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Record of thermoluminescence in sea sediments in the last millennia

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Summary. — The profile of thermoluminescence (TL) has been measured in the Ionian shallow-water core GT89-3 with a resolution of 3.096 years (corresponding to a sampling interval of 2 mm) during the last 1800 years. A similar TL record was previously obtained in the core GT14, taken from the same area, with a resolution of 3.87 years (corresponding to a sampling interval of 2.5 mm). We present here the comparison of the TL profiles. We confirm the presence of the centennial and the decennial cycles earlier identified in the TL signal, corresponding to cyclicities appearing to exist in the solar-activity records. We discuss the origin of the TL signal by comparison with the cosmogenic isotopes ^{14}C and ^{10}Be records.

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1. – Introduction

In previous papers [1-5] we have studied the TL profile of the core GT14, extracted from the Ionian Sea (Gulf of Taranto) in the Gallipoli Terrace. This core was dated with high accuracy by radiometric and tephroanalysis methods [6] and shows a sedimentation rate uniform within 1%. The core, spanning the time interval from 170 AD to 1977 AD, was sampled at consecutive intervals of 2.5 mm corresponding to 3.87 years. The power spectrum of the TL series showed a line structure, with few dominant components at 10.8, 12.06, 22, 28.5, 59 and 137.7 y, possibly related to solar variations by their similarity to the well-known solar cycles [4]. In order to confirm the existence of these periodicities, we have measured the TL profile in another core named GT89-3, taken from the same area and showing the same sedimentation rate as the GT14 core [7]. For the GT89-3 core, we used a different sampling interval of 2 mm, corresponding to 3.096 years, to be able to understand possible sampling effects in the time series spectra. Moreover, we have adopted different TL experimental procedures for the preparation of the samples in order to test the reproducibility of the results and to throw light on the origin of the TL signal.

2. – The TL experimental procedures

Two different procedures were used to prepare the samples of the core GT89-3: *a*) the powder technique (previously used in the preparation of the GT14 samples) and *b*) the disk technique.

Visible and UV light alter the original signal [8] and therefore the preparation of the samples was performed in red light. One gram of the still wet material was taken from adjacent intervals of 2 mm and washed in NaOH, H_2O_2, H_2O and acetone. After drying in oven at 40 °C overnight, the powder was sieved to sort out only the grains of dimension $d < 44\ \mu\text{m}$.

a) Powder technique: the powder was weighted in 4 samples of 15 mg for each interval and uniformly distributed on a little platinum holder for measurements.

b) Disk technique: the powder was weighted in samples of 10 mg and dispersed in acetone using an ultrasound apparatus; the uniform suspension in acetone was introduced in four vials and deposited on aluminium disks of 1 cm of diameter by drying overnight.

The samples obtained by procedures *a*) or *b*) were measured using the TL analyser, described by Miono and Otha [9]. The samples were heated up to 420 °C at a heating rate of 5 °C/s, in an atmosphere of pure nitrogen. The glow curves were registered by an *X*-*Y* recorder as a function of temperature. Both preparation methods preserve the original composition of the minerals releasing the TL signal stored in carbonate, quartz and feldspar crystals of the sediment, but could influence the collection of light or the uniformity of the signal. In the next paragraph we will show that the methods are almost equivalent.

In order to understand which component of the material carries the TL signal, we have prepared 146 consecutive samples in two different ways: half of the material has

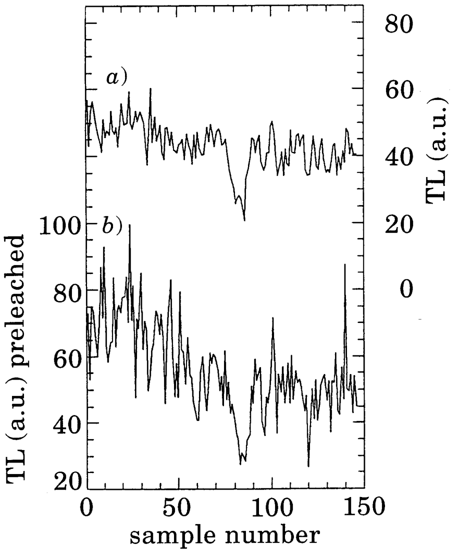

Fig. 1. – Comparison of the two TL records (146 samples) obtained by *a*) powder technique, *b*) HCl preleaching.

been treated as described in *a*) and the other half has been pretreated with HCl in order to remove the carbonates. The results of this test are shown in fig. 1*a*) and *b*). The overall similarity of the two records suggests that 1) the TL signal principally comes from the silicate component, 2) the fluctuations are strongly enhanced in the leached material and 3) the profile is almost unaltered removing or not the carbonates.

