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DIVERGENCE IN THE TL DATING RESULTING FROM DIFFERENT METHODS OF EQUIVALENT DOSE DETERMINATION

JAROSŁAW KUSIAK[1], MARIA ŁANCZONT[1], ANDRZEJ BOGUCKYJ[2], and JÓZEF WOJTANOWICZ[1]

[1]*Department of Physical Geography and Palaeogeography, Maria Curie-Skłodowska University, Akademicka 19, 20-033 Lublin, Poland (e-mail: paleog@biotop.umcs.lublin.pl)*

[2]*Department of Geomorphology and Palaeogeography, Ivan Franko National University, Doroszenka 41, 290 000 Lviv, Ukraine*

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Abstract: This paper focuses on problems connected with equivalent dose (ED) estimation. The ED results obtained by using two methods: regeneration (R) and total-bleach (TB) were compared. To this end, ten loess samples were taken from about 20 m thick Zahvizdja profile (western Ukraine).

The obtained results show that the ED values are strongly dependent on the applied technique. When using the regeneration method, the maximum ED value observed is 1051±158 Gy. The total-bleach procedure gives considerably different results, because the gradual increase of ED values is recorded up to 2833±896 Gy. TL ages obtained by the TB method were confirmed by palaeomagnetic investigations.

1. INTRODUCTION

Aeolian sediments such as loess are particularly suitable for the application of thermoluminescence dating method, enabling determination of time passed since the last exposure to sunlight. Wintle and Huntley (1980) used this method for the first time in dating of detrital minerals. Since that time several research groups have conducted tests on accuracy, using different techniques of equivalent dose (ED) determination. The published materials contain conflicting reports on the reliability of TL dating and the upper dating limit (e.g. Berger *et al.,* 1992. Pillans *et al.,* 1996; Frechen *et al.*, 1997 and 1999). Nowadays, this method also arouses controversies among the Quaternary geologists, and research works are still continued to establish a reliable TL sediment-dating procedure.

In this paper we present the TL dating results for 10 samples from the Zahvizdja loess profile (western Ukraine). We compare their TL ages obtained by using two methods of equivalent dose determination: regeneration (R) and total-bleach (TB) methods.

2. LOCATION AND DESCRIPTION OF THE ZAHVIZDJA PROFILE

The studied loess site occurs in the central part of the East Carpathian Foreland. It is situated to SSE of Lviv, near Ivano-Frankivs'k (**Fig. 1**). The East Carpathian Foreland extends between the Carpathian margin in SW and the Dnister river valley in NE. Rivers draining this area rise in the Carpathians. They dissect the Carpathian Foreland on quite a few flat and wast plateau-ridges which are mostly covered by fluvial gravels, evidencing old flows of the Dnister River during the Early Quaternary (Villafranchien), i.e. when the Dnister valley was forming. These plateau-ridges covered by gravels are termed the Lojova planation surface. Valley-sides of the Dnister River and its tributaries have a typical step like arrangement because the Pleistocene terraces occur at different altitudes. Both the interfluve plateau-ridges and the valley sides are covered with loesses.

The Zahvizdja profile is exposed in a large brickyard excavation, so spatial extension of its particular elements could be studied in detail. The profile is connected with a high terrace rising about 80 m above the valley floor of the Bystrycja Solotvyns'ka River. This terrace was formed as a result of dissection of the Lojova planation surface, reaching 360-380 m a.s.l.

The profile is about 22 m thick and it is built mainly of loesses and silty-clayey deposits overlying the marine Krakowiec clays from the Miocene. Three main lithological units can be distinguished in the profile. The upper unit (0-14.5 m) consists of loesses and loess-like deposits; a paleosol occurs just near the ground surface, because of exploitation works. The middle unit (14.5-19.0 m) is silty-clayey, and the lower unit (19.0-20.5 m) is built of fluvial deposits of channel facies (gravels and sands), and flood facies (muds). All these sediments are carbonate-free.

A series of eight paleosols of high stratigraphic rank occurs in the Zahvizdja profile. They represent the Upper Eopleistocene and Mesopleistocene (Łanczont *et al.*, in press).

