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RARE ALKALI ELEMENTS IN THE HUNGARIAN GEOLOGICAL FORMATIONS: A COMPREHENSIVE STUDY.

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ABSTRACT

The authors of the scientific report "Comparative study of rare alkali elements (Li, Rb, Cs) distribution in magmatic, metamorphic, and volcano-sedimentary formations in Hungary" (Kubovics et al. 1992a) prepared within the framework of NSRF (OTKA) No. 728/87 intended to answer the questions raised for more than 40 years ago by SZÁDECZKY-KARDOSS (1955).

The rare alkali element contents of geological formations in Hungary were systematically studied.

It has been established that the highest Li-concentrations in Hungary are linked to lithiophorites found in bauxites and Mn-ores.

The differentiation products of the granites in the Velence Mts namely aplites and orthoclase from pegmatitic nests are most enriched in Rb.

The enrichments of Cs in various rock types - independently of their origin, e.g. in lamprophyres, basalts, andesites, rhyolites, and their pyroclastics - proved to be the result of postmagmatic alteration that resulted in zeolitisation which is the principal host of Cs.

INTRODUCTION

The alkali metals form a group of extremely reactive elements with simple electronic configurations ns^1. They have large atomic (and ionic) radii and low ionization energies compared with other elements. These properties lead to low melting point, boiling point, density and low heats of sublimation, vaporization and dissociation. The alkali metal compounds are generally characterized by high solubility in water, their hydroxides are the most basic of all ones. Their oxidation state in compounds never exceed +1.

Their chemical properties are quite similar, except for lithium in some respects, which is the smallest element in the group and has the highest ionization energy, melting point and the lowest density.

The small size of lithium ion frequently causes special properties in its compounds as well. It shows many geochemical similarities to Mg because of the similarity in ionic size of the two elements: $r(Li^+)$76pm, $r(Mg^{2+})$72pm (SHANNON-PREWIT-type ionic size in 6 coordination number, in: GREENWOOD and EARNSHAW 1984). These "anomalous" properties are for example: lower solubility of some of its compounds, and occurrence in natural ferromagnesian minerals where it partly replaces magnesium.

The distribution of alkali elements in the various geological formations has been well studied. It is particularly true for the two common alkali metals, Na and K. The distributional

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charasteristics of the rare alkalis - considering that these elements have become important industrial raw materials in the recent decades - are also relatively well-known.

PREVIOUS STUDIES

The first comprehensive results on the rare alkali concentrations of geological formations in Hungary were published by SZÁDECZKY-KARDOSS (1955). He emphasized that Li was most probably enriched in the Velence and Mórágy Mts granitoids and especially pegmatites (pegmatitic mica). He also underlined the possibility of a secondary enrichment of Li-bearing mica in the surrounding sedimentary deposits. Beside these he stressed some possible Li-enrichment in the marine iron ore deposit in the Mecsek Mts as well as the glauconite-bearing sandstones in Nógrád.

In connection with Rb and Cs he emphasized that both elements were expected to be enriched in high-K rocks, like Mórágy Mts granitoids and especially pegmatites, Mecsek Mts phonolites /$K_2O \approx 4\%$/, and Telkibánya K-metasomatites ($K_2O \approx 10$-12%).

The preliminary results of the Nation-wide Rare Element Research Program co-ordinated by the Hungarian Geological Institute (1966-1970) were summarized by FÖLDVÁRI-VOGL (1970). No significant Li-deposit was found during this regional study, however, this element was relatively enriched in the hydrothermally altered slate around the Velence Mts granite. There were also no significant enrichments in coal ashes, only some minor elevated concentrations were found in the coals of Tatabánya and Pusztavám.

As to Rb, this element was found to show no enrichments of economic importance in the formations of Hungary.

According to the results of this study there was Cs-enrichment in the range of about 80 ppm in the grey granite porphyry out of the granitic rocks in the Velence Mts. The principal Cs-bearing mineral was biotite, while a small proportion was in the feldspar. The informations on the Cs-distribution in the geological formations of Hungary, however, remained scarce after the study because of methodological difficulties.

Among the previous works it is important to mention the results of VARGA et al. (1975) on the Rb-contents of some volcanic rocks in the Mátra Mts. The rocks studied were all K-enriched with high adularia and sericite. The spectral analyses yielded K/Rb-ratios, on the basis of which the Rb-content could be calculated. The highest values obtained are summarized in Table 1.

