Geochronology Beyond Radiocarbon: Optically Stimulated Luminescence Dating of Palaeoenvironments and Archaeological Sites

Constantin D. Athanassas¹ and Günther A. Wagner²

1 Centre de Recherche & d’Enseignement de Géosciences de l’Environnement CEREGE, Européole Méditerranéen de l’Arbois, Aix-en-Provence, France
E-mail: athanassas@cerige.fr
2 Geographisches Institut der Universität Heidelberg, Germany
E-mail: gawag-wagner@web.de

This article reviews optically stimulated luminescence (OSL) dating as used on Quaternary sediments and for archaeological dating. The underlying physics is summarized and the laboratory method itself is described. Examples of OSL dating illustrate its use in palaeoenvironmental and archaeological contexts, although problems associated with the technique are also addressed. Finally, we discuss long-range variants of OSL that may help date deposits currently considered too old for OSL to be applied.

Keywords: long-range luminescence dating, sediments, Quaternary geology, anthropogenic deposits

INTRODUCTION

The last few decades have seen a significant impact of geoscience techniques and concepts on palaeoenvironmental and (geo)archaeological studies. Geoarchaeology and palaeoenvironmental studies both require a solid chronological framework by which to recognize causes and effects, as well as rates of processes. For this purpose, in addition to archaeological and geomorphological reasoning, radiocarbon dating (¹⁴C) is used to date organic materials. But in view of its ineffectiveness to date materials older than 50 ka and the omnipresence of (inorganic) clastic sediments in most depositional contexts, optically stimulated luminescence (OSL) dating and its long-range dating alternatives are indispensable.

Optically stimulated luminescence was pioneered in the 1980s by Huntley et al. (1985) as the successor to thermoluminescence (TL). Rapid signal resetting of OSL under direct or indirect sunlight (bleaching) rendered OSL pivotal in sediment dating because it can date the time elapsed since the last exposure of natural minerals to daylight. It soon became evident that OSL could be applicable to dating numerous types of sedimentary environments—aerolian, coastal, fluvial, glacial, and lacustrine—thereby enhancing our understanding of Earth’s surface processes (Rhodes 2011; Roberts et al. 2015) and of human evolution (Jacobs et al. 2008; Wagner et al. 2010).

Recent luminescence methods are collectively known as long-range dating variants, and these include thermally transferred optically stimulated luminescence (TT-OSL), post-infrared infrared stimulated luminescence (pIR-IRSL), multi-elevated temperature post-infrared infrared stimulated luminescence (MET-pIR-IRSL), violet stimulated luminescence (VSL), and infrared-radiofluorescence (IR-RF). These new methods have allowed chronometric dating on sedimentary deposits that goes beyond the usual distal age limit of conventional OSL dating, which is estimated to be about 200,000 years (Rhodes 2011; Roberts et al. 2015).

This paper aims to inform geoscientists, archaeologists, and geoarchaeologists about the physical background, methodology, and applications of optically stimulated luminescence dating. The case studies illustrated here are mainly drawn from the authors’ own work and provide applications of OSL dating and its long-range variants to best advantage, as well as giving examples that challenge their performance.

FUNDAMENTALS OF OSL DATING: TECHNIQUES AND LIMITING FACTORS

Dating by optically stimulated luminescence builds on the phenomenon that light is emitted from ionized geological minerals when undergoing optical stimulation. Natural ionizing radiation is universal and includes radioactivity—expressed by the emission of α, β, and γ rays from decaying atomic nuclei of ²³⁵U, ²³⁸U, ²³²Th, and ⁹⁰K—and cosmic rays. Natural ionizing radiation excites the mineral to a prolonged excitation state in which electrons accumulate in lattice defects inherent to the mineral; the longer the ionization over geological time the larger the amount of trapped charge.

