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A SEDIMENTATION STUDY OF ST. ANA LAKE (ROMANIA) APPLYING THE ^{210}Pb AND ^{137}Cs DATING METHODS

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Abstract: The biggest problem with most lakes that have no contact with other water sources and are being charged by precipitation is the massive eutrophication. The aim of this work was to determine the sedimentation rate in order to evaluate the progress of eutrophication for St. Ana Lake (Ciomad Mountain near the Băile Tuşnad in Harghita County (Romania)). The concentration of ^{210}Pb was determined by means of high resolution gamma spectrometry as well as derived from ^{210}Po activity which was measured through alpha spectrometry; values obtained are in good agreement. For the excess ^{210}Pb activity values between 4.0±0.5 Bq/kg and 218±20 Bq/kg have been found. As an alternative method, the ^{137}Cs dating method was applied as well. Calculated mass sedimentation rates are in the range of 0.06±0.01 to 0.32±0.05 g/cm^{2}year with a mean of value of 0.15±0.02 g/cm^{2}year. Linear sedimentation rates yielded much higher sedimentation values (between 0.5±0.1 and 7.9±0.7 cm/year with a mean of 2.4±0.6 cm/year), due to the predominant organic matter composition and the long suspension time of the sediment. This is an indication for the process of eutrophication which will probably lead to the transformation of the lake into a peat bog.

Keywords: ^{210}Pb dating, alpha spectrometry, gamma spectrometry, ^{137}Cs, sedimentation rate.

1. INTRODUCTION

Information stored in the natural archives of lake sediments is used in a wide range of environmental studies, such as: determination of the history of changes in lake water quality associated with problems such as eutrophication; monitoring atmospheric pollution by heavy metals, radioactive emission from nuclear installations and other contaminants and the assessment of changing erosion rates in catchments arising from disturbances such as afforestation, deforestation or changes in agricultural procedures. One of the most important methods for dating recent sediments is the ^{210}Pb method (Arnaud *et al.*, 2006). The principle of the method is based on quantifying the disequilibrium in sediments between ^{210}Pb and its parent isotope in the series, ^{226}Ra. The disequilibrium arises through diffusion of the intermediate gaseous isotope ^{222}Rn. A fraction of the ^{222}Rn atoms, produced by the decay of ^{226}Ra in soil, escapes into the atmosphere where they decay to ^{210}Pb through a series of short-lived radionuclides. This is removed from the atmosphere by precipitation or dry deposition, falling on the land surface or into lakes (Appleby, 2001). The lead which falls directly into the lake is transported by the water column and deposited at the bottom of the lake. This is called excess ^{210}Pb, and it is added to the supported ^{210}Pb continuously formed in the sediment from the decay of uranium series.

This method can provide dates ranging from 0 to 150 years, the maximum and minimum values respectively, depending on the atmospheric flux of ^{210}Pb and other

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geological parameters. As ^{210}Pb emits a gamma line of weak intensity (4%) and low energy (46.5 keV), the estimation of its concentration by means of gamma spectrometry is not trivial, in many applications ^{210}Po being measured by means of alpha spectrometry instead. However, in this case the minimum obtainable age is 2 years, the time span necessary for ^{210}Po and ^{210}Pb to reach secular equilibrium (Sikorski and Bluszcz, 2008). The method can be successfully applied in stable environments with a constant sedimentation rate, where the calculation model is well defined (Schmidt *et al*., 2007; Begy *et al*., 2009; Tylmann, 2004). However, the method can also provide accurate results in the case of non-uniform sedimentation rates, though in this case, the difficulty of finding appropriate sedimentation models exists (Sikorski and Goslar, 2003). Usually, two simple models, which are called the CRS (constant rate of supply) (Appleby and Oldfield, 1978) and CIC (constant initial concentration) (Robbins and Edgington, 1975) are applied.

Another possibility for dating the lake sediments is by using the artificial radionuclides. The fission product ^{137}Cs is present in all parts of the Romanian environment, brought by atmospherically currents. The most important sources are the fallout from atmospheric nuclear weapons tests and fallout from the Chernobyl accident. Thus the presence of ^{137}Cs is a time marker that can be correlated to the results obtained by applying the ^{210}Pb dating method (see e.g. Carroll *et al*. 1999a, 1999b, Saravana Kumar *et al*. 1999, Robbins and Edgington, 1975, McDonald and Urban 2007)

2. STUDY SITE

St. Ana Lake is localized Ciomad Mountains (central Romania), near Băile Tuşnad in Harghita County (**Fig. 1**). It is a volcanic lake situated at 950 m altitude. The lake has an approximately circular shape, being 620 m long, 460 m width and having a maximum depth of 7.1 m. The lake has no direct contact with any water source and is being charged by precipitation. The water passes away by underground.

