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Research paper

# Chronostratigraphy of a sediment record from the Hajar mountain range in north Oman: Implications for optical dating of insufficiently bleached sediments

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**Abstract**

In this study we report on optical stimulated luminescence (OSL) ages of quartz extracted from a sedimentary record in the N-Oman mountain range. Equivalent dose ($D_E$) distributions derived from single aliquot measurements (SAR) of small aliquots (ca. 200 grains) were investigated to test whether the skewness and broadness of the dose distribution can be used as criteria for the identification of insufficient bleaching. Furthermore, the methods proposed by Lepper and McKeever [2002 An objective methodology for dose distribution analysis. Radiation Protection Dosimetry 101 (1–4), 349-352]. Singhvi [Juyal, N., Chamyal, L.S., Bhandari, S., Bushan, R., Singhvi, A.K., in press. Continental record of the southwest monsoon during the last 130 ka: evidence from the southern margin of the Thar Desert, India. Quaternary Science Review] and Fuchs and Lang [2001 Fuchs, M., Lang, A., 2001. OSL dating of coarse-grain fluvial quartz using single-aliquot protocols on sediments from NE Peloponnese, Greece. Quaternary Science Review 20, 783–787.] to derive $D_E$'s from insufficiently bleached sediments were compared. At first, the investigations were carried out on artificially bleached, irradiated and mixed quartz material from the Oman study area to simulate insufficiently bleached sediments. Then, the various statistical methods for identifying insufficient bleaching and $D_E$ derivation were applied to the natural samples from the study area, where 18 samples were measured. For the identification of insufficient bleaching the preferential parameter is the broadness of a distribution. For $D_E$ calculation, both the Singhvi method and the Fuchs and Lang method produce similar results, which are consistent with the stratigraphic order. A drawback of both methods is their sensitivity to low outliers. The Lepper and McKeever method was not applied to the natural samples, due to limitations in its application to a small number of aliquots and due to the ambiguous identification of the rising limb of the dose distributions.


*Keywords:* Sedimentary record; Optical dating; OSL dating; Insufficient bleaching; Small aliquots; Quartz; N-Oman

## 1. Introduction

Optical dating of sediments prerequisites the zeroing of the latent luminescence by exposure of the mineral grains to daylight during the last process of sediment reworking, transportation and deposition (cf. Aitken, 1998). An incomplete resetting of the luminescence signal is associated with residual luminescence, which causes an age overestimation. In the case of wind-borne sediments, resetting of the latent luminescence signal to a sufficient near-zero residual value occurs almost always (cf. Hilgers, et al. 2001). However, for waterlain sediments incomplete bleaching is frequent (cf. Jain, et al. 2004; Olley, et al. 1998) due to the filtering and attenuation characteristics of the solar spectrum within the clear or turbid water column or due to short transport distances (cf. Ditlefsen, 1992; Fuchs et al., 2005). Thus, the detection of insufficient bleaching and the derivation of the true depositional age-representative equivalent dose ($D_E$) for fluvial sediments are necessary requirements and challenges in luminescence dating.

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There are several suggestions of how to detect insufficient bleaching, and how to derive the correct $D_E$ from an insufficiently bleached sample (Wallinga, 2002a). One way to detect insufficient bleaching is the analyses of $D_E$ dispersions based on single aliquot or single grain analysis. Li (1994) and Clarke (1996) used the coupled parameters $D_E$ dispersion and luminescence intensity derived from multiple grain single aliquot measurements, but they took not into account the important aspects of aliquot sizes. While looking at the $D_E$ distribution and shape of the distribution, Olley et al. (1998, 1999) recognized the shortcoming of large aliquots and reduced the number of grains per aliquot. Fuchs and Wagner (2003) followed this concept of small aliquot measurements but suggested an upper and lower limit of grains per aliquot to reduce the unwanted effect on the $D_E$ distribution even for well bleached sediments. Recent developments in instrumentation (Bøtter-Jensen et al., 2003) enables even the routine measurements of single grains, but the problematic effect of microdosimetry (Kalchgruber et al., 2003) could lead to a high $D_E$ dispersion even for well bleached samples (Thomsen et al., 2005).

