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THE INFLUENCE OF CESIUM ACTIVITY ON THE ANNUAL DOSE FOR OSL DATING

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Abstract: Introduction of single aliquot protocols dramatically improved the accuracy of absorbed dose measurements, so now dating of very recent events, which ages are of the order of tens of years or even single years, becomes possible. The overall accuracy of the OSL age is now limited by the correct assignment of the dose rate. Certain types of recent sediments may contain fall-out radioisotopes that increase the average annual dose. In this paper we demonstrate that the contribution from fall-out radioactivity may be significant and the way to take it into account by using a modified age equation.

1. INTRODUCTION

Optically stimulated luminescence (OSL) can be used to estimate the time elapsed since buried sediment grains were last exposed to daylight. This method of sediment dating makes use of the fact that light releases charge from light-sensitive traps in crystals of minerals such as quartz or feldspar. Establishing the OSL age of sediment basically requires values of two quantities, the absorbed dose of ionising radiation and the dose rate. While the former is measured by an amount of OSL it induced, the latter depends on sample's radioactivity, water content, grain size and on amount of grain removed during etching. Usually cosmic rays contribute also to the dose received by grains, and this contribution depends on geographic co-ordinates, altitude, water content, rock types and burial depth.

Introduction of single aliquot protocols dramatically improved the accuracy of absorbed dose measurements, so now the overall accuracy of the OSL age is mainly limited by the correct assignment of the dose rate.

Very often the annual dose rate is determined by means of gamma spectrometry where the concentrations of particular radioisotopes are measured. The main contribution to the dose rate originates from natural radioactive series (^{235}U, ^{238}U, ^{232}Th) and from ^{40}K (with a small contribution of ^{87}Rb usually added), and also from cosmic radiation. Contributions from other radioisotopes are usually not included. However, recent surface sediments, which were deposited within the last 50 years, were also exposed to the radioactive fallout containing a number of short and medium lived radioisotopes and the radiation released by them may significantly contribute to the total dose absorbed by mineral grains. One of still present and easily detected fallout radioisotopes is ^{137}Cs. Our paper examines the influence of this radioisotope on the dose absorbed by quartz grains in recent sediments. We do not deal with contributions from other fallout isotopes, but their shares may be evaluated in a similar way as presented herein.

^{137}Cs has got to atmosphere mainly as a result of nuclear weapon tests (contributing to a worldwide fallout) and also as a result of nuclear power plants accidents (a regional or continental scale fallout). A significant flux of ^{137}Cs in the atmospheric fallout began in November 1952, and thus our investigation has been applied to samples of sediments deposited within the last 50 years. In this paper we try to answer the question: Is the influence of ^{137}Cs activity on the dose rate significant and how to evaluate it for young sediments?

2. FALLOUT OF ^{137}CS

^{137}Cs became present in natural environment since the first nuclear test on 16th of July 1945, but the first significant amounts of ^{137}Cs released to atmosphere occurred in November 1952. The highest intensity of nuclear weapon tests took place between 1956 and 1965. After this period the fallout of ^{137}Cs is gradually decreasing. The fallout of ^{137}Cs depends on geographical latitude, on the Northern Hemisphere the fallout is greater than on the Southern Hemisphere. The maximum value of ^{137}Cs fallout appears at about 45° latitude North, minimum appears at the equator and is strongly correlated to precipitation (Ritchie and McHenry, 1990). Cesium is transported to soil in several ways: a direct deposition from atmosphere, a wash-off from vegetation, a turnover from vegetation, a redeposition of eroded soil particles and deposition from water on floodplains and coastal regions. Cesium is strongly and readily adsorbed to soil particles, especially to the colloidal fraction. The migration of ^{137}Cs in soil caused by chemical and biological processes is limited. Main processes, which result in movement of ^{137}Cs within soil, are physical processes, the soil erosion or ploughing, for instance. In non-eroded and non-mixed soil ^{137}Cs concentration profiles show a rapid decrease with depth within the first 10 – 15 cm (Poręba *et al*., 2003).

3. OSL DATING

Optically stimulated luminescence (OSL) can be used to determine the time elapsed since certain minerals, such as quartz and feldspar, were last exposed to daylight. It is now widely used in the dating of geological sediments such as aeolian, marine and fluvial sands and muds, loess, and colluvial sediments over the last 200 kyr.

