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Tertiary magmatism within the Republic of Macedonia: a review

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Abstract

Widespread Tertiary magmatism of both orogenic and within-plate signatures developed within the Macedonian part of the Dinarides. Orogenic magmatites (predominantly volcanic rocks) are present in 5 areas (from east to west): Osogovo-Besna Kobila, Kratovo-Zletovo, Bučim-Borov Dol, Dojran and Kožuf. The age of the igneous rocks decreases in the same direction: from Priabonian-Early Oligocene at Os ogovo-Besna Kobila area, Early Oligocene-Miocene in Kratovo-Zletovo and Bućim-Borov Dol to Late Miocene-Late Pliocene in Kožuf.

The Osogovo-Besna Kobila area contains only acid volcanics (trachydacites to dacites), occurring mainly as subvolcanic to hypabyssal bodies. They have collision-related chemical characteristics. Latites and andesites to dacites predominate in the Kratovo-Zletovo area, but a monzonite pluton is also be found. Only the shoshonite series (from latites to trachyrhyolites) is present in Bučim-Borov Dol, and trachytes in Dojran area. The Kožuf area, which consists of two series; (1) shoshonitic (high-Mg shoshonites, latites and trachydacites) and (2) andesitic series (low-K andesites and high-Fe rhyolites), seems to be the most complex volcanic field. Magmatites in the last three areas have island arc signatures.

The within-plate volcanics rocks are high-K to ultra-K mafic igneous rocks (phonotephrites) accompanied by some absarokite, shoshonite to latite occurrences. Their age is Upper Miocene-Pliocene. These rocks are grouped in 4 areas: Kumanovo, Skopje-St.Nikole, Štip and Demir Kapija. The high Mg# and the high Cr-Ni contents of the phonotephrites, together with Fo-rich olivine phenocrysts suggest their asthenospheric origin.

1. Introduction

Extensive volcanic activity, accompanied by intrusion of minor plutons, took place in the Macedonian part of the Dinaride system during the Late Tertiary. These magmatic rocks represent part of a 1200 km long magmatic zone extending from southeastern Austria to Turkey (Harkovska et al., 1989). Here mainly orogenic igneous rocks are exposed, followed by within-plate alkaline mafic rocks.

The magmatic rocks outcrop essentially in the Serbo-Macedonian massif and in the Vardar zone (Fig. 1). The Serbo-Macedonian massif represents a collage zone (Dabovski et al., 1991) consisting of high-grade metamorphic terrains with exotic origin, probably derived from the Gondwana (Ricou, 1994). It became a part of the Eurasian plate due to subduction processes that occurred during different time periods before the Jurassic-Early Cretaceous. The Vardar zone represents the remains of the Tethyan Ocean, closed here in the Late Jurassic.

The aims of this paper are to summarize all available literature data about the age, petrography and geochemistry of the Tertiary magmatism in Macedonia. These data are complemented by the results of our new studies on the Tertiary plutonic rocks. Based on the petrochemical data, the classification of the volcanic rocks is defined more precisely then earlier and, where possible, differentiation mechanisms of the rocks series are outlined. The geodynamic setting of the magmatic manifestations is discussed and a genetic model is proposed.

2. Description of the orogenic magmatic rocks

Five orogenic Tertiary magmatic (mainly volcanic) areas are known in Macedonia (Karamata et al., 1992) (Fig. 1). One of them extends in the Serbo-Macedonian massif (Osogovo-Besna Kobila), the second along the border between the massif and the Vardar zone (Kratovo-Zletovo). The others extend along the Vardar zone (Bučim-Borov Dol, Dojran and Kožuf). Important ore fields are located in all magmatic areas (except for Dojran area): mineralizations of Pb-Zn in Osogovo-Besna Kobila and in Kratovo-Zletovo, of Cu in Bučim–Borov Dol and of Au-Sb-As-Tl-Pb-Zn in Kożuf. Besides these volcanic rocks, Arsovski (1997) supposed the occurrence of isolated igneous bodies in the western part of the Pelagonian massif, but their Tertiary age has not yet been proved.

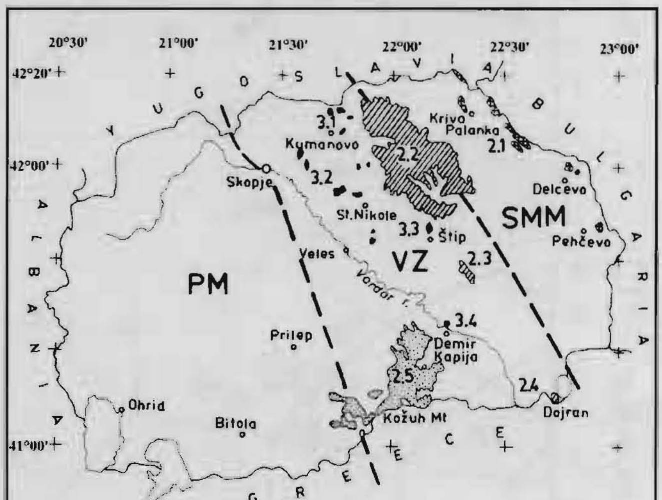

Fig. 1 – Areas of the Tertiary volcanic rocks in Macedonia. Orogenic volcanic areas: Priabonian-Oligocene collision-related (2.1, Osogovo-Besna Kobila); Oligocene-Miocene (2.2, Kratovo-Zletovo, 2.3, Bućim-Borov Dol and 2.4, Dojran); Miocene-Pliocene island-arc area (2.5, Kozuf). Within-plate volcanic areas (Miocene-Pliocene): 3.1, Kumanovo, 3.2, Skopje-St.Nikole, 3.3, Štip, 3.4. Demir Kapija. Tectonic units (according to Arsovski, 1997): SMM, Serbo-Macedonian massif; VZ, Vardar zone and PM, Pelagonian massif.

2.1. *The Osogovo-Besna Kobila (Sasa-Toranića) area*

This area represents NW trending magmatic zone, hosted in metamorphic and plutonic pre-Tertiary rocks of the Serbo-Macedonian massif. It extends for about 100 km on both sides of the Bulgarian-Macedonian border, continuing in Yugoslavia to the North (Simić, 1997). It has a great metallogenic importance. The magmatic zone contains several dykes, up to 50 m thick, strike 260°, rarely perpendicularly to this direction. They are of subvolcanic to hypabyssal facies. According to Pendjerkovski (1965), they are microquartzdiorites and microgranodiorites. In the Deve-Bair and Pehčevo localities (Aleksandrov, 1992) as well in some regions of southwestern Bulgaria (Harkovska et al., 1989 and references therein), pyroclastic rocks and volcanic domes can be also found. They are interbedded with or overlie Priabonian sediments; others volcanic bodies crosscut Early Oligocene sediments (Arnaudova, 1973). K-Ar data from the dykes of Bulgarian part of this magmatic zone yield age of 32-33 Ma (whole rock analyses, Harkovska et al., 1989), in Yugoslavia – 32.56-29.47 Ma (K-Ar age of biotites-Simič, 1997).