If insufficient bleaching is suspected, several models exist to derive the possible true depositional age-representative $D_E$ from a dose distribution. All of these models assume that the true $D_E$ is represented by lower values of a dose distribution, due to a more or less complete resetting of the latent luminescence signal, whereas $D_E$'s of higher values are caused by an additional residual dose. Based on this assumption Olley et al. (1998) derived in their example from known-age fluvial deposits from Australia correct $D_E$'s by arbitrarily considering the lowest 5% of their positively skewed dose distributions. Based on the assumption that insufficient bleaching is related to asymmetrical and positively skewed distributions, Lepper and McKeever (2002) suggested a model, the so-called *leading edge* method, where the true $D_E$ is derived from the inflexion point of a Gaussian curve which is fitted to the rising limb of the distribution. Fuchs and Lang (2001) proposed a site specific but easy to apply model especially designed for small numbers of aliquots. Hereafter, the $D_E$ is derived from a sorted low to high value data set, calculating an arithmetic mean and its corresponding coefficient of variation ($\nu$) by starting with the two lowermost $D_E$ values and adding to each calculation step one more $D_E$ value, until a certain threshold of $\nu$ is exceeded. The threshold of $\nu$ is derived from an artificially bleached and irradiated sample from the sample locality, representing the best achievable precision for natural samples of the studied site. In this mode, larger scatter is thought to be due to heterogeneous bleaching.

In this study we investigate samples from a sedimentary record near the oasis of Maqta in the Hajar mountain range, north Oman (N 22.83°, E 59.00°), where the sediment structure suggests a waterlain origin, thus indicating that insufficient bleaching was likely. To test OSL criteria for insufficient bleaching, the skewness and broadness of the $D_E$ distribution is empirically investigated on artificially bleached, irradiated and mixed quartz material from the Oman study area in order to simulate insufficiently bleached sediments. Furthermore, the difference between the methods suggested by Lepper and McKeever (2002) and by Fuchs and Lang (2001) in analyzing $D_E$ derivation from insufficiently bleached samples was investigated using the simulated insufficiently bleached sample material, while the methods proposed by Fuchs and Lang (2001) was modified by using not the unweighted but the weighted mean. Additionally, a statistical method proposed by Singhvi (Juyal et al., in press) representing a simplified minimum age model of Galbraith et al. (1999) was included in this comparison. Thereafter, the $D_E$ is derived from an insufficiently bleached sample by calculating the weighted mean of a $D_E$ range. This range is defined by a lower limit represented by the lowest $D_E$ of a distribution, and an upper limit defined by the sum of the lowest $D_E$ plus two times the highest $D_E$ error of the lower part of the dose distribution plus an instrumentation error of 4% ($D_{E\min} + 2\sigma_{\max} + 4\%$).

Two of the methods, the method proposed by Fuchs and Lang (2001) and by Singhvi (Juyal et al., in press) were then applied to 18 natural samples from the study area, where except for three samples only a limited number of maximum 24 aliquots per sample were available. $D_E$ estimates based on both methods show good agreement within errors and the resulting OSL ages are in stratigraphic order. The proposed method by Lepper and McKeever (2002) was not applied to the natural samples, due to limitations in its application to a limited number of aliquots and due to the ambiguous identification of the rising limb of the dose distributions (Woda and Fuchs, submitted for publication).

