2013), single-grain dose recovery experiments were performed by heating samples to 350 °C for 120 s (to empty the OSL source traps) after which a beta dose similar to D_e (the "known dose") was applied. The "recovered dose" was then measured, using an identical measurement sequence to that used for measuring the equivalent dose. Single-grain equivalent dose, dose recovery and OD values are presented in Table S6. The "dose recovery ratios" (recovered dose/known dose) ranged from 0.96 ± 0.01 to 1.00 ± 0.01, with a mean value of 0.99 ± 0.01, indicating that the measurement parameters used in this study are appropriate for the Al Wafa samples. OD_{rec} values ranged from 6.3 ± 0.6 % to 9.7 ± 1.1 %, with a mean value of 7.6 ± 1.2 %. OD_{β} values ranged from 15.8 ± 1.8 to 27.1 ± 1.6 %, with a mean value of 22.6 ± 4.1, while OD_{De} values ranged from 18.5 ± 1.5 % to 28.2 ± 1.5 %, with a mean value of 23.9 ± 3.6 %.

The wide range of OD_{β} values from heat retainers from a single location, and derived from a single geological unit, suggests that the effects of beta microdosimetry is highly sample dependant. This implies that, even for multiple samples from a single location, the well-bleached OD required for the application of several statistical models may need to be determined for each individual sample. In addition, only one of the six samples measured yielded an OD_{De} value below 20%. Several studies (e.g. Armitage et al., 2011) have used an OD_{De} value of 20% to distinguish between well-bleached, undisturbed samples (OD_{De}<20%) and samples which have been subject to partial bleaching or post depositional mixing (OD_{De}>20%). Our results contribute to a growing body of literature (Guérin et al., 2015; Thomsen et al., 2016) which does not support the use of a 20% OD_{De} threshold. Instead a site-specific, or possibly even a sample specific, OD_{De} estimate for a well-bleached sample is required. In the absence of such an estimate, partial bleaching or post depositional mixing should be diagnosed based upon a knowledge of the sample's depositional context and burial history. This approach risks confirmation bias (the tendency to observe effects which are expected to be present), and highlights the need for a robust method for estimating the OD expected from a well-bleached sample.

5 Extending the OSL dating technique to non-carbonate lithologies

Where heat retainers are composed of carbonate rock, extraction of dateable material is easily achieved using progressive HCl dissolution to remove the outer light-exposed portions of the rock (e.g. Armitage and King, 2013). Conversely non-carbonate heat retainers have been subject to more

complicated preparation procedures. For example, Rhodes et al. (2010) removed the light exposed portion of the rock using a geological hammer, followed by two cycles of mechanical crushing (one producing pebble sized fragments, and the other yielding sand sized fragments) and a short HF etch to remove surface crushing effects and feldspar. This latter approach has the disadvantage of being time-consuming and, depending upon the mineralogical composition of the parent rock, potentially yielding a polymineral sample with unpredictable luminescence characteristics. An alternative approach is to remove the outer portions of the rock with a geological hammer, and crush the remaining dating fraction in a mill, preparing the resulting powder using the standard method for 4-11 μm silt. The advantages of this approach is that it is less labour intensive, is likely to yield a more predictably pure quartz separate, and presents the sample to the luminescence reader in a predictable geometry. As with the method of Rhodes et al, (2010), this method requires a realistic approximation of the mean grain size of the dosimeter before crushing to calculate an accurate dose rate. More significantly, a crushing induced increase in signal has been observed for burnt flint samples (Aitken, 1985), section 7.1), while recent evidence suggests that crushing or shearing may either reduce or reset the natural luminescence signal (Bateman et al., 2012; Swift et al., 2011).

To test the possibility that milling induces a luminescence signal, a selection of seven zero age quartz separates were measured in their crushed and uncrushed states. Similarly, to test the possibility that milling reduces the luminescence signal, a selection of five quartz separates from Holocene dune samples (D_e ranging from 2-20 Gy) were measured in their crushed and uncrushed states. Finally, to test the utility of crushing as a method for extracting dateable quartz from non-carbonate heat retainers, small chips from the core of the four heat retainers reported in Section 3 were crushed, and the De compared to that for material extracted via sequential dissolution in HCl. Crushed samples were prepared by loading ~2 g of material into a light-tight stainless steel grinding jar, and grinding at 20 Hz for 120 s using a Retsch Mixer Mill MM400. The 4-11 μm fraction was subsequently extracted from the milled material using the method described in Supplemental Material Section A. Results are presented in Supplementary Material Tables 3-5.

