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ALBERTO BENCINI, ANGELO PECCERILLO

**K, Rb and Sr distribution in calc-alkaline rocks from
Eastern Carpathians**

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Geochimica. — K, Rb and Sr distribution in calc-alkaline rocks from Eastern Carpathians. Nota di ALBERTO BENCINI(*) e ANGELO PECCERILLO(**), presentata(**) dal Socio G. CAROBBI.

RIASSUNTO. — È stato determinato il contenuto di K per fotometria di fiamma e il tenore in Rb e Sr per spettrofotometria di assorbimento atomico in 39 campioni di rocce provenienti dalla catena vulcanica Cenozoica dei monti Calimani-Gurghiu-Harghita (Carpazi Rumeni). In base allo schema di classificazione per rocce di zone orogeniche proposto da Peccerillo e Taylor (1976 b), i campioni in esame appartengono per la quasi totalità alla serie alcali-calcica. Pochi campioni appartengono alla serie alcali-calcica ricca in potassio e alla serie shoshonitica.

Il contenuto in potassio delle vulcaniti non mostra alcuna tendenza all'aumento durante i due stadi del vulcanismo dei Carpazi rumeni, ad eccezione dei prodotti finali shoshonitici caratterizzati da tenore in K_2O notevolmente più alto.

Nelle rocce alcali-calciche il tenore in Rb varia da 20 a 99 ppm con valori medi vicini a quelli trovati nelle serie alcali-calciche di arco insulare e di margine continentale. Lo Sr varia da 225 ppm a 605 ppm con valori simili ai tenori medi di rocce alcali-calciche di arco insulare.

Le rocce appartenenti alla serie alcali-calcica ricca in potassio hanno valori di Sr e Rb più alti rispetto ai campioni alcali-calcici tipici.

Le shoshoniti hanno tenore in Sr molto alto (2340-2600 ppm).

È stato osservato che non esiste relazione fra abbondanza di questi elementi e tipo di fenocristalli presenti nelle rocce. Infatti sia le rocce a pirosseno che quelle ad orneblenda hanno tenori in K, Rb e Sr molto simili. Questo fa ritenere che la distribuzione di tali elementi in queste rocce sia regolata dalla massa di fondo piuttosto che dal tipo di fenocristalli.

L'alto contenuto in K delle shoshoniti è da attribuire alla presenza di biotite.

Una caratteristica peculiare delle rocce oggetto del presente lavoro è data dal rapporto K/Rb che varia da 197 a 343 nei termini alcali-calcici, con valori medi più bassi di quelli trovati in altre serie alcali-calciche di catene vulcaniche attive. Questo carattere particolare può essere spiegato se si ammette, in accordo con Armstrong (1971), che una parte dei sedimenti della fossa oceanica sia trasportata nel mantello durante la subduzione e partecipi ai processi di fusione parziale che originano il magma alcali-calcico. Un altro processo che potrebbe abbassare il rapporto K/Rb nei magmi è la partecipazione alla fusione parziale di flogopite, che è stabile nel mantello fino a profondità di circa 200 km e che è caratterizzata da un basso valore del rapporto K/Rb.

INTRODUCTION

The genesis of the orogenic calc-alkaline series represents a major problem of igneous petrology. During the last few years a number of hypotheses have been put forth in order to account for the petrological and geochemical features of these rocks and their relation to processes of lithospheric subduction. Although it is generally accepted that calc-alkaline rocks are generated

(*) Istituto di Mineralogia, Petrografia e Geochimica dell'Università, Via Lamarmora 4 Florence (Italy).

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by the same genetic process regardless of the geological setting of their emplacement, some authors (Jakeš and White, 1971; Pichler and Zeil, 1972) believe that andesites erupted on Andean-type margins are generated by partial melting within the continental crust rather than in the mantle, as hypothesized for island arc calc-alkaline sequences (Green, 1972; Nicholls and Ringwood, 1973; Stern and Wyllie, 1973).

A useful tool for understanding whether or not more than one single model has to be invoked for the genesis of these rocks is a widespread geochemical investigation on calc-alkaline series coming from different geological settings.

The Carpathians are an intracontinental arc which shows differences from both island arcs and Andean-type continental margins. In fact, it lies on a continental type basement whose thickness, however, does not exceed 45 km (Soculescu *et al.*, 1964), which is much less than Andean margins. Accordingly, a geochemical investigation on Carpathian calc-alkaline volcanics can give some contributions to the understanding of the problem regarding the relationship between tectonic setting and calc-alkaline magma genesis.