## 2. Sample preparation and measurement procedure

Quartz from the studied site in Oman was used to carry out the $D_E$ distribution experiments on simulated insufficiently bleached samples, and for sediment dating. To extract the coarse grain quartz (90–200 μm), the material was first wet sieved, followed by a treatment with HCl and $H_2O_2$ to remove any carbonates and organics. Density separation with lithium-heteropolytungstate was used to separate the quartz from any heavy minerals ($>2.75\,g/cm^3$) and from feldspars ($<2.62\,g/cm^3$). Additionally, the sample was etched in 40% HF for 60 minutes to remove any potential feldspar contamination and also the alpha irradiated outer layer of the quartz grains. All sample preparations were done in subdued red light ($640 \pm 20$ nm).

Luminescence measurements were carried out on a Risø-Reader TL/OSL-DA-15, equipped with blue LEDs ($470 \pm 30$ nm) for stimulation, a Thorn-EMI 9235 photomultiplier combined with a 7.5 mm U-340 Hoya filter (290–370 nm) for detection. Irradiation were from a $^{90}Y/^{90}Sr$ $\beta$-source ($9.16 \pm 0.4$ Gy/min).

For $D_E$ determination the single aliquot regenerative dose protocol (SAR) proposed by Murray and Wintle (2000) was applied. To define the samples' dose–response, six regeneration cycles were used. Shine-down curves were measured for 20 s at elevated temperatures (125 °C) after a preheat of 240 °C (10 s) for the natural and regeneration signals and 160°C for the test dose signals. The integral of 0–0.4 s of the shine-down curves, after subtracting the background signal from the mean of the 16–20 s integral, was used for $D_E$ determination. Feldspar contamination of the aliquots was checked by stimulating the sample with infrared light (IRSL) after artificial dosing.

All measurements were carried out on small multiple grain aliquots containing ca. 200 grains per aliquots (Fuchs and Wagner, 2003), mounted in aluminium cups (12 mm diameter) using silicon oil.

The dose rate ($\dot{D}$) for OSL age calculation was determined by low level $\gamma$-spectrometry. Cosmic-ray dose rates were calculated according to Prescott and Hutton (1994). The water content of the samples was determined using the average value of the possible water content range, based on the porosity of the samples. An error for the water content value was chosen, which included the possible water content range. The values used for the water content were checked by measuring the in situ water contents of the samples, showing conformity within errors.

## 3. Experimental design

To generate artificial insufficiently bleached sediments, the quartz extract was first placed in a monolayer for several days behind window glass in a sunlit location and then put under a solar lamp (Hönle SOL2 filtered by a UV-filterglass) for 180 minutes to reset any potential luminescence signal and to create equal starting conditions. Afterwards, the material was divided into six portions, each of which was $\gamma$-irradiated ($^{60}Co$) with a different dose of 5.9, 7.8, 9.8, 12.7, 15.6 and 20.5 Gy, respectively. To simulate insufficient bleaching, the $\gamma$-irradiated quartz portions were mixed under subdued red light to get three different insufficiently bleached samples (Blend_1, Blend_2, Blend_3) with different $\gamma$-irradiated compositions (Table S1). In addition to these simulated insufficiently bleached samples, a non-mixed sample (Ref_1) composed of grains that had received only one dose was used as a reference for a well bleached sample.

## 4. Results and discussion

### 4.1. Dose recovery test

First of all, the quartz extracts from the study area were tested for their OSL suitability. Therefore, a dose recovery test was carried out on four natural and bleached samples, each given a different $\beta$-dose between 6.0 and 22.5 Gy (Table S2). 24 aliquots were measured for each sample. As Table S2 shows, for every given dose the $D_E$ could be recovered within their 1σ errors with ν in the range of 5-9%. Thus, despite the problem of insufficient bleaching, the quartz sediment from the study area seems suitable for OSL dating.

