

## ADVANCES IN THERMO- AND OPTO-LUMINESCENCE DATING OF ENVIRONMENTAL MATERIALS (Sedimentary Deposits). Part I: Techniques

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### ABSTRACT

This review refers to the state-of-the-art of the rapidly developed luminescence dating methods on environmental non-burnt materials formed mainly during the past quarter of a million years. It focuses in particular on the sedimentary deposits, including materials related to cultural activities. It also critically discusses the limitations and potential applications of these luminescence dating methods on solar set crystal clock.

Sample selection is less straightforward in unheated sediments. Suitable samples are: fine (distal) loess, thin clayey layers in lacustrine deposits, clayey muds in off-shore or near-shore marine sediments and organic-rich paleosols developed in loess, lacustrine, fluvial or dune deposits. For all such unheated sediments, the "partial bleach" method is the preferred and prudent technique to use.

The use of Thermoluminescence (TL), Optical Stimulated Luminescence (OSL) and Infra-Red Stimulated Luminescence (IRSL) measurements related to accurate determination of the equivalent dose (ED) for dating ( $\text{age} = \text{ED} / \text{annual dose-rate}$ ) has been promoted as part of a combined dating technique of sediments and archaeological materials. Recent single-aliquot protocols for accurate determination of ED conclude the considerable potential in many applications. Many, if not all, of the changes in OSL previously attributed to charge transfer may in fact be the result of changes in luminescence recombination probabilities (sensitivity).

Finally, tests on IRSL and quartz signal loss are investigated.

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

### INTRODUCTION

The determination of formation time of environmental materials, which are related to environmental events derived by natural or man-made

causes during the past quarter of a million years, is of profound importance for the study of many disciplines, e.g. human evolution, paleoenvironmental reconstruction, palaeoclimatology, pala-

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

eoecology, archaeology, to mention a few.

This paper is a critically assessed review of the much promised optical and thermoluminescence (TL) dating techniques of sedimentary deposits. The determination of the total dose by the traditional thermoluminescence (TL) method is complemented by the optical stimulated luminescence methods (OSL), involving green and infrared light for stimulation.

Several ways have been demonstrated for isolating and controlling several major sources of error, such as the type of TL/OSL instability, known as anomalous fading, as well as the effects of uncertainty about the degree of zeroing of the luminescence signal in certain depositional environments. Because of different sensitivities to light of the TL and OSL of quartz and feldspars, the latter have been shown to be the preferred mineral component for dating most unheated sediments.

A luminescence age is proportional to total ionizing radiation absorbed, divided by the annual dose-rate. The absorbed dose is estimated from luminescence measurements, whereas, the dose-rate is usually determined from radioactivity analysis. Here, particular emphasis will be placed on luminescence measurements.

For sediments, relatively less error generally arises from dose-rate determination than from luminescence measurements. Accuracies of better than 5 to 10% in the dose-rate estimates can now be achieved routinely for many sediments. But, inaccuracies of the same order in luminescence dating of sediments obeying certain criteria regarding zeroing of luminescence signal at deposition, are recently achieved for many sediments. This review includes greater detail in the description of the dating methodologies employed, discusses critically the principles on tests of mainly the OSL 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.

Only the sunbleached event or process is considered recorded in unheated detrital material. The central difficulty with these processes is that the effectiveness of solar resetting mineral clocks is highly variable. Among samples which are suitable materials for recording the last exposure to sun-

light are: oceanic and lacustrine sediments, loess, subaqueous silt clays, sand dune or beach deposits, glacial sedimentary deposits, dust incorporated in the ice of a glacier itself, fluvial and fluvio-glacial sediments, though the latter need further study (Fuller *et al.*, 1994; Duller, 1994b, c).

The mineralogy of polymineral samples has shown predominant minerals of K-feldspar, plagioclase and quartz. Despite different regions of origin, TL glow-curves from sediment are fairly similar. Because of its brightness, the TL from feldspar dominates over that of quartz, which in terms of quantity is usually more plentiful. The former is more rapidly bleached and it is, therefore, likely to be better set to zero; on the other hand there is the possibility of anomalous fading. It may, therefore, be desirable to measure the quartz signal separately after mineral separation (or by infrared shining of the polymineral sample, see below). However, with quartz from old samples there are complications due to its non-linear dose response.

The present review comprises of two parts. PART I describes the age equation, the dose-rate determination common for both TL and OSL dating of geoarchaeological materials, and the processes involved to solar resetting zero-age luminescence. PART II refers to various methods with their versions for the determination of the equivalent absorbed dose accumulated, characterizing the totally or partially sun bleached material, of the geological or archaeological event that took place in the past.

## **PART I LUMINESCENCE DATING EQUATION - GENERAL PRINCIPLES**

A useful crystal clock must:

- a) store "information" at a known received rate,
- b) behave as a closed system,
- c) have a useful range, and
- d) have a well-understood zeroing process.

For luminescence dating, the "information" stored is the number of electrons that migrate to traps (particular kinds of impurities and crystal defects) as a result of absorption of the energy of  $\alpha$ ,  $\beta$ ,  $\gamma$  and cosmic radiation by the crystal. The  $\alpha$ ,  $\beta$  and  $\gamma$  radiation arise mainly from the ambient U and Th isotopes and their decay products, as well as from  $^{40}\text{K}$ .

Luminescence is emitted during laboratory heat-

ing or shining, when the electrons are liberated from thermally sensitive (for TL) or optically sensitive (for OSL) traps and some recombine with opposite charges (positive holes) at luminescence centers (e.g. trace impurities).

This natural luminescence intensity of mineral is a measure of the past radiation absorbed dose and, when combined with an estimate of the past dose-rate and the sample's sensitivity to ionizing radiation, yields the time elapsed since the traps were last emptied.

The luminescence age,  $t$ , equals post-depositional accumulated absorbed dose (or palaeodose) divided by the dose-rate, eq. (1).

$$t = ED / (k d\alpha + d\beta + d\gamma + dc) \quad (1)$$

where, ED, the equivalent dose, is the laboratory  $\beta$  or  $\gamma$  dose that produces the same luminescence intensity as the palaeodose,  $d\alpha$  is the effective alpha particle dose-rate,  $k$ -factor is the TL sensitivity from betas/TL sensitivity from alphas, and the other components correspond to the  $\beta$ ,  $\gamma$  and cosmic ray dose-rate. The units are gray (Gy) for ED and gray  $\text{Ka}^{-1}$  for dose-rate, where 1 Gy = 1 Joule  $\text{kg}^{-1}$  = 100 rads.

The cosmic ray (mostly myons below 0.5 m soil depth) component  $dc$  is usually minor (<5% of the total dose-rate for many sediments) (Prescott and Stephan, 1982; Prescott and Hutton, 1994).

The closed system assumption can be violated in several ways, principally by draining of electrons from traps by agents, such as light (optical bleaching), heat, or even perhaps pressure and chemical changes (the effects of these last two agents have not been vigorously studied).

In a sense, the type of instability of TL or OSL, known as anomalous fading, which occurs in some feldspars, is a violation of the closed system assumption because electrons escape from traps. Loss or gain of K, U or Th (or their decay products) will also violate this assumption, changing the dose-rate in the process, although, excluding evident translocation of fine-grains, this is expected to be a problem only for U and its decay products because of the leachability of U and Ra and mobility of radon (Rn) gas.

The zeroing process of the luminescence clocks is not well understood when the event being dated is the last exposure to sunlight. The problem is that the non-zero luminescent residual in the grains at

deposition can be a highly variable fraction of the total luminescence intensity measured in the laboratory, depending on subtle conditions at deposition. Even for samples older than tens of thousands of years, this "inherited" residual or "starting-level" luminescence can correspond to a large fraction of the natural luminescence signal. In principle, the fraction that determine the amounts of illumination penetrating each grain at deposition are unique to each sample and cannot be determined exactly, but only by inference.

Nevertheless, several tests of this zeroing problem have been conducted and attention is given to this matter below.

## DOSE-RATE

The analytical techniques for determining the several components of the age equation can be quite elaborate, although by modern physical research standards the cost and complexity of instrumentation is low.

Regarding relevant laboratory practices and used apparatus for TL and OSL the information is given by Aitken (1985), Galloway (1991), Botter-Jensen and Duller (1992).

The principles and practices of dose-rate evaluation have been reviewed by Mejdahl and Wintle (1984) and Aitken (1985), therefore, only a brief overview is presented here.

The  $\alpha$ ,  $\beta$ ,  $\gamma$  dose-rate components are measured with various methods:

- a) High purity Ge gamma and alpha spectrometry (Murray and Aitken, 1982; Liritzis and Danalitsis, 1985; Saro and Pikna, 1987),
- b) neutron activation analysis,
- c) plastic scintillator (Galloway and Liritzis, 1991),
- d) Geiger - Muller (GM) proportional counter (Botter-Jensen and Mejdahl, 1985),
- e) alpha particle counting "pairs" technique (Huntley and Wintle, 1981; Liritzis and Galloway, 1982c), and potassium determination by XRF, atomic absorption or other nuclear or chemical techniques,
- f) TL dosimetry, employing phosphors e.g.  $\text{CaSO}_4:\text{Dy}$ ,  $\text{CaSO}_4:\text{Mn}$ ,  $\text{CaF}_2$ ,  $\text{LiF}$ ,  $\text{SiO}_2$  (Mejdahl, 1972; Liritzis and Galloway, 1980, 1982b, c; Liritzis, 1985; Michael *et al.*, 1984), and
- g) portable  $\gamma$ -scintillometer (Liritzis and Galloway, 1981).

The *in situ* dosimetry of (f) is not practical for sed-

iments but for Class I heated materials.

The relative uncertainty in dose-rate is kept well below 5%, but the effective final error can be between 5-7%. This depends upon assumptions and corrections made regarding:

- a) water content, which attenuates  $\alpha$ - and  $\beta$ -particles and  $\gamma$ -rays by factors of 1.50, 1.25 and 1.14 respectively (Zimmerman, 1971),
- b) U-series disequilibrium occurring mainly in the decay products of Th-230, Ra-226 and Rn escape (Kokkoris and Liritzis, 1997; Wintle, 1978; Wintle and Huntley, 1980),
- c) radon emanation,
- d) k-value (or the later introduced a-value and b-value, Aitken, 1985), and
- e) possible migration of radioisotopes.

