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VULCANISMUL NEOGEN AL LANȚULUI MUNTOS CĂLIMANI-GURGHIU-HARGHITA

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CONTENT

POSITION OF THE CĂLIMANI–GURGHIU–HARGHITA AREA WITHIN THE NEOZOIC VOLCANIC ZONE OF THE ALPINE REGIONS

BY

DAN P. RĂDULESCU[1]

The northern and eastern border of the Pannonian Basin – with its eastern extension the Transylvanian Basin – was during the Neogene time the scene where ample volcanic phenomena have occurred (to a much lesser extent they have also taken place either in other regions of this basin or in previous moments). The strain developed in these contact regions between the rigid block from the basement of the basin and the folded or in the course of folding parts of the alpine orogene – which closely joined with the rigid block, – had determined the occurrence of a large zone of weakness along which magmas have migrated toward the surface. The volcanism has developed either on the continent or frequently at the periphery of the basin, where it generated several islands, which later attached themselves to land.

The Călimani-Gurghiu-Harghita Mountain Chain represents the south-eastern outermost part of this large volcanic region, which practically extends, without any interruption, up to north of Budapest, through the sub-Carpathian Ukraine, Czechoslovakia and Hungary. Although this chain belongs to a clearly outlined petrological province, its various parts display numerous distinctive features; most of them derive from differences relating to the start moment and the duration of the period of the volcanic activity.

Age. Nowadays the concept on the migration of the Neozoic volcanism broadly from west and north towards east and south-east (K u t h a n, 1948; R ă d u l e s c u, in print) is sufficiently argued. From

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this point of view it may be observed that the Călimani-Gurghiu-Harghita Mountain Chain comprises besides older products the most recent ones and the best preserved volcanic structures; these two features cannot be compared to those in other districts of the volcanic region.

If our information about the upper limit of the volcanic period is somewhat more ample, numerous most essential elements concerning the beginning of the activity are as yet but incompletly known. At present it is out of question to establish with precision the relationships between the volcanism of the Călimani-Gurghiu-Harghita area and the tuff horizons both of the Pannonian Basin and the Dacic—Euxinic Basin; this accounts for the fact that the researches were also directed towards the sedimentary deposits from adjacent areas. The study carried out by Popescu on the Pannonian deposits along the eastern margin of the Transylvania Depression, in the close neighbourhood of the mountain range, has proved that volcanic pyro- or epiclastic elements are occuring in sedimentary rocks hardly in the terminal part of the Pannonian s.s. (Meotian) [2]. It is of interest to point out that the lack of epiclastic volcanic particles in the rest of Pannonian deposits excludes the alternative implying that the corresponding time should represent a calm interval between the periods of volcanic activity. The existence of volcano-sedimentary deposits, which form a lower compartment below the one built up of the nowadays conspicuous volcanic super-structures, proves that the erosion of some volcanic formations when it occurred was immediately recorded in the sediments of neighbouring areas.

However, if we would admit that an older volcanism had also existed, it is to be situated much lower on the stratigraphic scale, at least, before the Sarmatian in order to explain the total disappearance of the volcanogenous material until the beginning of the Pannonian; as we have to deal with a small-sized basin we would rather expect, a preservation of such a material in sediments during a longer time interval, than its rapid disappearance. A volcanism so old would not represent, however, but another „stage" of the one under discussion; it would be something different, and its correlation with the already identified volcanism, and that from the neighbouring areas, is to be examined under other angles.

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[2] A. Popescu (1966) Studiul mineralelor grele din depozitele pannoniene situate între valea Mureșului și valea Gurghiului. Manuscript. Arh. Inst. Geol. Likewise subsequent manuscripts.

The continuance of the volcanic activity until very recent moments was suggested by numerous researchers relying on the good preservation of volcanic apparata (Rădulescu et al., 1964), as well as on the identification, either in drillings or at the surface of some horizons of volcanic material intercalated in Quaternary deposits (Pricăjan, 1961; Liteanu, Mihăilă, Bandrabur, 1962; Ghenea, 1967); these observations have determined some scientists to accept the continuance of volcanism up to, at least, the Middle Pleistocene (Liteanu, Ghenea, 1966; Peltz 1971).

The absolute age determinations carried out these last years (Rădulescu, Pătraşcu, Bellon, 1972) allowed to make some essential specifications, without exhausting this problem (Rădulescu, in print). The age of 3.92×10^6 years for rocks, which closed the volcanic activity in the largest part of the Harghita Mts, renders mostly improbable if not completely excluded a much younger volcanic activity. On the other hand, the ages of 7.08 and 7.37×10^6 years, for older rocks, also pertaining to the upper compartment of the volcanic structure, are in perfect agreement with the beginning of the volcanic activity in the terminal part of the Pannonian (= basis of the Pliocene at about 12×10^6) years.

Another fundamental concept documented by absolute ages is that the migration of volcanism may be recognized even within the Călimani-Gurghiu-Harghita area, where both the beginning and the closing of volcanic phenomena have succeded from north towards south (Rădulescu in print).

The conclusion which has therefore compelled the recognition is as follows: the whole volcanic activity from the Călimani-Gurghiu-Harghita area, whose products are today conspicuous, is very young and represents the terminal part of the subsequent alpine magmatic activity.

Released material. Both the late moment of starting of volcanism and its shorter displaying period had profound repercussion as regards the nature of the material having been expelled. By contrast with all other areas of the alpine region showing subsequent Neozoic volcanism — wherein the petrographical and chemical variety displays in most cases a wide range — within the Călimani-Gurghiu-Harghita area only andesitic magmas closely related as to their chemical features, were released. The totality of products has a pronounced basic character, preserved during the whole activity; the quartz-bearing forms of andesites and the dacites are recognized only incidentally and their spreading is very limited.

As it may be observed the normal and complete development of processes during the subsequent volcanism — alternation of andesitic and rhyolitic + dacitic stages — cannot be recognized in this area. As regards some areas, e.g. the Apuseni Mountains or the Oaş-Gutăi area, the difference consists only in the lack of this alternation, fact which would eventually allow to presume that in the Călimani-Gurghiu-Harghita area it is only the last andesitic phase that is represented (the difficulties as to accept such a hypothesis have been, however, mentioned); with respect to other areas, such as central Slovakia, the difference consists in the nature itself of the last phase, which is there a rhyolitic one (Forgac et al., 1968).

This remarkable petrographical and chemical uniformity of the volcanic material may have been mostly originated by the type of the differentiation magmatic processes maybe due to their short time of development, which did not essentially affect, the primary features of magmas; nevertheless the homogeneity of magmas, correlated with the immense bulks of rocks they have built up, does not fail to suggest their deep-seated origin.

Volcanic activity. The chemical nature of magmas has determined the general type of the volcanic activity. As compared to other areas, the clear-cut separation in time of the dominantly explosive activity from that with a dominantly effusive character, proves to be specific here; the last part of the activity (upper compartment) is practically lacking on pyroclastics, whereas in the first part (lower compartment) and in the sedimentary deposits of corresponding age from adjacent regions, they are very well represented. Although the type of the mixed activity and that of the stratovolcanic structure is still characteristic of the Călimani-Gurghiu-Harghita area, nevertheless it marks a rather obvious individualization as compared to other areas, especially for the last moment of the volcanism from this area, the upper compartment.

The very rapid rhythm of the development of phenomena is likewise characteristic; the building up of the volcanic structures and their destruction during the first stage, as well as the building up of volcanic edifices during the second stage of activity corresponds to relatively short time intervals. From this viewpoint both stages of volcanic activity over the Călimani-Gurghiu-Harghita area represent, in comparison with other areas, only two isolated although very strong paroxysmal manifestations.

In the light of the so far expounded ideas relating to the nature of the released material, as well as to the development of the volcanic activity, the hypothesis about the relations between the volcanism of the Călimani-Gurghiu-Harghita area and the formation of the tuff horizons from neighbouring regions, appears more clearly. As previousley noticed, this problem cannot be rised, but only beginning with the Upper Pannonian s.s. deposits. Or, very significant is the fact that by contrast with the frequency and thickness of the tuffs in the pre-Pannonian deposits, in deposits Pannonian in age the tuffs occur but exceptionally always bearing an andesitic character and of reduced thickness, and only in the outer part of the Carpathian Arc; more frequently the pyroclastic material occurs as associated with the epiclastic one in hybrid rocks. This situation is quite corresponding with the features of volcanism so as they were sketched for the Călimani-Gurghiu-Harghita area: namely exclusively andesitic, and consisting of two distinct moments of activity, the first predominantly explosive, and the second predominantly effusive.

Basement. The basement of the Călimani-Gurghiu-Harghita area is characterized by the existence, at rather small depths, of metamorphic rocks. Excepting the southern outermost part — where the presence in depth of Mesozoic deposits showing a large thickness and pertaining to the folded structure of the Carpathians — within the whole area, the metamorphic massif is overlain only by Neogene deposits, and to a lesser extent, by the Paleogene ones; their thickness is variable east-westwards occasionally reaching 1500 m. This situation is, if excepting some isolated points from the Apuseni Mountains and from the area with subvolcanic structures of the East Carpathians, absolutely uncommon within the region showing a subsequent Neozoic volcanism, and its repercussion on the development of phenomena must not be overlooked. It is most probable, for instance, that the purity of andesitic magmas and the homogeneity of their features could be partly at least explained by the insignificant thickness of the sedimentary deposits pierced during their ascent; on the other hand, the same argument points out, as mostly presumable, the fact that the volcanic products forming the southern outermost part of the Harghita Mts area should have been submitted to influences due to the sedimentary basement they pierced. Differences of this nature which have been discerned between the volcanic rocks from the southern part of this area and the rocks from the median and the northern ones have confirmed this hypothesis (Rădulescu, Dimitriu, this volume).

From the structural point of view it is noteworthy that the area, where the volcanic activity took place tallies partially with a presumable deep-seated dislocation line, along which basaltic magmas have subsequently achieved the piercements of the basement (Rădulescu, 1962). This fact has certainly contributed to the ,,fixation'' of volcanism within a very narrow surface of a linear character. In most other areas the position of volcanic edifices seems to have been controlled by complex systems of directional and transversal dislocations that make the distribution of volcanic apparata rather complicated and variable. Within the Călimani-Gurghiu-Harghita area the distribution of volcanic edifices is almost perfectly linear. Obviously we are not dealing with a single dislocation but probably with several dislocations disposed ,,en coulisse'', hence both the parallelism and the very reduced distance between them contribute to give a general impression of ,,liniarity'' over the whole zone. Although this aspect is determined by the distribution of apparata from the upper compartment of the structure, it is probable that also the previous edifices corresponding to the lower compartment — volcano-sedimentary formation — had a similar position since none indications related to the presence of some roots of volcanoes outside the axial zone from this area do exist.

Age, released material, type of activity, basement traversed — there are the chief factors owing to which the Călimani-Gurghiu-Harghita area marks its individualization as compared to other areas with subsequent Neozoic volcanism, along the Alpine-Carpathian Range; they have determined the distinct petrographic, structural, paleogeographic features, permitting to consider the Călimani-Gurghiu-Harghita Mountain Chain as a perfectly outlined geological-volcanic unit in this province.

The succint analysis carried out here elucidates, however, the approach of the very frequently discussed problem referring to the correlation of areas with subsequent volcanism over the territory of Romania or over the whole Alpine-Carpathian territory. In our opinion within the Călimani-Gurghiu-Harghita area there exist distinctive features even in essential characters of volcanic and geological phenomena, which allow correlations only at a general level. In spite of the desiderata and attempts of the author himself to find terms which would correspond as to time, volcanic activity type, petrographical, chemical, metallogenetic features for various areas, we had to reach the conclusion that for the Călimani-Gurghiu-Harghita area its appurtenance to the province of the subsequent Neozoic volcanism is solely reflected in its lessentia features upon which numerous peculiar elements have grafted.

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LOWER COMPARTMENT OF THE STRUCTURE OF THE CĂLIMANI, GURGHIU AND HARGHITA MOUNTAINS : THE VOLCANO-SEDIMENTARY FORMATION

BY

DAN P. RĂDULESCU.[1], SERGIU PELTZ[2], ANTON POPESCU[2]

The basal part of the volcanic structure from the Călimani, Gurghiu and Harghita Mountain Chain represents an independent compartment — a very complex volcano-sedimentary formation — which crops out particularly in the peripheral zones of the region (Rădulescu et al. 1964) ; in proportion of about 45 per cent this compartment is overlain by the upper compartment of the structure — volcanic apparata with lavas and, subordinately, pyroclastics. The maximum extension east-westwards of the lower compartment is reached in the central and southern parts of the Gurghiu Mts (about 40 km). Its thickness of order of hundred metres, is variable and ranges most frequently from 300 to 500 m.

The basement overlain by the lower compartment is built up of sedimentary deposits, Tortonian, Buglowian (uncertain) Sarmatian and Pannonian s.s. in age. Among these deposits only the Sarmatian, and especially the Pannonian ones, are of interest as to their extension and possibilities as to have supplied clastic material for the volcano-sedimentary formation.

The Sarmatian has developed in the molasse facies and is slightly fossiliferous. It consists of a conglomeratic horizon (lower) and a marly horizon (upper).

The Pannonian occurs transgressively and unconformably over Sarmatian deposits, and is characterized by its lithological uniformity :

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grey marly clays within which sand or rarely gravel intercalations more or less consolidated do occur. Broadly three horizons —namely the lower clayey horizon, the median sandy horizon and the upper clayey horizon— whose cartographical separation presents some difficulties may be recognized; their thickness reaches approximately 1500 m. The most extended is the upper clayey horizon both as to its area and its thickness in the stratigraphical column (about 1000 m). In this prevailingly clayey packet frequent intercalations of grey fine sands, sandy clays, as well as coal seams of infinitesimal thickness are encountered. The sandy horizon is well developed on the western slope of the Gurghiu Mts where it reaches a thickness of 250—500 m. Southwards, on the western slope of the Harghita, the sandy and upper clayey horizons are replaced by a conglomeratic horizon of about 700 m thick with frequent, however, obviously subordinated intercalations of clays, sands and sandstones.

The Volcano-Sedimentary Formation

Terrigenous and volcanic rocks differing as regards their grain size, facies, petrographic type take part in the building up of the volcano-sedimentary formation. The andesite material builds up the background of the formation, the participation of the nonvolcanic one ranging commonly from 5 to 10 per cent.

The petrographical components of the volcano-sedimentary formation fall into three basal categories : non-volcanic, volcanic and hybrid ; neither the presence of the three categories of rocks, nor the presence of all petrographic types from each category are absolutely necessary ; the volcano-sedimentary formations display various aspects according to the association mode of these components.

In the figure 1 an attempt to systematize these possibilities on purpose to achieve a most exact description and definition of various situations is presented. The conventional notation of petrographic components leads to synthetic descriptive formulae which contain qualitatively the specific features of the formations.

The so far carried out researches have evidenced the fact that within various regions of the Călimani-Gurghiu and Harghita Mts, the petrographical components of the formations develop in a differential mode, without, however, to give rise to essential differences between the three parts of this chain. Our present knowledge of the volcano-sedimentary formation allows to distinguish within it three sequences with regional extension. The composition of the formation is schematically rendered in the lithological columns of the figure 2, particularly according to the situation in the western part of the eruptive chain, where the most frequent outcrops are encountered. The petrographical classification and nomenclature used here are presented in the table.

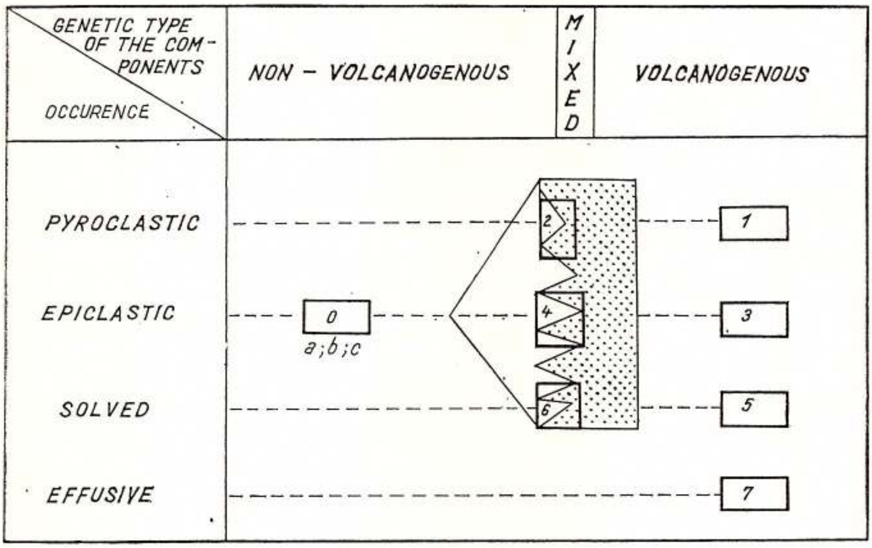

Fig. 1. — Constituents of the volcano-sedimentary formation from the Călimani, Gurghiu and Harghita Mts.
a, b, c mark successively the coarse, median and fine grain-size.

TABLE

Terminology utilized for petrographic types

Grain-size mm		Non-volcanic	Hybrid	Volcanic	
				Pyroclastic	Epiclastic
Over 100		Conglomerate	Breccia, hybrid conglomerate	Pyroclastic breccia	Volcanic conglomerate
32–100		Microconglomerate	Microbreccia, hybrid microconglomerate	Pyroclastic microbreccia	Volcanic microconglomerate
0.1–32	4–32	Sandstone	Hybrid sandstone, tuffite	Lapilli tuff	Volcanic sandstone
	1–4			Coarse ash tuff	
	0.1–1			Fine ash tuff	
Under 0.1		Pelite		Dust tuff	Volcanic pelite

The *lower sequence* comprises products of the first volcanic manifestation from the region, and epiclastic deposits wherein volcanic components [3] appeared for the first time. Its thickness ranges from 30 to 100 m. The deposits are conspicuously exposed at the periphery of the volcanic zone, especially in the neighbourhood of Sovata, Ocna, Păuleni, Deda, Răstolița and in the Gurghiu Valley, where the relationships between the underlying sedimentary deposits, and those with which the volcano-sedimentary formation begins, could be minutely established.

Along the western border of the volcanic chain, the transition from the clayey-detrital Pannonian deposits, completely devoid of volcanic components (C/D Zone) to the volcano-sedimentary ones was progressively achieved, through a reddish sandy packet, guide horizon, at whose level the first indices of the Neogene volcanic activity from this sector of the East Carpathians were identified. Extending over a length which exceeds 150 km ,,the red sand horizon'' is composed of yellow-reddish fine-grained (median diameter 0.12–15 mm) sands, whose thickness varies between 5 and 25 m. In the heavy fraction of these sands, representing less than 1 per cent of the whole arenaceous material, the amounts of volcanic minerals, mainly hypersthene and green hornblende, are ranging from 0.5 to 45 per cent. Their quantitative and qualitative variation is presumably reflecting the existence of several volcanic edifices, which have supplied with ash material. The participation of the volcanic component is usually discernible only by means of a microscopical examination; it is represented not only by heavy fraction particles but also by iron from the hydroxides which give its colour to the deposit. Alternations of epiclastic and pyroclastic deposits are less frequently encountered (Hodac, along the Gurghiu Valley).

Due to the faunal assemblage existing at the level of red sands (*Congeria subglobosa* Partsch and Unionides with a robust shell) these deposits are referable to the basal part of the E Zone of the Pannonian, thus the moment marking the beginning of the volcanic activity in the Călimani-Gurghiu-Harghita Chain corresponds to the transition from the C/D Zone (clayey-detrital Pannonian, fauna with *Congeria banatica* R. Hoern.) to the E Zone (volcano-sedimentary Pannonian, fauna with *Congeria subglobosa* Partsch).

