

# Radiothermoluminescence Dating and Applications to Pleistocene Sediments

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**Abstract.** The article sets out the main tasks facing researchers studying RTL dating and suggests various ways of accomplishing them, that have been developed in the departments of radiochemistry and geomorphology at Moscow State University. The questions under discussion are the formation of the age RTL lightsum in sedimentary rocks and its functional connection with dose-rate and zeroing of paleodosimeters. Methods of RTL dating developed and used by the authors are discussed in detail. The version the RTL method suggested is illustrated by the data concerning the age of the Dniestr terraces. The investigations lead to the conclusion that natural quartz can be employed as a thermoluminescence paleodosimeter for the determination of ages ranging from tens of thousands to 1–2 million years by using the high temperature peaks of the glow curve.

## Introduction

In the recent years most researchers have believed that thermoluminescence (TL) dating or the radiothermoluminescence (RTL) method has a comparatively low upper limit of application (100–200 thousand years). This fact has been more than once discussed at various symposia, conferences and congresses (Vlasov et al. 1980b, 1982, 1984; Vlasov and Kulikov 1983, 1985, 1986). However, there is still controversy on this subject.

In our opinion rapid quartz saturation and the resulting low upper limit of RTL dating obtained by different groups of researchers are the results of individual features of different dating techniques. We find it necessary to describe our methods in great detail, since they allow dating of geological objects within a wider range (between  $n \cdot 10^6$  and  $10^6$  years). Speaking at the latest symposium the authors (Vlasov and Kulikov 1983, 1985, 1986) formulated the main tasks facing the researchers and suggested ways of solving them. They noted that to apply the RTL method of geochronology successfully it is necessary:

1. to develop a physical model for the paleodosimeter under natural and laboratory conditions with regard to the efficiency of the accumulation of the lightsum on exposure to different sorts of radiation on the one hand and on the other, the concentration, migration and distribution of radionuclides in geological environments as well as the concentration, distribution and size of paleodosimeters etc.

2. to develop a physical model for the determination of the quantitative and qualitative factors which control lightsums in sediments.

3. to develop a geological model of the formation of sediments of different genetic types from the point of view of determining the level of "residual" TL at time-zero (level of "Zero-moment") and suitability for RTL dating.

4. to develop a physical model for the description of the factors determining the measured effect (RTL lightsum) under natural and laboratory conditions.

5. to determine the RTL parameters of paleodosimeters and research the methodology of correct calibration in laboratory tests.

The problems raised have been dealt with, to various extents, by different researchers. The relevant information can be obtained, for instance, from the review by Wintle and Huntley (1982) and in Aitken's monograph (1985). This paper presents a method of resolving these problems that has been developed in the Department of Radiochemistry and Geomorphology of Moscow State University between 1977–1986.

## Formation of Age RTL-Lightsum in Sedimentary Rocks

The size of the RTL-lightsum ( $S$ ) is a fundamental parameter, which that provides gives the clue to the age of the sedimentary rock in the RTL dating; it can be measured absolutely (quanta per gram) as well as relatively (relative unit per gram). The paleodosimeter is usually a single mineral fraction, for instance, of quartz with definite average grain size. Consequently, the observed value of the lightsum is the additive value:

$$S = \sum_{i=1}^n S_i \quad (1)$$

where  $S_i$  is the lightsum of selected grains,  $n$  the number of grains in a sample. Obviously, one and the same value of  $S$  can be realized under different distributions of the grains of the paleodosimeter in lightsums within the range  $S_i=0$  to  $S_i=S$ .

The lightsum saturation ( $S_\infty$ ) is an important property of a paleodosimeter, being the limiting value of the lightsum, that cannot be exceeded for a given level of dose rate (Vlasov and Kulikov 1977). The saturation level ( $S_\infty$ ) for a quartz dosimeter is achieved by a dose of about several M rad ( $10^6$  rad), which corresponds to a period of radiation of several million years under natural conditions. Consequently, in bedrocks, paleodosimeters are at the dose dependant saturation level, corresponding to the dose rate found in bedrocks. Figure 1 shows the lightsum formation in sediments. It is assumed that in original bedrocks, a paleodosi-

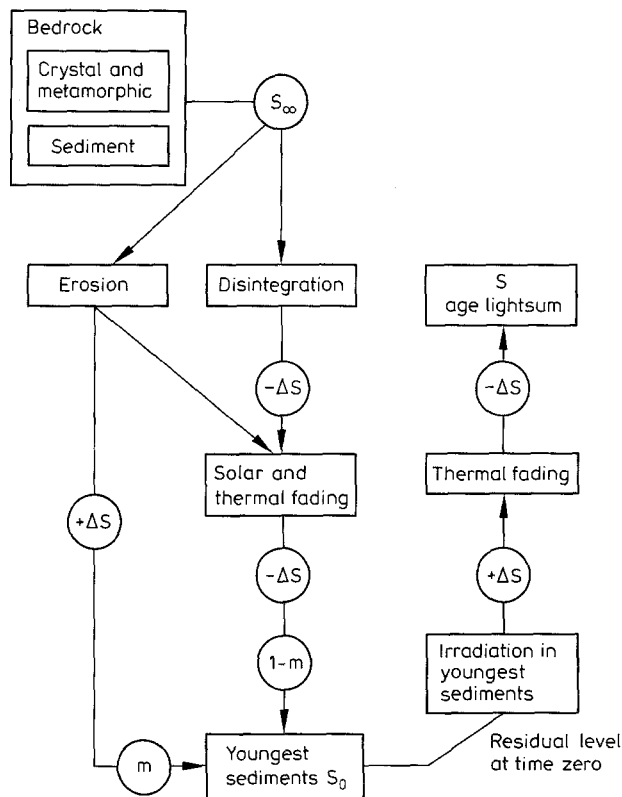


Fig. 1. The formation of the age lightsum in sedimentary rocks.  $-\Delta S$  lightsum loss;  $+\Delta S$  lightsum accumulation;  $m$  fraction of material with saturation lightsum;  $S_0$  lightsum during residual formation