Of the seven zero age samples measured (Table S7), four showed a significant increase in equivalent dose upon crushing, while three yielded indistinguishable values. The mean increase in equivalent dose due to crushing was 0.25 ± 0.14 Gy, ranging from 0.08 ± 0.05 Gy for a Libyan desert dune sand (Armitage et al., 2007) to 0.93 ± 0.33 Gy for a South African coastal dune sand (this study). From these data it is apparent that crushing can induce a significant increase in D_e, but that the effect is neither universal nor uniform. None of the five Holocene samples yielded crushed/uncrushed ratios distinguishable from unity (Table S8), suggesting that any signal increase due to crushing is either

negligible or masked by other sources of D_e scatter. Crushed material from the cores of the four heat retainers yielded equivalent doses consistent with the range of values obtained from uncrushed material (Tables S2-S5). However, to determine an age, the mean grain size of the uncrushed quartz must be known to determine the applicable dose rate. Without this information i.e. assuming mean grain size of the dosimeter to be somewhere between 4 and 210 μm, a large uncertainty term needs to be used to account for the ~15 % variation in environmental dose rate between these grain sizes. In the case of the four heat retainers measured here, the effective grain size of the quartz prior to crushing may be determined by calculating the dose rate required for the equivalent dose of the crushed material to yield the mean age determined using the various grain size fractions presented in Section 3. This analysis indicates that heat retainers contained grains with a mean effective size of 40-60 μm (n=2), 90-150 μm (n=1) and >210 μm (n=1). For the samples in question, environmental dose rates decrease by 7.7 ± 0.8 % from 40-60 to 180-210 μm. Consequently, estimation of the mean grain size either by visual inspection or petrography may yield acceptably precise estimates of the environmental dose rate. These data suggest that crushing is a viable technique for extracting dateable mineral separates from heat retainers with non-carbonate lithologies.

6 Conclusions

Heat retainers from ancient hearths offer the opportunity to test our understanding of OSL signal accumulation and measurement processes. We measure equivalent doses from a range of grain sizes commonly adopted in luminescence dating studies. For each hearth stone, different size fractions yield similar ages, indicating that for our samples at least, the instrumental and environmental dose rate correction factors used in luminescence dating yield internally consistent (accurate?) results. In addition, since mineral grains within heat retainers cannot be subject to partial bleaching or post firing mixing, overdispersion in single-grain data from these samples is due to measurement processes and beta microdosimetry. The latter may be isolated by subtracting quadratically the single-grain overdispersion obtained from a dose recovery experiment on the same sample. This approach yielded values for overdispersion resulting from beta microdosimetry ranging from 15.8 ± 1.8 to 27.1 ± 1.6 %. Our results suggest that the application of a standard overdispersion threshold (e.g. 20%), above which processes such as mixing or partial bleaching may be diagnosed, may not yield accurate results. Lastly, the possibility of extracting dateable material from heat retainers via mechanical crushing was explored. A small but significant increase in equivalent dose was observed for a number of quartz separates from zero-age samples after crushing. However, it was not possible to distinguish the equivalent dose from crushed and uncrushed fractions of five Holocene samples. These results suggest that crushing is a viable method for extracting dateable material from a rock, but we note that

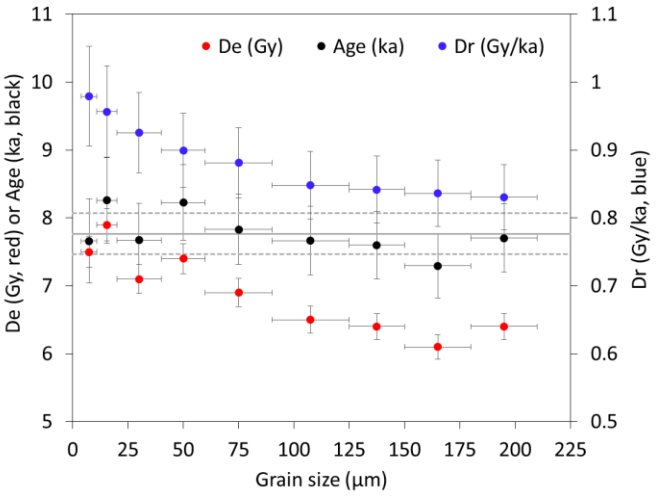
some knowledge of the dosimeter's mean grain size is required for calculation of an accurate environmental dose rate.

7 Acknowledgements

SJA's contribution to this work was partly supported by the Research Council of Norway, through its Centres of Excellence funding scheme, SFF Centre for Early Sapiens Behaviour (SapienCE), project number 262618. GEK acknowledges support from Swiss National Science Foundation grant number PZ00P2-167960.

Figures

Figure 1: A small hearth at Al Wafa. The hearth mound is ~1 m in diameter.



290

291 Figure 2: Equivalent dose (D_e), environmental dose rate (D_r) and calculated age for different grain
292 size fractions of quartz from heat retainer sample FZ84. The thick grey line represents the mean
293 age, calculated using data from all grain sizes, while the dashed lines represent ± 1 standard
294 deviation.