[3] A. Popescu (1966). Studiul mineralelor grele din depozitele pannoniene situate între valea Mureșului și valea Gurghiului. Manuscript Arh. Inst. Geol.

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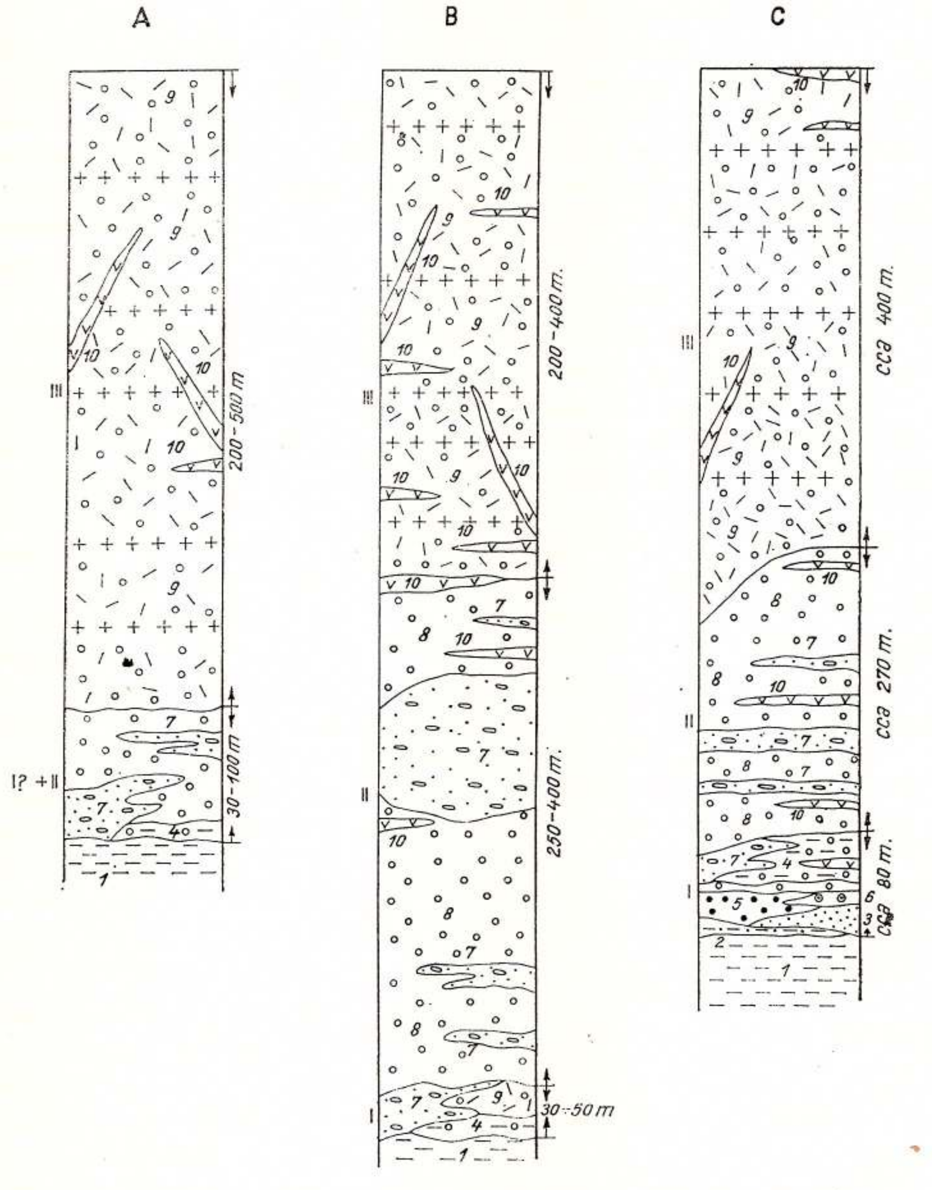

Fig. 2. — Synthetical lithological columns in the volcano-sedimentary formation.

A. North-western part of the Călimani Mts (Peltz, 1965, unpublished data). *B*. Mureş Defile Zone (Peltz and Peltz, 1963, unpublished data; Rădulescu et al., 1964; Peltz, 1965, 1969). *C*. South-western part of the Gurghiu Mts (Peltz and Peltz, 1970).

I lower sequence; II intermediary sequence; III upper sequence.

1, marls, sandy marls, clays; 2, hybrid agglomerates; 3, epiclastics, 4, 5, 6, 9, 10, pyroclastic breccia, pyroclastic microbreccia, tuffs; 7, basalt andesite; 8, andesite.

The red sands are often overlain by grey compact slightly marly clays, devoid of arenaceous intercalations, 4—12 m thick, wherein the content of volcanic minerals is, likewise considerably increased. The development of these clays proves to be discontinuous, frequently reduced to the order of decimetres, or even centimetres at the uppermost part of the red sands. In these upper clays scarce and weakly preserved forms, pertaining to the fauna with *Congeria banatica* R. Hoern. ascertain the persistence of this form up to the level of the fauna with *Congeria subglobosa* Partsch.

Deposits which are directly overlying the red sands or the clays are slightly different from one point to another. Usually there is a pyroclastic and epiclastics alternation with fragments of 4—12 cm in diameter, accumulated under subaquatic conditions. The non-volcanic material is subordinate; the andesitic epiclastics amount, however, about 10 per cent even in rocks with aspect of volcanic breccia. From the point of view of grain size and shape of the fragments, the rocks are homogeneous; they frequently present a bedding, and an advanced alteration degree of the tuffaceous binding. In other cases epiclastic horizons are absent the material of this nature occurring subordinately in hybrid rocks.

An alternation of microconglomerates, breccias, pyroclastic breccias and microbreccias, tuffs, hybrid conglomerates and sandstones, tuffites is deposited in continuity of sedimentation. Within the hybrid rocks the predominance of volcanic minerals is evident, however, contents of 5—10 per cent of metamorphic minerals (almandine, zoisite, clinozoisite, epidote, zircon) are also characteristic. Such deposits were generated under subaquatic conditions. At the terminal part of the sequence coarse pyroclastics with small-sized intercalations of tuffs or epiclastics are recognized.

From the point of view of their petrographic nature, volcanic fragments of the lower sequence are hornblende (green or brown) -andesites and different varieties of pyroxene- and hornblende-bearing andesites.

The main features of the lower sequence seem to be as follows: *a*) heterogeneous constitution determined by the association ofthe volcanic material (pyroclastic and epiclastic) with the non-volcanic one; *b*) wide developement of the subaquatic facies; *c*) strongly explosive nature of the volcanic activity supplying material; *d*) lack of lava flows and intrusive bodies. According to the codification presented in the figure 1, the variety of rocks that enters into the composition of the sequence, may be expressed as follows (the frame around some notations indicates a subaquatic sedimentation; when materials with different grain size do coexist, the dominant size class is noted at the numerator):

	2a	1a
	2b	1b
[0c]	4b	[$1\frac{a}{b}$]
	[$4\frac{b}{c}$]	
	[6b]	

The *intermediary sequence* presents a more complex lithology than the lower one. Lava flows and intrusive bodies, always of andesitic nature, are to be found together with pyroclastics and epiclastics; there are absent rocks built up exclusively of nonvolcanic components showing in general a very strictly localized distribution. The thickness of the sequence ranges — as it may be stated in the south-western part of the Gurghiu Mts and in the Mureş Defile—from 250 to 400 m.

In the north-western part of the Călimani Mts the intermediary sequence is not sufficiently individualized. The deposits which may be referred to it are pyroclastic breccias and microbreccias with elements of pyroxene-andesites and pyroxene-hornblende-bearing andesites, comprising intercalations with epiclastics showing the same lithological composition.

In the Mureş Defile Zone (southern part of the Călimani Mts and the northern one of the Gurghiu Mts), the intermediary sequence is very well developed (fig. 2, B). In the lower third of the sequence, breccias and pyroclastic microbreccias with intercalations of tuffs and epiclastics are encountered. Fragments of all deposits belong to pyroxene andesites, and subordinately to hornblende- pyroxene-bearing andesites. This complex contains flows of basaltic andesites. In this part of the sequence the subaquatic facies predominates too as in the lower sequence. The subordinate participation of lavas points out that the volcanic processes, contemporaneous with the building up of this part of the formation, have been predominantly explosive; the petrographic features of lavas indicate a subaerial volcanism.

An important moment in the lithogenesis of the intermediary sequence from this territory is marked by the activity of lacustrine basins, namely Andreneasa, Lunca Bradului, Neagra and Toplița located within inner depressions of the mountain chain. In these basins, deposits showing the same volcano-sedimentary features, have accumulated. The most characteristic development of these complexes may be observed in the central part of the Toplița Basin, where alternations of conglomerates, microconglomerates, sands, sandstones, and subordinately clays with coarse and fine pyroclastics, are encountered. The deposits are characterized by a normal rhythmic bedding or, occasionally a crossed one, the variety of grain size and mineralogical composition. In epiclastic deposits rests of ligneous vegetation or of the herbaceous one, that have undergone the incarbonization or opalization processes are found. The constituent fragments of deposits are hornblende-andesites, and subordinately pyroxene-hornblende-bearing andesites or pyroxene-andesites.

The deposits of these basins reflect locally a quiet period of the volcanic activity without, however, its complete ceasing.

In the synthetical column of the formation from the Mureș Defile Zone (fig. 2, B) the deposits of these basins are ,,symbolically'' represented by epiclastics since the latter constitute the type of the predominant rock. At the upper part of the sequence breccias and microbreccias, tuffs with intercalations of epiclastics and andesitic lavas do reappear.

In the southern part of the Gurghiu Mts, the basement of the intermediary sequence — consisting likewise of an alternation of pyroclastics and epiclastics with fragments of similar petrographic nature, comprising andesitic lavas — presents clear-cut features of subaquatic deposition; by contrast, the upper part was predominantly subaerially accumulated. Conglomerates and microconglomerates from the lower part of the sequence are composed here of boulders and well-rounded pebbles. In the constitution of rocks from the lower part of the sequence there participated 70 per cent of volcanic material — of the same nature as in the rest of the region — and 30 per cent of pebbles of black quartzites, white quartzites, gneisses etc. The matrix of conglomerates is porous, occasionally friable, grey-yellowish displaying a variable grain-size. Sandstones consisting exclusively of volcanic material, often with impressions of plants are added to coarse rocks. The pyroclastics are well represented both in the basal part and in the upper one of the sequence by rocks showing all types of grain-size.

The intermediary sequence may be characterized as follows: *a*) a subordinate and very limited participation of nonvolcanic components; *b*) an important participation of epiclastic deposits; *c*) a wide development of the subaquatic facies; *d*) the presence of andesitic lavas. Its composition may be expressed by the formulation:

1a
1b
| 1b |
| 4a | 3a
| 3a |
| 3b |
| 3c |
7

The *upper sequence* presents a lithology perfectly correlable over the whole territory of the volcanic chain. This situation is schematically illustrated in the synthetical lithological columns of the figure 2. Broadly, the upper sequence is composed of pyroclastic breccias and microbreccias and tuffs which alternate, and to which lavas and intrusive andesitic bodies are added. The constituent elements of the clastic rocks are hornblende-andesites, pyroxene-hornblende-andesites and basaltic andesites. Fragments occasionally reaching considerable sizes with angular and subangular shapes are encompassed within a grey compact or porous matrix either microbreccious, lapillic or tuffaceous. The thickness of the sequence reaches sometimes 300–400 m.

The chief features of the upper sequence are: *a*) relatively simple petrographic composition; *b*) the accumulation of material exclusively under subaerial conditions; *c*) rised frequence of lava flows and intrusive bodies; *d*) quite subordinate occurrence of epiclastic components. Its codified expression is:

1a
1b
3b
7

The present phase of knowledge relating to the volcano-sedimentary formation does not allow to determine with precision the upper limit of the terminal sequence.

It is quite possible that the pyroclastics which were included at the upper part of this sequence, would represent the beginning of the volcanic activity corresponding to the upper compartment. No conclusive observation, which would justify such an interpretation, has been so far reached.

Massive volcanics

The presence of massive volcanics within the volcano-sedimentary formation ascertains the continuation with a reduced intensity of intrusive and effusive processes (besides the explosive ones) during the whole period of its building; if for the intercalated lava flows, the simultaneity with the volcano-sedimentary formation is beyond any doubt, for intrusive bodies, however, the presumption of their appurtenance to the activity period, corresponding to the upper compartment, cannot be always excluded.

The thickness of the lava flows is of the order of metres, more seldom of tens of meters, and they crop out over restricted areas; only in the Călimani Mts such rocks are more developed, particularly innerside of the Călimani caldera, in the Poiana Negrei region etc. In many points of the mountain chain such lava sheets were identified by drillings.

The intrusive bodies are known only in the northern part of the volcanic area; they occur both within the volcano-sedimentary deposits and the subjacent sedimentary deposits. Their presence over extended areas in the Călimani Mts allowed some researchers to presume that they would pertain to large intrusive masses emplaced before the beginning of the volcanic processes and, therefore, have played the part of the basement for the products of volcanic processes. The groundlessness of this standpoint was proved by the demonstration of the structural connection between these bodies and the volcanic superstructures (Rădulescu, 1960).

Besides sills, dykes and small-sized necks, there were described laccoliths and domes displaying larger sizes. In many cases the rock presents a coarser granulation as compared to normal forms of volcanics, however, the maintenance of the nomenclature of the latter was preferred on purpose to emphasize their connection with volcanic products.

The dacites are recognized in some points of the northern part of the volcanic chain; they are obviously found at the basement of the whole volcanic structure, nevertheless they do not seem to be a component of the volcano-sedimentary formation, but rather to have a distinct still incompletely cleared up position. They form flows and sometimes probably, necks; fragments of this petrographic nature were not encountered in the volcano-sedimentary formation. The rocks are grey-yellowish with scarce femic components, which are represented by biotite-(Drăgoiasa) or green hornblende often total resorpted (Voivodeasa).

Hornblende–andesites are widespread over the whole area occupied by the lower compartment. They occur as lavas and intrusions and participate in the composition of varied types of clastic rocks.

From the point of view of their mineralogical composition the andesites comprise numerous petrographic types characterized by these participation in different proportions of hornblende (green or basaltic) sometimes resorpted, and of pyroxenes. The extreme types—hornblende-andesites, hornblende-pyroxene-andesites, basaltic hornblende-andesites, resorpted, hornblende-pyroxene-andesites. hornblende-pyroxenes-andesites—can be microscopically separated, but their spatial development can not be always mapped. The rocks are massive, seldom slightly porous, bearing plagioclase phenocrysts (28–47% An) and hornblende; the pyroxene crystals are always smaller. Within the groundmass large amounts of magnetite may be often noticed. The colour is grey with variable hues.

Pyroxenes–andesites represent the second main petrographic type, whose presence in the volcano-sedimentary formation is observed over the whole region. Augite and hypersthene-bearing rocks are probably the most frequent; dominantly or exclusively augite- or hypersthene-bearing rocks are added to the former. Their black colour renders them usually distinct from hornblende-bearing andesites. Plagioclase (32—52% An), augite. hypersthene, titanaugite, diopside and pigeonite constitute phenocrysts; twinning and various intergrowths were recognized in the pyroxene crystals. The groundmass often contains besides microlites, essential amounts of glass.

Basaltic andesites display a limited distribution exclusively in the southern part of the Călimani Mts, and the northern one of the Gurghiu Mts, where they form small-sized bodies. The black colour and the conchoidal fracture may occasionally represent criteria of separation with respect to pyroxenes andesites.

The presence of basic plagioclases (45–65 An), and frequently of olivine is to be noted; pyroxenes are of course frequently encountered.

The distribution of massive volcanics within the sequences of the volcano-sedimentary formation presents, as previously noticed, some distinctive features; the fact that they have in general only a geometricalstructural significance, and only in the case of lavas an implication in the chronology of phenomena, is to be emphasized.

Recently the Pleistocene age was assigned to the volcano-sedimentary formation from the southern outermost part of the Harghita Mts (Peltz, 1971), and on this basis the above formation was separated from similar deposits within the Călimani-Gurghiu-Harghita Chain. Although this opinion tallies with the conception on the migration of volcanism north-southwards (Rădulescu, in print), nevertheless it is not consistent with the absolute age determined for hypersthene-augite-andesite, from the central part of the Harghita Mts ($3.92 \pm 0.2 \times 10^6$ years, ibid); although the age of deposits is probably variable north-southwards, it is obvious that from the structural point of view we have to deal with the same compartment in the whole region.

The lower structural compartment of the Călimani-Gurghiu-Harghita area has formed as a result of the erosion of certain previously existing volcanic structures, under the conditions of persistence of a reduced explosive and effusive activity. The deposits represent the product of rapid accumulation processes in the time interval between two paroxysmal volcanic periods (Rădulescu, this volume).

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GEOCHEMISTRY OF U, Th, K IN VOLCANIC ROCKS FROM THE CĂLIMANI–GURGHIU–HARGHITA AND PERŞANI MOUNTAINS

BY

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1. Introduction

These last years the Călimani-Gurghiu-Harghita area was the object of a geological-volcanological and petrological research work. These studies having in view the deciphering of the geological structure, the clearing up of the mode according to which the eruptive activity has developed and its age, the knowledge of petrographical features of volcanics have been followed by petrochemical and geochemical studies. Thus, an advanced degree in the knowledge of the important structural unit of the East Carpathians could have been reached.

The purpose of this paper is to achieve an elaborate and minute study on the distribution of trace elements in volcanic rocks. It is for the first time that U, Th and K distribution in neovolcanics from East Carpathians was investigated.

Considering the geochemical peculiarities of elements under discussion there are followed to the same extent, the involvements of a scientifical and practical nature and namely : U, Th and K contents in volcanics ; evidencing of petrotypes or volcanic complexes with anomalous contents ; U, Th and K distribution according to eruption phases and structural compartments ; U, Th and K variation in the magmatic differentiation processes ; U, Th, and K contents in rocks affected by secondary transformations.

Analyses carried out for U, Th and K completed the geochemical data relating to volcanics from the Călimani-Harghita area as well as

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the basalts from the Perşani Mts. Thus the collecting of samples for the above study was carried out, as far as possible, from the same sectors from which there have been previously taken samples for the studying of trace elements Pb, Cu, Zn, Cr, Co, Ni, V, Ga, Ba, Sr, (Peltz et al., 1971; Peltz et al., 1973), fact that will allow the further interpretation of some problems of a general geochemical interest.

Related to the area of about 5000 sq. km of the volcanic zone under investigation, the bulk of data may be considered as informative. It corresponds, nevertheless to the purpose of this study which aims at the obtaining of the first information as regards the U, Th and K geochemistry in the East Carpathians neovolcanics.

2. Analytical results

2.1. Working method. The field collected samples of the weight ranging from 1 to 3 kg were grinded at a grain-size of 1–3 mm by means of the grinding mill. After the homogenization approximately 700 g have been put in plastic boxes.

The U, Th, and K determination was carried out by gamma-ray using the pulse height analysers of 400 and 800 channels Intertechnique type. The scintillation detector was equipped with a NaI (TI) crystal with sizes of 40–50 mm.

For determining the radioactive elements the following energetic values were used : uranium – 350 KeV ; thorium –238 KeV ; potassium 1470 KeV. The width of a channel was of 10 KeV.

On purpose to compute the concentrations of these elements from the samples there were prepared standards by SiO_2 mixing with uranium and thorium ores, their dosage being chemically carried out. There was used potassium carbonate for potassium determination wherein the potassium was again determined by flamme photometry.

The establishment of the error in the concentration determinations was made by remeasurement of 14 samples randomly selected. The error obtained does not exced $\pm$ 10 per cent, for the three elements.

The detection limits for radioactive elements are : U – 0.2 ppm ; Th – 0.2 ppm ; K – 0.1 %, for a measurement time of 60 minutes.

