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Optical dating in archaeology: thirty years in retrospect and grand challenges for the future

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Abstract

In 1982, when Richard Klein first became one of the Editors of this journal, the luminescence dating community was embarking on a new phase of exploratory research. Attention was turning from the use of thermoluminescence (TL) dating to estimate the time of last heating of archaeological objects, such as pottery and burnt flint, to the TL dating of unheated sediments that had been transported by wind and then deposited on the landscape. This revolutionary development enabled the extension of TL dating to sedimentary deposits in a variety of environmental settings and to the multitude of archaeological sites that lack suitably heated artefacts. In sediment dating, the age of most interest is usually the time elapsed since grains of quartz or feldspar were last exposed to sunlight, as the energy of the sun's rays is sufficient to evict electrons from their light-sensitive traps. These traps are steadily refilled after sediment deposition and the longer the grains remain buried, the more TL they will emit when measured. In 1985, Huntley and colleagues proposed 'optical dating' as a simpler and superior means of stimulating the light-sensitive traps in Quaternary sediments, and this is now the principal luminescence-based method of dating geological and archaeological deposits. Optical dating is an umbrella term for an armada of acronyms, the most common in archaeological contexts being OSL (optically stimulated luminescence), TT-OSL (thermally-transferred OSL), IRSL (infrared stimulated luminescence) and pIRIR (post-infrared IRSL). All of these variants are founded on the same basic tenet - measurement of a light-sensitive signal to determine (typically) the last time that sediment grains were sun-bleached - but each approach has its virtues and vices. In this paper, we review this 'family' of luminescence dating techniques and look back on 30 years of optical dating in archaeology. Some of the more interesting and important achievements are highlighted, including the critical insights gained in the last two decades from OSL measurements of individual grains of quartz. We also look to the future of optical dating in archaeological contexts. Efforts to extend the age limits of optical dating to older hominin and archaeological sites will remain a key goal, and understanding how archaeological sites - of all ages - form and evolve over time could be improved greatly by combining micromorphology analysis with optical dating of undisturbed (intact) sediments. The latter poses a series of particularly formidable technical challenges, but if the past is any guide to the future, then we can expect optical dating to illuminate much more of human history before celebrating its Golden Jubilee.

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Optical dating in archaeology: thirty years in retrospect and grand challenges for the future

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Abstract

In 1982, when Richard Klein first became one of the Editors of this journal, the luminescence dating community was embarking on a new phase of exploratory research. Attention was turning from the use of thermoluminescence (TL) dating to estimate the time of last heating of archaeological objects, such as pottery and burnt flint, to the TL dating of unheated sediments that had been transported by wind and then deposited on the landscape. This revolutionary development enabled the extension of TL dating to sedimentary deposits in a variety of environmental settings and to the multitude of archaeological sites that lack suitably heated artefacts. In sediment dating, the age of most interest is usually the time elapsed since grains of quartz or feldspar were last exposed to sunlight, as the energy of the sun's rays is sufficient to evict electrons from their light-sensitive traps. These traps are steadily refilled after sediment deposition and the longer the grains remain buried, the more TL they will emit when measured. In 1985, Huntley and colleagues proposed 'optical dating' as a simpler and superior means of stimulating the light-sensitive traps in Quaternary sediments, and this is now the principal luminescence-based method of dating geological and archaeological deposits. Optical dating is an umbrella term for an armada of acronyms, the most common in archaeological contexts being OSL (optically stimulated luminescence), TT-OSL (thermally-transferred OSL), IRSL (infrared stimulated luminescence) and pIRIR (post-infrared IRSL). All of these variants are founded on the same basic tenet – measurement of a light-sensitive signal to determine (typically) the last time that sediment grains were sun-bleached – but each approach has its virtues and vices. In this paper, we review this 'family' of luminescence dating techniques and look back on 30 years of optical dating in archaeology. Some of the more interesting and important achievements are highlighted, including the critical insights gained in the last two decades from OSL measurements of individual grains of quartz. We also look to the future of optical dating in archaeological contexts. Efforts to extend the age limits of optical dating to older hominin and archaeological sites will remain a key goal, and understanding how archaeological sites – of all ages – form and evolve over time could be improved greatly by combining micromorphology analysis with optical dating of undisturbed (intact) sediments. The latter poses a series of particularly formidable technical challenges, but if the past is any guide to the future, then we can expect optical dating to illuminate much more of human history before celebrating its Golden Jubilee.

1. Ancestral flashbacks

The potential for luminescence dating in archaeological contexts can be traced back to Daniels et al. (1953), who were the first to suggest that the luminescence response of naturally occurring minerals to ionising radiation could be used as a tool for estimating the time since ancient pottery was last heated. Tests on archaeological ceramics followed within a few years (Grögler et al., 1960). For the next two decades, attention was focussed on heated pottery and ceramics from archaeological sites (for dating) and museum collections (for authenticity testing), as well as burnt flints, baked hearth sediments, oven stones from burnt mounds and other heated objects (Fleming, 1979; Aitken, 1985, 1990; Wintle, 2008).

In these pioneering studies, mineral grains were stimulated in the laboratory by heating them to 500°C, and the thermally-induced glow – or thermoluminescence (TL) – was detected by a photomultiplier tube. Unlike incandescence, the production of TL requires that the grains had received a prior dose of ionising radiation, which is derived mainly from local sources of environmental radioactivity – namely, the sediments surrounding the artefacts and chemical impurities inside the artefacts themselves.

Many minerals emit luminescence, but quartz and feldspar – the two most abundant minerals on Earth – have been used most often for dating. Grains act as tiny radiation dosimeters, absorbing the incoming radiation energy and storing a small fraction of it as trapped electrons at defects in their crystal lattices. These electrons can be released from their traps by heating the grains in the laboratory, with the intensity of the induced TL providing a measure of the time elapsed since the object was last heated. The *Journal of Archaeological Science* was quick to publish a review of TL dating in just its second year (Seeley, 1975).

In the early 1960s, it was noticed that the TL traps in calcite and sediment could be bleached by sunlight (Aitken et al., 1963), but Soviet scientists working on loess deposits in the Ukraine were the first to apply TL dating to unheated sediments (Shelkopyas and Morozov, 1965). They proposed that the trap-emptying mechanism was weathering, grinding by glaciers and exposure to sunlight. By the late 1970s, the light-sensitive TL traps in silt- and sand-sized grains of quartz and feldspar were being investigated more widely as a means of dating terrestrial and marine deposits (Dreimanis et al., 1978; Wintle and Huntley, 1979). In 1982 – the same year that Richard Klein became an Editor of this journal – a landmark review of these revolutionary developments was published in the first issue of *Quaternary Science Reviews* (Wintle and Huntley, 1982), and three other papers laid the groundwork for TL dating of sediments transported and deposited by wind and water (Prescott, 1982; Readhead, 1982; Singhvi et al., 1982).

Three years later, Huntley et al. (1985) proposed a more direct and effective means of accessing the light-sensitive electron traps: shine a powerful green laser on the mineral grains and measure the resulting optically-induced luminescence. They coined the term ‘optical dating’ for this new method, and reported the first-ever optical ages for archaeological sediments. Hütt et al. (1988) subsequently found that infrared photons were sufficiently energetic to stimulate luminescence from potassium feldspars, thereby enabling optical dating using inexpensive infrared light-emitting diodes (LEDs). By the mid-1990s, optical dating had replaced TL dating as the method of choice for sediments that had been exposed to the sun’s rays prior to deposition. Reviews of TL dating and the first decade of optical dating – with examples of archaeological applications – are available elsewhere (Feathers, 1996; Roberts, 1997; Aitken, 1998), including a horizon scan of promising new opportunities for optical dating that was published in this journal (Wintle, 1996).

Here we pick up the story in the late 1990s, when optical dating was undergoing a major transformation – the development of ‘single aliquot’ methods to measure the radiation energy stored in separate portions and individual grains of heated and unheated quartz. We also reflect on some current advances and future directions of optical dating that could further illuminate our human past. However, this paper is not intended as a comprehensive review of either the field of optical dating or the wide range of archaeological questions to which the technique has been applied. The choice of subject matter instead reflects our personal research interests in archaeology and geochronology, drawing extensively on case studies from our own investigations, and we appreciate that others may not share our views of the most important or promising developments. For example, we do not discuss applications of optical dating to heated artefacts,

anthropogenic structures or Holocene archaeological deposits, but we cite many other publications that readers can consult for information on such topics, such as the recent overview by Liritzis et al. (2013). Finally, although this paper is focussed on archaeological sediments deposited during the Pleistocene, optical dating also has many uses in much younger contexts and may be preferable to radiocarbon (^{14}C) dating of archaeological events and objects from the recent past.

2. Optical dating: the basics

How do we calculate optical ages? At its simplest, the age equation can be expressed in the following form:

$$\text{age} = \text{equivalent dose} / \text{environmental dose rate}$$

The equivalent dose is reported in gray (Gy), where 1 Gy = 1 J/kg of absorbed radiation energy. Mineral grains will absorb energy while buried and shielded from light, resulting in the gradual filling of vacant electron traps. So the equivalent dose is a measure of the amount of energy stored – and time elapsed – since the traps were last emptied by sunlight. The term ‘equivalent’ is needed because the number of trapped electrons depends on both the dose received and the type of radiation (Huntley et al., 1985). The equivalent dose is sometimes referred to as the palaeodose, but the term used should be ‘palaeodose equivalent’ because the actual past radiation dose is not determined (Huntley, 2001). The denominator in the equation – the environmental dose rate – is reported in Gy per unit time and represents the rate of delivery of all environmental sources of ionising radiation to the grains over the same time span. As the true dose rate is not measured, the term ‘equivalent’ applies to the denominator also, but is frequently omitted (Huntley, 2001; Lian and Huntley, 2001).

Optical ages are calculated directly in calendar years (or sidereal years, strictly speaking), so there is no need for subsequent calibration such as that applied to ^{14}C ages to convert ‘radiocarbon years’ into sidereal years. The denominator and numerator in the age equation are measured using different methods. Here we summarise only the main aspects of their measurement, as currently practiced, to provide background context for the current and future developments described later. Aitken (1998), Lian and Huntley (2001) and Duller (2008a) give additional details, written with end-users in mind, including practical guidelines on how to collect samples in the field. Other overviews of optical (and TL) dating with archaeological examples include Troja and Roberts (2000), Bøtter-Jensen et al. (2003), Feathers (2003), Lamothe (2004), Lian and Roberts (2006), Jacobs and Roberts (2007), Preusser et al. (2008), Rhodes (2011), Burbidge (2012), Lian (2013), Liritzis et al. (2013) and Wintle (2014).

