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ON THE HEAVY ELEMENTS CONTENT OF SEDIMENTS AND ROCKS FROM TWO SEMICLOSED ECOSYSTEMS: PROGLACIAL LAKE BÂLEA (FÂGÂRAŞ MOUNTAINS) AND CRATER LAKE ST. ANA (HARGHITA MOUNTAINS)[1]

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The content of seven heavy elements (natural Sc and Cr, Co, As, Sb, Br and Se as possible pollutants) was determined by the Neutron Activation Analysis (NAA) in 29 samples of sediments and surrounding rocks collected from proglacial Lake Bâlea (Fâgâraş Mountains) and crater St. Ana Lake (Harghita Mountains) to estimate the degree of anthropogenic pollution, as well as the sediments provenance. Consequently, we have found that, within experimental uncertainties, the content of the considered elements in sediments was close to those of surrounding rocks, and, at the same time, comparable with the normal environmental content as defined by the Romanian Regulations, which enabled us to consider them as normal, nonpolluting constituents of lake sediments. A further R-mode, Principal Component Analysis (PCA), performed for each lake, showed that Sc, Cr, and Co, on the one hand, and As, Sb, Br and Sb, on the other, form two distinct clusters, regardless of the lake, while the Q-mode analysis pointed to a different lake geochemistry with respect to sediments as well as rocks.

Методом нейтронного активационного анализа (НАА) определено содержание семи тяжелых элементов (природного Sc и Cr, Co, As, Sb, Br, Se как потенциальных загрязняющих веществ) в 29 образцах из донных отложений и окружающих пород, собранных в окрестности ледникового озера Былеа (горный массив Фэгэраш) и кратерного озера Святая Анна (горный массив Харгита). Целями исследования были оценка уровня антропогенного загрязнения и определение источника происхождения донных отложений. Найдено, что в пределах экспериментальных ошибок содержание рассматриваемых элементов в отложениях близко к их содержанию в окружающих

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породах и, в то же время, сравнимо с нормальным составом окружающей среды согласно румынским нормативам. Это позволяет нам считать их незагрязняющими компонентами озерных донных отложений. Дальнейшая обработка данных (R-mode, компонентный анализ PCA) отдельно для каждого озера показала, что Sc, Cr, Co, с одной стороны, и As, Sb, Br, Sb, с другой, создают два различных кластера для обоих озер. В то же время факторный анализ Q-mode указывает на разные геохимические свойства озер по отношению к донным отложениям и породам.

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INTRODUCTION

Bâlea Lake (0.046 km^2, Fâgâraş Mountains) [1] and St. Ana Lake (0.195 km^2, Harghita Mountains) [2] differ by origin and location. Bâlea Lake is a typical high-altitude proglacial lake, while Lake St. Ana, located in the Ciumatu extinct volcano caldera, is the only volcanic lake in Romania (Fig. 1). Both lakes are characterized by the absence of any source of upstream industrial activity, making them good indicators for any environmental screening. On the other hand, both lakes, due to their picturesque setting, are visited during summer time by a great number of tourists, so a certain degree of anthropic influence is expected to exist.

Both lakes collect pluvial water from relative restricted areas (0.234 and 0.147 km^2, respectively), so that weathering of surrounding formations should significantly influence the sediments geochemistry. Accordingly, the sediments of Bâlea Lake are expected to reflect the mineralogical composition of the Suru Formation, mainly consisting of metamorphic rocks

