

ADVANCES IN THERMO- AND OPTO-LUMINESCENCE DATING OF ENVIRONMENTAL MATERIALS (Sedimentary Deposits).

Part II: Applications

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ABSTRACT

This review is about the state-of-the-art of the luminescence dating methods on environmental non-burnt materials formed mainly during the past quarter of a million years. It is focused particularly on the applications on sedimentary deposits, including materials related to cultural activities. It describes some major applications of TL, but mainly of the OSL on sedimentary deposits, whereas particular reference is made to a special dating application in archaeology. Limitations and some problems of luminescence dating methods are also mentioned, as well as guidelines for useful application of these luminescence methods.

The new applications of thermoluminescence (TL) and optical stimulated luminescence (OSL) / infrared stimulated luminescence (IRSL) dating (tephra, ceramics, sedimentary deposits) are demonstrated and the notable progress in understanding the limitations to accuracy, as well as the development of new techniques is reported. Only a few types of quaternary deposits can at present provide reliable TL dates. Suitable samples include fine loess, thin clayey layers in lacustrine deposits, clayey mud in off-shore or near-shore marine sediments, and organic-rich palaeosols developed in loess, lacustrine, fluvial or dune deposits. Dune sands are very promising, but greater reliability has yet to be demonstrated. For all such unheated sediments, the "partial bleach" method is the preferred and prudent technique to use. The OSL and IRSL dating of ceramic material seems to be well established, the dating of carved megalithic stone building needs more dating results for reconfirmation and the OSL, TL and IRSL of quartz and feldspar in sediments require more applications of known age samples.

KEY WORDS: thermoluminescence, optical luminescence, minerals, quartz, feldspar, sediment, dose, bleaching, ceramics, megalithic

INTRODUCTION

The attractiveness of thermally and optically stimulated luminescence (TL and OSL) dating meth-

ods is that they can be applied directly to environmental deposits usually considered barren of datable material, as well as to deposits laid down

** in memoriam to late Dr Vagn Mejdahl, pioneer in dosimetry and dating, a wise and gentle colleague, friend and teacher.*

within the lower Quaternary time window (ca 40 to 300 Ka, 1 Ka=1000 years) not routinely accessible by other methods (e.g. ^{14}C , K-Ar, Fission Track). The latter is due to either the low half-lives of the C-14 radioisotope or the lack of suitable materials present.

Three classes of events can be dated by luminescence methods: the last cooling of a mineral, the last exposure to sunlight, and the growth of a mineral due to crystallization process.

Most progress has come from studies of known-age material, deposited under known conditions, either heated, sun bleached materials or deposited carbonates.

Within *Class I (heated materials)*, both distal and paroxysmal tephra deposits, as well as heated rocks have been dated, using TL techniques originally developed for pottery dating (Aitken, 1985; Berger, 1991; Berger and Huntley, 1983; Liritzis *et al.*, 1996a; Liritzis and Galloway, 1982a; Miallier *et al.*, 1983).

Within *Class II (sun bleached materials)*, loess, dunes, buried soils, waterlaid silts, glaciofluvial and oceanic sediments, and carved megalithic stone building have been successfully dated (Wintle and Huntley, 1980; Mejdahl, 1985, 1986; Berger, 1988; 1995; Berger *et al.*, 1987; Berger and Eyles, 1994; Singhvi *et al.*, 1982; Ollerhead *et al.*, 1994; Liritzis, 1994a, b; Liritzis *et al.*, 1996b, 1997b).

Class III includes various types of deposited carbonates (travertines, stalagmites, calcium nucleus or calcium fissures of old rocks etc.) (Debenham, 1983; Franklin *et al.*, 1988; Li Hu Hou, 1988; Wintle, 1978; Liritzis, 1989).

This review includes greater detail in the description of the dating methodologies employed, discusses critically the applications of mainly the optical stimulated luminescence technique of unheated sediments and various other major sources of error and means to minimize their effects, and emphasizes the potential luminescence dating of sediments, including the solar zeroing of luminescence in carved megalithic limestone blocks used in archaeological contexts. Within the past ten years, significant advances in procedures technology and understanding of the thermoluminescence (TL) and optical luminescence stimulated by green or infrared light (GLSL or IRSL, also abbreviated to OSL) behavior of minerals have been made that place lumi-

nescence dating techniques on the verge of widespread application to many Quaternary deposits and new archaeological materials.

Applications of TL methods to the dating of quaternary deposits have been reviewed by Singhvi and Mejdahl (1985), Mejdahl (1986), Berger (1988), Wintle (1993a) and on OSL dating by Wintle (1993b).

The quaternary scientist interested only in a reliable, routine geochronometer will find general guidelines for appropriate application of the methods.

APPLICATIONS OF TL DATING OF QUATERNARY SEDIMENTS

During the transportation of minerals perhaps high up in the atmosphere, there is ample opportunity for their TL to be severely sun bleached, constantly and continuing onwards from the time they lay on the surface, before being blown by the wind, prior to being covered.

The involvement of glacial action suggests there may be correlation with climate and for this reason its dating is of considerable interest in quaternary research; equally it is important in the dating of Paleolithic sites, either when remains of human occupation are found in the loess itself or in establishing the chronology of a stratigraphic sequence.

Sunlight is not, however, the only agent that has been suggested for erasure of previously acquired TL, weathering and glacial grinding are put forward as additional possibilities, by the initial researchers (Dreimanis *et al.*, 1978; Morozov, 1968). However, is currently taken for granted in nearly all laboratories that the effects of sunlight are at any rate dominant.

Below are given some examples of TL applications.

1) Wintle (1981) published the first comprehensive TL dating program on loess providing six dates on Pegwell Bay (Kent) loess deposits, confirming the late Devensian period. She used 4-11 μm grain size fraction applied the R- Γ method and an overall error of 20% was suggested.

2) The stratigraphic ages of some well-known and characteristic palaeosols in Austrian and Hungarian loess sections have been re-established by Zoller *et al.* (1994), who presented 41 new TL dates. For loess older than the last interglacial, quartz and feldspar inclusions (90-200

μm) yield more reliable TL ages than the polymineralic fine grain fractions.

The total bleach-regeneration method was applied for ED determination except of one sample, determined by partial bleach (preheat 180°C for 1 min). Some ages were supported, also, by archaeological means, amino-acid dating and biostratigraphical correlations.

Although apparent TL ages obtained from the polymineralic fine grain fraction tend to underestimate the ages of loess older than 50-100 ka, their possible usefulness for inter-regional correlations has been demonstrated.

Similar underestimated TL dates for loess have been reported by Packman and Grun (1992), Frechen (1992), Frechen *et al.* (1992).

3) Wintle (1993b) has summarized the reports of age underestimations, for the alkali feldspars from cover sands of western Europe, some by as much as 20-40 %, even for deposits younger than 100 ka.

4) Debenham (1985) observed too, consistent age underestimations for deposits mostly loess, thought to be older than 100 ka, using the total bleach regeneration procedures - a dose-response interpolation procedure and glow-curve signal integration, at $300\text{-}310^\circ\text{C}$, for all samples. He attributed this age underestimation to a hypothetical long-term loss of luminescence centers, which has been criticized by Berger (1988, 1989). Berger (1989) states that this hypothesis is valid only if age underestimations measured with the use of the regeneration methods are measured with all the TL dating procedures. The latter has not been observed, and so Debenham hypothesis seems incorrect.

5) Furthermore, newer TL ages for some arctic lake core Alaskan samples do not support the argument for the relevance of limited lifetime parameters (Berger and Anderson, 1994). Instead, there the TL ages agree well with C-14 ages, and TL has dated the whole core to 125 ka. It was used low energy ($>550\text{ nm}$) optical bleaching, $110\text{-}140^\circ\text{C}$ for 4-5 days pre-readout heat treatment and blue TL emissions, and the partial bleach R- β and dose-plateau plots for ED determination. Weighted saturating exponential regression and error analysis was used for the growth curves.

These results demonstrated that effective zeroing of light-sensitive TL has occurred in the younger

samples from Squirrel lake core. Based on the occurrence of generally similar sedimentology with depth, it was inferred that such zeroing has also occurred in the older samples and that therefore, all of these TL age estimates are reasonably accurate.

It was recognized that a plateau in ED values is a necessary (though not sufficient) condition for an accurate TL age. Also, it was stated that no pre-heating recipe is suitable for all samples in a given lake core. The reasons for such idiosyncrasies may lie in the polymineral/polygrain nature of the TL samples.

