TL age of Upper Pleistocene stratified deposits measured using the additive, regeneration and R\textsubscript{171} methods

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Stratified sands, which are components of the glacial and interglacial Vilkiškės and Tartokai profiles (Lithuania), were used as research material for experimental dating with the TL method. Sieve analyses of samples collected from stratified sands show varying grain-sizes in the samples, with domination by different fractions in different samples. Four aliquots of grains of the same diameter were selected from each sample for further research by means of the additive, regeneration and R\textsubscript{171} methods. The results showed some consistent patterns, which suggest a direction that future studies could take on these deposits samples. It is necessary, though, to examine grains which comprise a substantial percentage share in the total sample mass.

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INTRODUCTION

According to Malinauskas (1991), the deposits exposed at Vilkiškės in southeastern Lithuania accumulated by saltation of sand in a stream during the Merkinė Interglacial period. Satkūnas (1993), though, suggested that the sands accumulated in a lacustrine basin at the beginning of the Butėnai Interglacial. Detailed structural, textural and compositional study of the Vilkiškės and Tartokai deposits was carried out (Barzdžiuvičienė et al., 2000). Some investigators postulated a relation between older lacustrine sediments from the Butėnai Interglacial and younger ones from the Merkinė Interglacial (Gaigalas and Fedorowicz, 2002).

The Tartokai and Vilkiškės profiles are natural exposures in the sides of the Neris River valley. These exposures are close to each other (Fig. 1). Both sections are more than 30 metres high. They have a similar geological structure and age (Gaigalas et al., 2002, 2005).

In 2001–2003, twenty samples were collected for TL dating purposes from sand deposits found in both profiles: aeolian sand, lacustrine sand and fluvial sand. Figures 2 and 3 show dating results, which have been already published (Gaigalas et al., 2002, 2005) and presented at conferences in Vilnius (Gaigalas et al., 2002) and Ustroń (Gaigalas et al., 2004a). With the support of a research grant from the University of Gdañsk (BW 1230-5-0125-3) I undertook additional parallel dating of five Vilkiškės samples using the OSL (optically stimulated luminescence) method in the Gliwice laboratory (see Table 1). At present the Vilkiškės profile has 41 dates obtained using luminescence methods: TL (in Gdañsk and in Lublin), OSL (in Tallin and in Gliwice) and the \textsuperscript{14}C method (in Gliwice) — see Table 1. There are no significant differences between TL and OSL dates for the youngest aeolian deposits (depth: 2.15–4.70 m). TL and OSL dates are also relatively consistent in the bottom part of the Vilkiškės profile (below 22.4 m). Disparities between TL and OSL dates are found in the middle part of the profile at a depth interval from 13 to 21 m (Table 1). In this part of the Vilkiškės profile there is laminated lacustrine sand.

These disparities inspired the author to undertake methodological work in samples from horizontally laminated sands of both sections. In summer 2004, Gaigalas collected four samples of these deposits from the Vilkiškės (V) and Tartokai (T)
profiles (see Figs. 1 and 2). The grain size composition of near
by samples marked A to D are shown in Table 2.

GEOLOGICAL SETTING

The predominant Quaternary deposits in Lithuania are de-
void of datable organics. One of Lithuania’s characteristic
features is the occurrence of deposits from all glaciations
known in Eastern Europe, with as many as five interglacials
and nine independent till units being found. In the Pleistocene
cover of Lithuania there are seven metachronous till forma-
tions left during independent glaciations or their major stages
(Gaigalas, 1994). These tills are related to advances and re-
treats of the ice sheets of the Käterlai, Dzäkija, Dainava,
Žemaitija, Medininkai and Nemunas glaciations. The glacial
sediments are separated by deposits of normal aquatic (fluvial
and lacustrine) sedimentation which was formed during vari-
ous interglacials: Vind/Char9Eiãnai, Turgeliai, But/c197
nai, Snaigup/c197l and Merkin/c197 (Eemian) and interstadials of the Nemunas
(Vistulian) Glaciation. There occur lacustrine, boggy and al-
luvial sands, silts, gyttja, clay, sapropelite with rare mollusc
shells (Molodkov et al., 2002). The Vilkišk and Tartokai
sections are composed of Snaigup/c197l and Merkin/c197
Interglacial and Nemunas Glaciation deposits. The latter contain till
beds (Gaigalas and Fedorowicz, 2002).

