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Analysis of elements in lake sediment samples by PIXE spectrometry

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This work aims to determine the concentrations of several elements (e.g. Pb, Ni, Zn, Mn, Cr, and Fe) from lake sediments, in order to characterize their origin and evolution. Particle Induced X-ray Emission (PIXE) technique using the 3 MV Tandetron™ particle accelerator from National Institute for R&D in Physics and Nuclear Engineering "Horia Hulubei" (IFIN-HH), Magurele-Bucharest, Romania, was applied. Sediment cores from different salt lakes from Romania (i.e. Amara Lake, Caineni Lake, and Movila Miresii Lake) were collected, in August 2015. The content of Pb, Cr, Mn, Fe, and Ni from sediment samples show similarities with other data presented in literature and international regulation. The Zn was the only element with a higher content in all samples (e.g. maximum 401.7–517.3 mg/kg d.w.).

1. Introduction

The sediments from salt lakes provide information on the various processes such as sedimentation, water dynamics, sediment contaminant interaction, sediment-organism interaction and historical indicators as well as about origin of the lakes [1–3]. Concerning the chemical composition and origin of the salt lakes from Romanian Plain, there are several hypotheses, such as: remnants of the Pliocene Lake; water supply from underground source arising from Romanian Carpathian salty area; water supply from underground through capillarity involve the salts that are in the clays situated near of the soil surface [1,2]. Salt lakes from Romanian Plain are part of different genetic categories, such as, coasts (i.e. Caineni Lake, salinity max. 45 g/L), loess saucer (i.e. Movila Miresii Lake, salinity max. 76 g/L) and oxbow (i.e. Amara Lake, salinity max. 90 g/L) and shows anoxic hypolimnetic horizons and a density stratification, and the depth of these horizons being dependent by the intake of freshwater which coming from rainfall or temporary tributaries. In this work was chosen these three lakes (i.e. Caineni, Movila Miresii and Amara) because these are situated in endorheic region in which the underground supply intake and evaporation are the main component in the hydrological balance and could be exploited in therapeutical treatment due to sapropelic mud from bottom of lakes. These salt lakes accurately reflect the climatic conditions with a strong aridity character of the Romanian Plain. The sediments are mainly chemogenic and biogenic origin (input inflow are minimum) and any kind of change due to pollution may lead to unbalanced ecological lake system. The first stage in order to investigate the origins of these lakes was to determinate the elemental content of the collected sediments [4,5]. The influence of vertical distribution of different elements can be assimilated by the atmospheric and local domestic pollution (e.g. Amara Lake is the most used lake in therapeutical treatment and certainly, the shore line is affected by the anthropic activity).

2. Sampling and sample preparation

Sediments cores were collected from different Romanian Plain salt lakes by means of a floating platform and vibrocorer system. The submerged sediments have been extracted using one meter long piston with an acrylic core tube to avoid accidental contamination from external sources. The samples were carried in vertical position, in thermally insulated containers at 4 °C and stored in a freezer at −18 °C for 48 h. After visual checking, the sediment core was sliced in 1 cm pieces and, afterward, selected one representative sample (from 10 to 10 cm) in order to investigate the major and minor elements. Then, 30 significant samples (10 samples

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Table 1
Correlation between sample code, depth and salt lakes annotation in accordance with diagrams from Fig. 1.

Sample code	Depth [cm]	Salt lakes annotation		
		Caineni Lake	Movila Miresii Lake	Amara Lake
1	0–1	C1	M1	A1
2	10–11	C2	M2	A2
3	20–21	C3	M3	A3
4	30–31	C4	M4	A4
5	40–41	C5	M5	A5
6	50–51	C6	M6	A6
7	60–61	C7	M7	A7
8	70–71	C8	M8	A8
9	80–81	C9	M9	A9
10	90–91	C10	M10	A10

Table 2
Minimum, Mean and Maximum values of the metals concentrations [mg/kg d.w.].