295

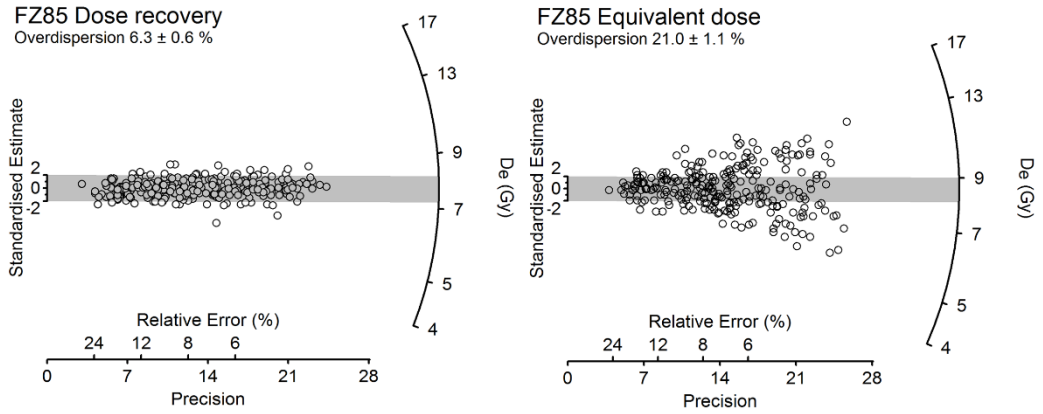


Figure 3: Radial plots of equivalent doses for individual quartz grains from heat retainer sample FZ85. The grey bars are centred at the equivalent dose calculated using the central age model (Galbraith et al., 1999). The left radial plot represents data from a dose recovery experiment, whereas the right panel shows the natural equivalent dose distribution.

References

Aitken, M.J., 1985. Thermoluminescence dating. Academic press.

Armitage, S.J., Bailey, R.M., 2005. The measured dependence of laboratory beta dose rates on sample grain size. Radiat Meas 39, 123-127.

Armitage, S.J., Bristow, C.S., Drake, N.A., 2015. West African monsoon dynamics inferred from abrupt fluctuations of Lake Mega-Chad. P Natl Acad Sci USA 112, 8543-8548.

Armitage, S.J., Drake, N.A., Stokes, S., El-Hawat, A., Salem, M., White, K., Turner, P., McLaren, S.J., 2007. Multiple phases of north African humidity recorded in lacustrine sediments from the fazzan basin, Libyan sahara. Quat Geochronol 2, 181-186.

Armitage, S.J., Jasim, S.A., Marks, A.E., Parker, A.G., Usik, V.I., Uerpmann, H.-P., 2011. The Southern Route "Out of Africa": Evidence for an Early Expansion of Modern Humans into Arabia. Science 331, 453-456.

Armitage, S.J., King, G.E., 2013. Optically stimulated luminescence dating of hearths from the Fazzan Basin, Libya: A tool for determining the timing and pattern of Holocene occupation of the Sahara. Quat Geochronol 15, 88-97.

Ballarini, M., Wintle, A.G., Wallinga, J., 2006. Spatial variation of dose rate from beta sources as measured using single grains. Ancient TL 24, 1-7.

Bateman, M.D., Swift, D.A., Piotrowski, J.A., Sanderson, D.C.W., 2012. Investigating the effects of glacial shearing of sediment on luminescence. Quat Geochronol 10, 230-236.

Bøtter-Jensen, L., Andersen, C.E., Duller, G.A.T., Murray, A.S., 2003. Developments in radiation, stimulation and observation facilities in luminescence measurements. Radiat Meas 37, 535-541.

Brennan, B.J., 2003. Beta doses to spherical grains. Radiat Meas 37, 299-303.

Brennan, B.J., Lyons, R.G., Phillips, S.W., 1991. Attenuation of alpha particle track dose for spherical grains. International Journal of Radiation Applications and Instrumentation. Part 18, 249-253.

Cunningham, A.C., Devries, D.J., Schaart, D.R., 2012. Experimental and computational simulation of beta-dose heterogeneity in sediment. Radiat Meas 47, 1060-1067.

Duller, G., 2007. Assessing the error on equivalent dose estimates derived from single aliquot regenerative dose measurements. Ancient TL 25, 15-24.

Duller, G.A.T., Bøtter-Jensen, L., Murray, A.S., Truscott, A.J., 1999. Single grain laser luminescence (SGLL) measurements using a novel automated reader. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms 155, 506-514.

Galbraith, R.F., Roberts, R.G., Laslett, G.M., Yoshida, H., Olley, J.M., 1999. Optical dating of single and multiple grains of quartz from Jinmium rock shelter, northern Australia: Part I, experimental design and statistical models. Archaeometry 41, 339-364.

Guérin, G., Jain, M., J. Thomsen, K., S. Murray, A., Mercier, N., 2015. Modelling dose rate to single grains of quartz in well-sorted sand samples: The dispersion arising from the presence of potassium feldspars and implications for single grain OSL dating. Quat Geochronol 27, 52-65.

