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K/Ar AGE DETERMINATION OF NEOGENE VOLCANIC ROCKS FROM THE GUTAI MTS. (EASTERN CARPATHIANS, ROMANIA)

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Abstract: K/Ar analyses were carried out on unaltered samples of volcanic rocks from the Gutai Mts., of Romania. The analysed samples, collected from the surface locations from the whole area represent lava flows. According to the previously performed geological mapping and existing lithostratigraphic relationships, the volcanic activity in the area developed during Badenian - Dacian time, (Borcos et al. 1973), with several eruption phases. Most of the volcanic rocks were considered Pontian and post-Pontian in age with a probable extension towards the Pliocene time (Borcos et al. 1973; Lang 1975). The newly obtained apparent ages range between 9 - 14 Ma printing out the andesitic volcanism in the Gutai Mts., started at the Badenian - Sarmatian boundary and unquestionably continued until the Upper Pannonian.

Key words: Eastern Carpathians, Gutai Mts., volcanic rocks, Miocene K/Ar ages, isochron, excess argon.

Introduction

The investigated area includes the whole Gutai Mts., (from the NW part of the Eastern Carpathians), which belong to the Neogene volcanic chain in the inner part of the Carpathian orogen. The region contains sedimentary rocks that, on the basis of paleontological data are Miocene in age. In some cases the Neogene volcanic rocks overlie older sedimentary sequences, or do not show any biostratigraphic relationships. Therefore their age can be obtained only by using radiometric dating.

However, several samples of fresh volcanic rocks were recovered from biostratigraphically well defined sequences (e.g. Sarmatian pyroxene andesites, Pannonian quartz bearing andesites). K/Ar dates on these samples offer a unique opportunity to calibrate the Paratethyan Neogene time scale.

Between 1974 and 1985 in the laboratory of the IFIN Bucuresti several K/Ar determinations were performed on fresh and hydrothermally altered rocks from the Gutai Mts. Some of these were published by Edelstein et al. (1977). The newly obtained data compared with the former ones show frequently significant differences.

Geological setting

In the Gutai Mts., the crystalline basement is situated at a depth of about 2000 m. It is overlain by Paleogene flysch deposits, of about 1000 m thick, belonging to the Transcarpathian Flysch.

The Miocene Molasse formations belong to the Central Paratethys and transgressively overlie the Paleogene flysch.

The presence of Badenian, Sarmatian and Pannonian stages were recognized on the basis of molluscs, foraminifers and/or ostracods fossil assemblages. The youngest deposits, with fresh water molluscs or fossil plants have been preliminary assigned to the Pontian and Dacian.

The volcanic rocks belong to the calc-alkaline series (Kovacs et al. 1992) and are represented by basalts, basaltic andesites, andesites and dacites; the andesites and basaltic andesites-especially the pyroxene ones-being predominant. The lava flows with interlayers of pyroclastic deposits and volcano-sedimentary sequences are widespread (Borcos et al. 1993; Edelstein et al. 1992).

As for mineralogical composition, the salic phenocrysts are exclusively represented by plagioclases (An 45-75) in the case of andesites and some pyroxene dacites, and plagioclases and quartz in quartz andesites and biotite dacites. The mafic phenocrysts are pyroxenes (orthopyroxenes and clinopyroxenes) present in all rock types and hornblende and biotite in some andesites and dacites. Glassy varieties occur especially in the case of dacites.

As for the chemical composition, the SiO_2 content varies in a large interval of 52.6 - 68.8 %, and the K_2O content lends a medium-K character (Kovacs et al. 1987).

Analytical techniques

Measurement of K/Ar ages was carried out in the Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI) Debrecen, Hungary.

Secondarily altered specimens were eliminated by microscopic inspection of thin sections. Heavy liquids and magnetic separation were used for obtaining mineral concentrates. The samples were degassed in a conventional extraction system using induction heating and measured by mass spectrometric isotope dilution with an ^{38}Ar spike using a mass spectrometer (magnetic sector type of 150 mm radius and 90° deflection) in the static mode. Recording and evolution of the Ar spectrum were controlled by a microcomputer. Potassium analyses were made using standard flame photometric techniques. K and Ar determinations were checked regularly by interlaboratory standards LP-6, GL-0, HD-B.1 and Asia 1/65.