2.2. Location of the samples. The study of U, Th and K distribution in the volcanics from the Călimani, Gurghiu, Harghita and Perşani Mts was made on the basis of a number of 369 samples. Their belonging to the volcanic massifs and petrographical types are presented in table 1.

TABLE 1

Appurtenance of analysed rocks to volcanic massifs and petrographical types

	Basalts	Basaltic andesites	Pyroxene-andesites	Andesites with pyroxene and hornblende	Hornblende-andesites	Biotite-andesites	Dacites	Diorites and microdiorites	Volcano-clastics	Rocks affected by secondary transformations
Călimani 118	—	16	35	13	7	—	12	5	18	10
Gurghiu 97	—	1	17	12	10	—	—	—	21	70
Harghita 81	—	—	10	8	6	10	—	—	4	42
Perşani 42	33	—	—	—	—	—	—	—	6	3
TOTAL 369	33	17	62	33	23	10	12	5	49	125

When examining the table 1, one may observe that all the types of rocks have been analysed, among which, the pyroxene-andesites, andesites with pyroxenes and hornblende and andesites with hornblende are better represented; they build up the most part of the volcanic area. Likewise the basaltic rocks (Perşani basalts, Călimani basaltic andesites) and dacites.

The products of both structural compartments, respectively of the chief stages from the development of volcanism are correspondently represented. This is resulting from the fact that from a number of 369 analysed samples, 185 pertain to volcanics from the upper compartment. Taking into account the large widespreding of volcanoclastics the number of analysed samples may be considered as informative.

From a totality of 369 samples, 125 represent rocks affected by secondary alterations. They proceed from areas of metallogenetical interest : Negoiul românesc, Stînceni, Şumuleu, Harghita-Băi, Sîntimbru-Băi.

2.3. U, Th, and K distribution in volcanics. Analytical data have been systemized according to volcanic massifs and petrographical types. Average values (table 2) have been computed for types with sufficient available data.

Considering the objects already traceable in previous study relating to the geochemistry of trace elements within the Călimani–Harghita Zone, the authors propose the following ones : 1) evidencing of anomalous contents and their explanation; 2) character of radioactivity distribution; characterize 3) U, Th, and K distribution in the course of the history of volcanism; to what extent these trace elements caracterize the volcanic structures; 4) behaviour of U, Th, K in differentiation processes; 5) radioactivity and secondary alterations; 6) comparative observations with rocks and volcanic sequences from other zones.

2.3.1. *Basalts*. Data we are examining are referable to olivine basalts from the Perşani Mts, as well as to their different varieties. They are likewise available for basaltic scoriae from the Heghieş Hill which represent the last products of the volcanism in this region.

The content of U is ranging from 0.8 to 3.1 p.p.m. for basalts and from 1 to 2.5 p.p.m. for scoriae. Higher average values may be noticed for the variety ,,cucuruz''. The content of Th for the varieties of compact basalt, ,,cucuruz'' and vacuolar ones reaches varied limits, their average being, however, almost the same (table 2).

TABLE 2

Average contents and variation range of radioactive elements in volcanics from the Călimani, Gurghiu, Harghita and Perşani Mts

	U. ppm.	Th. ppm.	K %	Th/U	n
1	2	3	4	5	6
		Basalts	(Perşani)		
Compact basalt	1.4 / 0.8–2.4	7.1 / 5.8–9.3	1.5 / 1.1–2.1	5.1 / 3.8–8.1	13
Cucuruz basalt	1.8 / 1.0–3.1	6.3 / 3.4–8.7	1.0 / 0.8–1.6	3.8 / 1.2–4.8	14
Vesicular basalt	1.3 / 0.8–2.5	6.6 / 5.8–17.4	1.2 / 0.6–1.6	5.6 / 5.0–8.2	6
Basaltic scoriae	1.5 / 1.0–2.5	6.7 / 6.2–7.2	1.0 / 8.8–1.2	4.0 / 2.5–6.8	6
Average basalts	1.6	7.8	1.2	4.9	33
		Basaltic andesites			
Călimani	1.9 / 1.2–3.2	6.9 / 3.4–10.1	1.5 / 1.0–2.2	3.4 / 2.4–5.0	16
		Pyroxene-andesites			
Călimani	2.2 / 0.5–5.5	7.1 / 2.3–17.3	1.5 / 0.3–2.8	3.2 / 1.4–6.8	35
Gurghiu	1.3 / 0.7–2.3	5.3 / 3.0–7.1	1.1 / 0.5–1.7	4.3 / 2.6–6.3	17
Harghita	2.3 / 1.3–3.1	9.0 / 6.1–10.5	2.0 / 1.5–2.8	4.0 / 2.7–5.1	10
Average	2.0	6.9	1.5	3.6	62
		Hydrothermalized pyroxene-andesites			
Sîntimbru	2.5 / 0.9–6.5	10.6 / 1.7–24.5	1.4 / 0.2–2.9	4.0 / 1.9–6.7	28

table 2

1	2	3	4	5	6
Harghita-băi	1.5 / 0.3–2.3	7.5 / 5.5–12.1	1.1 / 0.2–2.4	4.6 / 3.3–8.2	15
Average	2.1	9.5	1.3	4.2	4.3
		Pyroxene-hornblende-bearing andesites			
Călimani	2.0 / 0.6–4.9	5.6 / 2.6–14.4	1.4 / 0.6–4.0	3.2 / 1.6–7.4	13
Gurghiu	1.3 / 0.7–2.3	5.9 / 3.4–9.9	1.0 / 0.7–1.5	4.8 / 2.4–9.9	12
Harghita	2.0 / 1.3–2.8	8.5 / 5.0–10.4	1.5 / 0.7–1.8	4.4 / 3.7–5.9	8
Average	1.75	6.4	1.25	4.1	33
		Hornblende-andesites			
Călimani	1.1 / 0.6–1.6	3.7 / 2.0–8.7	1.0 / 0.6–2.0	3.6 / 2.5–4.8	7
Gurghiu	1.5 / 0.8–2.1	8.0 / 4.3–12.1	1.25 / 0.9–1.4	5.4 / 2.5–7.5	10
Harghita	2.1 / 1.5–2.8	8.8 / 6.6–10.7	1.7 / 1.6–1.9	4.3 / 3.5–5.0	6
Average	1.5	6.9	1.3	4.5	23
		Hydrothermalized hornblende-andesites			
Bătrîna	1.6 / 0.6–2.5	7.8 / 3.2–11.5	1.1 / 0.4–2.8	4.8 / 2.8–6.8	37
Şumuleu + + Seaca -Tătarca	1.3 / 0.1–2.0	5.4 / 4.0–7.3	1.1 / 0.3–2.6	4.0 / 2.0–5.3	14
Average	1.5	7.1	1.1	4.6	51

table 2

1	2	3	4	5	6
		Biotite-andesites			
Harghita	3.6 / 1.0—7.0	14.5 / 12.2—17.7	3.1 / 2.0—3.7	4.8 / 2.5—12.2	10
		Dacites			
Călimani	2.5 / 1.3—3.8	7.6 / 3.7—10.1	2.3 / 0.6—3.0	3.0 / 1.8—3.8	12
		Diorites, microdiorites			
Călimani	2.7 / 1.3—4.0	10.5 / 5.1—15.8	2.1 / 1.3—2.6	3.9 / 3.3—4.3	5
		Volcanoclastics			
Călimani	1.0 / 0.5—2.0	4.2 / 2.0—12.3	1.0 / 0.3—1.8	4.4 / 2.4—7.8	18
Gurghiu	1.1 / 0.5—1.7	5.5 / 3.0—9.8	1.0 / 0.6—1.8	5.1 / 2.1—7.0	21
Harghita	2.1 / 1.7—2.6	9.4 / 8.1—11.1	1.6 / 1.3—1.9	4.5 / 3.6—6.5	4
Average	1.5	5.3	1.05	4.75	43

As regards K the same variation of individual values and implicitly of content limits are noticed. By contrast with U and Th, the decrease of the K value from the compact basalt to scoriae is observed.

Anomalous values of U and Th have been dosed in the sample of the Bogata vacuolar basalt : 2.5 p.p.m. U, 17.4 p.p.m. Th. In comparison with these values, the fumarolized rock indicates an essential discrepancy of U and Th (0.7 p.p.m., respectively 1.8 p.p.m.). The lack of comparative data also for other situations prevent us from proceeding to a more detailed research in this direction. For the same reason we cannot observe if any changes in radioactivity from the old flows up to newest ones had taken place.

According to data from relevant literature, the Perşani basalts present contents in U close to those observed in the alkaline basalts from Hawai, and in olivine basalts from the Big Bend National Park. (table 3).

2.3.2. *Andesites.* The comparative examination of the U, Th, K average values evidences various contents, as well as differences among the Călimani, Gurghiu, Harghita volcanic massifs (table 2).

TABLE 3

Contents in U and Th in basalts (according to published data synthetized by J. M. Stussi, 1970)

Petrographical type	U Mg/g	Th Mg/g	Th/U Mg/g
Olivine basalts, Hawaï (Larsen et al., 1960)	0.41–0.47	1.3–1.4	2.7–3.4
Alkali-olivine basalts, Hawaï (Larsen et al. 1960)	1.8	5.4	3.1
Alkalic basalts., Japan (Heier and Rogers, 1963)	0.48–0.57	3.6–4.2	7.6
High-alumina basalts, Japan (Heier and Rogers, 1963)	0.13–0.28	0.45–1.10	3.7
Tholeiitic basalt, Japan (Heier and Rogers, 1963)	0.03–0.26	0.05–0.32	1.6
Tholeiitic basalt, Hawaï (Compston et al., 1968)	0.18	0.69	3.8
Olivine basalts Big Bend National Park (Gottfried, 1963)	1.2–1.9	2.6–3.9	2.1–2.4
General average (Heier et al., 1963)	0.59	2.7	4.5

For uranium the smallest average contents are observed at hornblende-andesites from the Călimani Mts (1.1. p.p.m.) and the highest ones for biotite-andesites from the Harghita Mts (3.5 p.p.m.). The average value of thorium rises from 3.7 p.p.m in hornblende-andesites, Călimani Mts to 14.5 p.p.m in biotite-andesites from the Harghita Mts. As regards the potassium the average values are ranging between 1.0 per cent in andesites bearing pyroxenes and hornblende from the Gurghiu Mts and 3.1 per cent in biotite-andesites from the Harghita Mts. A direct correlationaship is observed between the rising of contents in Th and K.

As a whole, the andesites present larger contents in K and U as compared to basalts with exception of the andesites from the Gurghiu Mts (table 2 and fig. 1). Likewise the andesites from the Harghita Mts display a more intense radioactivity than those from the Gurghiu and Călimani Mts (fig. 1).

Andesites bearing pyroxenes and hornblende from the Gurghiu Mts present lower U, Th, K contents in comparison with those from the Călimani and Harghita Mts. The average value of U is the same as that

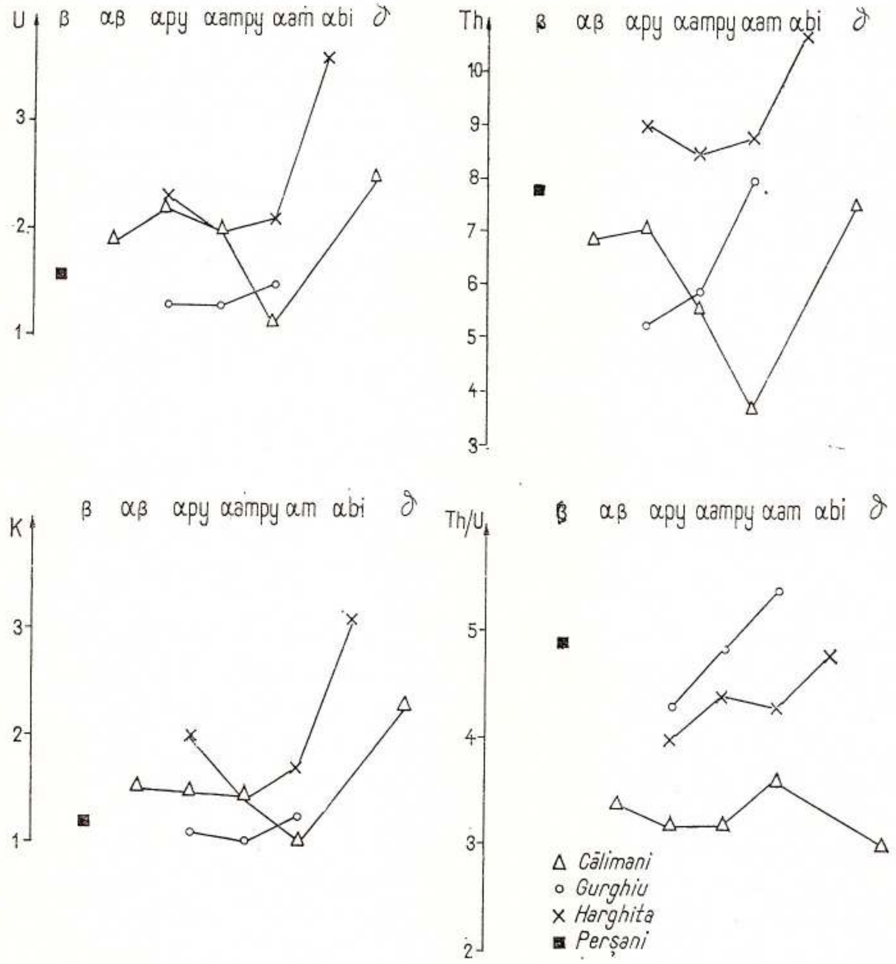

Fig. 1. — Variation of average contents of U, Th, K in volcanic rocks from the Călimani, Gurghiu, Harghita and Perşani Mts. 1, Călimani Mts.; 2, Gurghiu Mts.; 3, Harghita Mts.; 4, Perşani Mts. β, basalt; αβ, basaltic andesite; αpy, pyroxene-andesite, αpyam, hornblende and pyroxene-bearing andesite; αam, hornblende-andesite; αbi, biotite-andesite; δ, dacite

of pyroxene-andesites and andesites bearing pyroxenes and hornblende from the Gurghiu Mts.

Among the petrographical types with a regional widespread over the territory of the volcanic zone, the rising of the average values of

U, Th and K from north towards south is observed only for hornblende-andesites (table 2 and fig. 1).

As regards the biotite-andesites from the southern part of the Harghita Mts the average contents of 3.6 p.p.m. U, 14.5 p.p.m. Th and 3.1 per cent K are to be noted. In the light of our today knowledge these contents seem to be connected with biotite, a correlationship between the increase of K and that of U-Th being noticed. The biotite-andesites from the Harghita Mts offer a conclusive example of correlation between the radioactivity and the mineralogical characters of volcanic rocks.

When considering the succession in time of eruptions grouped into two stages one may notice that the andesites from the IInd stage display a higher radioactivity (table 4 and fig. 2). This rise of radioactivity is observed for all the three elements. On the other hand, the Th/U ratio is lower for products of the second stage.

2.3.3. *Andesitic volcanoclastics*. The radioactivity of these rocks presents two distinct situations : *a*) constituent elements i.e. lava fragments ejected in explosive phases present the radioactivity of previously commented andesitic rocks ; *b*) the cement presents lower U contents, thus : 0.5–1.3 p.p.m. U in cement as compared to 1.3–1.7 p.p.m. in rock fragments.

Although informative, the analytical data regarding the cement of pyroclastics ascertain the mobilization processes of Uranium.

It is possible that the leaching of uranium would have taken place subsequently to the formation of the volcanoclastic deposit, as it may be observed within the volcano-sedimentary formation from the Provincia Romana (Loccardi, Mittempergher, 1971)[2]. An as yet peculiar indication referring to lithotypes wherein U and Th might accumulate is furnished by andesitic epiclastics. Particularly in a silt level of andesitic composition, pertaining to the volcano-sedimentary deposits of the Toplița Basin, (Călimani Mts), contents exceeding the average radioactivity have been dosed.

As for basalts the lack of published data relating to U, Th, K in other neighbouring neovolcanic zones does not allow comparative observations on regional scale.

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According to data from relevant literature (table 5) the average contents of U and Th of the basaltic andesites are similar to those of the Madoc Area (USA) and Balhaş (USSR). Likewise the content limits

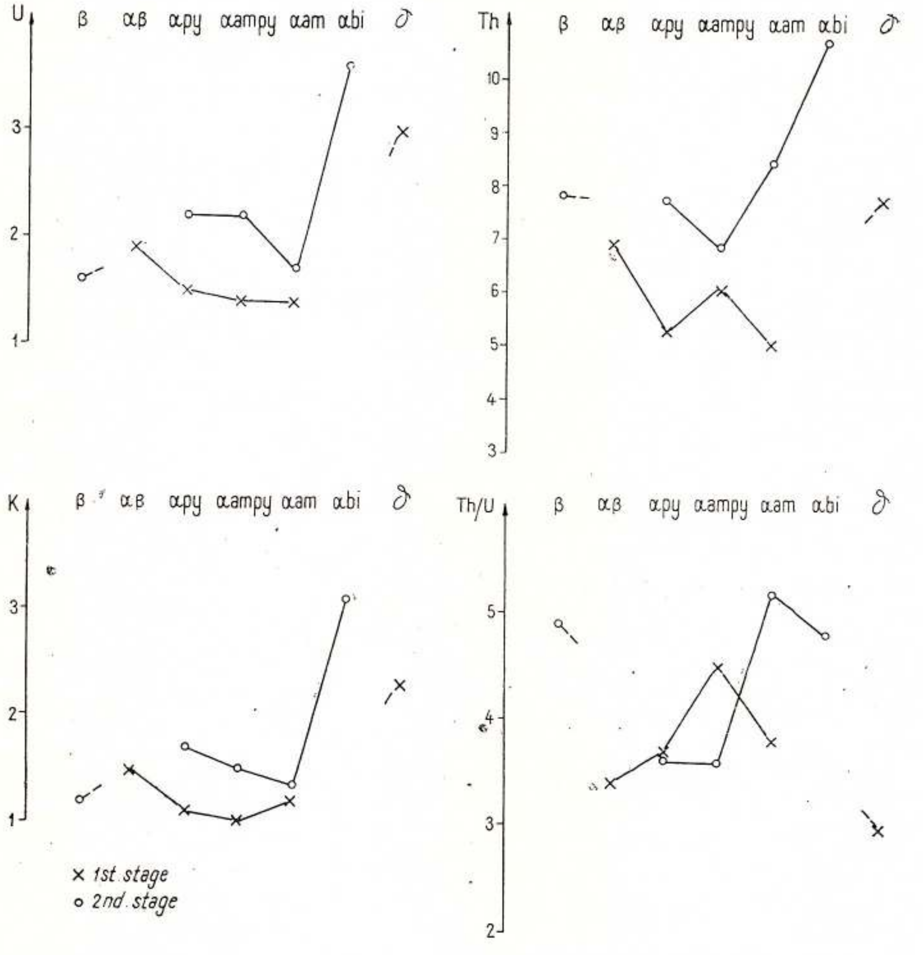

Fig. 2. — Variation of average contents of U, Th, K in volcanic rocks from compartments, stage I and II Călimani, Gurghiu, Harghita Mts. The same legend as for Fig. 1; a, I st stage; b, II nd stage.

of uranium in andesites under investigation, are comprised in limits dosed for andesites of the San Juan calc-alkaline province with "latitic" tendency (USA).

