SEDIMENT ACCUMULATION RATE IN THE RED LAKE (ROMANIA) DETERMINED BY Pb-210 AND Cs-137 RADIOISOTOPES

R. BEGY, C. COSMA, Z. HORVATH

Faculty of Environmental Science, Babes-Bolyai University,
Fantanele Str, 30, RO-400294, Cluj-Napoca, Romania,
E-mail: rbegy@phys.ubbcluj.ro

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This work presents the first estimates of the Red lake's sedimentation rate. The determination of the sediment accumulation rate was estimated by using two methods of recent sediment dating: the Pb-210 and the Cs-137 method. Both methods are based on using gamma emission radionuclides. The Pb-210 and Cs-137 concentration in the sediment were measured with a gamma spectrometer with a Hp-Ge detector, GMX type. There has been found activities between 41 ± 7 – 135 ± 34 Bq/kg for Pb-210 and 3 ± 0.5 – 1054 ± 150 Bq/kg for Cs-137 respectively. Using the CIC methods for the calculation of sedimentation rate, we obtained a 1.23 ± 0.6 cm/year value, which is an average for this lake.

Key words: Pb-210, Cs-137, radioisotopes, dating methods, sedimentation rate.

INTRODUCTION

In 1837 in the metamorphico-mesozoic zone of the Oriental Carpathians a relatively rare phenomenon happened, the slope sliding of the Ghilcos Mountain blocked the Bicaz River and formed a natural barrage lake which received the Red name later. The barrage resisted the vertical and regressive erosion as well as the static water pressure.

The lakes formed by natural barrage of the valley have often a short life, because the formed barrage erodes or are quickly silted. The Red Lake is a special case, because the accumulated water's volume after the valley's barrage was significant and the geology and consistency of the barrage did not allow the quick erosion of this. We don't have to forget that the barrage, even natural, constituted damage for the natural dynamic of that side.

The sedimentation is a consequence of the reestablishing tendency of this equilibrium. In the case of the Red Lake there is the high sedimentation rate, which is highly influenced by men made activities (G. Pandi, 2004).

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In the process of the phenomenon's analyze we have to consider that because of the formation of the Red Lake, it is considered like an accumulation lake regarding the sedimentation.

One of the most important recent sedimentation dating method is the Pb-210 method, which is the natural radionuclide result from U-238 series. In many cases, the method turned out to be accurate, especially in stable environment with constant sedimentation rate, where the calculation model is well defined. The method also provides accurate results for non-uniform sedimentation (where the sedimentation rate is not constant), the difficulty is the finding of an appropriate sedimentation model. Two simple models, which are called the CSR and CIC models (Appleby & Oldfield, 1978, Robbins, 1978), are usually applied.

Disequilibrium between Pb-210 and its parent isotope in the series, Ra-226, arises through diffusion of the intermediate gaseous isotope Rn-222. A fraction of the Rn-222 atoms, produced by the decay of Ra-226 in soil, escapes into the atmosphere where they decay, through a series of short-lived radionuclide to Pb-210. This is removed from the atmosphere by precipitation or dry deposition, falling on the land surface or into lakes.

The lead which falls directly in the lake is transported by the water column and deposited to the bottom of the lake together with the sediment.

Another possibility for dating the lake sediments is the artificial radio nuclides. The two sources of this are the nuclear weapon test between 1953 and 1963, and the accident of Chernobyl atomic plant power station from 1986. The radio nuclides which were deposited and are present nowadays are: Sr-90, Cs-137, Pu-239-240-241.

In the northern hemisphere the deposition reached a significant level until 1954 and grew rapidly later.

In 1958 it was noticed a drop and it was followed by an increase because of the restarting of the testing.

After the agreement from 1963, the Cs-137 deposition diminished, but at the beginning of the 70's it increased again, caused by the states which did not sign the agreement.

The Chernobyl accident from 26th April 1986 injected a lot of artificial radionuclides in the atmosphere, this process continued during May, 1986. The principal radionuclides were I-131, Cs-137, Cs-134. The depositions were at thousands of km from the accident place, in the northern hemisphere, these depositions are estimated to 10^{17} Bq compared to $4{,}3\text{x}10^{17}$ Bq which was provided from nuclear testing (Cambray *et al.*, 1987).

The radionuclides distribution was non-uniform and greatly controlled by winds and rains. In some parts of northern Europe (Scandinavia, Great Britain) and central Europe the deposition in the first days of Cs-137 was higher than the total depositions provided by nuclear weapon.

MATERIALS AND METHODS

Fig. 1 shows the shape of the Red Lake and the places where the sediment samples were gathered from. The profile depth is also marked. The columns were divided in 4–6 layers of 8–10 cm each.