Atomic constants suggested by Steiger & Jäger (1977) were used for calculating age. All errors represent one standard deviation (i.e. 68 % analytical confidence level).

Details of the instruments, the applied methods and results of calibration have been described elsewhere (Balogh 1985).

Methods

For valid age determination, it is necessary that at the time of formation of the rock, when the radioactive clock was set, the system contained no extraneous ^{40}Ar and subsequently remained a closed system accumulating radiogenic ^{40}Ar, i.e. the system did not gain or lose either ^{40}Ar or K. The validity of this assumption depends strongly on the geological conditions and each case must be decided separately. Thus it is essential to study a number of rocks in order to reach a significant result. On the other hand, the analytical ages can be regarded as geological ones only if their reality can be supported by stratigraphical, petrographical and chronological arguments as well (Dalrymple & Lamphere 1979).

The samples under consideration were collected in the whole region of the Gutai Mts., generally avoiding proximity to the hydrothermal zone, where secondary effects could have, or are known to have, rejuvenated the volcanic rocks. Owing to the mineralogy and texture of the investigated volcanic rocks, mainly whole-rock data have been reported.

On the basis of our experience the K/Ar data obtained on the suites of rocks belonging to the same group estimate the probability of age intervals for different volcanic events of the Gutai Mts. and these radiometric data could reflect the succession of the events (see Fig. 2).

On the other hand the discordant K/Ar data and the obvious disagreement of radiometric - and geological ages believed to result from the incorporation of varying amounts of excess argon or argon loss. Both events can usually be discovered by using the isochron method on cogenetic rocks and on fractions of different magnetic susceptibility or density of a single piece of volcanic rock (Shafiqullah & Damon 1974; Fitch et al. 1976) since uniform increase or decrease of age of all parts of a lava flow or a large-sized intrusion is highly unlikely. Since misinterpretation of the measurements because of excess argon or diffusion deficiency cannot be excluded in the area under consideration, the isochron method was used in the dubious cases (No. 1, 2, 3, 7 and 20).

Results and discussion

The simplified succession of the volcanic rocks from Gutai Mts. and analytical data are presented in Tab. 1.

According to the biostratigraphical data, acidic volcanic activity took place during the Badenian. However, due to the strong alternation these volcanic rocks have still not been radiometrically dated.