2.1 The denominator

The environmental dose rate is the sum of the individual alpha, beta and gamma dose rates, plus the contribution from cosmic rays, which is usually estimated from published equations (Prescott and Hutton, 1994). Cosmic rays typically account for only a small fraction of the total dose rate, with the majority supplied by the radioactive decay of uranium and thorium (^{238}U , ^{235}U , ^{232}Th and the daughter products in each of these chains) and potassium (^{40}K) in the materials surrounding the dated grains or artefacts. These radioactive elements are ubiquitous at low concentrations in the natural environment. For sand-sized grains of quartz (and sometimes feldspar), the external alpha-irradiated rinds (the outermost $\sim 25\ \mu\text{m}$) are effectively removed during sample preparation by etching the grains in hydrofluoric acid, leaving beta and gamma radiation as the chief contributors to the dose rate. For potassium feldspar grains, there is also a significant internal beta dose rate from the radioactive decay of ^{40}K and rubidium (^{87}Rb) inside the grains. Aitken (1985, 1990, 1998) and Duller (2008a) review the various contributors to the environmental dose rate and the methods most often used to measure them.

Ideally, the dose rate should have remained constant since the sample was last heated or buried. This requires that the uranium and thorium chains have been in secular equilibrium – that is, the parent and daughter radionuclides are present at equal activities – over this time span, or that the extent of any disequilibrium has not changed. The measured beta and gamma dose rates may need to be adjusted if

time-dependent changes are implicated (Krbetschek et al., 1994; Prescott and Hutton, 1995; Olley et al., 1996, 1997). A common form of disequilibrium in limestone environments is the leaching of uranium by percolating groundwater, and radon gas may also be lost to the atmosphere. But if these and other forms of disequilibria have operated continuously over the period of sample burial, then the measured dose rates do not require adjustment. Moreover, most time-dependent disequilibria in the ^{238}U series are unlikely to give rise to errors in the total dose rate of more than a few percent, unless only the parent uranium activity is measured (Olley et al., 1996, 1997).

The beta dose rate commonly contributes most to the total dose rate. Beta particles can penetrate up to ~3 mm through sediment, so differences in radioactivity over small distances can have a substantial effect on the calculated dose rate. The total beta dose rate can be measured directly by counting all nuclear disintegrations that emit a beta particle (Bøtter-Jensen and Mejdahl, 1988; Sanderson, 1988) or estimated by measuring individual radionuclide concentrations and then converting these to beta dose rates. The latter techniques include inductively-coupled plasma mass spectrometry (ICP-MS) and neutron activation, which measure the parent uranium and thorium concentrations, as well as potassium. High-resolution gamma spectrometry and alpha-particle spectrometry give additional information about the current state of (dis)equilibrium of the ^{238}U and ^{232}Th decay chains from the activities of individual radionuclides, but not all occurrences of disequilibrium in the past will have left traces that persist to the present day. Thick-source alpha counting – another emission-counting method – can also be used to determine uranium and thorium concentrations (Aitken, 1985), and these can be combined with measurements of the potassium content (using techniques such as atomic absorption or emission spectroscopy, inductively-coupled plasma optical emission spectroscopy or X-ray fluorescence) to calculate the total beta dose rate.

Each of these approaches has its merits and disadvantages. For samples affected by disequilibrium in the ^{238}U series, an unreliable estimate of the beta dose rate might be obtained if only the uranium concentration is measured and the parent is not in secular equilibrium with its daughters. In such instances, methods such as beta-counting and high-resolution gamma spectrometry should provide more accurate estimates of the beta dose rate for the entire chain (Olley et al., 1996, 1997). Another consideration when measuring the beta dose rate is the size of the measured sample and its exact location relative to the dated grains. The dosimetry sample is meant to give an accurate indication of the beta dose rate experienced by these grains. But this presents something of a conundrum for single-grain dating, as it is not currently feasible to reliably measure the beta dose rate at such small spatial scales. While equivalent doses can be determined for single grains, the dose rate to each grain is estimated from the bulk sample. To calculate true single-grain ages requires measuring and modelling *in situ* beta dose rates to individual grains. Ongoing efforts and future directions in this field are discussed in Section 4.

The gamma dose rate can also be estimated using these laboratory techniques, but field measurements are generally preferred as gamma rays can penetrate up to ~0.5 m through sediment and rock. Many archaeological deposits, especially in caves and rock shelters, have heterogeneous compositions at this spatial scale, so portable gamma-ray detectors are commonly used to measure the *in situ* gamma dose rate at each sample position. These measurements can take less than an hour, and can thus be conveniently made while excavations are in progress. Alternatively, luminescence dosimetry capsules – usually calcium sulphate doped with dysprosium ($\text{CaSO}_4\cdot\text{Dy}$) or aluminium oxide doped with carbon ($\text{Al}_2\text{O}_3\cdot\text{C}$) – can be placed in the deposit to measure the combined dose rate from gamma and cosmic rays. A drawback with capsules is that they have to be buried for several weeks or months, and it may not be feasible to position them in the exact same locations as the luminescence dating samples if excavations have continued in the meantime. Capsules record the total gamma dose rate from radionuclides in the uranium and thorium chains and ^{40}K , as do gamma-ray detectors when the ‘threshold’ technique is used (Mercier and Falguères, 2007), so both should provide a robust measure of the total gamma dose rate prevailing at the time of sample collection – even if the decay chains are not currently in secular equilibrium.

The external components of the environmental dose rate are influenced (to differing extents) by the presence of interstitial water in the deposit, as water absorbs some of the radiation energy that would otherwise have reached the grains. So an estimate must be made of the sample water content over its

entire period of burial, which may or may not be similar to the value measured for the modern-day sample. Most archaeological deposits have water contents that range from a few percent to near-saturated (20–30% mass of water to mass of dry sample). Some uncertainty must be assigned to each estimate, sufficient to cover all reasonable fluctuations in the *mean* value. The relative uncertainty may need to be larger for older samples, to allow for potential effects of climatic and hydrological changes over long time spans. But the water content has less influence on the final calculated age than is often appreciated: for sandy quartz sediments, the calculated age typically increases (or decreases) by only ~1% for each 1% increase (or decrease) in water content.

For some samples, the uncertainties associated with changes in water content and other complicating factors associated with the environmental dose rate, such as radionuclide migration, can be eliminated by using the ‘isochron’ or ‘subtraction’ technique (Mejdahl, 1983; Zhao and Li, 2002; Li and Li, 2008a, 2008b). The age is calculated from the differences in the equivalent doses and internal dose rates of quartz and/or feldspar grains of different sizes extracted from the same sample, assuming that all grains were deposited at the same time and thereafter experienced the same external dose rate. Li et al. (2011) reported reliable isochron ages for feldspar grains deposited up to ~250 ka ago. The technique is limited, however, to samples that contain feldspar grains in a range of sizes, with the stipulation that the grains were well-bleached at deposition and did not subsequently become mixed with older or younger grains.

2.2 The numerator

The ‘optical’ part of optical dating refers to the use of photons to stimulate the light-sensitive traps in quartz and feldspar, with the equivalent dose – commonly abbreviated as D_e – estimated from the intensity of the induced luminescence. Over the last 30 years, a variety of light sources have been used to stimulate these traps, resulting in a proliferation of terms (several of which are synonyms) and acronyms in the literature. Table 1 summarises the main terms encountered, and how they relate to each other.

INSERT TABLE 1 HERE

The key feature that differentiates these variants is the colour of the light – that is, the wavelength or energy of the photons – used to stimulate the electron traps. Blue or green light is most often used for quartz, and the induced ultraviolet OSL signal is selected for dating. Feldspars, unlike quartz, are very sensitive to infrared photons, so they are usually stimulated using infrared LEDs and the violet/blue IRSL emissions are chosen for dating. The contrasting infrared sensitivity of these two minerals can be exploited to check the purity of quartz and to stimulate feldspars in the presence of quartz. The OSL and IRSL signals are much too weak to be seen with the naked eye, so they are measured using sensitive instruments – normally a photomultiplier tube fitted with glass filters to restrict transmission to particular photons.

When grains of quartz are exposed to light – whether natural sunlight or photon stimulation in the laboratory – the traps giving rise to the OSL signal are quickly depleted. The most light-sensitive part of the quartz OSL signal, called the ‘fast’ component, decays exponentially to background within a few seconds (Fig. 1a and 1b), so it is the component of choice for sediment dating (Bailey et al., 1997; Galbraith et al., 1999; Wintle and Murray, 2006). Compared to the fast OSL component, the IRSL traps in feldspar bleach both more slowly and less completely (Godfrey-Smith et al., 1988; Thomsen et al., 2008) and the most light-sensitive IRSL traps also suffer from a malign phenomenon known as ‘anomalous fading’ (Wintle, 1973; Spooner, 1994), which refers to the leakage of electrons at a much faster rate than expected from kinetic considerations (Li and Tso, 1997; Huntley and Lian, 2006; Kars et al., 2013).

INSERT FIG. 1 HERE

Unless a fading correction is made, the calculated IRSL ages will be too young, with the shortfall increasing with the age of the sample (Lamothe and Auclair, 1999; Huntley and Lamothe, 2001; Wallinga et al., 2007). A correction method, based on laboratory measurements of the fading rate, is available for feldspars deposited within the last 50 thousand years (ka) or so (Huntley and Lamothe, 2001). This approach is not

valid for older samples, due to the dose dependency of the fading rate and other factors (Huntley and Lian, 2006; Kars et al., 2008; Li and Li, 2008), but procedures have been developed recently that can access traps that fade much less (Thomsen et al., 2008; Buylaert et al., 2009, 2012a) or do not fade at all (Li and Li, 2011; Li et al., 2014a, 2014b). These post-infrared IRSL (pIRIR) traps are not as light-sensitive as those responsible for the conventional IRSL or fast-component OSL signals (Poolton et al., 2002; Buylaert et al., 2012a), leading to a trade-off between minimising the extent of fading and ensuring that the pIRIR traps were emptied by sunlight in the past.

The D_e is determined in a similar way for both minerals. In practical terms, the D_e is the laboratory radiation dose required to produce the same OSL or IRSL intensity as that emitted by the natural sample, where the term 'natural' refers to grains that have not been exposed to ionising radiation, heat or light (except for the laboratory safelights) since the sample was collected. Historically, a single estimate of D_e was obtained by extrapolation of a calibration curve of the growth in luminescence intensity with added dose, using multiple (e.g., 20–50) sample replicates – or 'aliquots' – each composed of several thousand grains (Berger, 1995; Wintle, 1997; Aitken, 1998). But the benefits of measuring the D_e from a single aliquot of sample was recognised from the dawn of optical dating (Huntley et al., 1985) and single-aliquot additive-dose procedures were developed in the 1990s for potassium feldspars (Duller, 1991; Galloway, 1996) and quartz (Murray et al., 1997).