An OSL age is the ratio of the radiation dose to the rate at which it was absorbed. Sophisticated instruments known as “luminescence readers” can reset excited minerals to the ground state by stimulating them with artificial light and recording their luminescence. Among natural minerals, quartz and feldspar are the most suitable geochronometers to date by OSL: their luminescence is typically stimulated with blue (470 nm) and infrared (870 nm) light, respectively. In the case of feldspar, released luminescence is termed infrared stimulated luminescence (IRSL). However, feldspars frequently undergo loss of trapped electrons at ambient temperatures, a phenomenon known as anomalous fading (Wintle 1973), which, if not corrected, can result in severe age underestimation.

Luminescence intensity tails off with stimulation time and can be graphically represented by a decay curve (Figs. 1A, B). Luminescence intensity is more or less proportional to the dose. The laboratory equivalent of the natural dose, known as the equivalent dose ($D_e$), the SI unit for
which is the gray (symbol Gy; Gy = J·kg⁻¹). The Dc can be evaluated by interpolating the natural OSL signal intensity into a formula, either linear or exponential, that represents the best fit to an empirical plot containing pairs of known doses (regenerated doses) with their associated OSL signal intensities. Thermal treatments are interposed each time a dose is administered to remove electrons from geologically unstable crystal defects. Heating may induce changes to the luminescence efficiency (sensitivity) of the crystal, which should be monitored and corrected. Thus, natural and regenerated signals are normalized against that of a fixed dose (test dose). The resulting plot is termed a “growth curve” (Fig. 1C).

The above method of Dc evaluation is known as the single aliquot regenerated (SAR) dose protocol (Galbraith et al. 1999; Murray and Wintle 2000). The single aliquot regenerated dose protocol for the measurement of a dose is administered to remove electrons from geologically unstable crystal defects. Heating may induce changes to the luminescence efficiency (sensitivity) of the crystal, which should be monitored and corrected. Thus, natural and regenerated signals are normalized against that of a fixed dose (test dose). The resulting plot is termed a “growth curve” (Fig. 1C).

The above method (Dc) evaluation is known as the single aliquot regenerated (SAR) dose protocol (Galbraith et al. 1999; Murray and Wintle 2000). The single aliquot regenerated dose protocol for the measurement of a dose is administered to remove electrons from geologically unstable crystal defects. Heating may induce changes to the luminescence efficiency (sensitivity) of the crystal, which should be monitored and corrected. Thus, natural and regenerated signals are normalized against that of a fixed dose (test dose). The resulting plot is termed a “growth curve” (Fig. 1C).

Estimation of the dose rate involves determining radioelement concentrations either by standard analytical methods, such as neutron activation analysis (NAA) or inductively coupled plasma mass spectrometry (ICPMS), or techniques based on the emission of α, β, and γ rays from the decaying nuclei, such as alpha- and beta-particle counting, or gamma spectroscopy. Radioelement concentrations are then converted to dose-rate units (Gy·ka⁻¹) using conversion factors (Guérin et al. 2011) that deliver the dose rate per radioelement’s concentration (ppm). The dose rate can also be measured in situ by placing dosimeters on individual subsamples (aliquots), which facilitates recording the fluctuations in Dc for the sample.

Precision in OSL dating is a serious concern user. The degree of tolerance to precision is case-specific but, usually, anything less than 10% is satisfactory. Several factors may serve as sources of error. These include intrinsic ones related to the crystalline properties of the material, or extrinsic ones due to factors such as differential bleaching or post-depositional sediment mixing. Aliquots composed of multiple grains with different bleaching and burial histories can have an undesirable impact on the precision of the average Dc. However, such uncertainties do not affect the usefulness of OSL dating in certain applications where precision is not as critical.
LONG-RANGE OSL DATING METHODS

Long-range OSL dating methods have been invented for both quartz and feldspar. These methods take advantage of latent signals than manifest themselves under thermal treatment of quartz (pre-heat) after multielevated temperatures (MET) until a plateau of signals is made certain: pIR-IRSL signals are reset more slowly than IRSL (50°C) and quartz-OSL. Novel variants of luminescence dating can surpass that maximum, pushing its performance limits deeper into the Pleistocene.