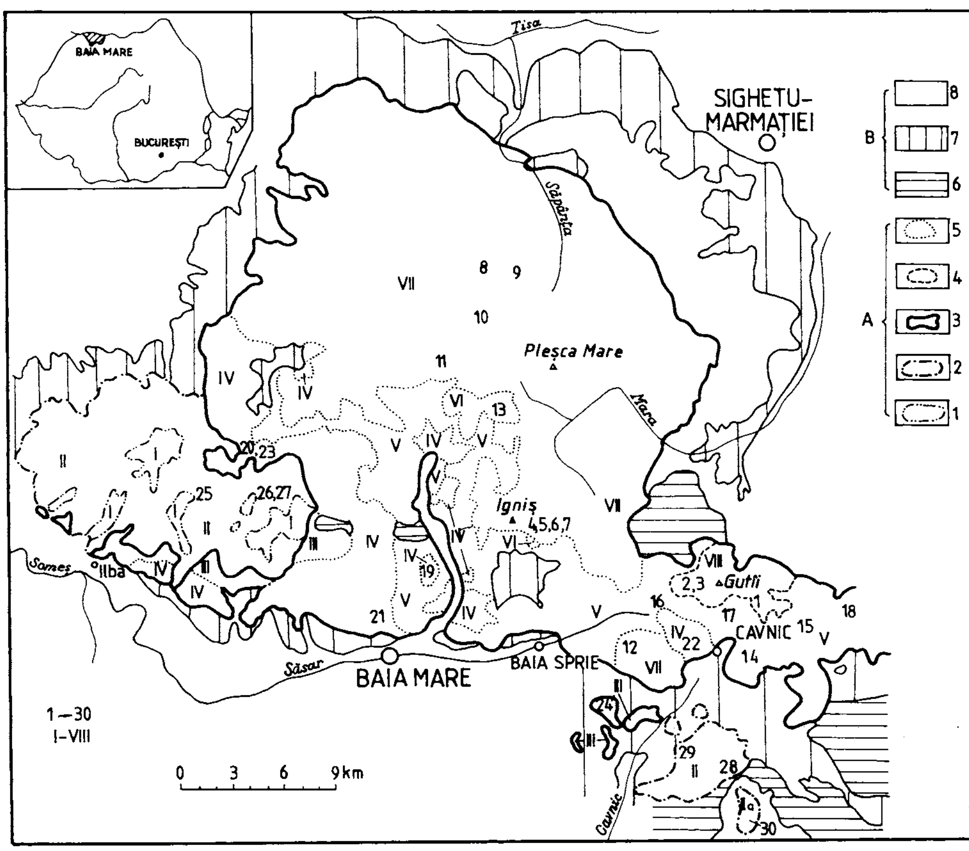

Fig. 1. Simplified geological map with samples locations. **A** - Volcanic rocks areas: 1 - Badenian; 2 - Sarmatian; 3 - Pannonian; 4 - Gutai Peak andesites; 5 - Volcanic rocks types in Pannonian (III-VII). **B** - Sedimentary rocks areas: 6 - Paleogene; 7 - Neogene; 8 - Quaternary. 1-30 Number of samples. **I-VIII** The succession of the volcanic rocks (see Tab. 1).

On the basis of radiometric data, the oldest intermediate volcanic rocks are Sarmatian in age and come from the western and south-eastern part of the Gutai Mts. (Fig. 1). A good agreement between the analytical data and biostratigraphical data was proved for the western area (13.3-12.1 Ma, II, No. 25-27). For the volcanic rocks from the south-eastern part (II, IIa, No. 28-30) the K/Ar datings suggest that they are younger volcanic products. Since the biostratigraphical data are not available it is felt that the concordant radiometric ages can be considered as geological ages.

The most reliable date was determined on fresh biotite (11.6 Ma) from the biotite dacites (III, No. 24) which fully agrees with the stratigraphical classification.

The concordant K/Ar ages measured on quartz andesites (IV, No. 19-23) are also Pannonian (11.3-10.5 Ma) and in good agreement with the paleontological data described in Fig. 2 (faunal association C). Excess argon influences are indicated by using the isochron method on sample 20. From isochron ages, the geological age can be inferred only with limited certainty since the error of I_1 age is great. It is highly probable that the average age counted for I_1, I_2 (10.7 Ma) should give a good approximation for the real geological age.

The analytical data confirmed the previous geological considerations regarding the quartz andesites and biotite dacites.

Against the previous biostratigraphical assumptions the radiometric dating proved that the volcanic rocks considered Pontian in age (V, No. 14-18) are Pannonian (10.9-10.1 Ma). Since the rock samples belonging to the V group were collected from a very large area, the highly concordant ages reflect the age of the eruptions of there andesitic rocks.

On the basis of some stratigraphical arguments, the rocks from the VI and VII groups (No. 4-13) were considered Pontian-Pliocene in age but these were not proved by the radiometric ages (10.3-9.0 Ma).

The volcanic rocks from the VI group were considered to be older than those from the VII group and different in their petrographical and petrochemical aspects. The analytical date (10.0 Ma) did not conflict with the presumed lower stratigraphical position within the "Pontian-Pliocene" sequence but taking into account the K/Ar age it must be considered Pannonian.

The rocks from the VII group are products of some different apparatus and can be also distinguished by petrochemical features.

Three of the four samples taken from Ignis Peak (No. 4, 5, 6) present very concordant ages. A more detailed age determination was performed on sample No. 7. On the opaque accessory minerals fraction a significant age distortion was detected (18.1 ± 1.5 Ma). Excess argon could be proved for the low K content mineral fraction. The effects on the whole rock ages can hardly be determined since the amount of excess argon varies between the different outcrops and perhaps within one occurrence, as well (Fuhrmann & Lippolt 1986).

