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Analysis of Optical Bleaching of OSL Signal in Sediment Quartz

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Highlights:

- Bleaching experiments on sediment quartz are performed.
- Blue LED light source incorporated in luminescence reader is used.
- New analysis of data measured by standard SAR OSL technique is proposed.
- The results are promising for recognizing and compensating for partial bleaching.

Abstract:

The aim of this work was to study the effect of the quality of optical bleaching on the results of OSL (Optically Stimulated Luminescence) dating method. The large aliquots of coarse quartz grains extracted from fluvial deposit were used in the study. The poor, medium and good bleaching were simulated in laboratory with help of Blue LED light source in series of experiments. Then the samples were irradiated with a common laboratory dose. The equivalent doses (DE) were measured by the help of standard Single Aliquot Regeneration (SAR) technique, but obtained DE distributions are analysed in a new way. The method for recognizing and compensating for partial bleaching is proposed. The conclusions for dating sediment quartz samples are presented and discussed.

Keywords:

Optically Stimulated Luminescence dating method,
sediment quartz,
bleaching.

1. **Introduction**

One of the main problems in the optically stimulated luminescence (OSL) dating of geological deposits concerns the completeness of reset of accumulated luminescence signal at the moment, which is subjected to the dating. It is assumed that for some types of

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sediments such reset can be stimulated by an exposure to daylight, which takes place prior to the deposition. However, the effectiveness of such optical bleaching is controlled by many conditions, which strongly depend on the environment of transport and sedimentation. The partial bleaching of sediment not only increases scatter of the obtained OSL results, but also ï what is even worse ï it makes the calculated age of the deposit older than it really is (Rittenour, 2008; Rodnight, 2008).

For dating partially bleached samples the single grain approach was proposed (Olley *et al.,* 1999). Some statistical models for determining the true burial dose were also developed (Galbraith and Roberts, 2012). Many of them are used only for analyzing single grain results (e.g.: Bailey and Arnold, 2006). Usually, it is assumed that for fluvial sediment samples the Minimum Age Model is the most appropriate (Olley *et al.*, 2004). However, single grain technique needs specially dedicated luminescence reader and such measurements are rather time consuming (Thomsen *et al.*, 2007 and references therein). This method also deals with specific problems, like microdosimetry effects and dark grains content.

The aim of this work was to study the effect of the quality of optical bleaching on the results of Single Aliquot Regeneration (SAR) OSL technique applied to large aliquots, consisting of hundreds of quartz grains. Our motivation was to elaborate analysis method enabling not only to recognize partial bleaching, but also to correct results of standard SAR OSL measurements in order to avoid overestimation of the equivalent dose. In case of geological deposits it is hard to be absolutely assured about material homogeneity and bleaching conditions in nature. Due to this obstacle SAR OSL measurements for this study were carried out on specially prepared samples, for which bleaching as well as burial dose were simulated in the laboratory.

The proposed method takes advantage of the idea of OSL dating method. The light, which stimulates signal, is used here also as the bleaching agent. Obviously blue LED light source in the reader is much stronger and covers only part of the daylight spectrum. Nevertheless full characteristic of light, which the natural sediment was exposed to, is never precisely known. Therefore blue light substitutes only for daylight, analogically as in standard techniques of luminescence dating laboratory radioactive source (usually alpha or beta or gamma with dose rate of the order of 100 mGy/s) is used as substitute for natural ionizing radiation (mixed and much weaker). The idea of the equivalent bleaching can be postulated similarly as the equivalent dose is defined in retrospective dosimetry.