While ages as young as a few decades can be estimated, the oldest age limit of OSL dating is controlled by the saturation of luminescence (i.e. limited response to the increasing dose). The rate at which luminescence becomes saturated is defined by the local dose rate: this restricts OSL dating in typical sedimentary environments to less than ~200,000 years (Rhodes 2011; Arnold et al. 2014; Roberts et al. 2015). Novel variants of luminescence dating can enhance Quaternary geochronology.

Alkali-feldspars have the potential for extending age ranges by making use of their radioluminescence (RF) properties. Radioluminescence in alkali-feldspars is emitted in the infrared (IR) region (865 nm) of the spectrum and results from electron transitions in the crystal induced by ionizing radiation. One use of IR-RF was in deciphering the chronology of Homo heidelbergensis in Europe (Wagner et al. 2010).

CASE STUDIES

Dating Palaeogeography

Athanassas et al. (2012) combined OSL and TT-OSL dating to work out a chronological outline for Upper Palaeolithic hominin activity and coastal palaeogeographic changes at Navarino on the Ionian Sea (southwest Peloponnese, Greece). Dating by OSL of archaeological layers that contain assemblages of Upper Palaeolithic chipped stone artifacts bracketed the chronology of hominin activity at Navarino between oxygen isotope stage 5 (OIS-5 at ~120 ka) and oxygen isotope stage 2 (OIS-2 at ~20 ka). Having established a chronological framework for the Mousterian (Neanderthal) layers, Athanassas et al. (2012) went further by correlating the OSL chronology with known Late Pleistocene sea-level stands, enabling a reconstruction of the regional palaeogeography.

Athanassas et al. (2012) demonstrated that the location of the Palaeolithic sites relative to the coast was framed by two palaeogeographic extremes developed during the Last Interglacial (LI = OIS-5) and the Last Glacial Maximum (LGM = OIS-2), respectively. The overall sea-level drop following the LI kept segments of the Ionian shelf exposed until the LGM. Palaeolithic exploration strategies would, therefore, have been affected by the shifting coastal geomorphology, compelling Upper Palaeolithic hominins to acclimatize to new environmental conditions.

Dating Peridesert Loess

The Canary Islands are located off the west coast of Africa in the Atlantic Ocean (Fig. 2) and are affected by the eoclimatic zones of West Africa, namely the Saharan and the Sahelian. The Canary Islands experience dusty winds from the east that derive from the Sahara Desert on a seasonal basis (Fig. 2A). Saharan dust fallout is an important factor in soil development on the Canary Islands (Menéndez et al. 2009).

Terrestrial sedimentary sequences from the Canary Islands are rich in Saharan dust; therefore, they can be used as proxies for the palaeoenvironmental evolution of the Sahara Desert (von Suchodoletz et al. 2010). Although the desertification history of northwest Africa is well documented for the Holocene and the Late Pleistocene (Petit-Maire et al. 1994), it is only intermittently known from the terrestrial records of the Middle and Early Pleistocene (von Suchodoletz et al. 2010). Fortunately, a well-preserved and continuous terrestrial record of “missing-links” from the north-African desertification history exists in the Canary...
Loessoid at Galdár, Gran Canaria. EOSDIS MODIS Rapid Response Team at NASA GSFC. Gran Canaria. A representative loessoid outcrop in Galdár geology is almost all mafic volcanics; its abundance in catchments on Gran Canaria (Menéndez et al. 2009). Component rich in quartz has been found in silted-up (von Suchodoletz et al. 2010). And a strong airborne-dust Occurrences of desert loess have been dated using OSL in Islands, which not only captures the latest parts of the Quaternary but also extends further back to the Pleistocene (Menéndez et al. 2009).

Occurrences of desert loess have been dated using OSL in the easternmost islands of Fuerteventura and Lanzarote (von Suchodoletz et al. 2010). And a strong airborne-dust component rich in quartz has been found in silted-up catchments on Gran Canaria (Menéndez et al. 2009). Because quartz is exotic to the Canary Islands (whose geology is almost all mafic volcanics) its abundance in palaeosols and loess-like deposits (loessoids) signals a Saharan provenance, evoking important chronological and palaeoclimatic implications.