Gross age inaccuracies such as mentioned above have hindered the acceptance of TL sediment dating as a valid tool for quaternary geology. However, a clearer view of the causative agents of these age underestimations is now emerging.

6) Berger *et al.* (1992) have reported TL ages for the 4-11 m polymineral fractions in a suite of loess samples from New Zealand and Alaska that have relatively well constrained independent ages up to ~ 800 ka. The results from use of dose-response extrapolation procedures (partial bleach R- β and total bleach method) have shown that the measured TL ages agree within 1σ with expected ages (only 2 out of 13 ages agree within 2σ).

The age underestimation in New Zealand loess using the "partial bleach-regeneration method" by Berger and Huntley (1989) is attributed to dose-dependent sensitivity changes (Wintle, 1985). Therefore, it was suggested that TL age by the regeneration method for feldspar dominant sample older than ~ 70 ka should be avoided. However, this has been recently overcome.

7) Another accuracy test was completed by Huntley *et al.* (1993), using $\sim 100\ \mu\text{m}$ quartz grains extracted from stranded beach-dune sands as old as 800 ka from south Australia with a regeneration procedure. They measured TL ages in close agreement with expected ages up to ~ 300 ka or ~ 650 ka, depending upon the tectonic uplift model used to calculate expected ages. Therefore, a global limitation to the accuracy of TL sediment dating methods for samples older than 100 ka does not exist, rather some sample-dependent and laboratory-dependent factors appear to play some role.

8) Berger and Easterbrook (1993) have further developed reliable procedures for accurate TL dating of quaternary waterlaid sediments; 6

glaciolacustrine, 3 glaciomarine drift and 8 floodplain deposits. They used the "partial bleach (R-b/G) technique" to fine-silt polymineral grains. Because of color and time-integrated intensity of natural light in sediment-laden water columns, greatly affect the degree to which light-sensitive TL in sample-dominant feldspars, is reset during transport before burial, low energy (560-800 nm) laboratory illumination (optical bleaching) was used (Berger, 1990). This region of light spectrum lies just on the low-energy side of the most persistent wavelengths in turbid water. The partial bleach (R-b/G) procedure and associated dose-plateau plots were used to measure ED values. Preheating to 75°C for 8 days or 110°C for 4 days was applied to remove the unstable TL from irradiated samples.

It was confirmed that from the glaciolacustrine sediments only the clayey laminae are likely to generally contain effectively zeroed light-sensitive TL. From the 3 glaciomarine-drift samples, two TL ages were overestimated and the third gave the expected result (177 ± 38 ka).

From the eight floodplain samples, it was proved that the overbank deposits are unsuitable for present TL procedures. On the other hand, fine-grain floodplain sediments believed to have been deposited in quiet, ponded water on the floodplain, (e.g. oxbow lakes, abandoned channels, associated marches and swamps) appear to be good candidates for TL dating up to at least 200 ka.

So far, there have been reported several OSL dating results of sediments older than 100 ka. (Duller, 1996; Berger, 1994, 1995; Mejdahl, 1989; Wintle *et al.*, 1993).

The above work showed that the results for sediments from the Whidbey Formation are consistent with its expected last-interglacial age, and those for three samples from stratigraphically correlated units of the pre-last-interglacial double Bluff Drift are consistent with deposition near or beyond 200 ka.

9) Moreover, Berger and Eyles (1994) have reported eight satisfactory TL ages of quaternary sedimentary deposits in Toronto area, Canada. The samples are clayey lamina, silt, peaty sand, from shallow water lacustrine deposits and glacial origin. They employed the partial bleach R-b TL technique to separate the post-burial light-sensitive TL signal from the total signal and to measure the ED value, 570-700 nm wavelength for

optical bleaching and 110°C for 4 days preheating.

It was proved, once more, that this preheating was effective - the same fractional decay in TL for 12-13 months storage at 20°C as resulted from the 110°C/4 days- while age underestimation was then unlikely. Almost all samples produced excellent ED plateau.

The TL ages ranged between 20-80 ka and significantly contributed to the chronostratigraphic sequence of the Toronto area and indicated the need for major revision of the existing time framework of central Canada.

SOME APPLICATIONS OF IRSL DATING OF SEDIMENTS

The target for the IRSL dating of sediments is the correct determination of incomplete bleaching the correct procedure for the measurement of equivalent dose (ED) and verification that the IRSL signal is stable for the geological time-scale. Incomplete bleaching of IRSL of coarse feldspar grains from colluvial sediments (Li, 1994) and glacial outwash deposits (Duller, 1994c) have been reported. In these cases, some grains are well bleached and others are not. This was detected by scatter in the ED values obtained when using the "single aliquot" method. For fine-grained alluvial sediments all the grains are likely to have experienced the same inadequate light exposure and hence no such scatter would be expected.

Aitken and Xie (1992) applied an approach using different parts of the IRSL decay curve. They subtracted later portions of the IRSL decay curves from the initial parts in an attempt to isolate the rapidly bleaching component.

For laser stimulated signals from sediment, Huntley *et al.* (1985) used different parts of the decay curves from irradiated aliquots to construct growth curves which were extrapolated to zero luminescence intensity to obtain EDs. Samples, which did not give a consistent ED value as a function of laser exposure time, were considered not to have received sufficient light exposure at deposition.

Below are given some application efforts for dating.

1) A useful use of IRSL signal is for scanning sediment cores. It has been applied to glacio-lacustrine and lacustrine cores (Duller *et al.*, 1992).

The association between major dark bands in the core and the IRSL variation is clear, the significant signal variations correlate with the laminae structure. The signal observed seems to be predominantly a measure of the exposure of the material to light during erosion and transportation. This analysis is very rapid and allows an instant appraisal of the nature of the luminescence signal due to feldspathic minerals. In the field, as in the laboratory, it would be invaluable in identifying stratigraphic breaks and episodes of reworking. For material in saturation, variations in signal level may reveal differences in the proportions of materials that produce IRSL. This approach could be applied to the determination of the CaCO_3 content of deep-sea cores since this has no IRSL signal. Thus, variation in signal level will be related to the ratio of CaCO_3 to other minerals that yield an IRSL signal, although this is not always the case.

However, two lacustrine cores yielded no IRSL signal above background and this may be due to the lack of a significant feldspar component in the cores, their young age or, most likely, the complete zeroing of the IRSL signal at deposition.

2) The IRSL spectra have been shown to be similar for a wide range of datable feldspars from loess. For dating applications, it is necessary to consider how to ensure that the IRSL signal measured is stable for the geological time-scale under consideration. In TL, various forms of 'plateau test' have been devised, such as, the ED versus glow-curve temperature, or, $(N+\beta)-I_0$ versus $(N-I_0)$, where N is the natural and I_0 is the residual signal. In OSL, empirical preheat procedures are employed, e.g. 220°C for 10 hrs or 140°C for 16 hrs, with a further 24 hr storage at room temperature (Wintle, 1993a,b).

Comparisons of IRSL and TL EDs have been made for fine-grain loess, for which preheat at 140°C for 16 hrs, as recommended by Li (1992), was used.

Samples from the last 130 ka from a section at Dolni Vestonice in the Czech Republic have given EDs which are very similar, although the ED from IRSL measurements is consistently less (15%) than that for TL. This could be explained by the preheat being insufficient to erase totally the unstable IRSL signal.

3) Tephra beds serve as time-stratigraphic marker horizons and can also provide a proxy record of

magma composition and eruption characteristics. Glass (4-11 m) samples from 15 volcanic ash deposits were examined for their suitability for optical dating, by Berger and Huntley (1994). The samples ranged in age from 8 to 580 ka. Only very weak luminescence was observed using 514 nm (green), 633 nm (red) and ~ 880 nm (IR) stimulation.

An equivalent dose of 29 Gy was obtained for Mazama ash using IRSL, which agrees well with TL giving an age of 7.7 ka.

Glass had been separated from the ash by floating at relative intensities in the range 2.45-2.50 (depending on the sample). The effectiveness of removal of almost all feldspar crystals was evidenced by significantly lowered TL intensities and by microscopic examination.

It was also found that high temperature preheating, 120°C for 4 days, could be used for OSL of old (>100 ka) glass shards. Such high temperatures would be preferred to remove effectively laboratory induced unstable luminescence, because optical dating lacks an internal stability test, such as that provided by the dose-response plateau test in TL dating.