2. Material and Methods

The sample KO20 originating from quaternary fluvial sediments was chosen to provide only initial material for the studies and the coarse quartz grains were extracted by standard technique routinely used in our laboratory (Weckwerth *et al.*, 2011). All the experiments (OSL measurements, as well as bleaching and burial dose simulations) were carried out on an automated model Riso TL/OSL-DA-20 reader (Bøtter-Jensen *et al.*, 2010) equipped with blue LED light source for stimulation (470 nm delivering 80 mW/cm^2 at the sample), and PMT with a Hoya U 340 filter (7.5 mm) for detection (290-370 nm). The incorporated beta source ^{90}Sr/^{90}Y delivering (140 +/- 9) mGy/s was applied for irradiations. From now on, all the dose values are expressed as the beta source irradiation time in seconds, in order to avoid taking into account systematic uncertainty of the beta dose rate. The samples were put on stainless steel discs in quantities of (2 +/- 0.2) mg per aliquot for measurements and (5 +/- 0.2) mg for simulations (for bleaching and burial dose alike).

The equivalent doses values D_E were determined by using the SAR-protocol (Murray and Wintle, 2000). For every sample 24 aliquots were measured by 40 s of blue light stimulation at 125°C, after preheat of 10s at 240°C. D_E was estimated by the linear interpolation applied for OSL signal integrals: from 0 to 0.8 s, background subtracted (Weckwerth *et al.*, 2011). The test (calibration) doses applied to correct for OSL sensitivity changes were fixed at 50 s of beta irradiation.

In laboratory the daylight bleaching was simulated by exposure to blue LED light in Riso reader. The quality of bleaching was controlled by setting the time of the light exposure.

3. Results

As it is shown in Fig. 1 the sediment sample KO20 exhibits relatively high level of D_E, with mean value of 3700 s and wide scattering: for 19 accepted aliquots standard deviation SD = 1300 s (36%). Such sample is thought to be good representative of rough deposit material, which has to be subjected to optical bleaching during geological processes of erosion, transportation and sedimentation. The effectiveness of stimulated bleaching was investigated by SAR OSL measurements of residual luminescence remaining in the aliquots.

Results obtained for the range of 0.55 s ï 1.45 s with resolution of 0.05 s are presented in Fig. 1 b.

In order to simulate various bleaching conditions 8 series of experiments were performed resulting in 8 different sub-samples (Table 1). Two types of bleaching time distributions were taken into account: Gaussian-like with SD = 40% (g series), and flat-shaped (f series). For simulating Gaussian bleaching the range of exposure times was delimited with width proportional to the central value. Inside this section a set of discrete values was determined by dividing the range according to the resolution specified in the Table 1. Then the number of aliquots was assigned to each value of bleaching time according to the Gaussian distribution. In Fig. 1 b the distribution designed for simulation g1 is presented as an example. For simulating flat like bleaching equal number of aliquots (usually two) was assigned to every value of 48 discrete bleaching times determined inside given range of constant width of 0.47 s with the resolution of 0.01s. It is worth to notice, that for flat simulation with long exposures (10 s and 40 s) the effectiveness of bleaching is almost constant within so narrow range.

In every bleaching simulation 96 individual aliquots were mixed together directly after bleaching. Then new set of 48 aliquots of such mixture was prepared for irradiation by laboratory beta source. In order to simulate little scatter of natural burial dose the beta source exposure times were governed by the Gaussian distribution: (100 +/- 4)s. Finally, the irradiated aliquots were mixed again, producing 8 subsamples for SAR OSL measurements.

Two examples of D_E distributions are presented in Fig. 2. Despite many histograms seem to be asymmetric in first approximation normal distribution was assumed to characterize the data: for g03 (Fig. 2 a): mean D_E = 726 s, standard deviation SD = 255 s (35%) and for g10 (Fig. 2 b): mean D_E = 120 s, standard deviation SD = 11 s (9%). In general, for both g and f series, the level and the scatter of D_E values increase with bleaching time decreasing. This dependency is presented in Fig. 3 a, where overestimation of D_E for short bleaching is clearly seen.