The upper unit is built of several loess layers separated by paleosols of forest type, usually with traces of denudation in their top parts. Stratigraphy of this part of the Zahvizdja profile was preliminarily defined on the basis of paleopedological criterion and geomorphologic situation, and also in relation to the typical loess profiles in the Podillia (Boguckyj, 1987; Boguckyj *et al.*, 1999). The stratigraphy of the discussed unit is rather complex. First two soils from the top of the profile are single ones; they were connected with the Lublinian (= Rügen) and Mazovian *sensu lato* (= Holstein) Interglacials. In the Ukrainian stratigraphic scheme these paleosols are named the Korshov = Kaydaki and Sokal = Zavadovka soils. A very complicated pedocomplex occurs at a depth of 8 m. It consists of two forest paleosols, the lower of which is better developed and distinguishable by red illuvial horizon; a thin loess layer or a series of fluvial deposits separates these paleosols. This pedocomplex can be correlated with the Ferdynandowian (= Voigtstedt) Interglacial, and it corresponds to the Solotvin = Lubny soil in the Ukraine.

Nature of the middle unit is different. It is almost wholly transformed by soil-forming processes, and consists of four paleosols lying in direct succession; the lowest one is formed on fluvial deposits (flood muds) of the lower unit. Genetic profiles of these paleosols are well developed, and their features indicate that they were formed in wet climate, warmer than the modern one. However, development periods of the successive soils were interrupted by cool stages during which strong cryogenic disturbances affected their A and E horizons. Brunhes/Matuyama paleomagnetic boundary was found within the illuvial horizon of the third paleosol from the top of this pedocomplex (Nawrocki *et al.*, in press).

3. METHODOLOGY

TL age (T_{TL}) is obtained from the equation:

$$T_{TL} = ED/Dr, \tag{3.1}$$

where equivalent dose (ED) is the laboratory dose which produces TL corresponding with natural TL in a sample; annual dose (Dr) is the effective dose received in a unit of time.

Two methods were used for evaluation of the equivalent dose (ED): regeneration (R) and total-bleach (TB) techniques. All measurements were carried out on the 45-63 µm polymineral grain size fraction sieved wet. Possibility of use of similar size fraction was presented by Wintle (1987) who wrote: „the grain size of loess usually has a silt (2-63 µm) content of 50-80% and this has resulted in the application of fine grain (4-11 micron) or intermediate (e.g. 50-56 µm) grain size techniques." The obtained aliquots were pre-treated with 10% HCl and 30% H_2O_2 to remove carbonates and organic material. Optical bleaching was performed using an UV-lamp

Fig. 1. Location of the Zahvizdja profile.

(Osram Ultra-Vitalux 300 Watt) for 16h (Frechen *et al.*, 1992). Laboratory irradiations were carried out at the Institute of Nuclear Chemistry and Technology in Warsaw using a ^{60}Co gamma source. The samples were irradiated with 9 additional gamma doses from 300 to 5000 Gy. Unstable thermoluminescence introduced into the artificially irradiated samples causes underestimation of the obtained TL ages, so the irradiated samples must be heated before readout of the TL glow curves (Berger *et al.*, 1992). Therefore, all aliquots were preheated at 160 °C for 3 hours. All samples were left at room temperature for 2 years after irradiation and for at least 3 days after preheating. The TL glow curves were recorded with use of the RA'94 TL reader-analyser with the EMI 9789 QA photomultiplier. Aliquots were glowed out in an argon atmosphere at a heating rate of 5 °C/sec up to 400 °C. The optical filter BG-28 (380-500 mm) was used (Berger *et al.*, 1992). Dose response was calculated with the FIT-SIM programme of Grün (1994) which is based on the simplex fitting procedures and analytical error calculation by Brumby (1992). Plateau test was carried out for each sample.

The annual dose (Dr) was calculated from the formula:

$$Dr = k\,a\,d_\alpha + d_\beta + d_\gamma + d_c, \quad (3.2)$$

where:
d_α, d_β, d_γ and d_c – doses coming from the α, β, γ and cosmic radiation, respectively; k = 0.14 – α-efficiency factor for grains ≤ 10 μm in diameter; a = 0.5 – a correction resulting from the fact that the 45-63 μm grain size fraction was used.

