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GAMMA SPECTROMETRY FOR OSL AND TL DATING OF LOESS DEPOSITS AT DYBAWKA AND TARNAWCE (SE POLAND)

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Abstract: **This article presents a comparison of results from the activity measurements of loess samples obtained with two methods in two laboratories. The methods are semiconductor gamma radiation spectrometry and gamma radiation scintillation spectrometry. Various measurement geometries and various sets of radioactivity standards were applied. Samples were collected from two loess profiles, namely Dybawka and Tarnawce. The results of activity measurements for natural radioisotopes of uranium, thorium and potassium obtained in the Gliwice and Gdańsk laboratories are in good agreement. Additionally, the activity of ^{235}U was measured in the analysed samples.**

1. INTRODUCTION

In addition to the radiocarbon dating method, optically stimulated luminescence (OSL) and thermoluminescence (TL) methods are at present widely used to determine the age of geological formations and archaeological objects (Bluszcz, 2001; Bluszcz *et al.*, 2001; Prescott *et al.*, 1997). In order to determine the age of a given sample with luminescence methods, it is necessary to know the equivalent dose (absorbed dose) and the annual dose (dose rate). The dose rate may be determined in a number of ways. The most frequent methods are TL dosimetry and gamma radiation spectrometry. TL dosimetry involves the use special dosemeters made of LiF, CaF_2, or Al_2O_3 (Bluszcz, 2000). The dosemeters are placed directly in the analysed deposit for a precisely specified time, but not shorter than 3 months (Bluszcz, 2000). The dose rate measurement with the use of the above mentioned dosemeters is a long-term process with a relatively high uncertainty (a dozen or so). Several years ago, LiF dosimeters were used in the Gdańsk laboratory for dose rate *in situ* measurements (Fedorowicz, 1990 and 2003). A dose rate may be also determined with the alpha radiation spectrometry method. However, despite the fact that this method allows the determination of activity in radioactive series, it is not possible to specify the activity of ^{40}K potassium in the sample (Bluszcz, 2000).

To determine the dose rate the gamma radiation spectrometry method may be used. This method allows a relatively easy determination of the natural activity in sample. The activities of ^{238}U, ^{235}U and ^{232}Th series can be determined and also the activity of ^{40}K. Compared to alpha radiation spectrometry, the important advantage of this method is the lack of tedious chemical extraction. Gamma radiation semiconductor spectrometry has the width of a photopeak of about 2 keV, compared to 50 keV for gamma radiation scintillation spectrometry. Thanks to a better energy resolution, it is possible to measure an isotope activity in more detail despite the presence of the Compton continuum, and to specify the degree of disequilibrium in the radioactive series due to the possibility of determination of the activity of respective elements of radioactive series.

In respect to luminescence dating methods, in practice it is assumed that isotopes of uranium and thorium series are in radioactive equilibrium. Radioactive equilibrium disturbances are rather small in thorium series; however, significant disturbances of equilibrium may be noted in the case of uranium series (Olley *et al.*, 1996). The main source of disturbance is the escape of ^{222}Rn, which may escape outside. Another source of disequilibrium in the uranium series is a relatively high mobility of easily dissoluble compounds of ^{238}U and ^{234}U (Hercman, 1999; Oczkowski *et al.*, 2001).

Regardless of the fact which measurement method is selected, the specification of radioisotope activities is subject to measurement uncertainty. If such activities are specified with a gamma radiation spectrometry method,

uncertainty is mainly related to the uncertainty of radioactivity determination and also to the uncertainty of radioactive standards and to the uncertainty of the background counting.

In order to obtain more precise results of isotope activity measurements in the samples to be dated with luminescence techniques, two laboratories: the Gdańsk laboratory from the Department of Geomorphology and Quaternary Geology at the Gdańsk University and the Luminescence Dating Laboratory in the Department of Radioisotopes in the Institute of Physics at the Silesian University of Technology in Gliwice, conducted interlaboratory comparative measurements of samples dated with the OSL and TL methods. For purposes of this study, the laboratories measured the natural radioactivity of samples from two different geological profiles: Dybawka and Tarnawce. The measurements were carried out with two various gamma radioactivity determination techniques and with the application of various radioactivity standards and various measurement geometries.