TABLE 4

Average content and variation range of radioactive elements in volcanic rocks pertaining to two stages

Petrographical type. Volcanic massif	Stage	n	U	Th	K	Th/U
1	2	3	4	5	6	7
			Basalts			
Perşani	I	—	—	—	—	—
	II	33	1.6 / 0.8—3.1	7.8 / 3.4—17.4	1.2 / 0.6—2.1	4.9 / 1.2—8.2
			Basaltic andesites			
Călimani	I	16	1.9 / 1.2—3.2	6.9 / 3.4—10.1	1.5 / 1.0—2.2	3.4 / 2.4—5.0
	II	—	—	—	—	—
			Pyroxene-andesites			
Călimani	I	14	1.7 / 0.8—2.8	5.2 / 2.3—8.3	1.2 / 0.5—1.8	3.2 / 1.4—6.8
	II	21	2.6 / 0.7—5.5	8.6 / 2.5—17.3	1.9 / 0.3—3.1	3.3 / 1.4—5.1
Gurghiu	I	5	1.1 / 0.7—1.6	5.8 / 3.0—7.1	1.0 / 0.5—1.4	5.1 / 4.3—6.3
	II	12	1.3 / 0.9—2.3	5.1 / 3.4—6.6	1.1 / 0.6—1.7	3.9 / 2.6—5.1
Harghita	I	—	—	—	—	—
	II	10	2.3 / 1.3—3.1	9.1 / 6.1—10.5	2.0 / 1.5—2.8	4.0 / 2.7—5.1
			Pyroxene-and hornblende-bearing andesites			
Călimani	I	5	1.1 / 0.6—2.1	3.7 / 2.6—5.5	0.8 / 0.6—1.0	3.6 / 1.6—5.0
	II	8	2.5 / 0.8—4.9	6.8 / 3.7—14.4	1.7 / 1.2—4.0	3.0 / 1.6—7.4

table 4

1	2	3	4	5	6	7
Gurghiu	I	7	1.3 0.7–2.2	6.1 3.5–9.9	0.9 0.7–1.2	5.1 3.4–9.0
	II	5	1.4 0.9–2.3	5.6 3.4–6.8	1.1 0.9–1.5	4.4 2.4–6.6
Harghita	I	6	1.8 1.3–2.1	8.2 5.0–10.1	1.3 0.7–1.7	4.6 3.8–5.9
	II	2	2.6 2.3–2.8	9.5 8.6–10.4	1.6 1.4–1.8	3.7 3.7–3.7
			Hornblende-andesites			
Călimani	I	7	1.1 0.6–1.6	3.7 2.0–8.7	1.0 0.6–2.0	3.6 2.5–4.8
	II	–	–	–	–	–
Gurghiu	I	–	–	–	–	–
	II	10	1.5 0.8–2.1	8.0 4.3–12.1	1.25 0.9–1.4	5.4 2.5–7.5
Harghita	I	3	2.0 1.5–2.8	8.0 6.6–9.8	1.7 1.6–1.9	4.2 3.5–5.0
	II	3	2.2 1.7–2.6	9.7 8.4–10.7	1.7 1.6–1.9	4.4 3.8–4.9
			Biotite-andesites			
Harghita	I	–	–	–	–	–
	II	10	3.6 1.0–7.0	14.5 12.2–17.7	3.1 2.0–3.7	4.8 2.5–12.2
			Dacites			
	I	12	2.5 1.3–3.8	7.6 3.7–10.1	2.3 0.6–3.0	4.0 1.8–3.8
	II	–	–	–	–	–
			Diorites and microdiorites			
Călimani	II	5	2.7 1.3–4.0	10.5 5.1–15.8	2.1 1.3–2.6	3.9 3.3–4.3

2.3.4. *Dacites.* The most acid volcanic rocks of the zone investigated present average contents of U, Th and K exceeding those of andesites (excepting biotite-andesites). For Th the content limits and the average value are the same as those for andesites.

TABLE 5

Contents in U and Th within andesites of alkalic and calc-alkalic volcanic series (according to synthesized data of revelant literature by J. M. Stussi, 1970)

Petrographical type	U mg/g	Th mg/g	Th/U mg/g
Andesites, Hawaï (Larsen et al., 1960)	1.2–1.3	3.6–5.3	2.8
Andesites, N. Zeeland (Taylor and White, 1966)	0.24–1.4	0.51–4.7	2.7–4.8
Andesites, Madoc Area U.S.A. (Gottfried et al., 1958)	1.4–1.9	—	—
Andesites, basaltic andesites, San Juan (Larsen et al., 1958)	1.9–32	—	—
Andesites, Auvergne (Goldstein et al., 1961)	2.3	—	—
Andesites, Balhaş (Kazmin, 1966)	1.7	6.0	4.8
Andesites, Lessen Park (Adams, 1955)	1.0–1.9	—	—
Circumpacific andesites, Saipan (Gottfried et al., 1963)	0.48–1.1	0.4–1.2	0.4–2.9

Values exceeding 3 p.p.m. U are observed for biotite dacite from Drăgoiasa, as well as the hornblende-dacite from Voivodeasa Valley. They are lower than those recorded for biotite-andesites from the south of the Harghita Mts (table 2).

2.3.5. *Dioritic rocks* from the central part of the Călimani Caldera present also higher contents in U, Th, K as compared to andesites. It is an indication relating to the tendency of increase or radioactivity under subvolcanic conditions.

3. Abundance of uranium, thorium and potassium.

Clarke of uranium and thorium of volcanics (according to Vinogradov, 1963) is ranging in following limits: acid effusive, U 2–7 p.p.m., Th 9–25 p.p.m.; basic effusive U 0.2–4 p.p.m., Th 0.5–10 p.p.m. The analysed rocks are comprised within these limits (table 2).

The abundance of U, Th and K in the neovolcanic zone from the East Carpathians as a whole, according to stages and volcanic massifs or more detailed according to petrographical types is resulting from tables 2 and 4. These data are compared to the distribution of trace elements in Earth according to Heier, Rogers, 1963 (table 6).

TABLE 6

Distribution of K, Th, and U in Earth

	K %	Th. ppm.	U. ppm.	Th/U
Crust	2.1	7.8	2.1	3.7
Oceanic	0.87	2.8	0.64	4.4
Continental	2.6	10.0	2.8	3.6
Mantle	0.11	0.08	0.016	5.0

It results that the content limits for the analysed volcanics are close to the values indicated for the crust.

The statistical distribution of radioactivity in the chief petrographical types was studied by means of histograms.

While examining the plate I two opposite tendencies of the distribution are observed, namely, the uniform distribution of U and Th in basalts and a uniform distribution of K in andesites and volcanoclastics, excepting the hornblende and pyroxene-andesites.

For the territory as a whole , the tendencies which have been stated for various petrographical types are not more so distinct, a distribution close to the uniform one for each of the three elements being stated.

In histograms there is also pointed out that the maximum contents statistically pertain to intervals from table 7.

TABLE 7

Intervals with maximum contents in U, Th, K

	Basalt	Basaltic andesite	Pyroxene-andesite	Hornblende and pyroxene andesites	Hornblende-andesites	Volcano-clastics
U. ppm.	1–1.9	1.2–1.7	0.7–1.3	0.7–2.7	1.6–1.9	0.6–1.4
Th. ppm.	6–69	3.4–5.5 și 7.8–9.9	5.6–7.7	3.8–9.4	1.8–3.5 și 5.4–7.1	3.2–4.7
K %	0.88–1.09		0.8–1.99	0.56–1.67	0.72–1.43	0.75–0.99

Affinities and differences among the petrographical types were also examined depending on their appurtenance to the volcanic unit by means of the U-K, Th-K, U/Th-K diagrams. Among these, the U-Th diagram is presented as an example (plate II).

Differences between basalts and basaltic andesites are observed, they being evidenced by the position and the various tendency of the correlation cloud. As regards the correlative relations between U, Th, and K, no differences as to andesites may be observed among various types, excepting biotite-andesites, and likewise no differences as for their appurtenance to the Călimani—Gurghiu—Harghita massifs, may be noticed.

It may be concluded that the correlative diagrams illustrate differences between the radioactivity of basalts, andesites and biotite-andesites. Within the group of andesites (excepting the biotite-andesites) the differences are less important.

The plotting of the U, Th, K values from andesites and biotite-andesites in the Th-K and U-K diagrams (according to H e i e r and R o g e r s, 1963) gives following indications (fig. 3) : location of andesites in the field of intermediary rocks and with a tendency to the acid ones ; different contents in Th and K of the biotite-andesites places them above the value which represents the average of granites.

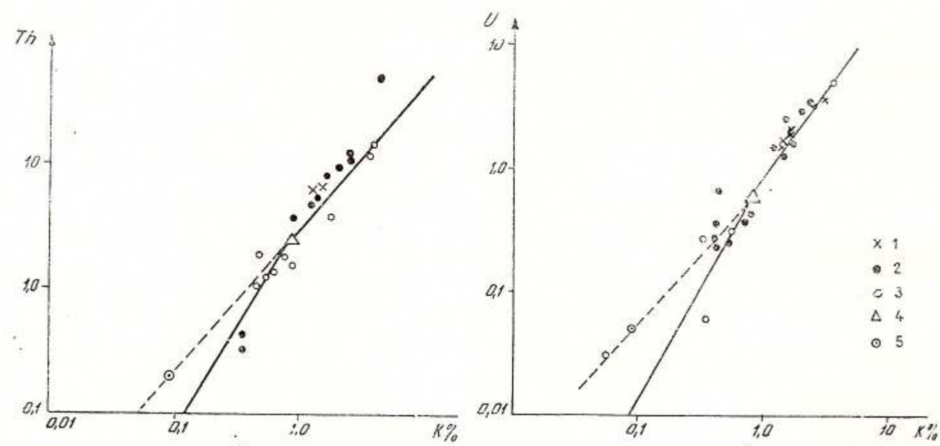

Fig. 3. — Thorium vs potassium (a) and uranium vs potassium (b) in andesites, biotite-andesites and various geologic materials (according to H e i e r and R o g e r s, 1963). 1, andesites ; 2, biotite-andesites ; 3, average of 166 granites. 4, average of 23 basalts and gabbros ; 5, average in meteorites, L e v i n et al., (1956)

On purpose to establish the correlation degree among the analysed radioactive elements, the correlation coefficients for various types of rocks have been computed. The table 8 which synthesises the results obtained, shows a closer correlation between the uranium and thorium in comparison with that between the U-K or Th-K for all types of andesites. An exception is presented by pyroxene-andesites for which the higher correlation values (even the maxima) were obtained for U-K (+ 0.93) and Th-K (+ 0.91) values exceeding that of U-Th (+ 0.83).

TABLE 8

Correlation coefficients among the radioactive elements for andesites

Nr.	Petrographical type	n	U–K	Th–K	U–Th
1	Basaltic andesites	16	+0.44	+0.39	+0.75
2	Pyroxene-andesites	62	+0.93	+0.91	+0.83
3	Hornblende and pyroxene andesites	33	+0.62	+0.72	+0.64
4	Hornblende-andesites	23	+0.79	+0.69	+0.89
5	Biotite-andesites	10	−0.04	+0.19	+0.98
6	Volcanoclastics	43	+0.84	+0.79	+0.81

Among the analyzed types of andesites, the biotite-andesites which have very low correlation coefficients among U-K and Th-K, stand out. The latter practically indicate the lack of correlation among these elements, but they present the maximum value (+ 0.98) for the correlation coefficient between U-Th.

Over the volcanic area on the whole, a rise of activity from basalts to dacites is observed. Considering the evolution trend of volcanism and the succession in time of eruptions, it may be stated that the radioactivity had been more intense at the beginning of volcanism. This general scheme is, however, intricate also owing to petrogenetical factors of another nature; thus a rise of the radioactivity, exceeding the one for dacites towards the end of the unfolding of volcanism, at the level of the eruption of biotite-andesites, is recorded.

The explanation of this "anomaly" is linked to the clearing up of the magma genesis of the biotite-andesites from the south of the Harghita Mts.

It is probably that the enrichment in U and Th might be explained by a pneumatolytic differentiation phase in the evolution of magma.

Concomitantly the effect resulting from the contamination of magma could also be taken into consideration. It is obvious that the geochemical peculiarities of this andesite as compared with andesites over the territory, are connected to the different geological environment wherein the magma had evoluated towards the surface.

4. Variation of uranium, thorium and potassium in magmatic differentiation

During the history of volcanism the radioactivity has recorded variations as a function of the character of magmas and the differentiation processes which took place in the course of the stages . The correlation of the U, Th and K distribution with petrogenetical and volcanological features points out the dependence of the distribution of these elements on various processes of magmatic differentiation and certain peculiar situations (biotite-andesites). All the previously presented data lead us to this essential conclusion.

The variation diagram of the average contents of U, Th, K in volcanics of the compartments from the Călimani Mts (fig. 4) illustrates this fact. In the I-st stage there is to be observed a marked decrease of radioactivity from dacites to hornblende-andesites, corresponding to the transition from the dacitic magma to the andesitic one, and subsequently the rise of the radioactivity in the cogenetical andesitic suite

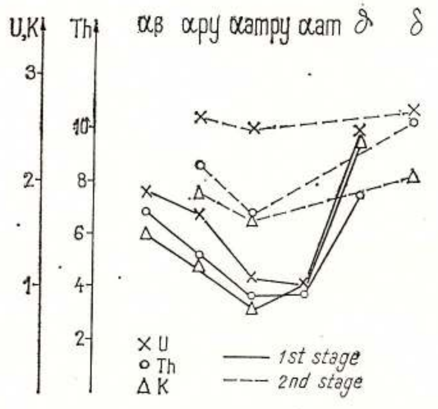

Fig. 4. — Variation of average contents of U, Th, K in volcanites of the two compartments from the Călimani Mts. 1, uranium; 2, thorium; 3, potassium; α β, basaltic andesite; αpy pyroxene-andesite; αpyam, hornblende and pyroxene-bearing andesite; α am hornblende-andesite; δ, dacite.

according to their differentiation sense hornblende-andesite, pyroxene-andesite, basaltic andesites. The same differentiation direction also appears for andesites of the IInd stage. The diagram also points to the fact that the andesites of the IInd stage are more radioactive.

5. Radioactivity and secondary alteration processes

On purpose to study the behaviour of U, Th and K in the secondary transformation processes, which affected some volcanics of the region, there were collected samples from outcrops, mining works and drillings as follows: *a*) experimental exploitation for sulphur Negoiul Românesc (Călimani Mts); *b*) outcrops of the hydrothermalized hornblende-andesite from Zebrac Valley-Stînceni (Călimani-Mts); *c*) outcrops of hydrothermalized pyroxene- and hornblende-bearing andesites Şumuleu and Seaca-Tătarca craters (Gurghiu Mts); *d*) hydrothermalized andesites, drillings from the Fîncel—Lăpuşna Caldera (Gurghiu Mts); *e*) Harghita-Băi and Sîntimbru-Băi drillings (Harghita Mts) for pyroxene-andesites affected by secondary alteration processes. The contents in U, Th, K of the rocks altered were compared to those of the corresponding petrographical type, in a fresh state, with a view to obtain some indications regarding the behaviour of these elements in hydrothermal and fumarolic processes.

The study of samples from the area of the sulphur deposit has evidenced that the solfatatric processes from the Călimani Caldera have caused a mobilization of U, Th and K. Thus, as compared to averages of 2.2 p.p.m. U, 7.1 p.p.m. Th and 1.53% K, an increase of radioactivity for the silicified rock and the leaching of U and K for the sulphur-bearing rocks is observed.

In the propylitized hornblende-bearing andesite of the Zebrac Valley the hydrothermal process led only to the increase of the content in K. The U and Th values are close or even equal with the average values of fresh andesites.

Hydrothermalized andesites intercepted by drillings from the Gurghiu Mts indicate lower contents in K. The latter are ranging from 1—20 per cent in comparison with average values exceeding 1 per cent. The radioactivity of the hydrothermalized andesite shows variations expressed in values averaging from 1.3 to 1.6 p.p.m. for U, and 5.9 to 8 p.p.m. for Th. The quartziferous amphibolic andesite from Seaca-Tătarca is showing higher values, 3 p.p.m. for U and 9.5 p.p.m. for Th at a depth of 706 m, as well as 2.5 p.p.m. for U and 6.6 p.p.m. for Th at a depth of 904 m. Likewise the diorite identified in the Şumuleu drilling at a depth of 1.171 m presents contents of 2.8 p.p.m. for U and 9.5 p.p.m. for Th. The contents up to 11.5 p.p.m. for Th were dosed in the hydrothermalized hornblende-andesite from the sector of Bătrîna.

The argillized pyroxene-andesite from Harghita-Băi shows lower contents in U and Th in comparison with the fresh andesite, 1–1.7 p.p.m. U in altered andesite, and an average of 2.3 p.p.m. of U in Harghita pyroxene-andesite ; 5.5–7 p.p.m. of Th, respectively an average of 9 p.p.m. of Th in pyroxene-andesites from Harghita.

The contents of potassium vary around a value of 1.97 the average for the fresh pyroxene-andesite. Nevertheless many samples with contents ranging from 0.19 to 0.75 p.p.m. are noticed.

In the Sîntimbru-Băi area as compared to the Harghita-Băi area, the rise of the radioactivity is differing. Thus the argillized and silicified andesite presents contents of U and Th exceeding the average values in pyroxene-andesites (table 2). The varied values of K indicate a behaviour similar to that observed in the Harghita-Băi area.

The relevant analytical data allow only to tackle the above mentioned problems. They are, however, sufficiently significant to prove that the secondary alteration processes have led to the modification of the geochemical balance of U and Th as a function of the composition of the „active gas'' and of hydrotherms, an increase or decrease of radioactivity thus resulting.

6. Conclusions

This paper furnishes the first information concerning the geochemistry of Th, U and K in neovolcanics from the East Carpathians.

The number of 369 analyses may be considered as representative since they proceed from all the volcanic massifs, pertain, to an almost equal extent, to the two main stages related to the unfolding of volcanism and their products.

The radioactivity of basalts differs from other types of radioactivity likewise within this group the radioactivity is differing for basalts and basaltic andesites.

The Perşani basalts show contents of U close to those observed for alkaline basalts from Hawai and North America.

Andesites present higher contents of U and K in comparison with basalts. Within the group of andesites one may observe as follows : *a*) rise of the radioactivity according to the differentiation hornblende-andesite → basaltic andesites, andesites from the IInd stage being more radioactive ; *b*) differences between the radioactivity of hornblende and pyroxene-bearing andesites from the Gurghiu Mts and the same andesites from the Călimani-Harghita Mts ; *c*) along the whole volcanic chain the radioactivity increase north-southwards, the Harghita andesites being richer in Th, U, K than those from Gurghiu and Călimani. From these the most radioactive are the biotite-andesites; it is probable that the enrichment in U and Th might be explained by a pneumatolytic differentiation during the evolution of the magma; *d*) the cement of volcano-clastics presents contents of U lower than those of lava fragments, whereas an accumulation of U and Th in fine-grained epiclasticis noted (Toplița).

Andesites from the Călimani-Gurghiu Mts present average contents of U and Th similar to those of andesites of USSR and USA.

It may be concluded that differences between the radioactivity of basalts, andesites and dacites do exist; over the volcanic area, as whole, a rise of radioactivity from basalts to dacites is observed.

The correlation among the U, Th and K distribution and the petrographical and volcanological features point out the dependence of the distribution of these elements on the magmatic differentiation processes (which took place during each of the two stages) and on certain peculiar situations namely the case of the biotite-andesites.

The study of volcanic products which have undergone secondary alteration processes yields following data: in sectors with fumarolisations the radioactivity is higher for silicified rocks and lower for the argillized ones; in sectors with hydrothermal alterations, a variation of radioactivity about guide mark values (average content in the fresh-appeared rock) is observed. Increases of the U and Th values occur for more acid rocks such as andesites and amphibolic microdiorites.

The results obtained demonstrate the usefulness of radiometric study with a view to complete our knowledge regarding the geochemistry of neovolcanics and relying on it to proceed to a thorough study of petrogenetic problems. There were likewise obtained indications related to the contribution of the radiometry to detect zones of secondary enrichment, as well as to point out the differences between the fumarolic processes and the hydrothermal ones, fact presenting involvements of metallogenetical nature.