Guérin, G., Mercier, N., 2012. Preliminary insight into dose deposition processes in sedimentary media on a scale of single grains: Monte Carlo modelling of the effect of water on the gamma dose rate. Radiat Meas 47, 541-547.

Martin, L., Mercier, N., Incerti, S., Lefrais, Y., Pecheyran, C., Guérin, G., Jarry, M., Bruxelles, L., Bon, F., Pallier, C., 2015. Dosimetric study of sediments at the beta dose rate scale: Characterization and modelization with the DosiVox software. Radiat Meas 81, 134-141.

Mayya, Y.S., Morthekai, P., Murari, M.K., Singhvi, A.K., 2006. Towards quantifying beta microdosimetric effects in single-grain quartz dose distribution. Radiat Meas 41, 1032-1039.

Murray, A.S., Wintle, A.G., 2000. Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. Radiat Meas 32, 57-73.

Rhodes, E.J., Fanning, P.C., Holdaway, S.J., 2010. Developments in optically stimulated luminescence age control for geoarchaeological sediments and hearths in western New South Wales, Australia. Quat Geochronol 5, 348-352.
Rhodes, E.J., Fanning, P.C., Holdaway, S.J., Bolton, C.C., 2009. Archaeological surfaces in western NSW: stratigraphic contexts and preliminary OSL dating of hearths. Terra Australis 28, 12.
Swift, D.A., Sanderson, D.C.W., Nienow, P.W., Bingham, R.G., Cochrane, I.C., 2011. Anomalous luminescence of subglacial sediment at Haut Glacier d'Arolla, Switzerland - a consequence of resetting at the glacier bed? Boreas 40, 446-458.
Thomsen, K.J., Murray, A.S., Bøtter-Jensen, L., 2005. Sources of variability in OSL dose measurements using single grains of quartz. Radiat Meas 39, 47-61.
Thomsen, K.J., Murray, A.S., Buylaert, J.P., Jain, M., Hansen, J.H., Aubry, T., 2016. Testing single-grain quartz OSL methods using sediment samples with independent age control from the Bordes-Fitte rockshelter (Roches d'Abilly site, Central France). Quat Geochronol 31, 77-96.

Supplementary material for "Optically stimulated luminescence dating of heat retainer hearths from the Sahara: Insights into signal accumulation and measurement." Armitage et al.

A. Sample preparation procedure

Having removed the light exposed layers of the heat retainer, dating samples were prepared using standard laboratory techniques. Briefly, carbonate was removed by immersion in 1.16 M HCl, followed by immersion in H_2O_2 to oxidise organic material. Samples FZ84, 85, 95 and 96 were sieved at 40 µm, and the material which passed through the mesh was subsequently Stokes settled to 4-11, 11-20 and 20-40 µm. Material retained in the mesh was sieved to yield 40-60, 60-90, 90-125, 125-150, 150-180 and 180-210 size fractions. Quartz was extracted from the <90 µm fractions via prolonged immersion in H_2SiF_6. Quartz was extracted from the >90 µm fractions via density separation at 2.62 and 2.75 g/cm^3, and 1 hour immersion in 23 M HF. Following H_2SiF_6 or HF treatment, samples were immersed 11 M HCl for a minimum of 12 hours to remove fluoride precipitates. The <60 µm fractions were settled from suspension onto 10 mm diameter aluminium discs, resulting in a monolayer of quartz across the entire upper face of the disc. The >60 µm fractions were mounted on aluminium discs using Silkospray silicone oil applied via a 5 mm mask, resulting in a monolayer of grains across the middle 5 mm of the disc. Single-grain analysis was performed on 180-210 µm grains mounted in Risø single-grain sample holders, containing a 10x10 array of 300 µm diameter holes. Sample preparation and mounting methods are summarised in Table S1.

Table S1. Summary sample preparation and mounting methods for the various grain size fractions reported in this study.

Grain size (µm)	Size separation method	Quartz purification method	Sample mounting method
4-11	Stokes settling	H_2SiF_6 + HCl	Settled from suspension
11-20			
20-40			
40-60	Sieve	Density separation + HF	Silkospray silicone oil via a 5 mm mask
60-90			
90-125			
125-150			
150-180			
180-210			

B. Single-grain rejection criteria.

Grains were rejected where one or more of the following conditions are met: (1) the T_n signal from the grain is too low to distinguish it from the variability in the background signal (the difference between T_n and the average count from the background interval is less than three times the standard deviation of the counts in the background interval, determined using the "sig. >3 sigma above BG" rejection criterion in Luminescence Analyst, (Duller, 2007)); (2) the recycling ratio (Murray and Wintle, 2000) differs from unity by greater than 10%; (3) the OSL IR-depletion ratio (Duller, 2003) is greater than two standard errors below unity; (4) recuperation is high (i.e. L_x/T_x for the 0 Gy regeneration point is greater than 5% of L_n/T_n,(Murray and Wintle, 2000)); (5) L_n/T_n does not intercept the growth curve (Armitage et al., 2000; Bailey et al., 2005) – no grains failed this criterion due to the low D_e of the samples, or (6) the dose response curve shape precludes the generation of a meaningful equivalent dose. Unlike the other five criteria, criterion six requires user-judgement and hence cannot be applied mechanistically. It was used where the signal does not increase monotonically in response to increasing dose, or where L_n/T_n intercepted the dose-response curve at the point where growth ceased, with the former being more common.