In the next section we present the experimental results obtained by the methods *a*) and *b*) in the core GT89-3. The profile using the leaching procedure will be the object of further studies.

3. – The TL profiles

The core GT89-3 was sampled with continuity at 2 mm intervals corresponding to 3.096 years; 584 samples were taken in order to cover the same period spanned by the GT14 core. The material was treated with the two procedures described in the previous paragraph. Each TL value was obtained by averaging 4 or 5 different glow curves taken from the washed material of the same layer: each profile results therefore from about 2600 glow curves. The TL profile of the samples measured on disks is shown in fig. 2*a*) and the profile obtained on the powder samples is shown in fig. 2*b*). The two profiles span the time interval from 170.9 AD to 1979 AD. On the original data, 11-point running averages are drawn by thick lines in order to easily compare short-term variations in the profiles. Similar features are evident, indicating the good efficiency of the TL signal collection with both methods. The average value M of the disk profile is $M = 81.6$ (a.u.) with standard deviation $\sigma = 12.0$ and of the powder one is $M = 73.2$ (a.u.) with $\sigma = 11.2$. The ratio $(\sigma/M)_{\text{pow}} = (\sigma/M)_{\text{disk}} = 0.15$ is identical for the two methods. In order to compare the new results obtained on the core GT89-3 with those previously

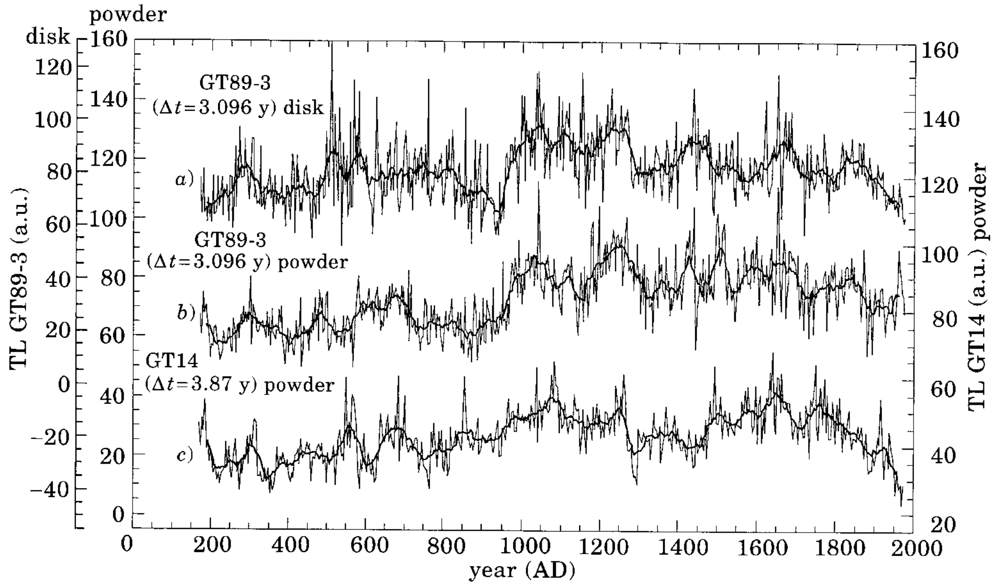

Fig. 2. – TL original series: GT89-3 core *a*) by disk technique, *b*) by powder technique, *c*) GT14 core by powder technique.

obtained in the core GT14, we show in fig. 2*c*) the TL profile previously published [8]. We recall that TL measurements were taken by the powder technique and by sampling at 2.5 mm spacing. We note that the ratio $\sigma/M = 0.13$ is very similar to those quoted above. The time series of GT14 was given by 467 data points.

Many common features are clearly visible like the step increase occurring at about 950 AD and the following decrease around 1300 AD; the peaks at 300 AD and at 1650 AD. But we may note also few differences between the three records.

4. – Spectral properties of the TL profile

The TL signals of the two series of the core GT89-3 were centred (means removed) and normalized by their standard deviations. In order to reduce statistical fluctuations, we have averaged the two time series. The resulting profile is shown in the inset of fig. 3. In this figure we report the power spectral density of the average TL signal of the GT89-3 core. The background level $\overline{P}$ of the spectrum, obtained by a linear fit of the spectral values in a logarithmic scale, is also drawn. The Fourier spectrum shows strong power in a band between (1/226) and (1/180) y^{-1}, and shows a well-defined line structure at periods of 139, 22, 15, 12, 11.4 years. We note that the first four lines were also found in the core GT14, as mentioned in the introduction. We have performed a detailed analysis of the band structure by carefully applying the method of superposition of epochs to the time series. In this approach the normalized time series is partitioned into consecutive segments of fixed length T that are then superposed and averaged. By least-square fitting a sinusoid with period T to the superposed data, we