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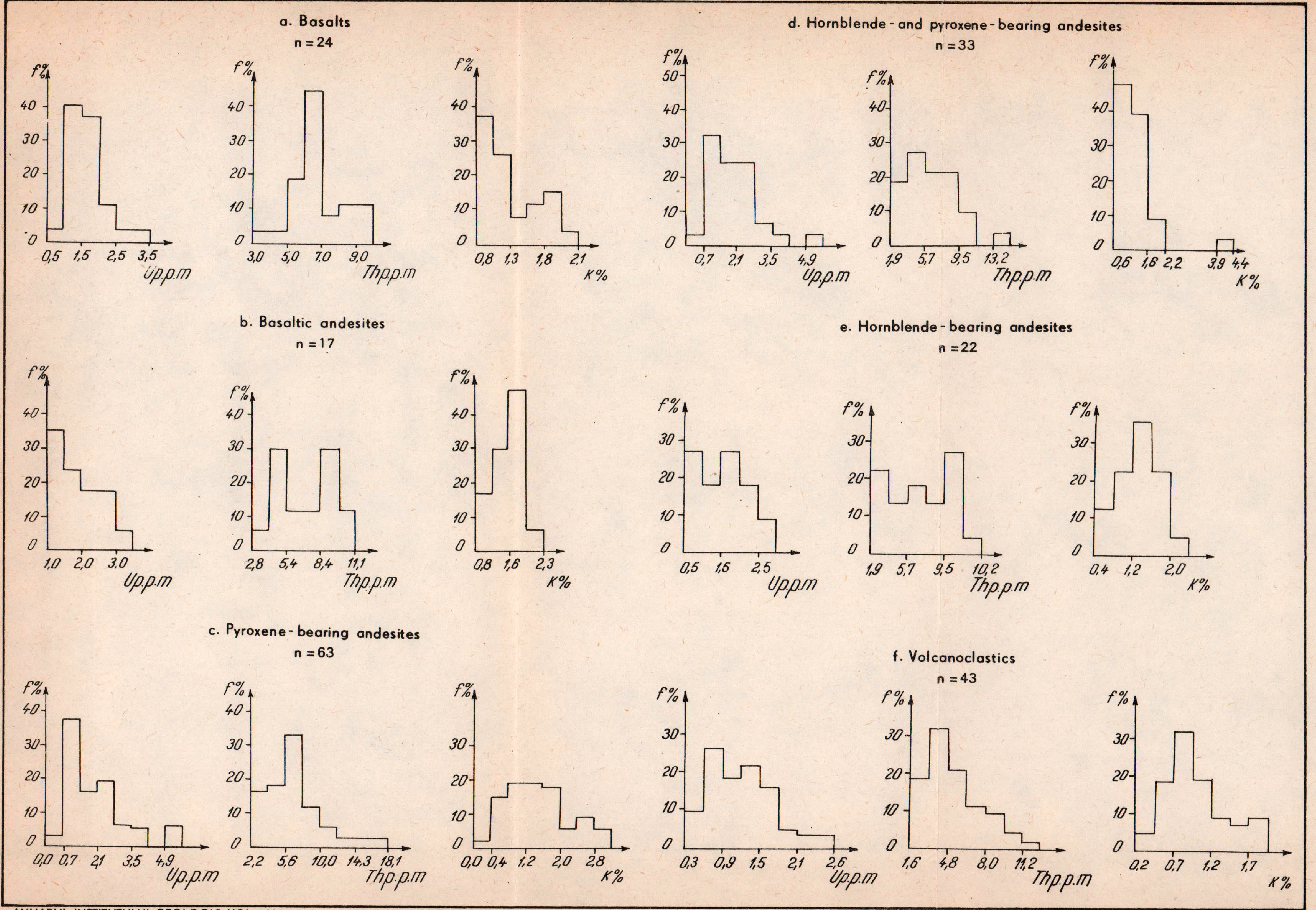
PARTIAL HISTOGRAMS



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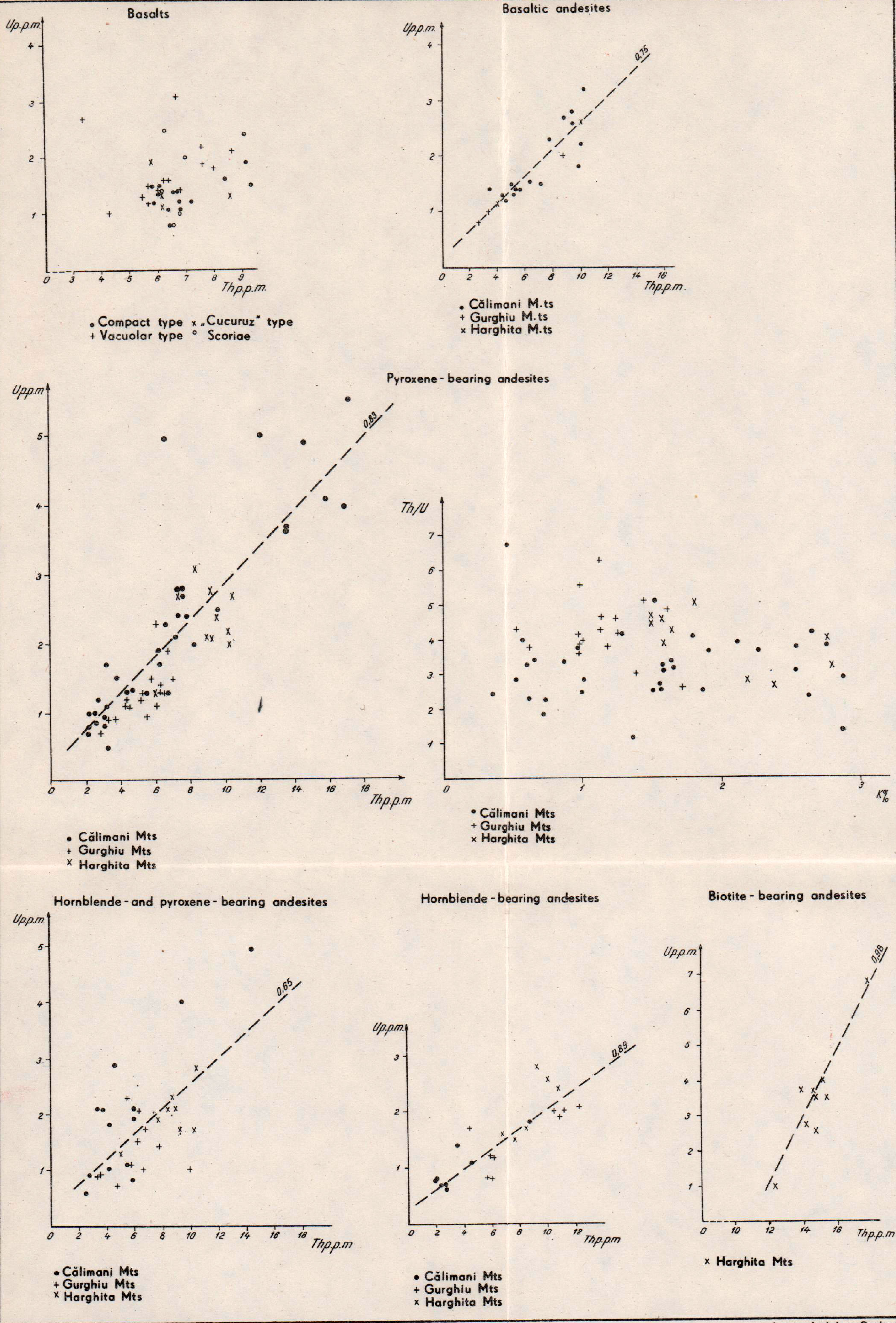
U VS. TH AND TH/U VS. K IN BASALTS AND ANDESITES



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CONSIDERATIONS ON THE EVOLUTION OF MAGMAS DURING THE NEOGENE VOLCANISM IN THE CĂLIMANI, GURGHIU AND HARGHITA MTS

BY

DAN P. RĂDULESCU[1] and AL. DIMITRIU[2]

The Neogene volcanics from the Călimani, Gurghiu and Harghita Mts are rather well-known from the chemical point of view, and petrographical considerations on this basis either of a regional or general character were presented in several previously published papers (Rădulescu, 1963; Rădulescu, Borcoş, 1968 etc.). The massive increase of the chemical information — as regards both the major components and, to a less extent, the minor components — allows today a statistical analysis of the petrological hypothesis.

The results thus obtained are integrating into the present-day outlooks on the evolution of volcanism from this region; they partly substantiate the already expressed concepts as a result of geological and petrological studies, but they largely represent on the other hand, quite new ideas.

Primary information and used statistical methods

Our present-day knowledge of the Neogene volcanism from the Călimani, Gurghiu and Harghita Mts does not as yet satisfactorily argue the answers to two fundamental questions :

1. Which are the relationships among the volcanic manifestations of the Călimani, Gurghiu and Harghita areas ?

2. Which are the relationships among the evolution stages of volcanism both within each of the above three areas and also comparatively among them ?

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Geological-petrological and volcanological criteria have not only allowed to answer these questions but also to draw a general picture of the geological evolution of this region. However, the answers obtained comprise, besides numerous objective elements, also a large hypothetical part, fact that renders them liable to permanent improvement from the argumentation and interpretation points of view.

As the way these two questions may be answered allows to understand the evolution of the volcanism in this region, and since they are very well suited to be examined with the aid of chemical data and by means of statistical methods, we will focus our attention on them. The analytical data are concerning both the contents in major components (SiO_2, Al_2O_3, Fe_2O_3, FeO, CaO, MgO, Na_2O, K_2O, TiO_2, P_2O_5) (152 samples) and the minor components (Pb, Cu, Zn, Ni, Co, Cr, V) (310 samples); they are mostly supplied by the papers of Rădulescu (1961) and Peltz et al.[3] As it may be stated from tables 1 and 2, all the petrographical

TABLE 1

*Relative frequency f% of petrographical types pertaining to the Neozoic volcanism from the Călimani, Gurghiu and Harghita Mountains, studied as for their composition of major chemical elements**

Petrographical type	Călimani			Gurghiu			Harghita		
	I	II	I+II	I	II	I+II	I	II	I+II
Dacites	8.8		6.4						
Hornblende-andesites	26.7	11.2	22.2	28.6	43.9	41.7		29.8	27.9
Hornblende-pyroxenes andesites	2.2	5.6	3.2	14.3	7.3	8.3	37.5	9.6	11.5
Pyroxenes-andesites	24.4	72.2	38.1	42.9	43.9	43.7	50.0	57.0	56.6
Basaltic andesites	35.6		25.4				12.5		0.8
Diorites, microdiorites		5.6	1.6						
Gabbros, microgabbros	2.2	5.6	3.2						

* In all Tables, the two volcanic stages from each region are noted with I and II

constituents of the region under investigation are satisfactorily represented in the analysed material. Our discussion is relying on fundamental data resulting from the geological research, particularly (*a*) on the sepa-

[3] Peltz S., Vasilescu Al., Vasiliu Cecilia, Udrescu Constanța (1969). Corelarea formațiunilor eruptive masive din lanțul Călimani—Gurghiu—Harghita. Mansuscript, Arh. Inst. Geol. București.

ration in 3 areas displaying volcanic phenomena, and (*b*) on the separation within each of these three areas of two major display stages. In order to obtain more confident conclusions there have been utilized uni- and multivariate statistical analyses, some among these methods having been for

TABLE 2

Relative frequency f % of petrographic types pertaining to the Neozoic volcanism from the Călimani, Gurghiu and Harghita Mountains studied as for their composition in minor chemical elements

Petrographical type	Călimani			Gurghiu			Harghita		
	I	II	I+II	I	II	I+II	I	II	I+II
Dacites	9.8		6.0						
Hornblende-andesites	19.5	11.5	16.4	50.0	34.5	37.8		47.7	41.1
Hornblende-pyroxenes-andesites	4.9	11.5	7.5	12.5	31.0	27.0	28.6	25.0	25.4
Pyroxenes-andesites	29.3	50.0	37.3	25.0	34.5	32.4	57.1	27.3	31.4
Basaltic andesites	36.6	7.7	25.4	12.5		2.7	14.3		2.0
Diorites, microdiorites		11.5	4.5						
Gabbros, microgabbros		7.7	3.0						

the first time applied to the study of igneous products. The conclusions that were reached are relying not only on the values of either statistical parameter or on the indications of a single multivariate test — elements which separately have only a limited significance — but on the general image which is forming as a result of totalizing and confronting of characteristics suggested by all criteria.

The characteristical features of the chemical composition of each rock association have been established by means of the statistical distribution parameters of major components (arithmetic mean $\overline{x}$, standard deviation $S_{\overline{x}}$ and the coefficient of variation $V = (S_{\overline{x}}/\overline{x}).100$). Their estimation was achieved in agreement with the law of normality which controls, as a rule, the distribution of major chemical components in igneous rocks (Rodionov, 1964). In cases when the number of analytical values $n \leqslant 10$, the standard deviation has been evaluated by the Dixon's and Massey's (1957) functions. The values of parameters are figured in table 3 being supplemented in table 4 by the range $\lambda = x_{max} - x_{min}$ of contents in SiO_2.

The study of chemical relations and implicitly the genetical ones among the rock associations was made by two methods which utilize the comparison of existent data, the first using their totality, and the second, selection of them.

TABLE 3

Statistical parameters of major chemical eleements

Localization	n	SiO_2			Al_2O_3			Fe_2O_3			FeO			MgO			CaO			Na_2O			K_2O		
		$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V	$\bar{x}$	$S_{\bar{x}}$	V
Călimani I	50	58.47	5.19	8.8	18.29	1.76	9.6	3.69	1.77	47.9	3.15	2.12	67.3	3.68	1.93	52.4	7.29	1.82	25.0	3.24	0.65	20.1	1.54	0.72	46.7
Călimani II	17	57.12	2.95	5.1	18.45	1.08	5.8	2.60	1.04	40.0	3.62	1.11	30.7	3.70	1.26	34.0	6.56	1.02	15.5	3.74	0.62	16.6	2.12	0.54	25.5
Călimani I+II	67	56.57	4.69	8.3	18.33	1.60	8.7	3.40	1.68	49.4	3.27	1.91	58.4	3.68	1.77	48.1	7.09	1.68	23.7	3.37	0.12	3.5	1.70	0.72	42.3
Gurghiu I	6	59.42	4.67	7.8	18.32	1.16	6.3	4.66	1.85	39.7	1.72	1.29	75.0	2.43	1.15	47.3	6.43	1.44	22.4	3.48	0.30	8.6	1.42	0.24	16.9
Gurghiu II	31	56.32	2.36	4.2	18.67	1.54	8.2	4.72	1.48	31.4	2.44	0.92	37.7	3.23	1.02	31.6	7.10	0.97	13.7	3.64	0.49	13.5	1.29	0.25	19.4
Gurghiu I+II	37	56.86	3.04	5.3	18.61	1.47	7.9	4.72	1.52	32.2	2.38	1.01	42.2	3.09	1.07	34.6	6.99	1.05	15.0	3.61	0.46	12.7	1.31	0.25	19.1
Harghita I	7	57.76	1.26	2.2	18.49	0.57	3.1	2.26	2.17	96.0	3.38	1.18	34.9	3.83	1.79	46.7	7.17	1.04	14.5	3.09	0.22	7.1	1.61	0.44	27.3
Harghita II	44	59.27	3.20	5.4	18.37	1.47	8.0	2.97	1.37	46.1	2.11	1.43	67.8	2.94	1.18	40.1	6.24	1.42	22.8	3.69	0.65	17.6	2.15	0.80	37.2
Harghita I+II	51	59.05	3.03	5.1	18.39	1.37	7.4	2.86	1.51	52.8	2.30	1.46	63.5	3.08	1.30	42.2	6.38	1.40	21.9	3.60	0.76	21.1	2.07	0.77	37.2

The Bondarenko's method (1968) worked out according to the thesis that silica shows a relatively high inertness in differentiation of magmas suggests that the testing of consanguinity of some rocks should be done in the framework of samples with close contents in silica,

TABLE 4

SiO_2 range

Localization		n	x min.	x max.
Călimani	I+II	67	48.69	69.15
Călimani	I	50	48.69	69.15
Călimani	II	17	49.89	60.80
Gurghiu	I+II	37	52.26	64.73
Gurghiu	I	6	53.32	64.73
Gurghiu	II	31	52.26	61.45
Harghita	I+II	51	54.39	67.11
Harghita	I	7	55.40	59.28
Harghita	II	44	54.39	67.11

which would define a reference moment in the evolution of magmas. In this intersection zone of SiO_2 ranges, the differences among the contents of other major chemical components can or cannot be essential. When investigatig the 6 rock associations from the region (Călimani I, Călimani II, Gurghiu I, Gurghiu II, Harghita I, Harghita II), it results that the intersection zone would be $\Delta\ SiO_2\% = \{59.28, 55.40\}$ .So as in the case of rock associations from the 3 areas (Călimani I + II, Gurghiu I + II, Harghita I + II) the intersection zone is $\Delta\ SiO_2\% = \{64.73, 54.37\}$ (Fig. 1). This study was carried out both for associations taken as a whole (fig. 1, tables 5 and 6) and for associations taken as pairs (Plate I, tables 7 and 8).

The analysis of differences as to the chemical composition of sub-collectivities, displaying a common silica range, is achieved for each chemical element taking into account a pair of rock associations, initially through a graphical procedure. The variation diagram of every element X_j with respect to the SiO_2 ($X_j = Al_2O_3$, $Fe_2O_3 \ldots$) is represented for a rock association so as it is exemplified in fig. 2. The differences between the distribution of a chemical element within the two rock associations are evaluated by the difference Z_i (see fig. 2). The average dispersion variance of the Z variable (when presuming that the actual average of

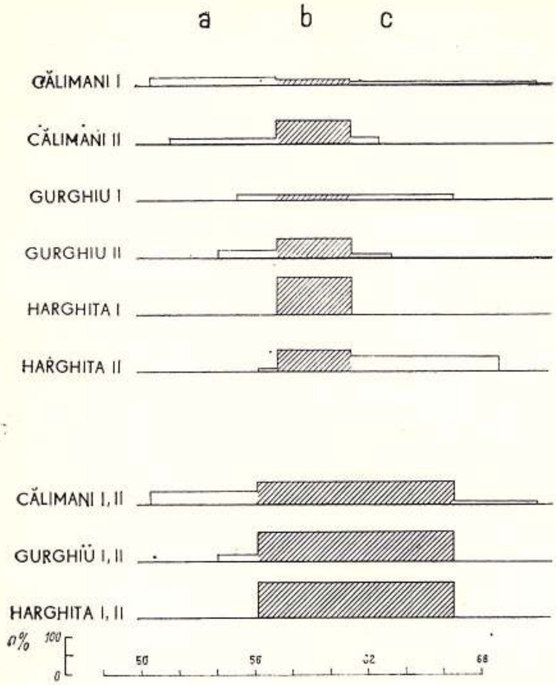

Fig. 1. — Relative participation of samples (n%) in common SiO_2 range (b), respectively lower (a) and higher (c) for testing consanguinity of rocks pertaining to formations on the whole.