The I_2 isochron age (10.1 ± 0.5 Ma) datum is a better approximation for the age of the rock than I_1 (10.9 ± 0.4 Ma) because the incorporation of more atmospheric Ar might have caused the shift of points in the $^{40}Ar/^{36}Ar$ - $K/^{36}Ar$ diagram resulting in an older analytical age.

The interval established for the VII group is more than 1Ma (10.3 - 9.0 Ma) suggesting that the different volcanic centers in the area were active at different moments.

A striking feature of the Pannonian volcanic activity is its great spatial extension (Fig. 1) with numerous petrographic varieties. Although it is sometimes possible to distinguish different an-

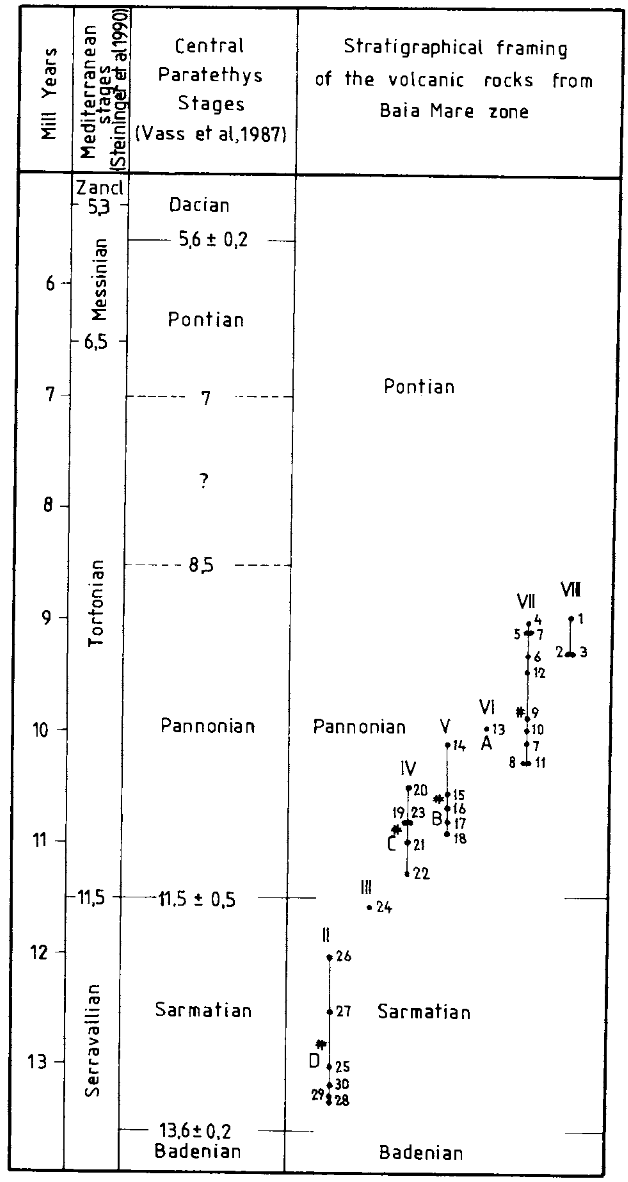

Fig. 2. The position of the volcanic rocks from the Gutai Mts., in the chronostratigraphic scale. Volcanic rocks: **II.** Pyroxene andesites, pyroxene basaltic andesites and pyroxene dacites (IIa) (Sarmatian); **III.** Biotite dacites (Pannonian); **IV.** Quartz bearing andesites (Pannonian); **V.** Pyroxene andesites and pyroxene basaltic andesites; **VI.** Hornblende pyroxene andesites; **VII.** Pyroxene andesites and pyroxene basaltic andesites; **VIII.** Pyroxene, hornblende, biotite, quartz, bearing andesites (Gutai Peak). Biostratigraphical data: **A** - Fossil flora in Chiuzbaia area-biozone with *Cupressus sempervirens, Fagus pliocenica, Ulmus cochii* (Givulescu 1990). **B** - *Congeria zsigmondyi* Hal., *C. Czjecki* Hoernes, *Cardium doderleini* Brus, *Limnocardium fatioi* Brus; *Monodacna viennensis* Papp, *Pisidium costatum* K-B, *Leptocythere monotuberculata* Sokač, *Candona (Pontoniella) unguiculus* (Reuss), *Pontoniella pontica* Agalarova (Bernad et al. 1987). **C** - *Congeria ramphophora* Brus, *C. ramphophra vösendorfis* Papp, *C. partschi partschi* Čzjek; **D** - *Cardium gleichenbergense* Papp, *C. (Cerastoderma) lithopodolicum* Dubois, *C.* aff. *C. barboti, C. pium pestis* Zhizh., *Ervilia dissita dissita* (Eichw), *Sarmysis sarmaticus* (Khol), *Ammonia beccarii* (Linné); (Marinescu 1964). The fossiliferous points position A - D is represented according to the field relationship between the sedimentary rocks and the dated samples. *Average age.