Fig. 1. A — Vilkišk outcrop with results of TL dating (Gaigalas et al., 2002), B — location of the Vilkišk and Tartokai outcrops

G — maximum extent of Scandinavian glaciation — Grüda (Branderburgian, Lesznian) stadial of the Late Nemunas Glaciation; TL ages in ka BP
STRATIGRAPHY

The stratigraphic framework of the Lithuanian Pleistocene has been elaborated by Gaigalas and Satkunas (Gaigalas, 1994). Gudelis’ palaeogeographic studies (1961), Riškienė’s (1979) palaeobotanical data, Kondratienė and Riškienė (1983) and Livrand (1991) contributed significantly to that framework.

Deposits of the last two glacial periods and interglacials are particularly well exposed in Lithuania. These are: the Snaigupė Interglacial (=Lublin), the Medininkai Glaciation (=Warta), the Merkinė Interglacial (=Eemian) and the Nemunas Glaciation (=Vistulian).

Snaigupė Interglacial deposits have been identified in Lithuania. The TL age (250 000±20 000 and 248 000±25 000 years BP) of the Snaigupė Interglacial was determined on the basis of samples collected from fine-grained pale grey lacustrine sand (at a depth of 3 and 9 metres) in the Gvildžiai exposure near Klaipėda in West Lithuania (Gaigalas et al., 2001; Gaigalas and Fedrowicz, 2002). The analysis of Snaigupė Interglacial sections shows that the lake sediments recognized in East and West Lithuania may most likely be attributable to the Drenthian–Warthian ice-free time interval of isotope stage 7. The flora of these deposits has been examined (Livrand, 1991). Palynological examination of the deposits was conducted by Kondratienė (1959, 1996), who concluded that these deposits were younger than the Butnai Interglacial and older than the Merkinė Interglacial. The deposits were correlated with the 7th MOIS (Marine Oxygen Isotope Stage) level. Medininkai Glaciation sediments are represented by boulder clay in Lithuanian profiles. Lacustrine and boggy sediments of this interglacial are widespread in Lithuania and reliably identified. Palynological diagrams have been developed for them (Riškienė, Kondratienė). Eemian deposits from Lithuania have been dated using TL, OSL and ESR methods (Gaigalas and Hütt, 1996; Gaigalas, 2000; Molodkov et al., 2002). The dates obtained with these methods indicate the time span of the interglacial as 120–70 ka BP (Gaigalas, 1994).

Thanks to the numerous TL, OSL and ESR (electron spin resonance) dates and palynological studies, it has been possible to prepare a chronostratigraphy for the last interglacial and last glacial phases (Gaigalas, 1994). The Nemunas Glaciation

For other explanations see Figure 1, TL ages in ka BP
was divided into three parts: an early (70–55 ka BP), middle (55–30 ka BP) and late interval (30–10 ka BP). Initially, the Nemunas interval was nonglacial in Lithuania. The Scandinavian ice sheet moved into Lithuania as late as during late Nemunas time (Gaigalas, 1994), the latter being represented by two stages: Grãda and Baltija.

METHODS

The samples collected from the profiles were dried and then sifted (Table 2). Genetic interpretation is taken from an earlier study (Gaigalas et al., 2002, 2004).

Vilkškės outcrop:
V-D (depth 22.50 m): fine-grained sand with admixture of coarse-grained silt (lacustrine origin, lateral zone);
V-C (depth 19.60 m): coarse-grained silt, sandy (lacustrine origin, slope zone);
V-B (depth 17.70 m): fine-grained sand with admixture of coarse-grained silt (lacustrine origin, lateral zone);
V-A (depth 12.50 m): fine-grained sand, silty (lacustrine origin, lateral zone);
Tartokai outcrop:
T-D (depth 27.00 m): fine-grained sand, silty (lacustrine origin, lateral zone);
The results of grain size analysis (Table 2) show that it is possible to differentiate one or two dominant fractions in all the samples. These fractions refer only to segments in which the grain diameter falls within the range of 80–250 μm. Grains of this size are most frequently used in the luminescence dating procedure. Polish laboratories analyse grains of the following diameters: 80–100 μm (Gdańsk), 125–200 μm (Gliwice), and 45–63 μm (Lublin).