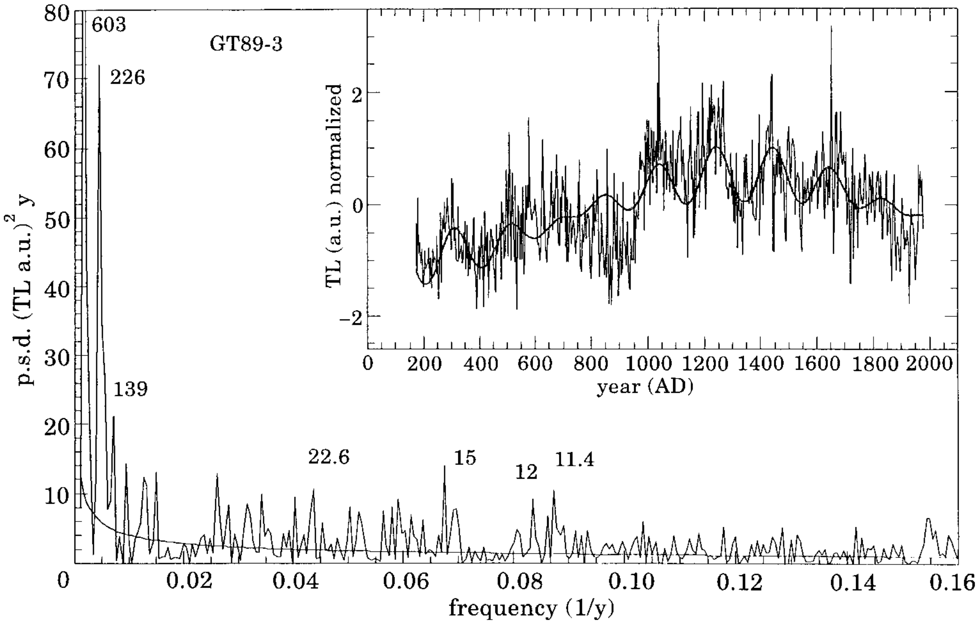

Fig. 3. – Power spectral density of the average TL signal of the GT89-3 core. In the inset the normalized TL series is shown; the superposed curve is the summation of the three waves of period $T = 2400$, 189.7, 225.4 y.

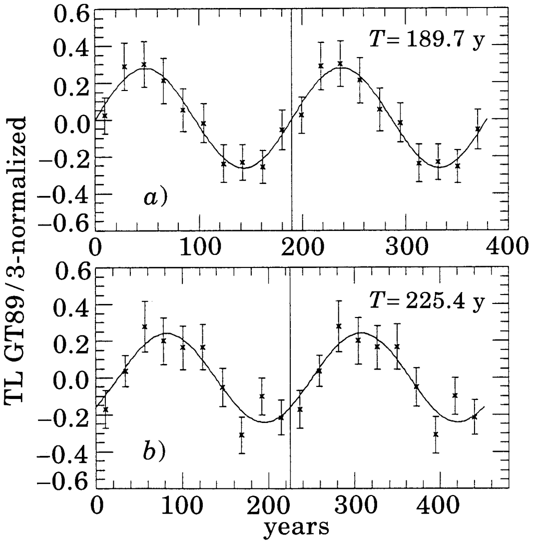

Fig. 4. – Superposed data and least-square fit sinusoid for the normalized TL signal of the GT89-3 core as given by the method of superposition of epochs; *a*) $T = 189.7$ y, $A = 0.27$ normalized units, $\varphi = 6.25$ rad, $r^2 = 0.99$; *b*) $T = 225.4$ y, $A = 0.24$ normalized units, $\varphi = 5.59$ rad, $r^2 = 0.98$.