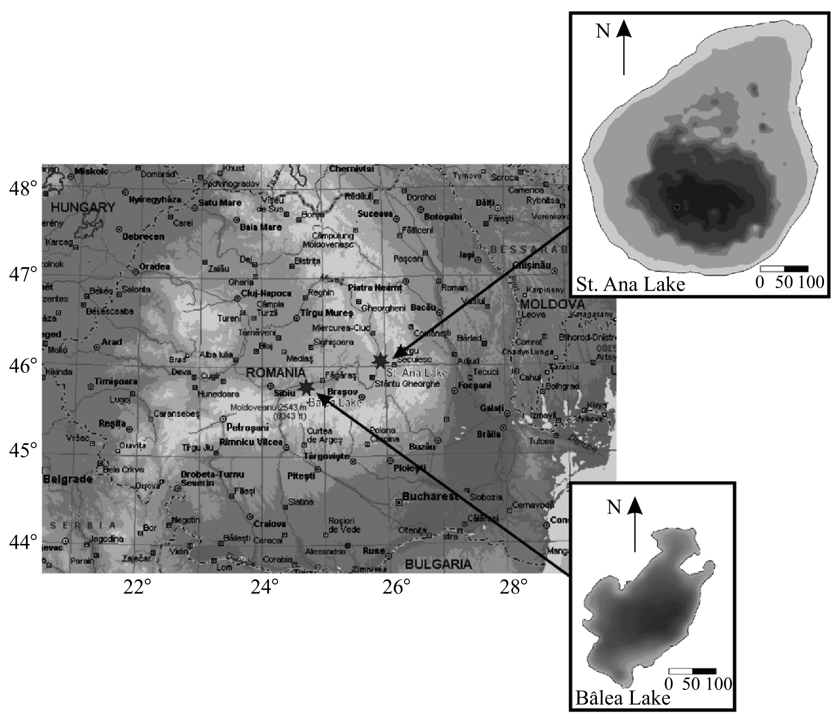

Fig. 1. The map of Romania illustrating the location of St. Ana and Bâlea lakes

such as amphibolitic schists, quartzo-felspatic gneisses and mylonites as well as limestones, all of them with a medium to high weathering potential [3], while the sediments of St. Ana Lake, which occupies the bottom of now extinct Ciumatu volcano, are composed of fragments of weathered andesite together with an appreciable amount of phytodetritus.

The Instrumental Neutron Activation Analysis (INAA), a highly sensitive analytical method which allows determining the content of about 45 elements at concentrations of up to μg^{-1} without any previous samples processing, represents one of the most suitable techniques in studying the heavy elements environmental pollution [4, 5].

For this reason, we have used INAA, in combination with the Principal Component Analysis (PCA), to investigate the content distribution of the seven heavy elements, i.e., naturally occurring Sc and possibly polluting Cr, Co, As, Se, Br and Sb in sediments and surrounding geological formations of the above-mentioned lakes. The results of this project are presented and discussed below.

1. INVESTIGATED AREAS

1.1. Lake Bâlea. The proglacial Lake Bâlea is located in the homonym depression, at an altitude of 2050 m, on the northern slope of the Fâgâraş Mountains. The entire Bâlea Depression is occupied by an association of paragneiss + amphibolites + crystalline limestones which form the Suru Formation belonging to the Fâgâraş Series [6] and involving Fâgâraş metamorphics more in alpine tectonic events as a nappe structure. Pana and Ricman [7] evidenced above this unit a mylonitic level whose preretromorphic content can be ascribed to an almandine amphibolites facies. Middle grade parageneses, rich in garnet, muscovite, biotite, are preserved too. The main lithological association is constituted of chlorite-sercitous schists, amphibolic schists and limestones representing an area affected by the retromorphism rebalanced under low grade conditions. This is why, Bâlea Lake sediments consist of two fractions: a coarser one deriving from the gnessic and amphibolic fraction and a finer, sandy one, originating from the mylonitic and limestone fraction.

With a maximum depth of 11.3 m [8] and a surface, as previously mentioned, of 0.046 km^2, Bâlea Lake is one of the greatest proglacial lakes of Romanian Carpathians. By taking into account that the ratio between the lake surface and its catchment area is equal to 0.0196 and that 92.3% of its water supply comes mainly from streams and springs, while about 98% of water flows directly through Bâlea spring [1], Bâlea Lake can be regarded as a minerotrophic one with an active water circulation, typical for high-altitude lakes.