Few studies exist on the St. Ana Lake. Some water dynamics and sedimentation studies are being carried out at the Faculty of Geography of Babeş-Bolyai University of Cluj, however they have not been published. The mean annual rainfall is between 600 and 700 mm/year, the contribution of slopes is approximately 1000-1500 mm/year and the evaporation is about 500 mm/y (Slăvoacă and Avramescu, 1956; Pandi and Magyari-Saska, 2007)**.** Based on these data a rough calculation would result in the conclusion that the water level should increase annually, however, in 1867 the lake depth was 12 m, in 1907 8.5 m, and in 2006 6.5 m (Orban, 1869; Pisota and Nastase 1957, Pandi and Magyari-Saska, 2007). The water level decreases due to underground discharge and sedimentation.

The samples were collected from 6 different points of the lake **(Fig. 2)** using a gravity flag corer. The sampling points have been chosen in a way to allow identifying the regions of the lake where increased sedimentation rates exist in order to infer the source and direction of the incoming sediment. Coordinates and characteristics of sampling points are presented in **Table 1**.

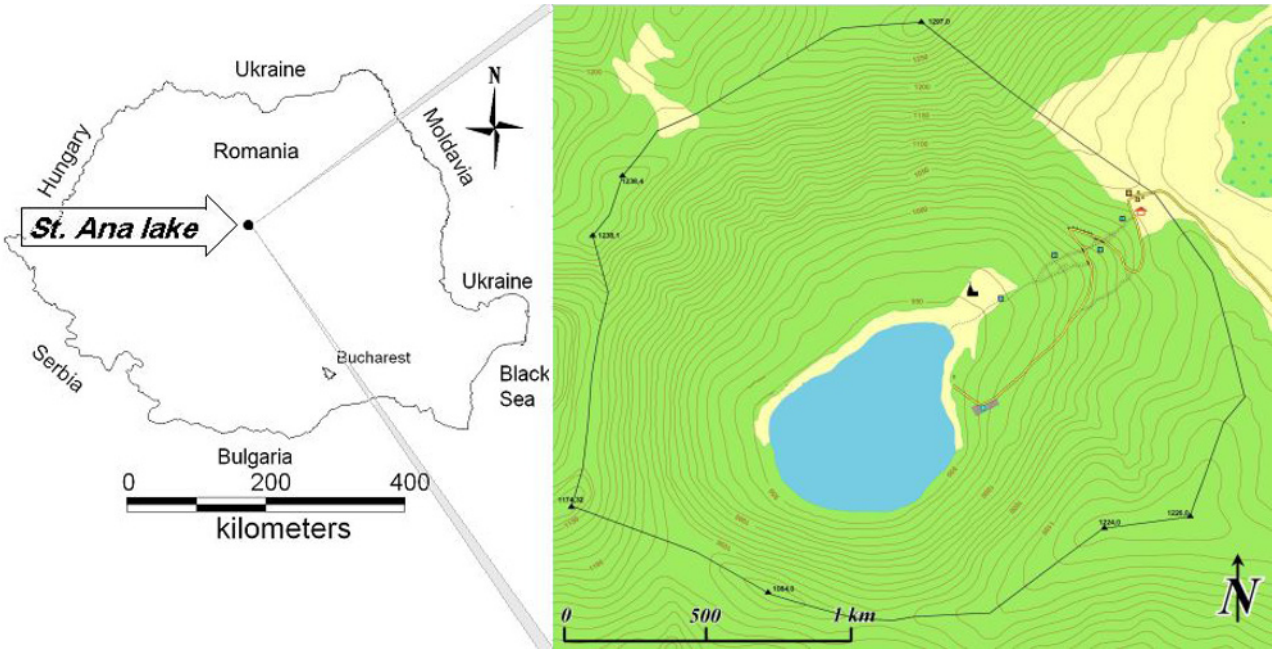

Fig. 1. *Map of Romania showing the location of the study site (St. Ana Lake)*

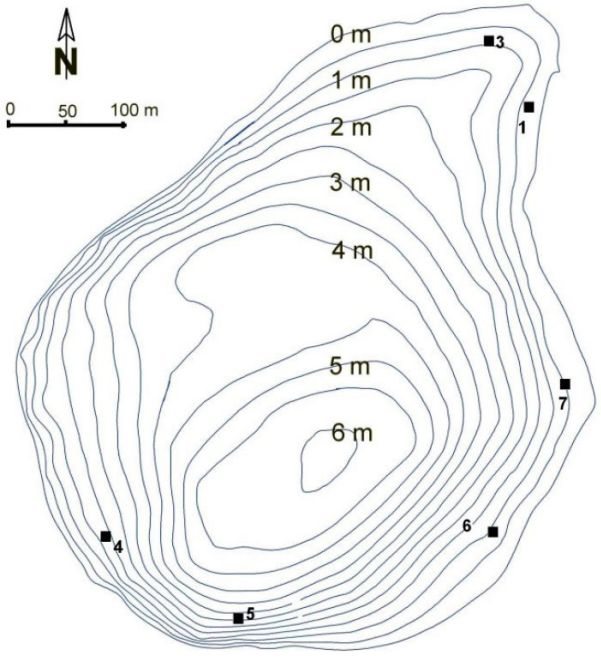

Fig. 2. *Bathymetric map (Pandi and Magyari-Saska, 2007) with sampling points indicated).*