In the case that U, Th, K and Rb concentrations are individually determined (usually Rb is computed from K-content), instead of obtaining direct dose-rates, conversion factors are used to calculate the dose-rate per ppm of element. Updated dose-rate conversion values are listed in Table 1 (Liritzis and Kokkoris, 1992; Liritzis *et al.*, 1998). The Rb content cannot be ignored, since together with K-feldspars inclusions, which inhere an internal beta-particle source from potassium-40, can provide an appreciable fraction of the

total dose-rate.

The grains used for luminescence dating are either of the fine (2-11  $\mu\text{m}$ ) or coarse (inclusions 100-200  $\mu\text{m}$ ) size. The  $\beta$ ,  $\gamma$ -radiations penetrate the inclusions and the  $\alpha$ ,  $\beta$ ,  $\gamma$  the fine grains. The average range of alpha radiation in unconsolidated sediment is about 20  $\mu\text{m}$  much less than the averages of 2 mm and 30 cm for  $\beta$  and  $\gamma$  radiations respectively (Liritzis, 1986). For example, for a uniform spread of grains between 5 and 10  $\mu\text{m}$ , the average reduction in the external  $\alpha$ -dose will be about 10% compared to that for negligibly small grains. But, because  $\alpha$ -particles are less effective than  $\beta$ - and  $\gamma$ -rays in producing luminescence, this 10% reduction may produce only 1 to 2% effect in the total dose and can often be ignored. For coarse grains, the attenuation of both the  $\alpha$ - and  $\beta$ - dose components becomes significant. In this case, for quartz and feldspar, the effect to external  $\alpha$ -irradiation is removed by HF acid etching. However, the reduction in the  $\beta$ -dose component through the coarse grain must be calculated (Mejdahl, 1979, 1983). For example, for beta dose an attenuation of 5-25% for 100-300  $\mu\text{m}$  radioactivity-free grains (e.g. most quartz) is calculated; this depends upon the source of the  $\beta$ -rays (partitioning of K, U and Th).

Table 1. Alpha-particle, beta-particle and gamma-ray dose rates for isotopic equilibrium, in mGy yr<sup>-1</sup> (1) per 1 ppm by weight of Oxide parent isotope (UO<sub>3</sub>, ThO<sub>2</sub> and Rb<sub>2</sub>O) and per 1% of K<sub>2</sub>O. For conversion from Element Oxide to ppm Concentration values, divide UO<sub>3</sub> with 0.8322, ThO<sub>2</sub> with 0.8788, Rb<sub>2</sub>O (%) with 0.9158 and K<sub>2</sub>O (to K) with 0.8301 (Liritzis and Kokkoris, 1992) (2).

Oxide of Parent Isotope	Alphas	Betas	Gammas
UO <sub>3</sub>	0.6423	0.0241	0.0444
ThO <sub>2</sub>	2.3564	0.1222	0.0922
K <sub>2</sub> O	0	0.6824	0.2042
Rb <sub>2</sub> O	0	0.000464	0

(1) The annual dose is calculated with the equation:

$$D[\text{mGy y}^{-1}] = 1.602 \times 10^{-10} [\text{mGy MeV}^{-1}] \times \lambda [\text{y}^{-1}] \times N \times E[\text{MeV}] \quad \lambda [\text{y}^{-1}] = \log_e 2 / \tau = 0.693147 / \tau$$

where  $\lambda$  = decay constant,  $\tau$  = half time,  $N$  = number of atoms of the nuclei.

For the U-series:

$$D[\text{mGy y}^{-1}] = D(^{238}\text{U for 1 ppm}) \times 238 / 237.9782 \times 99.274 \% + D(^{235}\text{U for 0.00711 ppm}) \times 0.83212$$

(2) Corrigenda: In the original publication (Liritzis and Kokkoris, 1992) the gamma dose-rates of 11.09 for UO<sub>3</sub> in their table 2 was overlooked, it was for U concentration values instead of UO<sub>3</sub>; thus it should be 9.22 instead of 11.09. As a result, in their table 4 the value of ( $\alpha + \beta + \gamma$ ) for UO<sub>3</sub> is 257.1 instead of 258.9. The Liritzis and Kokkoris (1992) dose-rates conversion tables, considering the above overlooking, are today valid. In comparison with other tables there are differences 1-2% due to slightly different values of isotopic data used by the authors. In conclusion there is a satisfactory agreement.

The HF acid etching assumes uniform removal of a surface layer. In fact, natural minerals are etched along defect channels and imperfections producing etch channels that penetrate the grains. Such anisotropic effect has not significant effect into calculations of the dose-rate in most cases. Large feldspar or quartz grains ( $>100\ \mu\text{m}$ ) in waterlaid sediments are associated only with deposits that have been transported in short distances or at depth, therefore, they are bound to possible restricted long exposure to sunlight, being left with an unknown substantial residual luminescence at deposition or zero-time.

### "ZERO-AGE" LUMINESCENCE BY SOLAR RESET-TING AND BLEACHING

Exposure to sunlight prior to deposition causes "bleaching" to near zero of previously acquired luminescence, and when the sunlight is cut off by later sediment being deposited on top, the luminescence re-accumulates in the same way as in pottery after firing. Ideally the luminescence signal should be zero for grains in a modern sedimentary deposit. For example, negligible signals for recently deposited grains have been found for quartz from a mixed mineral beach sand (Huntley *et al.*, 1985, 1991), modern intertidal sands (Godfrey-Smith *et al.*, 1988; Stokes and Rhodes, 1989), and aeolian sands (Stokes, 1992).

For a sample exposed to the sun, there is a certain probability per incident photon that an electron will be evicted from its optically sensitive trap and so the rate of eviction will be proportional both to this probability and to the number of trapped electrons, as well as to the intensity of the light (non-linear order from retrapping could be involved making the process more complex). This simple model leads to the expectation that the number of electrons remaining trapped at time,  $t$ , will be exponentially dependent on  $t$ , similar to the case of radioactive decay.

However, experimental evidence indicates that this is not the case and that as the 'bleaching' proceeds, the probability decreases (Fig. 1) (e.g., for calcites, Liritzis and Bakopoulos, 1997).

Thus, we have the concept of the 'easy-to-bleach' TL electron traps being removed first followed in turn by TL traps which is harder and harder to bleach, until eventually an unbleachable residual component is left. Thus, TL signals from feldspars and quartz (also, calcite) are never zero, even

after prolonged exposure to light, but reach an equilibrium level. The parameters required to remove all but this component depends upon the light exposure time, the intensity of the light exposure, the spectrum (wavelength) of the bleaching illumination, and the susceptibility to bleaching of the minerals present in the sample, which refers to the total trapped charge distribution within each crystal.

The rapidity of signal reduction of the optically sensitive traps by sunlight exposure for feldspar and quartz is considerable within the first 15-30 sec, during which time no loss of TL signal is seen for either mineral. The speed of the OSL signal reduction also enables the technique to be applied to mineral grains, which did not receive long light exposure prior to deposition, such as hill wash or fluvial sediments. IRSL dating studies on fine grained hill wash sediments have given much younger ages than TL measurements on the same sample discs (Wintle *et al.*, 1993a, b; Spooner *et al.*, 1990). However, complications can result from the presence of electrons in traps, which are bleached more slowly, e.g. those electrons that are responsible for part of the TL signal. Godfrey-Smith *et al.* (1988) have shown that when the light intensity was reduced to only 10% of the level on a sunny day, bleaching of the OSL signal from quartz proceeded 10 times more slowly.

Simple experiments designed to observe the results of bleaching by visible wavelengths of the TL from K-feldspars have demonstrated that the TL signal loss occurred at all visible wavelengths (Kronborg, 1983). He also showed that the TL signal from such grains could be bleached through 7 m clear water. Bleaching of TL signals in feldspars as the result of exposure to sunlight have shown the complexity of the processes taking place in crystals under natural illumination. In their IRSL studies Bailiff and Poolton (1991) demonstrated that three of their four feldspar specimen, which exposed to wavelengths of 450-1,000 nm using a lamp and monochromator, showed significant bleaching for wavelengths around 880 nm. Much greater signal loss, however, is recorded for bleaching with wavelengths of less than 600 nm. Exposure to wavelengths of less than 350 nm (violet) did not cause bleaching of the IRSL from feldspars, but resulted in an increase in the IRSL signal. This was interpreted as being the result of charge transfer from deeper traps into traps, which

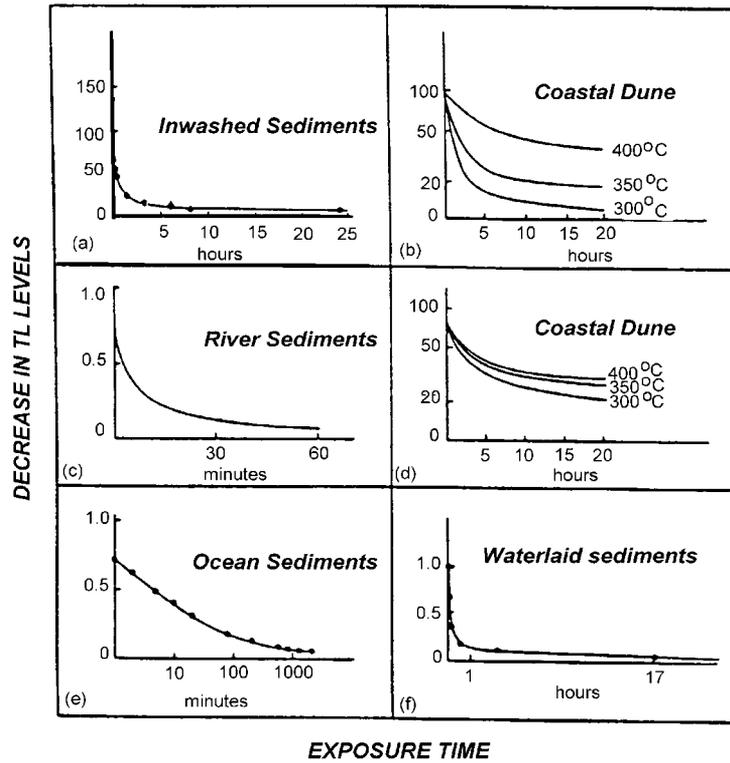


Figure 1. a) Bleaching with different lamps of TL in various sediments (adopted from Singhvi and Mejdahl, 1985).