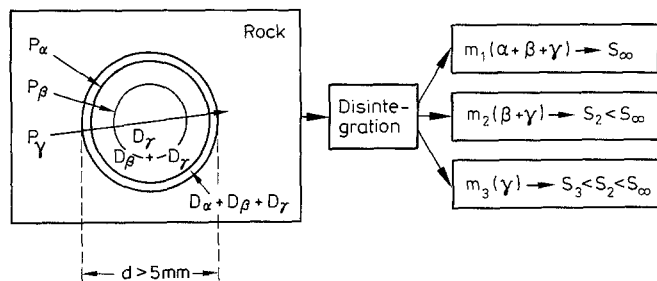


Fig. 2. Lightsum loss during paleodetector disintegration.  $m_1, m_2, m_3$  the masses of disintegrated material, exposed to various types of radiation under natural conditions

meter with grain size  $d \gg R$  (where  $R$  the maximum range of the beta particle emission) is saturated relative to the existing dose fields in bedrocks. The relative material flows  $m$  and  $(1-m)$  have different average lightsums. Lightsum loss at different stages is denoted as  $(-\Delta S)$ , and introduction without lightsum loss, for instance, during erosion or its accumulation by irradiation in sediments is denoted as  $(+\Delta S)$ .

1. Disintegration of Bedrocks (Fig. 2)

The destruction of relatively large quartz inclusions in bedrocks occurs under the impact of mechanical, thermal and chemical factors. In any of these cases reduction in average size of inclusions occurs, which leads to differentiation of

quartz in the saturation lightsum. The difference in the saturation lightsums is linked by the difference in the effective dose rates to quartz inclusions with  $d > 5$  mm due to different ranges of the alpha and beta particles emissions.

During the break down of quartz inclusions from a size of  $d > 5$  mm to  $d = 0.1 \div 0.2$  mm the relative reduction of the lightsum for the given fraction occurs due to the change of radiation conditions (without additional factors), i.e.:

$$f(\text{dis}) = \frac{S(\text{dis})}{S} < 1 \tag{2}$$

Depending on the effective dose rate before and after disintegration, the initial size of the inclusion and paleodosimetric parameters, the average value of  $S(\text{dis})$  lies between 0.5 to 0.75. It should therefore be noted, that during the disintegration process, effective reduction of RTL lightsum occurs without any additional factors. This conclusion was first made by the authors and is of great significance for the comprehension of processes of initial lightsum formation in sediments with high speed accumulation (moraine, proluvium).

2. The Lightsum Loss by Insolation

Previously we showed that in laboratory and natural conditions; solar fading as well as thermal fading for individual regions are the main zeroing factors (Vlasov et al. 1979). The reduction of the lightsum (fading) depends on the intensity and the duration of the zeroing process.

In the first approximation, without taking into account the lightsum accumulation under solar irradiation and without a second capture of delocalised charge carriers, fading can be described as follows:

$$S = S_{\infty, n} \exp(-\eta t) \tag{3}$$

where  $S_{\infty, n}$  is the lightsum saturation of the paleodosimeter under natural conditions;  $\eta$  effective fading rate;  $t$  exposure time of erasing factors. Approximations taken for fading description do not affect further conclusions. It showed, that under natural conditions the residual lightsum at time zero is a function of the sediment's genesis and the dosimetric parameters of paleodosimeters (Vlasov et al. 1981 a). The age equivalent of the residual (progenetic) lightsum depends on the genesis of the sediments and ranges between several thousand (alluvium and loess) and several hundred thousand (moraine, proluvium) years (Vlasov et al. 1977, 1979, 1980 b).

It should be noted, that the idea concerning the role of genesis in residual formation in paleodetectors is shared by our colleagues from the Institute of Geology of the Academy of Science of the Estonian Soviet Socialist Republic. Raukas et al. (1985), who arrived at similar conclusions while dating various genetic sediments of Estonia and Tadzhikistan.

3. Erosion of Bedrocks

Various types of erosion (water, wind) cause erosion products with different zeroing factors affect mg newlyformed sediments. Obviously, the materials can be distinguished by sedimentation rate and hence, by transportation time and time of exposure to the zeroing process. The mass of material introduced into a sediment with mean rate  $V_i$  is:

$$m_i = V_i t_i \tag{4}$$

where  $t_i$  is the time of sediment accumulation, hence the average lightsum in a newly formed sediment equals:

$$S = \frac{\sum_i S_i m_i}{\sum_i m_i} = \frac{\sum_i S_{\infty i} V_i t_i \exp(-\eta_i t_i)}{\sum_i V_i t_i} \quad (5)$$

During high rate in material deposition ( $t$  is not enough) we can observe a high level of residual light sum, and vice versa, during low rate the residuals of the material ( $t$  is too large) we can observe a low level of the lightsum. These correlation can be seen in natural conditions (Vlasov et al. 1981).

#### 4. Accumulation of the Lightsum of Paleodosimeter in Sedimentary Rocks

The exposure of the non-saturated paleodosimeter to ionizing radiation of natural radionuclides and cosmic radiation results in the formation of the new lightsum RTL in sedimentary rocks. We have earlier investigated this problem in detail in a number of works (Vlasov and Kulikov 1977, 1985, 1986; Vlasov et al. 1979, 1982).