Quartz grains were separated from the sediment. First, they were treated with 4% HCl, organic substances and carbohydrates being cleared within 24 hours. Next the grains were treated with 2% NaOH solution for a similar time. After multiple rinsing with distilled water, the grains were treated with 40% HF solution. This acid removed the external envelope of grains and dissolved grains of other minerals (Bluszcz, 2000).

Portions of grains were prepared from the treated material to measure equivalent doses with three methods: additive method, regeneration method and partial bleach method (RI).

The Vilkiškės section samples were dated in five laboratories in the period 2000–2004 (Table 1). The research was based on luminescence (TL and OSL) and 14C methods. Different types of grains of various diameters were used for luminescence dating. The equivalent dose was measured by a variety of means. The TL method was employed in the Gdański laboratory, which analysed quartz grains with the multiple-portion regeneration method. The Lublin laboratory used the polymerineral fraction which was examined with the partial bleach method. Applying the OSL method, the Gliwice laboratory used quartz grains and the single-portion regeneration method (SAR). The laboratories run by Hütt and Molodkov in Tallin used feldspar and the RI method.

The additive method is the oldest TL procedure. It was used by the Lublin laboratory several years ago. It is not used nowadays; however, it provides a basis for the construction of the “growth” line for the RI method. Figure 3 illustrates the determination of the equivalent dose with the additive method (line “a”) and the partial bleach method — RI (lines “a” and “b”).

The construction of “a” growth line consists in marking the natural thermoluminescence (NTL) level on the chart. This level is determined by the energy stored in grains separated from the sample under analysis. Points on the growth line are derived from the results of “artificial thermoluminescence” (NTL+γ). The γ radiation values are selected by the researcher. The Gdański laboratory exposes samples to cobalt bomb rays. These are measurements of energy obtained by the grains as a result of additional gamma radiation with known doses. The equivalent dose (ED1) is selected as an intersection point of the “a” line with the dose line.

The “a” line is an element for the read-out of the equivalent dose in the RI method (Wintle and Huntley, 1980). In order to determine it, it is necessary to construct the next growth line “b” (Fig. 3). For this purpose all grain portions prepared for the measurement with the additive method should be exposed to a short-term simultaneous treatment with an ultraviolet lamp (UV). This will reduce the energy stored in the grains. The “b” line is constructed in the similar way to the “a” line. The resulting ED2 value is an intersection point of the “a” and “b” growth lines.

In the regeneration method the equivalent dose is specified by measuring the thermoluminescence of grains exposed to a few hours’ exposure to the ultraviolet lamp (UV). This process is called optical bleaching. It results in the maximum reduction of energy in the grains to the “residual thermoluminescence level” (NTL+UV). During growth line construction, the results of residual thermoluminescence and natural thermoluminescence (NTL) are first placed on the chart. Next, thermoluminescence measurements of grains which were earlier bleached and radiated with γ radiation doses of known values (NTL+UV+γ) are represented in the chart. In this way it is possible to reconstruct the signal which was originally contained in the grains analysed (NTL). The growth line (linear or exponential function) allows one to reconstruct the value of energy stored in the grains. The dose absorbed by the grains under laboratory conditions is assumed to be the ED (Fig. 4).

In order to specify the dose absorbed by the grains in the Vilkiškės and Tartokai samples they were radiated with cobalt-bomb rays of 8 values: 50, 100, 150, 200, 250, 300, 350...
and 400 Gy. The same values were used to construct the growth line for all the measurement methods used in the research. The upper value of 400 Gy was selected on the assumption that it would be approximately twice as high as the one stored by the oldest grains. Results of equivalent doses (ED) were divided by dose rates (Dr). The result of this quotient is a TL date. The values of the dose rate Dr (Gy/ka) and TL dates (ka BP) are shown in Table 3.