C. Equivalent dose, dose rate and calculated age for different size fractions from Al Wafa heat retainers.

Equivalent dose (D_e), dose rate and calculated age data for different size fractions from Al Wafa heat retainers are presented in Tables S2-S5. In these tables, "Mean 4-210" denotes the mean age determined for grain size fraction in the range 4-210 μm. The "effective grain size" is calculated as the grain size with a dose rate which yields the mean 4-210 μm age for the 4-11 μm quartz produced by crushing the bulk sample (Section 5). Dose rates and ages are calculated using a mean water content of 5±2 %, a burial depth of 30 cm (yielding a cosmic dose rate of 0.235±0.020 Gy/ka) and partitioning the gamma dose between the heat retainer and underlying sediments (Armitage and King, 2013). For alpha dose rates, an a-value of 0.04 ± 0.02 was used throughout. Instrumental beta dose rates were corrected for grain size using the values measured by Armitage and Bailey (2005, Table 1).

Table S2: Heat retainer FZ84

		Dose rates (Gy/ka)				
Grain size (μm)	De (Gy)	Alpha	Beta	Gamma	Total	Age (ka)
4-11	7.5 ± 0.2	0.10 ± 0.05	0.41 ± 0.04	0.23 ± 0.02	0.98 ± 0.07	7.7 ± 0.6
11-20	7.9 ± 0.2	0.08 ± 0.04	0.41 ± 0.04	0.23 ± 0.02	0.96 ± 0.07	8.3 ± 0.6
20-40	7.1 ± 0.2	0.06 ± 0.03	0.40 ± 0.04	0.23 ± 0.02	0.93 ± 0.06	7.7 ± 0.5
40-60	7.4 ± 0.2	0.04 ± 0.02	0.40 ± 0.04	0.23 ± 0.02	0.90 ± 0.05	8.2 ± 0.6
60-90	6.9 ± 0.2	0.03 ± 0.01	0.39 ± 0.04	0.23 ± 0.02	0.88 ± 0.05	7.8 ± 0.5
90-125	6.5 ± 0.2	-	0.39 ± 0.04	0.23 ± 0.02	0.85 ± 0.05	7.7 ± 0.5
125-150	6.4 ± 0.2	-	0.38 ± 0.04	0.23 ± 0.02	0.84 ± 0.05	7.6 ± 0.5
150-180	6.1 ± 0.2	-	0.37 ± 0.04	0.23 ± 0.02	0.84 ± 0.05	7.3 ± 0.5
180-210	6.4 ± 0.2	-	0.37 ± 0.04	0.23 ± 0.02	0.83 ± 0.05	7.7 ± 0.5
Mean 4-210	-	-	-	-	-	**7.8 ± 0.5**
		Effective grain size 40-60 μm				
4-11 (crushed)	**7.0 ± 0.2**				**0.90 ± 0.05**	**7.8 ± 0.5**

Table S3: Heat retainer FZ85

		Dose rates (Gy/ka)				
Grain size (μm)	De (Gy)	Alpha	Beta	Gamma	Total	Age (ka)
4-11	10.7 ± 0.3	0.10 ± 0.05	0.37 ± 0.04	0.22 ± 0.02	0.93 ± 0.07	11.5 ± 0.9
11-20	10.8 ± 0.3	0.09 ± 0.04	0.36 ± 0.04	0.22 ± 0.02	0.91 ± 0.07	11.9 ± 0.9
20-40	10.1 ± 0.3	0.06 ± 0.03	0.36 ± 0.04	0.22 ± 0.02	0.88 ± 0.06	11.5 ± 0.8
40-60	9.6 ± 0.3	0.04 ± 0.02	0.35 ± 0.04	0.22 ± 0.02	0.85 ± 0.05	11.3 ± 0.8
60-90	9.4 ± 0.3	0.03 ± 0.01	0.35 ± 0.04	0.22 ± 0.02	0.84 ± 0.05	11.3 ± 0.7
90-125	8.6 ± 0.3	-	0.34 ± 0.04	0.22 ± 0.02	0.80 ± 0.05	10.7 ± 0.7
125-150	8.6 ± 0.3	-	0.34 ± 0.04	0.22 ± 0.02	0.80 ± 0.05	10.8 ± 0.7
150-180	8.3 ± 0.3	-	0.33 ± 0.04	0.22 ± 0.02	0.79 ± 0.05	10.5 ± 0.7
180-210	8.9 ± 0.3	-	0.33 ± 0.04	0.22 ± 0.02	0.79 ± 0.04	11.3 ± 0.7
Mean 4-210	-	-	-	-	-	**11.2 ± 0.8**
		Effective grain size 40-60 μm				
4-11 (crushed)	**9.5 ± 0.3**				**0.85 ± 0.05**	**11.2 ± 0.8**