Some of the principal features of the determination of effective dose accumulation in paleodetectors, used in this paper, are:

1. The  $\gamma$ -radiation dose rate was determined with gradual distribution of  $\gamma$ -radiating radionuclides of uranium and thorium, and  $^{40}\text{K}$  in quasihomogeneous media with multiple dispersion of  $\gamma$ -radiation taken into account. The impact of the dispersed radiation on the detector was allowed for together with dose accumulation factors for infinite media ( $B_{D, \infty}$ ). The calculation method applied gives some results, that are higher in value than the data, obtained by Nambi and Aitken (1986).

2. The  $\beta$ -radiation dose rate was determined by use of medium  $\beta$ -radiation energy of radionuclides under the condition that the detector's diameter is less than the maximum  $\beta$ -radiation run.

3. The average  $\alpha$ -radiation dose for a spherical detector ( $\bar{D}_\alpha$ ) was determined in supposition on the assumption that  $\alpha$ -radiation dose is formed only at the full absorption level of  $\alpha$ -particles ( $R_\alpha$ ). Under equal distribution of  $\alpha$ -radiators in the media surrounding the detector ( $\alpha$ -radiator's concentration in the detector was negligibly small), we have:

$$\bar{D}_\alpha = D_\alpha \cdot K_\alpha = D_\alpha \left[ 1 - \left( 1 - \frac{R_\alpha}{r} \right)^3 \right] \quad (6)$$

where  $D_\alpha$  is the radiation absorbed dose in the  $R_1$  layer of a spherical detector with  $r$  radius.

For example, when  $R_\alpha = 23$  mkm, and  $r = 100$  mkm, we have  $K_\alpha = 0.54$ . The results obtained are in good agreement with experimental data (Vlasov and Kulikov 1977). It should be noted, that when  $r < R_\alpha$ ,  $\alpha$ -radiation dose is equally distributed in the detector.

4. Under real conditions detectors with relative maintenance concentration sample in  $C_1$  are dispersed in quasihomogeneous radiating media, which have a considerable number of defects that are free of radionuclides, cavities, for instance (pores). When such defects are concentrated,  $C_2$ , the  $\alpha$ -radiation dose rate determined for a continuous flawless medium ( $R_\alpha$ ) must be corrected:

$$P_{\alpha(\text{corr})} = P_\alpha \cdot K_1 = P_\alpha [(1 - C_1)(1 + C_2)]^{-1} \quad (7)$$

For example, having detectors in the sample,  $C_1 = 0.05$  and concentrating defects (porousness),  $C_2 = 0.5$ ,  $K_1 = 0.70$ .

5. The relative effectiveness of different types of ionising radiation while affecting RTL detectors of one type, may be determined experimentally and calculated theoretically. Theoretical calculation is based on stochastic model of lightsum accumulation in RTL detectors (Vlasov et al. 1981 b). It was shown theoretically and confirmed experimentally, that relative effectiveness of ionising radiation on RTL detector is determined by the linear energy transition ( $L$ ):

$$K_3 = \exp \left[ -\frac{l}{\Delta E} (L_2 - L_1) \right] \quad (8)$$

where  $L_2$  and  $L_1$  linear transition of the radiation types being compared;  $l$  linear size of elementary energy emission area ( $\Delta V$ );  $\Delta E$  energy absorbed in microvolume  $\Delta V$ .

Determination of  $K_3$  value by equation (8) for  $\beta$  and  $\gamma$ -radiation shows  $K_3 = 1$ , and for  $\alpha$ -radiation with the energy 5–6 MeV  $K_3 = 0.1$ , which are not inconsistent with the adopted values (Aitken 1985).

In all cases the value of the correction coefficient is less than 1 and leads to a reduction of the partial contribution of  $\alpha$  irradiation, i.e.

$$P_{\text{eff}} = P_\gamma + K_1 P_\beta + K_1 \cdot K_2 \cdot K_3 \cdot P_\alpha + P_{\text{CR}}$$

where

$K_1$  – relative content of paleodosimeter in the rock

$K_2$  – relative contribution  $\alpha$ -saturated layer in paleodosimeter

$K_3$  – efficiency of the accumulated lightsum from  $\alpha$  irradiation.

For example, the absorbed dose for a medium with the (average) amount of radio nuclides in equilibrium with the decay product equals (by components):

$U(3_{\text{ppm}})$	$P_\alpha = 8.01 \text{ mGy} \cdot \text{a}^{-1}$
	$P_\beta = 0.34 \text{ mGy} \cdot \text{a}^{-1}$
	$P_\gamma = 0.35 \text{ mGy} \cdot \text{a}^{-1}$
$Th(10_{\text{ppm}})$	$P_\alpha = 7.42 \text{ mGy} \cdot \text{a}^{-1}$
	$P_\beta = 0.26 \text{ mGy} \cdot \text{a}^{-1}$
	$P_\gamma = 0.38 \text{ mGy} \cdot \text{a}^{-1}$
$K(1.2\%)$	$P_\beta = 0.72 \text{ mGy} \cdot \text{a}^{-1}$
	$P_\gamma = 0.27 \text{ mGy} \cdot \text{a}^{-1}$
	$P_{(\alpha, \beta, \gamma)} = 17.75 \text{ mGy} \cdot \text{a}^{-1}$

Effective dose rate calculation for quartz grains with average diameter 0.2 mm and for corrective coefficient of  $K_1$ ,  $K_2$  and  $K_3$ , given above is:

$$P_{\text{eff}} = (0.35 + 0.38 + 0.27) + (0.34 + 0.26 + 0.72) \times 0.70 + (8.01 + 7.42) \cdot 0.70 \cdot 0.54 \cdot 0.1 = 2.50 \text{ "mGy"} \cdot \text{a}^{-1}$$