The annual dose was measured with a MAZAR spectrometer (Fedorowicz, 2003). The $^{40}$K, $^{226}$Ra, $^{232}$Th concentrations were converted into $\alpha$, $\beta$, $\gamma$ radiation doses with the use of coefficients specified by Aitken (Aitken, 1983). Adjustments were also made to account for deposit humidity (Aitken and Xie, 1985).

### RESULTS

Table 3 gives 12 results of TL dates for each sample analysed. The ED was specified for each of the four grain size fractions with the additive (A), regeneration (R), and $R^*$ methods.

1. The age of sample portions shows a positive correlation with the grain diameter (in 11 out of 18 cases).

2. Most frequently, the date specified for a given grain diameter shows the largest values using the additive method and the lowest values for the regenerative method.

3. The majority of results for a given grain diameter obtained with the three methods do not differ significantly. This shows that the grains did not change their sensitivity.

4. It was not possible to date the samples with the $R^*$ method in 8 cases because the calibration curve drawn for the additive method showed saturation. In particular, the saturation condition was shown by the V-C sample. The dose rate was the highest for this sample compared to the others and it amounted to 2.41 Gy/ka.

5. The results obtained for the grains whose percentage was minimal in the entire sample are marked by the high measurement uncertainty, e.g., grains of the 125–160 $\mu$m fraction in the T-D and T-Cs samples. In respect of the T-D sample, the age departs sharply from the remaining results and it is significantly higher (ca. 17%). In respect of the T-Cs sample, it was not specified precisely.

One of the samples (V-C) showed a saturation condition for all the methods and for each fraction.

### DISCUSSION AND CONCLUSIONS

The measurements presented in this article were conducted on samples collected from stratified deposits, the age of which should not exceed 250 000 years. This limitation was to eliminate reservations concerning the upper limit of the equivalent...
The equivalent dose did not exceed 400 Gy for all the samples. Hence, it was three times lower than the one indicated by Frechen (1997) as the limit of regeneration method applicability. The Table 3 results do not show a tendency of consequent age underestimation if the regeneration method is applied to samples older than 80 ka BP (Berger et al., 1992). It is difficult to discuss the results of V-B and T-C samples due to indefinite age values. The remaining results show that the selection of grain fraction for luminescence dating should not be accidental.

TL age results are usually subject to measurement uncertainty which does not exceed 15%. Sometimes differences between the results of one sample divided into four fractions (Table 3) substantially exceed these values. According to the author, prior to the commencement of luminescence dating, a sieve analysis should be the first activity conducted during the examination of sediments of water origin. The second activity should be to eliminate such fractions from further research that have a minor percentage representation in the entire sample. Further examination should focus on the predominant sample in the fraction. The results in Table 3 show that if the sample has two dominant fractions in terms of their percentage share, their results are very similar.

The growth of age along with the increase of a sample grain diameter (see Table 3) may be explained as follows. Larger and heavier quartz grains go down faster than smaller and lighter grains. For this reason, grains of a larger diameter might have had a shorter exposition to solar radiation which was reducing the energy stored in them earlier. Thus, the age of larger-size grains may be longer than that of smaller grains.

It is loess that is the most frequently used material in luminescence methods. The above cited reservations concerning the applicability of regeneration method also apply to loess. The world literature most frequently quotes loess dating results. The largest number of methodical works and studies concern loess. Other types of deposits enjoy much lower interest. Stratified (lacustrine and fluvial) deposits are not always highly regarded as a luminescence dating material by researchers who conduct dating. Dating is most frequently carried out by physicists.

This methodical work was inspired by long discussions of the author (physicist) with the geologist (Gaigalas). The physicist conducted dating and prepared the results. The geologist will determine their value. The grain-size analysis and geochemical analysis were already conducted for the Vilkšės and Tartokai sections. As a result, six lithocomplexes were distinguished, which allowed for the correlation in both outcrops of the Anthropeogen and the Nemunas Glaciation (Rokai section). As a result, six lithocomplexes were distinguished, which allowed for the correlation in both outcrops. The Fifth Baltic Stratigraphic Conference. Basin stratigraphy — modern methods and problems. Vilnius, September 22–27, 2002. Abstracts: 41–43.


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