TABLE 1

Calculated Rb content of some K-rich rock in the Mátra Mts. (VARGA et al. 1975)

	K_2O %	Rb %
Bányabérc road cut	10.78	0.34
Bányabérc shaft	11.13	0.37
Korlát-hill	12.85	0.46
Korlát-hill, south edge	13.08	0.47
Kőmorzsás-peak	11.71	0.30
Felsőállás-peak	9.91	0.32
Fekete-leak	12.00	0.52

AIMS OF THE STUDY

New types of Li, Rb and Cs enrichments were found during the recent years mainly in volcano-sediments (e.g. in Usbekistan and Kazakhstan) but in perlites as well (YANEV 1994). So we also considered timely the evaluation of the Hungarian geological formations from this point of view.

Within the framework of the survey we studied the distribution of Li, Rb and Cs in the wide range of magmatic, metamorphic and volcano-sedimentary formations in Hungary using optical emission and atomic absorption methods considering foreign analogies.

METHODS OF INVESTIGATIONS

Atomic emission spectroscopy (AES)

More than 300 selected samples were analysed by emission spectrographic method. Because of Li, Rb and Cs concentration values are commonly below detection limits of conventional AES methods, one author developed a special AES technique for determining rare alkali elements concentration that is more sensitive than has been previously used (HOFFMANN and NAGY-BALOGH 1990, NAGY-BALOGH and HOFFMANN 1991, NAGY-BALOGH 1992a,b).

Li, Rb and Cs were determined using alternating current arc excitation. Matrix effects were minimized by buffering the sample with high purity NaCl. 25 mg of a 1:1 homogeneous mixture of powdered sample and NaCl were loaded into high purity aluminium electrode, dropped with 2 drops phenol-formaldehid rezin and heated at 160°C for 30 minutes. The loaded and heated electrodes were excited by arc at 5 amps for 100 seconds. International standard samples were treated in the same manner.

Spectra were detected by grating spectrograph PGS-2 (Zeiss, Jena) on type 1-N (Kodak-Pathe) infrared sensitive photoemulsion. The wavelengths used for analysis were: Li 812,6; Rb 780,0 and 794,7; Cs 852,1 nm; reference lines Na 818,3 and

819,4 nm. The Na lines had extremely high intensity and it was out of the range suitable for densitometry. Decrease of Na line intensity was accomplished by a neutral filter in front of Na wavelength position. Less than 3% Na-content of rock samples does not disturb the use of Na as reference element.

Quantitative evaluation was made by digital microphotometer connected to a microcomputer.

Detection limit for Li is 4 g/t, Rb 1g/t, Cs 1 g/t, 8-10% relative standard deviation.

Neutron activation analysis (INAA)

In the course of earlier studies more than 100 samples were analysed for rare elements, inclusive Cs, by INAA.

INAA method was used also for control analyses in the low, medium and anomalously high Cs-concentration ranges. The results of AES and INAA were in a good agreement, systematic error was not noticed. The INAA analyses were made at the Nuclear Technical Institute of Technical University, Budapest.

Flame atomic absorption and emission spectrometry was the control method for Li and Rb analysis. After acidic ($HF+HClO_4$) digestion Varian AA-475 instrument, Y-capillar and standard addition technique was used for Li and Rb determination.

RESULTS AND DISCUSSION

Beside our new analyses rare alkali determinations found in theses prepared in the Department of Petrology and Geochemistry, Eötvös University as well as the results of the above mentioned Nation-wide Rare Element Research Program were also used for evaluation.

The location of sampling points is displayed in *Fig. 1.*

Our results have been summarized according to the rock types studied (Table 2).

The contents of the rare alkali elements in *ultramafic and lamprophyric rocks* correspond to their average clarke values except for those samples that suffered some kind of postmagmatic alteration.