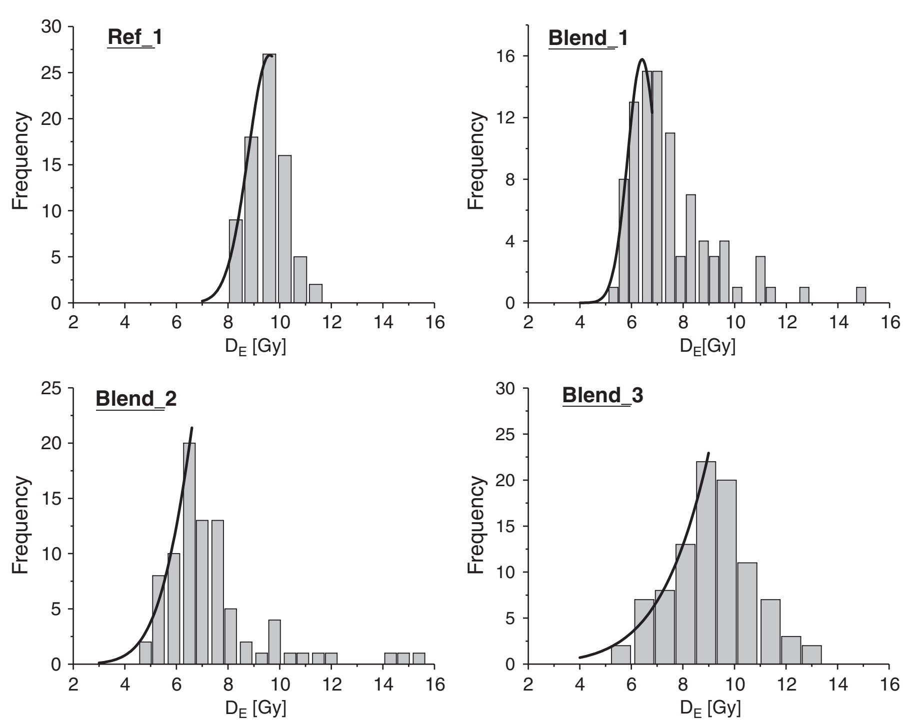


Fig. 1. $D_E$ histograms for the artificially generated insufficiently bleached samples and for the simulated well bleached reference sample (Ref_1). The black curve indicates the Gaussian fit to the rising limb of the dose distribution using the model of Lepper and McKeever (2002).

### 4.2. Simulated insufficiently bleached samples

$D_E$ distributions for the simulated insufficiently bleached samples, and for the well bleached reference sample (Ref_1), based on ca. 100 measured aliquots for each sample are shown as histograms in Fig. 1. The well bleached sample, Ref_1, shows a tight normal distribution with $\nu = 8\%$ (Table S3), confirming results reported earlier by for example Wallinga (2002b), who derived tight $D_E$ distributions for well bleached quartz material. For the simulated insufficiently bleached samples the dose distributions are positively skewed in the case of sample Blend_1 and Blend_2, but normally distributed for sample Blend_3. $D_E$ distributions are broad for all of the insufficiently bleached samples with $\nu$ larger than 18% (Table S3). Thus, insufficient bleaching does not necessarily manifest itself in a positively skewed distribution as assumed by Lepper and McKeever (2002), but can also cause a rather symmetrical but broad distribution compared to a well bleached sample (Fuchs and Wagner, 2003).

Comparing the $D_E$'s derived from applying the methods proposed by Lepper and McKeever (2002), Singhvi (Juyal et al., in press) and Fuchs and Lang (2001) to the simulated insufficiently bleached samples, the lowest dose fraction of 5.9 Gy from the $\gamma$-dosed mixtures (Table S1) was derived within $1\sigma$ errors for sample Blend_1 and Blend_2 by all of the methods. In case of Blend_3, the lowest dose of 5.9 Gy could only be derived within $1\sigma$ errors from the Fuchs and Lang (2001) method, whereas the Lepper and McKeever method (2002) shows a large $D_E$ overestimation. This is due to the fact that Blend_3 shows a broad and symmetrical distribution, where the inflexion point of the Gaussian fit lying beyond the $D_E$ with the highest frequency (Fig. 1). Thus, the Lepper and McKeever method (2002) is not applicable for non-positively skewed distributions. Taking into account the $2\sigma$ errors, again all methods were successful in deriving the lowest dose of 5.9 Gy. However, for sample Blend_2 and Blend_3 the $D_E$ error calculated with the Lepper and McKeever method is, on the $1\sigma$ level, already at 20% and 45%, respectively, thus no reasonable