have determined the phase Ω and the amplitude A of the waves, in steps of 0.1 y, for T varying from $T = 180$ y to $T = 240$ y. The correlation coefficients r^2 of the sinusoids to the folded experimental data were determined. The best-fit sinusoid is obtained when the maxima of r^2 and A are reached. This was found for the frequencies $(1/189.7)\,\text{y}^{-1}$ ($A = 0.27$, $\varphi = 6.25$ rad, $r^2 = 0.99$); $(1/225.4)\,\text{y}^{-1}$ ($A = 0.24$, $\varphi = 5.59$ rad, $r^2 = 0.98$). The results are shown in fig. 4*a*) and *b*). The phases are referred to 247.4 AD, as in all our previous analysis [8]. We note that the amplitude of these waves in per cent of the original TL average signal is $\sim 6\%$ (peak to peak). The sum of the two century scale components of the TL signal produces a carrier wave $\sin(2\pi t/206 + \varphi_{206})$ with an amplitude modulation wave $\sin(2\pi t/2400 + \varphi_{2400})$; there are two possibilities of $\varphi_{206} = 5.92$ rad if $\varphi_{2400} = 1.9$ rad or $\varphi_{206} = 2.78$ rad if $\varphi_{2400} = 5.03$ rad. The best fit of a sinusoidal wave to the original data, with a period of 2400 years, has a phase of $\varphi_{2400} = 5.00$ rad. If this is the modulating wave, the corresponding phase of the 206 y component is $\varphi_{206} = 2.78$ rad. In the inset of fig. 3 we show the summation of the 2400 y fit with the two waves of periods 189.7 and 225.4 y. The spectral features described so far account for nearly 65% of the standard deviation of the original times series.

We now remind that the TL spectrum of the core GT14 shows [8] two decennial lines at 12.06 y ($\varphi_{12.06} = 4.84$ rad) and 10.8 y ($\varphi_{10.8} = 0.94$ rad). The sum of the two components produces a modulated wave train with a carrier period 11.4 y and an amplitude modulation period 206 y [3]. The phase of the carrier and of the modulation wave were determined up to a shift of $\pm\pi$. Now, with the new results of the core GT89-3 we have determined the phase of the 206 y wave $\varphi_{206} = 2.78$ rad and therefore we unambiguously fix the phase of the carrier wave $\varphi_{11.4} = 6.03$ rad. In the core GT89-3 the power spectrum shows the line at 11.4 y and by the method of superposition of epochs the phase has been found to be $\varphi_{11.4} = 5.7$ rad, in good agreement with that inferred for the core GT14.

Having observed the correspondence of the TL signals in the two cores, in order to reduce the statistical fluctuations of the measurements, we compute a composite series of the GT89-3 and GT14 records by the following procedure: we fix the starting time of the composite series (170.9 AD) and the sampling interval ($\Delta t = 3.096$ y) both coincident with those of the GT89-3 core. Due to the experimental procedure, the measured values $\mathrm{TL}_{\mathrm{GT89\text{-}3}}(t)$ and $\mathrm{TL}_{\mathrm{GT14}}(t')$ of the three series of fig. 2 are an average over the respective time intervals $T = (t, t + \Delta t)$ and $T' = (t', t' + \Delta t')$, where $\Delta t = 3.096$ y and $\Delta t' = 3.87$ y. Therefore, the composite value $\mathrm{TL}(t)_{\mathrm{compos}}$ in the interval T may be computed by the average

$$\mathrm{TL}(t)_{\mathrm{compos}} = \frac{1}{3}\left\{\mathrm{TL}^{\mathrm{disk}}_{\mathrm{GT89\text{-}3}}(t) + \mathrm{TL}^{\mathrm{powd}}_{\mathrm{GT89\text{-}3}}(t) + \sum \mathrm{TL}_{\mathrm{GT14}}(t')\cdot\left[\frac{\tau(t')}{\Delta t}\right]\right\},$$

where the summation is over those values of the GT14 series corresponding to intervals T' with not null superposition $\tau(t')$ with the considered interval T and where the weights $\tau(t')/\Delta t$ are the percentages of superposition. In our case, each sample of GT89-3 can overlap with one or two samples of GT14.

The composite TL series is shown in the inset of fig. 5. The corresponding power spectral density is shown in the same figure. We observe in particular the presence of power at frequency $(1/11.4)\ \mathrm{y}^{-1} = ([1/10.8 + 1/12.06]/2)\ \mathrm{y}^{-1}$. This corresponds to the average Schwabe solar cycle. We note also the presence of the band centred at frequency $(1/206)\ \mathrm{y}^{-1} = ([1/10.8 - 1/12.06]/2)\ \mathrm{y}^{-1}$. The amplitudes of the 206 y and of the 11 y cycles are, respectively, ~ 4% and 2% of the original TL signal; this estimate is