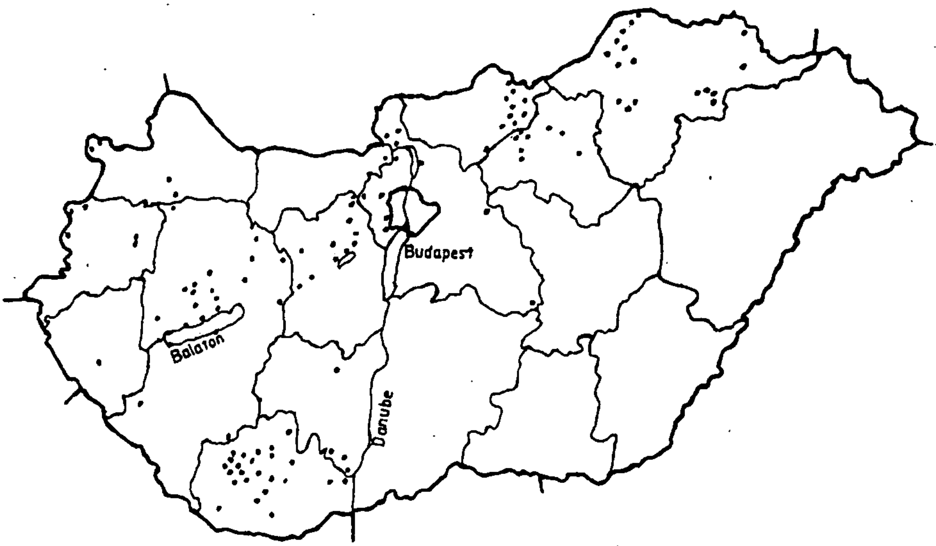

Fig. 1. Sampling points for rare alkali elements determinations in Hungary

TABLE 2

Formations measured for rare alkali element concentrations

– lamprophyres
– basaltic rocks
– andesite, dacite, trachyte, rhyolite and their pyroclastics
– intrusive magmatic rocks
– metamorphic rocks
– sedimentary rocks (bauxite, Mn-ore, glauconitic sandstone, limestone, gypsum containing aleurolite, alginite, etc.
– rocks of hydrothermal origin (zeolitised rhyolitic tuffs, clinoptilolite, mordenite, analcime (Csódi-hill), hydro-muscovite, fluorite, barite
– monomineral fractions
– soils
– mineral and medicinal waters

TABLE 3

Alkali element contents of Hungarian basalt samples

	Na %	K %	Li g/t	Rb g/t	Cs g/t	K/Rb	K/Cs	Rb/Li	Na/K	Rb/Cs
Somoskõ	4.21	2.01	8.4	52	0.28	390	72500	6.6	2.1	186
Salgó	3.66	1.87	9.3	45	0.56	416	33393	4.8	2.0	80
Salgó	3.97	1.89	8.8	44	0.66	430	28623	4.8	2.1	67
Kis-Salgó	4.08	1.84	9.3	45	0.85	409	21647	5.0	2.2	53
Tóti-hill	3.05	1.64	10.7	42	1.00	390	16400	3.9	1.9	42
Halyagos SW quarry	3.81	1.36	9.3	45	0.94	302	14468	4.8	2.8	48
Gulács	2.77	2.05	9.75	41	1.22	500	16803	4.2	1.4	34
Mindszentkálla	3.11	2.24	8.4	46.6	0.94	481	23830	5.5	1.4	50
Halyagos Lukács-mine	3.58	2.01	9.3	47.5	1.32	423	15227	5.1	1.8	36
Badacsony Lábdi hill	3.53	2.01	9.3	51.2	1.13	392	17780	5.5	1.8	45
Nógrád average of 4 samples	3.98	1.91	9.0	46.5	0.59	410	32300	5.2	2.09	78.8
Balatonfelvidék average of 6 samples	3.31	1.88	9.5	45.6	1.19	413	18851	4.8	1.76	38.3
average of 10 samples	3.58	1.89	9.3	46	0.9	413	26068	5	1.89	64.1

Out of the *basic volcanic rocks* those of Pannonian age are characterized with Li, Rb and Cs contents typical for continental alkaline basalts (Table 3). Basaltic rocks with older age contain more Rb and Cs than their average clarke-values for this rock type because of postmagmatic alteration to clay minerals and/or zeolites. Exceptionally high Cs-content of average 150 ppm was detected in both the outcrop and drill core samples from the leucitic lamproitic rock (PUSKÁS et al. 1993) near Bár, South Hungary. This can be explained by the loose framework crystal structure of leucite that may host the large Cs-ion.

In the *intermediate volcanic rocks* (andesite, dacite and their pyroclastics) rare alkali concentrations corresponding to their average clarke values were found, i.e. a little higher values than in the basic rocks. Specific geochemical significance can be attributed to the relatively high Cs-contents of neogene volcanics: this appear to exclude their island arc origin (STAVROV 1978). Island arc andesites and dacites, similarly to basalts, contain Cs under the 1 ppm level, while in our case the average measured values were around 5 ppm, corresponding to continental margin position of the volcanics. In the case of hydrothermally altered rocks the Cs-content may even attain 30 ppm, e.g. in the analcime containing andesite-dacite of Csódi hill, Visegrádi Mts.