Table 1: Analytical data and simplified succession of the volcanic rocks from Gutai Mts.

No. of Samples	No. of K/Ar lab.	Dated fraction	K cont wt. (%)	$^{40}Ar_{rad}$ (%)	$^{40}Ar_{rad} \times 10^{-7}$ (cc STP/g)	Apparent age (Ma)	Isochron age (Ma)	Eruptive rock succession (previous stratigraphical framing)
1.	2204	w. r.	2.52	35.2	8.815	9.0±0.4		Quartz, biotite, hornblende, pyroxene andesites (Pontian-Pliocene) VIII
		w. r.	2.52	24.6	9.190	9.3±0.6		
		p. m.	2.53	24.2	9.434	9.6±0.6		
		h. fr.	1.59	37.3	5.992	9.6±0.5		
		0.6 A	1.69	37.5	7.369	11.2±0.5	I_1=9.0±0.8	
2.	2396	w. r.	2.40	38.3	8.667	9.3±0.4	I_2=9.7±0.5	
		p. m.	1.40	44.2	7.166	13.1±0.6		
		f. p.	0.45	29.9	1.489	8.5±0.5		
3.	2397	w. r.	2.22	43.4	7.998	9.3±0.4		
		p. m.	1.72	76.3	7.484	12.6±0.5		
4.	2116	w. r.	1.94	70.6	6.827	9.0±0.4		Pyroxene andesites and pyroxene basaltic andesites (Pontian-Pliocene) VII
5.	2121	w. r.	1.62	49.0	5.773	9.1±0.4		
6.	1866	w. r.	1.94	45.4	7.219	9.5±0.4		
		w. r.	1.94	24.6	7.063	9.3±0.6		
7.	2117	w. r.	1.63	66.5	7.381	11.6±0.5		
		p. m.	0.85	38.2	4.221	12.7±0.5	I_1=10.9±0.4	
		f. p.	1.64	47.1	7.400	11.6±0.5	I_2=10.1±0.5	
		h. fr.	0.30	17.6	2.135	18.1±1.5		
8.	2056	w. r.	1.51	43.3	6.092	10.3±0.5		
9.	2638	w. r.	1.82	55.4	6.970	9.8±0.4		
10.	2641	w. r.	1.45	47.5	5.583	9.9±0.4		
11.	2207	w. r.	1.43	36.0	5.772	10.3±0.5		
12.	2058	w. r.	0.78	43.0	2.903	9.5±0.4		
13.	2216	w. r.	2.73	47.7	10.589	10.0±0.4		Hornblende pyroxene andesites (Pontian-Pliocene) VI