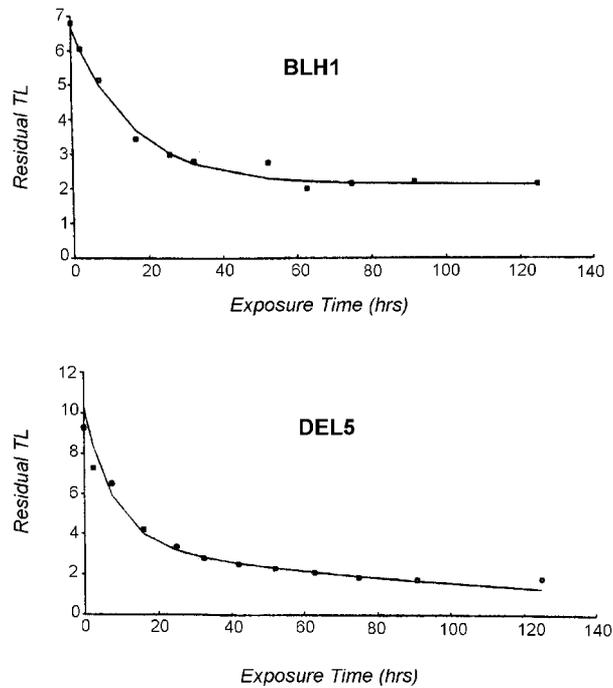


Figure 1. b) Residual curves of solar bleaching of TL for limestone (BLH1) and marble (DEL5) (Liritzis and Bakopoulos, 1997; Liritzis *et al.*, 1996).

take part in the IRSL process. This increase reached its peak in the ultraviolet at wavelengths of less than 300 nm. Wavelengths of less than 295 nm, although present in the solar spectrum, do not reach the Earth's surface because of absorption in the ozone layer. The ozone layer is concentrated in the stratosphere, about 25-28 km above the Earth's surface, and hence even the fine dust particles carried high in the atmosphere are not likely to have been exposed to these wavelengths. This means that even after sediments have been exposed to sunlight, it will still be possible to regenerate an IRSL signal by exposing the sediment to wavelengths <300 nm.

Ditlefsen (1992) has observed the reduction of both the TL and IRSL from K-feldspar grains after bleaching with a sunlamp filtered through a water column containing known concentrations of suspended sediment. The IRSL signals were reduced much more rapidly than the TL signals, but bleaching of the IRSL was slow for suspensions of greater than 0.05 g l<sup>-1</sup>, which are typical of proglacial rivers. This sediment load reduced the incident light intensity by x 10,000, indicating that much longer exposure times are required under these conditions to produce a well zeroed IRSL signal. Incomplete zeroing of IRSL for minerals in laminated sediments from a proglacial lake have been reported (Duller *et al.*, 1992). For sediments not deposited under direct sunlight, it is clear that thought must be given to the possibility of a non-zero IRSL level at deposition and a combined TL/IRSL approach has been suggested by Hutt and Jungner (1992). The production of an isochron (line of identical age) using different OSL signals from different minerals with different bleaching characteristics may be the best approach.

Lu Yanchou *et al.* (1987) and Gardner *et al.* (1987), however, have observed that almost all of the light-sensitive TL had been removed in the fine grains (mostly quartz) of scarp surface samples, yet the coarse-grain quartz from the same scarp surface samples retained a large fraction of their light-sensitive TL. None of the authors who have studied quartz from dunes suggested a cause for the observed ineffectiveness in the resetting of the TL for intensively insolated sands. One factor could be related to scattering and absorption of sunlight among a closely packed mixture of different minerals, another could be the TL of quartz can be insensitive to wavelengths greater than ca. 450 nm, or finally another factor may be the presence of surface

coatings on the grains (e.g. desert polish, iron staining) which are routinely removed by HF acid before TL studies. Amorphous iron oxide coatings such as goethite and limonite do not pass much visible light above about 450 nm (Hutt *et al.*, 1971; Townsend, 1987). Berger (1987) suggests that fine-grain clumping during transport, rather than pre-depositional iron staining, is the likely reason that a feldspar-dominated, "zero-age", eolian proximal silt retained a large, bleachable relict TL signal.

To summarize, sand-sized dune quartz and silt-sized proximal eolian feldspar grains can be screened from light before final burial, but the screening mechanisms probably differ and are poorly identified or understood.

Studies of the TL in waterborne sediments have suggested that the factors controlling the effectiveness of zeroing during water transport can be quite different from those outlined above for eolian grains. For example, with samples ranging from suspended river silts to marine mud, correct dose values could be obtained only when the partial bleach technique and long wavelengths of bleaching light (>550 nm) were used.

In cloudy or turbid water, the shorter wavelengths of visible light are relatively attenuated through absorption (primarily) and scattering by solid particles. Consequently, the ambient solar spectrum appears shifted toward the red, the effect being greater for more turbid water or water having a high organic content, such as estuarine waters. Clear water, in comparison, has only a small effect on the relative attenuation of different wavelengths in the region ca. 325 to 625 nm (Berger *et al.*, 1984; Jerlov, 1976; Huntley, 1985)

The practical question in evaluating palaeodose (ED) for all "solar set zero" dating purposes is whether, at deposition, the bleaching had been sufficient to remove all but this residual component. There are three states of bleached materials for palaeodose determination; the Partial Bleach, the Total Bleach and the Mixed Bleach. They can be approached by either the additive dose or the regeneration methods following the multiple or the single aliquot protocols.

For the total and mixed bleaching, the presumed circumstances of deposition are such as to ensure full removal of the bleachable component; more the TL is dominated by feldspar, the more is likely to be the case.

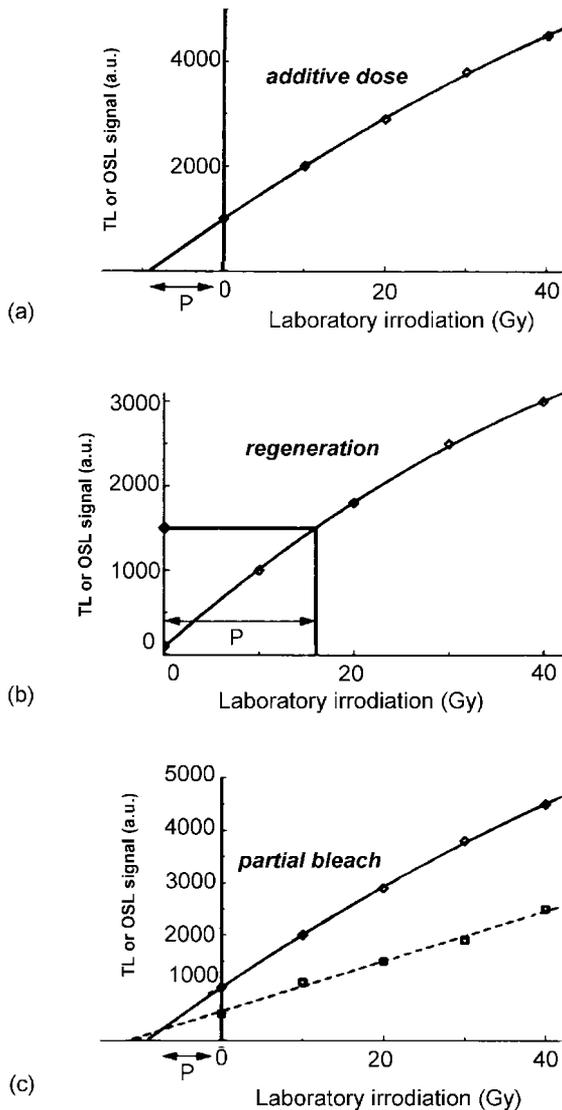


Figure 2. Diagrammatic representation of the three main TL or OSL luminescence methods used for equivalent dose (ED) or palaeodose determination: a) additive dose, b) regeneration, c) partial bleach (after Duller, 1996).

## PART II METHODS FOR THE DETERMINATION OF EQUIVALENT DOSE (ED) IN SUNBLEACHED SEDIMENTS BY TL TECHNIQUES TOTAL BLEACH METHOD Additive dose procedure

In this approach, the procedure is basically the same as used for pottery (Aitken, 1985; Liritzis and Galloway, 1982a; Michael *et al.*, 1997). A luminescence growth curve is constructed by

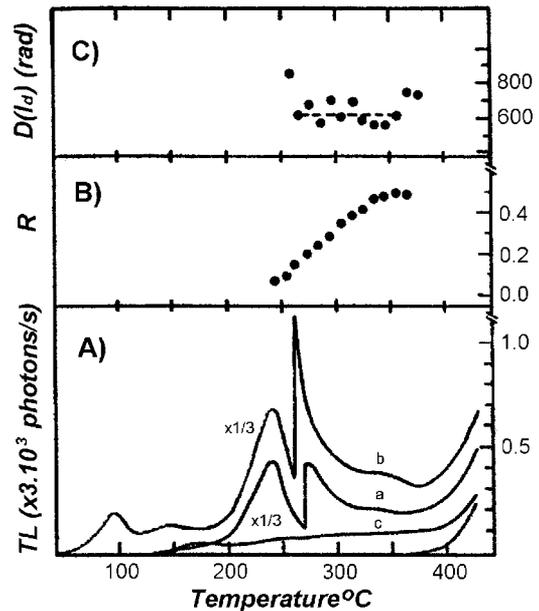


Figure 3. A plot of sediment ED versus glow curve temperature indicating plateau behavior. A) TL glow curves for Nehnar glacier dust sample in Kashmir, a) natural TL, b) natural + 6 Gy beta irradiation, c) natural + 1000 min sunlamp exposure. B) A plot of  $R = I_o / I_{nat}$ , where  $I_{nat}$  is the initial level of the sample and  $I_o$  the TL level reached after sunlamp exposure, versus glow curve temperature, and C) the plateau level (after Singhvi and Mejdahl, 1985).

adding doses to the natural dose ( $D_{nat}$ ), except that instead of reading off the equivalent dose (ED) from the intercept on the dose axis, it is read off from the intercept on a horizontal line through ( $R_o$ ) the latter being the level of TL remaining after a long laboratory bleaching (Fig. 2).