Taking into account, the contribution of cosmic radiation of  $P_{\text{CR}} \cong 0.2 \text{ mGy} \cdot \text{a}^{-1}$  we obtain:

$$\frac{P_{(\alpha, \beta, \gamma)} + P_{\text{CR}}}{P_{\text{eff}} + P_{\text{CR}}} = 7.96$$

The significance of this fact is obvious, since age is directly proportional to the dose and inversely to the dose rate, i.e. the sediment age would be underestimated by about 8 times taking into account the correcting coefficients. At-

tempts to introduce correcting coefficients were made earlier (Aitken 1985), but not all factors were taken into account. Effective dose rate can be defined "in situ" according to the measured deposit of gamma radiation in effective dose rate (Vlasov and Karpov 1981).

### Dose Effect Functional Link

Generally, the dose-effect functional link has a non-linear character. Non-linearity of dose-dependance and the possible factors causing it have often been discussed (Kaul et al. 1968, Kaufhold and Herr 1968, Zeller 1968, Tite 1971, Fleming 1970, Hutt and Raukas 1977, Hutt et al. 1977, Aitken 1985). However, this question cannot be considered absolutely clear and demands further research. In practical terms, the non-linearity of dose dependence that always makes it difficult to determine the age, derives from theoretical and experimental difficulties in determining dependence parameters that are possibly due to comparatively small accumulated doses; in determining the age of geological objects, the principle of linear approximation is not acceptable.

With our co-authors we (Vlasov and Kulikov 1977, 1979, Vlasov et al. 1981 b) have developed physical models of lightsum accumulation in RTL detectors with due account taken of the rate of accumulation and the rate of loss under natural and laboratory conditions. The model is based on the principle of correlation of probabilities within the limit of microvolume of crystal-phosphorus ( $\Delta V$ ) in an elementary energy emission act (spur) and the presence of an electron hole centre of the given type (in traps or thermoluminescence centre). The process of localization of the charge carriers in traps for this model is described by the equation:

$$\frac{dn}{dt} = (n_0 - n)^2 \sigma_1 P_{\text{eff}} + n^2 \sigma_2 P_{\text{eff}} - n\eta \quad (9)$$

where

$n_0$  – initial concentration of vacant trap centres (traps) in charge carriers;

$n$  – concentration of filled trap centres for the time  $t$  (or for dose  $D$ );

$\sigma_1$  – cross section of charge carriers traps;

$\sigma_2$  – cross section of charge carriers delocalization under ionizing radiation;

$\eta$  – thermal delocalization rate (thermal fading) of localized charge carriers.

The measured response rate of the RTL detector – lightsum ( $S$ ) is proportional to the concentration of the filled charge centres ( $n$ ) and recombination cross section ( $c$ ):

$$S = cnN \quad (10)$$

One can suppose, that the functional dependence of dose-effect for natural RTL detectors depends more on the concentration of the localized charge carriers ( $n$ ) and is far less dependent on the concentration of recombination centres ( $N$ ), because in the case of paleodetectors,  $N \gg n$ .

In this instance equation (9) determines dose-effect and can be applied to certain individual cases. For example, assuming that radiation delocalization after the filling of the traps can be neglected ( $\sigma_2 = 0$ ), we have:

$$n = \frac{(B-A)(1 - e^{-At})}{2P_{\text{eff}}\sigma_1 \left[ 1 - \left( \frac{B-A}{B+A} \right) e^{-At} \right]} \quad (11)$$

It should be noted, when  $t \rightarrow \infty$  ( $D \rightarrow \infty$ ), we have:

$$n_{\infty} = \frac{B-A}{2P_{\text{eff}}\sigma_1} \quad (12)$$

where

$$A = (4n_0\sigma_1P_{\text{eff}}\eta + \eta^2)^{1/2};$$

$$B = 2n_0\sigma_1P_{\text{eff}} + \eta;$$

Equation (11), taking into account equation (10) and (12) can be transformed into:

$$t = A^{-1} \ln \left( \frac{2S_{\infty}n_0\sigma_1P_{\text{eff}} - S(B-A)}{2S_{\infty}n_0\sigma_1P_{\text{eff}} - S(B+A)} \right) \quad (13)$$

where

$S_{\infty}$  – lightsum saturation at dose-rate  $P_{\text{eff}}$ , as under natural conditions ( $D \rightarrow \infty$ );

$S$  – lightsum of the paleodetector at dose  $D > 0$ .

Equation (13) can be used to determine the age of both geological objects by RTL method. If the paleodetector when introduced to the object of dating has a residual lightsum ( $S_0 > 0$ ) correction must be introduced to the age calculation.

In this case equation (13) is as follows:

$$t_0 = A^{-1} \ln \left( \frac{2S_{\infty}n_0\sigma_1P_{\text{eff}} - S_{\infty}f(B-A)}{2S_{\infty}n_0\sigma_1P_{\text{eff}} - S_{\infty}f(B+A)} \right) \quad (14)$$

where  $f = S_0/S_{\infty}$  is determined experimentally for modern objects (sediment) of the same genetic type as the age sample.