These original single-aliquot procedures had several shortcomings, however. The crucial breakthrough was the development of a single-aliquot regenerative-dose (SAR) procedure, which incorporated a means of monitoring and correcting for the sensitivity changes that accompanied the repeated cycles of laboratory bleaching, irradiation and heat treatment (or 'preheat') given to each aliquot or grain (Murray and Roberts, 1998; Galbraith et al., 1999; Murray and Wintle, 2000; Wallinga et al., 2000). Current SAR procedures use the OSL or IRSL signals induced by stimulating a small test dose given repeatedly during the measurement sequence as the sensitivity-correction device, and a 'preheat plateau test' – in which the natural grains are subjected to a range of alternative preheats – is sometimes used to check if the D_e values show any dependence on the combination of preheat temperature and duration (Aitken, 1998). The first applications of the SAR procedure were on quartz grains from archaeological sites (Murray and Roberts, 1997; Roberts et al., 1997, 1998a, 1998b, 1999; Galbraith et al., 1999; Murray and Mejdahl, 1999) and subsequent methodological improvements have often taken place in archaeological contexts.

To estimate the D_e , the natural signal (usually abbreviated as L_n) is compared to the signals observed when the same aliquot or grain is given a series of radiation doses in the laboratory (L_x). The test dose signals measured in the natural and regenerative dose cycles (T_n and T_x , respectively) are used to calculate the ratios L_n/T_n and L_x/T_x . The latter are plotted against the corresponding regenerative doses and the data points are fitted with a mathematical function (commonly a saturating exponential, with an additional linear component in some cases) to construct a sensitivity-corrected dose-response (growth) curve for each aliquot or grain (Fig. 1 inset plots). The L_n/T_n ratios are then projected on to their respective growth curves to obtain the D_e values by interpolation. This approach has been standard practice since first being applied to 57 single grains of quartz from sediment samples collected at an archaeological site in northern Australia (Galbraith et al., 1999; Roberts et al., 1999) and then to multi-grain aliquots of heated and sedimentary quartz from Africa and Europe (Murray and Wintle, 2000).

The fast component of quartz OSL typically saturates at doses of 150–250 Gy, whereas the thermally-transferred OSL (TT-OSL) and violet stimulated luminescence (VSL) emissions from quartz – and also the non-fading pIRIR signals from feldspar – saturate at much higher doses and, hence, have the capacity to date much older events (see Section 5). The 'characteristic saturation dose' (denoted as D_0) is one of the parameters of a saturating exponential function, and D_0 values are commonly used to assess the long-range dating potential of luminescence signals. Following Wintle and Murray (2006), some researchers use twice the D_0 value ($2D_0$) as an upper limit for reliable D_e estimation. For example, this value was used to calculate the minimum age of 430 ka for an engraved shell at Trinil on Java (Joordens et al., 2015). The $2D_0$ value represents the dose at which the growth curve attains 86% of the saturation intensity, but there is no mathematical impediment to estimating the D_e at higher D_0 values if the shape of the growth curve can be

characterised accurately (Galbraith and Roberts, 2012). The doses at 90% and 95% of the saturation level correspond to 2.3 and 3 times the D_0 value, respectively. D_e values will be estimated with lower precision at higher doses, because the growth curve becomes flatter as it approaches saturation, but reliable D_e estimates of up to 3 times the D_0 value have been obtained for feldspars using the non-fading pIRIR signals (Li and Li, 2012; Li et al., 2014a, 2015).

The SAR procedure incorporates several quality-assurance checks of sample suitability. In the 'recycling ratio test', an identical regenerative dose is given near the start and end of the measurement sequence to check that the sensitivity correction has performed successfully. The latter is judged from the statistical consistency of the repeat L_x/T_x measurements: if this recycling ratio differs significantly from a value of 1, then the sensitivity correction has failed and those aliquots or grains may justifiably be removed from the data set. Another routine check is the 'dose recovery test', in which some grains are deliberately bleached and then given a known laboratory dose; the SAR procedure is then applied to verify that the correct (known) dose is recovered (Galbraith et al., 1999; Wallinga et al., 2000). The dose recovery and preheat plateau tests are used to select the most suitable measurement parameters for each sample, including the optimal combination of preheat temperature and duration, but neither takes into account a host of other possible complications, such as non-identical field and laboratory conditions. The outcomes of these tests should thus be viewed as 'best case' scenarios (Galbraith et al., 2005), rather than as assurances that the D_e values and optical ages for the natural samples will necessarily be accurate – the latter can best be gauged by comparison with independent chronologies (Rittenour, 2008; Li et al., 2014a).

A 'recuperation test' is also regularly included as a zero-dose cycle in the SAR sequence. After measuring the T_x signal in the preceding cycle, each aliquot or grain is preheated (without first receiving a regenerative dose) and the L_x signal is then measured. As the OSL or IRSL traps should be vacant, this test provides a check on the extent to which they are refilled by electrons thermally transferred from deeper traps and/or by differential sensitivity changes in the various OSL components of quartz, which can result in a progressive increase in the amount of 'slow' component (Bailey et al., 1997) remaining after each SAR cycle (Arnold et al., 2008; Gliganic et al., 2012; Jacobs et al., 2015). The presence of a dominant fast component in the quartz OSL signal can be assessed using the 'fast ratio' (Durcan and Duller, 2011), and a plot of D_e against stimulation time used to examine the degree to which the traps responsible for the different OSL components were bleached before sediment deposition (Bailey et al., 2003; Singarayer et al., 2005). The fast component can also be separated mathematically from the less light-sensitive components (Bailey et al., 1997; Galbraith et al., 1999; Bulur et al., 2000; Singarayer and Bailey, 2003; Cunningham and Wallinga, 2009) – albeit with caution (Adamiec, 2005) – or measured directly using a post-infrared OSL procedure if the emissions are sufficiently intense (Bailey, 2010).

For aliquots or grains that satisfy these internal tests, their D_e values are then combined to obtain a final D_e estimate for age determination. In the simplest (and most desirable) case, the individual D_e values will be statistically consistent with some average D_e value and can, thus, be represented by the mean or weighted mean (Galbraith et al., 1999). Many archaeological deposits are more complex, however, and require single-grain analyses – coupled with a detailed understanding of site formation and sample context at scales ranging from landscape to microfacies – to obtain reliable ages. In the next section, we show how single-grain measurements can yield valuable information about site integrity and the fidelity of optical ages, especially when combined with studies of the sediment micromorphology (Jacobs and Roberts, 2007; Marean et al., 2007; Feathers et al., 2010; Jacobs et al., 2011b; Araujo et al., 2013; Jankowski et al., 2015).

3. Single grains as timekeepers and tracers

In the last two decades, many studies have shown the benefits of measuring individual sand-sized grains of quartz to determine the D_e value for a sample, rather than using single aliquots composed of tens, hundreds or thousands of grains (Roberts et al., 1998a, 1999; Olley et al., 1999, 2004; Adamiec, 2000, 2005; Jacobs et al., 2003, 2006b, 2008c, 2011b; Duller, 2006, 2008b; Feathers et al., 2006, 2010; Porat et al., 2006; Bateman et al., 2007; Demuro et al., 2008, 2013; Arnold and Roberts, 2009, 2011; Tribolo et al., 2010; Arnold et al., 2012, 2013).

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367 There are three main reasons to analyse single grains. First, an individual grain is the smallest fundamental
368 unit of analysis in optical dating (Jacobs and Roberts, 2007; Duller, 2008b), providing detailed information
369 on the behaviour of the grains that comprise the sample of interest. Second, individual grains may respond
370 differently to identical measurement conditions, so a multi-grain aliquot will likely contain some grains with
371 physical properties ideally suited to the SAR procedure and others that are not. As a result, aliquots can be
372 compromised by the presence of ill-suited grains, especially if they dominate the OSL signal. Third, the
373 distribution pattern of single-grain D_e values for a sample offers insights into the extent to which the grains
374 were uniformly sun-bleached before deposition, and the legacy of subsequent disturbance by humans
375 (anthroturbation), soil biota and other factors that can jeopardise the stratigraphic integrity of a deposit.

376

377 If grains were adequately bleached by sunlight before deposition and remained undisturbed thereafter,
378 then multi-grain aliquots may yield accurate ages – provided the aliquots are not contaminated by grains
379 with aberrant physical properties. But many archaeological sites are complex depositional environments
380 that demand single-grain analyses to construct robust chronologies. Indeed, archaeological questions have
381 motivated many of the key developments in single-grain optical dating (Roberts et al., 1997, 1998a, 1998b,
382 1999; Henshilwood et al., 2002; Jacobs et al., 2003, 2006b, 2008c, 2013).

383

384 *3.1 Discarding the junk*

385

386 To obtain accurate ages, it is important to first weed out grains that are unsuitable for D_e determination.
387 These grains can be identified and discarded before age estimation using a series of objective ‘rejection
388 criteria’ as internal quality-assurance checks of sample suitability (Galbraith et al., 1999; Yoshida et al.,
389 2000; Duller, 2003; Jacobs et al., 2003; Thomsen et al., 2005). The standard set of criteria used for quartz
390 (Jacobs et al., 2006a) includes the rejection of grains with OSL signals that are too dim to distinguish from
391 background; grains that fail the recycling ratio test or the recuperation test; grains with natural signals that
392 are saturated or do not intercept the dose-response curve; and grains that respond to infrared stimulation
393 (Duller, 2003). The latter observation is generally attributed to feldspar inclusions, but some quartz grains
394 are sensitive to infrared photons at room temperature (Godfrey-Smith and Cada 1996; Jacobs et al.,
395 submitted). These five criteria are applied routinely to all samples, with the addition of further criteria to
396 reject problematic grains in specific samples. Duller (2012) found that application of the fast ratio was
397 advantageous for grains with OSL signals close to saturation, while Gliganic et al. (2012) introduced two
398 additional rejection criteria – based on the D_0 value and the degree of sensitivity change of individual grains
399 – to deal with the same issue.

400

401 It is not unusual to reject 80% or more of the measured quartz grains using the standard criteria, although
402 the proportion of rejected grains varies from sample to sample. In our experience, grains are most often
403 discarded because their OSL signals are obscured by the photon counts from scattered stimulation light and
404 instrument noise. To gain further insights into the general behaviour of the grains and help identify any
405 biases introduced, it is common practice to tabulate how many grains were rejected from each sample and
406 for what reasons. Jacobs et al. (2006a) tested the efficacy of applying the rejection criteria to two sets of
407 quartz grains (from Sibudu Cave and Blombos Cave in South Africa), using both controlled laboratory
408 experiments and natural samples. They showed that the rejection of aberrant grains did not bias or
409 adversely affect the resulting D_e distributions, but that if these criteria were not applied then the degree of
410 scatter in D_e values might be misinterpreted as resulting from external factors influencing the sample
411 during its burial history. These findings have since been corroborated by further tests of single-grain dating
412 procedures at Blombos Cave (Jacobs et al., 2013) and other archaeological sites, such as the recent studies
413 of Douka et al. (2014), Jacobs et al. (2015, submitted) and Jacobs and Roberts (2015).