Most of the analyzed volcanic rocks in the Osogovo-Besna Kobila are strongly hydrothermally altered. According to the unaltered samples these are dacites, or more rarely, trachydacites (Fig. 2A and Table 1). They are rich in phenocryst

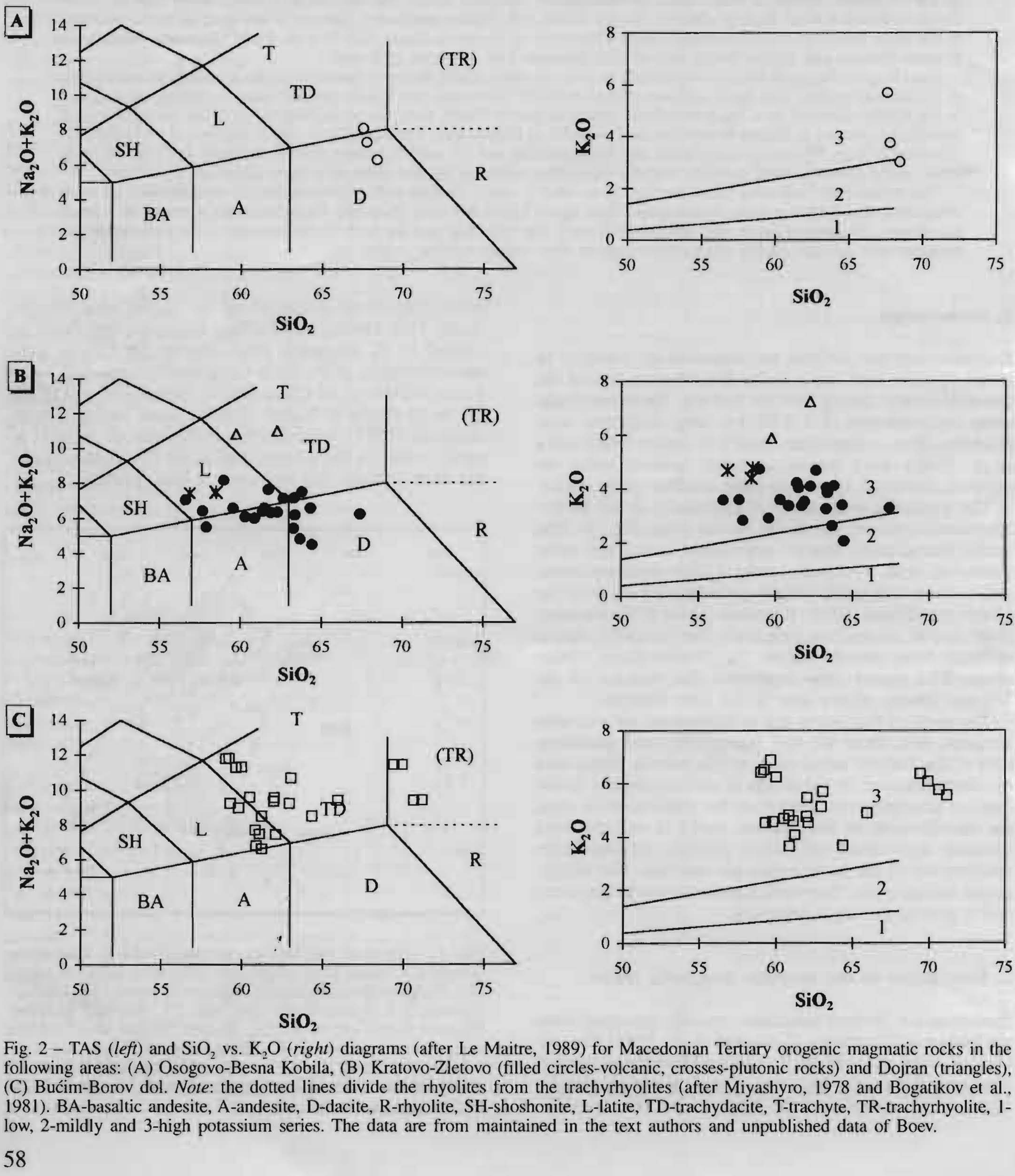

Fig. 2 – TAS (*left*) and SiO_2 vs. K_2O (*right*) diagrams (after Le Maitre, 1989) for Macedonian Tertiary orogenic magmatic rocks in the following areas: (A) Osogovo-Besna Kobila, (B) Kratovo-Zletovo (filled circles-volcanic, crosses-plutonic rocks) and Dojran (triangles), (C) Bućim-Borov dol. *Note*: the dotted lines divide the rhyolites from the trachyrhyolites (after Miyashyro, 1978 and Bogatikov et al., 1981). BA-basaltic andesite, A-andesite, D-dacite, R-rhyolite, SH-shoshonite, L-latite, TD-trachydacite, T-trachyte, TR-trachyrhyolite, 1-low, 2-mildly and 3-high potassium series. The data are from maintained in the text authors and unpublished data of Boev.

(about 30%). The rocks contain the K-feldspar (sometimes up to 5-6 cm in size), poorly zoned plagioclase (An_{35-40}) (15-16%), quartz (2-3%), amphibole, biotite, rare clinopyroxene, and accessories such as apatite, titanite and zircon. The groundmass is holocrystalline, but glassy varieties can also be found.

According to the Rb contents of the volcanic rocks in Bulgarian (Arnaudova, 1973), they can be considered as syn-collisonal.

2.2. *The Kratovo-Zletovo area*

The Kratovo-Zletovo is the largest magmatic area in Macedonia with a surface of 1200 km^2. It is also a very important mining area with big Pb-Zn deposits. Many authors have studied this area (Stojanov, 1974; Stojanov & Denkovski, 1974; Stojanov & Radović, 1974; Stojanov & Serafomovski, 1990) and the petrographic characteristic and chronology of the volcanic events are presented here according to these studies.

Volcanic deposits predominate in the area, but plutonic rocks are also be found. The oldest volcanic occurrences are the ash-bearing sediments in the Preabonian flysch near the village of Talasmanći (Arsovski, 1997). Effusive volcanic activity occurred in 4 phases, but only the first two ones, of Early Oligocene age, have been dated radiochronologically. The latites of the first phase at Novo Ketenovo yielded the age 33.3-33.5±0.5-0.6 Ma[1], those of Kundinovo are 32.9±0.7 Ma[1], and the volcanic rocks of Rugínće are 29-32±2 Ma (Karamata et al., 1992). The latites of the second phase at Zdravči Kamen yielded an age of 30.9±0.6 Ma[1]. The age of the other phases were determined based on their geological relationships, e.g. products of the third phase are deposited within Early Miocene sediments (Arsovski, 1997) and those of the fourth phase intersect these sediments. The andesite dyke in the Plaviča locality (16 Ma, Tomson et al., 1998) probably belongs to fourth phase. According to Arsovski (1997), andesitic volcanoclastic rocks were deposited in limnic Pliocene sediments, but they may be epiclastites.

The brief description of the rocks of the different phases is the following:

First phase: latites and andesites, consisting of phenocrysts of plagioclase (An_{45-55}), amphibole, biotite, some clinopyroxene, and accessories such as magnetite, apatite, and rare zircon. They are accompanied by dacites to trachydacites with sanidine phenocrysts of 1 cm in size.

Second phase: andesite-dacite ignimbrites, consisting of fine-grained (3-4 mm) porphyroclasts of plagioclase (20-30%), sanidine (up to 10%), quartz, some amphibole, biotite and clinopyroxene. They often contain dacite and andesite lithoclasts, sometime small metamorphic and sedimentary clasts from the basement. Ignimbrites are crosscut by subvolcanic bodies of coarse porphyritic dacites containing sanidine and plagioclase phenocrysts (up to 3 cm in size), some biotite and amphibole. According to Cizars & Rakić (1956), one dacite body crosscuts ore vein No 1 in the Dobrevo Mine.