For quantitative merit of the bleaching quality the coefficient Q was introduced. It is defined as the ratio of the true burial or laboratory dose (here 100s) to the equivalent dose D_E estimate:

$$Q = \mathit{True\ Dose}\ /\mathrm{mean}\ D_E \qquad (1).$$

Theoretically Q values are limited to the range: from 0 (the poorest bleaching) to 1 (total bleaching). In Fig. 3 b the D_E scatter is plotted against bleaching quality coefficient Q. It derives that the over-dispersion is characteristic for poorly bleached samples. Recalling SD value of natural sediment (36% - see Fig 1 a) it suggests that KO20 sample can be partially bleached. However, it is not possible to determine the D_E overestimation nor the true burial dose yet.

4. Analysis and Discussion

In addition to the histograms, D_E values are also plotted against initial (natural) OSL values I_0 recorded in first SAR cycle (Fig. 4). To make such plots comparable for different samples, both D_E and I_0 were normalized to their mean values. Hence, for equally bleached material one can expect that all data points in the graph should be randomly distributed around mean values of $D_E=1$ and $I_0=1$, with no correlation between D_E and I_0. However, instead of forming circle around point (1,1) the experimental results clearly show linear dependence. Strong correlation between D_E and I_0 can indicate poor bleaching (Li, 1994). As a matter of fact poorly bleached sample g03 (Fig. 4a) clearly demonstrates wide-range dependency $D_E(I_0)$ in contrast to the moderately bleached sample g10 (Fig. 4b), where such correlation is limited to narrow range only.

Following the predictions for the ideal, totally bleached sample, we propose to find the most flat part of the $D_E(I_0)$ data plot, which should give the best estimate of D_E. This region can be defined by the postulated equation:

$$D_E = const(I_0) = R, \qquad (2)$$

where R is the only fitting parameter. The fitting procedure is started from the lowest I_0 value and it is progressively repeated $n-1$ times (when n equals to the number of data points). In every step next data point with successive I_0 value is included (Fig. 4).

As an outcome of such analysis we obtained a sequence of R results accompanied by $\boldsymbol{G}^2$ values, which characterize the quality of the fit. By plotting R values against $\boldsymbol{G}^2$ (Fig 5 a-b) we found, that the data focus in the region of minimum of $\boldsymbol{G}^2$ values. The corresponding R value was chosen as the optimal value of R parameter, representative for the whole data set: 0.72 for g03 (Fig. 5 a) and 0.94 for g10 (Fig. 5 b). Analogically, the optimal R value of 0.83 for natural sample KO20 was determined.

Next we plotted optimal values of R parameter estimated for all subsamples against their values of bleaching quality Q (Fig. 5 c). We found that this dependency can be fitted by linear function:

$$\text{Optimal } R \text{ value} = A + B\,Q\,, \tag{3}$$

where $A = 0.69$ +/- 0.01 and $B = 0.29$ +/- 0.02, and correlation coefficient is 0.98.

When the bleaching characteristics (3) of the sample is established, it can be used to determine the quality of bleaching in nature: Q_{nat}. In case of KO20 $Q_{nat} = 0.48$ (Fig. 5 c). Once the Q_{nat} value is already known, the true burial dose can be derived using the formula (1). For sample KO20 it turns to be of the order of 1790 s.

In case of difficulty with determining the optimal value of R for natural sample we suggest to proceed with additional analysis. It comprises of plotting the sequence of parameter R values against number of points used for fitting (Fig 6). Comparing such graphs obtained for simulations with plot produced for natural sample it enables to estimate the bleaching simulation, which is the most equivalent to the natural one. In case of KO20 sample it is g1 (Fig 6a) and f08 (Fig 6b), both presenting optimal R parameter value of 0.83. Hence, it confirms the result obtained from Fig. 5 c.

5. **Conclusions**

The new method of analysis of the SAR OSL results obtained from large aliquots was proposed. It was demonstrated that such analysis can provide quantitative information on the quality of bleaching under assumption, that scatter of D_E values is dominated by incomplete bleaching and other reasons (e.g. post-depositional mixing or dose-rate heterogeneity) can be neglected. Although no particular model of bleaching is assumed, but aliquots presenting low D_E values are favored, as they are thought to be bleached better than other.