2. MATERIAL AND METHODS

The following methods were applied: gamma radiation scintillation spectrometry and gamma radiation semiconductor spectrometry. In the case of gamma radiation scintillation spectrometry, the following devices were used: MAZAR-95 and TUKAN. Both MAZAR and TUKAN have NaJ/Tl scintillation detector type SSU 70. MAZAR is a three-channel scintillation spectrometer, which automatically converts counts in the ^{40}K, ^{226}Ra, ^{228}Th channels into concentrations of such elements and alpha, beta, gamma radiation doses. The TUKAN scintillation spectrometer has 4096 channels. Its analog-to-digital converter has a 12-bit resolution. It conducts compensatory conversions with the 10-bit averaging of channel width. Marinelli containers with a capacity of 0.5 dm^3 (a cylinder with r=0.4 dm, h=1 dm) were used as measurement containers. For purposes of gamma radiation semiconductor spectrometry, a coaxial germanium detector of high purity (HPGe) CANBERA type P and of 35% efficiency was used. In this case, samples were measured with the application of two various measurement geometries: Marinelli with a capacity of 0.6 dm^3 and the medium box of 130 cm^3.

Three sets of radioactive standards prepared by CLOR (Central Laboratory for Radiological Protection in Warsaw, Poland) were used as calibration standards. The two sets of standards were used for the measurements with the semiconductor spectrometer, separately for each measurement geometry, and one set of standards was used for the scintillator spectrometers. The measurement time was different for various detectors and ranged from 25,000 to 86,000 s. In the case of MAZAR-95 system, twenty measurements of 2,000 s each were conducted. Prior to the measurements, all the samples were dried in the laboratory dryer and sifted, and then placed in the containers, tightly closed, and seasoned for the period of 30 days to obtain an equilibrium in radioactive series.

Comparative activity measurements were conducted for 9 samples from the two geological profiles: Dybawka (DA) and Tarnawce (TA). Both profiles are located in the vicinity of Przemyśl (SE Poland). The samples analysed are loess formations (Łanczont and Fedorowicz, 2004). Both loess profiles are in close proximity at a distance of approx. 2 km. The TA and DA profiles are outcrops of approx. 20 m thickness. The stratigraphy of the profiles was determined on the basis of their geological examination (Łanczont, 1991). Profiles differ with the time of sediment deposition. Dybawka loess is Vistulian (Upper Pleniglacial), while Tarnawce loess shows a larger time span (from Vistulian loess to Wartanian loess).

Table 1 presents sample weights for various measurement containers and techniques applied. **Table 2** shows the activity of radioactive standards used in the experiment. The isotope concentration of natural radioactive series ^{238}U, ^{232}Th and the concentration of ^{40}K were measured. The activity of ^{235}U isotope was also specified in the samples covered by the analysis.

Table 1. *Masses of samples used in the experiment.*

Sample name	Mass for the Marinelli container, HPGe detector (g)	Mass for the Medium container HPGe detector (g)	Mass for the Marinelli container scintillator detectors (g)
DA-2	806.9	162.72	621.4
DA-5	717.2	168.02	548.6
DA-6	752.7	168.90	609.4
DA-9	853.7	179.41	612.4
TA-1	838.0	163.04	613.4
TA-2	679.7	171.21	496.4
TA-4	816.1	160.72	642.4
TA-14	832.4	169.96	603.3
TA-16	825.6	175.72	625.5

Table 2. *Activity of radioactive standards used in the experiment (in Bq/kg).*

	Detector		
Isotope	HPGe, Marinelli container	HPGe, Medium container	Scintillator detector
^{238}U	2,970	221.2	2,760
^{232}Th	1,016	65.6	939
^{40}K	15,300	885.7	15,823

3. CALCULATION OF ACTIVITY

In the case of gamma radiation scintillation spectrometry, the activity of deposit samples was measured in three intervals of gamma radiation photon energy. Photons from the ^{40}K potassium isotope of 1.46 MeV energy were registered in the first interval (1.26-1.65 MeV); photons from the ^{214}Bi isotope of 1.76 MeV were registered in the second interval (1.65-2.30 MeV) and photons from the ^{208}Tl isotope of 2.62 MeV energy were registered in the third interval. The ^{214}Bi and ^{208}Tl activities that were specified may be adopted as activities in the uranium and thorium series if they are in secular equilibrium.