TABLE 5

Testing of the hypothesis as regards the consanguinity among the volcanics in the three regions (W^2)

Stages	Chemical element	Gurghiu I + II		Călimani I + II	
Harghita I+II	Al_2O_3	0.57		0.03	
	Fe_2O_3	0.75		0.82	
	FeO	0.01		2.23	
	CaO	0.18	13.68	1.16	7.36
	MgO	3.85		0.01	
	Na_2O	3.69		2.37	
	K_2O	4.63		0.74	
Gurghiu I + II		Al_2O_3		0.30	
		Fe_2O_3		3.39	
		FeO		1.95	
		CaO		1.39	18.63
		MgO		3.96	
		Na_2O		2.04	
		K_2O		5.60	

TABLE 6

Testing of the hypothesis as regards the consaguinity among the volcanics pertaining to various stages of activity (W^2)

Stages	Chemical elements	Harghita II	Gurghiu I	Gurghiu II	Călimani I	Călimani II
Harghita I	Al_2O_3	3.99	1.06	0.60	1.10	1.21
	Fe_2O_3	0.38	0.11	0.29	0.68	1.88
	FeO	0.09	0.03	0.03	0.10	3.30
	CaO	3.57 10.56	0.48 8.06	3.54 9.57	3.59 11.74	5.79 14.72
	MgO	1.20	0.02	0.01	0.04	0.47
	Na_2O	0.00	2.29	0.08	2.17	0.93
	K_2O	1.33	4.07	5.02	4.06	1.14
Harghita II	Al_2O_3		0.17	4.96	0.31	1.50
	Fe_2O_3		0.01	0.58	0.25	6.53
	FeO		0.20	0.00	0.24	8.19
	CaO		2.88 6.23	0.02 14.04	1.32 9.15	0.42 30.51
	MgO		0.33	4.44	0.53	8.34
	Na_2O		1.57	0.27	2.57	1.17
	K_2O		1.07	3.77	3.93	4.36
Gurghiu I	Al_2O_3			0.07	0.04	0.23
	Fe_2O_3			0.43	0.15	2.41
	FeO			0.04	0.16	1.40
	MgO			0.35 8.06	0.00 5.16	0.99
	CaO			2.18	1.55	3.37 15.47
	Na_2O			3.49	2.13	3.56
	K_2O			1.50	1.13	3.51
Gurghiu II	Al_2O_3				1.03	0.15
	Fe_2O_3				0.20	6.73
	FeO				0.02	6.52
	CaO				0.70 9.21	0.14 21.94
	MgO				1.43	0.21
	Na_2O				5.18	0.61
	K_2O				0.65	7.58
Călimani I	Al_2O_3					1.14
	Fe_2O_3					11.78
	FeO					8.67
	CaO					3,56 42.86
	MgO					3.69
	Na_2O					4.21
	K_2O					9.81

differences is zero), together with the number of readings define a W_j^2 function (see table 12, where the computations are illustrated by an example). It is observed that the value of the W^2 function is estimated for each chemical element (W_j^2) which by totalizing of all chemical elements

TABLE 7

Testing of the hypothesis as regards the consanguinity among the volcanics in the three regions taken as pairs (W^2).

Stages	Chemical element	Gurghiu I + II		Călimani I + II	
Harghita I + II	Al_2O_3	0.01		0.22	
	Fe_2O_3	0.30		6.24	
	FeO	0.73		3.98	
	CaO	0.45	22.53	0.24	25.25
	MgO	0.01		3.40	
	Na_2O	1.31		0.01	
	K_2O	19.72		11.16	
Gurghiu I + II	Al_2O_3			0.07	
	Fe_2O_3			4.13	
	FeO			2.03	
	CaO			4.03	15.68
	MgO			0.04	
	Na_2O			0.38	
	K_2O			5.00	

informs about differences displayed by the whole chemical composition of rock associations under study.

W_j^2 and W^2 are controlled by chi-square distribution. The hypothesis as to the comagmatism relations is admitted as true for $W^2 < \chi^2_{\text{m.d.f.: 0,95 level of significance}}$, whereas the hypothesis implying non-essential differences as regards the distribution of a j element is accepted for $W_j^2 < \chi^2_{\text{1.d.f.; 0,95 level of signif.}}$ There results that the B o n d a r e n k o's method allows both the test of the comagmatism (by W^2) and the accurate determination of chemical elements which define the deviations from the magmatic consanguinity (W_j^2). In tables 5–8 the

TABLE 8

Testing of the hypothesis as regards the consanguinity between the volcanics of activity stages taken as pairs (W^2)

Stages	Chemical element	Harghita II	Gurghiu I	Gurghiu II	Călimani I	Călimani II
Harghita I	Al_2O_3	5.86	2.76	0.45	3.41	2.25
	Fe_2O_3	1.77	0.12	1.18	0.40	1.32
	FeO	0.01	0.18	0.00	0.98	3.64
	CaO	1.01 9.48	0.09 11.85	7.03 17.87	0.12 8.72	5.76 13.60
	MgO	0.00	0.46	0.63	0.00	0.10
	Na_2O	0.79	3.12	1.03	2.38	0.04
	K_2O	0.04	5.12	7.55	1.43	0.49
Harghita II	Al_2O_3		0.01	0.88	1.02	0.23
	Fe_2O_3		0.04	0.92	1.13	11.77
	FeO		4.00	2.64	0.75	15.20
	CaO		0.01 13.01	5.40 29.59	1.30 36.40	4.59 44.73
	MgO		0.01	7.39	4.16	11.88
	Na_2O		5.00	0.01	5.77	0.01
	K_2O		3.94	12.35	22.01	1.05
Gurghiu I	Al_2O_3			1.28	0.22	0.51
	Fe_2O_3			0.03	0.09	4.46
	FeO			0.54	0.09	7.43
	CaO			0.07 8.99	0.05 1.31	0.74 28.19
	MgO			1.18	0.67	0.51
	Na_2O			5.85	0.03	6.60
	K_2O			0.04	0.16	7.94
Gurghiu II	Al_2O_3				1.72	0.98
	Fe_2O_3				0.02	9.92
	FeO				1.42	14.46
	CaO				0.23 11.70	2.34 45.04
	MgO				0.72	0.32
	Na_2O				7.01	0.31
	K_2O				0.58	16.71
Călimani I	Al_2O_3					0.03
	Fe_2O_3					6.84
	FeO					9.92
	CaO					6.39 39.84
	MgO					1.32
	Na_2O					7.74
	K_2O					7.60

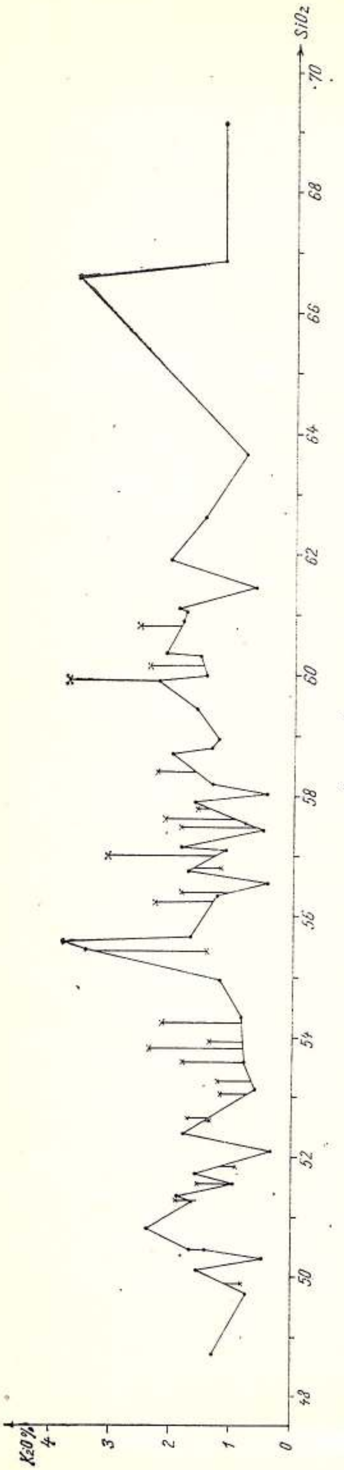

Fig. 2. – Estimation of Z_i value for testing the consanguinity by the W_j^2 function; for (e.g. Călimani Mts volcanic rocks pertaining to stages I and II, considered as pairs).

W^2 and W_j^2 values for cases when the comagmatism hypothesis is rejected or when one element has a different distribution in the rock associations taken into account, 'were figured in a frame.

By contrast with the testing of the similarity of the chemical composition only by samples, pertaining to the common range of silica, there has been considered as useful the checking of equivalence likewise without any restrictions for the silica range; in this mode there are also included the peripheric portions of the differentiation domain where the most specific features of phenomena are very frequently materialized. Consequently, there have been utilized the Wilks' Miller and Kahn, 1962) and Rodionov's (1968) methods applied in various researches relating to the test of the appurtenance of two or several subcollectivities to a general collectivity (subcollectivities of rocks, ores, faunal populations) (Ianovici, Dimitriu, 1968).

The Wilks' V criterion is a logarithmical function of determinants of W and S covariance matrixes, the last matrix being obtained, assuming that the differences among the averages of the chemical composition of the pairs of considered rock associations are non-essential

$$V = -\left(n_1 + n_2 - 1 - \frac{p+2}{2}\right) \ln \frac{|W|}{|S|}$$

where n_1, n_2 = number of chemical values of compared rock associations,

(1) respectively (2); p = number of characteristics (chemical components); $|W|$, $|S|$ = determinants of W and S matrixes.

The V has chi-square distribution with p degrees of freedom. There will be accepted as true the hypothesis on the chemical composition equivalence of rock associations under study for $V < \chi^2_{p\,d.f.;\ 0.95}$

The chemical composition has been represented, as usually by SiO_2, Al_2O_3, Fe_2O_3 + FeO, CaO + MgO and Na_2O + K_2O.

As regards the Rodionov's method the chemical comparison of two rock associations (noted by T_s and T_k) is carried out by means of the $V(T_s, T_k)$ whose distribution is likewise chi-square with p degrees of freedom

$$V(T_s, T_k) = \frac{N-1}{N(N-n_s)n_s} \cdot \sum_{J=1}^{p} \frac{\left[n_k \sum_{t=1}^{n_s} x_{tj} - n_s \sum_{t=1}^{n_k} x_{tj} \right]^2}{\sum_{t=1}^{N} x_{tj}^2 - \frac{1}{N} \left(\sum_{t=1}^{N} x_{tj} \right)^2}$$

where $N = n_s + n_k$

TABLE 9

Testing of the equivalence of the chemical composition of volcanics by the V function (according to Wilks)

		Călimani II	Gurghiu I	Gurghiu II	Harghita I	Harghita II
A	Călimani I	21.42	2.11	3.37	4.70	
	Călimani II		22.54	7.09		26.77
	Gurghiu I			9.31	12.06	49.46
	Gurghiu II				2.93	21.43
	Harghita I					12.37

		Gurghiu I + II	Harghita I + II
B	Călimani I + II	3.34	109.31
	Gurghiu I + II		21.13

NOTE: Chemical compounds introduced into calculations: SiO_2, Al_2O_3, Fe_2O_3 (total), CaO + + MgO., Na_2O + K_2O. ($\chi^2_{5d.f.;\ 0.95} = 11.10$)

$\boxed{V}$ — the hypothesis Ho: $M_{\Xi_1} = M_{\Xi_2}$ is rejected.

The assumption as to the equivalence of the chemical composition of rock associations is admitted as true for $V(T_s, T_k) < \chi^2_{\text{m.d.f.}: 0.95}$. In the calculation of the $V(T_s, T_k)$ function, all the chemical components both the major and the minor taken individually have been introduced [3].

TABLE 10

Testing of the equivalence of the chemical composition of volcanics — major elements — by the $V(T_s, T_k)$ function according to Rodionov.

		Călimani II	Gurghiu I	Gurghiu II	Harghita I	Harghita II
A	Călimani I	**42.95**	**9.69**	21.27	19.82	**67.28**
	Călimani II		58.30	**71.62**	**60.10**	**78.15**
	Gurghiu I			15.14	**25.95**	17.24
	Gurghiu II				**45.60**	**80.09**
	Harghita I					**29.15**
B	Călimani I + Gurghiu I	**49.27**		**20.09**	**22.40**	**66.50**
C	Călimani I + Gurghiu I + II	**68.23**			**30.25**	**88.68**

	Gurghiu I + II	Harghita I + II
Călimani I + II	**37.98**	**64.81**
Gurghiu I + II		**71.88**

Note: Chemical components introduced into calculations: SiO_2, Al_2O_3, Fe_2O_3, FeO, MnO, CaO, MgO, Na_2O, TiO_2, P_2O_5, H_2O.

$\chi^2_{12\ \text{d.f.}: 0.95} = 21.00$

Ho: $M_{\sqsupseteq 1} = M_{\sqsupseteq 2}$

[3] The algorithm and a FORTRAN IV program of the method were given by Zamfirescu, Dimitriu, Chirilă, Dănescu (in print).

The difficulties encounterd in the calculation have limited the applying of the W i l k s' method to the investigations on the equivalence

TABLE 11

Testing of the equivalence of the chemical composition of volcanics-trace elements by $V(T_s, T_k)$ function.

		Călimani II	Gurghiu I	Gurghiu II	Harghita I	Harghita II
A	Călimani I	21.14	13.76	45.03	14.30	133.86
	Călimani II		16.31	46.79	19.94	150.74
	Gurghiu I			16.21	12.54	51.69
	Gurghiu II				30.32	67.25
	Harghita I					48.81

		Căliman II				
B	Gurghiu I + Harghita I	20.44	15.31	26.32		67.05

	Gurghiu I + II	Harghita I + II
Călimani I + II	45.50	169.55
Gurghiu I + II		76.72

Note: Chemical components introduced into the calculations: Pb, Cu, Zn, Ni, Co, Cr, V.

$\chi^2_{7 d.f.;\ 0.95} = 14.10$

$\boxed{V(T_s, T_k)}$ is rejected Ho: $M_{\Xi_1} = M_{\Xi_2}$

of the chemical composition, only for major components of the volcanic products (table 9), whereas the R o d i o n o v's method has been used both for the major components (table 10), for the minor ones (table 11) and for the research according to the type of rocks (table 12).

TABLE 12

Computation of the W_j^2 function, utilizing the Z_i values from Plate I

Z_i	Z_i
−0.30	1.30
0.20	1.15
0.55	0.20
−0.27	0.65
0.00	−0.10
−0.10	1.50
0.30	0.90
0.45	0.25
0.55	0.70
1.00	
1.55	
0.55	
1.30	
−1.95	
0.00	
0.95	
0.70	
−0.45	
1.70	

$$\overline{Z} = \frac{\sum_{i=1}^{n} z_i}{n} = \frac{13.28}{28} = 0.47$$

$$S^2 = \frac{\sum_{i=1}^{n} z_i}{n-1} = \frac{21.89}{27} = 0.81$$

$$W_j^2 = \frac{\overline{Z}^2 . n}{S^2} = \frac{0.22.28}{0.81} = 7.60$$

Relationships among the volcanic manifestations from the Călimani Gurghiu, Harghita Mts

The so far expressed ideas referring to the problem, or those which may be inferred from the carried out research work, are grouped into two categories :

In the framework of the first, the acceptance of a general similarity among the 3 areas (from all the viewpoints) leads to the concept of the simultaneity of volcanic processes ;

In the framework of the second, the admittance of the existence of some differences among the 3 areas leads to the concept of a succession of phenomena, the advance of the volcanism from N towards S.

The analysis of this problem is evidently made more minutely than correspondently of notions ,,province'' and volcanic ,,epoch''; the conclusions that will be reached would not contradict, on the one hand, the existence of these geological units — already well proved — and would not aim, on the other hand, to argue again problems already satisfactorily substantiated.

The general impression emanating from the analysis of the carried out statistical processing is that each of the above 3 areas marks a rather clear-cut individualization. This may be particularly stated în cases when no restrictions as regards the compared SiO_2 range are intervening, because even the ampleness of the latter represents the reflection of the evolution degree of magmatic masses, and this degree is itself an element which is to be compared; from the standpoint of evolution of magmas, the peripheric zones of the SiO_2 range would represent the most interesting and significant parts.

The statistical parameters of the content in major elements (table 3) show clear-cut differences among these 3 areas, in particular, for Fe_2O_3, CaO and K_2O; the significance of differences cannot be attenuated by any of the similitudes of values, which moreover do never concern all the 3 areas (FeO, MgO, Na_2O for Harghita and Gurghiu; Al_2O_3 for Harghita and Călimani; SiO_2 for Călimani and Gurghiu), and consequently are not essential from this point of view. Equally significant there are the variation ranges of the parameter values, so as it may be for instance, stated for SiO_2, from the table 4; the above 3 areas are very clearly individualized. The results of the multivariate analysis also plead in that sense. The W^2 criterion (table 7), as well as the $V(T_s, T_k)$ criterion (table 10) point out essential differences among the 3 areas for the major chemical elements.

Differences among the 3 areas are not obvious only with respect to major components, but, to the same extent, the minor ones; the $V(T_s, T_k)$ criterion points out for this time too, essential differences (table 11).

Another way the differences among the 3 areas are displaying, consists probably, in the modification of the variability degree of different features. The statistical values from tables 3 and 4 suggest a tendency for „stabilization" of this variability from N toward S within the Călimani – Gurghiu – Harghita succession. However, such an observation is not supported by all the data, and within the same parameters contradictory observations may be also forwarded (table 3).

A more minute analysis of various values points out that within these differences, the 3 areas do not display an identical behaviour. All data indicate that the Harghita area occupies a special position as the characteristics of its products may be opposed both to those from the Călimani area and to those from the Gurghiu area.

In some cases the differences between the Harghita and Gurghiu areas and between Harghita and Călimani areas are readily discernible, by contrast with resemblance between the Călimani and Gurghiu areas (Wilks' criterion, table 9). In other cases the situation is not so easily discernible. Thus, most surprizingly with respect to the previous affirmation, the W^2 criterion indicates clear resemblance between the Harghita and Călimani areas and between Harghita and Gurghiu areas (table 5). Nevertheless, as (*a*) the differences among these 3 areas are precisely indicated by other parameters (tables 3, 4, 7, 10, 11), and as (*b*) they suggest a non-simultaneity of volcanic manifestations it may be presumed that in the Harghita area supplementary elements had intervened, which have secondarily created a similitude that is revealed only by some parameters. This supplementary intervention represents itself a characteristic feature for the Harghita area.

The individualization of the Harghita area by means of a subsequent intervention of characteristic elements is also indicated by the fact that the general SiO_2 range is established hardly during the second stage of the evolution, whereas in the Călimani and Gurghiu areas this range is defined still from the first stage (table 4). The same explanation might be given also for the attenuation of the tendency to stabilization of the characters within the Călimani—Gurghiu—Harghita succession (previously mentioned) which in the Harghita area, was displaying also during the second stage of volcanism.

In conclusion it would seem, that after having statistically examined the distribution of the major and minor chemical components of volcanic rocks, we may state that, in the framework of the general similarities defining the petrological province, there exist numerous minute differences among the volcanic products of the Călimani, Gurghiu, Harghita areas which allow to delimit each of them as regards the chemical evolution of magmas. Such a conclusion suggests some discrepancy in time between the volcanic manifestations of the above 3 areas. On the background of differences, which may be considered as having been generated by normal magmatic differentiation processes a somewhat especial position is occupied by the Harghita area where it would seem that a disturbance had occured in the late moments of the magmatic evolution.

Relationships between the evolution stages of volcanism within the 3 areas

Almost all parameters indicate the similarity of the first stages of volcanism from the 3 areas. The Wilks', Rodionov's and W^2

criteria (tables 8, 9, 10) for major components like the $V(T_s, T_k)$ function (table 11) for minor components do completely agree.

The differences among the 3 areas, prove to have been generated during the second stage of the volcanic activity. Most criteria point to the lack of similitude between these stages both as for the major components (tables 8, 9) and as to the minor ones (table 11).

The examination of relationships between the two stages has evidenced firstly the peculiar position of the Harghita area. Here a clear-cut individualization is displayed by the the second stage, which shows the features of a disturbance as compared to the inter-stage relations in the Călimani and Gurghiu areas.