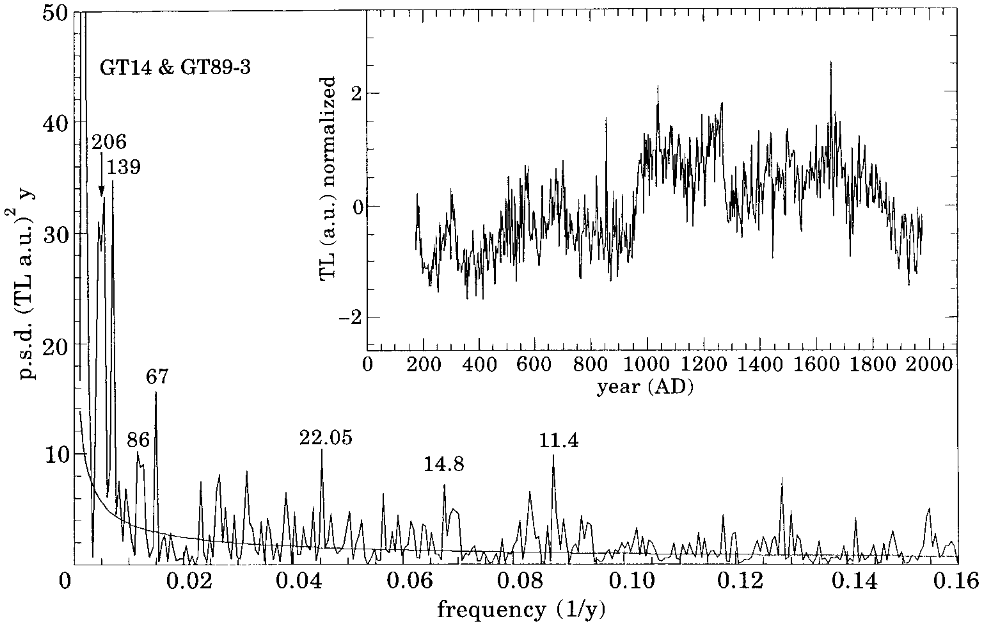

Fig. 5. – Power spectral density of the composite series of the GT89-3 and GT14 TL records ($\Delta t = 3.096$ y); the main periodicities are indicated above the peaks. The composite series is shown in the inset.

derived by the method of the superposition of epochs. The Hale and the Gleissberg solar cycles appear as well in the spectrum together with the line at (1/139) y^{-1}, the origin of which is most probably also solar, being detected in the historical aurorae records [10]. The TL signal seems mostly forced by the solar output variations, especially if a connection of the 200 y periodicity with solar variability is not merely conjectural in the radiocarbon tree ring record.

5. – Discussion and conclusions

This is the first evidence for the simultaneous presence of the periodicities of 11 y and 200 y in the power spectrum of a terrestrial time series recording past solar-terrestrial relationships. The most recent results on ^{10}Be and ^{36}Cl concentrations in Dye 3 ice core [11, 12] confirm the process of interplanetary solar modulation of galactic cosmic rays through the 11 y variability of cosmogenic isotopes in the ice record during the last 550 years. The length of the record does not allow to identify the 200 y period as a line in the spectrum, but the Gleissberg cycle is evident, as happens in the spectrum of the sunspots [13]. The ^{14}C cosmogenic isotope, on the other hand, although produced by GCR as the previous ones, is a chemically active element and major terrestrial factors are involved in its time variability: as a result of dilution of radiocarbon in the atmospheric CO_2, the 11 y in the tree ring series is damped. However, it is commonly accepted that tree ring radiocarbon records the history of solar activity, reflected in the solar wind, backward ~ 10 ky BP. The lines in the power spectrum at ~ 200 y and at the Gleissberg period (*e.g.* [14, 15]) are commonly ascribed to the presence of solar components in the radiocarbon signal. In the TL time series power spectrum we have the simultaneous evidence for the same components of solar origin found in ice and tree rings cosmogenic isotopes.

The comparison of the composite TL series with the decadal ^{14}C in tree rings (*e.g.*, given in [14]) during the last two millennia and with the annual ^{10}Be in Dye 3 ice core [16] during the last 550 years give both correlation coefficients ^{14}C-TL and ^{10}Be-TL $r \sim 0.4$, with TL leading by ~ 60 y the ^{14}C and by ~ 12 y the ^{10}Be records. We must remember that the correlation coefficient between the annual records of ^{14}C and ^{10}Be is of the same order ($r = 0.42$). The low degree of correlation between all these records is due to the fact that the signals are embedded in terrestrial noises of different origin. We think that these results confirm our working hypothesis that the TL signal is controlled by solar activity: the radiation dose stored in the crystals deposited in the sediments may be affected by variations in the ionizing component of cosmic rays starting from the lowest energies and/or modulated by the light to which the crystals are exposed before their deposition.

In conclusion, we have achieved a satisfactory confirmation of our earlier findings and interesting new results, by performing an entirely new experiment on a different core, using a different sampling interval and different experimental procedures for preparing the TL samples.

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