414

415 Although the rejection criteria remove most of the ill-suited grains, there invariably remains some scatter in
416 D_e values above and beyond that explained by all known sources of measurement uncertainty – even under
417 controlled laboratory conditions, such as a dose recovery test, where there are no complications from
418 partial bleaching, post-depositional mixing or spatial variation in beta dose rate. This unexplained scatter is
419 known as ‘overdispersion’ (Galbraith et al., 2005; Galbraith and Roberts, 2012). Overdispersion reflects,

among other things, grain-to-grain differences in luminescence sensitivity and behaviour, which are innate in natural minerals (Fig. 1b) due to the idiosyncratic properties of lattice defects within each crystal and their variable responses to heating, bleaching and irradiation.

3.2 *Patterns, context and statistics*

Once grains with aberrant physical properties have been discarded, the remaining grains are then assessed for any external factors that may contribute to the scatter in D_e values – bearing in mind the inherent overdispersion. As each grain may have a unique bleaching and burial history, the measurement of individual grains is particularly advantageous in archaeological contexts where there are concerns about post-depositional mixing (Feathers et al., 2006, 2010; Jacobs et al., 2006b, 2008a; Bateman et al., 2007; David et al., 2007; Armitage et al., 2011), the effects of beta microdosimetry (Jacobs et al., 2008b, 2008c), the possibility of roof spall contamination (Jacobs et al., 2011b) and other forms of inhomogeneous bleaching (Roberts et al., 1998a, 1999; Li, 2001). The distribution pattern of single-grain D_e values for a sample provides information on the degree to which grains were impacted by one or more of these depositional and post-depositional processes, whether the mineral is quartz or feldspar (Lamothe et al., 1994; McFee, 1998; Feathers and Tunnicliffe, 2011; Neudorf et al., 2012; Reimann et al., 2012). To date the event of interest – usually the most recent bleaching event – the D_e values obtained from individual grains must be compared and then combined in a suitable way.

As a first step, the distribution of D_e values should be examined using a histogram or, better still, a ‘radial plot’ (Fig. 2). Radial plots are particularly informative because they enable D_e values to be displayed simultaneously with their precisions and automatically sorted by relative precision (Galbraith, 1988, 2005; Galbraith et al., 1999). Other important features can also be assessed at a glance, including any scatter among the D_e values exceeding that expected from the measurement errors (overdispersion) and any patterns in the distribution, such as the existence of discrete D_e components. Galbraith (2010) and Galbraith and Roberts (2012) review some of the most popular approaches to plotting D_e values.

Next, the D_e values for the population of grains related most closely to the event of interest need to be combined appropriately. A variety of well-established statistical ‘age’ models developed originally for fission track analysis (Galbraith, 2005) are available for this purpose (Galbraith et al., 1999; Roberts et al., 2000; Galbraith and Roberts, 2012). These frequentist models are more often applied to D_e values than to optical ages per se. This is the case for single grains of quartz because the environmental dose rate specific to each grain is not measured (see Section 4). For feldspars, these models are typically applied to the optical ages to account for different rates of anomalous fading, but D_e values for single grains of feldspar can be modelled directly if they are measured using the non-fading pIRIR signals.

No age model should be used as a ‘black box’, judged exclusively on statistical criteria or applied in isolation from the depositional context of the sample and the distribution pattern of D_e values. Rather, the choice of age model should take into account the likely bleaching and burial history of the grains, stratigraphic considerations, independent age control and any other relevant information (Galbraith, 2005; Galbraith et al., 2005). Bayesian methods have potential in this regard – the key being to incorporate appropriate ‘prior’ information in the model – and have been used previously to evaluate D_e distributions (Sivia et al., 2004; Greilich et al., 2006; Cunningham and Wallinga, 2012; Cunningham et al., 2015). When working on statistical problems such as these, it is advisable to collaborate with professional statisticians (Galbraith, 2010, 2015; Galbraith and Roberts, 2012).

So, how do we interpret the distribution patterns of D_e values and how does this inform our choice of model? The ideal situation is when grains are fully bleached by sunlight at the time of deposition and not mixed subsequently. Fig. 2a shows the D_e distribution for a ~100 ka sample from Pinnacle Point site 13B in South Africa (Jacobs, 2010). The D_e values are overdispersed by 19%, which is typical of samples that have been well-bleached and then lain undisturbed after burial (Arnold and Roberts, 2009). To combine these D_e values for final age determination, the central age model (CAM) of Galbraith et al. (1999) – or a modified

version for young samples (Arnold et al., 2009) – is commonly used to calculate the weighted mean D_e ; both versions take account of any overdispersion.

INSERT FIG. 2 HERE

In other situations, grains may have been partially or heterogeneously bleached prior to deposition. This is common for fluvial deposits because sunlight is less effective at emptying the OSL traps in grains transported by water – especially if turbid. Alternatively, grains could be transported into a cave in a fully bleached state, and then become mixed with pre-existing older deposits in the darkness of the cave. In both settings, grains with the smallest D_e values are likely to represent those exposed to sufficient sunlight most recently. An example is shown in Fig. 2b for a sample collected from Kudjal Yolgah Cave, a palaeontological site in Western Australia. This D_e distribution is overdispersed by 65% and exhibits the well-defined base line characteristic of partially bleached samples, with a cluster of D_e values between 7 and 10 Gy along the leading edge of a dose continuum extending to 100 Gy. The minimum age model (MAM) described by Galbraith et al. (1999), and by Arnold et al. (2009) for young sediments, is suitable for combining the individual D_e values of such samples; the grey band in Fig. 2b is centred on the MAM estimate of D_e . For partially bleached samples, the MAM should be applied to single-grain D_e values, as multi-grain aliquots may give rise to age overestimates if they contain grains with high residual doses (Olley et al., 1999, 2004).

At some archaeological sites, grains that were well-bleached at deposition can be disturbed after burial, resulting in mixing between different-age layers. With single-grain dating, it is possible to identify the extent of such mixing, and to account for this in the final age determination (Roberts et al., 2001; Jacobs et al., 2006b, 2008a, 2011b; David et al., 2007; Feathers et al., 2010; Armitage et al., 2011). Fig. 2c shows the D_e values obtained for individual grains of quartz from a sample collected near the junction of the uppermost Middle Stone Age layer (35 ka) and the overlying Iron Age layer (1 ka) at Sibudu Cave. The D_e distribution is overdispersed by 174%, but two discrete populations can be visually identified, with few intermediate D_e values. This ‘wheel spoke’ appearance is characteristic of samples composed of distinct populations of grains with very different ages, in contrast to the continuous spectrum of D_e values obtained from samples that suffer from partial bleaching (Fig. 2b) and those that have suffered from significant anholoturbation or wholesale mixing (Fig. 2d). The smaller the difference in D_e between the populations of grains mixed together, the harder it is to statistically separate them. The finite mixture model (FMM) of Roberts et al. (2000) can be used to objectively identify discrete D_e components in a mixture, and then combine the individual D_e values for each component. Worked examples are provided by David et al. (2007) and Jacobs et al. (2008a, 2011b). The FMM can only be applied to single-grain D_e distributions; multi-grain aliquots can result in the creation of ‘phantom’ populations that are meaningless as regards the depositional age of the sediments (Arnold and Roberts, 2009; Arnold et al., 2012).

Distinguishing between samples that contain partially bleached grains and those composed of well-bleached grains that were mixed after burial is not always straightforward. In such cases, sample context is a particularly critical element in the decision-making process (Galbraith and Roberts, 2012). In our experience, partial bleaching of sediments is rarely a concern at human occupation sites in caves and rock shelters or at many open-air settings. But the possibility of insufficient bleaching of grains should always be borne in mind, especially when dating archaeological sites located on floodplains and in other depositional environments where the sediments may not have been fully bleached at deposition. Holocene sediments deposited by water frequently exhibit a clustering of data points near the leading edge of the D_e distribution, with a decrease in density at higher doses (Olley et al., 2004; Arnold et al., 2009). There may be occasions when well-bleached sediments are buried and then intruded by older grains, generating D_e distributions that are similar in appearance to those of partially bleached samples. If the majority of grains happen to cluster near the lower edge of the D_e distribution, then the MAM and FMM will give similar results, but any contextual information about processes of site formation should always be taken into consideration.

It is sometimes not feasible to date the archaeological event of interest using single grains, multi-grain aliquots or any of the existing statistical models. Mixtures of partially bleached sediments and deposits that have been massively disturbed by anthropurbation or other processes (Bateman et al., 2007; Tribolo et al., 2010; Chazan et al., 2013) fall into this category; serious concerns must exist about the stratigraphic integrity of archaeological materials found therein. Such samples pose the same challenges to any method that infers the antiquity of artefacts and human remains by association, rather than by direct dating. But single-grain measurements are still useful as particle tracers: they can confirm or refute the stratigraphic integrity of a deposit, and thus play an important role in elucidating the post-depositional processes operating within a site and the broader landscape (Heimsath et al., 2002; Bateman et al., 2007). Fig. 2d shows the single-grain D_e distribution for a sample from Klipfonteinrand in South Africa. The D_e values are scattered randomly between 10 and 150 Gy, with no obvious clusters. As with Fig. 2b, the distribution has a solid base line and a similar degree of overdispersion (70%). Although the D_e values are not concentrated along the leading edge of the distribution, it could plausibly be interpreted as a partially bleached sample with relatively few fully bleached grains. By combining these OSL data with our contextual knowledge of the deposit, however, the spread in D_e values can confidently be attributed to anthropurbation – the digging of pits by Later Stone Age people into the underlying Middle Stone Age deposits. It is not possible to obtain a meaningful optical age from such a D_e distribution, but for other parts of this site we obtained distributions with discrete D_e components, indicating that mixing is localised. Single-grain analysis can, thus, provide insights into the stratigraphic fidelity of archaeological deposits, as well as their ages.