This effective removal of unstable luminescence for quartz has been approached by another method (Liritzis, 1995). Here, instead of preheating, the unstable luminescence at room temperature corresponding to a small additional beta dose to the natural signal is measured. Subsequently, the unstable signal, which corresponds to the natural luminescence and has already decayed at environmental temperatures, is defined. The additive dose growth curve is constructed for a single aliquot, for the natural and subsequently added doses, β_1 . Proportional subtraction of the unstable component of the $(N+\beta_1)$ data point from all data points of the curve brings to another line, which represents the additive dose growth curve of the natural or stable component of the sample. Extrapolation of this line to system background level, determines the equivalent dose (Fig. 1). This proposed method is still at the testing stage.

4) Mineral grains transported by rivers can experience a wide variety of light exposure conditions before deposition and eventual incorporation into an alluvial sedimentary deposit. Different grain sizes are likely to have different histories of light exposure, with fine grains being carried nearer to

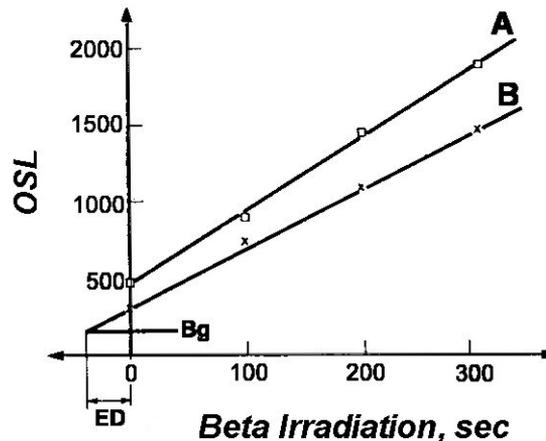


Figure 1. Green OSL signal of archaeological quartz versus beta dose irradiation time in seconds (100 sec = 26.6 Gy). Curve A is the corrected (due to fading and preheat) readings for stable plus unstable component. Curve B is the residual from the subtraction of the unstable component from curve A. Curve B (of the form $OSL=323+3.5T$, $r=0.99$, prob. level=0.0067) intersects background B_g and defines the ED (after Liritzis, 1995).

the top of the water surface and coarse grains being moved by saltation close to the riverbed. Some grains will be deposited on mid-channel bars and those on the surface will be exposed to direct sunlight. Berger (1988, 1990) has reviewed effects of light intensity variation in depth of water-laid sediments. He concludes that the best approach to determine ED and the age is the partial bleach method, when only light having wavelengths above 550 nm should be used for the optical bleaching; this would avoid overbleaching in the laboratory by wavelengths not experienced at deposition.

5) Fuller *et al.* (1994) tested the "partial bleach" methodology applied to the IRSL on an alluvial sediment from the Danube river in Romania. Experiments were carried out on 4-11 m grains collected from overbank fines deposited directly above skeletons from a burial site at Schela Chdovei on the east bank of the river. The age of the skeletons is thought to be Mesolithic, which in this part of Europe lasted from about 8.4 ka until 7.3 ka calendar years ago.

The "additive dose" IRSL method was used with linear extrapolation to system background level, as well as the "total bleach" TL method using a residual level after 24 hr-bleach with an unfiltered light source. Finally, IRSL growth curves were constructed for bleaching times greater than 400 sec. Those curves intersect the curve for the

unbleachable data set to give the ED as a function of bleaching time (Fig. 2). The four shortest bleaching times show a trend of increasing ED with bleaching time, probably due to insufficient bleaching. The three longest bleaching times were however averaged to produce a mean $ED=34.6\pm 4$ Gy, which indicates that the sediment was well bleached at deposition. This agrees with the values given previously for the "total bleach" method of 31.5 ± 6.3 Gy and the "additive dose" method of 36.3 ± 3.6 Gy. The corresponding age is therefore 8.16 ± 1.29 ka, which corresponds with the Mesolithic age of deposition suggested by archaeological evidence.

Simulation tests of this work suggested also that the IRSL partial bleach methodology is not appropriate for sediments, which have not had their IRSL set to zero at the time of deposition. It would be better to compare the IRSL EDs with those obtained for other luminescence signals, such as the TL or green OSL in quartz. Such a comparison has been made successfully for ceramics (Liritzis *et al.*, 1997a).

6) Fluctuations in eustatic sea level, combined with tectonic uplift and tilting of the region around Wanganvi in south of north island New Zealand have created a series of raised marine terraces rising 300 m in altitude. Tephra and Aeolian sand units were preserved on the top of these terraces in a series of loess. Dates had been

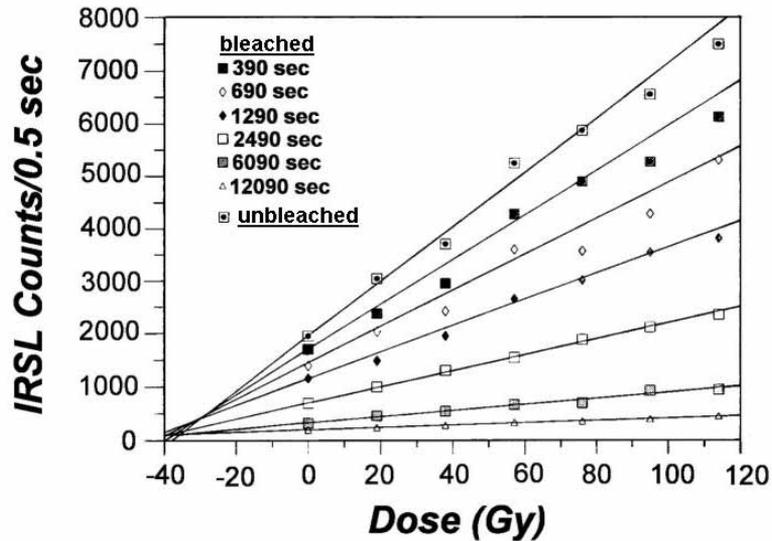


Figure 2. Partially bleached growth curves, derived from fine quartz grains data of alluvial sediment deposited from the Danube. Due to insufficient bleaching various bleaching times were used between 390 sec to 12090 sec with 0.5 sec IRSL shines applied successively on each disc after every bleach. The additive dose procedure was used and all curves intersected at a point giving an average (after Fuller *et al.*, 1994).

produced by amino-acid racemization, fission track and luminescence of fine grains.

Duller (1994b) has applied IRSL and TL of multiple aliquots on feldspar inclusion (150-195 m) grains to determine EDs by the "total bleach" and "regeneration" methods, for a suite of 12 such dune sands. The ages span from recent to 350 ka. The preheating was 140°C/62 hrs, and the bleaching made with SOL-2 solar simulator.

In general, there is good agreement between EDs for TL and IRSL measurements, and between geological age estimates and the luminescence ages, for samples younger than 30 ka. Some underestimated ages are explained from: i) the uncertain water content, ii) contamination due to translocation of younger material down to profile, iii) uranium decay chain disequilibria caused by ground water mobilization, iv) TL and IRSL signals of older samples are close to saturation, v) high percentage of mafic minerals inhibited bleaching or have not exposed sufficiently to sunlight, vi) for other underestimations in ages for >30 ka are due rather to uncertainty in the age control or the long term fading of feldspars (Mejdahl, 1989).

Withstanding the uncertainties in the age control in three out of 12 samples, the conclusion is the

reliable luminescence ages may be produced up to 130 ka, using the methods described.

7) Ollerhead *et al.* (1994) have indicated the great promise of the OSL dating method, from five OSL dates (250-300 years on K-feldspars, employing the additive dose method with IRSL and 6-7 hrs sun bleaching), compared to two TL dates following the additive dose linear plots and the partial bleach R-G method. The ages ranged from present day to 765 ± 45 ka for the oldest dune. The ages and their resolution exhibited the expected seriation from youngest to oldest and are consistent with historical evidence coupled with an assumed constant sediment aggradation rate. They have produced readout curves of ages versus OSL signals.

8) Duller (1994b) in his dating of poorly bleached sediments discriminate between two types of poorly bleached sediments; type A, where all the grains have been equally but incompletely bleached at deposition, due to the duration of exposure or restricted bleaching spectrum, and type B, where there exists a mixture of grains exposed to daylight at deposition for very different periods of time, some were totally bleached and others partially bleached, but still retain a large luminescence signal at deposition e.g. due to

very short transport distance (colluvial deposition), Aeolian (well-bleached) material mixed with fluviially or glacially transported.