Harshness of the alpine climate, a typical alpine tundra vegetation together with a fairly young age no greater than 10 ka, combined with a relatively diminished transport of mineral detritus, determined a very slow sediments accumulation whose thickness was between 20 and 80 cm [9, 10].

1.2. Lake St. Ana. The crater St. Ana Lake (Harghita Mountains), the only one of its kind in Romania, occupies the southern crater of the Ciumatu volcano, whose last eruption, as determined by ^{14}C, took place 28 000 BP [11], generating andesitic lava, now exploited in some quarries, not far away from Ciumatu Mountain. At present, Ciumatu Caldera consists mainly of friable pyroclastic material, covered with a dense mixed coniferous and deciduous forest (see Fig. 1).

Situated at a medium altitude of 950 m, St. Ana Lake has a total surface of 0.0195 km^2, a maximum depth of 6.4 m, a ratio between surface and catchment area of 0.150, and receives water only from precipitation, which confers a more ombrotrophic character. It is worth mentioning that due to the high friability of the caldera pyroclastic material, the St. Ana Lake sediments present an appreciable thickness, reaching in some places more than 4 m.

2. MATERIALS AND METHODS

2.1. Samples. During 2008 and 2009 campaigns, five cores with unconsolidated sediments from Bâlea Lake (three cores) and St. Ana Lake (two cores) as well as a number of rock samples were collected. Then the cores, kept all the time in vertical position, were X-rayed, and longitudinally split into two halves, one of them being cut into more segments and dehydrated at 105 °C. For INAA investigations, we have used no more than 1 g extracted from each segment.

Regarding Bâlea Lake rocks, we have selected six samples of different kinds of paragneisses (paragneisses, paragneisses associated with chloritic schists, quartzo-feldsparic paragneisses with garnets and paragneisses associated with amphibolitic schists) and one of amphibolites, all of them characteristic for the Suru Formation. In the case of St. Ana Lake, due to a relative uniformity of geological conditions, our selection includes only fragments of andesite, collected from different locations within the volcanic caldera.

Finally, for our study we have selected 29 specimens: 12 samples of sediments and three andesites belonging to St. Ana Lake location as well as six samples of sediments and eight of different rocks from Bâlea Lake (see table).

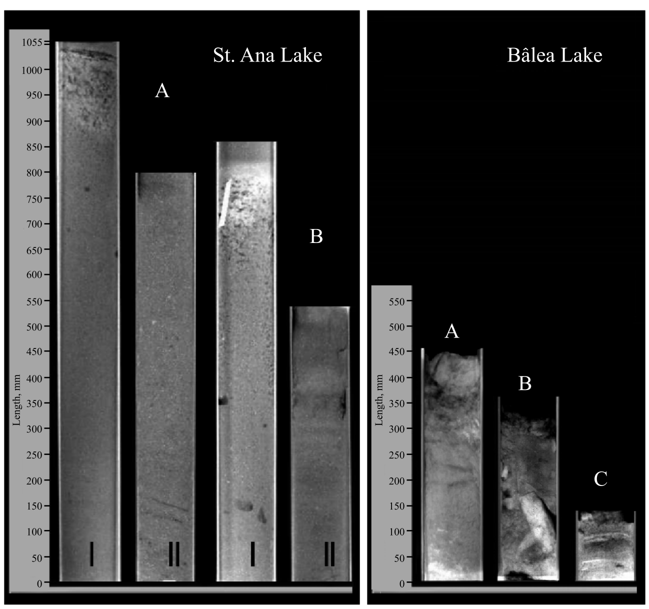

Fig. 2. Digital radiographs of investigated cores. Due to their length, St. Ana Lake cores (left) were sectioned into two fragments (I and II), while Bâlea Lake cores (right), significantly shorter, were investigated uncut