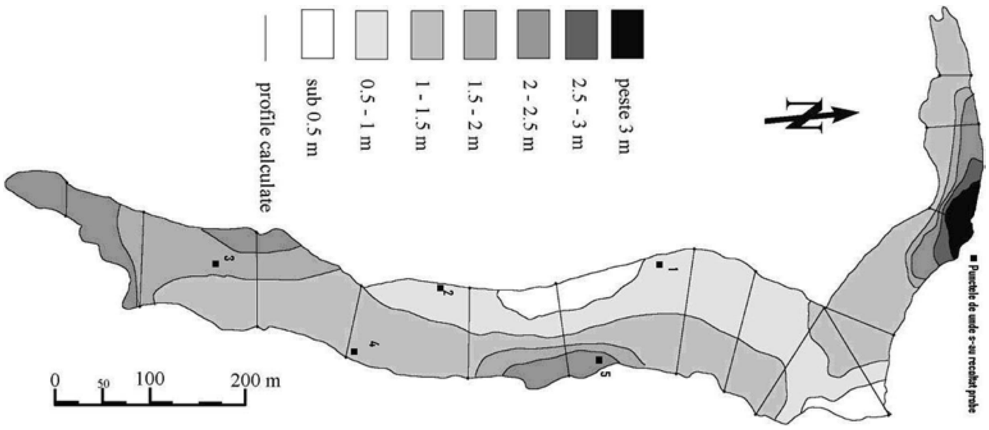

Fig. 1 – Some lake characteristics and sampling points.

Excess Pb-210 in sediment, over that in equilibrium with the *in situ* Ra-226 in sediments, decays in accordance with the radioactive decay law,

$$C_{\mathrm{Pb}} = C_{\mathrm{Pb}}(0)e^{-\lambda t} + C_{\mathrm{Ra}}(1 - e^{-\lambda t}) \tag{1}$$

(where λ is the disintegration constant of Pb-210). This equation can be used for sedimentation dating.

In the lakes where the erosion process from catchments area and residence time in water column are constant, the result is a constant sedimentation rate.

It can be supposed that each sediment layer has the same quantity of atmospheric Pb-210 activity (excess Pb-210) at the sedimentation moment.

Therefore the sediment from a certain depth m (dry deposition in g/cm^2) will be of the age:

$$t = \frac{m}{r} \tag{2}$$

where r is sedimentation rate (g/cm^{2}y) and "atmospheric" Pb-210 activity will vary with the depth after:

$$C_{(m)} = C(0)e^{-\lambda m/r} \tag{3}$$

where λ is the constant for Pb-210 (0,03114 year^{-1}) and

$$C(0) = P/r \tag{4}$$

is the atmospheric Pb-210 activity in the superior part of sediment core and *P* denotes the mean annual rate of supply of fallout Pb-210 to the sediments.

When it's represented in logarithmical scale the activity for Pb-210 *versus* depth gives a linear profile. The sedimentation rate average r can be determined from the graphic (J. Carrol *et al.*, 1999).

The measures were made using an ORTEC Digidart spectrometer with a HpGE GMX type detector, it has 1.92 keV resolution at 1.33 MeV line of Co-60 and the relative efficiency of 34.2%. The lead shielding of the detector with 40 cm diameter, with 8 cm wall, inside the lead shielding is a Cu layer of 3 mm. The shielding is very important especially in the case of environmental samples when the sample activity is smaller than the background activity. The used geometry was of "Sarpagan" type, a cylindrical box with adequate dimensions. The minimal time of acquisition was of 24 hours. The activity concentration of lead 210 is determined using 46.5 keV gamma line with relative intensity of 4% and the Cs-137 from 661 keV peak (Tanner *et al.*, 2000).

Ra-226 is determined using the radon peaks after a month of storage, when it reaches the equilibrium between Ra and Rn progeny. The detection limit is calculated using the formula below (J. Uyttenhove, 2003):

$$MDA(E_{46}) = \frac{A_m}{\varepsilon_{46} \cdot f \cdot t_m}\left(\sqrt{2bR_{46}B_{46} + A_m^2 / 4} + A_m / 2\right)$$

where A_m = 5 for an error of 20%

ε – detector efficiency;

f – relative intensity of peak at 46 keV;

t_m – measuring time (s);

b – shape factor = approx. 2 for symmetrical Gaussian peaks;

R – energy resolution of 46 keV peak (channels);

B – background (counts on channels).

For Pb-210, in our conditions, this value is $8 \pm 3(2\delta)$ Bq/kg.

RESULTS AND DISCUSSION

The values obtained for the radionuclide concentration in sediment layers can be seen in Figs. 2–6. In these figures we can notice "the problem of the first layer". This means that during the collection of the samples a big problem is to maintain the first layer in the initial form. In many cases there are losses of material from the first layer of the sediment.