Optical dating is based on specific properties of quartz and feldspars that depend on the existence of defects within mineral crystals and the interaction of electrons with these defects. When the mineral is exposed to ionising radiation, some electrons are ejected from their usual states and some of these subsequently become lodged at specific defects. The larger the dose of radiation, the larger the number of electrons trapped at these defects. Traps fill until all the traps are full or until some other process leads to a state of dynamic equilibrium, or until a sunlight exposure or heating empties them.

Knowing the dose of radiation, to which the sample has been exposed since some event (a paleodose) and the radiation dose, to which it is exposed per year as a result of the radioactive decay of radioisotopes present in the sediment and the dose, which come from cosmic radiation, the age of the sample can be expressed by the simple equation:

$$Age = \frac{Paleodose}{0.90 D_{\beta} + D_{\gamma} + D_{c}} \quad , \tag{3.1}$$

where D_β, D_γ, D_c are beta, gamma and cosmic dose rates respectively. The numerical factor of 0.90 arises because of attenuation of the beta contribution (Aitken, 1998) in coarse fraction grains. If dose rates are expressed as annual doses in Gy per year and the paleodose in Gy than the result is OSL age in years.

Practically annual doses are determined by means of gamma spectrometry or by *in situ* dosimetry. Usually the presence of fallout ^{137}Cs (as well as other radioisotopes – see **Table 5**) will cause underestimation of the average annual dose value (though the underestimation is less severe in case of TLD measurements it still may be significant), and, if not accounted for, may lead to the overestimation of an age.

Presence of ^{137}Cs is easily detected by gamma spectrometry, so below we will estimate the ^{137}Cs dose contribution to the total dose absorbed by the recent exemplary sample.

To calculate values of annual doses we followed an approach presented by Adamiec and Aitken (1998) and used their data. For the radioactive fallout isotopes we used data from the NuDat data base (Dunford and Kinsey, 1998).

Dose rate D (in μGy/yr), when the specific activity of parent in the sample c (Bq/kg) is given, is

$$D = 5.056 \cdot E_p \cdot c \quad , \tag{3.2}$$

where E_p is energy expressed in MeV released in the series per parent disintegration. The numerical factor is based on the relation 1 MeV = $1.602 \cdot 10^{-13}$ J and 1 year = $3.156 \cdot 10^{7}$ s.

Table 1 compares annual dose values (μGy/yr) in a sample with the specific activity of each parent equal to 1 Bq/kg. The value given for full series is for the case of secular equilibrium, i.e. the disintegration rate of each daughter is equal to that of the parent, except where it has been modified by branching. This and other types of disequilibrium have been discussed by Krbetschek *et al*. (1994) and Olley *et al*. (1996).

In further calculations we assume that the mean value of the annual dose of cosmic radiation is 280 μGy/yr (Oczkowski *et al*., 2000).

Table 1. *Annual dose values given in μGy/yr (micrograys per year) for the specific parent activity of 1 Bq/kg. Data for ^{238}U, ^{232}Th and ^{40}K according to Adamiec and Aitken (1998); for ^{137}Cs authors' calculations*

Parent	**Dose rate (μGy/yr)**	
	beta	gamma
potassium[1]	2.70	0.79
thorium	6.72	11.72
uranium[2]	11.32	8.76
cesium	1.24	2.86

[1] Values given for potassium include also ^{87}Rb in a proportion of 50 ppm natural rubidium per 1% of natural potassium.

[2] Uranium data are given for natural uranium containing 0.71% ^{235}U and 99.29% ^{238}U and related to 1 Bq/kg of ^{238}U.

4. ANNUAL DOSE CALCULATIONS

The annual dose contributions from the very long-lived radioisotopes are constant over the range of OSL dating method, but half-lives of fallout radioisotopes are in the range of several decades so the respective dose rates vary significantly within few years. If the initial annual dose is D_{f0} and the half-life of the radioisotope is $\tau_{1/2}$ then the actual annual dose after time T is

$$D_f(T) = D_{f0} \exp\left(-\frac{\ln(2)}{\tau_{1/2}} T\right), \tag{4.1}$$

The average annual dose over this time is then

$$\overline{D_f}(T) = \frac{1}{T}\int_0^T D_f(t)dt = \frac{\tau_{1/2}}{\ln(2)} \frac{D_{f0}}{T}\left[1-\exp\left(-\frac{\ln(2)}{\tau_{1/2}}T\right)\right], \tag{4.2}$$