Several geochemical studies have been carried out on these rocks (e.g. Peltz *et al.*, 1973 a, b; Boccaletti *et al.*, 1973; Peccerillo and Taylor 1976 a). Data on major and many trace elements are at present available, however data on Sr and Rb are still lacking for some large stretches of the Calimani-Harghita Mts. which represent the southern part of the Carpathian arc. The petrological interest of these two elements lies in the fact that owing to its large ionic radius, Rb is camouflaged in the potassium minerals and Sr is enriched in alkali feldspars and plagioclases (Berlin and Henderson, 1969). It is well known that all these minerals play an important role in magma genesis and differentiation.

In this paper data on K, Rb and Sr for 39 samples coming from the Calimani-Harghita volcanic chain are reported with the primary aim of providing new data on East Carpathian rocks and discussing their genetic significance.

VOLCANIC SETTING

Since a detailed description of the geological and volcanic setting of the Calimani-Harghita Mts. has been given by several authors (e.g. Radulescu and Borcoş, 1964; Peltz *et al.*, 1973 a), only a brief summary following these authors is given here.

The Calimani-Gurghiu-Harghita belt represents the southern stretch of the Carpathian volcanic arc. It runs for a distance of about 160 km from Vatra Dornei in the north to Tuşnad in the south (fig. 1). The volcanic activity of the Calimani-Harghita belongs to the third cycle of the volcanism of the Rumanian Carpathians. This cycle developed in two stages: the first one started during the Pannonian with eruption of a small amount of dacitic lavas