The alpha particle efficiency can be measured for fine grain samples using a laboratory alpha source, but the contribution of alpha particles to grains over 10 μm in diameter is more difficult to measure but may be estimated – e.g. computer calculations predict that a 50 mm quartz grain would experience only 50 % of the alpha dose received within the same sediment by a grain of 10 μm or less. The beta and gamma doses would be the same for both grain sizes (Wintle, 1987).

Doses d_α, d_β and d_γ were calculated on the basis of concentration measurements of the natural radionuclides: ^{40}K, ^{226}Ra, ^{228}Th, with use of the three-channel γ spectrometer of MAZAR-95 type, assuming equilibrium in the decay series. Radionuclide concentrations (in Bq/kg) were converted to α, β and γ dose rates (in Gy/ka) on the basis of data published by Aitken (1983). Cosmic dose rate was determined on the basis of data published by Prescott and Hutton (1988). Corrections for deposit moisture were taken after Berger (1988). Average moisture of samples in the studied profile was 15±5 %.

4. DATING RESULTS

Ten samples were taken from the studied profile for TL dating: seven from the upper unit and three from the middle unit. TL ages were obtained for nine samples; age of a sample from the lowest paleosol was not determined.

The results of TL analysis are presented in **Table 1**. The TL age of the first paleosol from the top of the profile (sample Lub-3680 from a depth of 1.0 m) obtained by the regeneration method (R) is 150±27 ka. It suggests the Eemian age of this soil. The TL age obtained by using the total-bleach method (TB) is 234±47 ka, and it indicates that this paleosol was formed in the Lublinian (= Rügen) Interglacial.

The TL age of Lub-3681 sample (from a depth of 6.0 m) obtained by using the R procedure (268±42 ka) indicates that this loess was accumulated during the Odranian (= Drenthe) Glacial. The TL age obtained by using the TB method is 532±101 ka, therefore the deposition period of this loess can be correlated with the Sanian 2 (= Elsterian 2) Glacial. Next sample, Lub-3682, was taken from a depth of 8.7 m, i.e. from the third paleosol from the top of the profile. The TL age obtained by using the

Table 1. *Results of TL dating.*

No. Lab.	Depth [m]	Annual dose Dr [Gy/ka]	Method	Plateau [°C]	Equivalent dose ED [Gy]	TL age [ka]
3680	1.00	3.660±0.40	R	250-310	550±77	150±27
			TB	240-380	856±144	234±47
3681	6.00	3.448±0.26	R	250-300	925±130	268±42
			TB	315-400	1831±327	532±101
3682	8.70	2.554±0.23	R	300-400	940±132	368±63
			TB	295-370	1497±269	586±117
3683	9.30	3.166±0.41	R	250-400	980±108	310±53
			TB	270-400	1969±479	622±174
3684	10.50	2,918±0,36	R	290-360	1051±158	360±68
			TB	290-350	1762±460	604±174
3675	12.70	3.356±0.30	R	290-340	940±113	280±42
			TB	280-370	2478±669	738±207
3676	13.70	3.376±0.24	R	300-350	1050±221	311±68
			TB	310-380	2833±896	839±277
3677	15.30	2.917±0.35	R	300-360	950±105	326±52
			TB	295-400	2577±1126	883±398
3678	18.30	3.166±0.38	R	290-350	970±204	306±73
			TB	290-400	2386±916	754±301

regeneration technique is 368±63 ka and corresponds to the Zbójnian (= Domnitz) Interglacial. The TL age obtained by using the total bleach method (586±117 ka) indicates the Ferdynandowian (= Voigtstedt) Interglacial.

Samples Lub-3683 and Lub-3684 were taken from the depths of 9.30 and 10.50 m, i.e. from this part of the profile, which occurs between the third and fourth paleosol from the top. The results of the R method for Lub-3683 sample (310±53 ka) and for Lub-3684 sample (360±68 ka) indicate the Zbójnian (= Domnitz) Interglacial. The TL ages of both samples obtained by using the TB method (622±174 ka and 604±174 ka) can be correlated with the Ferdynandowian (= Voigtstedt) Interglacial.