What we measure in the laboratory is the present activity of the given radioisotope D_{fp}. In this case the activity, and the dose rate, in the past were greater than now. Thus, using the present day value instead of the initial one leads to another equation for the average value

$$\overline{D}_f(T) = \frac{1}{T}\int_{-\min(T,T_0)}^{0} D(t)dt = \frac{\tau_{1/2}}{\ln(2)} \frac{D_{fp}}{T}\left(\exp\left[\frac{\ln(2)}{\tau_{1/2}}\min(T,T_0)\right]-1\right), \tag{4.3}$$

where $\min(T, T_0)$ is a smaller of two values: T – the age of the sediment and T_0 – the age of deposition of the radioisotope.

As an example we have chosen a sediment sample with the following values of specific radioactivities: ^{238}U – 25.2 Bq/kg, ^{232}Th – 20.7 Bq/kg, ^{40}K – 410 Bq/kg. We assumed six different values of initial ^{137}Cs specific radiation: 0, 10, 25, 50, 75, and 100 Bq/kg. Calculations have been made for dry sediment but the relative differences between samples containing and not containing cesium depend little on the water content. We further assume that sediment is homogenous with respect to Cs distribution and thick enough to neglect the finite mean range of gamma rays in it (diluvial sediments for example).

The results of our calculations are given in **Table 2** and shown in **Fig. 1**.

As a second example we have chosen a diluvial sediment profile in Złoty Stok (the Sudetes Mts., Poland). A series of eight samples was collected from the exposure to the depth of 80 cm. ^{137}Cs was detected to the depth of about 60 cm. We have calculated annual doses for this samples using Equations (**3.2**) and (**4.3**), and values of measured concentrations of radioisotopes. Calculations of the average ^{137}Cs dose rate require that the age of the sediment is known. For this purpose we have assumed that the sediment at 50 cm is 50 year old and that the sedimentation rate was constant at 1 cm/yr. **Tables 3** and **4** give details of measured concentrations and calculated annual doses. It may be seen from the presented numbers that including ^{137}Cs in the dose rate calculations rises the annual dose values by about 5%.

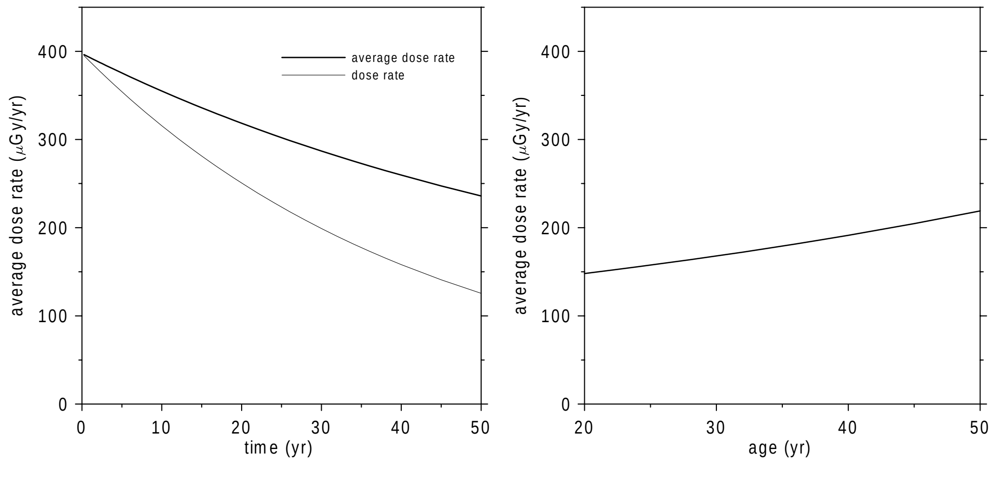

Fig. 1. *Average annual dose contribution of ^{137}Cs. Note that abscissa axes have different meaning on both plots. Left. Changes in the average annual dose when the initial value of specific activity is 100 Bq/kg ^{137}Cs. Lower thinner line shows the actual dose rate. Right. Changes in the average annual dose when the present day value of specific activity is 31.58 Bq/kg ^{137}Cs (i.e., 100 Bq/kg 50 years ago)*