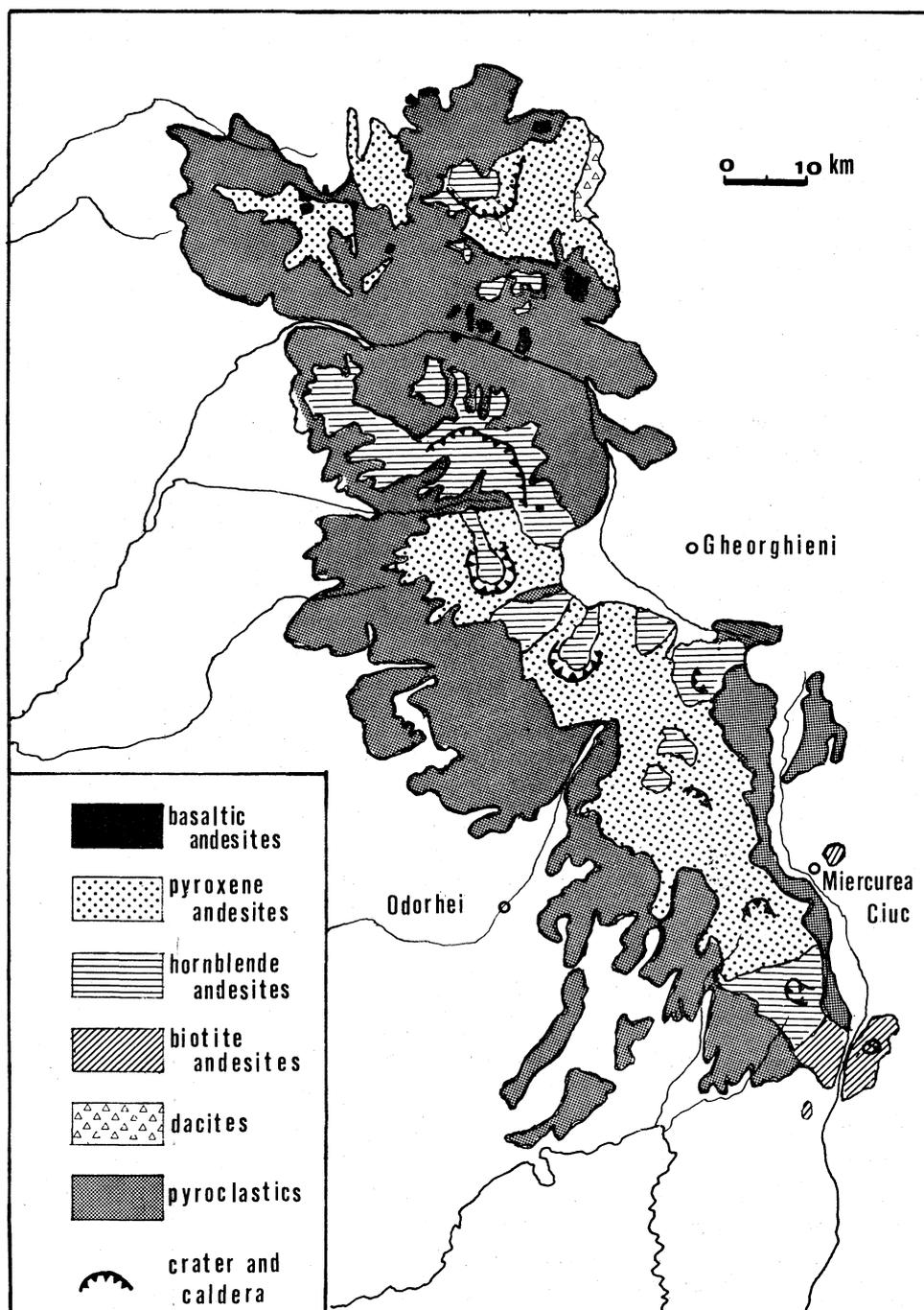


Fig. 1. - Simplified geological map of the calc-alkaline volcanics from the Calimani-Gurghiu-Harghita mountains (from the Geological Map of Rumania, scale 1:200,000).

followed by hornblende, pyroxene and basaltic andesites which are the latest products of this stage. The volcanic activity was prevalingly subaqueous and the products are essentially pyroclastic with a large occurrence of effusive events as well. The second stage started at the end of the Pliocene after a period of volcanic quiescence. The first products are hornblende andesites followed by pyroxene, biotite and basaltic andesites. The activity took place essentially on landmasses.

The latest activity of the Carpathian volcanism occurred in the Perșani Mts. situated on the maximum bend area of the Carpathian arc, where the subduction is still apparently active (Roman, 1970).

ANALYTICAL METHODS

For the determination of K, Rb and Sr 500–1000 mg of rock powder were attacked with HF-HClO₄, evaporated to dryness, dissolved in HCl and diluted to 50 ml. K was determined by classical flame photometric methods. Sr and Rb were analyzed by atomic absorption spectrophotometry using a Perkin-Elmer 303 apparatus with a three-slot burner, equipped with a Hitachi-56 recorder. A standard addition procedure (Cioni *et al.*, 1971) using La as masking agent was employed for Sr determination. Rb was determined basically using the method of Roelandts (1972), adding K and La directly to an aliquot of the attack solution for suppressing the interferences. Care was taken in checking the HCl concentration in both samples and standard solutions. The precision of the method is $\pm 3\%$ for both Rb and Sr for the range of values found in our samples. SiO₂ was estimated by X-ray fluorescence analysis directly on powder pellets using several international rock standards for curve calibration. The accuracy is poor (about 1.5%) so the silica data will be used in order to have the SiO₂ range of the samples under study and to have a rough picture of trace element variation within this range.

RESULTS

The data on the analyzed samples are reported in Table I, together with the main phenocryst mineralogy and the magmatic stage for each sample. Fig. 2 shows the SiO₂ versus K₂O relationship for the analyzed samples. The classification is from Peccerillo and Taylor (1976 b). The boundaries on both SiO₂ and K₂O divisions are an attempt to divide a continuum and are not to be considered as rigid. Accordingly, some samples, especially those plotting close to the boundaries, can actually belong to either one of the adjoining fields, considering also the accuracy of the SiO₂ values.

Almost all the samples are calc-alkaline andesites with minor basaltic andesites and dacites. Few samples fall into the field of the high-K calc-alkaline series. Samples R 107 and R 105 have the high K content typical of shoshonitic rocks.