A first observation, the one relating to the definitization of the limits of the SiO_2 range, has been already made (table 4). The peculiar situation of the second stage in Harghita Mts is indicated also by the increase of the $\bar{x}$ value for most major components from the first to the second stage in the Harghita area by contrast with its decrease in the Călimani and the Gurghiu areas (table 3). The spreading of $S_{\bar{x}}$ displays likewise a peculiar behaviour in the Harghita area as compared to the other two areas (table 8). For some criteria the particular position of the second stage of the Harghita area is reflected either in a constant difference between it and the other stages of the 3 areas, by contrast with the variable character of relationships between the latter, or by the fact that a stage appears with affinities with respect to all anothers, excepting the second stage of the Harghita area, with which it marks an essential difference (table 9).

It is noteworthy that for some criteria the values indicate a similarity between the two stages of the Harghita area, but it is only apparent. Thus, for instance, in the case of the W^2 criterion (tables 6 and 8) the differences for SiO_2 do not appear owing to the nature of the criterion, but they exist for Al_2O_3; despite the similarity indicated by this criterion for the global chemical composition, one must consider as more significant the differences for these two essential components.

The so far presented data are not to be considered as indications that the relationships between the stages of the Călimani and Gurghiu areas, being different from those of the Harghita area, are still very much alike between them. In the various cases the relations are not completely homogeneous, but the relationships among the stages, their „filiation" in each of the two areas is out of question. The relations among the stages of the Gurghiu area, show more constancy than those of the Călimani area. The first suggestions in this respect are given by the average values for Al_2O_3, Fe_2O_3 etc. (table 3). However, as very significant may be considered the values of the W^2 (table 8), Wilks' (table 9) Rodionov's (table 10) criteria which are pointing out an obvious relationship between the two activity stages from the Gurghiu Mts.

The examining of relations among the stages of volcanic activity has outlined in addition two general ideas. The first is concerning the occurrence of differences among the 3 areas only during the second volcanic stage, starting from a homogenous background in the first stage. The second is concerning the causes of these differences; for the Harghita area the interference of an external perturbant factor is specified, whereas the existence of ,,filiation'' in the other two areas allows to explain the global differences by the non-simultaneity of processes.

Conclusions

The image of the unfolding of volcanic processes over the area of the Călimani, Gurghiu and Harghita Mts, resulting from the study effected, is essentialy detailed and more precise as compared to the previous ones. The existence of some distinctive features for the 3 areas was already pointed out (Rădulescu, 1963), still the analyses did not tackle the detecting of the mechanism and the causes of these differences. Today we may forward the following assumption which, developed on the basis of chemical data, is consistent with all the already known geological elements.

The magmatic activity over the whole territory is characterized at its beginning by the chemical homogeneity of its products. Thus, it is suggested that the magma masses had not undergone longlasting evolutions within their various subcrustal magma chambers, which might have conferred them independant features. We may rightly consider that the ascent of magmas toward these high levels of the crust did not precede for a long time the beginning of the volcanic activity. The development of the volcanic activity was stimulating — by discharges of thermic and mechanic energy, by extraction of considerable bulks of substance — the differentiation processes; having certainly began still some time before the subaerial activity they marked an accentuation in these moments determining not only the variation of products from this first activity stage but especially creating premises for the variability of the following stage.

If we take the assumption that the initial magmatic material displayed common characters in all the 3 areas, then it may be supposed that the timeinterval of the differentiation was the chief factor responsible for the variability of the petrographical characters during the second stage of activity. Thus the hypothesis is suggested that volcanic manifestations did not simultaneously occur in the 3 areas; it is true that this hypothesis does not present the single possibility to explain the situation, but it corresponds to numerous suggestions given in this sense likewise by the geological characteristics.From the point of view under discussion, it is impossible to make an affirmation as to the beginning of the supracrustal activity. Elements of chemical nature — corresponding to elements of geologic nature — compel us to accept the non-simultaneity of the limit between the two stages and of the closing moments of the activity in all the 3 areas (R ă d u l e s c u, in print). Consequently there would have been periods of coexistence of various stages in these 3 areas, assumption supported by the lack of fundamental opposition from the chemical point of view between the upper stage and the lower one from adjacent areas in the Călimani—Gurghiu—Harghita succession. However, we are not inclined to afford a too large extension those periods of coexistence.

The unfolding of the differentiation processes has led to essential differences among the products of the second stage in all the 3 areas. In the Călimani and Gurghiu Mts the evolutive character of the transition from the first stage to the secon1 one is obvious. Nevertheless, it is most probable that in the Harghita Mts the peculiarities proceeding from the differentiation have been superposed by an external strong perturbation factor. The wide pronounced extension of the SiO_2 range during the second stage, and the constant differences indicated by the statistics of the Al_2O_3 contents from these two stages make us to presume an interference of a silico-aluminous material — may be an assimilation of sedimentary rocks — to constitute the cause of perturbance. This hypothesis corresponds with the existing differences as regards the nature of the basement between the southern outermost part of the eruptive chain, on the one hand, and the median and northern sectors, on the other hand (R ă d u l e s c u, this volume).

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RELATIVE PARTICIPATION OF SAMPLES (n%) WITHIN THE COMMON (b), RESPECTIVELY LOWER (a) AND HIGHER (c) SiO_2 RANGE ON PURPOSE TO TEST THE CONSANGUINITY OF ROCK ASSOCIATION PAIRS

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CONSIDERATIONS ON THE ORIGIN OF MAGMAS OF THE NEOZOIC SUBSEQUENT VOLCANISM IN THE EAST CARPATHIANS

BY
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A problem of major interest for the geology of the East Carpathians — the origin of magmas, whose subaerial or subcrustal consolidation gave rise to the volcanic zone — was but rarely tackled. H. Stille (1953) is practically the single scientist who has discussed this problem with more details, and has endeavoured to prove the exclusively lithogenous origin of Neozoic magmas from the Carpathians. His conception was later adopted by most investigators of the above region, this standpoint corresponding with the general tendency displayed by the evolution of ideas on the origin of magmas in the orogenic region (1940—1960). Concomitantly with the accumulation in these last years of observational data relating to the Romanian Carpathians, the acceptance of this hypothesis has met with ever more difficulties; without denying the local formation of magmas by melting of pre-existent rocks, we have the impression that nowadays the question ,,how were generated magmas in the course of the Neozoic within the Romanian Carpathians ?'' is to be otherwise answered. Almost unanimous in considering the melting as the single process for the formation of andesitic magmas, the researchers of the last decades had nevertheless essentially different opinions even as for the main features of this phenomenon; the differences of opinions have always reflected the chief difficulties encountered in the working out and acceptance of this hypothesis even by its most devoted supporters.

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These last years some doubts that many researchers have had as regards the lithogenous nature of andesitic magmatism were confirmed. It is sufficient to mention a situation as that from the northern part of the central America, where the impossibility to accept such a hypothesis has been recently proved (W e y l, 1966, 1967). The volcanism which had developed here during the Neogene – although showing petrographical and chemical features of a subsequent magmatism – does not correspond with any close in time orogenesis to which it could be referable. Its development over an oceanic basement, the sialic crust lacking, and the absence of any indications with respect to the sinking of more recent sediments, are excluding the possibility of a lithogenous nature for magmas. Very strong arguments for the interpretation of the genesis of andesites have been then obtained owing to the investigation of the distribution of trace elements both in andesites and in other volcanic rocks (D o e et al., 1968; T a y l o r, 1968). The massive presence of andesites during the geosynclinal magmatism, displaying characters perfectly corresponding to those of rocks from the subsequent volcanism, is also particularly significant (D i c k i n s o n, 1964); in such cases too the lithogenous formation of andesitic magmas seems to be out of question. Later on numerous andesite occurrences, without any connection with the sialic crust, have been identified outside the orogenic regions (K u n o, 1968).

Observations of this nature are as yet relatively scarce but it is obvious that the problem relating to the origin of andesitic magmas is to be reconsidered. Today attemps are made to specify criteria which would permit the separation of lithogenous andesites – whose existence is not out of question – from the hypogene ones. The study of the distribution of trace components, and that of the isotopical composition of some elements leads not only to significant results but, occasionally to contradictory ones. It would seem that, as yet, solely the geological criteria may be considered as having a general validity, whereas the other ones may by useful according to the case under study.

However, it is doubtless that the most important element in discussing this problem would be today the plate tectonics concept. Although the mechanism of the unfolding of processes and their details are far to be as yet known, nevertheless it would seem that no impediment of geological and geophysical nature should exist as to derive volcanic phenomena in orogenic areas from the subduction of oceanic crust. The immediate implication resulting from the acceptance of this hypothesis consists essentially in admitting a deep-seated source for andesitic magmas with eventual contaminations during the ascent and stops at intermediary levels of the crust.

The reticences that the researchers of the Neozoic volcanic regions from Romania have had as regards the lithogenous origin of magmas, became more accentuated these last years concomitantly with the accumulation of observational data; further on we will present some principal aspects of the problem where the incompatibility with the hypothesis relating to the lithogenous nature of the andesites is obvious. Partially they do correspond to a rather rich observational material, and partially they represent only comments of a quite general character. Owing to the nature of this problem a part of considerations hereafter do not only refer to the Călimani—Gurghiu—Harghita area, but may be also applied to the other regions with Neozoic volcanism from the East Carpathians.

Association of andesites with basalts

Although the products of the subsequent igneous activity cover in Romania the whole range of petrographical types characteristic of calc-alkaline magmas, its specific feature is a clear-cut predominance of andesites over rhyolites, dacites and basalts. This predominance is mostly conspicuous within the Călimani—Gurghiu—Harghita area where there are practically present exclusively andesitic rocks and their extremely basic forms. It is to be emphasized that, here the basalts are perfectly typical from all points of view; if we also take into account the fact that this area is partially located along the trajectory of a deep fracture — on which have raised, at least, in some moments, basic magmas, which have generated basalts (Rădulescu, 1969) — then the presence of the hypogene magma is out of question. To what extent, however, the andesites might be its products?

The chief observation to be made is that a large part of andesites display pronounced basic features; the silica content may be of 51 per cent without any essential petrographical changes, whereas occasionally the existence of olivine does concretize also from the mineralogical standpoint, their basic character. Such rocks are abundant in more recent activity phases, however, they never lack in the oldest ones. They are described in the Romanian relevant literature under the name of basalt-like or basaltic andesites. Both the basalts proper, and particularly the basic forms of andesites are present in the whole Călimani—Gurghiu—

Harghita area. The chemical composition and the areal distribution of basalts and basalt-like andesites justify the presumption of their genetical relationship.

On the other hand, however, the basalt-like andesites cannot be separated from the andesites s.s. which build up the most important mass of volcanic products; the chemical, petrographical, geological features display their close relationship. The petrochemical analysis of the whole petrological province has proved that one of its essential features is that the basic and intermediary domains are closely related, whereas the acid domain appears as separated from the intermediary one (Rădulescu, 1961, 1963). Thus the products with an intermediary and basic composition prove to have a genetically common nature. The coexistence of andesites and basalts within the same petrologic province is occasionally used as an argument for the acceptance of their common genesis (Kuno, 1968); the situation in this region is more favourable since their relationship is chemically and petrologically proved.

Chemical composition of volcanics and their evolution

The numerous results, obtained in the research on the chemical composition of rocks, are likewise significant. Although the lack of some precise criteria in order to separate the hypogene material from the lithogenous one prevent from obtaining certain conclusions, some comments can be forwarded relying on the very abundant chemical material.

In the case when magmas were formed by melting of preexisting rocks it might be expected that the ,,non-magmatic'' nature of the initial material has been to some extent reflected in final products, even if its complexity and the subsequent evolution of the melting would have determined the gain of new characters. Really, both the distribution of major elements and that of trace elements tally quite satisfactorily with the fundamental features of the hypogene magmas; although as yet insufficient in number, nevertheless particularly the results of researches on the distribution of trace elements lead to such conclusions (Rădulescu, Stiopol, 1964, Ianovici et al., 1968); for instance, the high Ni-content in all the rocks is to be stressed (Peltz, Bratosin, 1971).

The petrochemical characters of the Neozoic magmatic province in the innerside of the Carpathian Arc over the territory of Romania, have been examined in details (Rădulescu, 1963; Rădulescu, B o r c o ş, 1968), and some data of this nature may be used in discussing the genesis of magmas.

The evolution in time of the composition of magmas has broadly developed from acidic to basic, and we presume that this fundamental character of the magmatic province is inconsistent with the hypothesis on the formation of magmas by fusion. If magmas would be lithogenous, they would have undergone, after their genesis, differentiation processes there resulting more and more acid products. Accepting as indiscutable the fact that the basalts and — according to the above expounded reasoning — the basalt-like andesites would represent the hypogene magma which closed the subsequent igneous activity, it would be necessary, that at least until the occurrence of basalt-like andesites, this evolution trend of the chemical composition of magma should be observed. Really the Neozoic magmatism starts with rhyolites and evoluates through dacites up to normal andesites, even before the occurrence of basalt-like andesites. On the other hand, there is no ground to presume that at the beginning of the subsequent volcanism, magmas were exclusively of lithogenous nature, and later on the possibility of a hypogene supply to have occured. The hypogene character of basalts being doubtless, it is more normal that this mechanism of genesis to have been accepted for the whole subsequent period.

Giving up the hypothesis on the lithogenous character of magmas, the chemical composition may be explained by the succession of supplies of magma from depth in varied contamination and differentiation stages. The differences which were observed among various parts of the Călimani—Gurghiu—Harghita area (R ă d u l e s c u, D i m i t r i u, this volume) are perfectly consistent with this way to envisage things.

Presence of metalliferous ore deposits

In the Carpathians, important ore deposits of native gold and silver, lead and zinc sulphides (as well as subordinately some other elements) are associated with the subsequent magmatism; they display homogeneous features over the whole area.

The possibility to separate the metallic elements from the pre-existing rocks submitted to a melting process, and their concentration up to the form which would permit the genesis of ore bodies is certainly possible. In our opinion it would, however, seem that in the case under discussion, most of the features of the occurrence, do rather correspond to a hypogene source of material. Particularly the constancy as to the occurrence of ore deposits, and the constancy of parageneses and of main chemical elements are facts that could be but hardly explained in the case when the formation of magmas would has taken place by the meltings of a preexistent material; the latter would have doubtlessly presented essential variations in its composition within regions as extended as those affected by the Neozoic magmatism. On the contrary, the uniformity of the chemical features of hypogene magmas — even if they were separated in numerous chambers showing a different evolution — provides a much more fitting explanation for the existent situation.

On the other hand, even in light of the rather uncertain data at our disposal on the unfolding of intratelluric magmatic phenomena the factor ,,time'' may be discussed. It would seem difficult to prove that the time interval comprised between the presumed fusion of pre-existing rocks and the appearance of ore deposits, has been sufficiently long for the development of the differentiation and concentration processes which would lead to ore forming fluids.

Petrographical characteristics of zones of presumed melting

The Neozoic subsequent magmatism has shown a particularly homogeneous character along the Carpathians Mts, both over the Romanian territory and outside it; the petrochemical and petrographical variation is achieved within normal limits without affecting this homogeneity. Such an affirmation may be forwarded, as previously stated with respect to the associated ore deposits too. On these grounds it is to be presumed that the material from which the rocks have formed had shown the same chief features in its whole mass. Such a presumption is perfectly consistent with the hypothesis of a hypogene magma; nevertheless it would be necessary to prove, in a particular way, that the lithogenous processes would have allowed to obtain magmas with so homogenous features in such an extended region.

If we should examine, no matter how summarily, the information at our disposal about the deep zones, which would had been submitted to melting along the Carpathians — Romania, USSR, Czechoslovakia, Hungary — the existence of essential petrographical differences could be stated : although, the melting processes are to be envisioned as affecting large masses of rocks and therefore, with the possibility of totalizing various chemical features, it would be normal that in resulted magmas, some of the original differences should reappear, even under attenuate forms.

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The ideas commented here, represent in our conception elements of inconsistency between the hypothesis on the lithogeneous nature of magmas and the actual situation in the Romanian Carpathians; today no decisive argument in our opinion could be forwarded to support this point of view. It is much more probable that the origin of large masses of andesitic material is to be looked for in the modifications undergone by the hypogene magma as a result of the differentiation processes, and eventually the contamination ones.

Recently an attempt aiming at the interpretation of the structure of the Carpathian area in the light of the plate tectonics concept (Rădulescu and Săndulescu, 1973) was made. The fact that the totality of geological features may be satisfactorily framed in this hypothesis, as well as the fact that the decisive part of the underthrust processes in the building up of the structure of the Carpathians had been long ago recognized, have logically led to accept the subduction of an oceanic-type crust from E towards W beneath the present structure of the Carpathians; it would have constitute the source of basic material, which in the course of the ascent, due to the contamination and differentiation processes, would have become magmas released in the framework of subsequent volcanism from the innerside of the Carpathians (Stille, 1953, presumed the sinking of sialic masses and the generation of magmas by their melting). Although it is difficult to estimate to what extent the magmas were contamined we presume that from the quantitative viewpoint the sialic material has always remained subordinated, and even if it has occasionally determined readily recognizable modifications did not succeed to affect the fundamental features of magma and its homogeneity. The existing petrographical variation — andesites of diverse types, dacites, rhyolites — is due first to differentiation phenomena, and to a less extent, to qualitative and quantitative differences as for the assimilated material.

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TENTATIVE PALEOGEOGRAPHICAL RECONSTITUTION OF THE CĂLIMANI–GURGHIU–HARGHITA AREA DURING THE NEOZOIC VOLCANIC ACTIVITY

BY

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Both the general lines and some details of the paleogeographical evolution of the Călimani–Gurghiu–Harghita Chain have been already cleared up (Rădulescu, 1968; Rădulescu, Peltz, 1970). The main still existing uncertain points are chiefly due to the impossibility to state the precise moment of the beginning of the volcanic activity.

All attempts related to the dating of the volcanic activity have so far represented mainly geological interpretations of a general nature, and to a less extent, they rely on objective information specific to this region. The examining of the stratigraphical position of the cinerite horizons from adjacent regions could not lead to conclusive results; if we exclude the Transylvania Depression — in whose deposits the pyroclastic material supplied by the eastern source (Călimani, Gurghiu, Harghita Mts) cannot be separated from that of the northern (Oaş–Gutîi Mts) and western sources (Apuseni Mountains) — outside the Carpathian Arc, the situation presents contradictory aspects. Within the Moldavian Platform deposits, the cinerite horizons are lacking on ante-Meotian deposits (excepting the horizon identified at Hudeşti whose significance cannot be, however, considered as clear-cut) fact that would correspond with the information recently provided by Anton Popescu[2] on the beginning of the volcanic activity in the Călimani–Gurghiu–Harghita area. However, cinerites are present and rather frequent in the deposits of the inner foredeep, both in the Eastern Carpathians and in the Getic Depression

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[2] A. Popescu (1966) Studiul mineralelor grele din depozitele pannoniene situate între valea Mureşului şi valea Gurghiului Manuscript Arh. Inst. Geol.

in deposits of Sarmatian, Tortonian, Helvetian and even Aquitanian age; which is their source? Which is the significance of their distribution only in the bend region of the Carpathian Arc and in the Getic Depression? Their concentration in the bend area of the Carpathians – if we would exclude from our discussion the Getic Depression, where the intervention of materials from other sources is quite probable – would seem to indicate the absence of a relationship with volcanic phenomena whose products are nowadays conspicuous in the Călimani–Gurghiu–Harghita Chain; their presence here in deposits of such ages, and even older, has already led to some speculations relating to the possibility of the existence of eruption centres in this region (Filipescu, 1944). However, on the other hand, the existence of cinerites within deposits Meotian in age from the same region is to be related to the activity of the Călimani–Gurghiu–Harghita Chain; hence must we reach the conclusion that the older cinerites too proceeded from the same source or, on the contrary, must we consider that in the course of time other sources as yet not evidenced had existed?