The average content (Aver) together with the corresponding standard deviations (St. Dev) (both in mg/kg) of investigated heavy elements in St. Ana Lake sediments, St. Ana Lake andesites, Bâlea Lake sediments as well as Bâlea Lake rocks. For comparison, the content of the same elements as defined by RR [17] together with UCC [14, 15] as well as N-MORB [16] contents are reproduced too. The number of analyzed samples is reproduced in parenthesis

	Sample	Sc	Cr	Co	As	Se	Br	Sb
Lake St. Ana	Sediments							
	Aver (12)	4.5	31	4.2	4.1	1.7	23.7	2.4
	St. Dev (12)	0.9	3	0.4	1.0	0.4	7.4	0.9
	Andesite							
	Aver (3)	4.5	29	3.6	2.4	1.2	28.3	1.3
	St. Dev (3)	1.2	10	1.0	0.7	0.3	16.4	0.0
Lake Bâlea	Sediments							
	Aver (6)	20.0	272	42.3	5.5	3.0	5.7	1.4
	St. Dev (6)	0.8	39	5.3	1.8	0.5	1.5	0.5
	Rocks							
	Paragneiss (3)	12.4	179	18.3	< 0.1	1.5	0.4	0.1
	Paragneiss + chloritic schists with garnets	31.5	693	69.0	4.3	2.5	0.2	0.1
	Amphibolite + amphibolitic schist	29.0	37	52.0	< 0.1	2.6	0.3	0.3
	Paragneiss + amphibolitic schist	52.0	480	60.0	< 0.1	< 0.5	0.6	0.7
	Paragneiss + chloritic schists	18.2	106	20.0	< 0.1	0.9	0.3	0.1
	Paragneiss + chloritic schists	19.1	114	16.0	< 0.1	< 0.5	0.8	0.1
	Aver (8)	24.9	291	39.3	4.3	2.0	0.4	0.2
	St. Dev (8)	16.0	274	24.5	—	0.8	0.2	0.2
Reference	RR normal	—	30	15	5	1	50	5
	RR alert	—	100	30	15	3	100	12.5
	UCC	11	35	10	1.5	0.05	2.1	0.2
	N-MORB	31.8	714.3	37.8	nd	nd	nd	nd

2.2. Digital Radiography. All digital radiographic images of cores were obtained by using a dedicated computer tomograph (CT) in the same experimental conditions, i.e., anodic potential of 110 kV, anodic current of 30 mA, scanning speed of 1 cm/s, at a vertical resolution of 0.5 mm and a horizontal resolution of 0.3 mm [12] (Fig. 2).

2.3. Activation Analysis. INAA was carried out at the WWR IRT research reactor of the Moscow Engineering Physics Institute. Samples weighing 100 to 200 mg, together with reference materials: IAEA-433, IAEA 140/TM, IAEA SL-1 and IAEA Soil-7, were irradiated with thermal neutrons in the vertical experimental channels at a fluence density of $9 \cdot 10^{12}$ $cm^{-2} \cdot s^{-1}$ for 15 to 20 h, and then kept between 5 and 10 days to allow the decay of short-lived radionuclides.

After that, both samples and reference materials were measured in three different phases, in accordance with the half-life periods of chosen elements.

All gamma spectra were recorded by using an ORTEC GEM 25185 HPGe detector with an energy resolution of 1.85 keV for the 1332 keV ^{60}Co line. It is worth mentioning that, by using IAEA SL-1 and Soil-7 samples, the differences between our values and certified values were between 1% for Rb and Ta and 14.7% for Nd, all of them in Soil-7. Further, for our study, we have selected the content of natural Sc and potential pollutants: Cr, Co, As, Sb, Br and Se in all samples.