Although it can be established that the laboratory bleaching is long enough for only the unbleachable component to remain, there is the risk that the exposure to sunlight prior to deposition was not sufficient to reach this level.

A check of this is given by the "plateau test" in which the equivalent dose is plotted against glow-curve temperature, (Fig. 3). As illustrated in Fig. 3, the degree of bleaching is dependent on glow-curve temperature and if the bleachable component had not been fully removed at deposition there will not be a plateau. This approach was

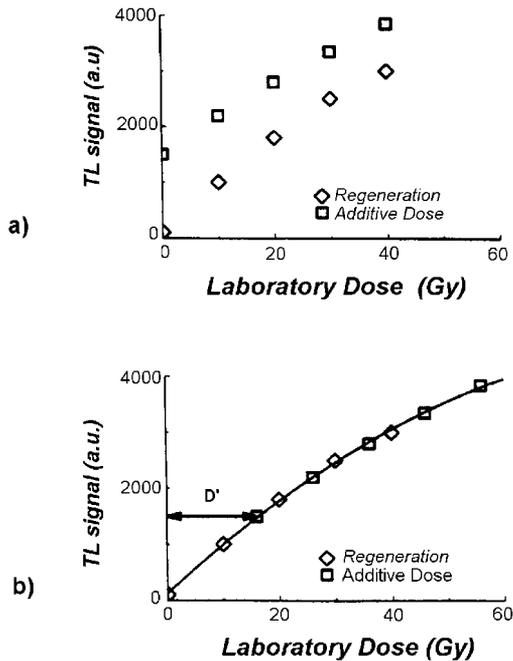


Figure 4. Diagrammatic representation of the slide procedure for ED determination: a) additive dose and regeneration datasets are collected, b) the additive dose data set is moved along the X-axis until it overlies the regeneration data set. The distance that the data have to be moved is equivalent to ED (after Duller, 1996).

developed by Singhvi *et al.*, (1982).

If  $R_{\text{nat}}$  is the natural TL level, then  $R = R_o/R_{\text{nat}}$  and  $R_{\text{nat}} - R_o = R_d$ , the TL due to the archaeological dose. The equivalent dose (ED) or  $D(R_d)$  can then be expressed simply as,

$$D(R_{\text{nat}}) = D(R_o) + D(R_d) \quad (2)$$

$$1 - R = (R_{\text{nat}} - R_o)/R_{\text{nat}} = R_d/R_{\text{nat}} \quad (3)$$

$$D(R_d) = (1 - R)D(R_{\text{nat}}) \quad (4)$$

with the pre-assumption of the proportionality between TL and dose.

Quantitatively speaking, the method is applicable to those materials, where sun exposure of a day or more can be assumed. In the laboratory, this bleaching is made for 10-15 hrs exposure with a solar simulator light source. For other materials,

overestimation of age due to overbleaching may occur.

The advantages of the method are:

- the method eliminates the problem of sensitivity change,
- the ED can be determined even for non-linear dose growth curve, provided that it is approximated by a known function e.g. a saturating exponential, and
- the experimental errors are somewhat less than those for partial bleach method, since the number of measurements are limited (see, below).

However, when the TL or multiple aliquot OSL techniques are employed, they both involve the preparation of many aliquots of the sample. This is inevitable, given the changes of sensitivity that occur upon heating during TL measurements. Each aliquot can only be measured once. Problems can arise concerning normalization between these different aliquots, and the large number that is required necessitates the processing of large amounts of sample. When the single aliquot OSL technique is used (Duller, 1991; Liritzis *et al.*, 1997a), one to two aliquots are only used.

In a novel application, this method has been extended to date the construction time of megalithic walls (Liritzis, 1994a, b). Here, it is reasonable to assume that the time required to cut and carve a large limestone, and place it in the wall, until another megalithic block overlies it, it is more than enough for the sun exposed surface to bleach down to a residual unbleachable TL level. This fast bleaching is not always the case, and the "dose-plateau" test of the partial bleach method is employed (Liritzis *et al.*, 1997b).

### Regeneration

In the regeneration technique suggested by Wintle and Huntley (1979) (Wintle and Proszynska, 1983), the sample gets a prolonged exposure to sunlight until the TL level is reduced to the unbleachable residual signal  $R_o$ . The TL growth curve is then regenerated by adding doses to the residual dose and the palaeodose ED is taken as that required to reproduce the natural level (Fig. 4).

It is important to verify that no change of TL sensitivity has occurred as a result of the bleaching, a rather critical point of the technique and practice has shown it occurs most of the times to a different extent for different glow curve temperatures. A great advantage of this method is that it can

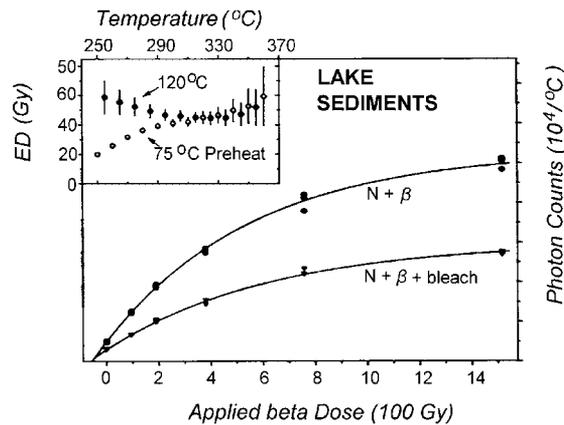


Figure 5. Dose-response curves and ED plateau plots for lake sediment sample, for bleaching of 3 days with low energy light to simulate light conditions in lake. The additive dose curves were made with preheat at 120 °C and on the TL peak of 310 °C. Two different preheatings were used, 75 °C and 120 °C. The ED was 40 Gy and this was also obtained for the dose-plateau test (inset) (after Berger and Easterbrook, 1993).

also be used for older samples where the natural TL is on the non-linear part of the growth curve. Smith (1983), however, introduced a regeneration method, which requires no optical bleaching for samples, showing post-bleach sensitivity change. In this method, the EDs for the older samples from a sediment section are obtained by comparing their natural TL levels with the (N+b) growth curve constructed with the youngest sample and by adding the ED of the youngest sample (Fig. 5). This ED determination holds under two assumptions: a) residual TL signals reached after solar bleaching in antiquity are similarly low, and b) the TL sensitivities of the samples are largely the same. Otherwise, the EDs are not correctly measured.

Recently, the application of the Single Aliquot Regeneration and Added Dose (SARA) (Mejdahl and Botter-Jensen, 1994), the Single-Aliquot Additive-Dose (Liritzis *et al.*, 1994; Murray and Roberts, 1997) and the Single-Aliquot Regenerative-Dose (SARD) (Murray and Roberts, 1998; Murray and Mejdahl, 1998) protocols to the estimation of ED, by optical stimulation methods, proved satisfactory.

### PARTIAL BLEACH METHOD

The partial bleach method was developed as a technique, which enables the TL level at the time of deposition to be derived, rather than assuming it to be that reached by an arbitrary laboratory

bleach. A detailed description of this has been reported by Huntley (1985). It assumed that the TL signal is made up of a number of components, each of which is characterized by a different sensitivity to light. This assumes that there is a number of electrons traps which are emptied by the equivalent - of a few minutes- exposure to direct sunlight and others, which require exposure time several orders of magnitude greater. The former might be expected to have been emptied during fluvial transport, but not the latter, unless they were on the surface of an exposed bar deposit.

### The regeneration-gamma (R-G) method

In this method (Wintle and Huntley, 1980), two sets of samples are irradiated with gamma or beta radiation to produce TL growth curves. Before the TL measurement, one of these sets is given a short sun or sunlamp bleach. In this way two dose growth curves; ( $I_{nat} + \text{gamma}$ ) and ( $I_{nat} + \text{gamma} + \text{sunlamp}$ ), with different slopes can be constructed. The sediment equivalent dose is then determined as the intersection between the two curves (Fig. 6). The basic philosophy is that the light exposure must be so short that only the TL level acquired since deposition is affected. This should, ideally, be checked for by constructing several ( $I_{nat} + \text{gamma} + \text{sunlamp}$ ) curves from the use of several irradiated sets and a range of short bleach times, and making sure that the intersection is the same. A con-

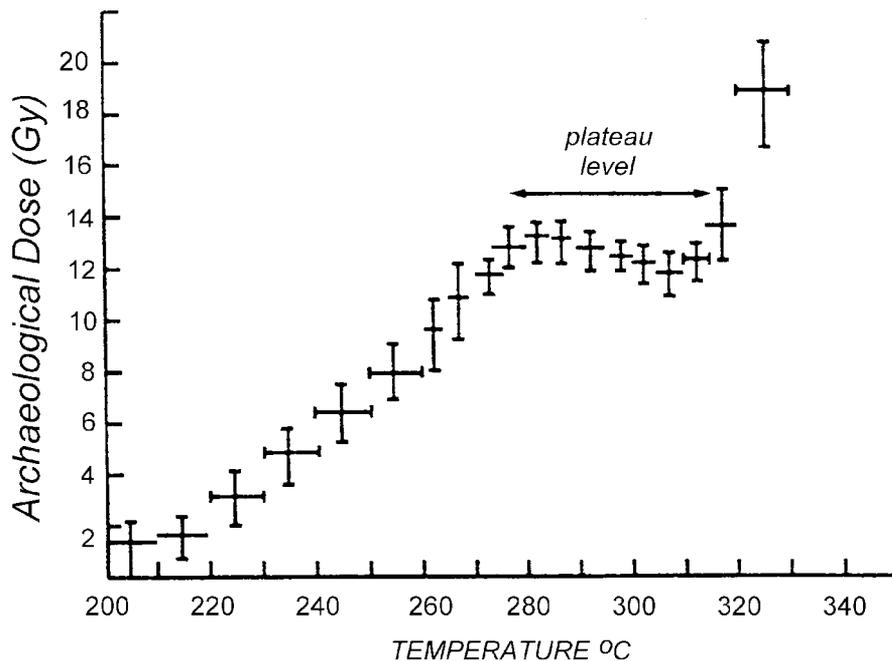


Figure 6. The "dose-plateau test" for the temperature region 200-330 °C, constructed per 10 °C intervals. The thermally stable electron traps correspond to the temperature interval that is 275-314 °C. This plateau ED value (in minutes of beta irradiation), is similar to that obtained from the extrapolation of the first build up curve (after Liritzis *et al.*, 1997b).

stant value of ED should then be obtained for a range of bleaching time shorter than the original bleaching. This procedure makes the method rather cumbersome.

