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### **Title**

Residual thermoluminescence for sun-bleached quartz: dependence on pre-exposure radiation dose.

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#### Abstract

Quartz grains zeroed by heat and subsequently irradiated - either in the laboratory, or naturally in volcanic deposits - were exposed to sunlight for several months prior to TL measurements. As a result, the residual TL, either Blue or Red, appeared to grow significantly with the radiation-dose experienced by the grains since zeroing. In another experiment, un-heated sedimentary quartz grains, sampled at the very surface of a coastal sand-dune, were irradiated and subsequently exposed to sunlight before TL measurements. The residual Red-TL still exhibited a strong dose-dependence, but at a level which was higher than the natural signal. Finally, it was concluded that the so-called “un-bleachable TL” is not a constant, intrinsic data, for a given quartz sample.

Because the residual TL is strongly correlated to the radiation dose experienced since the last zeroing by heat, it can theoretically be quoted in terms of “dose”, as sometimes done. However, this raises the question of its correct evaluation, and, on the other hand, there is a risk of misinterpretation of its physical meaning in the case of un-heated sediments.

In the context of sediment dating, the experimentally-estimated residual signal that is routinely subtracted from the total signal for palaeodose estimation is therefore higher than the signal that would have been measured prior to burial. Such effect induces an error which can be significant for Red TL. It can be corrected for, at least roughly, from experimental exploration of the dose-dependence of the residual TL.

Key-words : quartz, thermoluminescence , sun-bleaching, residual TL.

## 1 Introduction

For decades it has been observed that the TL of sediments increased with burial depth. This has early tentatively been used for chronological purpose (e.g., Morozov, 1968, Shelkopyas, 1971). However, TL was firmly assessed to be usable for dating sediments only when Wintle and Huntley (1979, 1980) realised that sun exposure was responsible of the weak TL of recent sediments.

Actually, un-heated sediments can be dated on the basis of the principles developed for heated mineral samples (e.g., Aitken, 1985), but for the restriction that the initial potential TL was not nil: numerous experiments have shown that sun exposure is not able to erase TL completely, leaving always an "un-bleachable" component or "residual TL". Erasing kinetics and residual TL depend on several factors including: nature of the minerals, exposure conditions and duration, light spectrum (e.g., Gemmel, 1985, Jungner, 1988, Spooner et al., 1988, Rendell et al., 1994). Recently, Westaway and Roberts (2006), using a regenerative-dose protocol, assessed that the residual Red-TL of quartz preliminary annealed by heating was also correlated to the radiation dose acquired before bleaching.

Because the residual TL has to be subtracted from the total measured TL for estimating the dose acquired since deposition (palaeodose,  $P$ , hereafter), its correct evaluation is a critical issue in sediment dating. However, the conditions of deposition can hardly be inferred, so that it is uneasy to assess precisely the amount of bleaching that had been attained when the sediment to be dated was overlapped by a layer of fresh sediment.

A typical bleaching curve (i.e., TL vs sun-exposure duration) will show an initial rapid fall (within a few hours for the "Blue-TL" or a few days for the "Red-TL"), followed by a more slowly decreasing curve (e.g., Aitken, 1985, Fig. 8.2), where the TL tends to stabilization. In practice, the residual level, experimentally evaluated by means of a "long laboratory bleaching" (i.e., several hours under a sun simulator) is generally taken as the initial signal to

be subtracted assuming that (i) the duration of initial exposure was long enough so that the bleachable TL was effectively bleached and (ii) the ratio [total TL]/[residual TL] is high enough so that errors on the residual are of negligible consequences on the palaeodose evaluation. Derived approaches, like the Partial Bleach Method" (Wintle and Huntley, 1980) and its refinements (Mejdahl, 1988) were proposed for experimental search of the initial level that was actually attained in the case of partial bleach.

Alternatively, a method, which is sometimes feasible, consists in estimating the residual from "zero-age" samples from the same origin, that is, for example, samples taken at the very surface of a sand dune (e.g., Singhvi et al, 1983).

In this context, the present work was devoted to further explore the correlation between the preliminary experienced radiation dose and the residual TL in the frame of the multiple-aliquot additive-dose technique. It was initiated for two reasons: (i) we wanted to test the assumption of Westaway and Roberts (2006) that the un-bleachable Red-TL could be linear vs dose. And, (ii), we were puzzled by the quoting of the residual in terms of "dose" in some publications (e.g., Singhvi et al., 1983, Lai et al., 2006) and we wanted to clarify this point.