TABLE I

Analytical data on East Carpathian volcanic rocks

Sample	SiO ₂ %	K ₂ O %	Sr ppm	Rb ppm	K/Rb	Rb/Sr	Volcanic stage	Phenocryst minerals
CALC-ALKALINE ROCKS								
Basaltic andesites								
R 177	52.3	1.00	310	30	276	0.097	II	<i>pl, horn, px</i>
R 50	52.7	0.87	225	24	300	0.107	I	<i>pl, horn, px</i>
R 239	53.3	1.07	310	45	197	0.145	II	<i>pl, px</i>
R 213	53.7	0.87	310	31	232	0.100	I	<i>pl, px</i>
R 165	53.7	0.98	390	30	271	0.077	I	<i>pl, horn</i>
R 52	54.0	0.79	260	20	327	0.077	I	<i>pl, horn, px</i>
R 60	54.4	1.29	325	39	274	0.120	I	<i>pl, px</i>
R 76	55.1	1.50	465	41	303	0.088	II	<i>pl, horn</i>
R 74	55.6	1.29	290	40	267	0.138	I	<i>pl, px</i>
R 127	55.9	1.15	295	34	280	0.115	II	<i>pl, px</i>
R 166	55.9	1.49	240	57	216	0.237	I	<i>pl, px</i>
Andesites								
R 237	56.0	0.71	305	23	256	0.075	II	<i>pl, horn, px</i>
R 89	56.6	1.32	430	39	280	0.091	II	<i>pl, px</i>
R 119	57.2	1.76	525	55	265	0.105	II	<i>pl, horn</i>
R 94	57.3	1.76	450	67	218	0.149	I	<i>pl, px</i>
R 133	57.5	1.69	315	63	222	0.200	II	<i>pl, horn</i>
R 304	57.5	1.25	280	44	235	0.157	II	<i>pl, horn</i>
R 87	58.0	1.38	390	44	260	0.113	II	<i>pl, px</i>
R 192	58.2	1.87	330	76	204	0.230	I	<i>pl, horn, px</i>
R 95	58.4	1.58	355	62	211	0.175	I	<i>pl, px</i>
R 116	59.0	1.85	520	62	248	0.119	II	<i>pl, px</i>
R 312	59.5	1.20	280	29	343	0.104	II	<i>pl, horn</i>
R 123	59.8	1.80	605	55	272	0.091	II	<i>pl, horn, px</i>
R 130	60.6	1.88	390	62	251	0.159	I	<i>pl, horn</i>
R 187	62.2	1.49	285	56	220	0.196	II	<i>pl, horn, px</i>

TABLE I (continued).

Sample	SiO ₂ %	K ₂ O %	Sr ppm	Rb ppm	K/Rb	Rb/Sr	Volcanic stage	Phenocryst minerals
CALC-ALKALINE ROCKS (continued)								
Dacites								
R 79	65.9	1.28	310	38	279	0.123	II	<i>pl, horn</i>
R 149	66.5	1.35	335	44	255	0.131	I	<i>pl, horn</i>
R 198	67.2	1.27	330	43	245	0.130	I	<i>pl, horn, px</i>
Rhyolite								
R 258	72.9	2.35	250	99	197	0.396	I	<i>alk-feld., q</i>
HIGH-K CALC-ALKALINE ROCKS								
Basalts								
R 157	49.2	1.32	295	48	228	0.163	I	—
R 183	51.9	1.85	405	51	301	0.126	I	—
High-K basaltic andesites								
R 153	52.6	1.93	300	71	225	0.237	I	—
R 143	54.1	1.77	470	55	267	0.117	I	—
R 220	54.7	2.06	355	96	178	0.270	I	<i>pl, horn</i>
High-K andesites								
R 234	57.0	1.93	355	75	213	0.211	II	<i>pl, px</i>
R 100	57.5	2.08	1450	57	302	0.039	II	<i>pl, px</i>
R 141	59.4	2.35	410	99	197	0.241	II	<i>pl, px</i>
SHOSHONITIC ROCKS								
R 107	56.4	3.87	2600	64	501	0.025	II	<i>pl, biot</i>
R 105	57.6	4.01	2340	70	475	0.030	II	<i>pl, biot</i>

Within the calc-alkaline series, K₂O shows a tendency to increase with increasing silica contents, but dacites have a smaller potassium content than andesites.

Sample R 258 appears to be a typical calc-alkaline rhyolite. The overall distribution of K₂O is approximately constant over the two magmatic stages except at the end of the volcanism when the shoshonitic lavas were erupted.