The method can be applied to sediments that were exposed only very briefly to sunlight. It cannot be applied, however, to samples with non-linear TL dose growth curve because of obvious difficulties in extrapolation. Also, the errors are generally large, since the difference in slope between the two lines will usually be small. Furthermore, the underlying assumption that the bleaching is independent of dose may not always be fulfilled, in particular for samples obeying second order kinetics.

The method has been used preferentially for fine-grain samples and has produced a considerable number of useful results as will be detailed later (Wintle *et al.*, 1984; Huntley *et al.*, 1983).

Kronborg (1983) found that the R-G method did not yield satisfactory results for large grains feldspars from fluvial material, but Jungner (1985) obtained good results for large grain quartz and feldspar from sand dunes.

### Residual subtraction and "dose-plateau"

Several aliquots are sunexposed for various exposed times the residual TL is measured. One interesting aspect is the variation of residual TL shapes as a function of exposure time. The different parts of the TL glow-curves - TL intensity versus temperature- are not bleached in a similar manner, being affected variably by the different solar wavelengths (Fig. 7). Therefore, plots of residuals after exposure of the geological TL, as a function of temperature are made. The correct ancient TL residual level can be defined as follows: subtraction of residual shapes from the natural TL shape and its additive growth curve, and construction of 'dose-plateau' curves (plots of Archaeological Dose (AD) versus temperature). Due to the TL residual shape variation with bleaching time, the correct dose value from the 'dose-plateau' curves is for that sun exposure, which yields an AD 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 max-

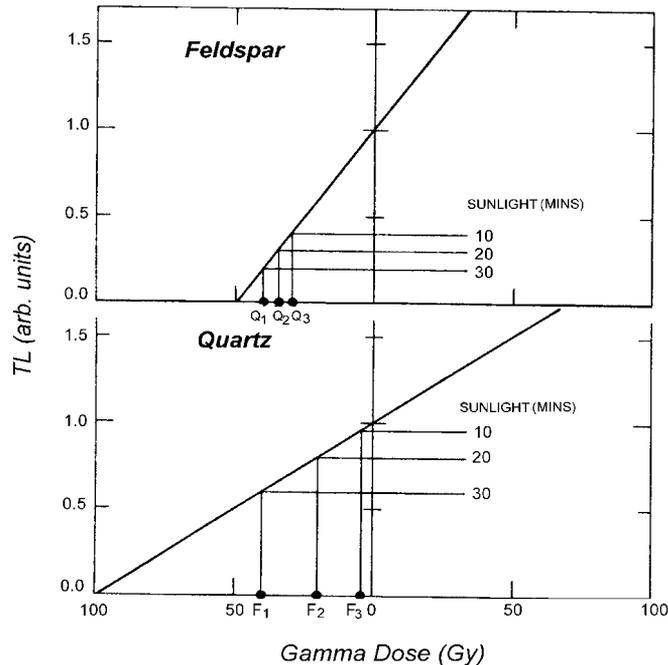


Figure 7. Principle of the quartz-feldspar method, for sun bleaches of 10, 20 and 30 min. The build up curves are intercepted with the residuals after bleaching to give corresponding sets of EDs,  $Q_1, Q_2, Q_3$  for quartz and  $F_1, F_2, F_3$  for feldspars. The correct sun exposure time taken as that producing a dose ratio equal to the dose-rate ratio. Then, the age is calculated from the feldspar ED (after Mejdahl, 1985).

imum length, too, (Fig. 6, Liritzis *et al.*, 1996, 1997b; Mejdahl, 1985, 1986, 1988; Wintle *et al.*, 1984).

It has been suggested for sediments (Mejdahl, 1986; Wintle *et al.*, 1984) and has been advanced and extended to calcites with successful results in the dating of megalithic wall in the classical Temple of Apollo in Delphi (Liritzis *et al.*, 1997b).

### The 'quartz-Feldspar' method

This method (Mejdahl, 1985) is still under development and uses large grains (0.1-0.3 mm) of quartz and potassium feldspar. Here, only natural samples are bleached; equivalent doses for quartz ( $Q_i$ ) and feldspar ( $F_i$ ) are then determined by intersection of the TL growth curves with horizontal lines (Fig. 7). The ratios  $F_i/Q_i$  are calculated and the correct sun exposure taken to be the one that produces a ratio  $r = F_i/Q_i$  equal to their dose-rate ratio of the grains ( $r > 1$  because of the inherent potassium contribution in feldspars).

The basis of the method is that quartz and K-feldspar have very different bleaching characteris-

tics; for sun exposure in air the TL level in K-feldspar is reduced about twice as fast as that in quartz (in temperature region 330-380 °C). This method has been tested with good results on recent (10-20 ka) aeolian sediments and shows promise for recent sediments, but so far it seems limited by the saturation level of quartz in 60-100 ka.

### Regeneration and 'dose-plateau' technique

In this case, various aliquots are exposed to light for different periods of time, and the growth curve is regenerated for each starting at their respective residual level. For each aliquot, the palaeodose is plotted as a function of glow curve temperature, and the correct residual level is taken as that producing the best plateau (Fig. 8).

The basis of the technique is that the shape of the curve obtained by plotting the residual TL as a function of glow curve temperature varies with bleaching time, a similar rationale to the section Residual subtraction and "dose-plateau" above.

As the accumulated dose must be the same for different temperatures, a palaeodose plateau is

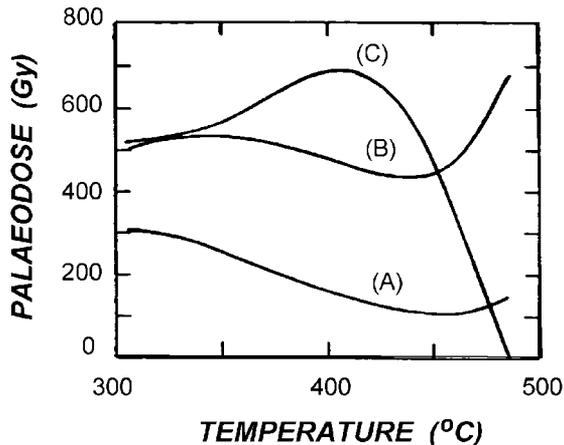


Figure 8. The regeneration technique applied to Na-feldspar from a partially bleached sediment from Sweden (R-821603). Palaeodose as a function of temperature for three different sunlight exposure times: a) 10 min, b) 20 min and c) 40 min. The preliminary results show that the true palaeodose is close to 500 Gy, where palaeodose values produce a plateau (after Mejdahl, 1986).

obtained only for that bleaching time which has reproduced the residual in this way present at the time of deposition. This method has been extensively used for dating fine grains from loess, but the fading effect limits the technique to about 150 ka. The growth curve in the 'additive dose' or 'regeneration' techniques is not always linear, but follows often a saturating exponential fit (first order kinetics) of the form  $I = I_s(1 - \exp[-C(D_e + D_a)])$ , or  $I = I_s[1 - \exp(-(D_a + D_e)/b)]$ , where  $I_s$  saturating intensity of TL or OSL,  $D_a$  the laboratory beta or gamma doses,  $D_e$  is the equivalent dose (ED) (the extrapolated intercept on the applied dose axis),  $b$  the exponent, and  $C$  is a constant term (Duller, 1994a, b; Liritzis *et al.*, 1997a).

However, in some cases several saturating exponential forms can be produced for a growth curve, with all varied parameters, providing different ED's. In such cases, the one with the smallest summed deviations for the least squares approach should be chosen. Nevertheless, although the rates of curvature of the TL build up curves are not usually known independently, a simple model can be used to justify extrapolations of additive dose growth curves and to obtain EDs (and  $b$ -values).

### OPTICALLY STIMULATED LUMINESCENCE (OSL) DATING

In 1985, a new promising technique, the optical dating, has been developed (Huntley *et al.*, 1985). In this technique, the palaeodose of quartz and feldspars of sediments is determined by photo-stimulated luminescence rather than TL, using different light sources and a broad band of the visible region including laser light (Ar, Kr, or dye lasers), a monochromatic beam from xenon spectrum and green (and infrared) light emitting diodes.

This radiation detraps electrons with far greater sensitivity than is possible with the conventional TL procedures mentioned earlier. Huntley *et al.* (1985) used green 514.5 nm line from an argon ion laser for stimulation and detected the OSL using a filter pack which had peak transmission at 400 nm. The possibility of stimulating luminescence with infrared photons was first demonstrated by Hutt *et al.* (1988), who observed IRSL at 400 nm for stimulation wavelengths of 800-1,000 nm.

Luminescence corresponding to the most light-sensitive traps is measured directly with the optical technique.

This optical dating technique is revolutionary, if only because it has the potential to provide rapid and automated read out of luminescence signals. Most of the optical luminescence work has been made employing green and infrared light on quartzes and feldspars.

With this technique, the two methods for ED determination described above, with their variants, can be realized in a very elegant and effective way and a quite large number of tests and applications have assessed the potential use of the optically stimulated luminescence technique.

The new luminescence readout technique involves a light source and suitable filters in front of the Photo Multiplier (PM) tube and in the incident light beam. This signal is distinct for different ages and for each sample decays rapidly within a few minutes (Fig. 9). All the other steps in the OSL dating process are similar to those used in conventional TL dating of unheated sediments.