The additive dose growth curves of multiple aliquots was employed, and the variation in ED versus bleaching time (plateau test) was used, based upon the rationale which uses the different rates at which the IRSL and TL signals are bleached, to define the period of time they were exposed to sunlight and hence to define the residual level to use for ED determination.

It was found that the TL and IRSL EDs are the same intersected, after 10 min or 3 min of bleaching, for the three samples studied.

Analysis of data scatter for multiple aliquot additive dose growth curves indicates the type A or B of the sediments. Type A sediment shows no clear change in scatter with added dose, in contrary to type B sediments. A similar conclusion for the three samples examined was reached employing the "single aliquot" method of IRSL and plotting ED with IRSL signal. Type A sediments that contained a large proportion of unbleached grains gave a large ED and a large luminescence signal. Conversely, a large proportion of grains well bleached gave smaller ED and a smaller luminescence signal. Also, a type B poorly bleached sample is expected to show a trend of lower EDs and lower luminescence signals, whereas a sample type A shows no variation in ED with luminescence signal. It was thought that for poorly bleached sediments a minimum estimation of ED is obtained by using the intersection of the regression line with the y-axis, but still overestimates the age for some samples.

Finally, it was suggested that for type A, where a large proportion of the TL signal was not removed at deposition, the "partial bleach R-I" and "plateau" methods might be applicable (Mejdahl, 1988). Dating of type B sediments involves further problems and the analysis should be treated with caution, since there is no safe way to confirm that any of the grains in the sample had the majority of their IRSL signal removed at deposition. The EDs determined, thus, should be treated as maximum values.

Analysis of "single aliquot" data or the pattern of scatter in a multiple aliquot additive dose procedure may indicate when the sediment is type B. Such samples containing at least a proportion of grains that had most of their IRSL signal removed

at deposition may be dated using the "single aliquot" additive dose method.

However, none of the methods described are able to date samples where the IRSL signal was not reduced to a low level at deposition in at least a proportion of the grains and these must be considered undatable using luminescence methods. The identification of samples in which no grains were well bleached at deposition remains a problem and hence it would seem prudent to view any date on type B sediment as a maximum value.

A similar conclusion that the cause of the scatter in the additive dose growth curves is not intrinsic to the samples, but is related to the variation in the stored dose between different grains (partial bleaching at deposition) has been reached by Rhodes and Pownall (1994), studying quartzes from young glaciofluvial sediments and by Rhodes (1990).

SOME MAJOR LUMINESCENCE DATING APPLICATIONS

In the established systems for OSL dating of both quartz and feldspar, an argon ion laser provides the green stimulating light (Huntley *et al.*, 1985; Smith *et al.*, 1986), and a 75 W halogen lamp light which provides a broad-band light source from which a suitable stimulation spectrum can be selected by using optical filters (Botter-Jensen and Duller, 1992). Later on (Poolton and Bailliff, 1989; Galloway, 1992, 1993, 1994) a system using green light emitting diodes (LED) for the OSL of quartz and feldspar was devised, comprising a ring of 16 green LED around the heating strip of the TL oven (Fig. 3). This system has been successfully used for tests and applications of single and multiple aliquot additive dose growth curves for dating (Galloway, 1993, 1994, 1996; Mejdahl and Botter-Jensen, 1994; Liritzis *et al.*, 1994; Liritzis, 1995; Liritzis *et al.*, 1996b; Murray and Mejdahl, 1998).

In particular, it was found that the green light stimulated luminescence (GLSL) on feldspars (orthoclase and microcline), following the "single aliquot" technique for ED determination can not be applied without the complication of making allowance for the change in sensitivity of the aliquot with re-use; a 7-10% change in sensitivity per re-use -bleached in the solar simulator SOL-2 or day light- and 3% for heating at 500°C, was found (Cutter *et al.*, 1994).

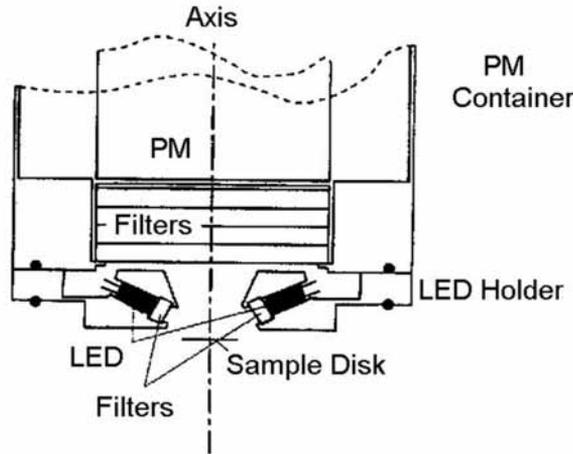


Figure 3. The OSL mounting assembly for the light emitted diodes (LED) and associated filters, shown in relation to the sample to be stimulated by the green light, the filters (GG475) used to separate the luminescence from scattered green light and the photomultiplier (PM) used to detect the luminescence (after Galloway *et al.*, 1997).

Some earlier applications to ceramics and sediments are reported in Berger (1995), Murray and Roberts (1997).

1) Two archaeological ceramics unearthed from the excavation at two Hellenic pyramids were dated by GLSL on quartz, with preheat at 200 °C/1 min, following the "additive dose" procedure,

and applying Duller's (1994a) and least squares correction (Liritzis *et al.*, 1997a, 2000). One result was compared to TL measurements. The ages of 3000±250 years B.C. and 650±200 years B.C. are in excellent agreement with the TL age result and typological grounds (Liritzis *et al.*, 1994) (Fig. 4).

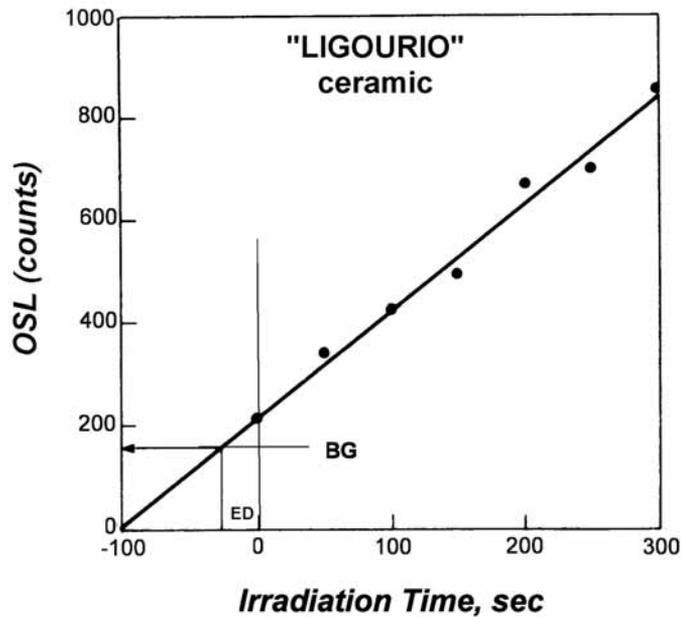


Figure 4. Green OSL versus irradiation time of beta source for ceramic quartz of Ligourio pyramidal, Greece. The background count intersects the curve, which provides an ED equal to 7.24 Gy (after Liritzis *et al.*, 1994).

2) A combined GLSL and IRSL study was made on nine ceramic sherds derived from 40 meters boreholes in the region of ancient Helice, northern Peloponnese (Liritzis *et al.*, 1997a, 2000). The aim was to date soil horizons, which bear very small ceramic sherds (1-2 cm²) in order to define the ancient occupational layer of the city of Helice, which, according to historical reports, had disappeared after a strong earthquake (submerged and strong tsunamis). The "additive dose growth curves" method was used, appropriately corrected applying Duller's (1991, 1994a) and Least Squares (Galloway, 1996) procedures, and the EDs were determined with extrapolations of saturating exponentials (Fig. 5). GLSL on quartz and on a mixture of quartz and feldspar grains was made. In fact, prior to these measurements, the luminescence of feldspars was erased by IRSL for 3000 sec and the ED for quartz by GLSL was measured. IRSL was also measured for feldspars producing ED similar to that from GLSL of quartz of the same sample. However, concerning the mixing quartz/feldspar ratio a useful indication has been devised by Duller (1995). It concerns the stimulation of mineral-specific GLSL signal ratio at 50°C to that of 300°C from multi-mineral samples, during the

monitoring of variation of GLSL as a function of sample temperature of these mixtures.