In this paper results of a new method are presented in details for only one sediment sample. Nevertheless, the method in its preliminary version already proved to be useful for recognition of partially bleached fluvial sediments investigated by Weckwerth *et al.* (2012), where it helped to explain inversion of apparent OSL ages.

Acknowledgments

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Bleaching quality	Q range	Gaussian with SD=40%				Flat with constant width of 0.47s			
		Sample code	Central value [s]	Range [s]	Resolution [s]	Sample code	Central value [s]	Range [s]	Resolution [s]
good	0.9 =< Q	g40	40	23 ï 59	2	f40	40	39.77 ï 40.24	0.01
medium	0.5 < Q < 0.9	g10	10	5.5 ï 14.5	0.5	f10	10	9.77 ï 10.24	0.01
poor	Q =< 0.5	g1	1	0.55 ï 1.45	0.05	f08	0.79	0.55 ï 1.02	0.01
		g03	0.3	0.15 ï 0. 45	0.02	f03	0.29	0.05 ï 0.52	0.01

Table 1

The list of blue LED light exposure times applied in the experiments for simulating: good, medium and poor bleaching. The ranges of coefficient Q (1) values were proposed to specify more objective criteria of the bleaching quality (compare Fig. 3 b). Note the differences in the width of time ranges between Gaussian and flat distributions.

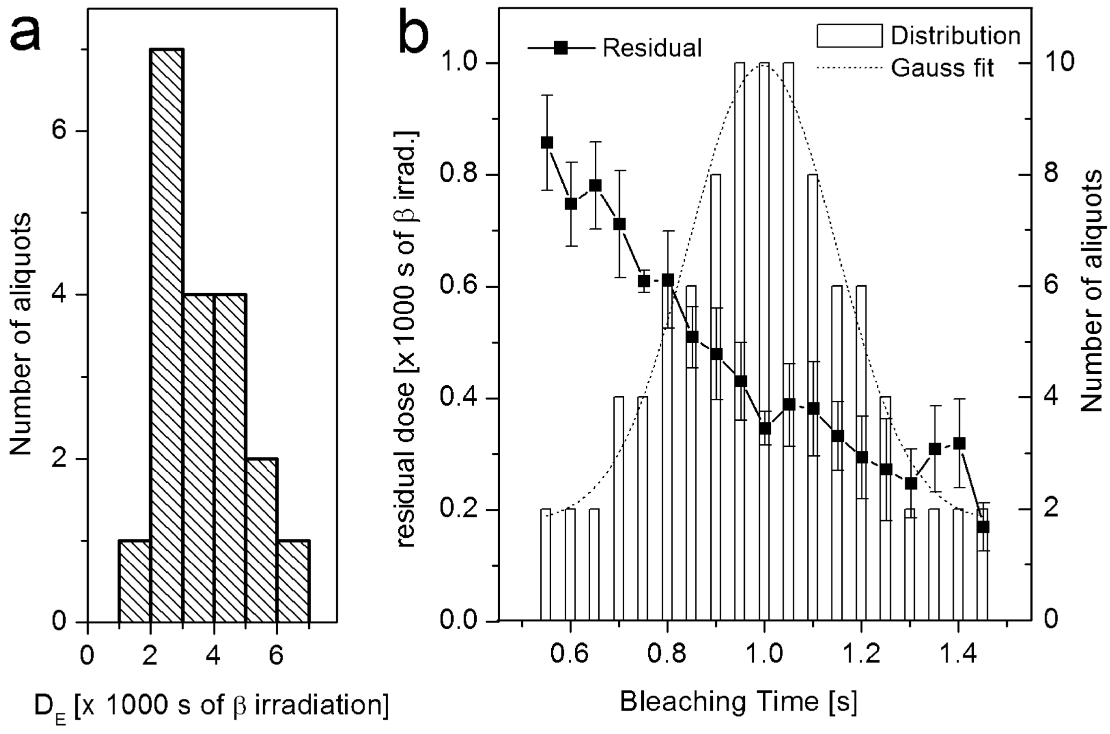

Fig. 1. a) The distribution of equivalent dose D_E values measured for natural sediment sample KO20 used in the study. b) The bleaching simulation with blue LED light ï the case of exposure times around 1 s: residual doses (left axis) against bleaching time and histogram presenting number of aliquots assigned to selected bleaching times as applied for preparing sample g1(right axis). The dose values are expressed as beta source irradiation time (in seconds).