3. MATERIAL AND METHODS

The sediment column was sliced in sub-samples of 3 to 9 cm thickness. Sub-samples (2-5 g) of the sliced core sections were used for measuring physical characteristics of the sediment sample, including bulk density, water content and porosity (Saravana Kumar, *et al*. 1999). The remains of the sliced core section were dried in oven at a temperature of 70°C and stored in the laboratory for radionuclide measurements (^{210}Po, ^{210}Pb, ^{226}Ra and ^{137}Cs) by means of gamma and alpha spectrometry.

Alpha spectrometry

Samples for alpha spectrometry were prepared by adding a combination of hydrofluoric, hydrochloric and nitric acid, and ^{209}Po as a yield tracer (30 mBq /sample) to 0.5 g of each sample and subjecting them to microwave digestion. For the removal of the acids and for a more efficient dissolution, the samples have been treated with HCl acid through repeated evaporation at 150°C. The small amounts of organic material remaining were removed by adding small amounts of hydroxyl peroxide. Finally, the samples have been rinsed with distilled water. ^{210}Po sources were obtained by spontaneous deposition from the solution on stainless steel disks, at a temperature of 82°C under continuous mixing for a period of three hours (Matthews *et al*., 2007).

Table 1. *Information regarding sampling points.*

Name	Longitude N	Latitude E	Sediment column high (cm)	Water depth (cm)
A.1	46.128316	25.889981	45	180
A.3	46.128491	25.889395	50	165
A.4	46.124487	25.884546	46	220
A.5	46.124061	25.885928	45	200
A.6	46.124419	25.888724	46	250
A.7	46.125548	25.889905	48	180

The measurements have been carried out using an ORTEC Soloist alpha spectrometry system with a PIPS detector (1200 mm^2 diameter) characterized by good resolution (19%), compact size, low background, excellent stability and low sensitivity to gamma radiation. The alpha radiation energy of ^{210}Po used was the 5304.5 keV line with a probability of emission of 99%. The activity of ^{210}Po was determined relatively to ^{209}Po, for a counting time of at least 90 ks.

Gamma spectrometry

For gamma spectrometric measurements samples have been pulverized and stored for 3 weeks in order for radioactive equilibrium to be reached in uranium series.

High resolution gamma ray spectrometry analysis was carried out using an ORTEC Digidart spectrometer with a GMX type HPGe detector having the following characteristics: active volume of 181 cm^3, 0.878 keV FWHM at 5.9KeV, 1.92 keV FWHM and 34.2% relative efficiency at 1332.5 keV. The detector is mounted in a lead castle and has a 0.5 mm beryllium window which makes it suited for measuring several U and Th decay products, down to ^{210}Pb with gamma ray energy of 46.5 keV. All measurements have been carried out in a cylindrical geometry, 1.7 cm high and 8 cm in diameter, the dimensions being chosen in order for self-attenuation to be reduced and a satisfactory counting statistics to be achieved in the same time. The minimum acquisition time was 24 hours. The measurement system was efficiency calibrated using IAEA standards (RGU-1, RGTh-1, and IAEA 375, respectively IAEA 312 and IAEA 327 radionuclides in soils). ^{226}Ra concentration was derived using the ^{214}Pb (^{222}Rn progeny) gamma line at 351.92 keV (relative intensity of 37.2%). The activity of ^{210}Pb was determined using the 46.5 keV gamma line (relative intensity of 4%) while ^{137}Cs concentration was calculated taking into consideration the 661.7 keV emission (relative intensity of 89%).

4. RESULTS AND DISCUSSION

Specific activities of ^{210}Pb, ^{210}Po, ^{226}Ra and ^{137}Cs from the sediment samples determined by gamma spectrometry and by alpha spectroscopy are shown in **Figs. 3** and **4**.