Third phase: andesites, containing plagioclase (An_{30-50}), biotite, amphibole and some pyroxene phenocrysts. They are accompanied by ignimbrites, consisting of partially broken phenocrysts of plagioclase, amphibole, clinopyroxene, biotite and some quartz (sometime up to 10%). The ignimbrites contain large quantities of andesite lithoclasts.

Fourth phase: fine-grained porphyritic andesites. The major phenocrysts are labradorite and clinopyroxene (Marić, 1952), together with small quantities of amphibole and biotite.

Volcanic activity, described above, migrated from NW to SE. It forms a petrochemical series transitional between shoshonitic (mainly the products of the first phase) and calc-alkaline one with a clear negative correlation between SiO_2 and alkalis (Fig. 2B and Table 1). The first three phases are rich in water – the major mafic minerals being amphibole and biotite. The products of the last phase are poor in water – the major mafic mineral is clinopyroxene. According to the variations of Ba and Sr contents relative to Rb (Fig. 3), fractionation of biotite predominates in the differentiation process, but the Rb vs. Y diagram indicates the participation also of pyroxene and plagioclase.

Some intrusive bodies also occur in Kratovo-Zletovo area (unpubl. data of Boev). One pluton is situated near Karlukovo village and one diorite-porphyrite dyke near Borovič village (the latter of 30.5±0.5 Ma). The Karlukovo pluton is quartz-monzonitic to monzonitic, composed of plagioclase, K-feldspar, biotite, clinopyroxene, and some quartz. Its chemical composition is identical to that of the latites of the first phase (Fig. 2B). Distribution of the normalized trace elements contents (Fig. 4) show high enrichment in LILE (e.g. up to 300 ppm Rb) and a progressive decreasing from HFS to Fe-Mg elements with very low contents of Ni and Cr. The negative Nb anomaly is characteristic of supra-subduction zones. The REE pattern (Fig. 4) gradually decreases from light towards heavy REE with a small negative Eu anomaly (except for one sample). The variations of Ba content vs. constant Rb content (Fig. 3) indicate that fractionation of K-feldspar and amphibole is the predominant differentiation process and the Eu anomaly – the limited participation of the plagioclase in this process.

2.3. *The Bučhim-Borov Dol area*

This is a small volcanic area of high metallogenic importance because it contains the large Bučim porphyry copper deposit. Several authors studied this area (Djordjević & Karamata, 1976; Djordjević & Knezević, 1980; Serafimovski, 1990) and the following petrographic characterization have been summarized based on their investigations.

This is a differentiated series where SiO_2 contents vary from 57 to 71% (Fig. 2C and Table 1) and contents of all major elements gradually decrease (not shown in the figures). Only K increases with increasing SiO_2, whereas Na content remains almost constant. According to the chemical composition, the volcanic rocks vary from latites, through trachydacites-trachytes to trachyrhyolites. The latites and trachytes rocks form necks and subvolcanic bodies (e.g. the Bučim copper deposit) and lava flows associated with the central volcanoes. The trachyrhyolites form dykes. The volcanic rocks of this area yield Early-Late Oligocene age: from 29.0±3.0 to 24.7±2.0 Ma (Boev & Lepitkova, 1991; Karamata et al., 1992).

The subvolcanic latites of the Bučim copper deposit are made up of coarse-grained phenocrysts of plagioclase (25-30%), amphibole, biotite (up to 15%), rare clinopyroxene,

[1] Unpublished data of Boev (analyzed in the Max-Planck Institut, Mainz, Germany).

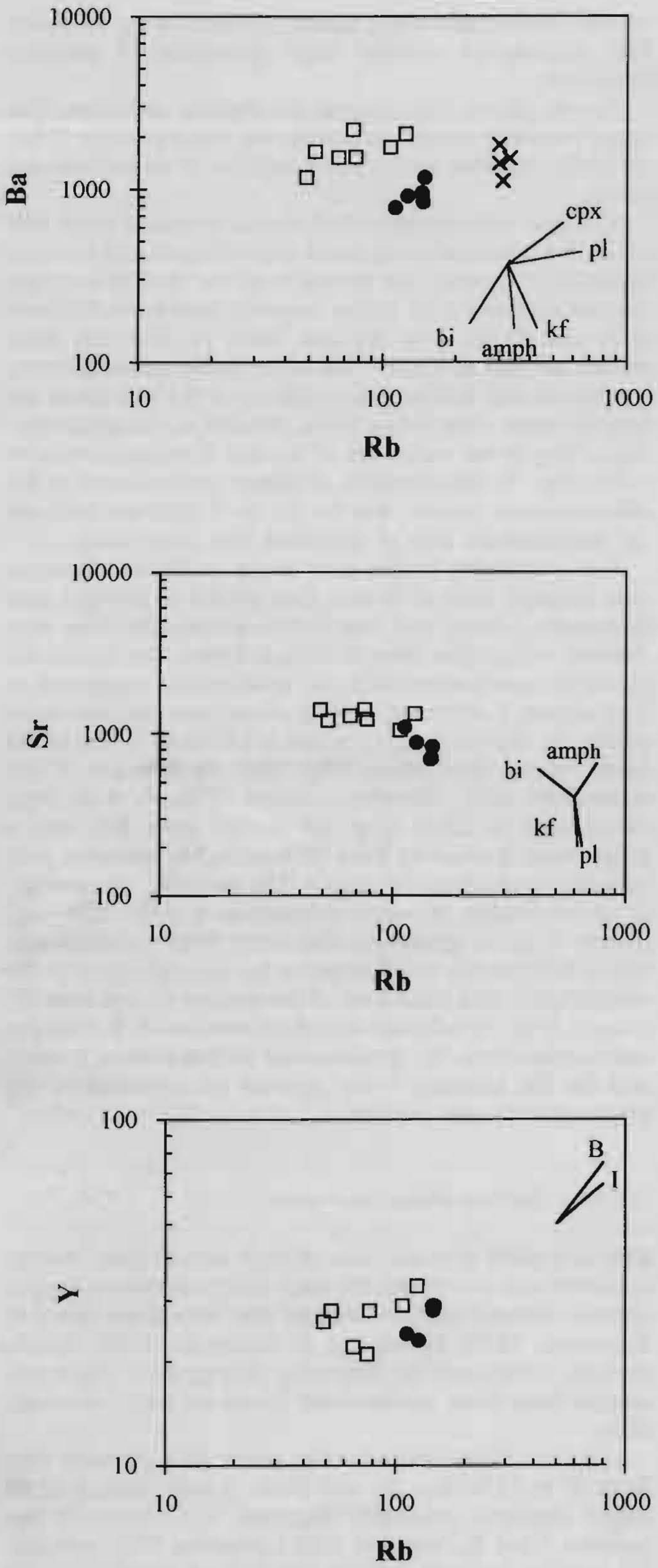

Fig. 3 – Rb vs. Ba, Sr, and Y contents of the Kratovo-Zletovo (filled circles-volcanic, crosses-plutonic rocks) and Boucim-Borov Dol (squares) areas.

In the right corner: modeled fractionation vectors in the Rb vs. Ba and vs. Sr diagrams for biotite, clinopyroxene, plagioclase, K-feldspar and amphibole (after Harris et al., 1983); theoretical fractionation vectors in basic (B) and intermediate (I) magmas for 50% crystallization of plagioclase+clinopyroxene+olivine assemblages in the Rb vs. Y diagram (after Pearce et al., 1990).