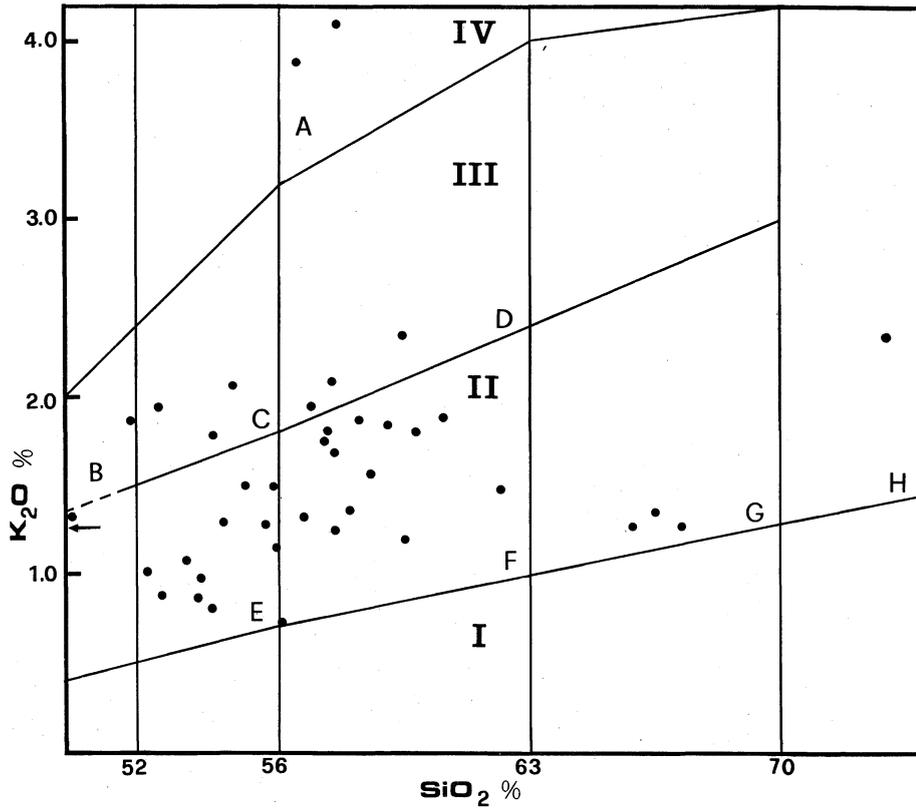


Fig. 2. - SiO_2 versus K_2O relationship for the samples analyzed. I: island arc tholeiitic series; II: calc-alkaline series; III: high-K calc-alkaline series; IV: shoshonitic series. A) banakite; B) basalt; C) high-K basaltic andesite; D) high-K andesite; E) basaltic andesite; F) andesite; G) dacite; H) rhyolite.