Continuation of Tab. 1

No. of Samples	No. of K/Ar lab.	Dated fraction	K cont wt. (%)	$^{40}Ar_{rad}$(%)	$^{40}Ar_{rad} \times 10^{-7}$ (cc STP/g)	Apparent age (Ma)	Isochron age (Ma)	Eruptive rock succession (previous stratigraphical framing)
14.	2057	w. r.	1.53	31.7	6.050	10.1±0.5		Pyroxene basaltic andesites pyroxene andesites and pyroxene hornblende andesites (Pontian) V
15.	2075	w. r.	1.41	17.3	5.832	10.6±0.9		
16.	2219	w. r.	1.39	32.7	5.786	10.7±0.5		
17.	2218	w. r.	1.40	24.7	5.897	10.8±0.7		
18.	2210	w. r.	1.63	56.8	6.946	10.9±0.5		
19.	2415	w. r.	1.64	40.7	6.909	10.8±0.5		Quartz andesites (Pannonian) IV
20.	2115	w. r.	1.51	49.7	6.701	11.4±0.5	I_1=10.9±1.7	
		f. p.	0.43	17.3	2.161	12.8±1.1		
		h. fr.	0.33	21.9	1.916	14.5±1.0	I_2=10.5±0.7	
21.	2206	w. r.	1.51	37.5	6.471	11.0±0.5		
22.	1865	w. r.	1.77	73.1	7.787	11.3±0.4		
23.	1863	w. r.	1.66	74.9	7.011	10.8±0.4		
24.	2217	b.	6.69	43.5	3.023	11.6±0.5		Biotite dacites (Pannonian) III
25.	2120	w. r.	1.29	20.1	6.624	13.1±0.9		Pyroxene basaltic andesites and pyroxene andesites (Sarmatian) II
26.	1861	w. r.	1.33	38.3	6.275	12.1±0.6		
27.	2118	w. r.		25.0	6.352	12.6±0.8		
28.	2076	w. r.	1.15	36.6	6.010	13.4±0.7		
29.	2119	w. r.	1.63	25.1	8.485	13.3±0.8		
30.	2205	w. r.	2.70	45.6	13.850	13.2±0.6		Pyroxene dacites (Sarmatian) IIa
		-	-	-	-	-	-	Rhyodacitic pyroclastic rocks (Badenian) I

Explanation: w. r. - whole rock sample; p. m. - permanent magnetic fraction; h. fr. - heavy fraction; f. p. - feldspar; b - biotite;
I_1 - ^{40}Ar / ^{36}Ar - K / ^{36}Ar isochron age; I_2 - $^{40}Ar_{rad}$ - K isochron age
0.6 A - dated fraction was separated at 0.6 A

desite types within the same series having similar radiometric ages, the time interval of the volcanic activity that produced these lavas cannot be clearly defined because the experimental error overlaps the life span of the volcanism.

Tab. 1 shows as was said above, no volcanites younger than 9 Ma were evidenced. The lower boundary of the Pontian stage is not older than 8.5 Ma, according to the existent stratigraphical scales (Vass et al. 1987; Vass & Balogh 1989; Steininger et al. 1990; Marinescu 1991 unpublished data; Berggren 1987; Pevzner 1987) and to the radiometric datings (Pevzner & Chikovani 1978; Semenenko & Pevzner 1979; Chiumakov et al. 1988; Andreescu pers. comm. 1992).

Considering the available radiometric data from the Baia Mare zone the authors can state that there are no Pontian volcanic rocks.

According to the geological data the youngest volcanic rock of the whole region is the andesite of the Gutai Peak. Regarding its stratigraphic importance, repeated and detailed age determinations were carried out on 3 samples. (VIII No. 1-3) Besides three dated whole-rock samples, different fractions separated from this andesite type were also measured. The K/Ar dates for different fractions exhibit considerable scatter and increase to anomalously high values with increasing of magnetic susceptibility of dated fraction. It can be observed, however, that the ages measured on whole rock samples are in good accordance with one another. The whole rock ages are supported by the ages determined on fractions of greatly different K content (No. 2204 and No. 2396).

The apparent ages of permanent magnetic fractions, (No. 2396, No. 2397) older than that of the whole rock samples must reflect the presence of an excess argon component. The experiences of the $^{40}Ar/^{39}Ar$ dating performed on magnetite prove the evidence of excess argon, as well (Özdemir & York 1990). Two analytical ages determined on permanent magnetic fractions (No. 2396, No. 2397) were eliminated from the final analyses, others combined to provide an isochron age. The representative points of andesitic rocks fit well on a straight line and the I_1 age (9.0 Ma) assigned to it, approximates the youngest ages measured till now in the Gutai Mts. On the basis of arguments described above it is felt that K/Ar dating supports the opinion that these andesites are the youngest intermediate volcanic rocks in the Gutai Mts., but conflicts with the view which regards them as the products of andesitic volcanism Pontian-Pliocene in age.