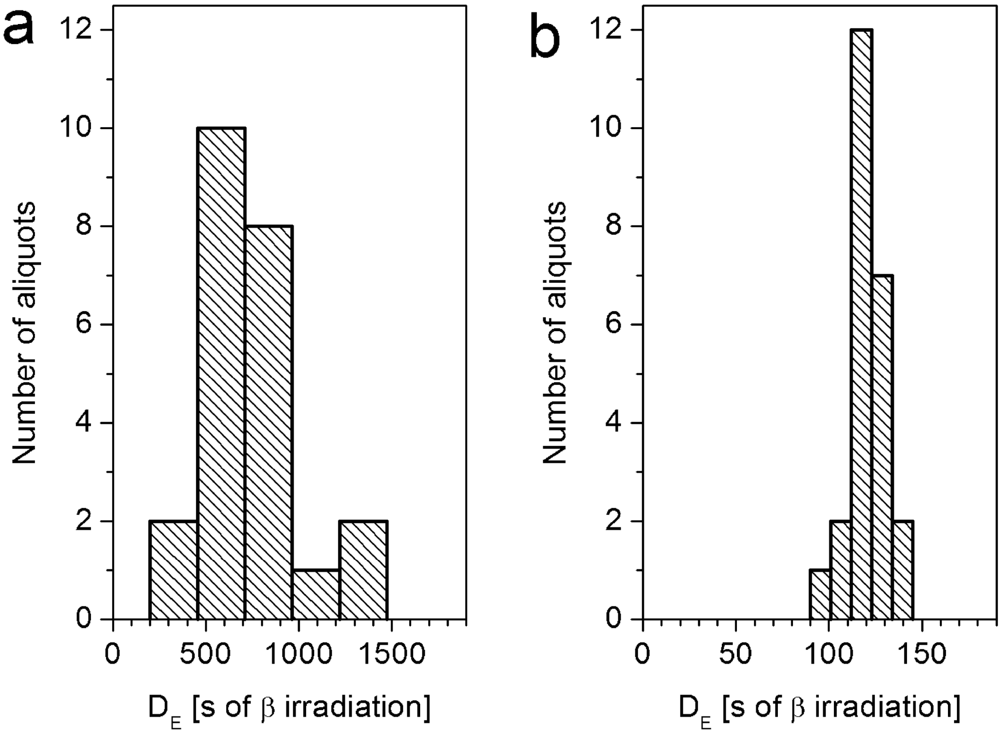

Fig. 2. The distributions of equivalent dose D_E estimates (expressed as beta source irradiation times) obtained from SAR OSL measurements performed on subsamples irradiated by beta laboratory source for 100 s after Gaussian bleaching: a) g03 and (b) g10. In both cases the number of accepted aliquots n =24.