Element	Min.	Mean	Max.	RSD [%]	Salt Lake
Pb	10.92	12.49	13.82	11.2	Amara
	17.57	17.92	18.33		Movila Miresii
	10.62	11.56	12.80		Caineni
Cr	10.17	12.39	15.42	12.2	Amara
	15.23	15.74	16.21		Movila Miresii
	10.97	11.71	12.71		Caineni
Mn	68.3	76.24	84.05	7.8	Amara
	82.38	89.88	94.23		Movila Miresii
	56.45	57.66	59.45		Caineni
Fe	32,011	34,545	39,093	6.4	Amara
	20,527	21,284	21,767		Movila Miresii
	22,773	23,290	23,889		Caineni
Ni	13.99	14.76	17.22	8.2	Amara
	18.79	19.62	20.87		Movila Miresii
	16.21	17.03	17.98		Caineni
Zn	315.7	398.5	517.3	6.7	Amara
	241.5	344.1	422.1		Movila Miresii
	221.4	322.8	401.7		Caineni

from each lake, Table 1) were dried at 60 °C for 48 h, powdered and sieved, in order to obtain 15 mm pellets for PIXE investigations.

3. The experimental set-up

The PIXE experiments were performed using a proton beam provided by the 3 MV Tandetron™ particle accelerator from the National Institute for R&D in Physics and Nuclear Engineering "Horia Hulubei" (IFIN-HH), Magurele-Bucharest. The pressure in the beam transport tubes of accelerator and in reaction chamber was 10^{-6} mbar during irradiation. The target current was between 3 and 5 nA in order to have a low count rate that implies a negligible dead time correction. The characteristic X-ray spectra of samples were detected with a HPGe detector model IGLET-X-06135-S with main characteristics: active diameter 6 mm, active depth 6 mm, beryllium entrance window of 0.0127 mm and energy resolution (FWHM) of 160 eV at X Kα 5.9 keV line of Mn, resulting from the radioactive decay of ^{55}Fe, EC decay type. The X-ray spectra were processed off-line using the GUPIX and LEONE softwares.

4. Results and discussion

The mean values of Pb, Cr, Mn, Fe, Ni, and Zn concentrations obtained by PIXE measurements are presented in the Table 2. From this table can be observed that the iron concentration, as major element, does not overtake the values reported in scientific literature [6,7]. One of the main reasons of high Fe content (correlated with early visual investigations) may be explained by redox process. The minor elements content, including Pb, Cr, Mn, and Ni, also, does not exceeded the values recorded in different studies [6,7] and international regulation. The high content of Zn has been determined in all analyzed samples (Table 2) and can be compared with values obtained in other study [8].

Fig. 1 shows high value for minor elements (i.e. Pb, Cr, Mn, and Ni) in samples collected from Movila Miresii Lake, comparative with the content determined in sediments from Amara and Caineni Lakes. Also, the Fe and Zn content from Amara Lake samples exceed the values recorded on the other two studied lakes and could be explained by the permanent anthropic pressure (tourism and domestic activities).

From Fig. 1(f) can be observed the fact that the Zn content suddenly decreases for all samples collected from 50 cm depth, comparative with all values obtained for other depth. This aspect can be explained by the capacity of Zn to migrate under different chemical states. Probably, in some climatic conditions, the mobile species of Zn migrate from sediments to surface lake water. The organic and mineral fraction present in lacustrine sediments of these three salt lakes is produced *in situ* because does not have extensive catchment area but, only small temporary tributaries and no outlet stream.