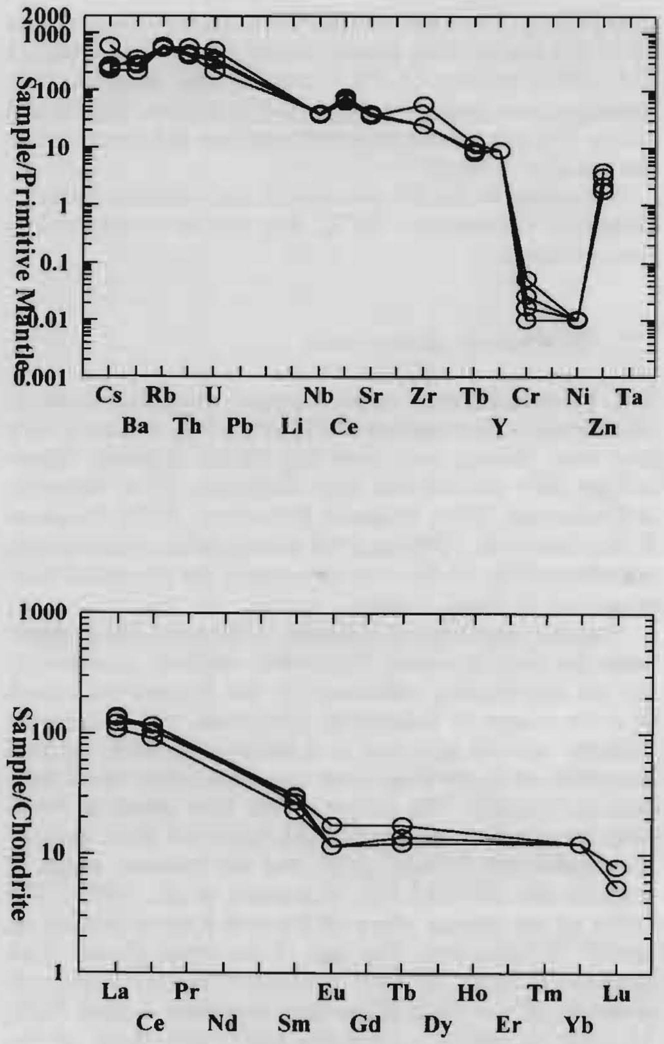

Fig. 4 – Trace elements distribution spidergram (top) and REE distribution pattern (bottom) of the Kratovo-Zletovo monzonites.

sanidine and accessories such as apatite, magnetite and allanite. The trachyrhyolites are fine-grained with phenocrysts (up to 35%) of quartz, sanidine, some plagioclase (2-9%) and remnants of amphibole, biotite, and accessories such as apatite and titanite.

With regard to petrochemistry, unlike the Kratovo-Zletovo area, the volcanic rocks of Bučhim-Borov Dol area belong to the shoshonite series only (Fig. 2C). According to the variations of Ba and Sr contents relative to Rb (Fig. 3), fractionation of clinopyroxene predominated in the differentiation process. The variation of Y content indicates that plagioclase also participated in this process.

The igneous rocks of Kratovo-Zletovo and Bučhim-Borov Dol areas using available trace elements data (Table 3) can be classified as subduction-related volcanic arc magmatic rocks (Fig. 5). The similarity with the trace elements contents of the Bučhim-Borov Dol rocks with the rocks of the active continental margins is indicated by Boev et al. (1992).

2.4. *The Dojran area*

This small area extends on the border between Macedonia and Greece. It contains (Fig. 2B and Table 1) several trachyte domes and dykes and rhyolites in Greece –

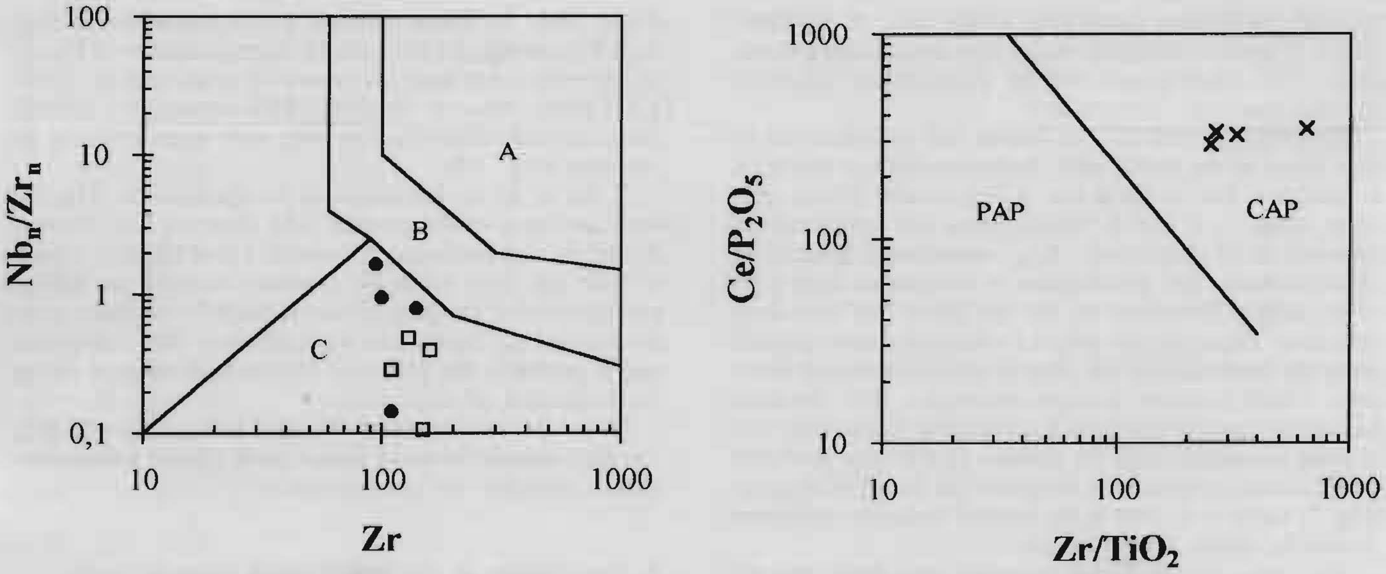

Fig. 5 – *Left*: Zr vs. Nb_n/Zr_n (after Thieblmont & Tegyey, 1994) discrimination diagram of the volcanic rocks of the Kratovo-Zletovo (filled circles) and Bucim-Borov Dol (squares) areas; A, within plate, B, collision-related and C, subduction-related. *Right*: Ce/P_2O_5 vs. Zr/TiO_2 discrimination diagram according to Müller et al. (1992) for the monzonites of Kratov-Zletovo area; CAP-continental arc, PAP-postcollisional arc magmatic rocks.

Doiranis locality (Majer, 1966). The age of the trachyte dome near Mrdaja is 33±5 Ma (Stoyanov & Sveshnikova, 1985). The trachytes are made up of zoned plagioclase (An_{25-42}), sanidine (up to 1 cm), clinopyroxene and amphibole, biotite, and coarse-grained titanite. Magnetite and apatite are also present as accessory minerals. The texture of the groundmass is trachytic.