Samples: Lub-3675 and Lub-3676 were taken from depths of 12.7 and 13.7 m, i.e. from silty loess-like deposit 2.50 m thick. The TL ages for Lub-3675 and Lub-3676 samples obtained by using the regeneration procedure (280±42 ka and 311±68 ka) indicate that this deposit was accumulated during the Odranian (= Drenthe) Glacial. The TL age obtained by the total-bleach method for Lub-3675 sample is 738±207 ka, and for Lub-3676 sample – 839±277 ka, so the deposits can be older than the Sanian 1 (= Elsterian 1) Glacial.

Two last samples – Lub-3677 and Lub-3678 – were taken from the pedocomplex occurring at a depth of 14.5-19.0 m. The results of the R procedures for Lub-3677 sample (326±52 ka) and for Lub-3678 sample (306±73 ka) correspond to the Zbójnian (= Domnitz) Interglacial. The TL age obtained by the TB method for sample Lub-3677 is 883±398, and for sample Lub-3678 – 754±301 ka, so the deposit can be correlated with warm periods preceding the Sanian 1 (= Elsterian 1) Glacial.

5. DISCUSSION

The presented results of TL analysis show how great are the interpretation problems of the TL datings. Two groups of TL ages were obtained for each sample. Each group can be correlated with different glacial or interglacial. For Lub-3680 sample the TL age obtained by the regeneration procedure of equivalent dose determination corresponds to the Eemian interglacial. For the rest of samples (from Lub-3681 to Lub-3684 and from Lub-3675 to Lub- 3678) all TL ages obtained by using the R technique are related to the period from the Lublinian (= Rügen) Interglacial to the Zbójnian (= Dömnitz) Interglacial. It is very important that despite the occurrence of eight paleosols of high stratigraphic rank, a distinct increase of TL age in the profile is observed only to a depth of 6.0 m (**Fig. 3**). From 6.0 to 18.3 m the R procedure gives chaotic results ranging from 268±42 to 368±63 ka. Lack of a systematic and distinct TL age increase with depth arouses our great doubts about reliability of the TL datings carried out using the discussed procedure. The results of complex investigations of the profile, and especially of the paleomagnetic analysis, confirm these limitations. Therefore, the TL ages obtained by using the regeneration method are accidental values, which are not in agreement with geological age of deposits studied (**Fig. 2**).