Table S4: Heat retainer FZ95

Grain size (µm)	De (Gy)	Dose rates (Gy/ka)				Age (ka)
		Alpha	Beta	Gamma	Total	
4-11	5.8 ± 0.2	0.07 ± 0.04	0.32 ± 0.03	0.22 ± 0.02	0.85 ± 0.06	6.9 ± 0.5
11-20	9.4 ± 0.3	0.06 ± 0.03	0.32 ± 0.03	0.22 ± 0.02	0.83 ± 0.05	11.3 ± 0.8*
20-40	4.1 ± 0.1	0.04 ± 0.02	0.31 ± 0.03	0.22 ± 0.02	0.81 ± 0.05	5.1 ± 0.3
40-60	4.8 ± 0.1	0.03 ± 0.01	0.31 ± 0.03	0.22 ± 0.02	0.79 ± 0.04	6.1 ± 0.4
60-90	4.8 ± 0.1	0.02 ± 0.01	0.31 ± 0.03	0.22 ± 0.02	0.77 ± 0.04	6.2 ± 0.4
90-125	4.3 ± 0.1	-	0.30 ± 0.03	0.22 ± 0.02	0.75 ± 0.04	5.7 ± 0.4
125-150	4.6 ± 0.1	-	0.29 ± 0.03	0.22 ± 0.02	0.75 ± 0.04	6.2 ± 0.4
150-180	6.2 ± 0.2	-	0.29 ± 0.03	0.22 ± 0.02	0.74 ± 0.04	8.4 ± 0.5
180-210	4.4 ± 0.1	-	0.29 ± 0.03	0.22 ± 0.02	0.74 ± 0.04	6.0 ± 0.4
Mean 4-210	-	-	-	-	-	**6.3 ± 1.0**
		Effective grain size 90-150 µm				
4-11 (crushed)	**4.7 ± 0.1**				**0.75 ± 0.04**	**6.3 ± 0.4**

*The age for 11-20 µm grain size has been omitted from the calculation of the mean age.

Table S5: Heat retainer FZ96

Grain size (µm)	De (Gy)	Dose rates (Gy/ka)				Age (ka)
		Alpha	Beta	Gamma	Total	
4-11	7.9 ± 0.2	0.07 ± 0.04	0.53 ± 0.06	0.24 ± 0.02	1.09 ± 0.07	7.3 ± 0.5
11-20	8.4 ± 0.2	0.06 ± 0.03	0.53 ± 0.06	0.24 ± 0.02	1.07 ± 0.07	7.9 ± 0.6
20-40	7.4 ± 0.2	0.04 ± 0.02	0.52 ± 0.06	0.24 ± 0.02	1.04 ± 0.07	7.1 ± 0.5
40-60	6.9 ± 0.2	0.03 ± 0.02	0.51 ± 0.06	0.24 ± 0.02	1.02 ± 0.06	6.8 ± 0.5
60-90	6.3 ± 0.2	0.02 ± 0.01	0.51 ± 0.05	0.24 ± 0.02	1.00 ± 0.06	6.3 ± 0.4
90-125	6.7 ± 0.2	-	0.50 ± 0.05	0.24 ± 0.02	0.98 ± 0.06	6.9 ± 0.5
125-150	6.2 ± 0.2	-	0.49 ± 0.05	0.24 ± 0.02	0.97 ± 0.06	6.4 ± 0.4
150-180	5.9 ± 0.2	-	0.48 ± 0.05	0.24 ± 0.02	0.96 ± 0.06	6.1 ± 0.4
180-210	5.9 ± 0.2	-	0.47 ± 0.05	0.24 ± 0.02	0.95 ± 0.06	6.2 ± 0.4
Mean 4-210	-	-	-	-	-	**6.8 ± 0.6**
		Effective grain size >210 µm				
4-11 (crushed)	**5.9 ± 0.2**				**0.87 ± 0.06**	**6.8 ± 0.5**

D. Single grain overdispersion values

Table S6: Single-grain equivalent dose, dose recovery and overdispersion data.