2.5. *The Kožuf area*

This area contains only the Kožuf volcanic massif (Boev, 1988; Boev et al., 1997) which extends along the Macedonian-Greek border. It is composed of lavas and various types of pyroclastic rocks (mainly debris and pyroclastic flow), deposited in the Pliocene Tikveš lake (the Vitačevo plateau). The volcanic activity in the area took place in Late Miocene to Late Pliocene (from 6.5±0.2 to 1.8±0.1 Ma; Boev & Lepitkova, 1991; Kolios et al., 1980). The volcanic rocks form a shoshonitic series, from high-Mg shoshonites and to trachydacites, including high-Ti latites to trachytes, too (Fig. 6 and Table 2). Another series is also present comprising some very small bodies of andesite to dacite and rhyolites with slight tholeiitic characteristics.

High-Mg shoshonites are composed of plagioclase (An_{42}), amphibole, biotite and clinopyroxene phenocrysts. The shoshonites are characterized also by their high H_2O and low Ti, Fe, P and Ca contents.

Latites are made up of phenocrysts of plagioclase (An_{40-47}) and sanidine, amphibole, biotite ($Mg^{\#}$ 0.64) and augite ($Wo_{48.9}En_{36.7}$; $Mg^{\#}$ 0.72). The groundmass consists of microlites of the above mentioned minerals. The accessory minerals are apatite, ilmenite, rutile and magnetite.

Trachydacites are the most common rocks in the Kožuf area and differ from latites by their more acidic plagioclase (An_{23-38}) and small quantity of mafic minerals represented mainly by amphibole (Si 6.37-6.68; $Mg^{\#}$0.55) and biotite.

In addition, the shoshonite series also contains high-Ti latites to trachytes, which form a neck. Most of these rocks

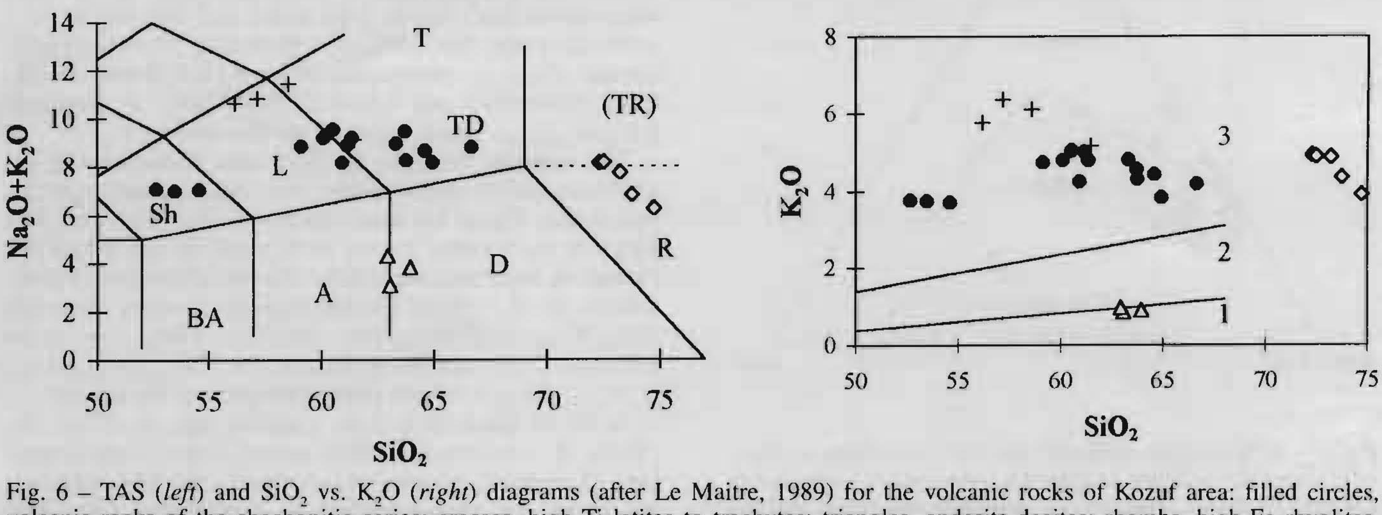

Fig. 6 – TAS (*left*) and SiO_2 vs. K_2O (*right*) diagrams (after Le Maitre, 1989) for the volcanic rocks of Kozuf area: filled circles, volcanic rocks of the shoshonitic series; crosses, high-Ti latites to trachytes; triangles, andesite-dacites; rhombs, high-Fe rhyolites. The data are from Boev (1988).

outcrops in Greece (according to the data of Soldatos, 1955). These are porphyritic rocks with crystallized groundmass. The phenocrysts are of plagioclase, sanidine, anorthoclase and clinopyroxene.

Furthermore, andesites to dacites and rhyolites can be also found in the Kožuf area. Andesites (with a transition to dacites – Fig. 6) have low K_2O contents, which classifies them as a low-K series. They are composed of phenocrysts of plagioclase (An_{50}), amphibole, biotite and clinopyroxene. The groundmass is vitrophyric with a lot of microlites. Rhyolites are the last phase and they form extrusion. These are vitrophyre rocks with coarse-grained phenocrysts of sanidine and plagioclase in a perlitic groundmass which contains feldspar microlites. The rhyolites have been classified as high K series (Fig. 6) but they also contain unusually high Fe content (4-8% wt), probably concentrated essentially in the glass. On the AFM diagram (Fig. 7) these rocks plot in the field of tholeiitic series and should be called ferrorhyolites.

The rocks of the Kožuf volcanic area show specific characteristics in the distribution of the major elements as follows (Fig. 8):

1. The more mafic variety of the volcanic series is rich in Mg and poor in Fe, Ca, Ti and P shoshonite;
2. The contents of the most of the major elements (Al, Fe, Ca, Na) in all volcanic rocks (except for rhyolite) are more or less constant. Only Ti and P decreases progressively (except for shoshonite);
3. The content of Mg in the Kožuf latite is lower then in the normal latites;
4. Unusually high Ti contents is observed in some latites (with a transition to trachytes); because of its high K content, they plot outside the trend of the Kožuf shoshonitic series (Fig. 8).

All these features cannot be explained by a normal crystallization differentiation and requires additional studies of their petrochemistry and phenocryst composition.

Trace elements were studied only in the most common rocks: latite-trachydacites and high-Ti latite-trachytes (Fig. 9 and Table 3). These volcanic rocks, regardless of their high K contents, do not contain large quantities of Rb (up to 210 ppm), but their Cs (about 40 ppm) and Sr (1100-1200 ppm) contents are high. REE-normalized patterns show normal differentiation with very weak negative Eu anomaly (Fig. 10).

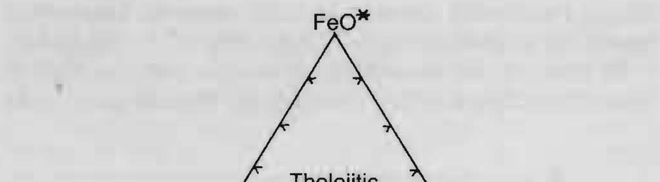

Fig. 7 – AFM diagram of Kozuf volcanics (according to Boev, 1988): asterisks, high-Mg shoshonites; open squares, trachydacites and high-Ti latites; open circles, andesites and latites and triangles, rhyolites.

Y, Ba or Sr vs. Rb diagrams for these rocks (Fig. 11) show increase of Rb content with more or less constant Sr and Ba and increasing Y content. These features together with the very weak Eu anomaly suggest the limited participation of the plagioclase in the differentiation process formed the latites and trachydacites. The clinopyroxene is probably the principal fractionated mineral during the formation of these rocks.