Liritzis *et al.* (1997a) have produced OSL and IRSL ages, which spanned from the 1st millennium A.D., through Roman period, Classical period, Dark Age, to early 2nd millennium B.C., with an average uncertainty of $\pm 10\%$ (Fig. 6).

In this work, it was reconfirmed the sensitivity change of minerals after bleaching by solar simulator SOL-2, daylight and heating (Fig. 7).

3) In the above work, a varved laminae sedimentary deposit at a depth of 13 meters, comprising of eleven alternated dark-light dands, was also dated by GLSL of quartz, following the "additive dose" growth curve procedure of "total bleach" method, and fitting the data with saturating exponentials. It was assumed that these layers were bleached by sun at the time of deposition. The mean age of four layers measured - 1st, 5th, 6th and 11th - was around 7500 ± 750 years B.C., displaced off the main chronostratigraphic sequence (ages versus depth) of the OSL, C-14 and archaeological ages of ceramic sherds. This implied the partial instead of total bleaching of the laminae, hence the obtained age being the maximum (Liritzis *et al.*, 2000).

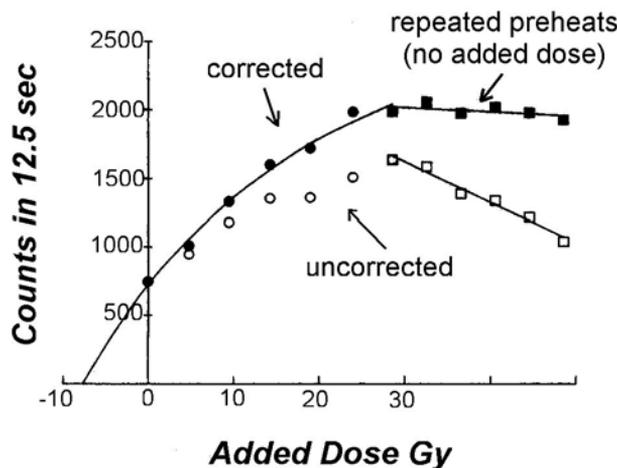


Figure 5. Single aliquot additive-dose data (uncorrected and corrected) for ceramic B42-1, with saturating exponential fit to the corrected points. Also shown, to the right of the figure, are the repeated preheat and real cycles which give the mean correction ratio (r) and the result of correcting these points by the mean ratio, which should ideally give a horizontal line (after Liritzis *et al.*, 1997a).

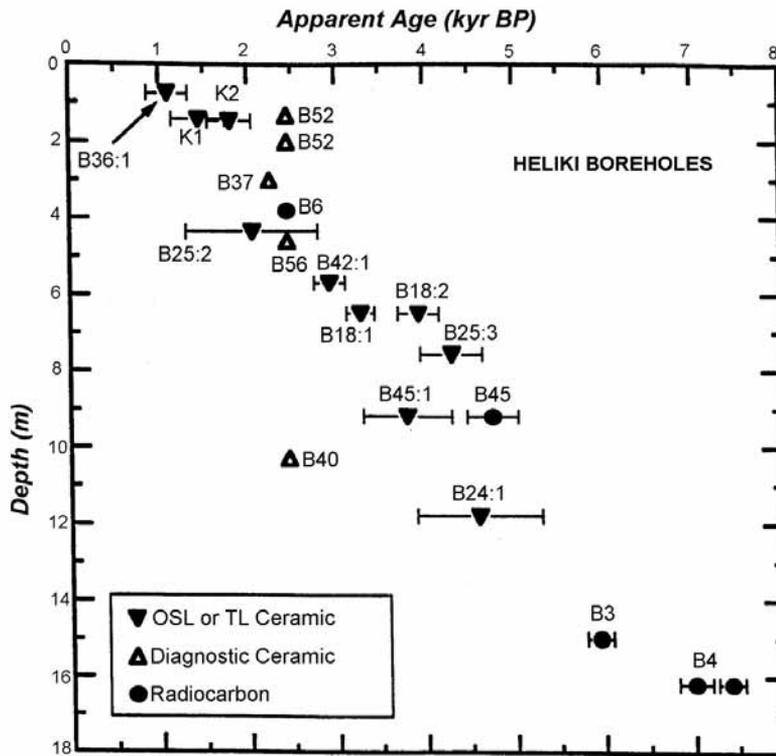


Figure 6. Apparent age by OSL or TL, supported also by diagnostic pottery and C-14 ages, as a function of depth, of tiny ceramic sherds recovered from boreholes in the search of ancient Helice (after Liritzis *et al.*, 2000).

4) OSL dating (420-550 nm of green light band) was applied on an archaeological ceramic from burnt stone (Sweden) following the "additive dose" method of multiple aliquots and fitting the data with exponential function. The obtained age of 854 years A.D. is similar within the error to 840 A.D. made by TL (Botter-Jensen and Duller, 1992).

5) One quartz dune sand from Sweden was dated by GLSL using the "additive dose" method, while the residual level was determined after 12 hrs exposure to sunlight. The OSL age of 9.5 ka is close to both the C-14 and clay-varve dates (Botter-Jensen and Duller, 1992).

6) In a TL dating project of dune sand at Puritjarra rock shelter in Australia's northern territory (Prescott and Fox, 1990), the TL ages, assuming total bleaching at deposition, provided consistent seriation with depth, but most of them suggest ages older than the most nearly corresponding C-14 age - the oldest C-14 age is $22,440 \pm 370$ years - by five to six thousand years.

Various repeats and tests concluded that the bleaching was incomplete for some samples. In this study, some samples were exposed in thin layers to Adelaide summer sunlight and were found not to be well bleached even after 21 days of summer sunlight. Others, though, bleached rapidly to low levels and the surface samples from these sites were found to have been already bleached, indicating long exposure to light in conditions presumed to reproduce the original conditions. It appears that solar resetting has been only partial, at Puritjarra, because the quartz used in the dating measurements is "shielded" from exposure to the sun by a thin layer of clay and/or iron oxide on the grains and because the orientation of the shelter virtually excludes direct sunlight. Here, the key question in dating bleached sediments, whether the materials (e.g. quartz) have been exposed to sunlight for 'long enough' to reset the TL clock, is revisited. Therefore, the "total bleach" assumption should be cautiously applied, as there are cases where wind-blown assessed sed-

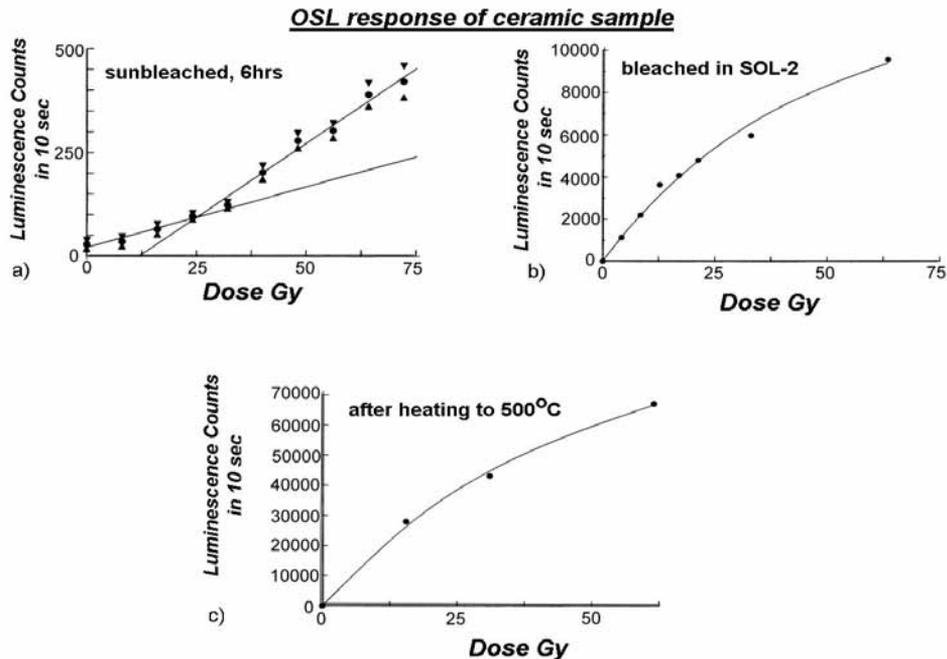


Figure 7. The response of ceramic sample K2 from Helice boreholes, Greece, after different bleaches: a) by daylight for 6 hours, showing a supralinear response at low dose and with an approximation to the response by two straight lines. The small non-zero intercept, if statistically significant, may be due to the recuperation phenomenon associated with bleaching and preheating. The result following bleaching for 1 hour in a Holn SOL-2 solar simulator is shown in (b) along with a saturating exponential fit to the measurements, luminescence = $12120 \times (1 - \exp(-0.024 \times \text{dose}))$. The response following bleaching by heating at 500°C for 1 min is shown in (c) along with a saturating exponential fit to the data, luminescence = $88800 \times (1 - \exp(-0.023 \times \text{dose}))$. The fit has essentially the same shape (exponent) in both (b) and (c), with (c) showing a higher luminescence sensitivity to dose (after Liritzis *et al.*, 1997a).