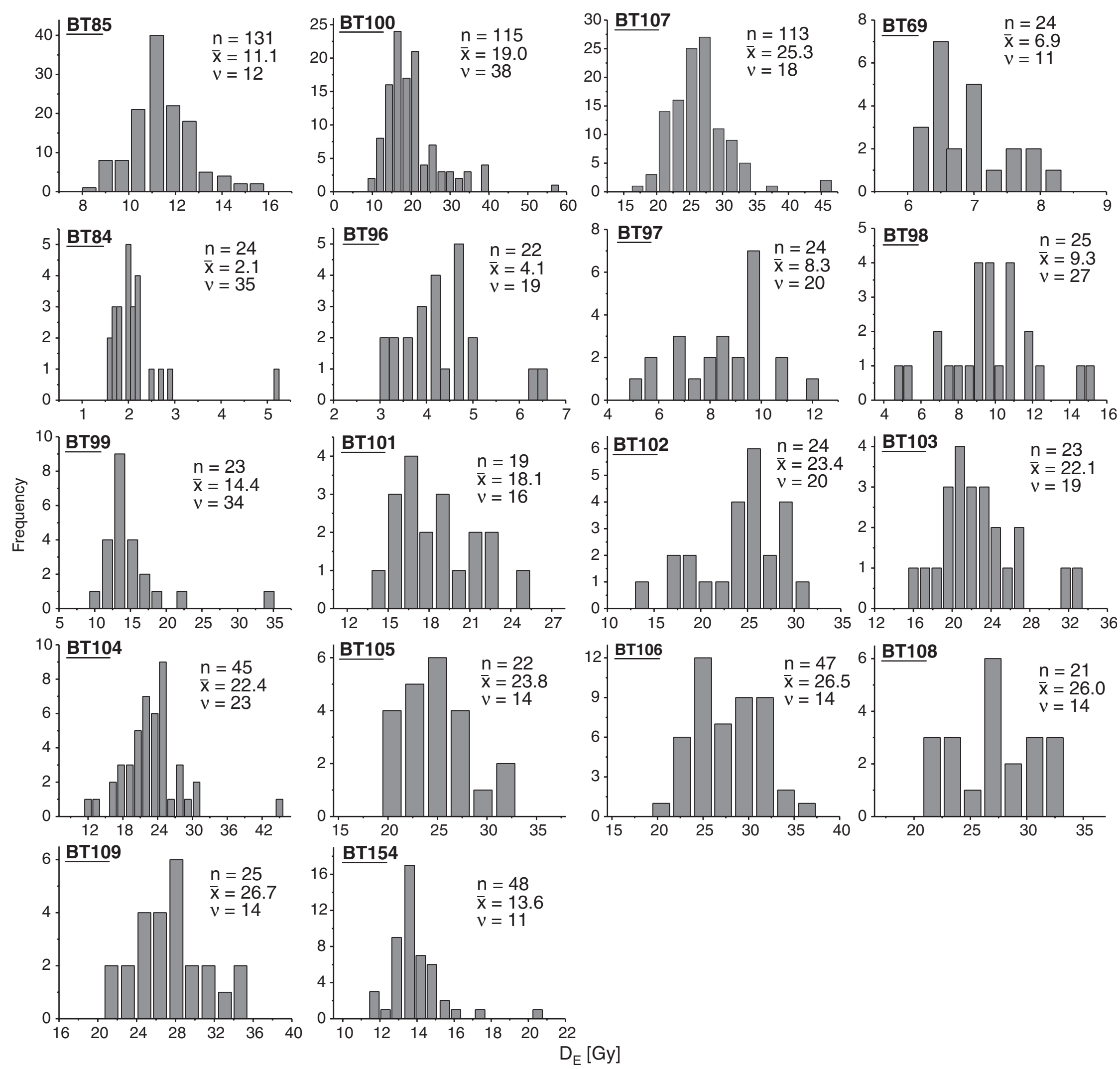


Fig. 2. $D_E$ histograms for all natural samples from the Oman study area. For every sample the number of measured aliquots ($n$), the mean $D_E$ in Gy ($\bar{x}$) and the coefficient of variation ($\nu$) are indicated.

information can be derived from these $D_E$ values because of their large errors.

### 4.3. Natural samples

Based on the experience gained from the artificially dosed samples, $D_E$ estimates were calculated for 18 natural samples from the studied Oman profile. For sample BT85, BT100 and BT107 more than 100 aliquots for each sample were measured. For the remaining 15 samples maximum 24 aliquots per sample were measured, because of the lack of sufficient quartz extracts.

In Fig. 2, the dose distributions of each natural sample are given as histograms with their mean $D_E$ and ν values, with ν always exceeding 11%. Thus, because $\nu > 8\%$ determined from the well-bleached sample, we interpret these natural samples as being insufficiently bleached (Table S2).

No distinct positive skewness (e.g. BT100) could be detected for the natural samples, even if the samples value ν exceeds 11%, thus indicating insufficient bleaching. Instead, the distributions often show a normal (e.g. BT98) or even negative skewness (e.g. BT102). For this reason, the applicability of the Lepper and McKeever method for $D_E$ calculation of insufficient bleached samples is problematic for the non-positively skewed samples. Furthermore, the