Great care, however, is needed during sample handling to minimize sample exposure to laboratory lighting, even to red lights.

Furthermore, because this technique empties electrons from both thermally stable and thermally unstable traps at the same time, irradiated samples

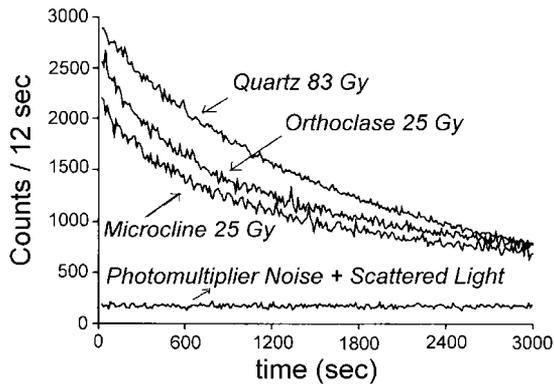


Figure 9. Time dependence of luminescence measured with the green Light Emission Diodes (LED) system for daylight-bleached quartz, orthoclase feldspar and microcline feldspar. Also shown is the system noise (after Galloway, 1994).

must be heated moderately before being illuminated by laser light, to remove any unstable signal. Various preheat treatments have been applied; for feldspars 10 min at 220 °C (Galloway, 1994; Li, 1991; Duller, 1991; Liritzis *et al.*, 1994, 1997a), or 140 °C for 16 hrs for fine-grained and 100 °C for one week for coarse-grained (Botter-Jensen *et al.*, 1991), or 140 °C for 16 hrs with a further 24 hr storage at room temperature (Li, 1992; Wintle *et al.*, 1993a), and for quartz 1 min at 200 °C (Galloway, 1994; Smith *et al.*, 1986; Liritzis *et al.*, 1994, 1997a) or 200 °C for 10 sec (Murray *et al.*, 1995). It should be pointed out that these short pre-heats will not overcome problems of anomalous fading, though monitoring of irradiated discs can be carried out over a period of months, with the same discs being used because of the 'non-destructive' nature of the measurements (Fig. 10).

The implication of this OSL phenomenon is that it can measure equivalent doses for minerals that have received short light exposures (minutes to hours) at deposition. The bleaching rate varies for the different minerals of quartz, feldspar, zircon, calcite, (Liritzis *et al.*, 1996; Morris and McKeever, 1993, 1994; McKeever and Morris, 1994; McKeever, 1991; Prescott and Fox, 1990).

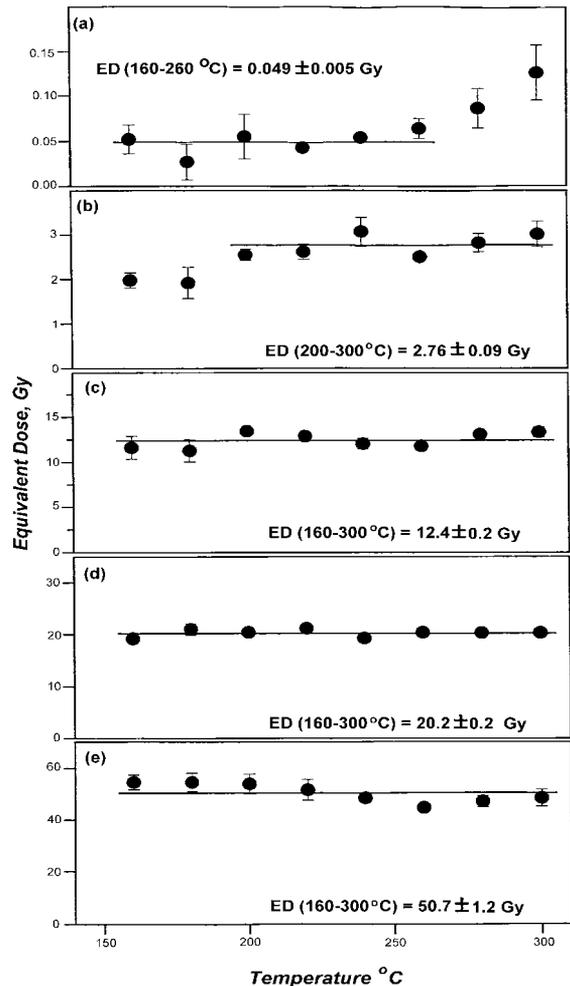


Figure 10. Preheat plateau for five samples, following the Regeneration dose for single aliquot, for different preheat temperatures (10 sec at each temperature). The mean (horizontal line) and standard error (bars) on each point based on three measurements of the ED are shown at each preheat temperature. The final ED was estimated for certain temperature range. During the preheat readings the 110 °C TL peak was measured and the OSL was normalized to this peak (after Murray and Roberts, 1998).

### ON THE OSL OF QUARTZ

Retrospective dosimetry using OSL on quartz extracted from e.g. bricks, needs to account for strong OSL sensitivity changes that are known to occur depending on the previous thermal treatment of the sample. In the area of luminescence

dating sedimentary quartz and quartz from heated archaeological samples show very different OSL sensitivities, the latter being in some cases orders of magnitude more sensitive (Aitken and Smith, 1988; Botter-Jensen and Duller, 1992; Botter-Jensen *et al.*, 1995). It has, also, been observed that the OSL versus dose curve is slightly supralinear for the annealed quartz, but linear for the unannealed samples, and that the dose response of the unannealed sample does not give zero OSL at zero added dose, in contrast to the annealed specimens. The non-zero luminescence observed at zero dose is believed to be due to incomplete emptying of the OSL traps during the bleaching phase, and/or recuperation of the OSL signal after the bleach (Aitken and Smith, 1988). In the annealed samples, however, all the levels are assumed to be emptied by the high temperature anneal. Certainly, it can be sure that the anneal has removed charge from most of the main OSL-producing traps, such as that responsible for the '325 °C' TL peak which correlates with the 514.5 nm OSL signal. Consequently, a zero OSL signal at zero dose is obtained (Botter-Jensen *et al.*, 1995). Fox (1990) has shown that this 325 °C TL peak in quartz emits light peaked at 380 nm. However, Prescott and Fox (1990) have found that for unannealed samples the 380 nm emission ridges outwards from more intense emission peaking at 480 nm; though after 40 sec bleaching with 350 nm light it has gone.

However, in general, annealing of quartz at temperatures up to 600 °C leads to dramatic alterations in the OSL sensitivity. Doses lower than 1 mGy have been measured by Green Light Stimulated Luminescence (GLSL) on fired quartz extracted from specimens in nature. Quartz has been used for GLSL dating as well as a dosimetric tool (Liritzis *et al.*, 1994; Liritzis, 1980; Botter-Jensen *et al.*, 1995).

Further, it has been observed that the TL sensitivity changes after bleaching and the residual level reached after long bleaching times depends upon the initial radiation dose.

Experimental observations indicate that the energies required to optically bleach the different TL peaks are not commensurable with the known thermal activation energies of these signals. If one assumes that TL bleaching occurs via a direct transition of charge from the trap into the delocalized band, then the expected ratio of optical-

to-thermal activation energies is estimated to be about 1.8 for quartz (Godfrey-Smith *et al.*, 1988). However, the observed ratio of optical-to-thermal activation energies for TL bleaching can differ greatly from this value. The onset of bleaching of the 100 °C peak, for example, occurs at 450 nm (Bailiff and Poolton, 1989). This corresponds to optical activation energy of 2.75 eV, and expected thermal activation energy of 1.5 eV. However, the observed thermal activation energy for the 100 °C TL peak is only 0.8 eV (Aitken, 1985; McKeever, 1985). Note that we distinguish between 'emptying of traps' and 'bleaching of a TL peak', which are not necessarily lead by identical mechanisms (Morris and McKeever, 1993).

Various models have been presented which concern the mechanisms of optical bleaching of TL signals from quartz, such as a) direct optical excitation of the trapped electrons, with associated retrapping, (Levy, 1982), b) direct optical excitation but with simultaneous filling of the trap with electrons from neutral recombination sites below the Fermi level, (Chen *et al.*, 1990), c) optically stimulated transfer of electrons from deep (thermally disconnected) traps, and not directly from the TL traps themselves, to the luminescent traps, including recombination of electrons and holes thereby reducing the recombination center concentration, (McKeever, 1991; Morris and McKeever, 1993, 1994). The optical transitions in question may or may not be thermally assisted (Aitken and Smith, 1988).

For the purpose of dating sediments the most important features related to the measurement of OSL and IRSL from quartz and feldspars are:

- a) portion of luminescence signal removal by sunlight,
- b) rate of bleaching by sunlight in different conditions of sediment deposition,
- c) luminescence signal sensitivity,
- d) luminescence signal stability, and
- e) correct method for determination of ED.

Prior to dating grains of a particular mineral, three characteristics need to be established: a suitable stimulation source, an observation spectral region away from the stimulation wavelengths, by choosing appropriate filters, and a reliable preheat procedure.

#### **DETERMINATION OF EQUIVALENT DOSE BY OSL**

There are two potential approaches to measuring

the ED; the multiple-aliquot and single-aliquot techniques. Although as the practice shows the single-aliquot techniques and associated protocols predominate in the applications, the multiple-aliquot techniques are occasionally used for comparison.

For the single-aliquot technique, two approaches are used: the additive-dose and the regenerative-dose. Murray *et al.*, (1997) demonstrated a workable single-aliquot additive-dose protocol for quartz. This was conceptually similar to the protocol proposed by Duller (1991) for feldspars, firstly used on archaeological quartz by Liritzis *et al.*, (1994), and later modified by Galloway (1996) (Liritzis *et al.*, 1997a; Galloway *et al.*, 1997).

The standard methods of equivalent dose (ED) determination for sedimentary material, the "Total bleach" and "Partial bleach" methods involve the preparation of many aliquots of the sample. Each aliquot can only be measured once, given the changes of sensitivity that occur upon heating during TL measurements. Problems arise, also, concerning normalization between these different aliquots, and the large number that is required necessitates the processing of large amounts of sample.