3.3 Chronological tests of accuracy and precision

Ultimately, comparisons with independently obtained ages are required to validate the accuracy of any dating technique. Several recent compilations of quartz OSL and feldspar pIRIR ages from around the world have been published (Rittenour, 2008; Rhodes, 2011; Buylaert et al., 2012a; Li et al., 2014a), as have comparisons on a site-by-site basis (Vogelsang et al., 2010; Aubry et al., 2012; Clark-Balzan et al., 2012; Dibble et al., 2012; Stewart et al., 2012; Jacobs et al., 2013; Richter et al., 2013). At Wollongong, we routinely check the accuracy of our single-grain techniques, including recent dating studies at five important archaeological sites: Haua Fteah (Douka et al., 2014), Les Cottés (Jacobs et al., 2015) and Pech-de-l'Azé I, II and IV (Jacobs et al., submitted). These sites have been investigated over many years, with detailed chronologies obtained using state-of-the-art methodologies across multiple techniques. The dated deposits collectively span the last ~100 ka and encompass a wide range of site-formation processes.

The ages obtained for these sites using the different methods are presented in Fig. 3, with the single-grain ages shown as red circles. Two important observations can be made. First, the various methods yield statistically consistent ages for any particular archaeological level. The sole exception is the Mousterian level at Les Cottés, which lies at or beyond the effective limit of ^{14}C dating and, hence, shows a divergence between the ^{14}C chronology and the luminescence (OSL and pIRIR) ages. Second, the single-grain ages have measurement uncertainties significantly larger than those of the ^{14}C ages, and this is true also for the pIRIR, TL and ESR ages. This is because OSL ages are calculated from many separate measurements, each of which has a random error and a possible systematic error; random errors can be reduced by averaging several measurements, whereas systematic errors cannot (Aitken, 1985). These errors are propagated through in quadrature (that is, as the square root of the sum of squares) to calculate the total uncertainty, which typically amounts to 5–10% of the age of the sample. Further details on error estimation and propagation are given by Taylor and Kuyatt (1994), Galbraith (2005), Galbraith and Roberts (2012) and Zou (2014).

INSERT FIG. 3 HERE

These three case studies represent typical, albeit complex, archaeological deposits. They illustrate that single-grain dating can produce ages that are both accurate and of useful precision if the sampling strategy, laboratory procedures and statistical analysis of the resulting data are designed and executed well. It is also of paramount importance to consider the broader context of the site and the microstratigraphic details of the deposit in any interpretation of the OSL chronology.

4. Chronologies in context: imaging intact samples

Despite the many archaeological and geomorphological insights gained from the optical dating of individual grains, the fact remains that these grains are analysed after disaggregating the sediment samples during laboratory preparation, resulting in the loss of essential contextual information. That is, mineral grains are extracted from the bulk sample and separated for measurement of the D_e , and the dose rate is determined from portions of the bulk sample that have been homogenised beforehand.

How much more might we learn if single-grain D_e values and grain-specific dose rates were measured *in situ* using intact blocks of sediment that could also be examined for microstratigraphic features and evidence of post-depositional disturbance and diagenesis? Such 'spatially resolved' measurements rank among the current grand challenges and next frontiers for optical dating, but some progress towards achieving these objectives has already been made.

4.1 Measuring and modelling beta dose rates

We mentioned in Section 2.1 that the beta dose rates to individual sand-sized grains of quartz or feldspar can vary considerably within a single sample because the location of radionuclides in sediments is not spatially uniform over distances of a few millimetres (the range of beta particles). The possible effects of such microdosimetry variations on grain-to-grain differences in D_e have been a topic of interest since the advent of single-grain optical dating. Olley et al. (1997) examined the archaeological sediments at Allen's Cave in South Australia and showed that the aeolian fraction (quartz grains with iron-oxide coatings, and clays) had a beta dose rate ~20 times that of the carbonate fragments derived from the surrounding limestone bedrock. They noted that this difference in dose rate was sufficient to explain the spread in single-grain D_e values observed by Murray and Roberts (1997), assuming that the largest and smallest D_e values were from grains surrounded entirely by aeolian sediments and carbonates, respectively.

The Allen's Cave deposit is deficient in heavy minerals (such as zircon and rutile) and potassium feldspar grains, but these and other minerals with high concentrations of uranium, thorium and potassium can act as potential beta dose 'hotspots', leading to grains in their proximity having higher-than-average D_e values; low-radioactivity carbonates will have the opposite effect. Over the past decade, the implications of such microdosimetry variations for interpreting D_e distributions have been explored in a variety of ways (Vandenbergh et al., 2003; Kalchauer et al., 2003; Mayya et al., 2006; David et al., 2007; Jacobs et al., 2008b; Bailiff et al., 2013), including the increasing use of Monte Carlo simulations to model the features expected of single-grain D_e distributions under a range of assumed conditions (Nathan et al., 2003; Chauhan and Singhvi, 2011; Cunningham et al., 2012; Guérin et al., 2012).

For simplicity, these simulations have commonly assumed that the beta dose hotspots are associated solely with randomly scattered grains of potassium feldspar, although the reality is that natural deposits are much more diverse and complex. The Australian sediment sample shown in Fig. 4, for example, contains almost no potassium feldspar; instead, heavy minerals rich in uranium and thorium are distributed randomly among the quartz grains. By contrast, the sample from Pech-de-l'Azé IV (Fig. 5) has a non-random distribution of potassium – the sediment matrix contains the highest concentrations, whereas the limestone, flint and bone have much lower potassium contents. Individual sediment samples vary greatly in the type and abundance of mineral and organic constituents, the extent of diagenesis that they may have undergone, and the existence of microstratigraphic features – all of which can affect the beta dose rates to single grains. These real-life complications impose limits on the value of extrapolating from idealised models in the absence of specific information about the burial environment of a sample, but simulations can provide a useful starting point for developing more realistic models of natural systems. Ultimately, this will require that modelling is combined with measurements of the dose rate for individual samples at the sub-millimetre scale.

INSERT FIG. 4 HERE

Monte Carlo simulations employ radiation-transport codes to predict the dose rate distribution in a virtual sample. The first stage involves constructing a computerised geometry of the sample, with some grains defined as 'sources' and others as 'detectors' – the latter represent grains for which the D_e values are of interest. It is necessary to specify the chemical composition, density, size and packing arrangement of the source and detector grains, together with the assigned radionuclide concentrations and how the radionuclides are distributed throughout the sample; a random distribution is often assumed. Beta particles are then emitted uniformly in all directions from the source grains, and their progress is tracked as they progressively lose energy through interactions with other grains in the virtual sample. By simulating millions of particles, the distribution of D_e values among the detector grains can be estimated.

Nathan et al. (2003) first demonstrated that this approach was feasible for simulated samples, using the MCNP radiation-transport code. Cunningham et al. (2012) used the same code, but with a simplified sample geometry, to run a numerical simulation of the potassium-derived beta dose rate to quartz grains in an experimental mixture. For their 'sand box' experiment, grains of sodium hydroxide were first bombarded with neutrons in a nuclear reactor to produce a short-lived radionuclide (^{24}Na), which has a similar beta energy spectrum to natural ^{40}K . These grains were then mixed with the quartz grains and left to irradiate them for 2 weeks, after which their D_e values were measured. The simulation reproduced the shape of the observed single-grain D_e distribution, but underestimated its spread.

More recently, the Geant4 toolkit (Agostinelli et al., 2003; Allison et al., 2006) has been used to simulate dose rates in sediments, incorporating a wider range of model parameters (Guérin et al., 2012; Martin et al., 2014). With further refinements, these simulations should be able to generate beta dose rate distributions for real sediment samples, if the concentrations of the relevant radionuclides and their spatial distributions can be incorporated as model parameters. At present, such data are assumed and not measured for each sample, so true single-grain ages cannot be calculated. To do so requires knowledge of the dose rate specific to each grain, rather than the overall distribution of dose rates in the sample as a whole, although the latter can shed some light on the distribution pattern of single-grain D_e values.

To achieve the ultimate goal of obtaining optical ages from individual grains, detailed information is needed about the microdosimetry of each sample while still intact. The distribution of radioactivity could then be measured in two – or preferably three – dimensions, and a model constructed based on those data. The current practice of disaggregating samples to extract quartz and feldspar grains for D_e determination sacrifices this spatial information. The dose rates to individual mineral grains can be influenced not only by microstratigraphic structures and diagenetic alterations, but also by their proximity to larger objects such as bones and artefacts, so sample disaggregation also limits the precision with which the dose rate distribution (and, hence, the age distribution) can be determined.

Measurements of the dose rate for intact samples present a series of major technical challenges, however, as most natural sediments have low levels of environmental radioactivity. For samples in which the uranium, thorium and potassium concentrations are sufficiently high, their spatial distribution can be mapped using neutron-induced fission tracks (Wagner et al., 2005; Tribolo et al., 2006), alpha and beta autoradiography (Haustein et al., 2003; Rufer and Preusser, 2009; Schmidt et al., 2013), laser-ablation ICP-MS (Schmidt et al., 2012) and energy dispersive X-ray spectroscopy using an electron beam for excitation (Baril, 2004; Godfrey-Smith et al., 2005; Greilich et al., 2005). For these experiments, rock and slag samples were cut into slices for analysis, while sediments were measured as loose grains, pressed pellets or as grains mounted in resin and then polished. Grainger (2009) and Bailiff et al. (2013) examined spatial variations in dose rate across the cut surfaces of archaeological brick and resin-encapsulated sediment samples using a variety of elemental analysis techniques, alpha autoradiography and Al_2O_3 : C grains, while two-dimensional images of radioactivity have also been generated using Al_2O_3 : C films and other OSL-based dosimetry systems (Akselrod et al., 2000; Ahmed et al., 2014; DeWitt et al., 2014; Yukihiro et al., 2014).

The resulting data from these studies range from qualitative to fully quantitative, and the latter might be used to guide the source geometry for a radiation-transport model to obtain spatially resolved estimates of dose rate at each point on the surface of the rock, brick or sediment sample. Converting from two-

688 dimensional measurements to dose rates in three dimensions requires additional information, which could
689 be obtained by measuring a series of successive slices to reconstruct the spatial distribution of
690 radionuclides throughout the sample.

691
692 In our laboratory, Jankowski (2015) has recently used X-ray computed microtomography (μ CT) to map the
693 three-dimensional distribution of heavy minerals inside a resin-cemented sediment sample that is
694 otherwise predominantly comprised of sand-sized quartz grains (Jankowski et al., 2015). The heavy
695 minerals can clearly be seen in thin section (Fig. 4a) and on the μ CT scan (Fig. 4b and 4c). Such minerals can
696 be highly radioactive. Jankowski et al. (2015) separated them from the other grains and measured beta
697 dose rates of more than 20 Gy/ka, which is 10 times the total dose rate of the bulk sample. Quartz grains
698 located near or next to these heavy minerals will receive enhanced beta doses, resulting in higher D_e values
699 and possibly increasing the spread of the D_e distribution.