Ultimately, the age ( $\Delta t$ ) is determined by the difference:

$$\Delta t = t - t_0 \quad (15)$$

Importantly, age calculation correction for the residual lightsum ( $S_0$ ) cannot be made as the difference of lightsum ( $S - S_0$ ). The parameters of paleodosimeter necessary for age dating ( $n_0\sigma_1; S_{\infty}; f; \eta$ ) can be obtained experimentally. (Vlasov et al. 1980a; Vlasov et al. 1981 b; Karbov et al. 1981; Vlasov et al. 1981 a). Here are the necessary comments:

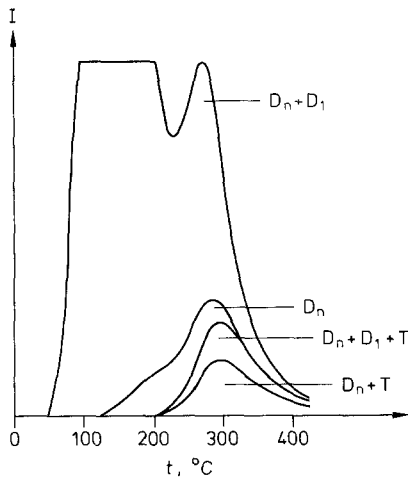
1. Equations (9–15) can be used only to describe the functional dependance of the elementary peak of thermoluminescence. The elementary peak can be determined among other things by the method of isothermal annealing. Figure 3 shows glow curve from quartz for various conditions of irradiation and annealing. Obviously, the glow curve from quartz of a paleodetector irradiated additionally in laboratory conditions by the  $D_e(l)$  dose represents the superposition of a large number of elementary peaks and can be used for its calibration with prior treatment. Isothermal annealing at  $T = 200^\circ \text{C}$  for 20 min makes it possible to determine the age peak at  $T_{\text{max}} = 310^\circ \text{C}$ .

The elementary peak can be verified by the method of ordinate relations (plateau test) or the Alentsev modified method (Karpov et al. 1981), which combines isothermal annealing and a variant of the plateau test.

2. When the detector is exposed to radiation in laboratory conditions, equation (9), taking into account equation (10), becomes:

$$S = S_{\infty, l} \frac{n_0\sigma_1 P_l t}{1 + n_0\sigma_1 P_l t} = S_{\infty, l} \frac{n_0\sigma_1 D_l}{1 + n_0\sigma_1 D_l} \quad (16)$$

where  $S$  is the lightsum of the elementary peak for initial conditions: at  $t = 0$ ,  $D = 0$ ,  $n = 0$  and  $n_0 = n_{\infty}$ .



**Fig. 3.** Thermoglow-curves of quartz paleodetector under different conditions of irradiation and annealing.  $D_n$  glow-curve for natural sample;  $D_n + D_1$  glow-curve for natural sample, exposed to  $D_1$  dose under laboratory conditions;  $D_n + T$  and  $D_n + D_1 + T$  the same samples, exposed to isothermal annealing at  $T=200^\circ$  during 20 min

When the paleodetector is exposed to radiation we have the detector irradiated in natural conditions with  $S_n > 0$  ( $n > 0$ ) and with the unknown  $D_n$  dose. Equation (16) for the particular case is as follows:

$$\Delta S = \Delta S_\infty \frac{n \sigma_1 D_1}{1 + n \sigma_1 D_1} \quad (17)$$

whereby

$$\Delta S = (S_t + S_n) - S_n \quad (18)$$

$$\Delta S_\infty = S_{\infty, t} - S_n \quad (19)$$

Equation (17) has the parameters  $n \sigma_1$ , and not  $n_0 \sigma_1$ .

For determining the parameters  $n_0 \sigma$  and  $S_\infty$  equation (17) reduces to:

$$S = a + b \frac{1}{D_1} \quad (20)$$

whereby

$$a = \frac{1}{\Delta S_\infty}; \quad b = \frac{1}{\Delta S_\infty \cdot n \sigma_1}$$

Taking into account that the concentration of the filled capture centres ( $n$ ) is linked with the initial concentration ( $n_0$ ) by equation (21)

$$n = n_0 \left(1 - \frac{S_n}{S_\infty}\right) = n_0 \frac{1}{a S_\infty} \quad (21)$$

Finally, we obtain:

$$S_\infty = \frac{1}{a} + S_n \quad (22)$$

$$n_0 \sigma_1 = \frac{a}{b} (1 + a S_n) \quad (23)$$

3. The coefficient,  $f = S_0/S_\infty$  is the relation of the residual lightsum level  $S_0$ , with which the detector is introduced to the sediment, to the saturation level under natural conditions ( $S_\infty$ ) at the dose rate of  $P_{\text{eff}}$  linked with the sediment's genesis. The mean values of  $f$  for the peak at  $T_{\text{max}} = 310^\circ \text{C}$  in quartz paleodetectors of 0.2 mm are given in Table 1.

**Table 1.** Mean values of  $f = S_0/S_\infty$  for different types of sediments

Type of sediments	$f$
Loess	$0.09 \pm 0.02$
Lacustrine	$0.13 \pm 0.02$
Alluvium	$0.15 \pm 0.05$
Soil	$0.16 \pm 0.05$
Moraine	$0.27 \pm 0.08$
Glacial	$0.27 \pm 0.04$

4. The rate of thermal fading at  $15^\circ \text{C}$  ( $\eta$ ) was determined by using the parameters of the capture for the peak with  $T_{\text{max}} = 310^\circ \text{C}$  (quartz), which we have obtained experimentally ( $\nu = 1 \cdot 10^{13} \text{sec}^{-1}$ ;  $E = 1.60 \text{eV}$ ) (Karpov et al. 1981). The parameters we have determined are in good agreement with the data for quartz, quoted by Aitken (1985).

Calibration of the paleodosimeter plays an important role in calculating the accumulation dose. Paleodosimeters for calibration must not possess any destroyed electron-hole capture (Vlasov et al. 1980, Vlasov and Kulikov 1977). Any treatment (heating, etching by hydrofluoric acid or ultraviolet irradiation) of the paleodosimeter is inadmissible, because any treatment changes the sensitivity to radiation. Change in sensitivity may lead to error in dose determination. It is practically impossible to take into consideration quantitative changes in sensitivity in the dosimeter draining.