The use of OSL offers a less destructive method of sampling the trapped charge concentration and of determining the ED using a single aliquot (a single disc) of grains.

Although Southgate (1985) attempted to use TL measurements on single feldspar grains and Smith *et al.*, (1986), mention the use of a single aliquot of quartz or zircon, for ED determination using OSL, no final results or details are presented. Rhodes (1990) pursued the possibility, but considered the method impracticable due to change in sensitivity during laser shining.

Since then, single quartz grains of known-age aeolian sediment thoroughly bleached before deposition have been OSL measured with success (Murray and Roberts, 1997), and of feldspars from marine sands (Lamothe *et al.*, 1994).

Duller (1991) described the use of a "single aliquot" of potassium feldspars and the non-destructive nature of infrared stimulated luminescence (IRSL), without apparently changing the sensitivity of the material. Such an approach has been made practicable using the IRSL add-on to the  $R_{iso}$  automated TL reader. After measurement of the natural IRSL signal for 0.5 sec, the

sample disc can be irradiated and measured several times to produce an additive growth curve of IRSL against dose. He applied this approach for ED determination following the "additive-dose" and "regeneration" techniques on materials from New Zealand dune sands and Dutch cover sands. As there is part of the IRSL signal, which is thermally unstable, a pre-heat procedure is employed before each IRSL measurement. A two-ways correction for the effect of pre-heating was devised, relying upon different assumptions concerning the way in which charge is distributed in a crystal when it is irradiated. His account addressed various of the potential pitfalls in a single aliquot approach and illustrated the success of the method by comparing the ED with different methods.

Equivalent dose determinations using regeneration, additive dose multiple aliquots and single disc IRSL gave in general good agreement within  $\pm 8\%$  for two of them, while for the other sample the methods showed great scatter.

Although the regeneration method seem to have several drawbacks associated primarily with changes in sensitivity during complete bleaching, the additive dose method appears more promising. However, it is possible that regeneration methods may be developed in the future, provided that the sensitivity of the stimulated luminescence is not affected by laboratory bleaching. A similar approach has also been mentioned in respect of quartz and zircon when stimulation is provided by argon ion laser (Smith *et al.*, 1986).

However, Duller (1994a) has found that the "single aliquot" correction method, based upon adjusting the magnitude of the measured luminescence signal gives underestimates for those samples whose luminescence response to dose is non-linear, and have a large ( $>80$  Gy) ED, and proposes a second correction method based upon adjusting the value of the total added dose (rather than the luminescence signal) that the sample has received.

The agreement between the new correction method with the standard multiple aliquot method is far better over the range 0.1-400 Gy. The validity of this approach awaits more applications to samples from different geographical areas, compared to standard methods.

The attractive "single aliquot" method of equivalent dose determination for IRSL of feldspars

introduced by Duller (1991) requires in fact a minimum of two aliquots. This has further been developed by Galloway (1996) who describes analysis procedure requiring only one aliquot, i.e. it uses measurements with no added dose to determine the correction factors for the entire data set. This was done on the fact that the decay factors due to preheating can be represented by a single function dependent on only one parameter by applying a least squares fitting procedure of the form  $f(n) = 1 - \alpha \ln n$  (Fig. 11), where  $n$  is the number of measurements on the aliquot,  $f(n)$  is the function of initial signal remaining and  $\alpha$  is a parameter which will be dependent on the pre-heating temperature and duration, on the duration of IR exposure and on the mineralogy of the particular feldspar. This  $\alpha$ -value is  $0.23 \pm 0.01$ , and the scatter in the corrected luminescence values is about  $\pm 2\%$ .

Application of the three above correction methods have been applied to archaeological ceramics, and, at present, the reliable ED value is taken as the average between the last two methods.

Recently, progress in the determination of ED of quartz by OSL has been made for heated quartz and sun bleached sediments (Mejdahl and Botter-Jensen, 1994; Murray and Roberts, 1997).

In the Single-Aliquot/Regeneration Added dose (SARA) technique (Mejdahl and Botter-Jensen, 1994) two aliquots are used. An additional beta dose is given to one, and both aliquots are heated to some preheat temperature, if required (Mejdahl and Botter-Jensen used 260 °C for 40 sec), before optical stimulation to measure the natural signal ( $L_N$ ) and the natural and added-dose signal ( $L_{N+\beta}$ ). Each aliquot is then given a series of three sequential regeneration dose/preheat/stimulation cycles designed to give regeneration light levels ( $L_1, L_2, L_3$ ), which encompass the observed  $L_N$  and  $L_{N+\beta}$  values and allow the interpolation of the calculated doses,  $C_N$  and  $C_{N+\beta}$ . In general,  $C_N$  is not identical to equivalent dose, ED, because of sensitivity changes, but by plotting the two values of  $C$  against the two known doses, 0 and  $\beta$ , the true ED can be obtained from the intercept on the added dose axis. Murray (1996) later showed formally that this protocol should successfully allow for low-dose non-linearity, if the non-linearity can be approximated by a single constant exponent. This also confirmed the requirement that any sensitivity changes be independent of dose.

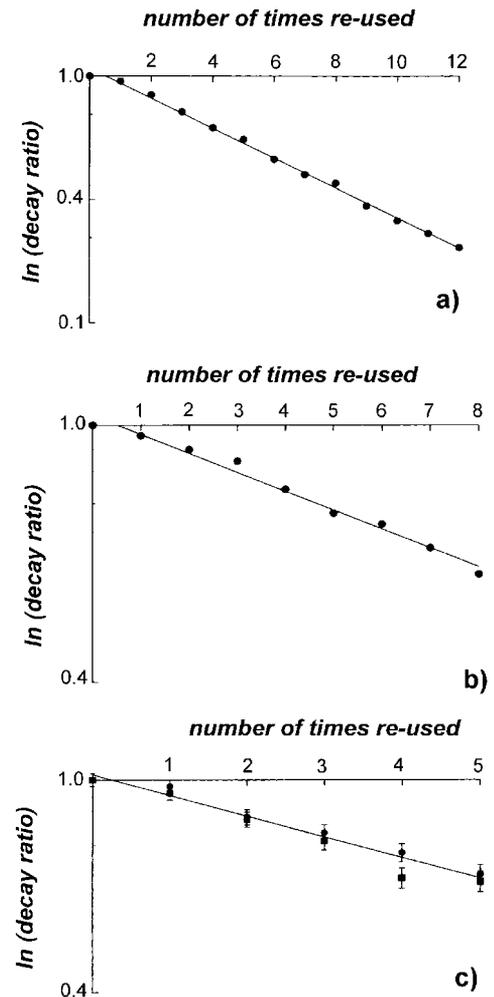


Figure 11. An example of the decay of OSL signal for quartz from ceramic sherds due to repeat preheat and reading cycle as used for correction of single aliquot measurements (6 to 13 points were used). a) for sample B42-1 with preheating at 220 °C for 1 min and 12.5 sec measuring time, b) sample B24-1 with preheating at 200 °C for 1 min and 100 sec measuring time, c) decay data for two aliquots of the same quartz B18-1. One aliquot was dosed and the decay ratios measured (circles), the decay ratios for the other aliquot were measured after making an additive dose set of measurements (squares). The line has the form,  $f(n) = 1 - \alpha \ln n$ . Here  $f(2) \sim 0.96$  is for quartz,  $n$  is the number of times used for an aliquot. Note that  $f(2) \sim 0.86$  for IR stimulation of feldspars (Galloway, 1996), (after Liritzis *et al.*, 1998).

In another version, the Single-Aliquot Regenerative-Dose (SARD) protocol, changes in sensitivity are accounted for with preheat temperature and regenerative cycle. Luminescence sensitivity changes are simultaneously monitored using the response of the 110 °C TL peak to a test dose.

Murray *et al.* (1997) and Murray and Roberts (1998) showed that the decay of OSL signals - resulting from repeated exposure to 0.1 sec of blue/green light at 110 °C, with a 10 sec preheat between stimulations- could be approximated by an exponential. Subsequently they developed an additive-dose single-aliquot protocol in which an aliquot was first preheated to some desired temperature, in a similar manner to Liritzis *et al.* (1997a). However, the observation that some samples could not be dated using the additive-dose single-aliquot protocol, because the decay of the OSL signal was not exponential, attributed to significant progressive sensitization of the luminescence recombination process during preheating, led to development of a regenerative-dose single-aliquot protocol (Murray and Roberts, 1998).

In a regenerative-dose approach, the natural OSL is measured, the sample is given a laboratory dose, and the OSL is measured again. The total OSL signal (rather than the initial signal) is measured during each cycle, so that there is essentially no blue/green light-sensitive trapped electron population prior to any laboratory dose. In the absence of sensitivity changes, ED can then be calculated from the ratio of the natural and regenerated OSL signals, plus a knowledge of the laboratory dose used to regenerate the OSL signal (Godfrey-Smith and Haskell, 1993; Olley *et al.*, 1998). Examination for sensitivity changes, usually present, is made observing the behavior of the OSL and 110 °C TL signals, with respect to both preheat regime and regenerative cycle. The use of the curve of the 110 °C TL peak area as a function of preheat temperature was made for sensitivity changes in both the natural and regenerated OSL signals resulting from different preheat temperatures - by dividing the OSL curve by the 110 °C TL sensitivity curve. Nevertheless, Murray and Roberts (1998) observed that the sensitivity-corrected estimates of ED are relatively independent of preheat temperature, which implies that thermal transfer of electrons is not important com-

pared to the prior population of electrons in the OSL trap.

They also showed that the preheat dependence of natural and regenerated OSL signals is very similar, after correction for sensitivity changes. This observation was independently supported by the wide preheat plateaus found in all the samples they examined.

The above indicate that now there is a significant amount of evidence, which suggests that charge transfer is a much less important phenomenon than previously thought.

Further, measurement of ED using single grains of quartz has been attempted, but the result of this method is not always reliable (Murray and Roberts, 1997).