From each sediment column was calculated an average sedimentation rate, at the sampling point. The sedimentation rates were calculated using CIC and CRS models. We applied both models and each gave the same results. With CRS model it can be seen that in the last 7–8 years the sedimentation process increased, in some cases the values reached double.

In CIC model, for Pb-210 and Cs-137 concentrations we used two mathematical fittings. The sedimentation rate was calculated from the fittings and from the mathematical model and it gave between 0.5-1.54 cm/year. Figs. 7–11 show the sediment dates versus depth, and the sedimentation rate of each sampling point.

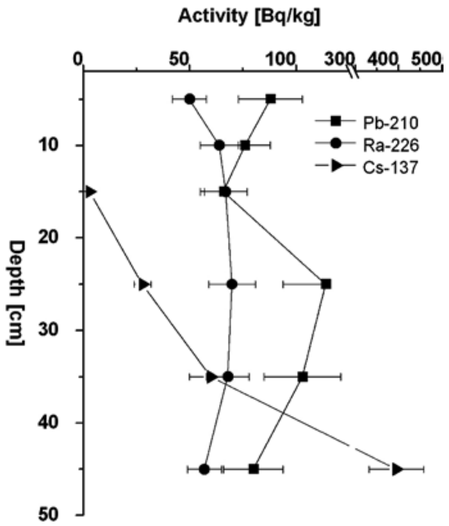

Fig. 2 – Radionuclide concentration in sediment-point 1.

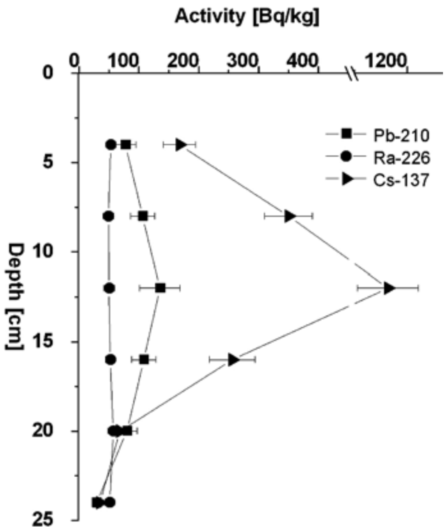

Fig. 3 – Radionuclide concentration in sediment-point 2.

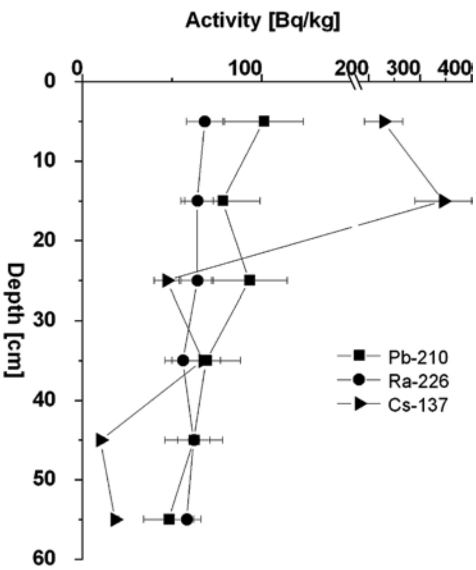

Fig. 4 – Radionuclide concentration in sediment-point 3.

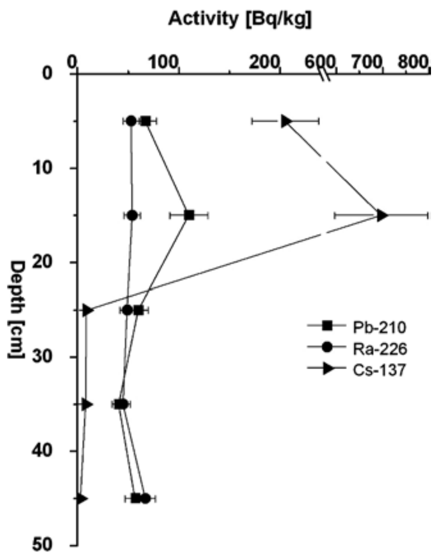

Fig. 5 – Radionuclide concentration in sediment-point 4.