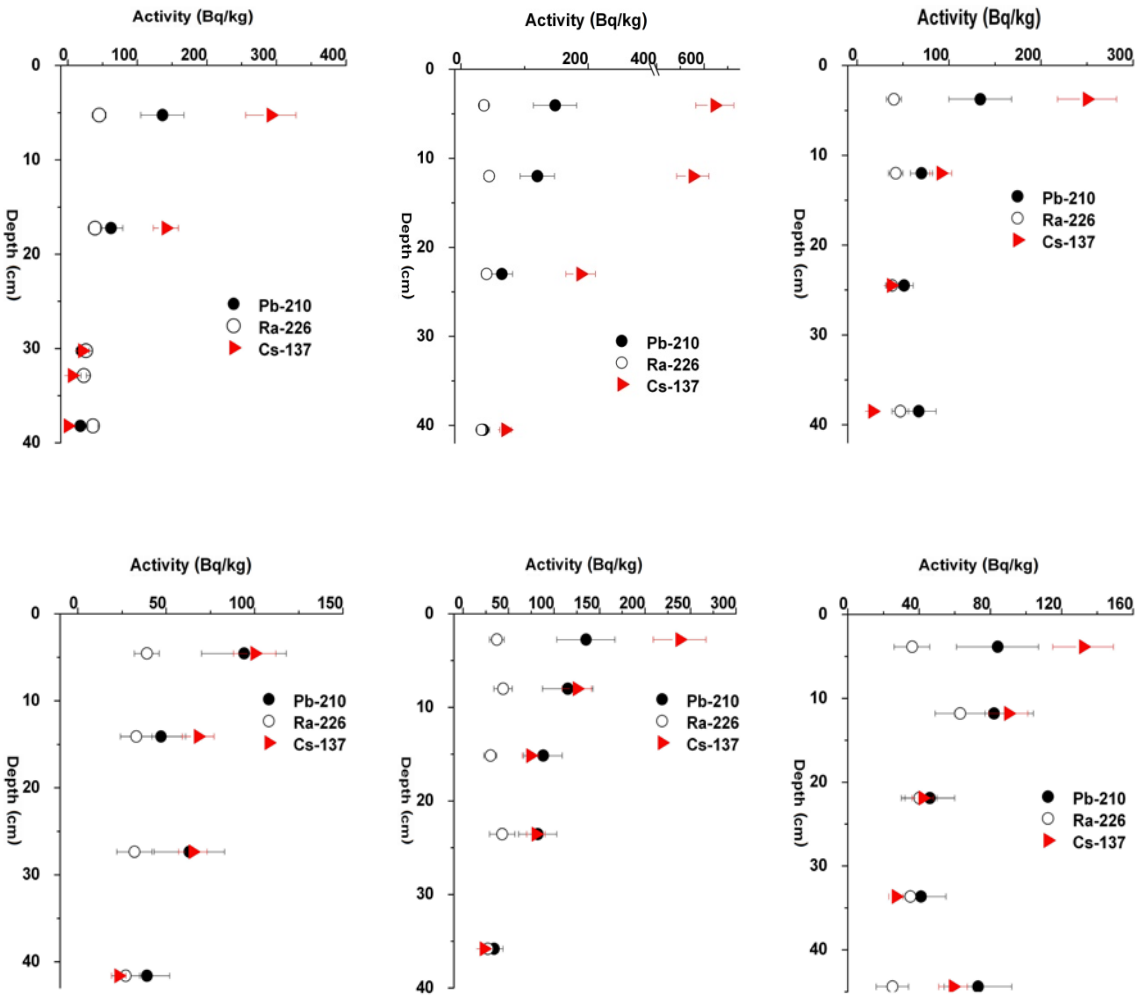

Fig. 3. *Distribution ^{210}Pb, ^{226}Ra and ^{137}Cs radionuclide specific activities determined by gamma spectrometry in columns A.1, A.3, A.4, A.5, A.6, and A.7.*

The advantages of applying alpha spectroscopy are its low detection limit and high accuracy, as well as the need for small quantities of material. The investigated samples contained large quantities of water and organic matter as indicated by **Fig. 5**. The porosity of the sediment from St. Ana Lake is high. After drying the samples the dry mass was low, and in the case of gamma measurements two sediment layers had to be used.

The CRS model was applied to calculate the age of each sediment layer. Calculations have been carried out using the ^{210}Po excess (alpha spectrometry) and ^{210}Pb activity concentration (gamma spectrometry). Changes in sedimentation rates can be observed (**Fig. 6**). Based on the ages, linear sedimentation (**Fig. 7**) and mass sedimentation rates have been calculated (**Fig. 8**). Average sedimentation rate for each sediment column is presented in **Table 2**.

Calculated mass sedimentation rates are in the range of 0.06±0.01 to 0.32±0.05 g/cm^{2}year with a mean of value of 0.15±0.02 g/cm^{2}year. Linear sedimentation rates yielded high sedimentation values (in between 0.5±0.1 and 7.9±0.7 cm/year with a mean of 2.4±0.6 cm/year), due to the predominant organic matter composition and the long suspension time of the sediment. This is an indication for the process of eutrophication which will probably lead to the transformation of the lake into a peat bog in approximately 300 years.