### TESTS OF INFRARED STIMULATED LUMINESCENCE (IRSL) ON FELDSPARS

Feldspars, with a wide range of major element composition giving rise to different crystalline types, are ubiquitous in sedimentary environment. It is not surprising that their IRSL signals will be widely used for dating. We should restate that the aim of the work on IRSL of feldspars is the correct determination of the equivalent dose since deposition or last firing event in the past. For the former, problems arise for the incomplete bleaching of grains.

Luminescence dating of feldspar samples from sediments using IR light from a xenon lamp for stimulation was first reported by Hutt *et al.* (1988). IR stimulation has the great advantage that the stimulating and emitted light wavelengths can easily be separated, and the work was quickly followed by many laboratories (Godfrey-Smith *et al.*, 1988, using Krypton ion laser and emitting diodes; Poolton and Bailiff, 1989; Spooner *et al.*, 1990, using emitting diodes; Hutt and Jaek, 1989a, b; 1990, using IR diode laser).

A very useful characteristic of IR is the erasure of luminescence signal in feldspars leaving quartz luminescence signal and TL signal virtually unchanged, thus it is possible to measure OSL of quartz only using for example green light, and TL on the same samples. This practicable attraction initiated the construction of diode units fitted onto the automated TL readers (Botter-Jensen, 1988; Botter-Jensen *et al.*, 1991). In fact, a signal of several thousand counts can be acquired for potassium feldspars in the first 0.1 sec of IR expo-

sure; this leads to an excellent signal-to-noise ratio, and it has only a small (<1%) effect on the total IRSL signal.

A variety of IR emitting diodes (LED) are commercially available (e.g. GaAl/As IR emitter TEMPT 484 emitting typically 100 W Sr<sup>-1</sup> (100 mA) at 800±80 nm, the total power delivered to the sample 40 mW cm<sup>-2</sup> at a diode current 50 mA; or type TSUS5402, emission peak 950 nm approx. power at sample 50 mW cm<sup>-2</sup>, transmittance peak 370 nm, 1%, 290-490 nm).

The luminescence process of IRSL of feldspars involves the eviction of electrons from donor traps, charge transfer, through the conduction band, and recombination at acceptor sites, each of these processes is, in general, thermally dependent, and leads either to enhancement or quenching of the luminescence with increasing temperature. However, the exact mechanisms of OSL in feldspars (and quartz) are not fully understood, and moreover, the thermal processes in feldspars appear to be much more complex at elevated temperatures and an activation energy of 0.2 eV was determined. Botter-Jensen *et al.* (1994) showed that in the case of Na-rich material, the thermal activation of OSL is strongly dependent on the excitation energy used, ranging from 0.06 eV under 450 nm illumination, to 0.15 eV at 1000 nm. It has been suggested (Poolton *et al.*, 1994) that this is due to the relationship between the thermal enhancement of both charge eviction from the donor centers and enhanced mobility of electrons in the conduction band.

A variety of luminescence transitions were examined in a range of both alkali and plagioclase feldspars and three distinct emission types were identified which displayed very different behavior (Poolton *et al.*, 1995).

Thermal quenching of the luminescence arises at higher temperatures because of the increased probability of non-radiative transitions from the excited to ground states. This effect is very important in choosing a reliable trap-depth TL peak for dose assessment.

Poolton *et al.* (1995) have demonstrated that OSL can be observed from the red emission in feldspars, and opens the possibility of making the measurements at low temperatures to enhance sensitivity. Also, they indicated that the UV-induced Photoluminescence (PL) excitation efficiency of Mn<sup>2+</sup> is low compared with other lumi-

nescence centers, probably due to the fact that optical transitions from the ground <sup>6</sup>A<sub>1</sub> (s) to the excited states are, in fact, spin forbidden (White *et al.*, 1986).

Interesting is the PL emission spectra for plagioclase feldspar recorded before and after heating to 500 °C. The new emission band created by this heating process at around 560 nm is probably Mn<sup>2+</sup>, while the emission in the red is definitely confirmed to be Fe<sup>3+</sup>, by virtue of its characteristic stimulation spectrum. Luminescence features are shown also to retain irreversible changes on heating, especially between 250-400 °C.

In the combined IRSL and TL study by Botter-Jensen *et al.* (1991) several samples exhibited a strong supralinearity, while others had a completely linear growth. Doses as small as 2.5 Gy were easily measured. The saturation dose for OSL was about the same as for TL that is around 3000 Gy. The dating results on glacio-lacustrine varved sediments are in good agreement with the TL or geological estimates. The preheat procedure used for IRSL was 120 °C for one week.

Studies of the relationship between TL and IRSL of potassium feldspars have been undertaken by Li and Aitken (1989), Duller and Wintle (1991), who have shown that both 270 °C and 330 °C TL peaks are reduced by long exposure to IR.

Huntley *et al.* (1988, 1991) have shown that for potassium feldspar the IRSL emission spectra at room temperature and the emission spectra for the TL signal integrated from 290 to 320 °C are similar, and hence that similar recombination centers are being used. However, Duller and Botter-Jensen (1993) have suggested that there is no correlation between TL signal lost during exposure to IR radiation and the IRSL signal generated, concluding that the traps responsible for generating the TL and IRSL signals are almost entirely different.

The thermal stability of the traps giving rise to the IRSL was measured by Duller (1994c) following analysis of irradiated and preheated samples using pulse annealing. It was observed that no IRSL was lost till a temperature of ~220 °C. His data analysis suggested that the traps containing charge, giving rise to the IRSL signal at room temperature (NB: the intensity of the IRSL from feldspars is affected by the temperature of the sample) and the high temperature (>250 °C) TL signal are similar, implying similar recombination centers involved.

There was no clear relationship between the low temperature TL signal and any part of the IRSL signal (Fig. 12). It appears though, that increasing the sample temperature causes the transfer of charge from non-IR sensitive to IR-sensitive traps. This becomes especially apparent when the sample has been bleached using IR at room temperature. When employing green stimulated light (GLSL) on feldspars, too, it was found that a large fraction of the GLSL and IRSL signals originate from common traps (Duller and Botter-Jensen, 1993).

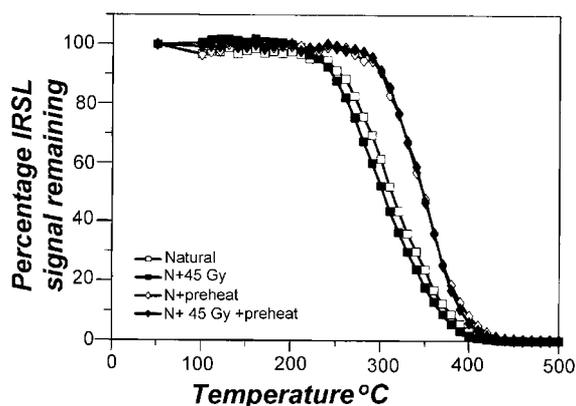


Figure 12. Pulse annealing for aliquots of dune sand sediment from New Zealand. Data have been plotted by calculating the percentage of the IRSL signal remaining after annealing at a given temperature. Aliquots have been treated as follows: Natural = natural dose; N + 45 Gy = natural plus 45 Gy beta irradiation; N + ph = natural preheated at 220 °C for 10 min; N + 45 Gy + ph = natural plus 45 Gy beta irradiation, and then preheated at 220 °C for 10 min (after Duller, 1994c).

### FADING OF IRSL OF FELDSPARS

Moreover, particular attention in the anomalous fading of IRSL from feldspars has been made. A variety of behaviors indicative of both athermal and thermal accelerable mechanisms were observed. The observation of apparently athermal anomalous fading casts doubt on the validity of the optimistic application of elevated temperature pre-fading treatments to the feldspar minerals. Subsequent to the work of Templer (1986) in which the localized transition model was found to

describe the anomalous fading seen in zircons, some studies on feldspathic materials (Clark and Templer, 1988) lent support to the elevated temperature approach for eliminating anomalous fading. Those findings, however, were not for 'pure' feldspar samples, but for polymineral fine grains. Measurements by Spooner (1994) suggested the anomalous fading of IRSL signal originated from one defect type only in all feldspars, a suite of 24 feldspar samples, assembled as representative of the major compositional regimes of the feldspar ternary diagram.

Anomalous fading has been detected in IRSL and OSL emissions from feldspar species of most significance to luminescence dating. Tunneling-type fading is shown to be the dominant mechanism, which necessitates monitoring.

A suitable anomalous fading monitoring procedure is the repeated short exposure to portions of a sample stored for several orders of magnitude of time at 10 and 100 °C; the resulting data should be plotted on a logarithmic time-scale of the form  $R = a + b \cdot \ln(\text{storage time})$ , where  $a$ ,  $b$  are parameters for the best fit equation to the remanent luminescence curves.

Alternatively, samples can be inspected for low-temperature fading phosphorescence immediately after laboratory irradiation.

The remanent IRSL and OSL for representative feldspars after 2-months storage at 10 °C showed a mixed stability behavior; stable, fading, intermediate (Spooner, 1994).

Non-fading behavior was observed only from examples of end-member (or near-end-member) species of the ternary system. Severe fading exhibited the high K-feldspars. In general, the implications from correlations of anomalous fading with composition (i.e. increased fading with increased calcium in the lattice, Akber and Prescott, 1985) and rapid cooling (i.e. fading from volcanic material, Wintle, 1974) are that the more disordered the lattice, the greater the proclivity to exhibit anomalous fading.

Huntley *et al.* (1988) demonstrated the use of a sensitive TL spectrometer for classification of emission spectra from twelve feldspars of various composition, seeking potential associations between degree of fading, emission spectra and feldspar character.

Detection of significant anomalous fading must be considered as a prerequisite criterion for sample rejection in sediment dating.

## 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 applying various techniques and their measured luminescence properties discussed.

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.

For all such unheated sediments, the "partial bleach" method is the preferred and prudent technique to use.

Several factors have been considered related to accurate determination 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.

By limiting the measurements to signals derived only from light-sensitive electron traps, however, the determination of equivalent doses (ED) by OSL has reduced the uncertainty in determining appropriate bleaching levels. The protocols themselves, esteemed by the single aliquot technique, 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, and a far greater involvement of geologists, geophysicists and archaeometrists with these luminescence-dating tools.

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