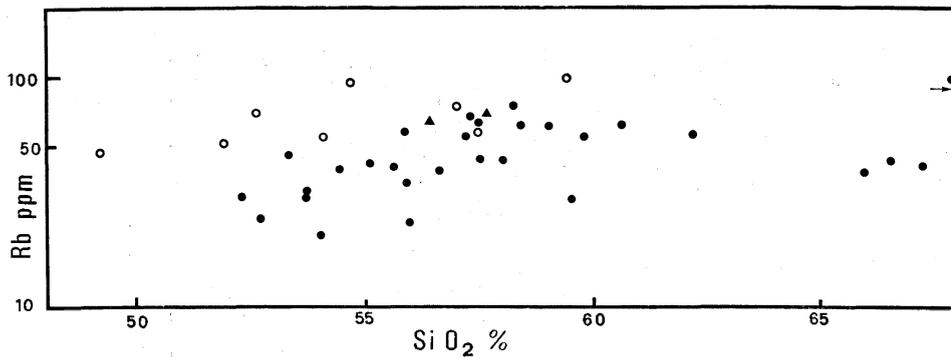
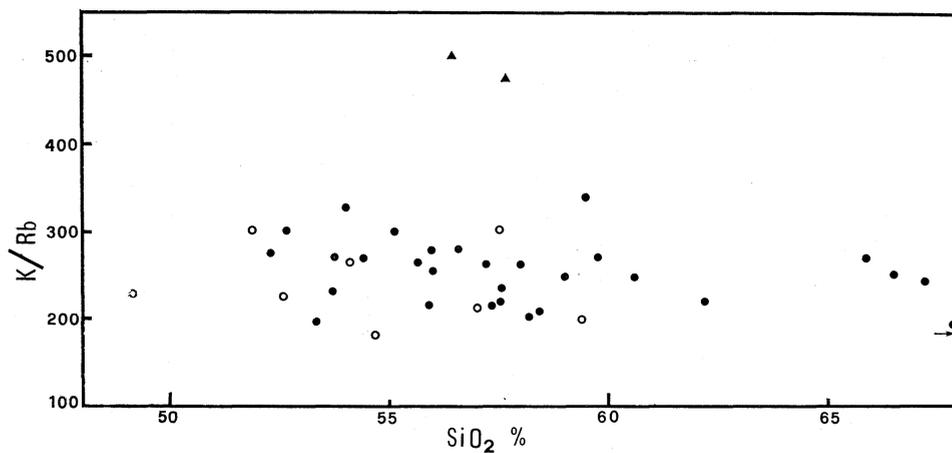
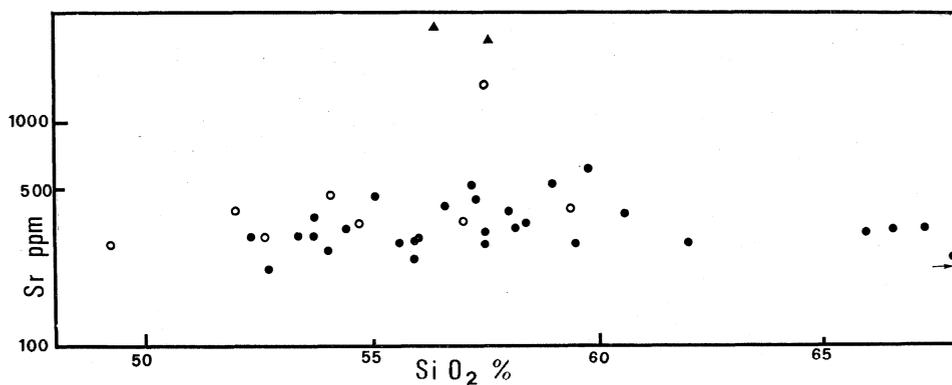
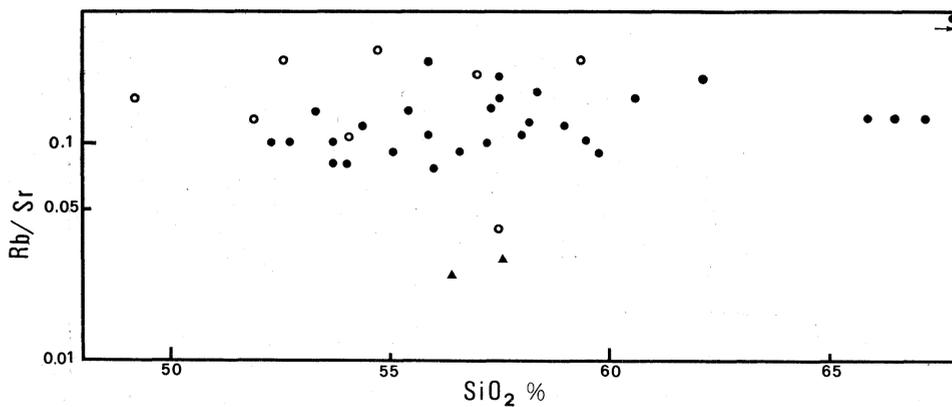


Fig. 3. - Rb versus SiO_2 diagram. Full circles = calc-alkaline samples; open circles = high-K calc-alkaline samples; triangles = shoshonitic samples.

Fig. 4. - K/Rb ratio versus SiO₂ diagram. Symbols as in fig. 3.Fig. 5. - Sr versus SiO₂ diagram. Symbols as in fig. 3.Fig. 6. - Rb/Sr versus SiO₂ diagram. Symbols as in fig. 3.

Rb values range from 20 ppm to 99 ppm in typical calc-alkaline samples, showing a poorly defined tendency to increase with increasing SiO₂ contents especially if dacite values are disregarded (fig. 3). The high-K calc-alkaline rocks show higher Rb contents in respect to typical calc-alkaline samples. The average Rb values found in the analyzed calc-alkaline samples are slightly higher than the average calc-alkaline content given by Jakeš and White (1972) for the same SiO₂ value.

K/Rb ratios are rather low and stay constant with increasing SiO₂ contents (fig. 4). The two shoshonitic samples have Rb contents falling within the range of calc-alkaline rocks with a relatively high K/Rb ratio.

Sr shows variable values. Within the calc-alkaline group it ranges from 225 ppm to 605 ppm and stays constant with increasing SiO₂ contents (fig. 5). The high-K calc-alkaline samples show similar Sr values which increase with increasing SiO₂ contents, although the pattern is strongly influenced by the high Sr contents of R 100. A similar trend has been found in the high-K calc-alkaline rocks from Papua (Jakeš and Smith, 1970). Sr shows very high values in the two shoshonitic samples.