iments are not always completely bleached at deposition. On the other hand, in the "partial bleach" method, one should assume the worst and base the dating on a component in the TL that bleaches easily. Spooner *et al.* (1988) confirmed that this is the 325°C, emitting light at 380 nm (Fox, 1990). The half-intensity bleaching time by full sun is about 20 sec for this peak and a few minutes of full sun is sufficient completely to remove it. It can therefore, be assumed that the conditions for "total bleach" were satisfied for this peak at the time of deposition and any TL ages determined from this peak by total bleach methods will be correct. A suitable filter combination for isolating this peak accepting 320-380 nm signals and largely rejecting other wavelengths that may not readily bleach, may offer a suitable new methodology to obtain more accurate dates (Franklin and Hornyak, 1990).

7) Stickertsson and Murray (1998) applied multi-

ple-aliquot and single-aliquot protocols to quartz from eleven late Pleistocene marine and freshwater deposits of known ^{14}C age, and from a historically dated Aeolian deposit. The three OSL aliquots- SARA, single-aliquot additive-dose, and SARD were used. The OSL ages ranged between 3000 years and 25000 years. The resulting dates are discussed in terms of absolute chronology and bleaching environments. They concluded that the application of preheat plateau tests following the single-aliquot additive-dose protocol to all samples was not universally applicable. But they proved that the single-aliquot, especially the SARD, protocols offer a rapid and efficient approach to estimating the ED in quartz taken from sedimentary sequences.

8) Roberts *et al.*, (1997) applied the multiple-aliquot additive-dose procedure and the single-aliquot additive-dose and the regenerative-dose protocols to date mud-nesting wasps, which

become petrified after abandonment. Mud-nesting wasps gather surface sediments from the margins of stream and pools, further exposing any quartz grains to sunlight during collection and transport of the mud. Final exposure to the sun occurs during construction of the nest in a rock shelter. Quartz grains embedded in a nest are hidden from sunlight and will accumulate the effects of the nuclear radiation flux to which they are exposed. Nests built by mud dauber and potter wasps in rock shelters in northern Australia often overlie and occasionally underlie prehistoric rock paintings. OSL of quartz embedded in the mud of fossilized nests shows that some anthropomorphic paintings are more than 17,000 years old.

9) A novel application of the TL dating, based upon the sunlight bleaching of light sensitive electron traps in carved calcites (marble, limestone), has been devised. It was initially developed to determine the construction age of two reduced size cyclopean-type pyramidal buildings of "Hellenikon" and "Ligourio" in Argolid, Greece (Fig. 8) (Liritzis, 1994 a, b; Theocaris *et al.*, 1997). The new principle of TL of optically bleached carved megalithic limestones and marbles is based upon the same principle to the optical (by sunlight) bleaching of electron traps of quater-

nary sediments (quartz, feldspar) during deposition on different environments e.g. oceanic, lacustrine, loess.

For carved megalithic stones, before their placing in the wall during preparation, it is accepted that presumably were left aside for at least one day. During this time the carved surface is exposed to sunlight, which sets the light-sensitive TL-clock to zero. In fact, there is a residual TL (insensitive to light) remaining, which serves as the zero level, or in other cases there exists an unknown residual TL level which must be defined, in order to accurately determine the ED for dating. The placing of the megalithic blocks to the wall overlaid by another block, the contact surfaces are in dark and the TL-clock of the light-sensitive traps are filled from that very moment with electrons produced from the ionization of calcitic lattice, by α , β , γ and cosmic radiation (Fig. 9). The sampling is made by gentle scraping the carved surface in the form of powder to a maximum layer of 0.5 mm. The TL-age = ED/dose-rate, where the dose-rate = $d\alpha_{\text{cal}} + \frac{1}{2} (d\beta_{\text{cal}} + d\beta_{\text{mor}}) + d\gamma_{\text{env}} + dc$, where, $d\alpha_{\text{cal}}$ is the dose of the alpha particles from the lower sampled calcite, $\frac{1}{2}d\beta_{\text{cal}}$ is the dose of the beta particles from the lower block, $\frac{1}{2}d\beta_{\text{mor}}$ is the beta dose from the mortar and $d\gamma_{\text{env}}$, dc the environ-

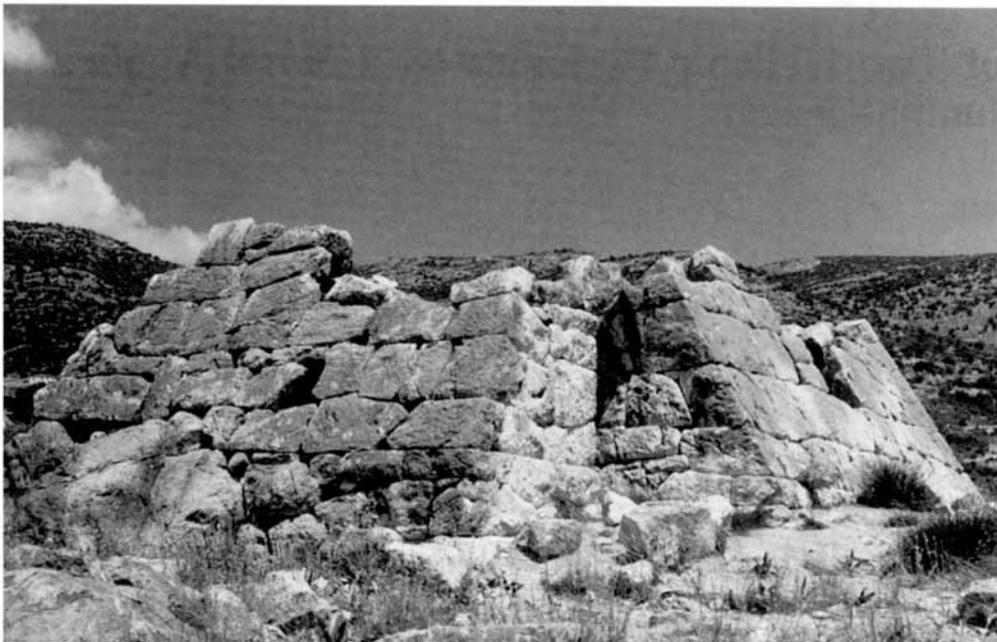


Figure 8. View from the east of the Hellenikon pyramidal building in Argolid, Greece. It was dated by the new method of optical thermoluminescence to ~ 2500 B.C. (Liritzis, 1994a, b; Theocaris *et al.*, 1997).