Sample	Equivalent dose data			Dose recovery data			
	D_e (Gy)	Grains (accepted/ measured)	OD_{De} (%)	Ratio (recovered/known)	Grains (accepted/ measured)	OD_{Rec} (%)	OD_{β} (%)
FZ40	9.2 ± 0.3	131/1000	18.5 ± 1.5	0.98 ± 0.02	135/1200	9.7 ± 1.1	15.8 ± 1.8
FZ47	7.3 ± 0.3	89/1200	24.9 ± 2.3	0.99 ± 0.01	164/1200	7.7 ± 0.7	23.7 ± 2.4
FZ84	6.0 ± 0.1	171/1200	26.5 ± 1.6	1.00 ± 0.01	242/1200	7.5 ± 0.7	25.4 ± 1.8
FZ85	8.6 ± 0.1	262/1200	21.0 ± 1.1	0.96 ± 0.01	276/1200	6.3 ± 0.6	20.0 ± 1.2
FZ95	4.5 ± 0.1	242/1100	28.2 ± 1.5	0.98 ± 0.01	281/1200	7.9 ± 0.7	27.1 ± 1.6
FZ96	6.0 ± 0.1	176/1200	24.5 ± 1.5	0.99 ± 0.01	195/1200	6.3 ± 0.8	23.7 ± 1.7

E. Equivalent doses for uncrushed and crushed quartz separates

Table S7: Sample description and equivalent doses for uncrushed and crushed fractions of zero-age quartzes.

Sample	Provenance	Equivalent dose (Gy)		Increase (Gy)
		Uncrushed	Crushed	(crushed-uncrushed)
BA5	Coastal dune (Armitage et al., 2006)	0.04 ± 0.01	0.23 ± 0.02	0.19 ± 0.02
CH62	Shoreline (Armitage et al., 2015)	0.01 ± 0.01	0.11 ± 0.02	0.10 ± 0.02
FZ16	Desert dune (Armitage et al., 2007)	0.04 ± 0.03	0.12 ± 0.04	0.08 ± 0.05
FZ32	Desert dune (Armitage and Bailey, 2005a)	0.08 ± 0.08	0.16 ± 0.08	0.08 ± 0.12
IN22	Coastal dune (Armitage et al., 2006)	0.01 ± 0.02	0.02 ± 0.01	0.01 ± 0.02
Risø quartz	Calibration sample (Hansen et al., 2015)	0.00 ± 0.00	0.09 ± 0.01	0.09 ± 0.01
SA10	Coastal dune (this study)	0.00 ± 0.00	0.93 ± 0.33	0.93 ± 0.33
			Mean	**0.21 ± 0.12**

Table S8: Sample description and equivalent doses for uncrushed and crushed fractions of Holocene quartzes.

Sample	Provenance	Equivalent dose (Gy)		Ratio
		Uncrushed	Crushed	(uncrushed/crushed)
CH22	Shoreline (Armitage et al., 2015)	3.72 ± 0.13	4.07 ± 0.19	1.09 ± 0.06
IN15	Coastal dune (Armitage et al., 2006)	5.35 ± 0.07	5.31 ± 0.06	0.99 ± 0.02
IN16	Coastal dune (Armitage et al., 2006)	2.34 ± 0.11	2.53 ± 0.12	1.08 ± 0.07
NG38	Shoreline (Armitage et al., 2015)	19.5 ± 0.2	20.3 ± 1.2	1.04 ± 0.06
NG39	Shoreline (Armitage et al., 2015)	19.1 ± 0.8	19.4 ± 0.5	1.02 ± 0.05
			Mean	**1.04 ± 0.02**

References

Aitken, M.J., 1985. Thermoluminescence dating. Academic press.

Armitage, S., Bailey, R., 2005a. The measured dependence of laboratory beta dose rates on sample grain size. Radiat Meas 39, 123-127.

Armitage, S., Duller, G., Wintle, A., 2000. Quartz from southern Africa: sensitivity changes as a result of thermal pretreatment. Radiat Meas 32, 571-577.

Armitage, S.J., Bailey, R.M., 2005b. The measured dependence of laboratory beta dose rates on sample grain size. Radiat Meas 39, 123-127.

Armitage, S.J., Botha, G.A., Duller, G.A.T., Wintle, A.G., Rebelo, L.P., Momade, F.J., 2006. The formation and evolution of the barrier islands of Inhaca and Bazaruto, Mozambique. Geomorphology 82, 295-308.

Armitage, S.J., Bristow, C.S., Drake, N.A., 2015. West African monsoon dynamics inferred from abrupt fluctuations of Lake Mega-Chad. P Natl Acad Sci USA 112, 8543-8548.

Armitage, S.J., Drake, N.A., Stokes, S., El-Hawat, A., Salem, M., White, K., Turner, P., McLaren, S.J., 2007. Multiple phases of north African humidity recorded in lacustrine sediments from the fazzan basin, Libyan sahara. Quat Geochronol 2, 181-186.

Armitage, S.J., Jasim, S.A., Marks, A.E., Parker, A.G., Usik, V.I., Uerpmann, H.-P., 2011. The Southern Route "Out of Africa": Evidence for an Early Expansion of Modern Humans into Arabia. Science 331, 453-456.

Armitage, S.J., King, G.E., 2013. Optically stimulated luminescence dating of hearths from the Fazzan Basin, Libya: A tool for determining the timing and pattern of Holocene occupation of the Sahara. Quat Geochronol 15, 88-97.