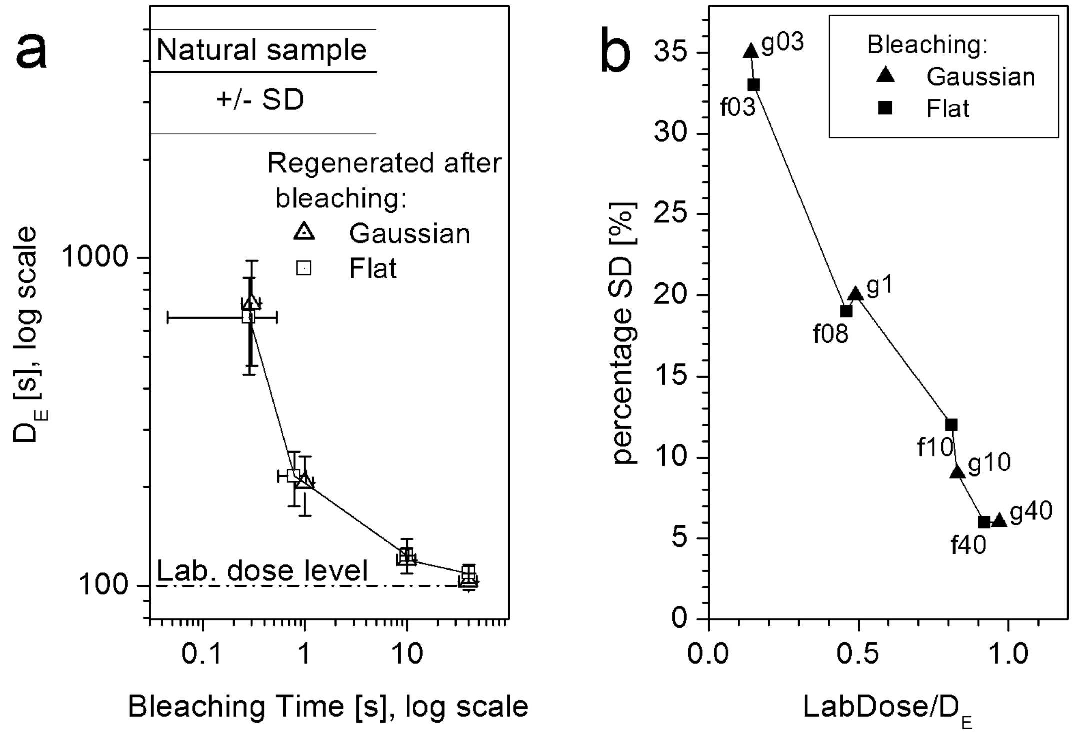

Fig. 3. The results of SAR OSL measurements obtained for various bleaching stimulations and common regenerative laboratory dose of 100 s: (a) dependence of equivalent dose D_E on applied bleaching time ï note double logarithmic scale and (b) dependence of percentage standard deviation SD of D_E on the bleaching quality coefficient Q, given by the formula (1). The subsample codes are indicated on the graph. The doses are expressed as beta source irradiation times (in seconds). Y bars represent SD of D_E, X bars represent SD of bleaching time for Gaussian simulation (triangles) and width of the range for flat simulations (squares).