In contrast to the scintillator method, the gamma radiation semiconductor spectrometry allows one to specify the activity of a larger number of radioactive isotopes. In respect of uranium series, it is not possible to determine the ^{238}U activity because this isotope has a line of a very small efficiency 0.064% of a relatively low energy of 49.6 keV (Papachristodoulou *et al.*, 2003). In addition, it is located relatively close to the line of the ^{210}Pb isotope with the energy of Eg=46.5 keV and efficiency of 4%. In order to specify the activity of ^{238}U series, the 1001 keV line from the ^{234m}Pa isotope may be used (Yucel *et al.*, 1998). The concentration of ^{226}Ra isotope was determined with the line of energy equal to 186.1 keV. The ^{226}Ra daughters activity were determined with the use of the following lines: 295.2 keV (19.2 % efficiency), and 352 keV (37.1 % efficiency) from ^{214}Pb and lines 609 keV (46 % efficiency), 1120 keV (15 % efficiency), and 1764.5 keV (15.9 % efficiency) from ^{214}Bi.

The activity in the ^{232}Th thorium series was specified with the use of ^{228}Ac lines (with energy of 338 and 911 keV and efficiencies of 11.3% and 26.6%, respectively), the radioactive equilibrium between ^{228}Ac and ^{232}Th being assumed. The activity in the ^{232}Th series after ^{220}Rn was determined with the use of ^{208}Tl lines (with energy of 583 and 2614 keV and efficiency of 30.6 and 35.8%, respectively).

Furthermore, the activity of ^{235}U isotope was determined. This isotope has several lines of gamma radiation: 143.8 keV, 163.3 keV, 187.5keV, and 205.3keV with the efficiency of 10.5%, 4.7 %, 53.0 % and 4.7 %, respectively. Besides, the ^{227}Th isotope of this series has lines with energy 93.9 keV and 236.0 keV, but of relatively low efficiency. In respect of ^{219}Rn daughter elements, a relatively efficient line of gamma radiation is the line derived from ^{211}Bi, with the energy of 351.1 keV and the efficiency of 12.9 %. It lies close to the line from ^{214}Pb, with the energy of 352.0 keV and efficiency of 37.1 %.

The activity of ^{40}K isotope in the samples was determined with the use of 1.460 keV line with the efficiency of 10.7 %. It is relatively distant from the other lines, and in particular from the intensive lines of uranium and thorium series isotopes.

4. RESULTS AND DISCUSSION

Figs 1-3 present results of activity measurements in the ^{238}U and ^{232}Th series, and of ^{40}K in soil samples as conducted by the two laboratories. **Table 3** presents the activities of the ^{235}U series isotopes.

Table 3. *The results of ^{235}U activity with the use of HPGe detector (Bq/kg).*

Sample name	Depth (m)	Activity, incl. uncertainty (Bq/kg)	^{235}U/^{238}U Ratio
DA-2	2.25	1.31±0.24	31.1
DA-5	3.45	1.28±0.33	31.6
DA-6	5.15	1.53±0.31	25.3
DA-9	7.95	1.44±0.25	24.9
TA-1	1.60	1.13±0.30	31.4
TA-2	2.80	1.25±0.36	30.0
TA-4	3.70	1.34±0.33	23.5
TA-14	10.50	1.14±0.29	25.1
TA-16	12.60	1.11±0.32	31.8