2.4. Statistical Assessments. To classify and interpret our data, we have used the Principal Component Analysis (PCA) [13] in combination with similar data as reported in [14, 15] for the Upper Continental Core (UCC), regarding the Normal – Mid-Ocean Ridge Basalt (N-MORB) [16] or established by the actual Romanian Regulations (RR) concerning the environmental pollution [17]. All statistical analyses were performed by means of StatSoft^{TM} Statistica 6.0.

3. RESULTS AND DISCUSSION

In investigating the distribution of the above-mentioned elements in both lakes, we have been interested in establishing to which extent these elements have an anthropogenic origin, and, at the same time, to evidence any relationship between sediments and the lithology of their drainage basins.

Accordingly, in the table we have reproduced the content of the seven considered elements. Further, in Fig. 3, we have reproduced the R-mode PCA plot of all the seven elements by considering samples as variables and elemental contents as cases, while Fig. 4 illustrates the Q-mode PCA plot when, by considering samples as cases, we have tried to evidence the interrelation between sediments and parent rocks.

As a general rule, both the table and Figs. 3 and 4 point to significant differences between the two lakes concerning the content of all the considered elements.

Owing to its extreme limited applications, the Sc can be regarded as a typical, non-anthropogenic element. In view of that, we have noticed, for each lake, a coincidence within experimental uncertainties of Sc content in sediments and surrounding rocks, but significantly different between lakes (table). This dissimilarity reflects, in our opinion, the existing difference between andesitic rocks, representing the mineral part of St. Ana Lake

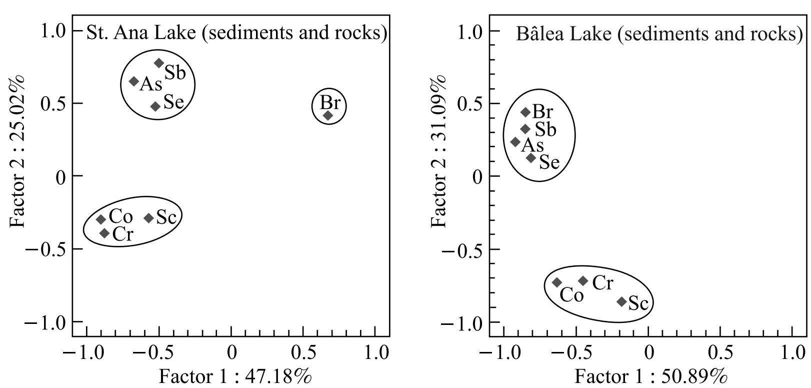

Fig. 3. PCA plot (factor 2 versus factor 1) regarding the relationship of investigated elements for each lake. On both plots, Sc, Cr and Co, on the one hand, and As, Se and Sb, on the other, form distinct cluster, while Br joins the latter only in the case of Bâlea Lake

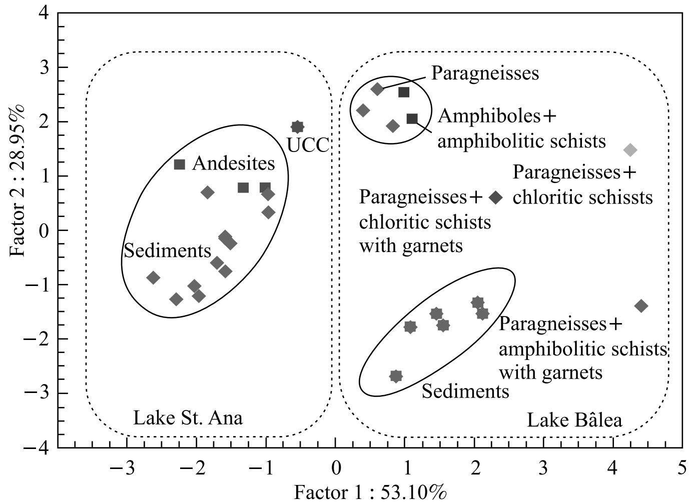

Fig. 4. PCA plot (factor 2 versus factor 1) regarding a classification of all samples with respect to the content of all the seven heavy elements. While all St. Ana Lake samples form a single cluster, Bâlea Lake sediments form a cluster distinct from all other adjacent rock samples, which, in their turn, seem more distributed, by forming few different clusters

sediments and the paragneiss + amphibolites + crystalline limestones association characteristic for the Suru Formation which hosts Bâlea Lake.