Bailey, R.M., Armitage, S.J., Stokes, S., 2005. An investigation of pulsed-irradiation regeneration of quartz OSL and its implications for the precision and accuracy of optical dating (Paper II). Radiat Meas 39, 347-359.

Ballarini, M., Wintle, A.G., Wallinga, J., 2006. Spatial variation of dose rate from beta sources as measured using single grains. Ancient TL 24, 1-7.

Bateman, M.D., Swift, D.A., Piotrowski, J.A., Sanderson, D.C.W., 2012. Investigating the effects of glacial shearing of sediment on luminescence. Quat Geochronol 10, 230-236.

Bøtter-Jensen, L., Andersen, C.E., Duller, G.A.T., Murray, A.S., 2003. Developments in radiation, stimulation and observation facilities in luminescence measurements. Radiat Meas 37, 535-541.
Brennan, B.J., 2003. Beta doses to spherical grains. Radiat Meas 37, 299-303.
Brennan, B.J., Lyons, R.G., Phillips, S.W., 1991. Attenuation of alpha particle track dose for spherical grains. International Journal of Radiation Applications and Instrumentation. Part 18, 249-253.
Cunningham, A.C., Devries, D.J., Schaart, D.R., 2012. Experimental and computational simulation of beta-dose heterogeneity in sediment. Radiat Meas 47, 1060-1067.
Duller, G., 2007. Assessing the error on equivalent dose estimates derived from single aliquot regenerative dose measurements. Ancient TL 25, 15-24.
Duller, G.A.T., 2003. Distinguishing quartz and feldspar in single grain luminescence measurements. Radiat Meas 37, 161-165.
Duller, G.A.T., Bøtter-Jensen, L., Murray, A.S., Truscott, A.J., 1999. Single grain laser luminescence (SGLL) measurements using a novel automated reader. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms 155, 506-514.
Galbraith, R.F., Roberts, R.G., Laslett, G.M., Yoshida, H., Olley, J.M., 1999. Optical dating of single and multiple grains of quartz from Jinmium rock shelter, northern Australia: Part I, experimental design and statistical models. Archaeometry 41, 339-364.
Guérin, G., Jain, M., J. Thomsen, K., S. Murray, A., Mercier, N., 2015. Modelling dose rate to single grains of quartz in well-sorted sand samples: The dispersion arising from the presence of potassium feldspars and implications for single grain OSL dating. Quat Geochronol 27, 52-65.
Guérin, G., Mercier, N., 2012. Preliminary insight into dose deposition processes in sedimentary media on a scale of single grains: Monte Carlo modelling of the effect of water on the gamma dose rate. Radiat Meas 47, 541-547.
Hansen, V., Murray, A., Buylaert, J.P., Yeo, E.Y., Thomsen, K., 2015. A new irradiated quartz for beta source calibration. Radiat Meas 81, 123-127.
Martin, L., Mercier, N., Incerti, S., Lefrais, Y., Pecheyran, C., Guérin, G., Jarry, M., Bruxelles, L., Bon, F., Pallier, C., 2015. Dosimetric study of sediments at the beta dose rate scale: Characterization and modelization with the DosiVox software. Radiat Meas 81, 134-141.
Mayya, Y.S., Morthekai, P., Murari, M.K., Singhvi, A.K., 2006. Towards quantifying beta microdosimetric effects in single-grain quartz dose distribution. Radiat Meas 41, 1032-1039.
Murray, A.S., Wintle, A.G., 2000. Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. Radiat Meas 32, 57-73.
Rhodes, E.J., Fanning, P.C., Holdaway, S.J., 2010. Developments in optically stimulated luminescence age control for geoarchaeological sediments and hearths in western New South Wales, Australia. Quat Geochronol 5, 348-352.
Rhodes, E.J., Fanning, P.C., Holdaway, S.J., Bolton, C.C., 2009. Archaeological surfaces in western NSW: stratigraphic contexts and preliminary OSL dating of hearths. Terra Australis 28, 12.
Swift, D.A., Sanderson, D.C.W., Nienow, P.W., Bingham, R.G., Cochrane, I.C., 2011. Anomalous luminescence of subglacial sediment at Haut Glacier d'Arolla, Switzerland - a consequence of resetting at the glacier bed? Boreas 40, 446-458.
Thomsen, K.J., Murray, A.S., Bøtter-Jensen, L., 2005. Sources of variability in OSL dose measurements using single grains of quartz. Radiat Meas 39, 47-61.
Thomsen, K.J., Murray, A.S., Buylaert, J.P., Jain, M., Hansen, J.H., Aubry, T., 2016. Testing single-grain quartz OSL methods using sediment samples with independent age control from the Bordes-Fitte rockshelter (Roches d'Abilly site, Central France). Quat Geochronol 31, 77-96.