In previous studies (Pandi, unpublished data), the north part of the lake is described by increased eutrophication and sedimentation. This data is based on the comparison of periodically made bathymetric maps. Modification in water depth means new deposited sediment. For the northern part of the lake data obtained in our study show increased sedimentation rate at the point

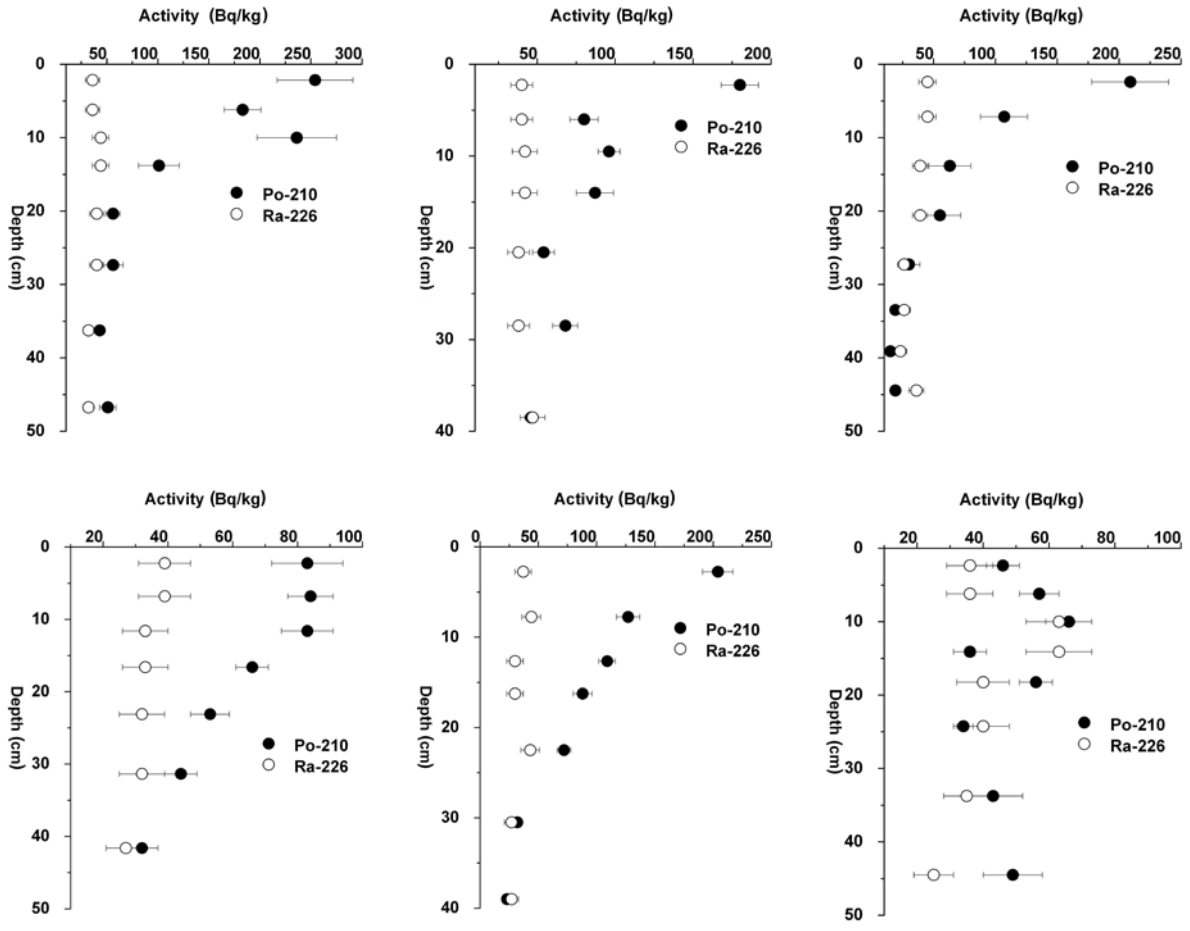

Fig. 4. *Distribution of ^{210}Po activity determined by alpha spectrometry and ^{226}Ra (measured by gamma spectrometry) in sediment columns A.1, A.3, A.4, A.5, A.6 and A.7.*