The next important point in the calibration is the correct determination of the dose absorbed by the paleodosimeter. For this purpose  $\gamma$ -irradiation by cesium-137 is best with  $E = 0.661 \text{MeV}$ . Use of other sources of ionising radiation (X-ray,  $\alpha$ -,  $\beta$ -radiation) makes the correct determination of absorbed and effective dose impossible.

The error, which can be equal to  $10 \cdot n$  percent will determine a proportional error in age dose determination and subsequently calculation of the age of the object.

Calibration of the dosimeter is by irradiation of a monomineral quartz fraction of a definite size without mechanical breaking. Irradiation is carried out in identical conditions for each portion of the paleodosimeter.

#### Age Determination of Pleistocene Sediments

The method of RTL dating developed by the authors, has been used in comprehensive analysis of the Issyk-Kul pit section, the Pamir, the Okhotsk sea coast, Moldavia, centre of Russian plain, the Black sea coast and other geographical regions.

To illustrate the potential of the suggested RTL dating method we offer the age determination data of the Dniestr terraces.

In association with Chepalyga (Institute of Geography of the Academy of Sciences, USSR) the authors have studied Pliocene-Pleistocene sediments in Moldavia. The RTL method has been used to date alluvium from the second to the ninth terraces on the Dniestr river. The analysis of the sediments in the second terrace in a section near the village of Karagash has produced 2 RTL datings of  $140 \pm 40$  thousand years and  $142 \pm 45$  thousand, referring to the lower part of the alluvium. The same layers contained residual fauna of Shkurlatov group mammals: Paleodoxodon antiquus, Bos trochocerus, Cerbus alaphus and Karagash

**Table 2.** Parameters of RTL detectors, used for age dating of sediments in Moldavia

Parameters of detectors and sample characteristics	Sample numbers-lab.			
	RTL-225 (TL-11)	RTL-209 (TL-9)	RTL-224 (TL-19)	RTL-230 (TL-20)
Grain size, mm	0.2	0.2	0.2	0.2
Quartz concentration %	97	97	95	72
Accumulative dose effective rate Cy/year	$4.2 \cdot 10^{-3}$	$2.5 \cdot 10^{-3}$	$1.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-3}$
$10^{-6}$ year $^{-1}$	0.2	0.2	0.2	0.2
$a \cdot 10^2$	0.3714	0.3050	0.4733	0.2241
$b$	0.5213	0.2475	1.7437	2.0419
$n_0 \sigma \cdot 10^8$ (Cy $^{-1}$ )	10.91	14.17	4.82	1.63
$A \cdot 10^6$ (year $^{-1}$ )	1.49	2.19	0.79	0.75
$B \cdot 10^6$ (year $^{-1}$ )	5.65	12.10	1.65	1.50
$S_{\infty, l}$ conventional unit	412	377	375	661
$S_{\infty, n}$ conventional unit	315	314	223	382
$S_n$ conventional unit	143	49	164	215
$S_n/S_{\infty, n}$	0.45	0.16	0.74	0.65
$f = S_0/S_{\infty, n}$	0.16	—	0.16	0.16
$t$ , thous. years	$140 \pm 40$	$25 \pm 6$	$1100 \pm 250$	$620 \pm 150$

mollusc Complex (according to Chepalyga) Corbicular fluminalis. The age of the layers bearing these fauna was dated as Mikulin interglacial period.

Alluvium of the third terrace with the Speisk heat-loving group: Carbicular fluminalis, Pseudinio r. speensis and Mammothus chosaricus has been dated  $235 \pm 60$  and  $260 \pm 60$  thousand years.

The age of the alluvium is referred to as the Usunlar Odintsov interglacial period.

The fourth terrace was dated in the Varnitsa section, where the age of alluvium is  $340 \pm 85$  and  $380 \pm 95$  thousand years (Lihvin).

The fifth terrace has been studied most thoroughly in the section near Tiraspol. Alluvium of the terrace contains residual mammalia of the Tiraspol group (Archidiscodon trogohterii, Equus cf siissentornensis, E. cf mosbachensis etc.) and heat-loving mollusc fauna: Pseudonio moldavica, P. robusta. Five datings have been obtained in this section:  $800 \pm 160$ ,  $735 \pm 160$ ,  $710 \pm 160$  thousand years for lower alluvium;  $620 \pm 150$  and  $630 \pm 150$  thousand years for upper alluvium.

The sixth terrace with the first species of Tiraspol mammalian fauna (with Archidiscodon trogontherii) was examined in the section at the village of Mikhailovka. Paleomagnetic analysis of the terrace alluvium, by Trubikhin (Guidebook A-7, C-7, 1982) revealed Brunnes-Matuyama inversion. Above the inversion zone the dating obtained was  $670 \pm 170$  and under it  $900 \pm 200$  thousand years.

The seventh terrace has been studied in the section near the village of Kitskany. Terrace alluvium with Taman mammalian fauna elements (Archidiscodon m. tamanensis) and Kosnitsk group of molluscs of upper Apsheron age (Chepalyga 1967) lying over the estuary lagoon clays with Haramilo paleomagnetic episode was dated  $940 \pm 200$  and  $1100 \pm 250$  thousand years.

Alluvium of the eighth terrace with Odessa mammalian fauna was studied in the section at the village Hadzhimus and dated  $1300 \pm 350$  thousand years.

Alluvium of the ninth terrace (the quarry of the village of Kobusk) with Haprov fauna was dated  $2300 \pm 500$  thousand years.