Based on trace elements discrimination diagrams (Fig. 12) the volcanic rocks of Kožuf show typical subduction-related volcanic arc characteristics.

3. Description of the within-plate volcanic rocks

In Late Miocene and in the Pliocene a within plate potassic mafic volcanism took place in Vardar zone. It is located in 4 volcanic areas: Kumanovo, Skopje-St. Nikole, Štip and Demir Kapija (Fig. 1). Kononova et al. (1989) classified these mafic rocks in two series based on the chemical composition and especially on the relationship between Al and Ca contents (Fig. 13): (1) tephrite-leucitic (Štip area) and (2) transitional series between tephrite-leucite and lamprophyre (Kumanovo and Demir Kapija areas) named by these authors a moderate-Al tephrite-leucitic series. According to the criteria of Foley at al. (1987) these are ultra-K rocks and because of the high $Mg^{\#}$ they fall on the field of the lamproite series of the same authors. Their high $Mg^{\#}$ is due only to their low Fe content. For this reason and their mineral composition, these rocks cannot be referred to as lamproites (Kononova et al., 1989). Both series are accompanied by volcanic rocks of the shoshonitic series (Fig. 14): absarokites (Skopje) and shoshonites to latites (in the other areas).

The volcanic rocks in the Kumanovo area (Kononova et al., 1989) are phonotephrites according to the TAS diagram (Fig. 14). They make up 16 small volcanic centers (with necks, flows, etc.) which crosscut and overlie Pliocene sands and clays. The lavas are accompanied by their pumice, breccias and tuffs. The body at Malo Nagoričane (Sveshnikova et al., 1986) contains phenocrysts of leucite, very rich in BaO (up to 3.12 wt%) and SrO (up to 0.87 wt%), diopside ($En_{7-10}Wo_{45-47}$ with 0.22-0.35 wt% Cr_2O_3), olivine ($Fo_{88.5-92}$), phlogopite (with 0.12-0.58%wt Cr_2O_3, 0.2-0.3%wt Ba0 and 1.63-2.07%wt TiO_2). Hyalophane ($Or_{48}Ab_{25}Cn_{26}$) has developed on the leucite.

The volcanic rocks in the Štip area (Kononova et al., 1989) are poorly differentiated and forms numerous necks and dykes. These are also phonotephrites (Fig. 14). The rocks of the locality Ezovo Brdo yield an age 5.5±5 Ma (Terzić & Sveshnikova, 1986). The latter contain (Sveshnikova et al., 1986) phenocrysts of leucite, diopside ($En_{10}Wo_{49}$), phlogopite, olivine ($Fo_{80.5-82}$) with 0.05%wt Cr_2O_3 and Ba-Sr plagioclase ($Or_{5.5-9}Ab_{38.5-47}An_{33-41.5}Sr_{4.5-12.3}Cn_{1-4.1}$), which pseudomorphosed the leucite.

A 20 m thick lava flow, yielding age of 9.5±2 Ma (Terzić & Sveshnikova, 1986), occurs in the Demir Kapija area (Kurešnička Krasta). According to the TAS diagram, the rocks are phonotephrites to shoshonites (Fig. 14). Because they are completely crystallized, these rocks are

Table 1 – Major elements content in the orogenic magmatic rocks of Osogovo-Besna Kobila, Kratovo-Zletovo, Dojran and Buèim-Borov Dol areas (in %wt)

	1	2	3	4	5	6	7	8	9	10	11	12	13
SiO_2	67.20	66.30	55.48	57.00	58.68	60.25	62.83	63.43	66.00	54.88	58.29	58.73	61.40
TiO_2	0.38	0.47	0.46	0.86	0.62	0.90	0.42	0.44	0.50	0.67	0.71	0.68	0.68
Al_2O_3	15.28	16.15	19.18	17.22	16.76	18.00	13.85	17.71	12.00	15.95	15.92	20.01	18.01
Fe_2O_3	2.96	0.75	5.83	3.50	4.93	4.76	6.63	3.94	5.80	6.13 tot	7.49 tot	2.42	0.30
FeO	1.12	2.05	1.04	4.50	2.15	0.54	2.06	0.79	1.20	n.d.	n.d.	0.97	1.60
MnO	0.21	0.32	0.15	0.22	0.21	0.07	0.36	0.06	0.50	0.16	0.17	0.08	0.06
MgO	1.30	1.51	1.10	3.00	0.88	1.65	1.94	0.85	0.80	4.72	3.57	0.92	1.51
CaO	3.36	2.78	7.56	6.08	7.52	5.42	5.20	4.70	4.80	7.12	5.87	3.72	3.86
Na_2O	3.20	3.45	3.38	2.80	3.62	2.12	2.16	3.14	2.90	3.01	3.06	4.87	3.72
K_2O	2.95	3.65	3.51	3.54	2.85	3.80	2.58	3.33	3.20	4.26	4.38	5.78	7.13
P_2O_5	0.26	0.38	0.29	0.12	0.30	0.09	0.61	0.18	0.20	040	0.29	0.16	0.33
CO_2			0.46			0.60	0.50						0.62
H_2O^+	1.40	2.05	0.88	0.80	0.78	0.70	0.10	0.95	1.00	0.74*	0.69*	1.20	0.88
H_2O^-	0.05	0.05	0.48	0.54	0.02	1.20	0.26	0.57	0.70			0.45	0.14
Total			99.80	100.35	99.32	100.10	99.50	100.09	99.60	100.23	100.44	100.04	100.24

1-2, Osogovo-Besna Kobila area (Karamata et al., 1992); Kratovo-Zletovo area: 3-9, volcanics and 10-11, plutonic rocks (unpubl. data of Boev); 12-13, Dojran area (Majer, 1966); 14-20, Buèim-Borov Dol area (Karamata et al., 1992 and unpublish. data). * L.O.I.

Table 1 (continuation)

	14	15	16	17	18	19	20
SiO_2	58.43	59.97	61.17	62.80	63.35	68.63	70.37
TiO_2	0.56	0.54	0.50	0.70	0.60	0.25	0.36
Al_2O_3	16.57	17.98	18.21	16.32	16.57	15.81	15.04
Fe_2O_3	3.23	4.50	4.35	2.42	2.77	1.80	1.14
FeO	1.47	2.28	1.69	1.44	1.10	0.13	0.39
MnO	0.10	n.d.	0.01	0.15	0.15	0.04	0.04
MgO	2.57	1.95	1.20	2.00	1.81	0.30	0.90
CaO	3.64	3.28	1.90	4.29	3.45	0.56	1.21
Na_2O	4.30	2.85	4.01	4.50	4.75	5.00	3.80
K_2O	6.75	4.75	5.40	3.60	3.60	6.30	5.50
P_2O_5	0.35	0.46	0.18	0.30	0.28	0.04	0.15
CO_2		0.03					
H_2O^+	1.80	1.23	1.10	1.50	125	1.27	1.11
H_2O^-	0.08	0.43	0.80	0.08	0.10	0.04	0.06
Total	99.85	100.25	100.52	100.10	99.79	100.17	100.07

referred to orendites (Sveshnikova et al., 1986). They contain leucite, pseudomorphosed by Na-orthoclase ($Or_{60}Ab_{34}An_5$), diopside ($En_{5-7.5}Wo_{45.5-46.5}$) and olivine (Fo_{86-88}).