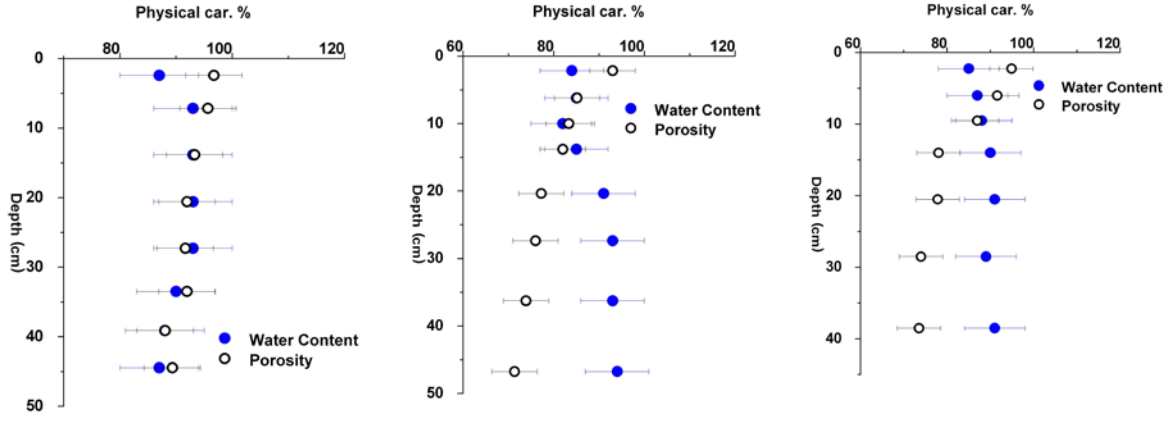

Fig. 5. *Physical characteristics of the sediment column in A.1, A.3, and A.4 sampling points.*

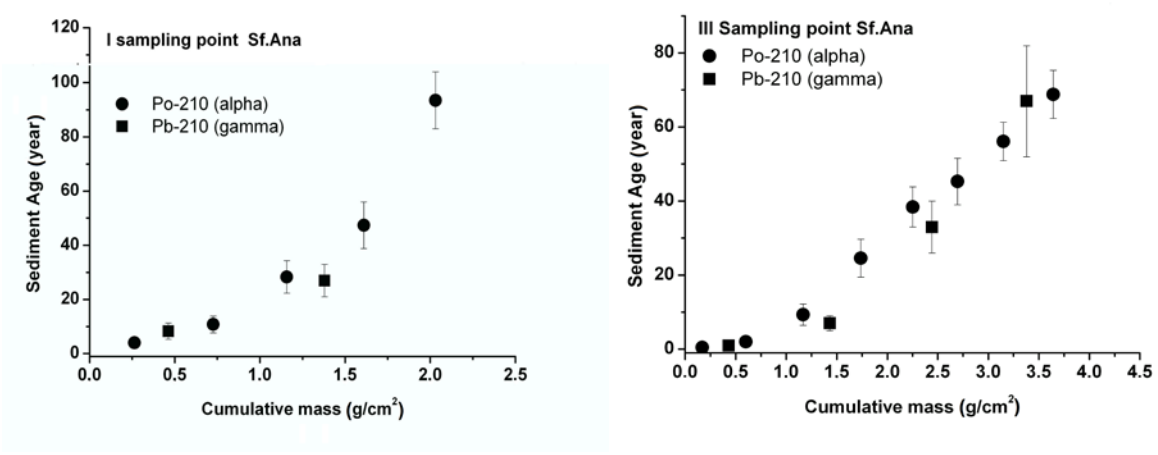

Fig. 6. *Comparison of sedimentation ages obtained from ^{210}Po and ^{210}Pb for sediment layers analyzed collected from sampling points A.1 and A.3.*