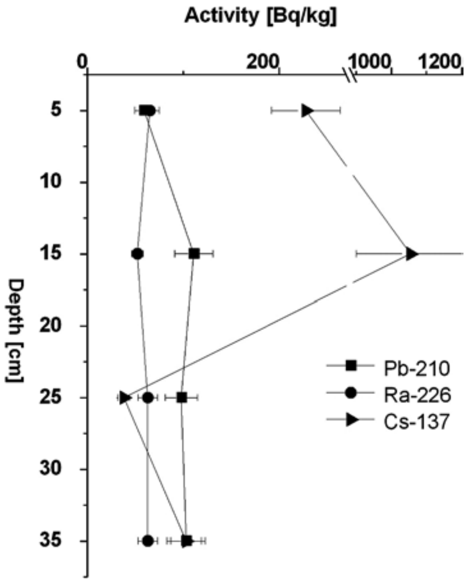

Fig. 6 – Radionuclide concentration in sediment from point 5.

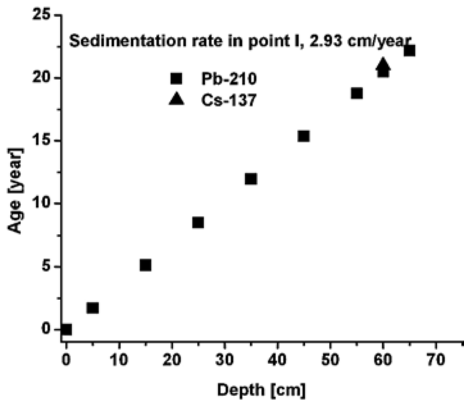

Fig. 7 – Sedimentation rate in point 1.

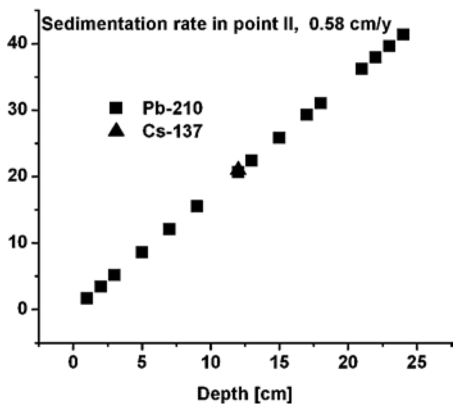

Fig. 8 – Sedimentation rate in point 2.

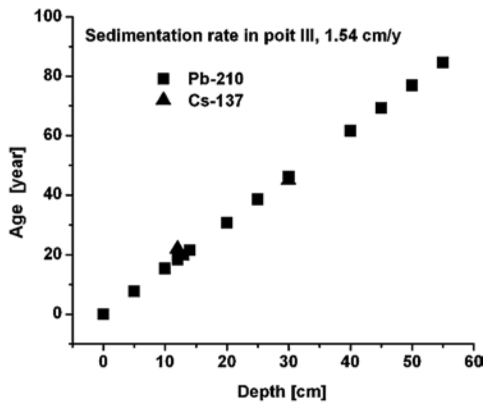

Fig. 9 – Sedimentation rate in point 3.

The results for the sediment layer age obtained from Pb-210 were compared to the ages resulted from Cs-137, and this gave very appropriate results as Pb-210. For the Red Lake sedimentation rate we can calculate an average value from all five cores 1.23 ± 0.6 cm/year. For the calculation of the total fill up of the lake were used the average values of the two furthest points (point 1 and 3). If we consider an average depth of the lake of 5.41 m, the total fill up will be in approximately 243 ± 70 year (G. Pandi, 2004). Determinations by other methods predict 257 years (G. Pandi, Cs Horvath). If the accentuated

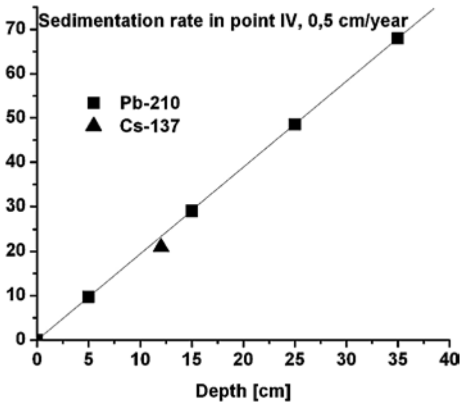

Fig. 10 – Sedimentation rate in point 4.

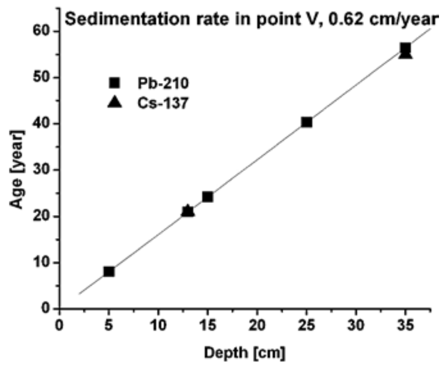

Fig. 11 – Sedimentation rate in point 5.

sedimentation is maintained to increase, the lake can disappear in less than 184 ± 30 years (CIC models for point 1).

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