The RTL datings obtained agree with the terrace levels of the Dniestr river fauna and paleomagnetic data. The large gap in the alluvium age of the 4<sup>th</sup> and 5<sup>th</sup> terraces led Chepalyga to assume the existence of an intermediate terrace.

The age calculation method is illustrated on the basis of special data for 4 different samples (Table 2):

1. RTL-209, modern alluvium
2. RTL-230, alluvium of the 5<sup>th</sup> terrace
3. RTL-225, alluvium of the 2<sup>nd</sup> terrace
4. RTL-224, alluvium of the 7<sup>th</sup> terrace

For RTL dating the sample was taken from the depth of 0.5 m; at the same place the dose-rate of  $\gamma$ -irradiation was measured for further calculation of the effective dose accumulation rate by methods earlier developed.

A polymineral fraction with average grain size 0.2 mm has been obtained from the sample and reached in hydrochloric acid from carbonate and ferrous contaminants, washed by water and alcohol. The quartz fraction was extracted by heavy liquids out of a dry sample. The quartz concentration was determined by mineralogical analysis. The relatively low concentration of quartz in sample RTL-230 deserves to be mentioned.

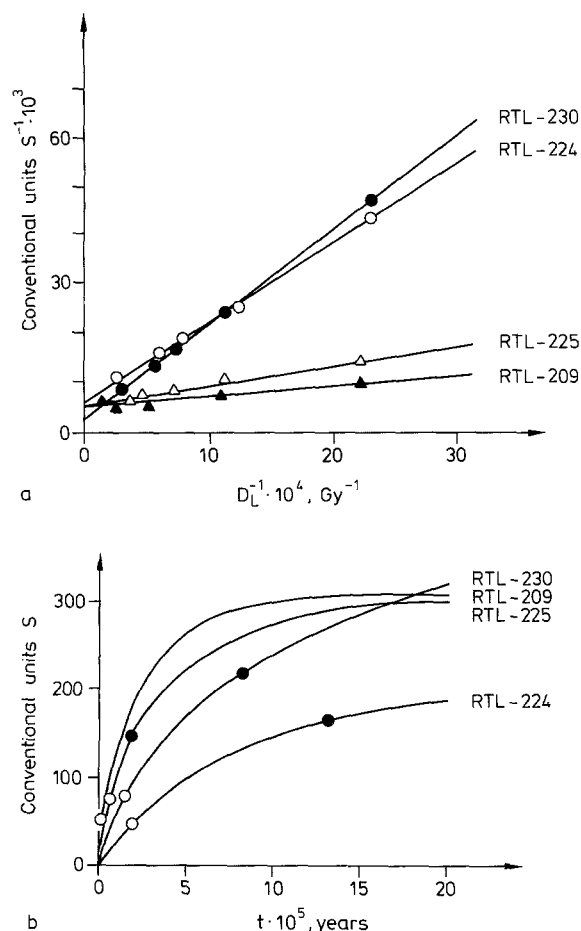
The mineralogic of analysis showed that besides quartz (72%) mica constitutes the main part of the impurities in this sample.

Despite the low quartz concentration, age calculation was also carried out on this sample.

Figure 4a shows the results of the this sample's zeroing according to equation (18). Figure 4b represents the lightsum accumulation calculations under natural conditions from equation (11) with the use of the given data (Table 2). The figure shows that all samples have different functions of the lightsum accumulation. The measured natural lightsum and the residual lightsum position can be seen on the diagrams.

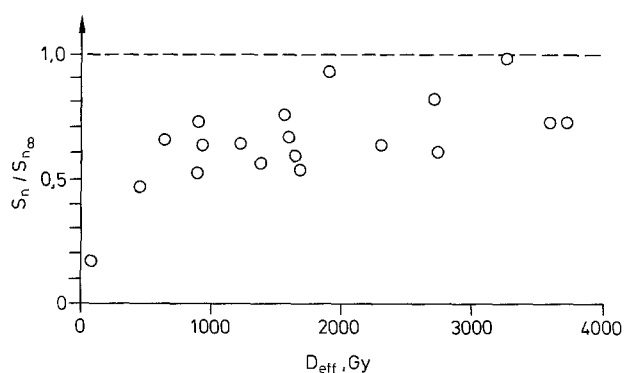
It is obvious that all samples would have an upper limit for age determination much larger than the observed age as the relative filling level of capture centres is far below saturation.

Figure 5 shows the level of relative filling of capture centres for different quartz samples (Moldavia, Pamir, the



**Fig. 4a.** The results of paleodetector zeroing for parameters calculation by equation (20) and that of age by equation (13)

**Fig. 4b.** The reconstruction of paleodetector dose dependence under natural conditions using the parameters obtained in the laboratory. ● the value of age lightsum; ○ the value of residual lightsum



**Fig. 5.** Relative filling of capture centres for various paleodetectors as a function of the effective dose, accumulated in natural conditions

Okhotsk sea coast, the Russian plain). It can be seen, that there is no clear interdependence between the filling level of capture centres and effective dose accumulation, which was to be expected taking into account broad variation of RTL parameters of quartz in nature. It is obvious, that the upper limit for dating is determined by relative filling

level of the captures centres. When  $S_n / S_{n_{\infty}} \approx 1$ , a sample cannot be dated by this method.

The experimental and theoretical research has convinced us that natural quartz can be used as a TL paleodosimeter for an age range of ten thousand (in rare cases – single thousands) to 1–2 million years ( $n \cdot 10^4 \approx 10^6$ ) using high temperature elementary glow curve peaks.