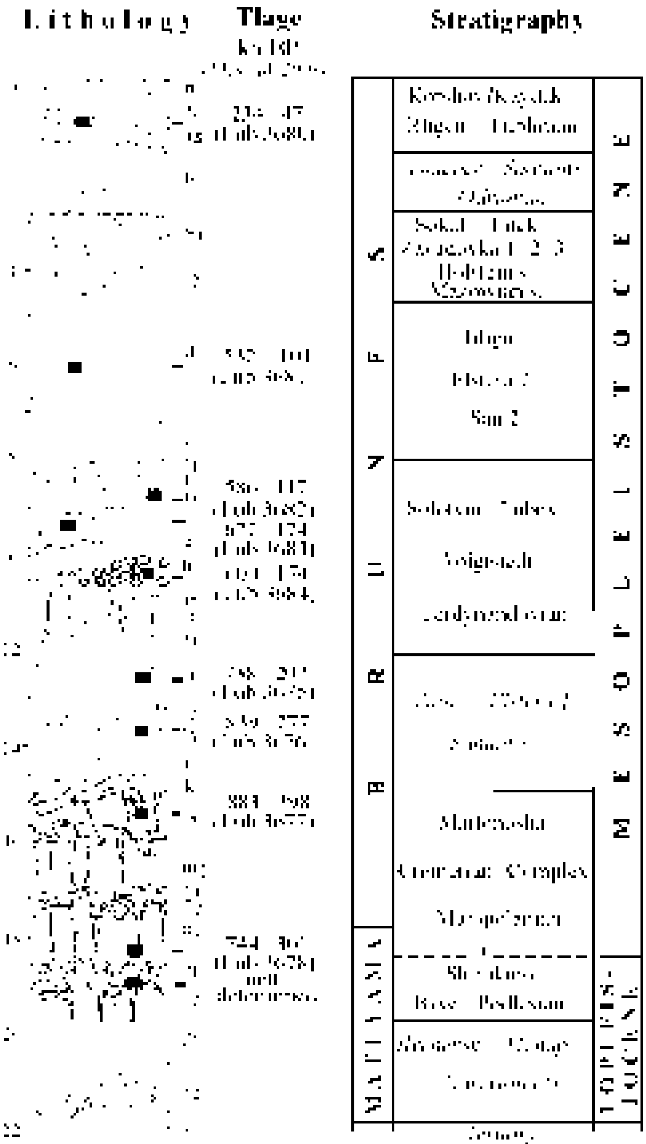

__Fig. 2.__ Lithology, TL ages and stratigraphy of the deposits in the Zahvizdja profile.

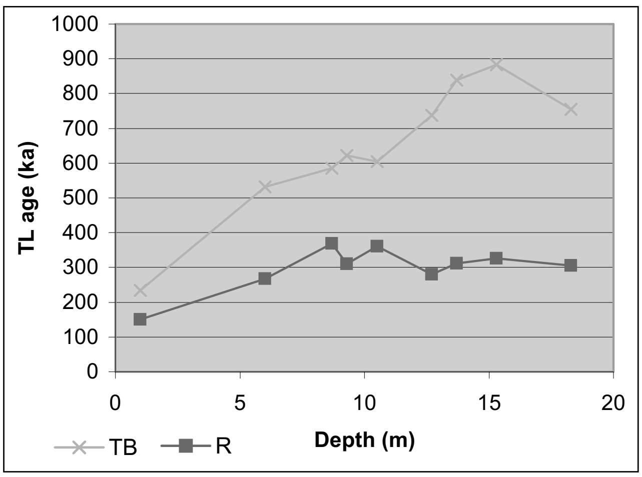

__Fig. 3.__ Comparison of obtained TL ages using different TL techniques.

In order to explain divergences between the deposit age estimated on the basis of paleomagnetic analyses, and also geomorphological, paleogeographical and paleopedological criteria, and its TL age obtained by the regeneration method we should make a thorough analysis of the equivalent dose determined for the particular samples. The ED values, just as the TL ages, distinctly increase with depth only to a depth of 6.0 m (**Fig. 4**). From 6.0 to 18.3 m the ED values obtained by the R procedure change chaotically from from 925±130 to 1051±158 Gy. It may suggest that the presented ED values correspond to the luminescence saturation level of deposits in the studied profile and demonstrate the apparent saturation age for the applied regeneration method. Age underestimation related to use of the regeneration method was noticed by several authors, among others by Berger *et al.* (1992) who found that the regeneration procedure gave age underestimation for samples older than 80 ka, with maximum TL ages of about 150 ka for samples of 500 to 800 ka age. Frechen *et al.* (1999) discovered that a systematic error in ED determination using the regeneration method resulted from a change in luminescence sensitivity of the material which was exposed to artificial or natural light. The upper limit of the ED value which can be obtained by the regeneration method is estimated at 1000-1200 Gy, so the corresponding TL age is 200-250 ka (Frechen *et al.*, 1997).

When estimating results obtained by the total-bleach method, we have quite different situation to that dealing with the regeneration technique. Lub-3680 sample was taken from the paleosol undoubtedly older than the Eemian Interglacial, as it is evidenced by geomorphological situation and paleopedological criterion. Therefore, the TL age obtained by the TB method (234±47 ka) can be correlated with deposition time of the studied sediment (Boguckyj *et al.*, 2001). The result of the TB for Lub-3681 sample (532±101 ka) is in a good agreement with its expected geological age corresponding to the Sanian 2 (= Elsterian 2) Glacial. Three samples – Lub-3682 (586±117 ka), Lub-3683 (622±174 ka) and Lub- 3684 (604±174 ka) – were collected from depths of 8.7, 9.3 and 10.5 m, respectively. They represent the pedocomplex of two forest soils occurring between 8.0 and 10.8 m.