700
701 Jankowski (2015) also examined resin-impregnated sediments from Pech-de-l'Azé IV, using a combination
702 of micromorphology and a portable X-ray fluorescence (pXRF) instrument to investigate the spatial
703 distribution of potassium. The sediment slice in Fig. 5a shows the chaotic nature of the anthropogenic
704 sediments, which contain bones burnt to various degrees, charcoal and ash, and limestone and flint
705 fragments. pXRF measurements were made every 3 mm, and the results are displayed as a contour map in
706 Fig. 5c. Areas with the highest potassium concentrations (red to yellow) are associated with the
707 sedimentary matrix, whereas patches of limestone, flint and bone have lower potassium contents (green to
708 blue). This map illustrates the spatial distribution of potassium, but it represents the average value for the 6
709 mm-diameter zone around each measurement position.

710
711 INSERT FIG. 5 HERE
712

713 A microphotograph (taken in cross-polarised light) of the area bounded by the red square in Fig. 5a is
714 shown in Fig. 5b; the white circle is 6 mm in diameter. The fabric of the deposit is highly variable at this
715 finer scale, too, and Fig. 5d shows the digitally extracted components in the circled area of Fig. 5b. Of
716 particular interest is the yellow coarse sand used for single-grain measurements: these grains are
717 surrounded by a variety of materials that will likely have different radioactivities and, thus, give rise to a
718 range of D_e values for grains deposited at the same time. The extent of this scatter will depend on the
719 relative radioactivities of the different components, which will differ within and between samples. For this
720 particular sample, the potassium concentrations are low and the range is small (mostly 0.20–0.35%), so
721 spatial variations in the potassium-derived beta dose rate should not add significant scatter to the D_e
722 distribution. Elements with low atomic number, such as potassium, can be difficult to measure reliably by
723 pXRF unless strict protocols are followed (Goodale et al., 2012; Johnson, 2014). Instrument calibration using
724 well-characterised and matrix-matched reference standards is essential, as are reproducibility tests of
725 instrument stability over time. Jankowski (2015) used soil and rock standards from the National Institute of
726 Standards and Technology, and several in-house sediment standards measured by laboratory-based XRF.

727
728 An alternative route to spatially resolved dose rates is to measure the radioactive emissions directly from
729 the surface of the sample. This approach co-opts autoradiography technology intended for biomedical
730 research, but dating applications require the detection of radiation at far lower levels, so signal collection is
731 challenging. At Wollongong, we are experimenting with Medipix and Timepix hybrid detectors (Llopart et
732 al., 2007; Ballabriga et al., 2011; Poikela et al., 2014) to provide two-dimensional images of the type and
733 intensity of radiation (alpha particles, photons or electrons) emitted from the sample surface in a 256 x 256
734 grid of 55 μ m square pixels (Fig. 6). Timepix with cadmium telluride semiconductor sensors can also detect
735 the energy level of each photon, enabling pixel-by-pixel identification of the radionuclides and their
736 locations across a surface area of 2 cm². We will also make *in situ* dosimetry measurements at high spatial
737 resolution using special metal oxide semiconductor field-effect transistor (MOSkin) detectors. These have
738 been developed in the Centre for Medical Radiation Physics at Wollongong (Rosenfeld, 2002; Kwan et al.,
739 2008; Metcalfe et al., 2013) and can discriminate between the radiation doses associated with alpha
740 particles and electrons. By placing Timepix and MOSkin devices on resin-impregnated slices of sediment,

our goal is to map spatial variations in radioactivity and to use these data to inform Geant4 simulations of alpha and beta dose rates to individual grains of quartz and feldspar in the same slices.

INSERT FIG. 6 HERE

4.2 *In situ* luminescence measurements

To obtain true single-grain ages requires the combination of these spatially resolved estimates of dose rate with single-grain D_e estimates for the same sediment slices. Instead of comparing the distributions of single-grain D_e values and the modelled dose rates for a sample as a whole – which is as much as can be accomplished from disaggregated sediment samples – paired measurements of D_e and dose rate for each grain would define the true distribution of single-grain ages. By taking account of the spread in D_e values due to spatial heterogeneity in the beta dose rate, this approach should enable any incompletely bleached, intrusive or otherwise anomalous grains to be more easily identified and discarded from the data set, thereby increasing both the accuracy and precision of the calculated optical ages.

Spatially resolved luminescence measurements of natural minerals have been in development since the 1970s. Initially, image intensifiers and imaging photon detectors were used to measure the TL emissions (Walton and Debenham, 1980; Smith et al., 1991; McFee and Tite, 1994; McFee, 1998). More recent systems have used highly sensitive charge-coupled devices (CCDs) to detect the TL, OSL and IRSL signals. CCDs consist of pixel arrays that give a digital image of the photon counts from each of the stimulated grains. Duller et al. (1997) developed the first such system for natural materials, and several custom-made instruments have since followed (Spooner, 2000; Baril, 2004; Greulich and Wagner 2006; McCulloch et al., 2011; Clark-Balzan and Schwenninger, 2012; DeWitt et al., 2014; Mundupuzhakal et al., 2014). Bailiff and Mikhailik (2003) took a different approach and developed an OSL scanning system, which has been applied to archaeological brick and sediment samples (Bailiff, 2006; Bailiff et al., 2013). Bailiff (2006) scanned the surface of two brick slices and, encouragingly, obtained *in situ* D_e values for individual quartz grains that compared well with those obtained for grains extracted from the disaggregated samples.

Electron-multiplying CCD systems are now available or in development by the two main commercial manufacturers of luminescence dating equipment (DTU Nutech and Freiberg Instruments), so we expect an expansion of research into two-dimensional luminescence imaging over the next few years. The digital images are several millimetres in diameter – comparable to the *in situ* dosimetry measurements – but their conversion into spatially resolved estimates of D_e is not straightforward. The chief difficulties for CCD systems concern the trade-off between sensitivity and resolution, and the software challenges presented by the huge data sets collected and their statistical analysis. Efforts to solve these technical and computational problems began more than a decade ago (Greulich et al., 2002, 2006).

If reliable D_e estimates for *in situ* grains can be combined with the grain-specific dose rates, then grain-by-grain dating of intact sediments and stone surfaces of archaeological interest (Greulich et al., 2005; Liritzis, 2011; Chapot et al., 2012) may finally become a reality. The integration of spatially resolved optical ages with microscopic and molecular analyses of the same samples would greatly enlarge our knowledge of the formation and post-depositional processes operating at archaeological sites. The costs of pursuing such an undertaking are substantial, as are the challenges faced, but it is a goal worth striving for.

5. Shining light deep into the Pleistocene

In their landmark study, Huntley et al. (1985) showed an increase in OSL intensity with depositional age for quartz grains from a sequence of stranded beach dunes ranging in age from modern to more than 700 ka. Optical dating has since been applied mainly to deposits younger than ~200 ka because of the saturation limit of quartz OSL (making it difficult to date older events unless the environmental dose rate is unusually low) and the anomalous fading of feldspar IRSL (for which reliable correction methods are available only for samples younger than ~50 ka). So optical dating currently covers most, if not all, of the time span of *Homo*

794 *sapiens* and contemporary hominins, but illuminating the earlier annals of human existence remains one of
795 the great challenges for the luminescence dating community.

796

797 Attempts to extend the time range of optical dating have exploited the fact that quartz and feldspar each
798 contain a variety of traps that are light-sensitive (to differing extents) and that saturate at different doses.
799 This opens the possibility of targeting those traps that saturate at the highest doses, and here we describe
800 some past and present efforts to probe these traps to develop reliable long-range chronometers.

801

802 5.1 Quartz

803

804 One suggestion to extend the optical dating limit of quartz arose from the observation (Roberts et al., 1999)
805 that the OSL dose-response curves of individual grains can vary considerably in their shape: some saturate
806 at doses of 50 Gy or less, while others continue to grow linearly to doses of more than 200 Gy. The latter
807 grains were dubbed 'supergrains' by Yoshida et al. (2000), who measured D_e values in excess of 400 Gy for
808 grains from the same Australian dune sequence as that studied by Huntley et al. (1985). This inherent
809 variability in OSL characteristics among single grains, and the ability of some to record exceptionally high D_e
810 values, has also been reported for other samples (Duller et al., 2000; Jacobs et al., 2003; Arnold and
811 Roberts, 2011). Ages of up to 700 ka have been obtained from supergrains (Yoshida et al., 2000), but such
812 grains are in the minority, so targeting them for dating requires that many thousands of grains are
813 measured, which can consume precious instrument time.

814

815 The slow component of quartz OSL offers another potential long-range dating signal. Several slow
816 components have been identified in quartz, one of which (component S3 of Singarayer and Bailey, 2003,
817 2004) is associated with traps that saturate at much higher doses than those responsible for the fast
818 component (Bailey, 2000; Bulur et al., 2000; Singarayer et al., 2000). For dune deposits in Morocco
819 containing Acheulian stone artefacts, Rhodes et al. (2006) obtained a D_e value of 700 Gy and an age of
820 almost 1 million years (Ma) from the S3 component. This signal has not been widely adopted for dating,
821 however, due to the need to separate it from the other OSL components and because the S3 traps are hard
822 to bleach, so they may not have been emptied at the time of sediment deposition. But this feature can be
823 utilised to provide an internal check on the extent of sediment bleaching: if the same D_e is obtained for the
824 S3 and fast components, then the grains must have been exposed to sufficient sunlight to empty even the
825 S3 traps (Singarayer et al., 2005).

826

827 Optically sensitive TL signals have also been explored as a means of increasing the time depth of quartz.
828 Jain et al. (2005, 2007) investigated the use of isothermal TL – the phosphorescence observed while holding
829 grains at a high temperature for an extended period (e.g., 500 s at 310°C) to evict electrons from the
830 bleachable traps. They found that the ultraviolet isothermal TL signal saturated at doses 10-fold larger than
831 the conventional OSL signal, whereas Huot et al. (2006) reported that sensitivity changes caused by the
832 initial measurement of the natural signal resulted in overestimates of the expected D_e for a variety of
833 sedimentary samples and in SAR dose recovery tests. Such complications continue to hinder the
834 development of a reliable isothermal TL dating procedure.

835

836 The red TL emissions from volcanically and archaeologically heated quartz grains have also been examined
837 for their long-range dating potential, after it was discovered that they saturate at high doses (Hashimoto et
838 al., 1986, 1987; Fattahi and Stokes, 2003a; Hashimoto, 2008). Fattahi and Stokes (2000) obtained a D_e of
839 ~2200 Gy and an age of 1.3 Ma for heated quartz from a New Zealand ignimbrite, but the optically sensitive
840 red TL signals appear to saturate at much lower doses and are harder to bleach than the fast OSL
841 component (Fattahi and Stokes, 2005; Lai and Murray, 2006; Westaway and Roberts, 2006).