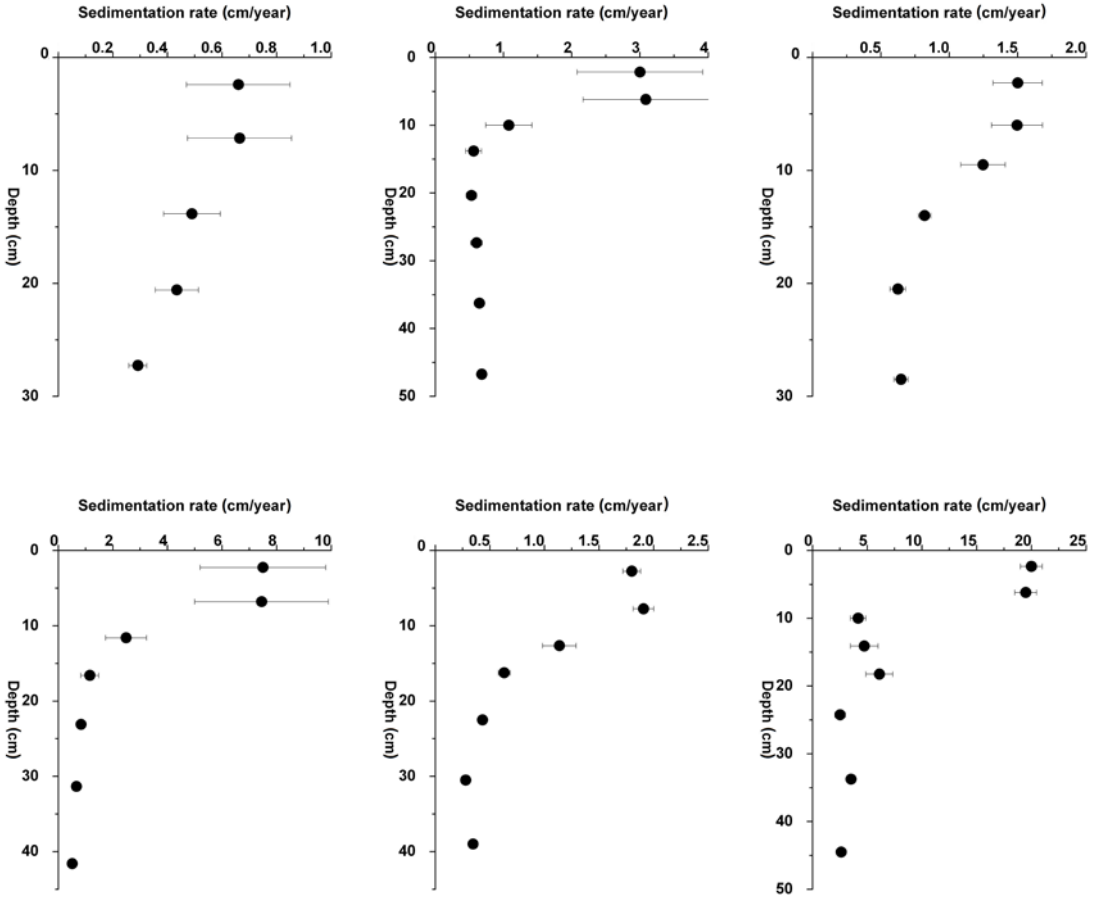

Fig. 7. *Linear sedimentation rates for sediment columns A.1, A.3, A.4, A.5, A.6.*

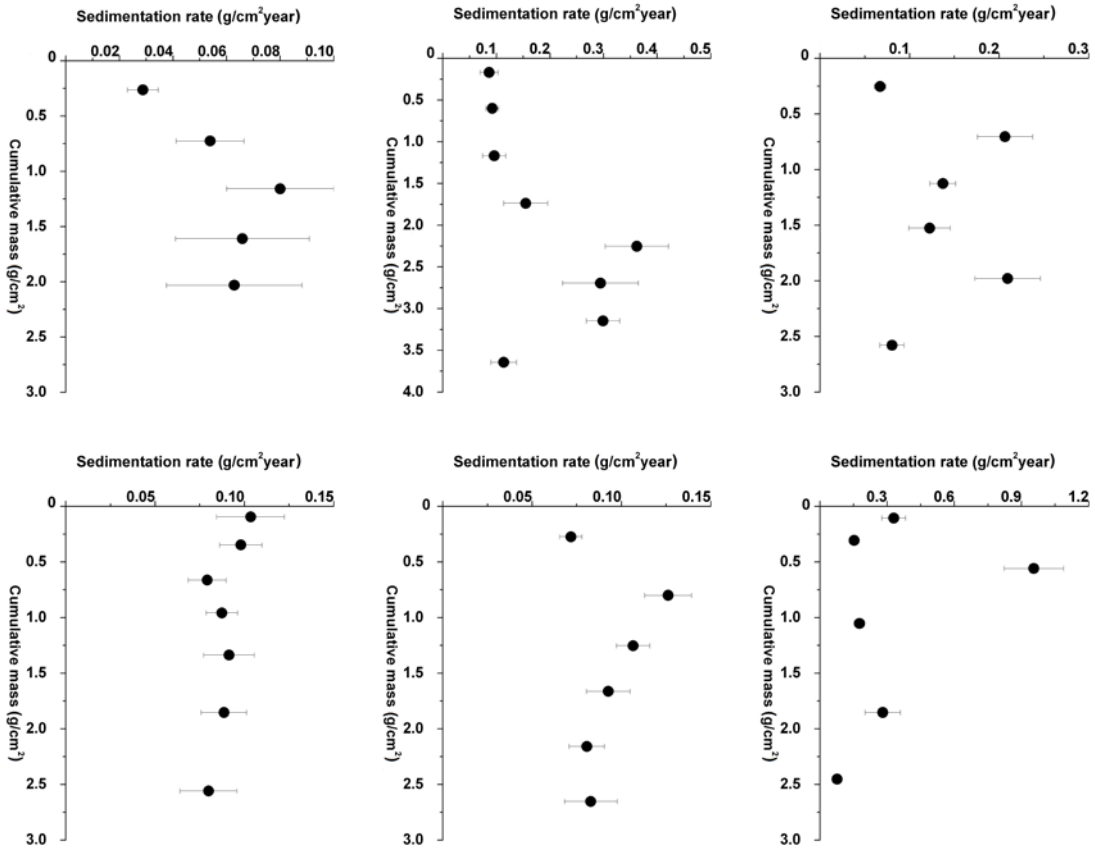

Fig. 8. *Mass sedimentation rate for A.1, A.3, A.4, A.5, A.6, and A.7 sediment columns.*

Table 2. *Sedimentation rates for St. Ana Lake. Quoted errors represent 1 sigma.*