Table 4. *Comparison of results of activity measurements in ^{238}U series.*

		Weighted mean (Bq/kg)		
Sample	Depth (m)	All laboratories	Semiconductor detectors	Scintillation detectors
DA-2	2.25	39.71±0.82	39.67±0.87	40.0±2.3
DA-5	3.45	38.11±0.85	38.60±0.91	34.4±2.5
DA-6	5.15	37.13±0.79	37.38±0.85	35.6±2.1
DA-9	7.95	35.02±0.68	35.41±0.77	32.1±2.0
TA-1	1.60	34.61±0.72	34.41±0.77	36.3±2.2
TA-2	2.80	36.98±0.87	37.33±0.92	34.4±2.5
TA-4	3.70	31.84±0.70	31.64±0.74	33.5±2.2
TA-14	10.50	28.65±0.67	28.57±0.71	29.3±2.0
TA-16	12.60	33.24±0.76	33.62±0.83	31.4±1.9

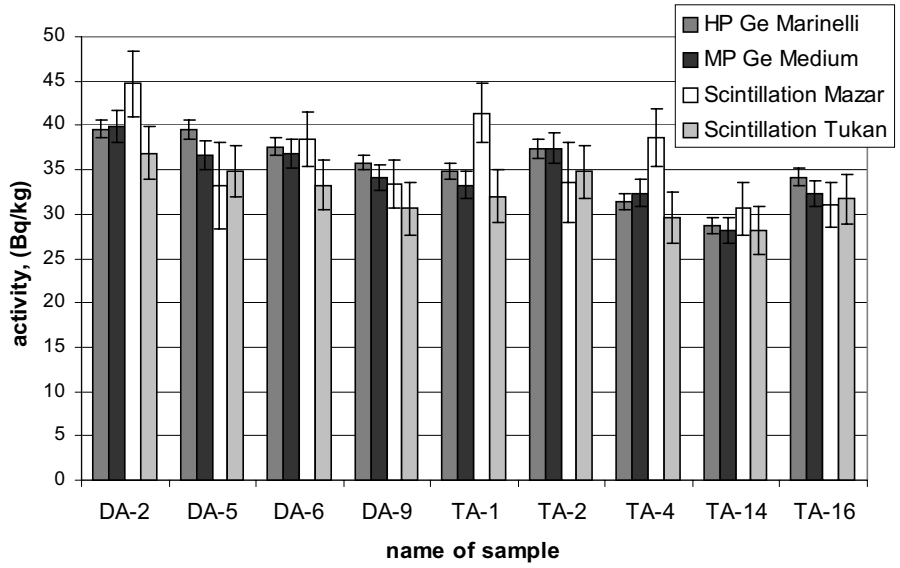

Fig.1. *The results of ^{238}U series activity measurement in the loess samples obtained with different methods.*

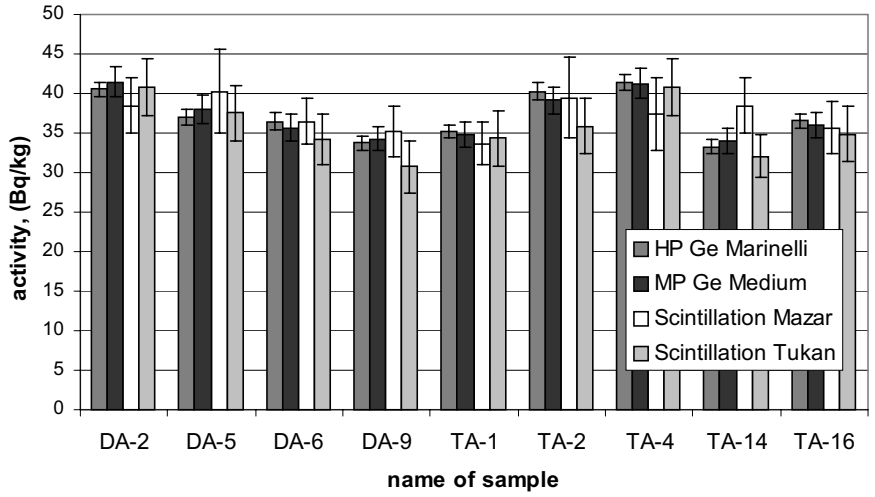

Fig.2. *The results of ^{232}Th series activity measurement in the loess samples obtained with different methods.*