According to the lithophile geochemical behaviour of alkali elements their concentrations in *acidic volcanic rocks* are higher than in the basic and intermediate volcanics. Outstanding enrichments, however, were found in the acidic pyroclastics altered by postvolcanic processes, namely Li=30-40 ppm, Rb=100-350 ppm, Cs=2,5-145 ppm.

Among the *intrusive magmatic rocks* basic ones contain rare alkali elements in low concentrations in accordance with their low clarke values in these rock types. In the granitoids of the Mecsek Mts the concentrations of rare alkali elements are higher as a consequence of their higher mica-content. The highest concentrations, however, were detected in the more evolved and differentiated Velence Mts granite. Conspicuously high Rb content was characteristic for their aplite (464-514 ppm) and pegmatitic orthoclase (797 ppm).

The rare alkali element contents in *metamorphic rocks* are principally constrained by their concentration in the pre-metamorphic protolith. We found elevated rare alkali concentrations in the amphibolite facies metamorphic rocks in the Mecsek Mts, the reason of which, however, needs further clarification.

Out of the *sedimentary rocks* only those have been involved into the scope of our measurements, in which some enrichments of any of the rare alkali elements were expected on theoretical or practical considerations.

Considerable Li enrichments were found in some bauxite samples. We supposed that Li was contained in the lithiophorite identified by BÁRDOSSY (1977).

Numerous measurements were carried out from the Mn-oxide crusts underlying bauxite bodies as well as the samples from the Úrkút Mn-ore body that were previously identified as lithiophorite. Comparing our results with those of VARENTSOV and GRASSELLY (1980) we found that the samples studied belong to the low-Li lithiophorite varieties. Their highest Li-content was 968 ppm.

Glauconite-containing sandstone and monomineral fractiones of glauconite thereof that SZÁDECKY-KARDOSS (1955) considered as most promising in view of their possible elevated rare alkali element contents proved to be not enriched in these elements. That is their higher K content was not followed by elevated Rb and Cs contents.

Out of the evaporites, we have analysed some gypsum-and anhidrite-containing aleurolite samples. Their rare alkali element contents corresponded to the average clarke values in these rocks.

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Our studies pointed to the importance of *post-volcanic processes* in the enrichment of rare alkali elements, so this problem has been dealt with in more detail. K-enriched volcanics originated by K-metasomatism have shown some Rb enrichment (to a max of 486 ppm), but this is far below the values supposed by VARGA et al. (1975). It has been established that there is no Rb enrichment concomitant with increasing K-content during the metasomatic process. This is also true in that case when K-mica predominates K-feldspar as is the case in Nagybörzsöny, Börzsöny Mts where the K-content of hidromica is 3%, whereas its Rb-content is merely 72 ppm.

The most significant Cs enrichments were discovered in zeolitised rocks (KUBOVICS et al. 1993).

The clinoptilolite and mordenite from Mád, Tokaj Mts contained 34 and 55 ppm Cs, respectively. The highest Cs-concentration in Hungary was found in the analcime from Dunabogdány Csódi hill, Visegrádi Mts (1153-1556 ppm).

The halloysite from the Gyöngyösoroszi ore field, Mátra Mts, contains 85-95 ppm Li in average. The clay minerals of the ore-containing dykes in Gyöngyösoroszi consist of illite, montmorillonite and halloysite. There is a max Li-content of 132 ppm, Rb-content of 218 ppm and Cs-content of 18 ppm in these vein-filling clay minerals. Rb and Cs are most probably fixed there by adsorption.

Studies on monomineral fractions

The monomineral fractions of some well-separable rock types (granite, granodiorite, gabbro, glauconitic sandstone) have also been analysed for rare alkali elements. In agreement with the previous data, biotites from granites and granodiorites were found to be enriched in Li, Rb and Cs as compared to K-feldspar. Out of our measurements we find worth of mentioning the Rb and Cs distribution inside the zoned microcline porphyroblasts from the granitoide near Pörböly, Mecsek Mts. The porphyroblasts there have a reddish core shifting towards a pinkish intermediate zone and then to a colourless outer rim. The Rb and Cs contents are 204 ppm and < 2 ppm in the core, while 213 ppm and 3 ppm in the intermediate zone and 306 ppm and 5,5 ppm in the rim. This change represents a certain shift in the rare alkali content of the crystallizing medium during porphyroblast formation.