These alkali mafic rocks are characterized (Kononova et al., 1989) (Table 4) by high contents of F (0.04-0.86%wt) and LIL elements such as Li, Rb (172 and 441 ppm respectively) and Cs (9-11 ppm of Štip area, 20-29 ppm of Kumanovo and 12-21 ppm of Demir Kapija), concentrated, most probably, in leucite. The contents of Sr and Ba are particularly high: Sr contents are very high in leucite phonotephrites (up to 3644 ppm) and varies in other rocks (between 900 and 1000 ppm); Ba goes up to 5800 ppm, varies between 200 and 4000 ppm. Sr is concentrated in feldspars and apatite, whereas Ba is concentrated in alkali feldspars, micas, occasionally in leucite (Sveshnikova et al., 1986). Among the HFS elements, increased concentrations of Zr (250-500 ppm) and Y (25-45 ppm) have been also found in these rocks.

In the trace elements discrimination diagrams the described mafic rocks plot in the field of within-plate basalts (Fig. 15).

4. Discussion of the genesis of the Tertiary magmatism in Macedonia

In order to obtain a more precise genetic hypothesis it is necessary to analyze the Paleogene magmatism of Macedonia against the background of magmatic evolution in the adjacent areas. We was studied a NE-SW transect (Rila plutons in Bulgaria, Osogovo-Besna Kobila in Bulgaria and Macedonia, Kratovo-Zletovo, Bučim-Borov Dol and Kožuf areas – Figs. 1, 16), perpendicular to the zone of remnants of the Tethyan Ocean. This transect groups all described orogenic magmatic rocks.

The Bartonian Rila granodiorite plutons (35-40 Ma, Kamenov et al., 1999) showing mildly alkaline petrochemistry, are situated in the most northeastern sector of this transect (in Bulgaria). To the SW, in the Osogovo-Besna Kobila area (in Bulgaria and Macedonia), hypabyssal-subvolcanic magmatism took place in the end of Priabonian and the Early Oligocene (Harkovska et al., 1989; Karamata et al., 1992). This magmatism is calc-alkaline (Harkovska et al., 1989) with lower K content than that in the Rila plutons. The distribution of Paleogene igneous rocks in this part of the transect is very similar to those of the Eastern Rhodopes-Western Thrace areas with a migration of magmatic activity towards the Tethyan suture to the SE and decreasing K contents in the same direction. In the Eastern Rhodopes-Western Thrace areas this is explained by the break-off of the subducted plate and magma formation from metasomatized mantle (Yanev et al., 1998).

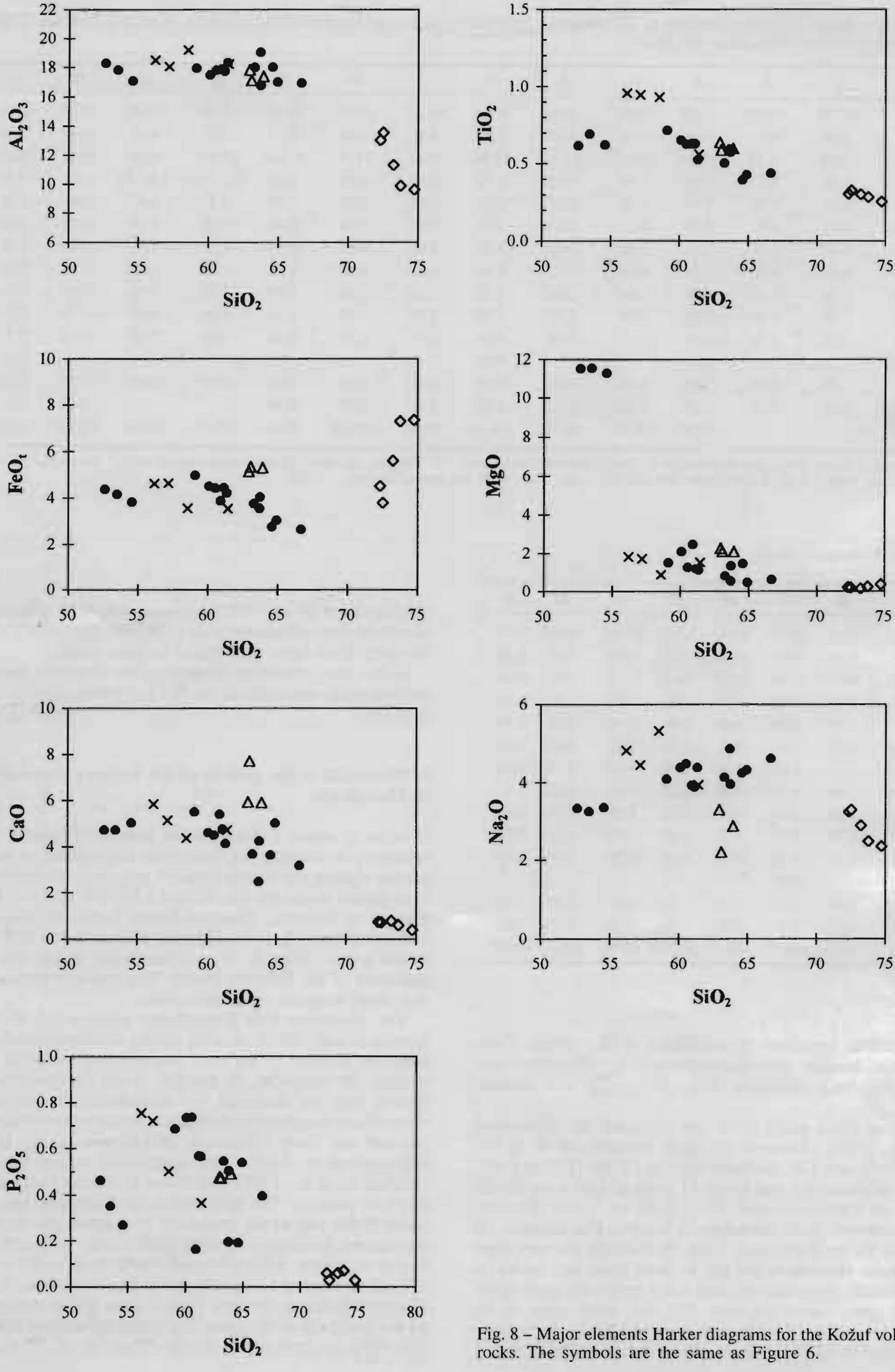

Fig. 8 – Major elements Harker diagrams for the Kožuf volcanic rocks. The symbols are the same as Figure 6.

Table 2 – Major elements content in the orogenic volcanic rocks of Kožuf area according to Boev et al. (1997) in %wt.