Table 2. *The influence of cesium activity on the average annual dose for the first 50 years*

Age of sample, (years)	**Initial activity of ^{137}Cs (Bq/kg)**									
	10		25		50		75		100	
	Average annual dose and relative difference									
	(µGy/yr)	(%)	(µGy/yr)	(%)	(µGy/yr)	(%)	(µGy/yr)	(%)	(µGy/yr)	(%)
5	2452	1.43	2504	3.50	2591	6.77	2677	9.82	2764	12.68
10	2450	1.35	2499	3.31	2591	6.41	2662	9.31	2744	12.04
15	2449	1.28	2295	3.14	2572	6.08	2649	8.85	2726	11.47
20	2447	1.21	2491	2.98	2564	5.78	2637	8.43	2710	10.93
30	2443	1.09	2483	2.69	2549	5.24	2615	7.65	2681	9.95
40	2441	0.99	2477	2.44	2537	4.75	2596	6.97	2656	9.09
50	2439	0.90	2471	2.22	2428	4.34	2580	6.38	2634	8.33

The annual dose without cesium is 2418 µGy/yr.

Table 3. *Specific radiation of concentrations of radionuclides measured in samples from Złoty Stok*

Sample name	Depth (cm)	Concentrations of radioisotopes in (Bq/kg)			
		U-238	Th-232	K-40	Cs-137
ZS 1	0-10	20.8±1.1	22.69±0.69	794.2±6.2	39.0±1.0
ZS 2	10-20	22.5±1.0	23.52±0.70	844.3±5.6	44.0±1.0
ZS 3	20-30	25.4±1.4	26.43±0.84	949.8±7.8	51.7±1.3
ZS 4	30-40	19.5±1.0	20.76±0.62	702.2±4.9	41.8±1.0
ZS 5	40-50	18.84±0.72	20.12±0.56	753.5±3.7	31.60±0.68
ZS 6	50-60	20.4±1.0	22.00±0.66	769.2±5.9	0.25±0.73
ZS 7	60-70	21.2±1.1	20.78±0.66	743.3±6.5	1.76±0.78
ZS 8	70-80	21.90±0.86	20.12±0.58	730.4±4.5	0.00±0.56

Table 4. *Average dose rates for samples from Złoty Stok*

Sample name	Depth (cm)	Dose rate with cesium (μGy/yr)	Dose rate w/o cesium (μGy/yr)	Difference (%)
ZS 1	0-10	3225	3092	4.3
ZS 2	10-20	3463	3300	4.9
ZS 3	20-30	3892	3686	5.6
ZS 4	30-40	2991	2811	6.4
ZS 5	40-50	3111	2961	5.1
ZS 6	50-60	3038	3037	0.0
ZS 7	60-70	2992	2983	0.3
ZS 8	70-80	2936	2936	0.0

Table 5*. Data on selected radioactive fallout isotopes*

Isotope	Half-life	Amount relative to ^{137}Cs[3]	Total energy released per parent disintegration (MeV)		Total contribution[4] to the dose rate (MeV)
			gamma	beta	
^{137}Cs	30.07±0.03 yr	1	0.565	0.245	0.786
^{90}Sr - ^{90}Y	28.79±0.06 yr	0.66/0.01 [1,2]	0.043	1.130	1.060
125Sb	2.75856±0.00025 yr	0.05	0.434	0.097	0.521
^{134}Cs	2.0648±0.0010 yr	0.61	1.555	0.164	1.703
^{106}Ru	373.59±0.15 d	0.26	0.000	0.010	0.009
^{131}I	8.02070±0.00011 d	2.95	0.382	0.192	0.555

[1] 0.66 for nuclear weapon testes deposition, 0.01 for Chernobyl deposition (Haak and Rydberg, 1998), [2] Total contribution is calculated as ,[3] were taken from Melleander (1986),[4] were taken from Haak and Rydberg (1998).

Cesium ^{137}Cs was not the only radioisotope present in the radioactive fallout. It is however one of the two relatively long-lived. The other is ^{90}Sr which, together with its daughter product ^{90}Y, emits only beta radiation, and thus is less easily detected in the sediment. There is also a number of medium-lived radioisotopes that were present in the fallout, and in the sediments, for up to about 10 years. The most relevant to our purpose information about these radioisotopes are collected in **Table 5**.