Inspired by the preceding dating studies on the eastern Canary islands (von Suchodoletz et al. 2010), Athanassas embarked on a mission to the Canary Islands to do research using OSL to define the chronology of loess deposits on Gran Canaria. A representative loessoid outcrop in Galdár was identified and sampled (Fig. 2B). The term "loessoid" designates peri-Saharan dust amassed at rates lower than typical loesses (Menéndez et al. 2009). A Middle to Late Pleistocene age had been speculated for this loessoid on the basis of geomorphology and modern dust accumulation rates on Gran Canaria (Menéndez et al. 2009).

Equivalent doses of Saharan quartz from the Galdár deposit were estimated using the standard SAR procedure for quartz by Murray and Wintle (2000). When the SAR dose-estimates were divided with the local dose rate, they returned a Late–Middle Pleistocene age (0.18 Ma) for Saharan dust deposition on northern Gran Canaria (Table 2).

This OSL age is consistent with the beginning of a chronological interval in the palaeoenvironmental history of the Canary Islands during which regional moisture underwent fluctuations between glacial and interglacial periods (von Suchodoletz et al. 2010). Specifically, modulations to the regional atmospheric circulation pattern during glacial stages forced antitrade winds (prevailing winds from the west to the east in mid latitudes) to shift southwards, allowing these anti-trade winds to convey their moisture over areas of North Africa (von Suchodoletz et al. 2010). An additional input of moisture into the mid-Pleistocene regional hydrological systems might relate to the African summer monsoon, brief spells of which affected the archipelago during the same period of time (von Suchodoletz et al. 2010).

**Dating Soil Erosion**

The basin of Phlious on the northern Peloponnese (Greece) has been populated since the Early Neolithic period. When the early farmers cleared the forests, the freshly exposed surface became vulnerable to soil erosion, in particular on the gentle slopes around the basin.

The soil material was washed downwards by the precipitation and was deposited as colluvium at the foot-slope. This process continued as long as agricultural activities went on. Therefore, by dating these sediments one can gain insights into changes of human land use and/or of climatic conditions at the time of their deposition. With OSL one can date the colluvial deposition directly, provided the feldspar and quartz grains were sufficiently bleached during their down-slope transport.

Quartz OSL ages indicate that colluvia were being deposited since the Early Neolithic (7th millennium BC). The age distribution allows calculation of sedimentation rates, which indicate the various cultural periods when land use was intensified (Fig. 3). The sedimentation rate was very low during the pre-Neolithic, but clearly increased at the beginning of the Neolithic. During the Chalcolithic and the Early Bronze Age, it was low again, but increased once more during the Middle and Late Bronze Age (Fuchs et al. 2004). Analogous patterns of colluvial formation since the Neolithic have also been observed at several sites in southwestern Germany (Lang et al. 1999). In all these cases, the alternating rates of sedimentation seem to correlate with cultural and population development.

**Dating Coastal Plain Evolution**

Theodorakopoulou et al. (2012) investigated the coasts of northeastern Crete and found that the coastal plains that bound the Gulf of Mirabello had been influenced by Holocene climatic events. Their multidisciplinary effort – synthesizing quartz-OSL dating and 14C dating of alluvial sediment cores, and micropalaeontological analysis – established a Holocene age for most of the coastal plain. Their research also demonstrated how well-documented climatic events affected the development of the alluvial plain and deteriorated archaeological settlement patterns.

Major defining points of Holocene climatic history were recognized within the local sedimentary record via abrupt changes in the sedimentation rate. Two well-separated events of substantial sediment accumulation were dated by OSL and 14C to around the Holocene Thermal Optimum at ~8–4 ka BP (before present) and the Roman Climatic Optimum at ~2.2–1.5 ka BP. Those events were each succeeded by episodes of no alluvial deposition, which can be correlated to dry-to-cool periods at ~4 ka BP and the Dark Ages Cold Period at ~1.5–1.1 ka BP, respectively. Of all of these episodes, the resurgence of sedimentation during the Roman Climatic Optimum seemed to have had a tremendous impact on the settlement pattern in the form of flash-flooding events, as deduced from sediment cores (Theodorakopoulou et al. 2012). This evidently diminished human habitation on the coastal plain.