The Rb/Sr ratio is variable and does not show any particular behaviour through the series (fig. 6).

As regards the relation between the K, Rb, Sr, K/Rb ratio and the phenocryst mineralogy of the analyzed samples it can be noted that, apart from a few anomalous values (e.g. Sr content of R 100 = 1450 ppm) the pyroxene-bearing andesites and the hornblende-bearing andesites have a large overlap of values. This could indicate that the phenocryst mineralogy does not affect the overall element abundances in the rocks and, therefore, that the bulk distribution especially of K and Rb is conditioned by the groundmass. The only clear relation is found for biotite-andesites with high K contents.

COMPARISON WITH OTHER CALC-ALKALINE SERIES

Bleahu *et al.* (1973) suggested that the Carpathian arc is a continental arc characterized by a series of structural elements, i.e. trench, folded arc, magmatic arc, retroarc basin, which are typically associated in many modern island arc systems.

Lexa and Konečný (1974) have pointed out that a number of differences both of petrological and of tectonic type do exist between Carpathians and active island arcs. These differences were interpreted as evidence against a close direct relationship between magmatism and subduction zone. In this context we believe that it could be of interest to compare the geochemical data of this work with that found in other calc-alkaline series coming from active magmatic chains.

In Table II, K, Rb and Sr values of the samples which plot in the field of calc-alkaline series of fig. 2, are compared with data of calc-alkaline rocks coming from active island arcs and continental margins. In order to make a comparison between groups of rocks with the same K₂O and SiO₂ range

TABLE II
Comparison of Rb, Sr, K/Rb and Rb/Sr values of Calimani-Harghita volcanics with those of other calc-alkaline series

Zone	SiO ₂ (%)		K ₂ O (%)		Rb (ppm)		Sr (ppm)		K/Rb		Rb/Sr	
	av.	range	av.	range	av.	range	av.	range	av.	range	av.	range
1) Indonesia	54.5	48.5-60.0	1.23	0.48-1.86	31	8-70	402	217-585	367	211-672	0.083	0.020-0.322
2) New Zealand . . .		basalt-andesite	1.25	0.32-2.34	40	4-87	271	144-508	325	214-649	0.17	0.01-0.5
3) Aeolian arc . . .	54.7	50.5-65.8	1.4	0.8-2.7	42	30-88	442	324-690	295	223-350	0.06	0.03-0.15
4) Bougainville (Fiji).	57.8	54.0-62.7	1.58	1.25-1.95	25	16-35	683	445-810	525	457-657	0.036	0.024-0.05
5) North Chile . . .	57.6	52.5-61.5	1.70	1.1-2.0	42	20-80	743	580-1500	397	207-747	0.145	0.027-0.386
6) Basalt (average) .					10		330		340			
7) Andesite (average)	59.5		1.60		31	19-44	385	215-570	430		0.08	
8) Calimani-Harghita	56.6	52.3-62.2	1.37	0.71-1.93	46	20-76	355	240-605	255	197-343	0.134	0.075-0.237

Data from: 1) Whitford (1975); 2) Ewart and Stipp (1968); 3) Keller (1974); 4) Taylor et al. (1969); 5) Siegers et al. (1969), Picher and Zeil (1972); 6) Jakes and White (1972); 7) Taylor (1968); 8) Present work.

the data taken from the literature were first plotted on a diagram like that of fig. 2 and only the samples with a K_2O % and SiO_2 % falling inside field II were considered for the computation of ranges and averages of Sr, Rb, K/Rb and Rb/Sr.

Compared with the average andesite of Taylor (1968) and average island arc calc-alkaline values of Jakeš and White (1972) the Rb contents of the Calimani-Harghita volcanics are slightly higher whereas Sr is about the same. The Andean andesites (Siegers *et al.*, 1969) have higher Sr and K contents in respect to our samples, but a similar Rb concentration. Higher Sr is shown also by Bougainville andesites (Taylor *et al.*, 1969) which have lower Rb in respect to Calimani-Harghita andesites. Indonesian (Whitford, 1975), Aeolian (Keller, 1974) and New Zealand (Ewart and Stipp, 1969) volcanics all show slight differences in Rb and Sr contents from the Calimani-Harghita andesites. As regards the K/Rb ratios it can be noted that the analyzed samples show lower mean values in respect to both island arc and Andean andesites, and this appears to be the only noteworthy feature of these rocks. In fact, as regards the absolute abundance of these elements, it falls well within the range of many other calc-alkaline series.