Conclusions

The andesitic volcanic activity from Baia-Mare zone (Gutai Mts.) lasted about 4 - 5 Ma. It was initiated in the Middle Miocene, around the Badenian-Sarmatian boundary. The volcanism continued in the Pannonian and ceased around the Pannonian-Pontian boundary.

The Sarmatian volcanic activity is restricted to the southwestern and southeastern parts of the Baia-Mare volcanic region, while the Pannonian andesitic volcanism shows a significant spatial extension; however, the dominant volcanic phase belongs to the Pannonian.

The Sarmatian and the Pannonian volcanic phases were easily distinguished by K/Ar dating but within the Pannonian no significant age differences were detected. The ages could reflect a rapid succession of the volcanicity.

The supposed Pontian and Pliocene activity was not proved. In fact the youngest radiometrically dated andesitic rock of Gutai Peak is Pannonian (9 Ma), as well.

The radiometric age determined on adularia (8.7±0.4 Ma) from hydrothermalized andesites from the Baia-Sprie ore deposit (previously considered as Pontian) confirms the ages older than 9 Ma obtained for the fresh rocks.

It is worth noticing that the volcanic activity in Gutai Mts. is partly contemporaneous with that in the adjacent areas (Oas, Tokay, Vihorlat Mts.) and differs from the Calimani-Gurghiu-Harghita.

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ACTIVITIES OF SLOVNAFT

THE SECOND YEAR OF ACTIVITY OF THE ASSOCIATION OF INDUSTRY AND THE PROTECTION OF NATURE

SLOVNAFT

In number 3 of our periodical *"GEOLOGICA CARPATHICA"* from June 1993, we published information about the origin of the Association of Industry and the Protection of Nature (AIPN). This association includes industrial enterprises, economic organizations, the city of Bratislava as a legal entity, the Slovak Agency for the Environment, and other smaller organizations and private companies. From the original 6 founding members, the membership base has widened to the present total of 15. A further 5 organizations have applied for membership.

From 12th to 14th October 1994, the AIPN held a seminar under the title: " The AIPN and INCA - functioning in practice in the conditions of Slovakia and England ". The aim of the seminar was to provide the participants with new possibilities for solving problems in the relationship between industry and the environment. Apart from the members of the AIPN, the seminar was also aimed at representatives of the industrial sphere, professional and voluntary defenders of the environment, the state administration and local government. Representatives of INCA from England Mr. Jeremy Russell and Mr. Howard Robinson were foreign guests of the seminar. The AIPN seminar was held under the patronage of the city of Bratislava, capital of Slovakia, the mayor, Mr. Peter Kresánek, the British Embassy Know-How Fund and the Ministry of the Environment of the Slovak Republic.

The seminar was opened and moderated by the spiritual father of INCA in the conditions of Slovakia and the founder of the AIPN, Ing. Jozef Žila. Experts from various areas of ecology, town planning, biology, botany, water management, hydrogeology and others participated in the seminar. The seminar also included excursions to visit projects of the AIPN: Hrušov Reservoir, Lamač - pond, Štrkovec Lake and the Biskupice river branch. Slovnaft Inc. of Bratislava, the main representative of the industrial sphere, also provided assistance with the technical and organizational side of holding the seminar. Mr. Ján Kavec, production deputy, representing the general director of Slovnaft, actively participated in the seminar. In the contribution, "The ecological policy of Slovnaft Inc., in relation to protecting nature, and the environment in general", he briefly informed the participants in the seminar, about the joint stock company Slovnaft Inc. of Bratislava, from various points of view: historical, productive, economic, commercial, research and developmental, and from the point of view of quality, organizational structure, work opportunities and the environment. We will look at the environmental aspect in more detail.

Mr. Kavec summarized a cross section of ecological problems in all areas of the environment: protection of the atmosphere, water and waste disposal, at present and in future years.

In protection of the atmosphere, Slovnaft has achieved significant results. While at the beginning of the 1980s, emissions of sulphur dioxide, the most important gas pollutant, was emitted to the atmosphere at a level of more than 43,000 t/year, by the last year less than 50 % of this amount (17,434 t/year) was