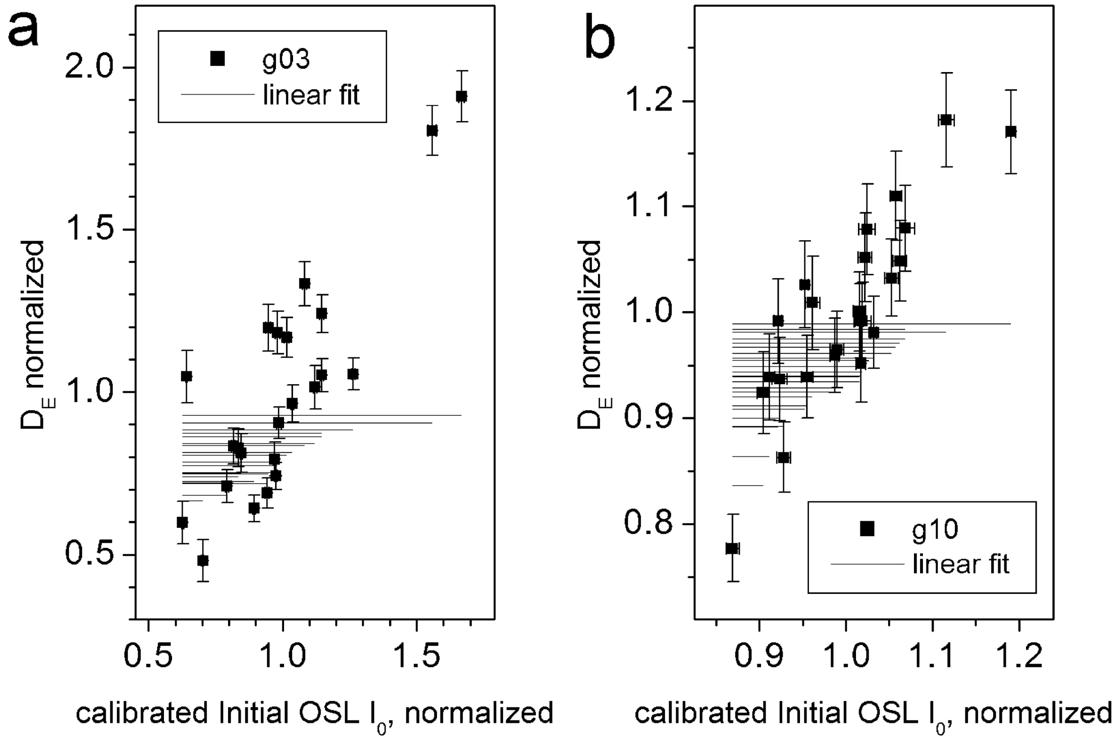

Fig. 4. Plots of normalized D_E estimates versus normalized initial OSL signals I_0 measured for subsamples irradiated by the same beta dose after Gaussian bleaching: (a) g03 and (b) g10. Note the different scales in (a) and (b). Horizontal lines represent successive linear regression fits of formula (2): $D_E = R = \mathrm{const}(I_0)$ ï see the text for details.