Table 5. *Comparison of results of activity measurements in ^{232}Th series.*

		Weighted mean (Bq/kg)		
Sample	**Depth (m)**	**All laboratories**	**Semiconductor detectors**	**Scintillation detectors**
DA-2	2.25	40.62±0.78	40.72±0.82	39.7±2.5
DA-5	3.45	37.30±0.83	37.21±0.87	38.4±2.9
DA-6	5.15	36.14±0.77	36.25±0.83	35.4±2.1
DA-9	7.95	33.78±0.67	33.84±0.70	33.1±2.3
TA-1	1.60	34.98±0.72	35.11±0.77	34.0±2.1
TA-2	2.80	39.71±0.87	39.98±0.92	37.1±2.9
TA-4	3.70	41.23±0.85	41.40±0.89	39.5±2.8
TA-14	10.50	33.55±0.73	33.43±0.77	34.5±2.1
TA-16	12.60	36.28±0.79	36.40±0.84	35.3±2.4

Table 6. *Comparison of results of ^{40}K activity measurements.*

		Weighted mean (Bq/kg)		
Sample	**Depth (m)**	**All laboratories**	**Semiconductor detectors**	**Scintillation detectors**
DA-2	2.25	578.3±9.1	577±10	587±22
DA-5	3.45	516.7±8.6	518.7±9.6	509±19
DA-6	5.15	510.9±8.6	512.1±9.6	506±19
DA-9	7.95	484.5±7.5	483.8±8.3	487±17
TA-1	1.60	500.2±8.1	499.0±9.1	505±18
TA-2	2.80	561.2±8.8	565±10	551±17
TA-4	3.70	525.1±8.3	526.5±9.6	529±16
TA-14	10.50	525.4±8.5	526.8±9.6	520±18
TA-16	12.60	485.2±7.8	481.5±8.9	498±17

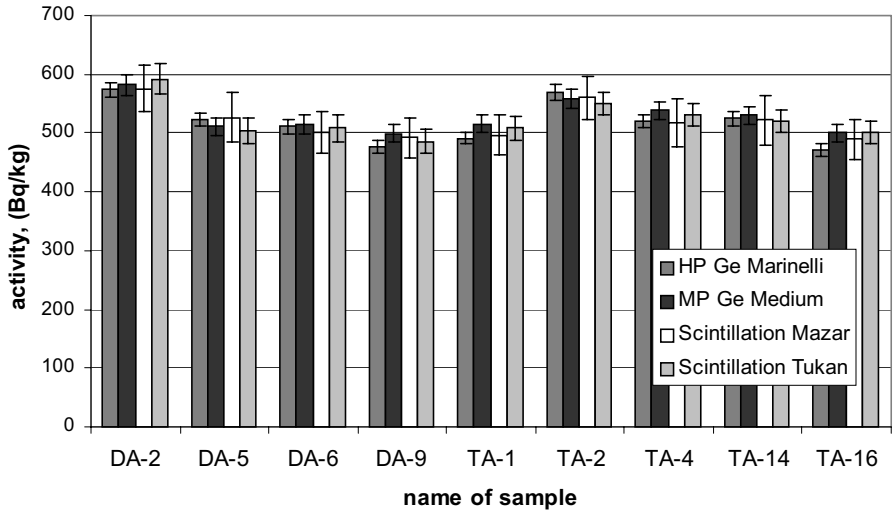

Fig.3. *The results of ^{40}K activity measurement in the loess samples obtained with different methods.*

Comparison of results obtained with various measurement methods

Tables 4, 5, and 6 present the comparison of results obtained in the Gliwice laboratory with the results from the Gdańsk laboratory. These tables present weighted averages for all the measurements of the same sample and weighted averages calculated separately for both laboratories (various measurement techniques).