5. MODIFIED AGE EQUATION

It is seen from **Table 4** that the presence of radioactive ^{137}Cs isotope in the recent sediments significantly affects the apparent dose rate. It is clear then that this additional contribution must be taken into age calculations. We propose that the age equation (**3.1**) is now rewritten in the following form:

$$\left(D_n + \overline{D}_f\right) T = D_e, \qquad (5.1)$$

where T is an age of the sample, D_n is the total effective annual dose calculated for naturally occurring radioisotopes, $\overline{D}_f$ is the average annual dose contribution of fallout isotope as defined by Equation (**4.3**), and D_e is the value of paleodose established through luminescence measurements. Substituting Equation (**4.3**) into (**5.1**) leads to

$$\left[D_n + \frac{\tau_{1/2}}{\ln(2)}\frac{D_{fp}}{T}\left(\exp\left[\frac{\ln(2)}{\tau_{1/2}}\min(T,T_0)\right]-1\right)\right]T = D_e, \qquad (5.2)$$

and finally to

$$D_n T + \frac{\tau_{1/2} D_{fp}}{\ln(2)}\left(\exp\left[\frac{\ln(2)}{\tau_{1/2}}\min(T,T_0)\right]-1\right) = D_e. \qquad (5.3)$$

The modified age equation (**5.3**) is a transcendental one and may be solved numerically with respect to T.

CONCLUSIONS

The results of this study suggest that one has to expect a significant contribution to the dose absorbed by coarse grains of minerals in recent sediments that contain ^{137}Cs of radioactive fall-out origin. Differences between annual doses with and without the cesium contribution are in the range 3-10%.

The proposed modified age equation (**5.3**) takes into account the transient contribution of fall-out radioisotopes and **Table 1** provides data supplementing Adamiec and Aitken (1998) data with respect to ^{137}Cs. A similar approach may be assumed to take into account the absorbed dose of radiation released by other fall-out isotopes like ^{90}Sr – ^{90}Y, for example.

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REFERENCES

Adamiec G. and Aitken M., 1998: Dose-rate conversation factors: update. *Ancient TL* 16(2):37-50.

Aitken M.J., 1998: *An Introduction to Optical Dating*. Oxford University Press, London: 41 pp.

Dunford C. L. and Kinsey R., 1998: *NuDat System for Access to Nuclear Data, IAEA-NDS-205 (BNL-NCS-65687)*. Information extracted from the NuDat data base, version of 17-Mar-2004, Using the PC version of the program NuDat. IAEA, Vienna, Austria.

Haak E. and Rydberg T., 1998: *Deposition, transfer and migration of ^{137}Cs and ^{90}Sr in Swedish agricultural environments and use of ^{137}Cs for erosion study*. *IAEA-TECDOC-1028,* IAEA, Vienna, Austria**:** 27-38.

Krbetschek M.R., Reiser V., Zõller R. and Heinicke J., 1994: Radioactive disequilibria in paleodosimetric dating of sediments. *Radiation Measurements* 23: 485-489.

Mellander H., 1986: Airborne gamma spectrometric measurements of the fall-out over Sweden after the nuclear reactor accident at Chernobyl, USSR. *Report No IAEA/NENF/NM-89-1*, IAEA, Vienna, Austria: 23.

Oczkowski H., Przegiętka K.R., Lankauf K.R. and Szmańda J.B., 2000: Gamma spektrometry in thermoluminescence dating. *Geochronometria* 18: 57-63.

Olley J.M., Murray A. and Roberts R.G., 1996: The effects of disequilibria in the uranium and thorium decay chains and buried dose rates in fluvial sediments. *Quaternary Science Reviews* 15: 751-760.

Poręba G., Bluszcz A. and Śnieszko Z., 2003: Concetration and vertical distribution of Cs^{137} in agricultural and undisturbed soils from Chechło and Czarnocin areas. *Geochronometria* 22: 67-72.

Ritchie J.C. and McHenry J.R, 1990: Application of Radioactive Fallout Cesium-137 for Measuring Soil Erosion and Sediment Accumulation Rates and Patterns. A Review. *Journal of Environmental Quality* 19: 215-233.