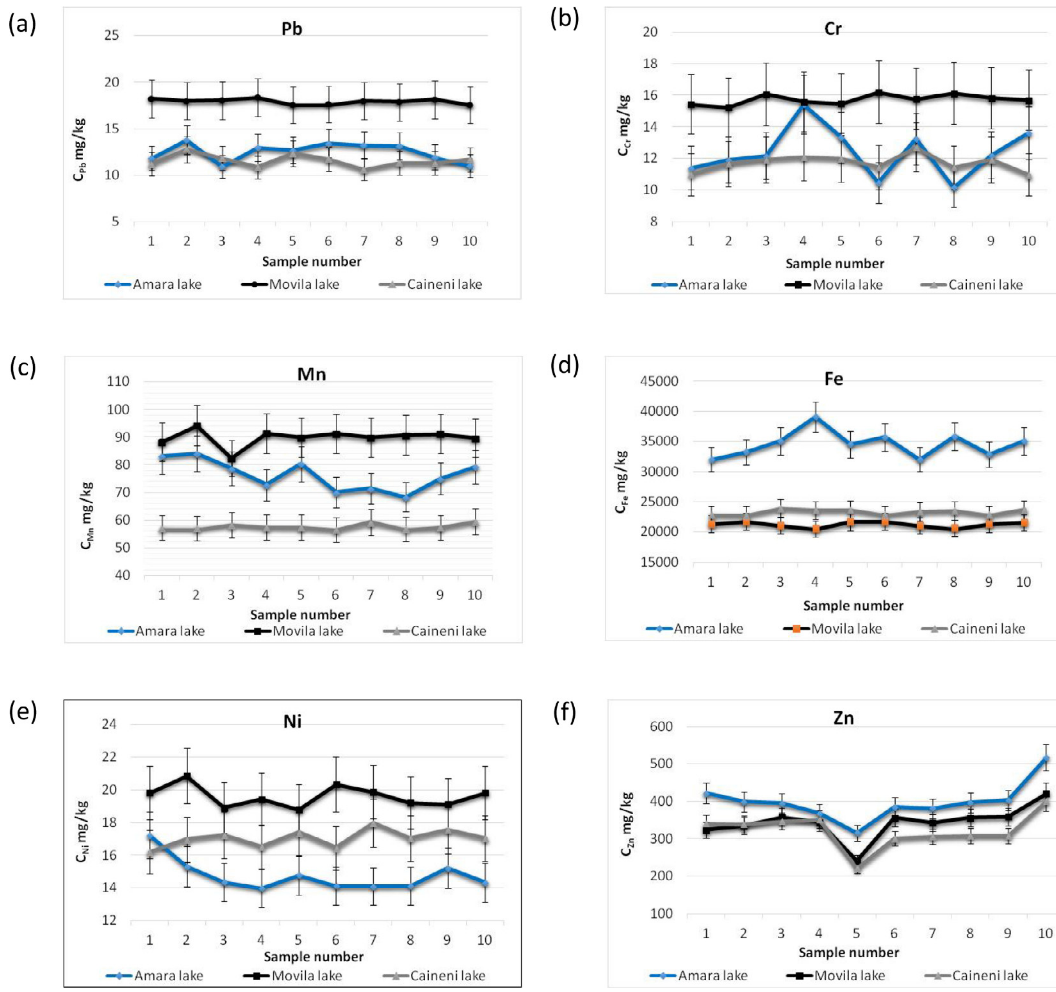

Fig. 1. Element content [mg/kg d.w.] for: Pb(a), Cr (b), Mn (c), Fe (d), Ni (e), Zn (f), in 10 representative samples from each studied salt lakes.

Metal distribution in sediment cores is influenced by the physical, chemical and biological/microbiological conditions of the lake; the layer absorption capacity is related to cationic species and organic sediment links, and also, by climatic condition changes in the catchment area [9].

5. Conclusions

The present work is a point of start for a future research, in order to establish the origin, current state and the evolution of the salt lakes from Romanian Plain. The elements content in the sediments collected from Caineni, Movila Miresii and Amara Lakes can provide important information about the environmental and climatic conditions with a strong aridity character of the Romanian Plain. The analysis of sediments can aid in reconstructing the history of changes, understanding human impact on the ecosystem, and suggesting strategies to maintain ecological balance.

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