**Dating Geomagnetic Reversals**

The Central Mediterranean exhibits archetypical Quaternary marine sequences, many of which are regarded as global stratotypes for the Early and Middle Pleistocene (Papanikolaou et al. 2011). On the island of Zante (or Zakynthos), in the Ionian Sea, there is an uplifted marine
sequence that has been studied by Papanikolaou et al. (2011). These authors used magnetostratigraphy to produce a chronostratigraphic framework for the uplifted sequence (Fig. 4), which captures the last geomagnetic reversal, known as the Brunhes/Matuyama boundary (0.77 Ma), and a subsequent geomagnetic excursion known as “17a” (0.66 Ma).

With the goal of assessing the age-range that can be attained by elevated-temperature IRSL dating methods, Athanassas tested sediments from the Zante marine sequence using the dating protocol of Thiel et al. (2011) against the well-defined magnetostratigraphy of the reference sequence. So far, only a few ages over 300 ka and only a single one over 600 ka have been reported (Arnold et al. 2014; Roberts et al. 2015 and references therein). Therefore, the Zante sequence poses a challenge to the methodology tested. The protocol used by Thiel et al. (2011) measures pIR-IRSL from feldspar at 290°C (pIRIR290), after having erased conventional IRSL at 50°C (TABLE 1). This method was tested on sediments from the Brunhes/Matuyama boundary and the 17a geomagnetic excursion (Fig. 4).

The calculation of accurate pIRIR290 ages is complicated by the difficulty in generating reliable equivalent doses over all of the sequence: equivalent doses appeared to be broadly spread (TABLE 2). It is known that post-infrared IRSL can experience residual doses that cannot be totally removed (Arnold et al. 2014). Intersample variations in hard-to-bleach large residual doses might explain the scatter seen here. Despite this, two samples with fading-corrected equivalent doses brought forth ages that coincide with the Brunhes/Matuyama (B/M) and 17a events at 0.72 ± 0.14 Ma and 0.62 ± 0.18 Ma, respectively (TABLE 2). This suggests that if low residual doses can be ensured, then pIR-IRSL can be exceptionally successful.

CONCLUSIONS

Palaeoenvironments influence major human evolutionary and cultural shifts. Optically stimulated luminescence (OSL) dating can link major environmental events with human cultural changes on timescales greater than 14C dating can manage. Sharp signal resetting in most sunlit environments has established OSL as the dominant technique in dating sedimentary deposits associated with the Earth’s surface and with anthropogenic processes during the last glacial-interglacial cycle. Even though the dose rate may confine conventional OSL dating to the Late Quaternary, novel OSL variants that make use of latent luminescence signals and that exhibit higher saturation levels can surpass that conventional boundary. Long-range OSL dating variants are continually being put to the test, and they continue to demonstrate their adaptability to different settings and periods of geological time. These long-range OSL techniques are enhancing the study of human evolution and landscapes in which they (hominins) lived, allowing us to peer deeper into the Pleistocene.

**CONCLUSIONS**

Palaeoenvironments influence major human evolutionary and cultural shifts. Optically stimulated luminescence (OSL) dating can link major environmental events with human cultural changes on timescales greater than 14C dating can manage. Sharp signal resetting in most sunlit environments has established OSL as the dominant technique in dating sedimentary deposits associated with

---

**FIGURE 3** Sedimentation rates of colluvia for various archaeological periods of the Phlius basin (northeast Peloponnese, Greece) determined by OSL from quartz. The error bars represent the uncertainty on the sedimentation rate estimates. The star on each graph represents minimum sedimentation rates, with true values being higher. Samples B1, A14, and B2 correspond to different study sites, and the differences between their corresponding plots indicate different sedimentation patterns. *After Fuchs et al. (2004).*