Soils

The investigation of representative soil samples from various parts of the country has confirmed the widely accepted experience that the rare alkali element contents of soils mainly depend upon its mineral composition. However rare alkali elements can also be enriched by adsorption in clay minerals and humus. The highest Rb and Cs concentrations in soil, however, are linked to zeolites, too. Exceptionally high Cs-contents of 67 to 81 ppm were measured in the soil formed on zeolitised rhyolitic tuff at Gerendásrét, Tokaj Mts (KUBOVICS et al. 1992b).

Waters

The Li-concentrations of mineral and medicinal waters in Hungary is known for 40 years (SCHULHOFF 1957). These measurements have been completed by the National Public Health Institute (OKI) with data referring to Rb and Cs contents of mineral and medicinal waters. Out of these data the Li, Rb, and Cs concentrations of the thermal water at Mezőkövesd are conspicuous (1880, 336 and 430 ppm, respectively). It is presumed that these high concentrations are genetically connected to the rhyolitic tuffs in Bükkalja (South Bükk Mts).

TABLE 4

Average cesium concentrations in the major hungarian geological formations (g/t)

	fresh rocks	*rocks altered by post-volcanic fluids*
Volcanic and sub-volcanic rocks		Transdanubian Mid-Mts:
ultramafic rocks and lamprophyres	< 1	7-37 Alcsútdoboz 2.1-3.2 Vál-borehole No. 3. max. 230 Budaörs-borehole No. 1.
mafic rocks		
Mesozoic basalt	< 1	1.4-13.1 (max. 36) Mecsek Mts (max. 80) W-Hungary
Neogene basalt	0.9-1.9 anomaly: K-rich basaltoid from Bar (150 g/t)	
intermediate rocks (andesite, dacite)	5-16 Balatonmária, Börzsöny Mts, Mátra Mts	30 analcim andesite Dunabogdány, Csódi hill
acid-rocks (rhyolite, rhyolitic tuffs)		
Paleozoic (Permian)	6 Mecsek Mts	110-147 zeolitised rhyolite tuffs
Mesozoic	14 Bükk Mts	23 uraniferous phosphatised rhyolitic tuffs, Bükkszentkereszt, Bükk Mts.
Neogene	10 Mátra Mts, Tokaj Mts	
Intrusive rocks		
gabbro (Mesozoic, Triassic)	< 1 Recsk, Mátra Mts	3 Mecsek Mts: Kurd-borehole No. 2.
granodiorite (Paleozoic)	3.3. Mecsek Mts	
granite (Paleozoic)	4.6 Velence Mts	
Metamorphic rocks		
shale	< 1 N-Hungary, Rudabánya	
mica schist	4.6 Mecsek Mts; W-Hungary	
amphibolite	3.4 Mecsek Mts	
Volcano-sedimentary rocks		
Neogene basalt-bentonite	3.2 Balaton Highland	
alginite	1.5 Balaton Highland	
Neogene tuffites	5 Mecsek Mts	
glauconite Schlier	10 Mecsek Mts	
glauconitic sandstone	5.6 N-Hungary; Rudabánya, Diósgyőr	
gypsic shale	<1-2 N-Hungary	
arkosa	24 Mecsek Mts	
Soils	1.1-81 different places in Hungary	
Mineral and Medicinal waters	2-23 μg/l (max. 430 μg/l Mezőkövesd)	
Monomineral fractions		
biotite from granite	2.7 Velence Mts 24-43 Mecsek Mts	
fluorite	<1 Velence Mts	
Cr-illite	7.5-20 Mecsem Mts from uraniferous ore	
hydromuscovite	5.1 Börzsöny Mts: Nagybörzsöny	
halloysite	4 Mátra Mts: Gyöngyösoroszi	
zeolites: clinoptilolite	34 Tokaj Mts: Mád	
mordenite	55 Tokaj Mts: Mezőzombor	
analcime	1153-1556 Visegrád Mts, Dunabogdány, Csódi hill	

As an example, the distribution characteristics of Cs, the most variable rare alkali element in the major Hungarian geological formations is displayed in Table 4.

Suggestions for further studies

On the basis of informations obtained during our comprehensive survey it is deemed advisable to carry out the following studies:

– to delineate regions with anomalously high Cs-concentrations, e.g. in the case of Cs-rich zeolitic rocks. It is presumed that even industrially important Cs-rich zeolitic deposits could thus be found.

– to investigate the possibilities of Cs-extraction from zeolites during their preparation for technical purposes.

– to carry out biogeochemical studies on areas with higher-than-average clarke values for Li, Rb, and Cs, as these data are practically absent for Rb and Cs, and even the biochemical role of Li is not unanimous. This kind of interdisciplinary studies would need the involvement of experts from different fields of science.

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