## 2 Experimental bases

Two different heated quartz samples from French Massif-Central, differing by their respective ages, were investigated. Sample C397 was extracted from a sediment baked by a lava flow around 40 ka ago and sample C437 was extracted from volcanic lapilli deposited around 15 ka ago (both from stratigraphical consideration). Additionally, we studied quartz from modern sediment sampled at the top surface of a coastal sand-dune at Le Pyla (Atlantic sea shore, France).

Quartz grains were prepared by the usual techniques (e.g., Aitken, 1985) in the grain size range 200-315  $\mu\text{m}$  (C397 was re-crushed from natural 850-1000  $\mu\text{m}$ ). Irradiations were routinely performed several weeks before measurements, by means of a  $^{137}\text{Cs}$  source delivering 39.5 mGy/s, on batches of around 100 mg. TL was measured using a home-made TL set, at 5.0°C/s, either in the blue or in the red region of the spectrum. Red TL was measured through a red glass filter (RG610 Shott), by a bi-alkali EMI 9635Qa PM tube. Such combination results in a maximum efficiency at around 620 nm (Miallier et al., 1991) and the ratio signal/noise allows measuring palaeodoses as low as 11 Gy for quartz (Montret et al., 1992). Blue TL was measured, using the same PM tube, through a BG12 filter (maximum around 400 nm).

Light exposure was made outdoors. Sun was preferred to laboratory solar simulator for making sure that all the samples exposed at a time, individually spread in 50 mm diameter Perspex boxes displayed on a  $\sim 0.25 \text{ m}^2$  area, protected from dust and hail by a sheet of 3mm ordinary glass, received exactly the same flux. Evidently, the light spectrum at the level of the samples was missing some UV light absorbed by glass; however, it was assumed that this could affect the kinetics of bleaching only (a little less rapid), and not the final residual TL.

### 3 Experiments

In a first step, measurements and data processing, for a given heated sample, were conducted the same way as for routine dating. This includes:

- construction of an additive dose-response-curve (ADRC) using the TL of aliquots that were previously irradiated at different doses.
- construction of a regenerated dose-response-curve (RDRC) with the TL of aliquots of fresh quartz (i.e., natural, not yet measured) irradiated after annealing in the oven (380°C/16h).

- evaluation of the equivalent palaeodose, P, using the slide technique LISA (Sanzelle et al., 1996).

Meanwhile, parts of each of the same quartz extracts (i.e., pre-irradiated, including natural + dose and annealed + dose), not yet measured, were exposed to sun for one month (during April and May). After this interval, they were also TL-measured. The TL-decay vs exposure-duration was not monitored, assuming that it had the same general features as usual, and particularly that the residual Red-TL was quite stabilized after one month exposure (Miallier et al., 1994).

The sand-dune sample from Le Pyla, was simply irradiated at increasing doses for simulating the natural irradiation of a sediment sample, which would be definitively buried after its last stay at daylight (i.e., the initial event to be dated). Then, some of the irradiated sample was exposed to sun for two months (during September and October) for checking at which extent the initial level can be recovered by experimental bleaching.

## 4 Results

### 4.1 Sample C437

The unexposed sample behaved like a "standard" young sample as concerns Red-TL for the shape of the glow-curves (Fig.1 A-B) and for the dose-response curves (Fig.2 A.) (see e.g., Montret et al., 1992, Pilleyre et al., 1992), resulting in  $P = 50.8 \pm 2.0$  ( $1\sigma$ ) Gy. Allowing for annual dose assessment (not developed here) this gives an age of  $14 \pm 1$  ka, in good agreement with the predicted age.

The sun-exposed samples presented glow-curves that were quite similar to unexposed ones, but at a lower scale (Fig.1 C-D). They did not show a constant level: the residual Red-TL was significantly increasing with the radiation dose experienced prior to sun exposure (Fig. 2 A).

Since the residual can be completely zeroed by heat, it was assumed that the dose-response-curves for the residual Red-TL would allow evaluation of the palaeodose P (still using the slide method LISA). It resulted in a value of  $60 \pm 4$  Gy, higher than the one estimated using the unexposed samples. The growth curves for the exposed and un-exposed samples exhibited shapes that were nearly similar in shape to each other, as well for laboratory-annealed as for non annealed samples (Fig.2 C). The average ratio exposed/unexposed was  $0.33 \pm 0.01$ , for the RDRC and  $0.42 \pm 0.02$  for the ADRC (Fig. 2C). Such similarity in shape for the dose-response-curves suggests that the number of residual trapped charges is, at first approximation, a given fraction of the trapped charges before sun-exposure. However, the fact that the percentage of bleaching was not the same for ADRC and RDRC was a preliminary indication that the phenomenon is not so simple.