842

843 More recently, the TT-OSL (Wang et al., 2006a, 2006b, 2007) and VSL (Jain, 2009; Ankjærsgaard et al., 2013)
844 signals from quartz have been tested as prospective Middle and Early Pleistocene timekeepers. The TT-OSL
845 signal is measured by first emptying the OSL traps and then heating the sample at an elevated temperature
846 to thermally transfer electrons from deep traps into the vacant OSL traps. For silt-sized grains of Chinese
847 quartz, Wang et al. (2006b) found that the TT-OSL signal continued to grow to many thousands of Gy,

848 enabling them to date back almost 800 ka (Wang et al., 2006a). Pickering et al. (2013) used TT-OSL to date
849 Early Pleistocene (1.0–1.1 Ma) cave deposits on the southern coast of South Africa, while Middle
850 Pleistocene TT-OSL ages have been obtained for other sites of archaeological and palaeoanthropological
851 interest (Arnold et al., 2013, 2014; Sun et al., 2013; Demuro et al., 2014; Hernandez et al., 2014). The traps
852 that give rise to the TT-OSL signal take weeks to months to be bleached by sunlight, and the measurement
853 procedures must be fine-tuned for each group of samples to obtain reliable ages (Jacobs et al., 2011a;
854 Rosenberg et al., 2011; Duller and Wintle, 2012), so more comparisons with known-age samples are
855 needed to validate the TT-OSL signal as a long-range chronometer.

856
857 VSL refers to the ultraviolet emissions from quartz when illuminated by violet photons, following an initial
858 bleach with blue light to empty the more light-sensitive OSL traps (Jain, 2009). VSL originates from deep
859 traps that are hard to bleach, but the initial part of the signal (component A) can grow to doses of ~6400 Gy
860 in the laboratory, which is 20 times higher than the saturation limit of the fast OSL component
861 (Ankjærsgaard et al., 2013). The exact relation, if any, between VSL and the other luminescence signals from
862 quartz that can extend to high doses has yet to be elucidated. Satisfactory agreement between OSL and VSL
863 (component A) ages of 60–330 ka was obtained for field samples with D_e values of up to 200 Gy
864 (Ankjærsgaard et al., 2013), but it remains to be seen at this early stage if the VSL signal is applicable to older
865 samples. In a recent study, six ancient soils in northern Israel yielded VSL ages ~60% younger than the
866 $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 0.7–1.5 Ma for the interstratified basalt flows (Ankjærsgaard et al., 2014).

867
868 These various long-range signals hold great promise for dating of Middle, and perhaps Early, Pleistocene
869 deposits. But it should be remembered that their light-sensitive traps are much more difficult to bleach
870 than those that give rise to the fast component of quartz OSL, so they are best restricted to sediments that
871 were fully bleached at deposition and are sufficiently old that any residual dose is a small fraction of the D_e .
872 Also, the performance of these signals can vary greatly from sample to sample, so further tests are required
873 on deposits with secure, independent age control to check the reliability of these experimental methods.

874 875 5.2 Feldspar

876
877 The luminescence intensity of feldspar is usually much greater than that of quartz and saturates at far
878 larger doses, so feldspars should be ideally suited to highly reproducible and precise measurements of D_e ,
879 especially for older samples. In addition, the high internal beta dose rate from the radioactive decay of ^{40}K
880 and ^{87}Rb inside sand-sized grains of potassium-rich feldspar reduces the effect of spatial and temporal
881 variations in the external dose rate (from beta, gamma and cosmic radiation, including any changes in
882 water content) on the total dose rate – and, hence, on the calculated age.

883
884 We mentioned in Section 2.1 that the isochron IRSL technique of Li et al. (2008a, 2008b) can eliminate the
885 uncertainties associated with the external contributions to the environmental dose rate for samples that
886 contain well-bleached and coeval potassium feldspar grains in a range of sizes. Li et al. (2008b) proposed
887 that the adverse effects of anomalous fading could also be avoided using this procedure, but they could
888 only speculate on a possible mechanism. Future improvements to the isochron technique could be
889 achieved by measuring D_e values using the non-fading pIRIR signals to simultaneously overcome the
890 problem of anomalous fading and any dose rate complications related to changes in water content or
891 radionuclide migration.

892
893 Although ultraviolet–blue IRSL emissions are normally used for optical dating of feldspars, many feldspars
894 also emit far-red TL which is not affected by fading (Zink and Visocekas, 1997; Fattahi and Stokes, 2003a;
895 Visocekas and Guérin, 2006). These TL traps are bleachable, but are emptied more slowly than the IRSL
896 traps and precautions must be taken to suppress the red glow from incandescence. To empty the optical
897 dating traps directly without interference from incandescence, infrared photons have been used to
898 stimulate far-red IRSL emissions (Fattahi and Stokes, 2003b, 2004; Lai et al., 2003). These traps saturate at
899 doses in excess of 2000 Gy, appear not to fade and the most light-sensitive component is bleached rapidly
900 (Stokes and Fattahi, 2003; Fattahi et al., 2004). This opens the door to dating feldspars deposited in the
901 Middle Pleistocene, and possibly the Early Pleistocene, without the need for a fading correction. The main

drawback is the need to use customised apparatus to detect the typically weak far-red signals – namely, an extended-range photomultiplier tube cooled to -20°C and fitted with appropriate optical filters.

Other feldspar signals have also been suggested to exhibit low to negligible rates of fading. These include time-resolved OSL (TR-OSL), which is the luminescence observed during and between short pulses of stimulation using visible or infrared photons (Sanderson and Clark, 1994; Tsukamoto et al., 2006; Jain and Ankjærgaard, 2011). Pulsed stimulation also offers a means of separating the quartz and feldspar OSL signals for samples that contain both minerals, as the TR-OSL decay of quartz is much slower than that of feldspar (Bailiff, 2000; Chithambo and Galloway, 2000; Denby et al., 2006; Ankjærgaard et al., 2010). Infrared radiofluorescence (IR-RF) dating of potassium feldspar has also been investigated as a means of circumventing anomalous fading (Trautmann et al., 1998, 1999a). IR-RF is the infrared luminescence emitted while applying an ionising radiation dose in the laboratory. The signal decays in intensity as a function of laboratory dose, and can be detected from individual feldspar grains (Trautmann et al., 2000). Good agreements between IR-RF and independent ages have been reported (Trautmann et al., 1999b; Erfurt et al., 2003; Novothny et al., 2010; Wagner et al., 2010), as well as disagreements (Buylaert et al., 2012b). IR-RF emissions have rarely been used for dating of archaeological or palaeoanthropological deposits, a notable exception being the type locality of *Homo heidelbergensis* in Germany, for which ages of ~ 600 ka were obtained (Wagner et al., 2010). Radiofluorescence is also emitted by quartz, but the ultraviolet to red signals have not yet been used to date archaeological events (Schmidt et al., 2015).

The latest efforts to overcome fading in feldspars involve the ultraviolet–blue pIRIR emissions from deep traps. It has long been known that the TL emitted by sodium feldspars at high temperatures (600 – 650°C) is not affected by fading (Valladas and Valladas, 1979; Guérin and Valladas, 1980; Guérin, 2006), and Jain and Singhvi (2001) found that infrared bleaching of feldspars at 220°C left a remnant population of electrons in deeper, more stable traps, which could be evicted using infrared stimulation at a higher temperature. This raised the prospect of isolating a non-fading IRSL component using a two-step procedure to induce the pIRIR emissions (Thomsen et al., 2008; Buylaert et al., 2009).

Grains are first bleached by infrared photons at a low temperature (50°C) to empty the shallow traps, which fade most, and the deeper traps are then stimulated at a raised temperature – originally 225°C , but 290°C has since been widely adopted to preferentially empty traps that suffer less from fading (Thiel et al., 2011; Buylaert et al., 2012a). The pIRIR traps do not bleach as rapidly as the conventional IRSL traps or those responsible for the fast component of quartz OSL, so an accurate estimate of the residual dose at deposition is critical for Holocene and Late Pleistocene feldspar samples to avoid age overestimation. For older samples, the need to correct for slight fading of the pIRIR traps is more important.

Fig. 7a is a recent compilation of pIRIR (290°C) and independent age estimates for 116 sediment samples (Li et al., 2014a). Thirty of the 41 samples older than 100 ka have pIRIR ages consistent with the independent estimates at 2σ , although the scatter is large. We caution readers that most comparisons of luminescence and independent ages are not done ‘blind’ – that is, in the absence of information about the expected age – and this shortcoming is compounded by the common tendency in science for ‘negative’ results not to be published, as well as replication bias (Ioannidis, 2005, 2012; Fanelli, 2012).

INSERT FIG. 7 HERE

Li and Li (2011, 2012) proposed a more elaborate pIRIR procedure to eliminate the need for fading corrections. Their approach involves multiple elevated temperature (MET) stimulations, with a plateau in D_e values at temperatures of 200°C and higher indicating that a non-fading signal has been isolated (Fig. 1c). Fig. 7b shows that the sensitivity-corrected MET-pIRIR (250°C) ages for 38 samples are in excellent agreement with independent ages over the last 300 ka, and 95% of the ages lie within 2σ of the independent estimates. As most of these samples are from northern China, the feldspar grains are likely to have similar luminescence behaviours, so further age comparisons should include samples from other geological provinces to test the wider validity of the MET-pIRIR procedure.

The pIRIR and MET-pIRIR signals commonly saturate at doses of ~1000 Gy (Buylaert et al., 2012a; Li et al., 2014a), which is equivalent to 350 ka at typical environmental dose rates for feldspars. This upper age limit can be extended by a further 50% – to encompass the entire Middle Pleistocene – by exploiting the strong ‘pre-dose’ dependence of the sensitivity of the MET-pIRIR signal, T_x , which can retain a ‘memory’ of the radiation dose received by the grains in nature (Li et al., 2013, 2014b). This signal can be bleached by sunlight, saturates at a much higher dose (~1500 Gy) than the sensitivity-corrected signal (L_x/T_x) and does not require a residual-dose correction. The high-temperature pMET-pIRIR traps also appear to be immune from fading, and ages consistent with independent estimates of up to 500 ka have been obtained from the T_x and L_x signals (Fig. 7b). The latter is measured with higher precision than T_x , but requires a residual-dose correction. Both signals show considerable promise as new long-range chronometers and, when used in tandem, provide an internal cross-check on the reliability of the pMET-pIRIR ages (Li et al., 2014b).