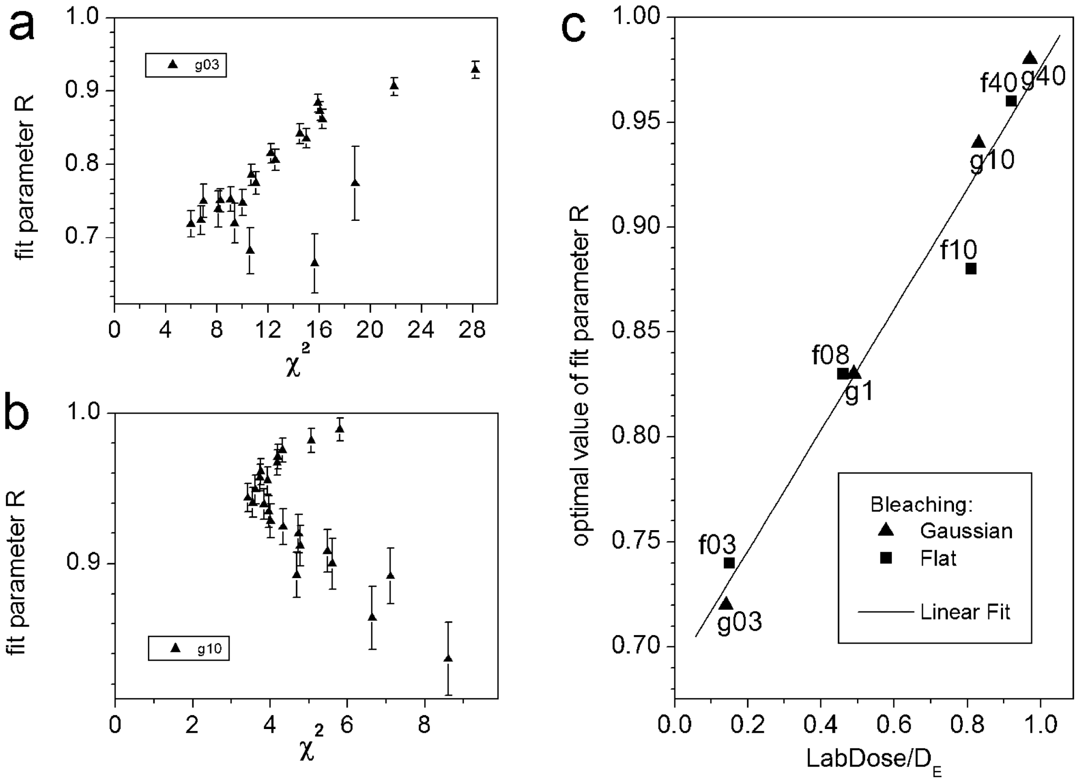

Fig. 5. Plots of values of fitting parameter R against corresponding G^2 values obtained for subsamples irradiated by the same beta dose after Gaussian bleaching: (a) g03 and (b) g10. c) The dependence of the optimal value of fitting parameter R on the bleaching quality coefficient Q. Linear fit (3) is also presented in the graph. The subsample codes are indicated and the level of natural sample is marked for estimating Q_{nat} value.

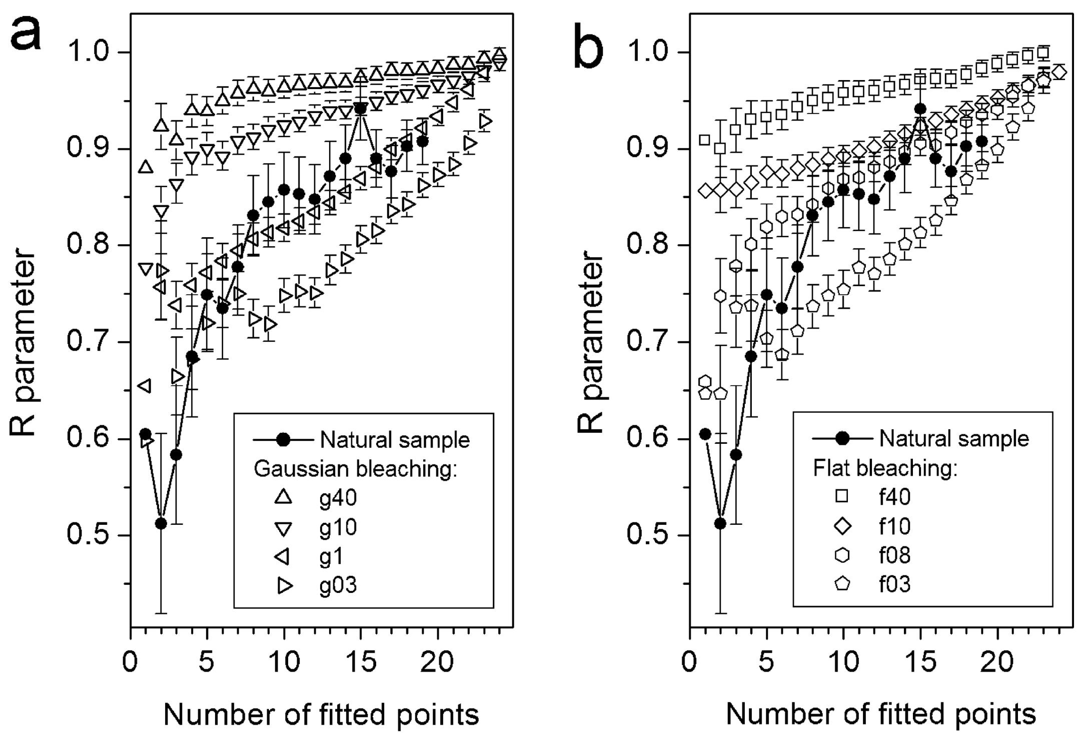

Fig. 6. The sequence of parameter R values plotted against number of points used for fitting (2). The results obtained for natural fluvial sediment sample used in the study (solid circles connected by solid line) are compared with the results of simulations (open triangles): (a) the series of Gaussian bleaching and (b) the series of flat bleaching.