For Blue-TL, only the natural signal was measured for the unexposed quartz grains. The exposed quartz grains were measured both for additive and regenerated samples. As a result, the natural TL fell to ~14% of its un-exposed value. Similarly to Red-TL, the residual Blue-TL was significantly increasing with dose, resulting in growing dose-response-curves both for ADRC and RDRC (Fig.3). The growth-rate of the dose-response-curves for the exposed samples is quite comparable for the Blue and Red-TL: for example, the ratio between the highest signal (N + 130 Gy) and the natural was ~ 2.5 for Red and ~ 2.9 for Blue respectively. It can then be concluded that similar mechanisms are involved both for Red and Blue TL.

#### 4.2 Sample C397

The Red-TL from the older heated quartz, C397, presented the same behaviour as for the younger one, C437 (Fig.1 B and D). Both the exposed ADRC and RDRC still show a strong correlation between residual level and radiation dose. The ratio between the exposed and



unexposed TL signals was nearly stable for the ADRC. It was increasing with dose for the RDRC (Fig.2 D), thus indicating a significant change in the shape of the dose response curve. It was not possible to derive P from the additive dose-response-curves because they differed in shape from the regenerated dose-response-curves so that the latter were of no help in reconstructing the ADRC by means of a slide technique. The above observations mean that the physical phenomena that are involved are becoming more complex with increasing dose and age.

#### 4.3 Quartz from Le Pyla coastal dune

The glow-curves and bleaching kinetics of this sample have already been shown (Miallier et al., 1994). In the present work, the initial level (i.e., as sampled) proved to be prone to additional loss of signal by around 8 % ( $\pm 5\%$ ) after further sun bleaching: Fig.4 B shows that the zero-dose ratio is slightly lower than unity (see also Fig. 4 A). However, the residual Red-TL finally increased with dose, just like for the annealed samples. When taking the natural TL signal - i.e. the actual zero-level for this sample if it would happen to be buried now - as a reference level, it can be verified that the residual Red TL for the sample that has received 300 Gy is significantly higher than the initial level ( $\Delta$  on Fig.4).

### 5 Discussion

#### 5-1 Correlation between residual TL and radiation dose.

For heated quartz samples - either naturally or in the laboratory - the "un-bleachable" component of TL appears to be not an intrinsic constant data as implicitly assumed in the routine procedures of sediment dating. It is strongly correlated to the radiation dose experienced prior to bleaching. Such correlation is shown by dose-response-curves, which are more or less similar to the ones for unbleached samples at low doses and tend to be different

when the dose is increased. These observations would incite one to suppose that, for some physical reason, a fraction of the initially trapped charges still remains trapped after sun-exposure. However, the experiments with the unheated quartz (Pyla sample) have suggested that the initial residual level (after natural bleach) could actually be the base-level of the further dose-induced growth of the residual TL derived from the experimental sun-exposure. Then, as a preliminary physical explanation, it can tentatively be assessed that two different trap families are able to provide released charges to the luminescent centres. One is supposedly light-sensitive and the other is not, both being zeroed by heat. Like the bleachable traps, the non-bleachable traps will be filled during irradiation - and possibly during sun-exposure by charges released by bleaching. Filling of these traps will be integrated on the whole "life" of the sample, including bleaching episodes, as long as no heating occurs, thus explaining the high residual for the Pyla samples.

Because the un-bleachable traps are not the same as the bleachable traps, it is logical that their dose-growth is not the same either, resulting in dose-response-curves that can be different in shape (Fig.2). For the same reason, the variation of the trap population with annealing is also different, resulting in different proportions exposed/un-exposed for TL between natural and laboratory-annealed samples (Fig.2). A similar explanation would go both for Blue and Red TL, but with distinct traps, inducing different number of traps and, finally, different apparent light-sensitivities.

The residual TL can theoretically be expressed as radiation-dose-equivalent, because it correlates to the dose received since the last zeroing by heat. The issue is in interpreting correctly its physical significance and estimating correctly its value. At least, it will be necessary to scale the residual TL with comparable signals, that is: signals from samples bleached at the same amount, after irradiation. When not doing so like, e.g, Singhvi et al.

(1983), Lai et al. (2006), quoting the measured residual as a “dose” is not relevant. In this case, the residual can only be quoted for its intensity or, alternatively, as a percentage of the natural signal.