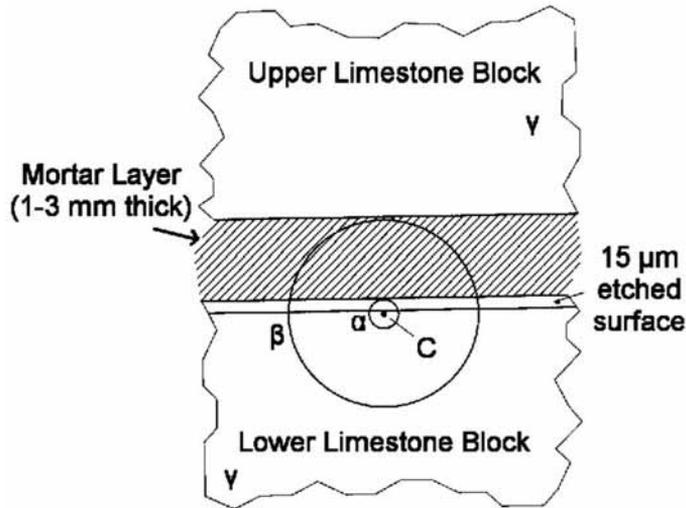


Figure 9. Diagrammatic illustration, inevitably not to scale, of the natural radiation dosimetry relevant to the dating process. The calcite for TL measurement is taken from the surface of the lower block after a layer of about $20\ \mu\text{m}$ thickness has been etched off the surface to ensure that clean calcite is used. C indicates a grain of the calcite used for dating in the diagram. It has been irradiated by alpha particles from within a sphere α of radius equal to the alpha particle range ($\sim 30\ \mu\text{m}$) that is primarily from alpha activity in the lower calcite block. It has also been irradiated by beta particles from within a sphere β of radius equal to the beta particle range ($\sim 1.5\ \text{mm}$) that is from a hemisphere within the lower calcite block and from a hemisphere within the mortar. In addition it has been irradiated by gamma rays from both limestone blocks and the surrounding environment, and by cosmic rays (after Liritzis, 1994 a, b; Theocaris *et al.*, 1997).

mental gamma and cosmic radiation. The major dose-rate component is the γ -radiation and sometimes the beta dose-rate from the mortar.

For the two pyramidal buildings and the Mycenaean wall the dated samples reached a residual TL level within the first 15-20 hours of sunlight exposure, thereafter the bleaching rate was very low to negligible. In these cases the "total bleach" method and the "additive dose growth curve" technique was employed; the residual TL signal at 15-20 hrs intersected the linear growth curves and determined the ED (Fig. 10). Further work has shown that the residual TL signal after about 20 hrs sun exposure is not always the rule, rather a slowly bleached component is present after the initial relatively rapidly bleached component.

In the case of Delphi (Temple of Apollo) sample this unbleachable or very slowly bleached component was present after the first 10 hrs of sun exposure, with a fast bleaching rate 6%-per-hr, followed by a very slow rate of 0.3%-per-hr.

In this case, it was devised a partial bleach

methodology for the actual sample and in simulated tests, based upon the rationale that the different parts of the TL glow-curve (TL intensity versus temperature) are not bleached in a similar manner, being affected variably by the different solar wavelengths. Therefore, plots of residuals after exposure of the geological TL, as a function of temperature (200 to 340°C) were studied. The correct ancient TL residual level is defined as follows: subtraction of residual shapes from the natural TL shape and construction of "dose-plateau" curves, i.e. plots of ED versus temperature. The correct dose value from these plots is for that sun exposure, which yields an ED plateau of maximum length.

Alternatively, a plot of [(natural-bleached residuals)/artificial TL] -versus-temperature, can produce the simulated residual at that curve with the maximum length, too (Fig. 11).

The TL ages for these megalithic structures were: a) 2730 ± 700 and 2260 ± 710 years B.C. for Hellenikon (Fig. 8) and Ligourio respectively,

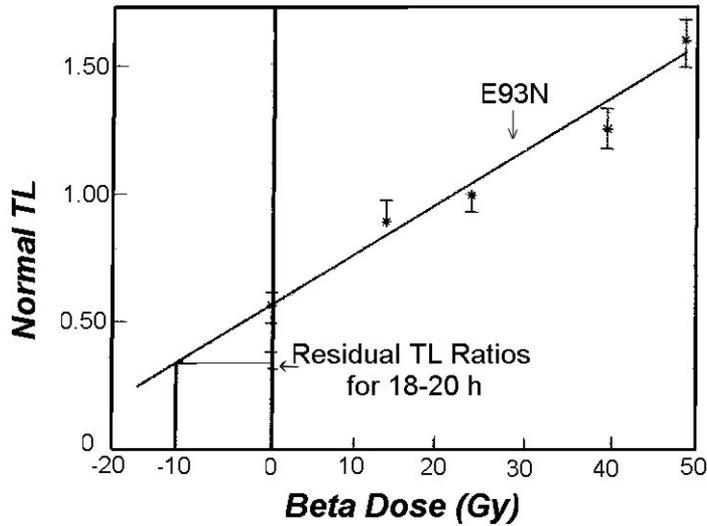


Figure 10. A typical additive-dose TL build up curve, in which the normalized TL is plotted against the added beta dose. The data refer to the Hellenikon pyramidal, sample E93N. The dose equivalent to the natural TL, that is the accumulated dose required for the age equation, is obtained by linear extrapolation to the residual level of TL remaining after prolonged exposure to sunlight (after Liritzis, 1994 a, b; Theocaris *et al.*, 1997).

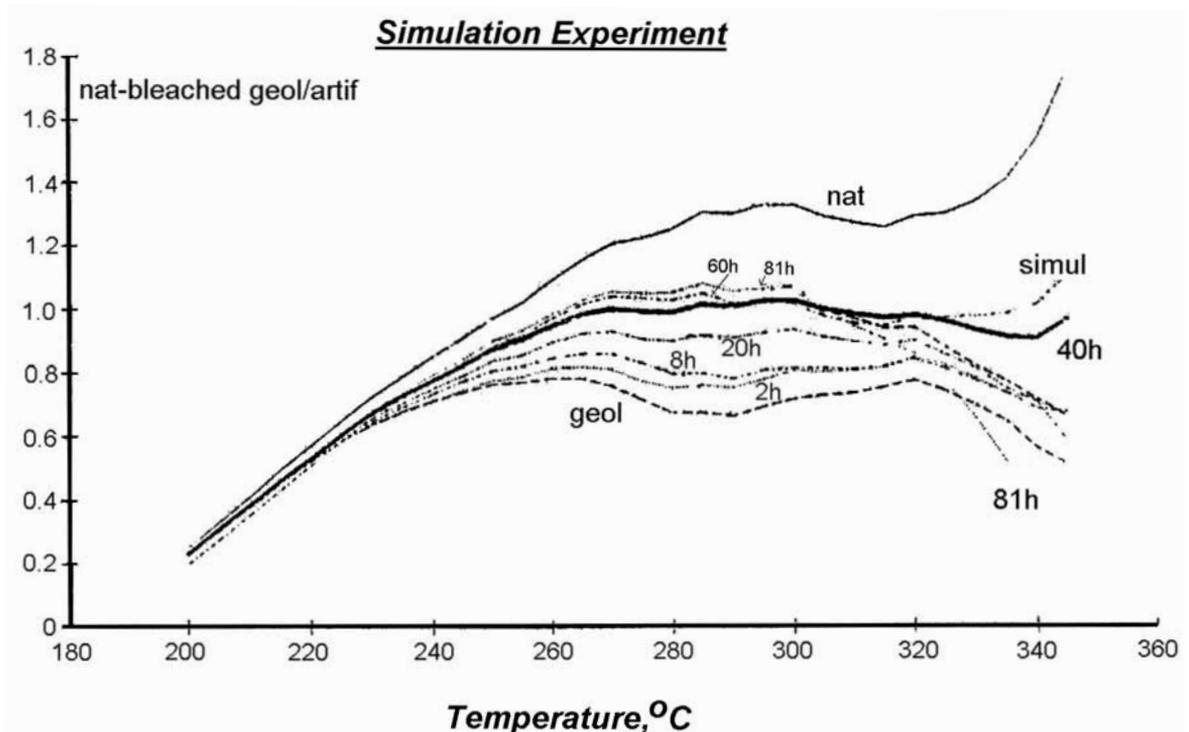


Figure 11. Simulation experiment for sample limestone E94N. Ratios of (natural minus bleached geological TL/artificial TL) versus temperature, for natural TL, geological TL, and 2, 8, 20, 40, 60 and 81 hours of sun exposure, as well as for the simulated (simul) residual of 35 h. The simulated 35 h (close to 40 h) curve produces the maximum plateau length (after Liritzis *et al.*, 1997b).

instead of the poorly attested archaeological estimation of 3rd to 4th c. B.C., b) 420 ± 300 years B.C. for the Delphi, concordant with its well known archaeological age of 550 B.C., c) 1100 ± 340 B.C. for the Mycenaean wall, similar to the archaeological estimation of 1280 B.C., d) the Efpalinion canal in Samos of c. 530 BC was dated to 570 ± 300 BC, and e) a Classical cyclopean wall in Amfissa, near Delphi, of the 5th century BC was dated to 480 ± 350 BC.