To compare the figures obtained in the two laboratories with different techniques, scaled deviations were computed for the activity obtained for respective radioisotopes in accordance with the following pattern:

$$Semi\_Scin\_dev = \frac{WM_{sem} - WM_{sc\,\mathrm{int}}}{\sqrt{\sigma_{sem}^2 + \sigma_{sc\,\mathrm{int}}^2}} \qquad (4.1)$$

where:
WM_{sem} – weighted mean for measurements obtained with the semiconductor detector, WM_{scint} – weighted mean for measurements obtained with scintillator detectors, σ_{sem} – standard deviation of weighted mean from measurements obtained with the semiconductor detector, σ_{scint} – standard deviation of weighted mean from measurements obtained with the scintillator detector.

Table 7 presents scaled deviations between the figures obtained with the two measurement techniques.

The results obtained for two laboratories were compared by using the t-Student test. There are no significant differences between the results obtained in those laboratories.

The comparison of weighted averages of results obtained in the two different laboratories with the two various measurement methods shows no clear systematic differences between the results obtained in the two laboratories. Therefore, it may be concluded that there are no significant statistical differences between the results obtained by the two laboratories.

Discussion of the Activity of Dybawka and Tarnawce Samples

The ^{232}Th isotope series in the samples analysed ranges from 33.55±0.73 Bq/kg to 41.25±0.85 Bq/kg (the values of weighted averages of all the values). It is larger than the average value for the area of Poland, which amounts to 20.7 Bq/kg (Jagielak *et al.*, 1998). It may be observed that in respect of the DA series, thorium series isotopes decrease in proportion to depth while no changes of activity in relation to depth were observed in the TA series.

The ^{40}K content ranges from 484.5±7.5 Bq/kg to 578.3±9.1 Bq/kg in the samples analysed and it is slightly higher than Poland's average which amounts to 410 Bq/kg (Jagielak *et al.*, 1998).

The ^{40}K content in the DA series samples shows a declining tendency in relation to the increase of depth. In respect of TA series, this tendency may also be observed, starting from the TA-2 sample.

Table 7. *Scaled deviations of results (see **Eg. 4.1**) obtained by two difference methods.*

		Scaled deviation		
Sample	**Depth (m)**	**^{238}U**	**^{232}Th**	**^{40}K**
DA-2	2.25	-0.129	0.399	-0.450
DA-5	3.45	1.572	-0.398	0.458
DA-6	5.15	0.777	0.361	0.274
DA-9	7.95	1.574	0.302	-0.193
TA-1	1.60	-0.785	0.507	-0.301
TA-2	2.80	1.101	0.967	0.687
TA-4	3.70	-0.818	0.624	-0.127
TA-14	10.50	-0.331	-0.467	0.314
TA-16	12.60	1.104	0.423	-0.885

Olley *et al.* (1997) observe that there is a correlation between the content of ^{232}Th (and its derivatives) and the ^{40}K content. This correlation is also observable in this study; however, it is not as clear as in the above mentioned article.

The content of ^{238}U series isotopes in the analysed samples ranges from 28.65±0.67 Bq/kg to 39.71±0.82) Bq/kg. It is higher than Poland's average, i.e. 25.3 Bq/kg (Jagielak *el al.*, 1998). A small decrease of ^{238}U activity in proportion to the depth may be observed for the both series of DA and TA samples.

The ^{235}U activities in the samples (**Table 3**) do not depart from average values.

5. SUMMARY

The above results present inter-laboratory comparisons of two various methods (scintillation and semiconductor gamma spectrometry) applied to determine a dose rate necessary for luminescence dating methods.

The results obtained with gamma radiation semiconductor spectrometry show lower uncertainty than those obtained with gamma radiation scintillation spectrometry.

The results obtained with the two measurement techniques display good agreement. In the case of ^{40}K and ^{232}Th activity in the samples, deviations between the results obtained with the various methods fall within the tolerance of measurement uncertainty. The difference between extreme values of activity for a given sample does not exceed 10%. It was only in the case of ^{238}U activity measurements for one sample that a slightly larger difference was observed.

Furthermore, the Student's tests did not show that the results of activity measurements in the samples were statistically important; on the contrary, the difference between the results obtained by the two laboratories is not proved statistically.

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