In the course of the present work, we had the hope that, according to the assumption of Westaway and Roberts (2006), the residual after-bleach TL for heated quartz could have a simple, linear response to dose, even in the case that the TL is approaching saturation (see Fig. A1 in Westaway and Roberts). This could have been a way for overcoming the difficulties in estimating the dose  $P$ , acquired since initial heating, for samples at the onset of saturation, e.g., in the context of ancient volcanoes. In turn, two aspects of the prediction were verified : (i) the un-bleachable TL is a growing function of radiation-dose and (ii) the corresponding dose-response-curve has not always the same shape as the one for the bleachable signal. Unfortunately, the “linear” assumption proved to be invalid for the quartz samples that were investigated in the present work. The fact that the dose-response-curves for both bleachable and un-bleachable can show roughly the same shape, at first glance, can mean that the traps, although different, are more or less correlated (e.g., linked to the same impurity) and/or that the TL growth is significantly controlled by the number of luminescence centres.

Finally, for old samples, estimating a palaeodose using the total TL or, alternatively, the un-bleachable TL face the same difficulties: using the additive dose-response-curve only, by means of extrapolation, is risky because its shape is unknown and, on the other hand, the regenerated dose-response-curve is of poor help, when using a slide technique, in the case its shape badly fits the additive one. It should be reminded that the present work was limited to the use of the additive dose technique. Developing it in the frame of other techniques like, e.g., SAR, needs more experiments and further discussion.

## 5-2 Consequences for unheated sediment dating

Since it is obvious that the initial TL level cannot be simply and securely recovered in the laboratory, such effect will induce errors in sediment dating. Errors will be more important for Red-TL, because its residual is significantly higher than for Blue-TL, as already observed (e.g., Miallier et al., 1994, Sholefield and Prescott, 1999, Fattahi and Stokes, 2004).

Sediment TL-dating most generally involves minerals that acquired an "infinite" dose prior to sun-exposure, without being annealed by heat. Such situation was tentatively simulated with the Pyla sample: for Red-TL, it was shown that the initial residual level - estimated from "long bleach" of the sample - was overestimated. For example, suppose the sample has received a natural dose of 300 Gy (simulated palaeodose, P). Subtraction of the measured residual TL can be made by shifting the horizontal axis at the level of the measured residual signal (dash-dotted line on Fig.4). It can be verified that it results in a dose-gap of around 40Gy. This will induce underestimation of P by 13%. For Blue TL, if the residual is comparatively ~3 times lower than for red TL ( $0.42/0.14 = 3$ ), then, roughly, the expected underestimation should be around 4-5%.

Therefore, trying to correct for this effect is justified. For that purpose, it is necessary to plot the dose-response-curve for the residual signal measured with samples bleached after irradiation (N+dose). Extrapolating such curve back along the dose axis, for a dose gap that corresponds to the previously estimated palaeodose P, will allow evaluation of the initial residual level (i.e., before burial). Then, P can be evaluated once again, using the best value of the residual, using an iterative method. Alternatively, the dose-response-curve can be extrapolated back until it reaches the extrapolated dose-response-curve for un-exposed samples; then, P is estimated to be the dose coordinate of the point, where the curves join each other. This is exactly the method that was proposed by Wintle and Huntley (1980) for

palaeodose evaluation in the case of partially bleached minerals, which is an important matter of concern in the field of sediment dating. These authors assumed that the amount of TL removed by partial bleach was a given fraction of the bleachable TL. At the light of the present work, it seems that the actual situation is more complex.

## 6 Conclusions

The residual TL of bleached quartz is a growing function of the radiation dose that was experienced before sun exposure. The basis level of such growth is nil for previously heated quartz, whereas it equals the initial residual level for naturally bleached quartz.

Such observations suggested that two different families of TL-traps can be involved in the observed TL signals, some being bleachable and the other not.

In the context of unheated sediment dating, such effect must be accounted for, because it can result in overestimation of the initial residual TL, in the case it is simply estimated from experimental “long bleach” of the sample to be dated.

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### Figure captions

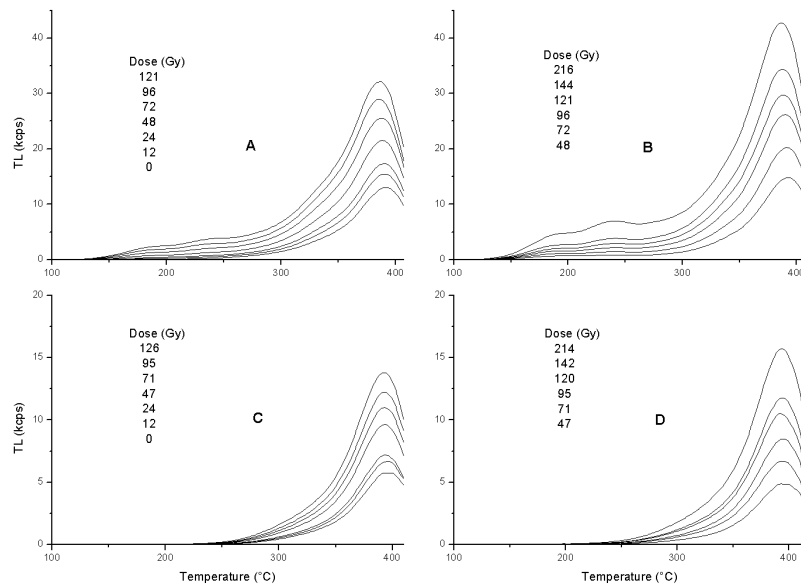


Figure 1.