This novel dating method opens new prospects in archaeological research, especially in the dating of prehistory. The focus at the moment is targeted on the OSL dating of carved megalithic buildings, which requires a much less material and effort for luminescence measurements, a most needed task due to the limited quantity one can extract from such surfaces. Preliminary work on OSL TL of ancient carved sandstone and granitic megalithic stone buildings from Egypt and Greece is most interesting. The presence of quartz and feldspar minerals in these rocks and the fast solar bleaching of their luminescence open new directions for the dating of prehistoric monuments.

10) Dating of wind-blown quartz from archaeological sediments, in South Australia has produced interesting results, with dates ranging between 300 to 68,000 years. The additive-dose total bleach procedure was followed (Roberts *et al.*, 1996).

PROBABLE CAUSATIVE FACTORS IN LUMINESCENCE DATING

For decades, modeled and observed luminescence behaviors of idealized solid-state crystals, as well as of synthetic and some simple natural crystals (e.g. fluorite) have been presumed to apply directly to TL results from geological samples, namely sediments.

TL results from polymineral and multi-grain sediment samples to the conceptual window of the idealized simple (single) crystals of solid-state physics has caused us to overlook other, more direct, explanations for the causes of e.g. recently measured age underestimates in samples older than ~ 100 ka.

More attention should be paid to the nature of the actual sample. Thus, microanalytical examination and grain identification is rendered necessary for the future TL or OSL dating application to

sediments and megalithic stones. The grain populations consist of individual grains with characteristic luminescence emission.

Major categories of causative factors for the unreliability of luminescence dating of sediments include:

Age control

Well-constrained independent ages should exist on sites that the luminescence dating is applied. The successful procedures, that provide excellent agreement between luminescence and expected ages, should provide a protocol for future applications of sediment dating.

Regeneration methods

These procedures are presently unreliable for feldspar-bearing sediments (>70 ka) probably due to dose-dependent sensitivity changes and to optical bleaching crystal sensitivity changes. The latter applies, also, to quartz.

Ultraviolet emissions

The color of TL signal is another important factor and blue TL emissions are employed, rather than UV TL emission. This change accounts for some age underestimates.

Peak integration and preheating

Use of a certain single temperature range for TL peak integration and computation of ED value or the dose-plateau plots is not yet fully justified. It is in the experimenter's choice to follow either procedure.

The choice of preheating can greatly reflect the shape and position of TL apparent peaks produced by the multi-grain samples. On the other hand it affects the OSL properties of the crystal too.

The underlying assumption of the existence of "uniform" TL emitters in the "single" TL peak is probably isolated in real samples. Such a polymineral fine-silt loess and density separates of 100-300 m quartz or feldspar grains from sands.

The preheating protocol is related to anomalous "fading" of feldspars, for TL and IRSL measurements.

Grain populations

The type and size of grains extracted from sediments is of critical significance to ED determination. As mentioned above, individual grains from

a sample have different intensities and luminescence emissions. Grun *et al.* (1989) extracted and measured single-grain glow curves of feldspars. They noticed a large variation in both intensities and in glow-curves shapes. About 10% of the fifty selected grains in this experiment, had TL intensities 5-10 times the intensities of the majority of grains. In such cases, one should question: what peak integration to use, how to choose preheating schedules, what use are kinetics-modeling computational exercises.

Thus, micro-TL/OSL or microluminescence studies should start up before further experimental results are accumulated.

Optical bleaching wavelengths

The natural light conditions that prevail during sediment deposition and the optical solar spectrum, which reaches the grains in waterlaid sediments, have a preferential bleaching on the light-sensitive electron traps. Therefore, the applied laboratory optical bleaching (SOL2 or sunlight) does not necessarily produce the same residual luminescence level as during deposition.

The practice is to use low energy light (550-700 nm) for a limited reduction in luminescence after laboratory bleaching. This procedure minimizes age overestimation.

Type of fitting function to growth curves

The exact form of a linear or saturating exponential fit to the dose growth curves from "partial bleach", "total bleach" or "regeneration" techniques, produce variable ED values, if the fitting parameters (exponents, least square test summed deviations) are not considered with care, and the experimental data points are a few.

Anomalous fading

The TL/OSL produced in volcanic or sedimentary feldspars by laboratory irradiation is unstable and decays. This presents a serious problem for dating because the short-lived unstable component is present only in the artificial (laboratory induced) TL/OSL, having mostly disappeared from the natural luminescence. Loess from Alaska (Berger, 1987) and waterlaid sediments from eastern Canada (Lamothe, 1984), the Gulf of Mexico and other sites in British Columbia (Berger *et al.*, 1987) have all exhibited 5-20% fading over 2-6 months.

In general, the occurrence or absence of fading in sediments has not commonly been reported.

Consequently, for fine-grained, polymineralic, feldspar-dominant samples, it is not known whether the detected fading represents a few grains that fade strongly, or whether all the grains have a short-term fading component.

Two general approaches to this perennial problem are available: utilization of non-fading minerals and exploitation of the luminescence behavior of the fading minerals.

Various attempts have been made to circumvent this problem, such as storage for a few days at elevated temperatures, related fading with feldspar compositions.

Quartz grains may be used, instead, due to the near absence of anomalous fading and the long mean lifetime of electron traps (ca. 10 million years for 325°C peak) (Wintle, 1977). However, it has been avoided, where possible, because of the onset of non-linearity in TL growth curves at relatively low doses (<200 Gy) compared to feldspars, and the resistance to optical bleaching that is shown by feldspars. The non-linearity can not be a problem if appropriate saturating exponential fitting (e.g. fit the data points approximately twice the actual ED value) is applied, while the 150-200 Gy dose region, for especially low-level dose-rate environments (e.g. limestone, sandy), can date sediments ~ 100 ka old.

Dose-rate accuracies

Several factors assumed may add up to a significant error in the corrected dose-rate value. Such factors could be, decay series U-disequilibrium (Radium-226 deficiency or excess, radon emanation, Th-230/U-234, U/Th), and mobility of such isotopes, water contents throughout the past. Inaccuracy in estimates of past pore-water content will be least (5 to 10%) for marine mud and greatest (30 to 50%) for terrestrial waterlaid sediments now above the water table. The effective past water content is usually <5% for coarse-grained Aeolian and beach deposits. Present practice is often to choose a mean estimate between the present in situ water and the saturation value.

CONCLUSION

The effectiveness of sunlight in reducing TL in sediments is now well known and forms the basis

for most of the dating applications discussed in the present review. With the objective of setting the available luminescence dates in a certain perspective various sedimentary deposits of geophysical, geological and archaeological significance have been dated and their measured luminescence properties discussed.

New applications of TL and OSL/IRSL dating (tephras, ceramics, sedimentary deposits) have been demonstrated in recent publications, and notable progress has been made in understanding the limitations to accuracy, as well as in the development of new techniques.

Only a few types of Quaternary deposits will presently provide reliable TL dates, and only if TL methods are applied prudently. Specifically, heated quartz in volcanic deposits and ceramics, and distal tephra can be dated reliably using the "additive dose" method.

Sample selection is less straightforward in unheated sediments. Suitable samples are fine (distal) loess, thin clayey layers in lacustrine deposits, clayey mud in off-shore or near-shore marine sediments, and organic-rich paleosols developed in loess, lacustrine, fluvial, or dune deposits. Dune sands are very promising, but greater reliability has yet to be demonstrated. For all such unheated sediments, the "partial bleach" method is the preferred and prudent technique to use.

In the above review the use of TL, OSL and IRSL measurements has been promoted as part of a combined dating technique of sediments and archaeological materials. Several factors have been considered related to accurate determina-

tion of ED for dating. Recent exploration on the sensitivity changes during exposure to light, with more applications on known age sedimentary deposits and testing, has been promising.

The OSL and IRSL dating of ceramic material seems to be well established, the dating of carved megalithic stone building needs more dating results for refinement, and the OSL, TL and IRSL of quartz and feldspar in sediments require more applications of known age samples.

However, by limiting the measurements to signals derived only from light-sensitive electron traps, the determination of equivalent doses (ED) by OSL has reduced the uncertainty in determining appropriate bleaching levels. The protocols themselves have steadily improved in precision, with uncertainties on average values of ED now routinely around 3%.

With these in mind the proposed luminescence dating techniques are expected soon to be set on a established basis, the time is auspicious for a far greater involvement of geologists, geophysicists and archaeometrists with these luminescence-dating tools.

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