**FIGURE 4** (left) Satellite images showing the location of the Quaternary marine sequence studied by high-temperature post-infrared infrared stimulated luminescence (pIR-IRSL). The sequence location (indicated by the yellow box on southeast Zante Island) is in the central Mediterranean off the coast of mainland Greece. (right) A simplified illustration of the Early–Middle Pleistocene stratigraphy for this marine sequence. The Brunhes–Matuyama boundary (B/M) and subsequent geomagnetic excursion known as “17a” are indicated. *Satellite images © Harris Corp, Earthstar Geographics LLC, SIO, © 2015 Microsoft Corporation.*
Table 2

<table>
<thead>
<tr>
<th>Origin</th>
<th>Sample</th>
<th>Geomagnetic event</th>
<th>Magnetometric stratigraphic age (Ma)</th>
<th>$U$ (ppm)</th>
<th>Th (ppm)</th>
<th>$K$ (wt%)</th>
<th>Dose rate (Gy ka$^{-1}$)</th>
<th>$D_r$ (Gy)</th>
<th>Age (Ma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Canary Islands</td>
<td>Galdár loess</td>
<td>N.A.</td>
<td>N.A.</td>
<td>2.9 ±0.1</td>
<td>9.3±0.1</td>
<td>1.05±0.01</td>
<td>2.98±0.20</td>
<td>524±53</td>
<td>0.18±0.02</td>
</tr>
<tr>
<td>Greece</td>
<td>Zante1</td>
<td>B/M reversal</td>
<td>0.77</td>
<td>1.5±0.1</td>
<td>4.7±0.1</td>
<td>0.95±0.01</td>
<td>1.5±0.1</td>
<td>1126±229</td>
<td>0.72±0.14</td>
</tr>
<tr>
<td>Greece</td>
<td>Zante2</td>
<td>17a excursion</td>
<td>0.66</td>
<td>1.1±0.1</td>
<td>1.6±0.1</td>
<td>0.16±0.01</td>
<td>0.64±0.01</td>
<td>716±87</td>
<td>1.12±0.14</td>
</tr>
<tr>
<td>Greece</td>
<td>Zante3</td>
<td>17a excursion</td>
<td>0.66</td>
<td>1.8±0.1</td>
<td>1.9±0.1</td>
<td>0.3±0.01</td>
<td>0.95±0.01</td>
<td>875±180</td>
<td>0.92±0.19</td>
</tr>
<tr>
<td>Greece</td>
<td>Zante4</td>
<td>17a excursion</td>
<td>0.66</td>
<td>2.5±0.1</td>
<td>4.6±0.1</td>
<td>0.99±0.01</td>
<td>1.96±0.01</td>
<td>1224±168</td>
<td>0.62±0.18</td>
</tr>
</tbody>
</table>

ACKNOWLEDGMENTS

The study by OSL of sediments from the Canary Islands was supported by an ESF-MedCLIVAR grant awarded to CDA in 2011. CDA was hosted by the Departamento de Física Universidad de Las Palmas de Gran Canaria (ULPGC) and he is thankful to Francisco José Pérez Torrado (GeoVol, ULPGC), Inmaculada Menéndez González (IOCAG, ULPGC) and Juan Francisco Betancor Lozano (Laboratorio de Palaeontología, ULPGC). Sampling of the marine sequence on Zante Island was guided by M. Papanikolaou (Ichron Ltd, UK). Material from the Canary Islands and Zante was measured by CDA at the OSL laboratory of NCSR DEMOKritos, Athens (Greece) during the period 2011–2012. We are thankful to RG Roberts (University of Wollongong, Australia) for his valuable comments on the manuscript. Radioelement concentrations were measured by ICPS at Acme Analytical Laboratories in Vancouver (Canada).

REFERENCES


Arnold L and 6 coauthors (2014) Evaluating the suitability of extended-range luminescence dating techniques over early and Middle Pleistocene timescales: Published datasets and case studies from Atapuerca, Spain. Quaternary International, doi: 10.1016/j.quaint.2014.08.010


Von Suchodoletz H and 5 coauthors (2010) Soil moisture fluctuations recorded in Saharan dust deposits on Lanzarote (Canary Islands) over the last 180 ka. Quaternary Science Reviews, doi: 10.1016/j.quascirev.2010.05.014