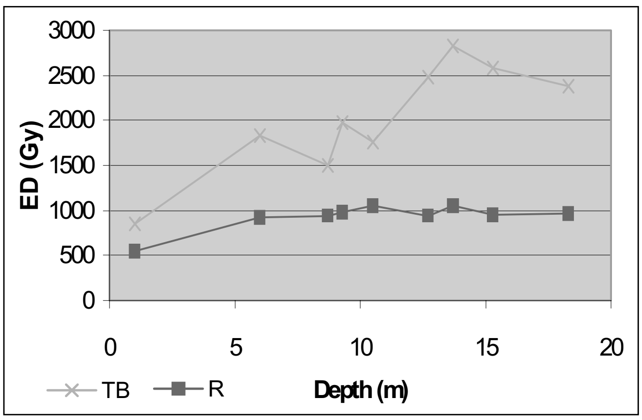

Fig.4. *Comparison of obtained ED results using different TL techniques.*

Lub-3682 sample was taken from the upper paleosol (third from the top of the profile) developed on muds or sandy-gravelly loams of a fluvial origin. Lub-3683 and Lub-3684 samples were taken from a muddy insert within deposits occurring between the third and fourth paleosol. These fluvial deposits are evidently a foreign component in the loess-paleosol profile. Their spatial extent is limited, and we can consider them as an infill of a small transversal valley, which once has developed on a slope and it probably has filled with material denuded from a plateau-ridge surface. The obtained TL ages are generally in a good agreement with geological estimates for the discussed part of the profile, i.e. they correspond to the Ferdynandowian Interglacial. Lub-3675 and Lub-3676 samples were taken from the lowest layer of the upper lithological unit, i.e. from deposit which can be correlated with the Sanian 1 (= Elsterian 1) Glacial. Lub-3675 sample from a depth of 12.7 m was dated at 738±207 ka, and Lub-3676 sample from a depth of 13.7 m – at 839±277 ka. Two next samples were taken from the middle lithological unit, i.e. the pedocomplex of four paleosols occurring between 14.5 and 19.0 m. Lub-3677 sample from a depth of 15.3 m was dated at 883±398 ka. The TL age obtained for Lub-3678 sample from a depth of 18.3 m is 744±301 ka. This sample was collected from the third paleosol from the top of this pedocomplex. Brunhes/Matuyama paleomagnetic boundary dated at 783 ka (Nawrocki *et al.*, *in press*) was found within the illuvial horizon of this paleosol. Therefore, the TL age of Lub-3678 sample obtained by the TB procedure is in a very good agreement with the deposit age determined by the paleomagnetic method. It indicates that the TL ages of three samples (from Lub-3675 to Lub-3677) are overestimated. It is possible to explain these discordances by two reasons: (1) too weak bleaching of mineral material resulting from short transport before sedimentation; (2) too low doses used for irradiation of samples. First reason may be connected with the fact that the studied loess sediments can contain an admixture of material coming from the underlying soils of the middle unit; the first paleosol from the top of this unit is strongly (in places even completely) denuded. Deflated or washed soil material was probably incorporated into the discussed part of the loess cover. Second reason may be connected with ED values for Lub-3675, Lub-3676 and Lub-3677 samples which are the highest in the whole profile and exceed 2500 Gy. Extremely low thermoluminescence intensity in these samples, and too low additional doses (up to 5000 Gy) caused relatively large errors in the ED determination.

6. CONCLUSIONS

The reports on the range and reliability of the thermoluminescence dating method of the Quaternary deposits are conflicting. Some authors think that the reliable and reproducible TL ages can be obtained only for the last 100 ka (e.g. Frechen *et al.*, 1997). Others are of opinion that TL dating may give accurate ages in the range from 100 to 800 ka (e.g. Berger *et al.*, 1992). These controversies induced us to make a series of TL datings with use of two procedures for the deposits of the Zahvizdja profile, which were also dated by other methods. The total-bleach method (TB) gave the results similar to the expected ones. With this method we were able to date samples in the range from 200 to 900 ka. In spite of these promising results, we should be prepared for difficulties when estimating the ED values higher than 2000 Gy because luminescence may reach the saturation level. We hope that further TL analyses planned to be carried out for the Zahvizdja profile will allow us to specify the preliminary results reported in this paper.

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