Name	Alfa determination		Gamma determinations	
	Mass sedimentation (g/cm²year)	Linear sedimentation (cm/year)	Mass sedimentation (g/cm²year)	Linear sedimentation (cm/year)
A.1	0.06±0.01	0.5±0.1	0.03±0.01	0.5±0.1
A.3	0.19±0.03	1.3±0.3	0.09±0.04	1.1±0.2
A.4	0.14±0.02	1.1±0.1	0.12±0.05	1.1±0.2
A.5	0.09±0.01	2.9±0.9	0.11±0.04	2.6±0.5
A.6	0.09±0.01	1.0±0.1	0.09±0.04	1.5±0.3
A.7	0.32±0.05	7.9±0.7	0.40±0.14	5.2±1.0
Mean	*0.15±0.02*	*2.4±0.6*	*0.13±0.05*	*2.0±0.3*

A.3 (**Fig. 9**); however, at point *A.1*, situated in the north-eastern part, we have obtained a lower sedimentation rate and evidence of massive eutrophication.

In the southern part of the lake, our dating method gives apparently contradictory results compared to those found in previous studies. These studies reported a very low sedimentation rate, justified by the presence of the forest near the southern bank. For this part of the lake, based on absolute dating methods, relatively low mass sedimentation rates (*A.4, A.5, A.6)* have been obtained; however the values for linear sedimentation rates are high. This can be explained taking into consideration the water dynamics. In this site, the lake's water is not perturbed by any anthropic activities and the sediment can remain for a long time in suspension, and by sediment focusing process that results in the sliding of the older sediment towards the centre of the lake. Based on nuclear dating methods, low sedimentation rates cannot be inferred, and the conclusions of bathymetric studies are not sustained. For point *A.7* it can only be concluded that the sediment layer is mixed as a result of the massive eutrophication and the higher sedimentation rate which was observed.

5. CONCLUSION

A sedimentation study of St. Ana Lake in Romania has been carried out by ^{210}Pb dating method. The concentration of ^{210}Pb was determined by means of high resolution gamma spectrometry as well as derived from ^{210}Po activity which was measured through alpha spectrometry. The use of gamma spectrometric methods for estimating ^{210}Pb specific activity is not trivial due to the low energy

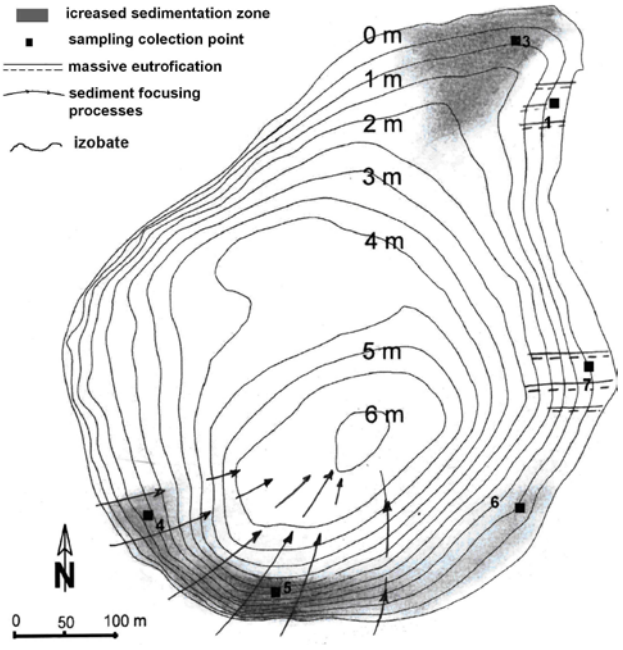

Fig. 9. *Map showing sedimentation processes in St. Ana Lake.*

emission of the most intense gamma line of this nuclide (46.5 keV, respectively 4%), which leads to the necessity of applying self-attenuation corrections and using a relatively large amount of sample. The obtained sedimentation rates using the two investigated methods are in good agreement. It was thus concluded that gamma spectrometry can be used with success for Pb-210 dating, though alpha spectrometry is a better method because of its higher precision and the need for less material thus leading to an improvement of the resolution in the sedimentation rate variation within a sediment column. Our findings also include the observation the linear sedimentation rates for St. Ana lake are high (up to ~7.9±0.7 cm/year) indicating that a process of eutrophication is taking place. Moreover, for the Southern part of the lake, sedimentation rates obtained by applying Pb-210 dating methods lead us to conclude that the information obtained through the comparison of periodically made bathymetric maps is not correct, meaning that sedimentation rates in this part of the lake are not necessarily low. Mass sedimentation rates obtained are indeed reasonably low, but the linear sedimentation rates are quite high. This was explained taking into consideration the water dynamics, as in this part of the lake the sediment can remain a long time in suspension, and by sediment focusing processes.

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