The RTL method should be used as a component of a combined analysis of the sedimentary rocks (nuclear physical methods, paleomagnetic, paleofaunistic, paleobotanic, lithomineralogic, etc.). The detailed dating of the type section is of greatest stratigraphic and geochronological importance.

## References

- Aitken MJ (1985) Thermoluminescence dating. Academic Press, New York, pp 1–359
- Chepalyga AL (1967) Anthropogene freshwater molluscs in the south of the Russian plain and their stratigraphic role. Geological Institute of Geology of the USSR Academy of Sciences 166
- Fleming SY (1970) Thermoluminescent dating: refinement of the quartz inclusion method. *Archaeometry* 12:133–146
- Guidebook of tours A-7, C-7 Moldavia and Odessa regions. The XI INQUA Congress. Moscow, 1982, p 45
- Hutt GI, Raukas AV (1977a) Perspectives of using the thermoluminescence method for determining the age of Quaternary deposits. *Bulletin of Commission for the study of the Quaternary Period*, Publishing House "Nauka", 47:77–83
- Hutt GI, Punning MJ, Smirnov AV (1977b) Thermoluminescence dating and its application to geology. *Estonian Academy of Science* 26:284–287
- Karpov NA, Tarasov MJ, Vlasov VK (1981) The analysis of thermoluminescence curve based on the modified Alentsev method. *Applied Spectroscopy Magazine* 34:243–247
- Kaul IK, Bhattacharga PK, Tolpadi S (1968) Factors in age determination by thermoluminescence of smoky quartz. *Thermoluminescence of geological material*. Academic Press, London, New York, pp 327–340
- Kaufhold G, Herr W (1968) Factors influencing dating  $CaF_2$ -minerals by thermoluminescence. *Thermoluminescence of geological materials*. Academic Press, London, New York, pp 341–354
- Methods of isotope geology (1983) Papers of All-Union Seminar, December 22–27, 1983, Zvenigorod, Moscow, Institute of geochemistry p 213
- Methodology of TL dating (1986) International TL workshop, April 20–24, 1986. Institute of geology Academy of Science of the Estonian SSR, Tallin, p 45
- Nambi KSV, Aitken MJ (1986) Annual dose conversion factors for TL and ESR dating. *Archaeometry* 28:202–205
- Raukas FV, Suslikov VN, Hutt GI (1985) Problems of Thermoluminescent dating of heterogenetic moraine sediments in Estonia and Tadzhikistan. In: "Quaternary geochronology". Papers of All-Union Conference, November 18–21, Moscow, p 109
- Tite M (1971) Some complicating factors and their impact on thermoluminescent dating. *Physics of minerals*, Moscow, Mir publishers, pp 120–133
- Vlasov VK, Kulikov OA (1977) The influence of radiation factors on absolute age determination by thermoluminescence method. *Vestnik Moskovskogo Universiteta. Seriya Geographiya* 6:95–103
- Vlasov VK, Karpov NA, Kulikov OA (1979) The boundaries of applicability of the thermoluminescent method of dating the youngest sediments. *Vestnik Moskovskogo Universiteta. Seriya Geographiya* 4:56–64
- Vlasov VK, Kulikov OA (1979) Additional notes to the methods of thermoluminescent dating of geological objects. *Izvestiya of the USSR Academy of Sciences. Seriya Geologiya* 11:90–97

- Vlasov VK, Karpov NA, Tarasov MJ (1980a) The model of ionizing radiation energy accumulation by solid-state dosimeters. *Physics Magazine* 50:2141–2145
- Vlasov VK, Kulikov OA, Karpov NA (1980b) Comparative analysis of RTL method of dating of geological objects. In: "Field and laboratory methods of studying of glacial sediments". Institute of Geology Academy of Science of the Estonian SSR. Tallin, pp 31–33
- Vlasov VK, Karpov NA (1981) Accumulation rate determination of the dose absorbed in thermoluminescence paleodosimeters. *Izvestiya of the USSR Academy of Science. Seriya Geologia* 9:92–97
- Vlasov VK, Karpov NA, Kulikov OA, Sudakova NG (1981a) Pleistocene sediments in age determination of glacial regions by RTL method. *Vestnik Moskovskogo Universiteta. Seriya geographiya* 6:110–117
- Vlasov VK, Karpov NA, Tarasov MJ (1981) Lightsum accumulation in thermoluminescence dosimeters. *Nucl Energy* 51:108–111
- Vlasov VK, Kulikov OA, Voskresensky SS (1982) Problems of radiothermoluminescence dating for geology. XI-th Congress INQUA, Moscow, Abstracts 2:348
- Vlasov VK, Kulikov OA (1983) Radiothermoluminescence method of dating for quaternary geology. All-Union seminar "Methods of isotope geology", Zvenigorod, Moscow, published by the Institute of geochemistry, pp 182–184
- Vlasov VK, Kulikov OA, Nikonov AA (1984) Age determination of the Pamir quaternary complex based on thermoluminescent analysis. XXVII-th International geological Congress, Moscow 1984, Abstracts 1:421–422
- Vlasov VK, Kulikov OA (1985) Importance of methods RTL dating during complex investigation series of basis sections USSR. The collection of works "Quaternary geochronology". Papers of All-Union Conference, November 18–21, Moscow, p 105
- Vlasov VK, Kulikov OA (1986) Radiothermoluminescent (RTL) method of dating. International TL workshop "Methodology of TL dating", April 20–24, Tallin, p 10
- Wintle AG, Huntley DJ (1982) Thermoluminescence dating of sediments. *Quaternary science reviews*, 31–53
- Zeller EJ (1968) Thermoluminescence of geologic material. Acad. press. Loyola College, Montreal, Canada, pp 519–523

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