We have noticed similar behaviour in the case of Cr and Co, two elements belonging respectively to the 6th and 9th groups of the third period of transition elements.

The average Cr content of St. Ana Lake sediments was (31 ± 3) mg/kg, coincident, within one standard deviation, with the content of (29 ± 10) mg/kg in neighbouring andesitic rocks. In Bâlea Lake sediments, Cr content was (272 ± 39) mg/kg, while the average content of all B samples was (291 ± 274) mg/kg. Regarding latter values, it is worth mentioning the significant differences we noticed between a paragneiss with chloritic schist and garnets (693 mg/kg) and amphibolitic schists (37 mg/kg). These results are in good agreement with those reported for UCC (35 mg/kg) and MORBs (254 mg/kg for the Indian Ocean, 714 mg/kg for the Pacific Ocean), by taking into account that andesites are closer to the UCC model [14].

Regarding RR, the Cr content in St. Ana Lake sediments is about the normal value of 30 mg/kg considered for soils, while in the case of Bâlea Lake sediments, the Cr content of $(272{\pm}39)$ mg/kg is very close to 300 mg/kg, value considered as intervention threshold [17]. As in both lakes the Cr content, although very different in values, appears very similar to parent rock, it can be considered rather a natural component of both sediments and rocks.

The same discrepancy between St. Ana and Bâlea lakes was noticed in the case of Co. Indeed, in the case of St. Ana Lake, both sediments and adjacent andesites showed, within one standard deviation, a coincident content of Co equal to (4.3 ± 0.4) mg/kg and (3.6 ± 1) mg/kg, respectively. Although for Bâlea Lake, these values were ten times greater, i.e., $42{\pm}5$ for sediments and $39{\pm}24$ for rocks, slightly exceeding the Romanian alert threshold of 30 mg/kg [17]. They were coincident within one standard deviation too, enabling us to consider, as in the previous cases, Co as a naturally occurring element.

Both As and Sb, highly chalcophilic and mobile in environment and belonging to the 15th group of elements, are strongly absorbed by clay and organic matter [18].

In our case, we have found in both lakes sediments close values, coincident within one standard deviation, i.e., (4.2 ± 0.4) mg/kg and (5.5 ± 1.8) mg/kg, respectively (see table and Fig. 3), higher than in surrounding rocks. Although about three times greater than those reported for UCC, these values are still close to the normal environmental values as RR states.

Antimony was found in both lakes at concentrations well below 5 mg/kg (see table), considered normal by RR and comparable with majority of soils as reported in [18], but significantly higher than 0.2 mg/kg assigned for UCC. Again in Bâlea Lake sediments, the average Sb content was about six times higher than those found in neighbouring rocks.

Since As and Sb are retained by clay, the higher concentrations found in sediments with respect to rocks could be explained by this peculiarity, sediments playing in this case a role of concentrator.

Regardless of these findings and judging on their content, both As and Sb could be considered in sediments as natural occurring elements, without any sizeable anthropogenic influence.

The next investigated element, Se, belonging to 16th group of elements, has a geochemical behaviour close to sulfur, with a content in UCC between 0.05 and 0.14 mg/kg [14, 15], but rarely exceeds 0.5 mg/kg in soil [18].

In our case, we have noticed for St. Ana Lake sediments an average value of (1.7 ± 0.3) mg/kg, slightly higher than in neighbouring andesites, but lower than (3.0 ± 0.5) mg/kg found in Bâlea Lake sediments. In the case of Bâlea Lake rocks, Se content varied between 0.5 and 2.6 mg/kg (amphibolites and amphibolitic schist) (see table and Fig. 3). All these values occupy an intermediate position between normal (1.0 mg/kg) and alert (3.0 mg/kg) RR values.