identification of a distinct rising limb of the dose distributions is not clear (Fig. 2) and the fitting routine for the Gaussian fit is very sensitive to the number of measured aliquots ($D_E$'s) included in the histogram, with only a ca. 20% probability of a successful Gaussian fit using ca. 24 aliquots (for details see Woda and Fuchs, submitted for publication). Due to the above-mentioned difficulties of the Lepper and McKeever method, we could not use this method for $D_E$ calculation of the natural samples.

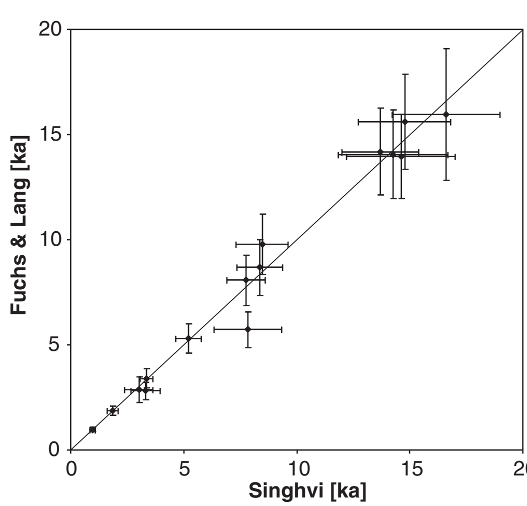


Fig. 3. Comparison of OSL ages based on $D_E$ value calculations after Singhvi (Juyal et al., in press) and Fuchs and Lang (2001), showing good agreement within errors.

We thus used the methods proposed by Singhvi and by Fuchs and Lang for $D_E$ determination and OSL age calculation. The resulting $D_E$'s, the $\dot{D}$-determination and the calculated OSL ages are shown in Table S4. For three samples (BT99, BT102, BT104), no $D_E$ could be calculated with the Fuchs and Lang method, because the experimentally derived ν-threshold was exceeded already using the first two $D_E$'s from the sorted low to high $D_E$ value data set. This is due to low outliers (Fig. 2), which makes this method, based on the quadratic deviation from the mean, very susceptible to failure. The use of either the mean deviation or the absolute deviation from the median (Huber, 2004) might be an improvement, which will be investigated in further studies.