6. Scales of analysis: towards a synthesis

Optical dating did not exist when Richard Klein first became an Editor of this journal. Over the last 30 years, the technique has matured into one of the chronological tools used most widely in Quaternary research, owing to the ubiquitous presence of quartz and feldspar grains in sediments deposited in a variety of environmental settings. In this paper, we have revisited some of the past achievements of optical dating in archaeological contexts, as well as some of the current challenges and future frontiers for the next two decades. Our current ability to measure the D_e values of individual grains extracted from disaggregated samples will, hopefully, be extended to age determinations of grains in their original spatial context – embedded in intact blocks of deposit – before we celebrate the Golden Jubilee of optical dating. And we are optimistic that new luminescence signals will be discovered, and procedures developed, that will facilitate dating of archaeologically relevant materials over the full time span of the Quaternary. By simultaneously expanding and integrating across these different scales of analysis – from the spatially minuscule to the temporally immense – much more of the human past will be illuminated in the future.

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1636

1637 **FIGURE CAPTIONS**

1638

1639 **FIGURE 1: (a)** Natural OSL decay curve and (inset) sensitivity-corrected regenerative-dose growth curve for
 1640 a single grain of quartz stimulated at 125°C using a green laser beam (Jacobs et al., 2012). The D_e for this
 1641 grain is ~137 Gy, estimated by projecting the natural signal (shown on the y-axis) on to the dose-response
 1642 curve. **(b)** OSL decay curves and (inset) dose-response curves for individual grains of quartz from a single
 1643 sample, illustrating the diversity of growth curve shapes for grains extracted from the same sample (Jacobs
 1644 et al., 2015). **(c)** Natural IRSL (50°C) and MET-pIRIR (100–250°C) decay curves for a single (multi-grain)
 1645 aliquot of potassium feldspar stimulated successively in 50°C steps using infrared LEDs (Li and Li, 2011). The
 1646 sensitivity-corrected dose-response curves (x-axis: dose in Gy; y-axis: sensitivity-corrected IRSL) and
 1647 corresponding D_e values are shown above each decay curve. Note the slower rate of luminescence decay
 1648 and the increase in D_e as the measurement temperature is raised, due to stimulation of progressively
 1649 deeper traps that are less light-sensitive but fade less.

1650

1651 **FIGURE 2:** D_e values displayed as radial plots for **(a)** a well-bleached sample, **(b)** a partially bleached sample,
 1652 **(c)** a mixed sample that can be resolved into two discrete D_e components, and **(d)** a mixed sample for which
 1653 a single archaeological event cannot be determined. Each of the filled circles denotes the D_e value and
 1654 relative standard error for a single grain. The D_e can be read by drawing a line from the zero-point on the
 1655 ‘Standardised Estimate’ axis (on the left-hand side), through the data point of interest, to intersect the
 1656 radial axis on the right-hand side. The point of intersection is the D_e . The relative standard error for this
 1657 estimate is read by drawing a vertical line from the same data point to intersect the horizontal axis at the
 1658 bottom. The ‘Relative Error’ (in %) is calculated as the standard error divided by the D_e and multiplied by
 1659 100, and the ‘Precision’ is the reciprocal of the relative error. Also indicated are the number of single-grain
 1660 D_e values in each plot (n) and the overdispersion (OD) value for each D_e distribution.

1661

1662 **FIGURE 3:** Multi-method age comparisons for three sites: **(a)** Haua Fteah, Libya (Douka et al., 2014), **(b)** Les
 1663 Cottés, France (Jacobs et al. (2015)), and **(c)** Pech-de-l’Azé IV, France, where MTA denotes the Mousterian of
 1664 Acheulian Tradition (Jacobs et al., submitted). The ages obtained from OSL dating of single grains of quartz
 1665 (SG-OSL) are shown as filled red circles in each plot. Also shown are post-infrared IRSL (pIRIR) ages for
 1666 potassium feldspars at Haua Fteah and Les Cottés, thermoluminescence (TL) ages for burnt flints and
 1667 electron spin resonance (ESR) ages for tooth enamel at Pech-de-l’Azé IV (EU, early uranium uptake; LU,
 1668 linear uranium uptake), closed system uranium-series/ESR (CSUS-ESR) ages for tooth enamel and
 1669 independently determined ages for three tephra at Haua Fteah, and radiocarbon (^{14}C) ages for bone (Les
 1670 Cottés and Pech-de-l’Azé IV) or charcoal, charred plants and terrestrial snail shells (Haua Fteah).

1671

1672 **FIGURE 4:** Resin-impregnated sample of quartz-dominated sand with scattered heavy minerals, from
 1673 MacCauley’s Beach, Australia (Jankowski et al., 2015). **(a)** Microphotograph of a thin-section under plain
 1674 polarised light. The white and translucent grains are quartz, and the dark grains are heavy minerals (zircon,
 1675 rutile, tourmaline and ilmenite). **(b)** X-ray computed microtomography (μCT) image of the same sample,
 1676 but over a larger field of view (144 mm²). White areas indicate heavy minerals. Measurements were made
 1677 at the Australian Centre for Microscopy and Microanalysis at the University of Sydney, using an Xradia

1678 MicroXCT-400. Experimental details are given in Jankowski (2015). **(c)** Same image as (b), but with the
1679 heavy minerals (indicated in black) extracted using ImageJ software.

1680

1681 **FIGURE 5:** Resin-impregnated sediment sample from Layer 8 at Pech-de-l’Azé IV, France (Jankowski, 2015).
1682 **(a)** Scan of thin-section showing different constituents: sediment matrix, limestone, flint, ash, charcoal, and
1683 bone fragments. The latter have been burnt to various extents (raw, browned, blackened and calcined). The
1684 superimposed grid is 3×3 mm. **(b)** Microphotograph taken in cross-polarised light of the portion of thin-
1685 section bounded by the red square in (a). The white circle (6 mm in diameter) is equivalent to the area
1686 analysed during pXRF measurements. **(c)** Contour map of potassium concentrations (in %) for the sediment
1687 block from which the thin section in (a) was manufactured, overlain with the same grid. Concentrations
1688 were measured at the corners of the each grid square using a pXRF analyser (Niton XL3t 950 GOLDD+). ‘Hot’
1689 colours indicate areas of relatively high potassium content (sedimentary matrix) and ‘cool’ colours denote
1690 areas with less potassium (limestone, flint and bone). (a) and (c) illustrate the non-random distribution of
1691 potassium over distances of a few millimetres. **(d)** Image of the components inside the white circle in (b),
1692 digitally extracted using JMicroVision software. Five groups of materials are distinguished: bone (black),
1693 coarse sand (yellow), fine sand (pink), matrix (teal) and void space (red). (b) and (d) illustrate that the
1694 coarse sand fraction isolated for single-grain OSL dating is juxtaposed by a variety of materials at the sub-
1695 millimetre scale.

1696

1697 **FIGURE 6:** Photograph of the **(a)** Medipix chip and **(b)** schematic design of the Medipix and Timepix hybrid
1698 detectors designed at CERN, the European Organization for Nuclear Research. The electronic chip and
1699 semiconductor sensor are both divided into a 256×256 array of $55 \mu\text{m}$ square pixels, with each of the
1700 pixels in the chip connected via bump bonds to a pixel in the sensor. Images courtesy of CERN.

1701

1702 **FIGURE 7: (a)** Comparison of pIRIR (290°C) ages with independent age estimates for 116 feldspar samples
1703 from Eurasia, Africa and North America ($n = 103, 12$ and 1 respectively) (Li et al., 2014a). **(b)** Equivalent plot
1704 of MET-pIRIR (250°C) ages for 38 samples (blue diamonds) and pMET-pIRIR (250°C) ages for 7 samples
1705 (orange circles and red squares) from Eurasia, with the majority (87%) from northern China (Li et al.,
1706 2014b). The MET-pIRIR ages are based on the sensitivity-corrected (L_x/T_x) signal, whereas the pMET-pIRIR
1707 ages are shown separately for the L_x and T_x signals. In both panels, the solid line indicates the 1:1
1708 relationship and the dashed lines demarcate the $\pm 10\%$ limits.

Table

Main terms (and minerals)	Common acronyms	Definitions and brief descriptions	Synonyms and related phenomena
Optical dating (all minerals)		Umbrella term for all luminescence dating techniques that use photons (rather than heat) to stimulate the electron traps.	Photonic dating
Optically stimulated luminescence (quartz)	OSL CW-OSL LM-OSL TT-OSL	<ul style="list-style-type: none">• Luminescence signal induced by optical stimulation of quartz. Occasionally used for feldspars.• CW prefix indicates continuous wave stimulation, in which the light source remains at a constant power (the standard approach).• LM prefix indicates linearly modulated stimulation, in which the power is steadily increased during the course of optical stimulation.• TT prefix refers to the thermally transferred OSL signal induced from deeper traps in quartz (by heating) than those that give rise to the conventional OSL signal. Sometimes referred to as recuperated OSL (ReOSL).	<ul style="list-style-type: none">• PSL, photon stimulated luminescence: this does not distinguish between visible and invisible (e.g., infrared) stimulating photons.• BGSL, blue/green stimulated luminescence: same as OSL, but specifying the colour of the stimulating photons.• GLSL, green light stimulated luminescence: same as above, but restricted to green photons.• VSL, violet stimulated luminescence: same as above, but using violet photons for optical stimulation.• OSP, optically stimulated phosphorescence: delayed OSL signal measured after optical stimulation has ceased. A form of time-resolved OSL or afterglow.
Infrared stimulated luminescence (feldspar)	IRSL pIRIR MET-pIRIR TR-OSL	<ul style="list-style-type: none">• Luminescence signal induced by infrared photons.• Post-infrared IRSL signal, stimulated at a high temperature after bleaching the sample with infrared photons at a lower temperature to empty the traps affected most by anomalous fading.• MET prefix indicates successive infrared stimulations at multiple elevated temperatures.• TR prefix refers to the time-resolved OSL measured during and between a series of brief pulses of optical stimulation (TR-IRSL for infrared stimulation).	<ul style="list-style-type: none">• IR-RF, infrared-radiofluorescence: emission of infrared photons (rather than stimulation by infrared photons) while an ionising radiation dose is being applied. Also referred to as radioluminescence (RL).• pMET-pIRIR, pre-dose MET-pIRIR procedure: exploits the dose-dependent sensitivity of hole centres, rather than electron traps.• POSL, pulsed OSL: restricted to the OSL measured only in the periods between stimulation pulses. TR-OSL and POSL are also used for quartz.

TABLE 1: Summary of principal terms and common acronyms applied to optical dating of quartz and feldspar. Also listed are some other acronyms that occasionally appear in the literature for the same or related luminescence phenomena.

Figure

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Figure**a.** **b.** **c.**

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