DISCUSSION

Data on SiO_2 and K_2O of the analyzed samples show that almost all the volcanic rocks cropping out in the Calimani-Harghita Mts. are calc-alkaline, but a few shoshonitic rocks occur too. Calc-alkaline volcanics have intermediate SiO_2 contents and only a few basic and acidic members occur, as found in many active island arcs. Fig. 2 shows that some andesites and basaltic andesites have a higher K_2O content and fall within the compositional field of the high-K calc-alkaline series. However, it has to be pointed out that this does not mean that they represent a separate group of rocks, since the boundaries on fig. 2 are not rigid and do not necessarily represent divisions between different rocks series.

The overall Sr and Rb contents fall within the range of values of other calc-alkaline series with Rb showing slightly higher values in respect to average values of intermediate calc-alkaline rocks published by some authors (Taylor, 1968; Jakeš and White, 1972). The only significant difference shown by the Carpathian rocks in respect to other calc-alkaline series is given by the lower K/Rb ratio which is closer to shoshonitic rather than to calc-alkaline values (Jakeš and White, 1970). This peculiarity of the Eastern Carpathian rocks can be explained in several ways. A possibility is that suggested by Armstrong (1971) who believes that a key role in the calc-alkaline magma genesis is played by the oceanic sediments dragged down along the Benioff zone and involved in the melting. This can explain the low K/Rb ratio in the analyzed rocks since the sediments generally have a low K/Rb ratio (Heier and Adams, 1963). But if we assume that the andesitic magma source region

is the mantle wedge overlying the Benioff zone, the process of involvement of sediments in the melting has to be ruled out. In this case the K and Rb sources have to be found in the mantle. Oxburg (1964) suggested that K and Rb in the mantle are concentrated in some minor phases as amphibole and phlogopite. Only the latter, however, has a low K/Rb ratio and its degree of involvement in the melting has a great effect on the K/Rb ratio of the magma (Beswick, 1976). In this context, the lower K/Rb ratio of the Carpathian andesites can be explained by a high degree of involvement of phlogopite in the partial melting which gave rise to the calc-alkaline magma.

Shoshonitic rocks R 105 and R 107 represent a different magma type, as shown by Peccerillo and Taylor (1976 a) on the basis of the REE distribution pattern. They are characterized by high K and Sr contents but a relatively low Rb and high K/Rb ratio compared with other shoshonitic rocks (Jakeš and White, 1970). This character can be explained, on the basis of the data at present available by assuming a magma source region with a high K/Rb ratio. This region could be the lower crust or the undergoing oceanic crust, both characterized by a high K/Rb ratio (Sighinolfi, 1969; Jakeš and White, 1970). Other processes such as accumulation of K-feldspars with a high K/Rb ratio (in order to explain also the high Sr contents of these shoshonites) have to be excluded since no positive Eu anomaly was found in the REE pattern of R 107 by Peccerillo and Taylor (1976 a).

CONCLUSIONS

The volcanic rocks cropping out in the Calimani-Harghita Mts. are characterized by the occurrence of predominant andesites with few dacites and sporadic rhyolites, as well as found in typical active island arcs. Small amounts of shoshonitic rocks occur too. The potassium content stays constant through the two magmatic stages except at the end of the volcanism when the shoshonitic rocks were emplaced.

The calc-alkaline rocks under study have K, Rb and Sr contents close to the average values of calc-alkaline rocks but show a significantly lower K/Rb ratio which could be explained by processes of involvement of sediments in the magma genesis (Armstrong, 1971) or by a high degree of involvement in the partial melting of phlogopite (Beswick, 1976). Shoshonites have high K and Sr contents and relatively low Rb with a high K/Rb ratio.

In short, these data together with that previously published by several authors (Peltz *et al.*, 1973 a, b; Boccaletti *et al.*, 1973; Peccerillo and Taylor, 1976 a) indicate that the calc-alkaline rocks from the Calimani-Harghita Mts. show many more similarities than differences in respect to other calc-alkaline series especially those coming from island arcs. This can be considered as evidence that the peculiar tectonic setting of the Carpathians has not played an important role in the processes of magma genesis which can be considered the same as in island arcs.

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