Fig. 3 compares the OSL ages based on the Singhvi and the Fuchs and Lang method, where both methods show for each sample consistent OSL ages within errors. An age versus depth profile using the $D_E$ calculation after Singhvi is shown in Fig. 4. Except for one sample in 6 m depth (BT98), the OSL ages are in stratigraphic order, which is an

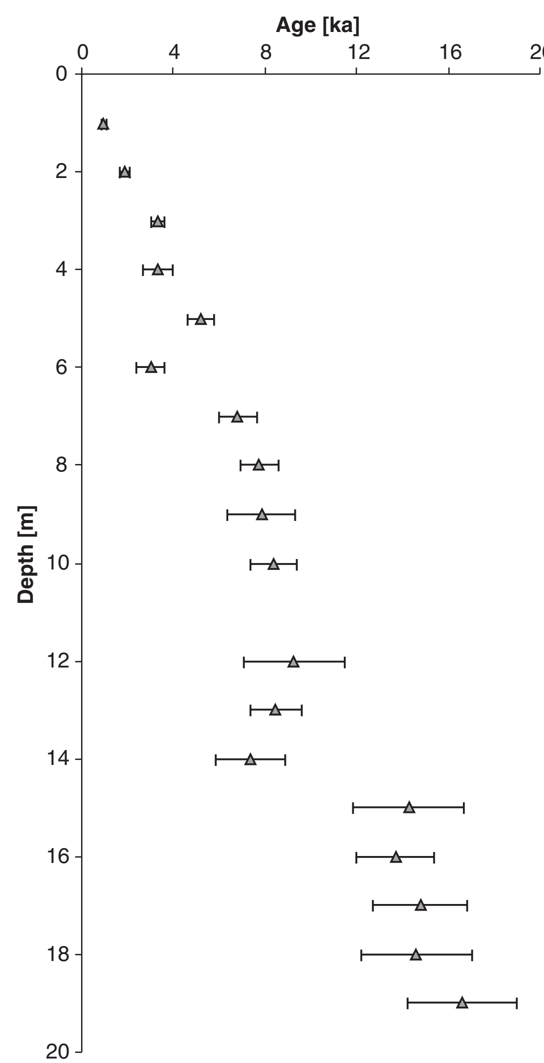


Fig. 4. Age versus depth plot. OSL ages are based on $D_E$ calculations after Singhvi (Juyal et al., in press).

indication of their correctness. The reason for the age underestimation of sample BT98, which could only be identified by intercomparison with the other OSL ages from the profile, is also due to low outliers (Fig. 2).

## 5. Conclusions

Contrary to the criterion of a positively skewed dose distribution, the site specific coefficient of variation $v$ seems to be a suitable parameter to detect insufficient bleaching. The parameter ν has to be derived individually for every locality (Fuchs and Wagner, 2003).

To derive the $D_E$ from an insufficiently bleached sample, the methods proposed by Lepper and McKeever (2002), Singhvi (Juyal et al., in press) and Fuchs and Lang (2001) were compared. Due to the large amount of aliquots needed, the problematic identification of a rising limb and the difficulties with the successful fitting of a Gaussian equation to the distribution, the Lepper and McKeever method does not seem universally applicable. Applying the two other methods, they provide same $D_E$ results within error, but seem to be sensitive to low outliers, resulting in one case in an OSL age underestimation. In three cases, a $D_E$ estimation after the Fuchs and Lang method was not possible due to low outliers. However, with both methods OSL ages were derived which are consistent with stratigraphic order, an indication of their general correctness. Nevertheless, as experiments on artificially dosed and mixed samples have shown, the lowest $D_E$ fraction could not always be derived. Thus, applying the proposed methods above, age overestimation cannot be ruled out if insufficient bleaching is detected.

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## Appendix A. Supplementary Materials

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.quageo.2006.03.006.

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