Red-TL glow curves for sample C437. A: Additive glow curves. B: Regenerated glow curves, for quartz annealed at 380°C prior to irradiation. C and D: same as A and B, respectively, samples exposed to sunlight for one month prior to TL measurements.

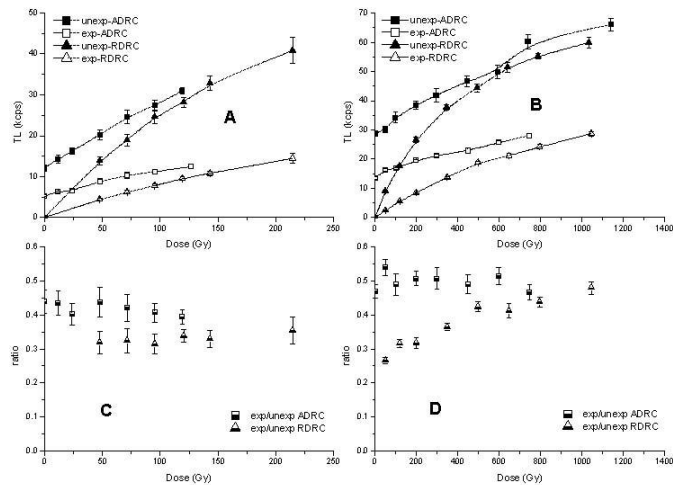


Figure 2

A and B: additive dose-response-curves (ADRC) and regenerated dose-response-curves (RDRC) for sample C437 (A) and for sample C397 (B). The plots show the Red-TL peak intensity vs radiation dose for quartz grains exposed for one month to sun after irradiation (exp) and for the same samples unexposed (unexp). Thin lines are spline fits to guide the eye. C and D: ratios between exposed and unexposed TL intensity derived from curves of A and B.

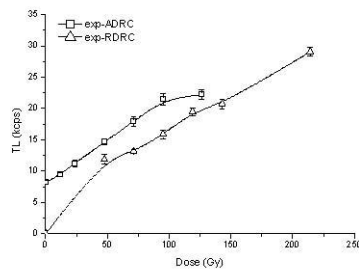


Figure 3

Same as Fig. 2A for Blue TL: dose-response-curves for sample C437 after exposure to sunlight.



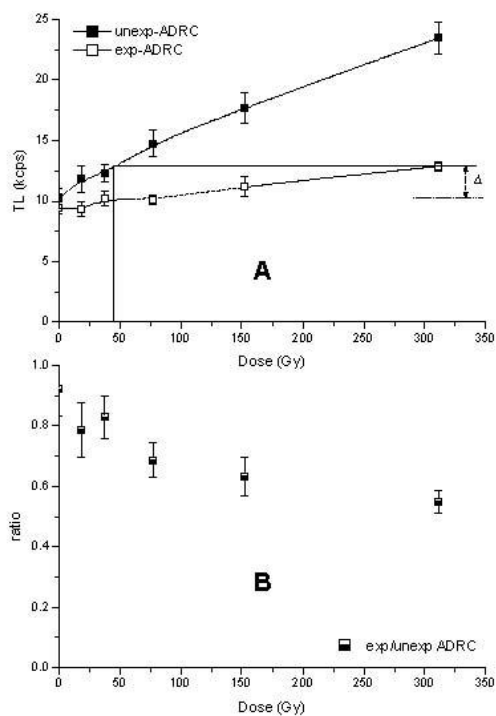


Figure 4

A: additive Red-TL dose-response-curves for quartz sampled at the top surface of a sea shore dune (Le Pyla, France). Open squares: samples exposed to sunlight for 2 months between irradiation and measurements. Black squares: unexposed samples.  $\Delta$  shows the TL-intensity of the sample irradiated at + 300 Gy, compared to the initial natural TL-intensity. The dash-dotted line indicates the radiation dose, measured using the unexposed additive dose-response-curve, that corresponds to the +300Gy residual TL (see text).

B: ratio between TL from exposed and unexposed samples.