Although, as in the case of As, the exact mechanism of Se enrichment in lake sediments is not well established, its presence in neighbouring rocks at comparable contents suggests a natural origin in both lakes, assumption sustained by a relatively restrained use of Se in industry, the total world production of Se in 2008 reaching 518 metric tons.

Bromine, the last heavy element investigated within this project, presents in UCC a content between 0.2 and 10 mg/kg, the highest content being recorded for volcanic soil [15, 18].

According to the table, the average content of St. Ana Lake sediments was (24 ± 7) mg/kg, close within one standard error to the value of (28 ± 16) mg/kg recorded for adjoining andesite, while for Bâlea Lake sediments the Br content was (5.7 ± 1.5) mg/kg, and about 14 times smaller for neighbouring rocks. Regarding this fact, it is worth mentioning that the Br concentration is relatively constant in all the considered samples, regardless of their lithology. As for both lakes the Br content falls well below the RR normal threshold, we can consider, as in the previous cases, that bromine has a natural, not an anthropogenic origin.

In the second part of our study we have examined, by means of R- and Q-mode PCA, the relationship between sediment and parent rocks (see Figs. 3 and 4). In Fig. 3, it can be noticed that Sc, Cr and Co, on the one hand, and As, Sb and Se, on the other, form two distinct clusters, this fact sustaining, in our opinion, the assumption that, regardless of their location, all these elements have the same behaviour. As, without a doubt, Sc and Se are natural constituents, it is reasonable to suppose that the As which enters in the same clusters has a natural, nonanthropogenic nature too. From this point of view, Br represents an exception as it forms a cluster with As, Sb and Se only in the case of St. Ana Lake, while in the case of Bâlea Lake, Br forms a single cluster by itself.

We have interpreted this peculiarity in the case of Br as due to the different lithology of the drainage basins of the two lakes: almost andesite in the case of St. Ana Lake and different associations of gneisses with amphibolitic schists and crystalline limestones in the case of Bâlea Lake.

A Q-mode analysis allowed us to group both sediments and rocks in agreement with the content of all the seven elements considered in this study. Consequently, Fig. 4 shows all 29 samples grouped into two main clusters, one of them containing all St. Ana Lake samples as well as the UCC one, while the other contains all Bâlea Lake specimens. Indeed, both St. Ana Lake sediments and andesites form a single large subcluster, close to UCC one and testifying to their common nature and composition. The second subcluster, more remote from the previous one, consists only of Bâlea Lake specimens, indicating their relative uniformity, but, at the same time, a different behaviour with respect to surrounding rocks, which form different subclusters with a relatively reduced number of members. Therefore, two samples of paragneisses and all amphibolitic schists form one subcluster, while all Bâlea Lake sediments are more remote.

In this way, we can suppose that the observed clusters and subclusters, based only on the content of the considered seven elements, reflect with enough precision the differences between the investigated samples regarding their natural provenance.

4. CONCLUDING REMARKS

The content of the seven heavy elements (Sc, Cr, Co, As, Sb, Br and Se) as determined by the Instrumental Neutron Activation Analysis in 29 samples of sediments and rocks of proglacial Bâlea Lake and volcanic St. Ana Lake was used to estimate to which extent these elements could be considered pollutants.

By comparing their contents with those of surrounding rocks as well as with the numerical values as stated by the Romanian Regulations concerning the Environmental Pollution, it was established that, although their average content was different for the two lakes, all of them could be considered as natural, nonpolluting elements.

The Principal Component Analysis performed both in R- and Q-modes showed that in the case of R-mode, all elements, regardless in which lake, form two similar clusters in concordance with their geochemical affinities, while in the case of Q-mode, all samples form few distinct clusters, rather reflecting their provenance.

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