The only objective data which might substantiate a conclusion are as follows: (*a*) within the Pannonian s.s. deposits in the neighbourhood of the Călimani–Gurghiu–Harghita Chain the volcanic constituents are completely absent beneath the E Zone (A. Popescu, op. cit.), and (*b*) the absolute ages of about 7 m.y. for rocks pertaining to the basal part of the upper compartment (Rădulescu, in print). In the light of these observations no connection, not even with the cinerites of the Lower Pannonian age from the Transylvania Depression (Bazna cinerites) nor with those from the outer side of the Carpathian Arc, in the Getic Depression, could have been presumed. Really, how could be explained the fact that at a large distance ash tuff deposits displaying essential thickness have accumulated, whereas in the close vicinity of apparata, volcanogenous components either pyro- or epiclastic are completely lacking? Since the object of our today researches is the observable volcanic material, conspicuous in the Călimani–Gurghiu–Harghita area, we will hence rely only on it for our conclusions; any connection with ante-Pannonian pyroclastics from neighbouring regions seems to be so far but mere speculations.

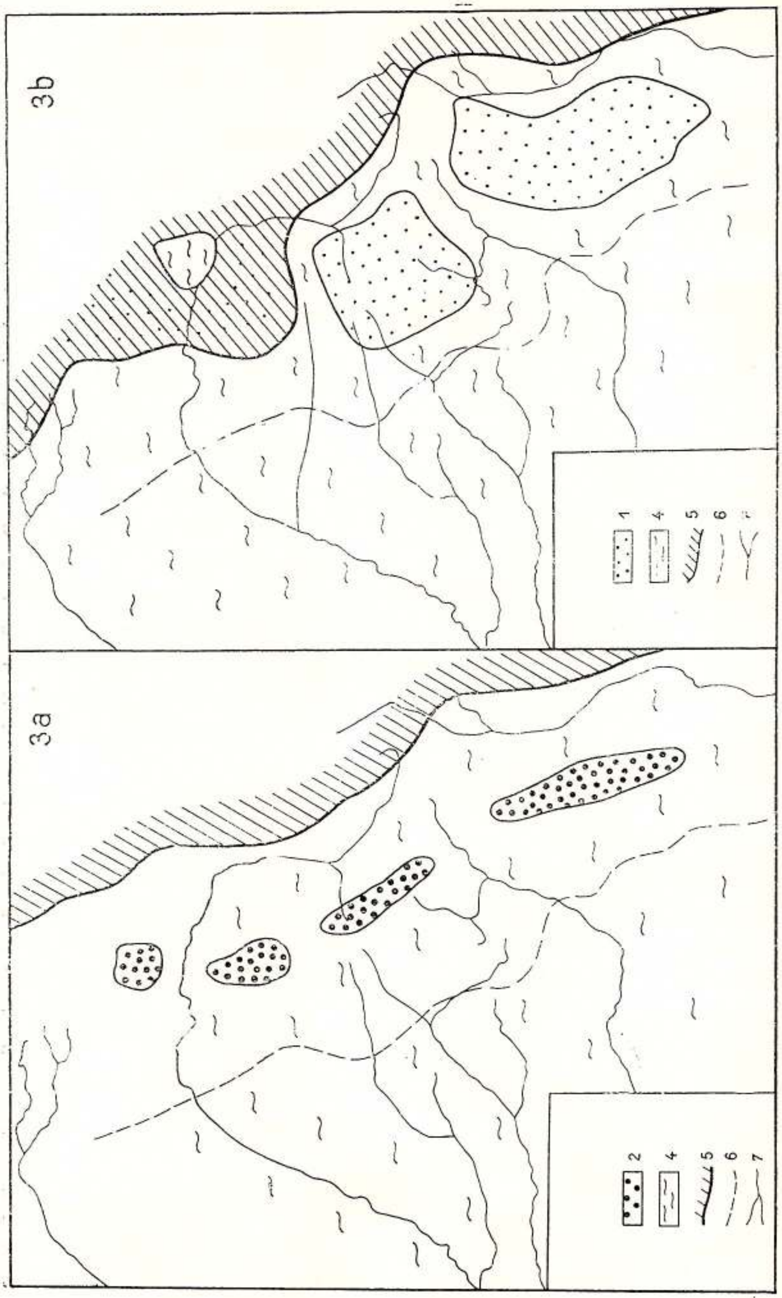
Nevertheless, in this discussion the interest for dating, the beginning of the volcanic activity is not only chronological, but particularly paleogeographical. The latest reconstitutions for the whole territory of Romania (Lithofacial map 1:2,000,000–1969) have pointed out that in the course of the Neogene there had existed a single short time interval when the waters of the Pannonian Lake have covered the region of the Călimani — Gurghiu — Harghita Chain, as far as the axial zone : Pannonian s.s. ; during the rest of the time it was a part of the land which separated the Pannonian Basin from the Dacic — Getic one. Hence, if we admit the beginning of the volcanism as having taken place in the initial or middle part of the Neogene time, we should be bound to imagine it as starting with a subaerial activity, whereas if we admit the beginning of processes as having occurred in the course of the Pannonian, they might initially have had a subaquatic character ; the latter is the viewpoint we expounded in previous papers (R ă d u l e s c u, 1968), and which in our opinion seems to be so far justified.

The Pannonian [3] is the time when the waters of the Pannonian Lake had extended eastwards covering — with exception of a small area in the northern part — the whole territory nowadays occupied by volcanic products. Relying on the argument yielded by A. P o p e s c u as to the occurrence of volcanic elements in the Pannonian deposits, we may connect the beginning of the volcanic activity with this moment. Other information as yet not generally valid may be also referred to in this sense ; it is for instance the case of volcanic rocks with features of subaquatic consolidation, pointed out in the northern part of the Călimani Mts [4].

The starting of the activity and the building up of the first volcanic structures were sudden and rapid (plate, moment 2). A row of volcanic islands has appeared nearby the eastern border of the lake ; probably more numerous at the beginning, they have finally formed some more important land masses separated by the arms of the lake (plate, moment 3a). If the localization of the piercement points was approximately preserved during the two phases of the volcanic activity, it may be expected that at least within the regions of the present Mureş, Gurghiu, Tîrnava Mare valleys a connection between the peripheral part (eastern) of the lake and its central part would have been achieved (fig. 1, moment 3a)

[3] Owing to lacks in our tknowledge of the absolute chronology of volcanic phenomena, special attention must be paid to the succession of events and not to their exact dating.

[4] C o s m a S t a n c i u, O. G h e r u c i (1958) Raport geologic asupra lucrărilor de cartări şi prospecţiuni în Mţii. Călimani — Bîrgău (Dornişoara — Fîntînele) Manuscript Arh. Inst. Geol.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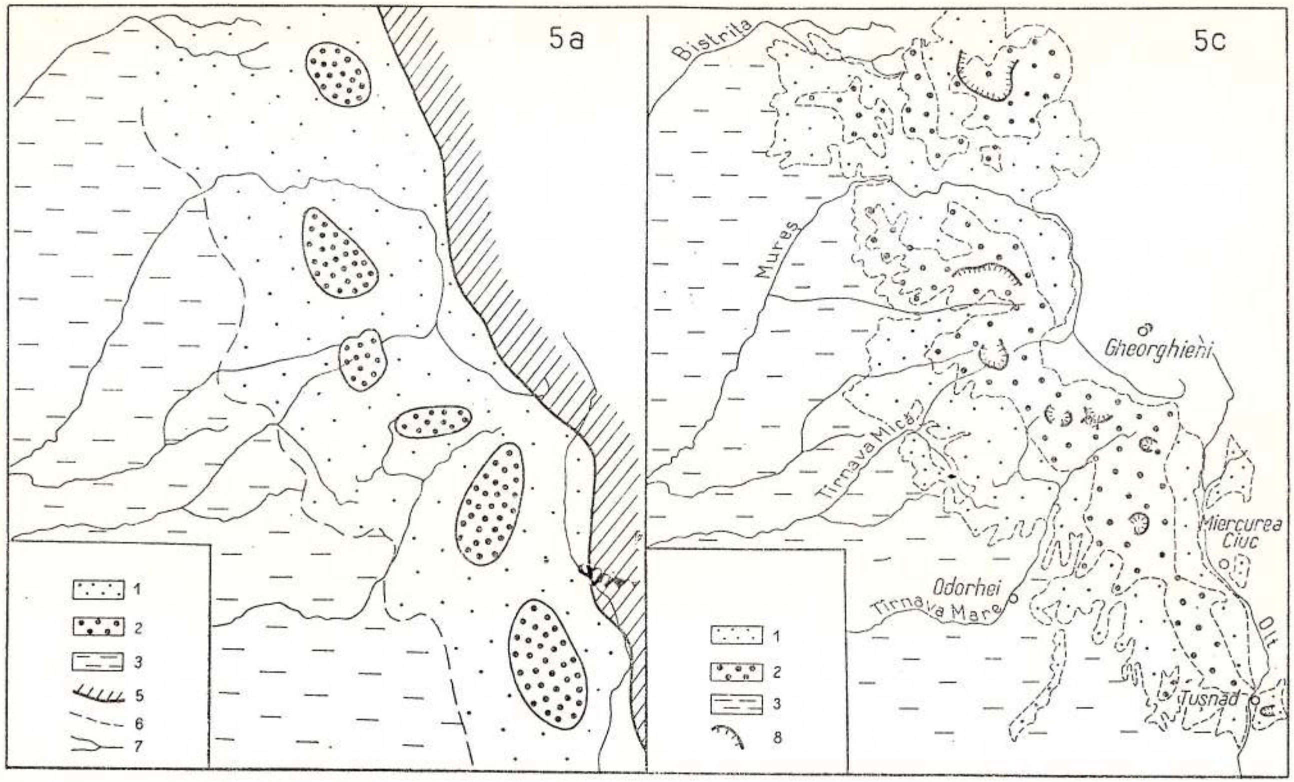

Fig. 1. Paleogeographical reconstitution for the chief moments of the geological evolution of the region (see plate).
1. Volcanic and volcano-sedimentary formations. 2. Volcanic formations. 3. Sedimentary formations. 4. Surfaces covered by waters. 5. Shore line of the old lake. 6 Rupture of slope in the lake bottom. 7. Present-day hydrographical network. 8. Main preserved volcanic apparata.

The phenomena thus imagined, one may presume the start of the volcanic activity had taken place in the middle or in the basal part of the Pannonian s.s., with a weak implication in the sedimentary deposits of the adjacent regions, probably due to the subaquatic character of processes. Hardly, at the limit between the C/D and E zones, the emersion of volcanic apparata and the occurrence of a strongly explosive character have determined the appearance of volcanic components in the sedimentary deposits.

The character of the subaerial volcanic activity has been at its beginning extremely explosive. The abundance of cinerites within Meotian deposits from the outerside of the Carpathian Arc constitues a conclusive proof; both the possibility of interference for other sources of pyroclastic material and the uncertain points existing as to the knowledge of the stratigraphy of Pannonian deposits from the Transylvania Depression do not allow us to identify here this moment.

If we take into account the bulk of epiclastic material, occurred subsequently as a result of the erosion of volcanic superstructures together with the pyroclastic one proceeding from the explosive destruction of the latter, we have to reach the conclusion that the sizes and number of volcanic apparata have been very large. The destruction of volcanic superstructures has probably begun very soon after their building, and the material resulted has constituted in its close vicinity the volcano-sedimentary formation. The surface of volcanic islands has considerably increased, in some regions their joining to the land zone and the forming of small-sized intermountain lacustrine basins having probably taken place (plate and fig. 1, moment 3b).

The genesis of the volcano-sedimentary formation cannot be conceived without the existence, at least partial, of an aquatic environment; if the destruction of volcanic structures was caused by a subaerial erosion, the deposition of the epi- and pyroclastic material has occurred, to a great extent, under subaquatic conditions. The deposits from the basal and middle parts of the volcano-sedimentary formation comprises horizons whose subaquatic sedimentation my be considered as doubtless; this fact signifies that its building had begun previously to the retreat of the Pannonian Basin waters when either the waters of this basin or locally those of smaller basins closed by volcanic heights covered this region. On the other hand, the petrographic features of deposits from the upper and partially middle part of the volcano-sedimentary formation ascertain that the processes have been developing in a subaerial regime.

The destruction of volcanic apparata and the genesis of volcano-sedimentary formation had presumably occurred during a rather long time interval in comparison with the previous stage of volcanic activity. The decreasse of the intensity in the volcanic activity, the intensification of the erosion and accumulation of volcanic detritus have taken place progressively until a moment of some equilibrium was reached. If we would attempt to evaluate the time interval for which available indications as to the presence of a lacustrine regim are at our disposal — from the beginning of the volcanic activity up to the middle of the sedimentation of volcano-sedimentary formation — we could state that it had been longer than it is admitted by the present day paleogeographical reconstitutions. If the volcanic activity had begun under its peculiar forms concomitantly with the sedimentation of deposits from the base of the E Zone, fact that in our opinion is proved, the middle part of the volcano-sedimentary formation cannot tally with the uppermost one of the E Zone (the transition from the Pannonian s.s. to the Pontian) when, according to the Lithofacial Atlas 1 :2,000,000, a complete filling of the lake had taken place. All these processes have certainly required a longer time interval than that corresponding to the E Zone; although we cannot estimate all the arguments of a stratigraphic nature, nevertheless, we have the feeling that the hypothesis as for the preservation of the lacustrine regime in the region of Călimani, Gurghiu and Harghita Mts must be accepted also subsequently to the end of the Pannonian s.s.

The volcanic activity during the genesis of the volcano-sedimentary formation is characterized by the fact that — besides the general decrease of its intensity — from the production of large amounts of cinerite material it will pass to the supply of coarse material and, to a lesser extent, of lavas thus revealing a modification in the type of activity. In the interval between the Pannonian s.s. and the Quaternary the cinerite material is almost completely lacking in the deposits found outside, as well as those inside the Carpathian Arc; very large masses of coarse pyroclastics are, however, accumulated over the territory of the Călimani—Gurghiu—Harghita Chain.

The areal distribution of deposits pertaining to the volcano-sedimentary formation is very significant for the understanding of the accumulation mode of the clastic material. The distances of its migration have no probably exceeded some 40 km, being in most cases much more reduced. The clear-cut limit of the volcano-sedimentary formation westwards, and the fact that it is corresponding today to a conspicuous rupture of slope, point out that even from the beginning (in the moment of the subaquatic regime), the spreading of the volcanic material, particularly westward has occurred up to a practically linear limit, parallel to the axial zone of the mountain chain. It is most probably that the loss of the transport capacity of lake waters was determined especially by the decrease of the bottom slope and that the present western limit of the volcano-sedimentary formation is marking the westward extension of the quasi-horizontal region of the bottom. The gradual decrease of depth in the lake and the forming of a horizontal bottom in its peripheral part by accumulation of volcano-clastic material have contributed to maintain a low transport capacity of waters, and hence the dispersion limit of particles. Thus the region covered by the volcanogenous material has undergone a continuous height increase stressing the initial subaquatic rupture of slope. The accumulation process has continued displaying the same general features in subaerial conditions determining the formation of a plateau sharply limited westward, but eastward joining, to a great extent, the lower parts of the mountainous zone consisting of crystalline schists (plate, moment 4 b). The draining of waters from this zone was made directly westward, towards the central part of former lake, through valleys carved into the surface of the plateau; however, it is not out of question, that in some points of this plateau prominences which represent obstacles in the east-westward way of the waster streams had been preserved. The end of the building of the plateau corresponds to a short break in the volcanic activity and the transition to a new type of activity.

The resuming of the volcanism took place under exclusively subaerial conditions, probably by the re-opening of the same conduits for magmas in the axial region of the newly formed plateau. Owing to a dominantly effusive activity some large-sized apparata have grown. In some points the emissions of lavas came sooner to an end determining a more simple petrographic constitution and allowing the beginning of erosion. In most cases the lava emissions — with essential mineralogical changes — have continued a longer time period leading to the occurrence of a mountainous zone with altitudes exceeding 2000 m.

As in the previous phase—and maybe even more pronounced—at the beginning isolated apparata or groups of apparata (plate and fig. 1, moment 5 a) did occur, which only subsequently were joining together either by their products or by newly formed apparata leading to the formation of the mountain chain. The hydrographical network concentrated in some more important water streams, which passed westward among the previously appeared volcanoes; later on, the connection with the central part of the Transylvania Basin is completely interrupted, and between the volcanic mountain chain and the Crystalline—Mesozoic Zone, intermountain depressions wherein lakes occur, have formed.

The closing of the volcanic activity is proceeding in climatic conditions favourable for an abundant vegetation, which has attenuated the erosion processes. The present day aspect of the region is due to the occurrence of some valleys cut into the mountain chain and through which waters of the intermountain basins were drained west- and southward (plate and fig. 1, moment 5 c).

The table totalizes the varied so far presented informational data; as it may be stated, in the evolution of various groups of considered phenomena discontinuities of different nature are individualizing.

From the viewpoint of the paleogeographical aspect there exist, for the period under study 4 main discontinuities — determined by: (*a*) occurrence of volcanic proeminence on the bottom of the lake in its peripherial eastern part, (*b*) occurrence of the archipelago of volcanic islands, (*c*) retreat of waters and setting in of the subaerial regime (*d*) disappearance of the mountainous volcanic relief over this land, — and 4 secondary discontinuities within the 3 last stages of the paleogeographical evolution of this region. They correspond — as it might be espected since it deals with determinant relationships between these phenomena — to the main discontinuities in the evolution of the volcanic activity and in the evolution of the genesis environments of geological formations over the territory under investigation. The same wide categories of phenomena have determined the evolution of the supply with clastic material (sources) whose discontinuities are likewise tallying with those of the paleogeographical evolution.

The 5 stages which may be distinguished in the paleogeographical evolution of the region comprise the time interval from the beginning of the Pannonian s.s. and until nowadays: the only age limit whose position in the evolution of the volcanic phenomena may be accepted with a satisfactory degree of certainty is that between the C/D and E zones of the Pannonian. The stages are defined by the distribution of waters and land areas as well as by the relief of the territory. The general sense of the evolution consists in the growth of the land on the expense of an area initially covered by waters, as a result of occurrence of volcanic apparata and of filling of the basin. The forms of relief mark a rapid evolution in this time interval corresponding to a „formation – destruction" cycle and covering the first half of a second one. The schematic profiles from the table attempt to suggest images which we could form as to the aspect of the region in the various stages of its evolution.

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SUCCESSION OF GEOLOGICAL EVENTS IN THE CĂLIMANI - GURGHIU - HARGHITA AREA DURING THE VOLCANIC ACTIVITY

DAN RĂDULESCU. Tentative Paleogeographical Reconstitution of the Călimani - Gurghiu - Harghita Area during the Neozoic Volcanic Activity

Geological time	Character of the genesis environment	Type of volcanic activity	Nature of material from sedimentary and volcano-sedimentary formations	Paleogeographical aspect	Profiles W - E	Moment of evolution
Present	subaerial (+ subordinately fluviatil)	lack	lack	land: mountain chain; intramountain depressions filled up		5 c
		subaerial dominatly effusive		land: mountain chain; intramountain depressions with lacustrine basins		5 b
				land: isolated mountains		5 a
		lack		land: plateau		4 b
		subaerial strongly explosive	volcanic epi - and pyroclastic	land: irregular relief		4 a
	subaquatic + subaerial			area of islands in increase; altitude in decrease; depth of lake in decrease;		3 b
Pannonian s.s E zone			pyroclastic + subordinately metamorphic	archipelago of volcanic islands		3 a
Pannonian s.s D zone	subaquatic (lacustrine)	subaquatic effusions and explosions	metamorphic + sporadically volcanogenous	subaquatic prominences		2
		lack	metamorphic	peripherical zone of the Transylvanian lake with low dipping and regular slope of botton		1

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