	1	2	3	4	5	6	7	8	9
SiO_2	50.12	59.20	58.67	60.86	62.72	65.08	55.82	60.12	72.49
TiO_2	0.65	0.60	0.71	0.52	0.50	0.43	0.95	0.55	0.30
Al_2O_3	16.70	16.80	17.81	18.20	17.84	17.04	18.41	17.84	11.22
Fe_2O_3	1.66	3.71	5.51 tot	4.64 tot	4.12 tot	3.39 tot	5.11 tot	3.86 tot	6.19 tot
FeO	2.39	1.50	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
MnO	0.07	0.06	0.11	0.11	0.08	0.08	0.15	0.09	0.12
MgO	10.80	2.12	1.50	1.11	0.79	0.47	1.81	1.51	0.14
CaO	4.42	5.60	5.48	4.10	3.64	5.04	5.81	4.62	0.78
Na_2O	3.05	3.10	4.05	4.35	4.09	4.34	4.80	3.86	2.87
K_2O	3.51	0.92	4.71	4.75	4.77	3.84	5.74	5.05	4.83
P_2O_5	0.33	0.45	0.68	0.56	0.54	0.54	0.75	0.36	0.06
L.O.I.	6.67	5.75	0.78	0.80	0.90	0.47	1.09	1.27	1.08
Total	100.07	99.81	100.01	100.00	99.99	100.72	100.44	99.13	100.08

1, high-Mg shoshonite; 2, andesite; 3-4, latites; 5-6, trachydacites; 7-8, high-Ti latites; 9, ferrorhyolite

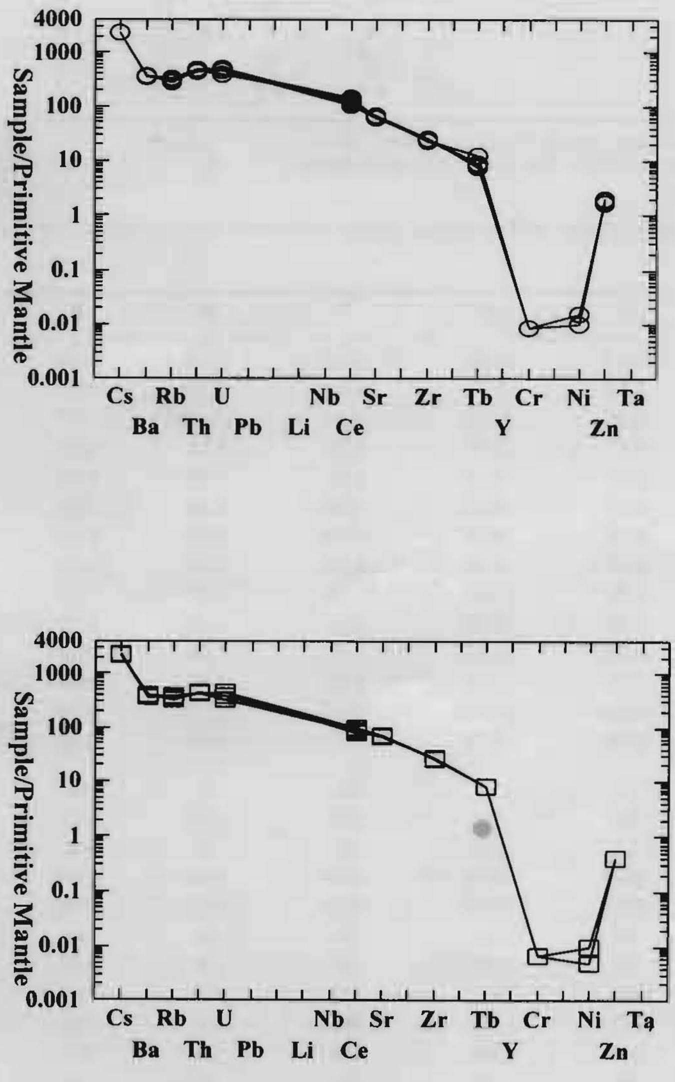

Fig. 9 – Trace elements distribution spidergrams of Kožuf volcanic rocks (according to Boev, 1988): *in the top* latites; *in the bottom* trachydacites.

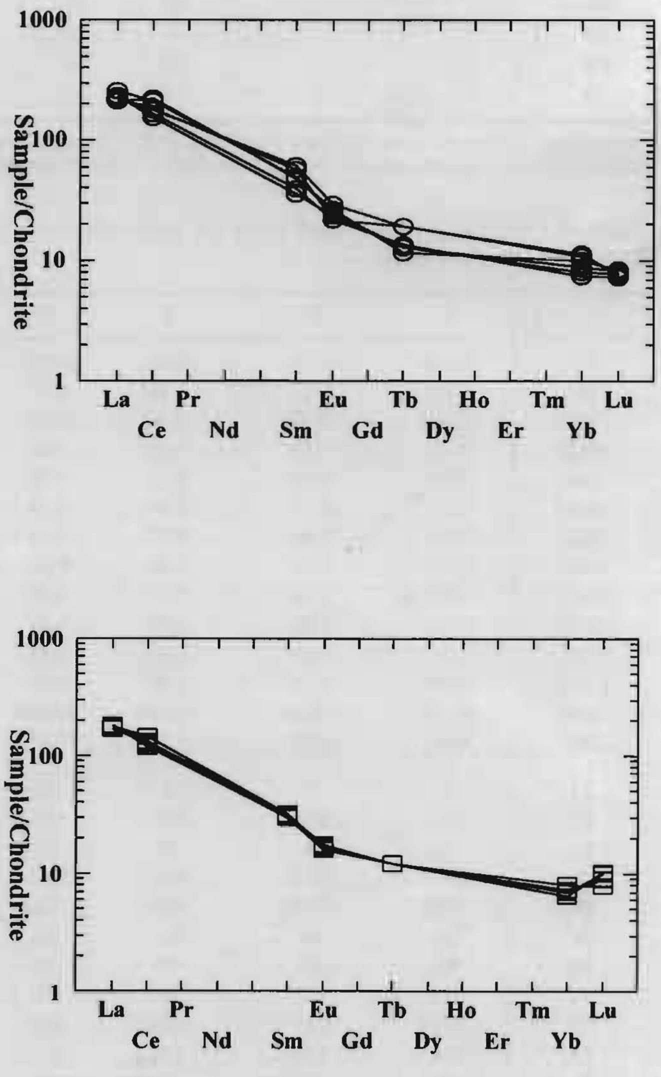

Fig. 10 – REE distribution patterns of the Kožuf volcanic rocks (according to Boev, 1988): *in the top* latites; *in the bottom* trachydacites.

Table 3 – Trace element content of the Tertiary orogenic magmatic rocks of Kratovo-Zletovo, Bućim-Borov Dol and Kožuf areas.

	Analyses from Table 1								Analyses from Table 2			
	4	5	7	10	11	14	15	17	3	4	5	6
Rb	113	100	146	300	330	49	149	108	174	180	190	200
Cs				11	5.1				42	41	40	39
Sr	1095	877	695	n.d.	n.d.	1392	830	1048	1100	1170	1250	1250
Ba	776	908	942	1400	1500	1161	1154	1734	1800	1760	1950	2100
Y	24	23	29	n.d.	n.d	26	29	29	34	34	26	23
Nb	1	6	7	n.d.	n.d.	n.d.	7	4	7	6	4	1
Zr	110	100	140	460	200	130	140	130	200	210	210	220
Cr				50	160				26	25	20	20
Zn				320	370				80	100	20	20
Ni				20	20				30	20	10	20
La				51	49				85	85	63	66
Ce				100	110				145	157	125	115
Sm				6.9	7.1				8.13	9.1	7.2	6.8
Eu				3	1				2.0	1.9	1.42	1.38
Tb				1	0.8				0.75	0.78	0.7	0.7
Yb				3	3				2.01	1.85	1.8	1.7
Lu				0.3	0.2				0.30	0.28	0.34	0.38
Th				27.4	28.8				28	31	28	28
U				5.9	6.5				8	9	6	8
Sc				28	27				15	10	15	10
Hf				7	10				6	5	5	4
Co				28	32				20	20	10	10
Ta				1.5	1.2				0.8	0.8	0.7	0

The numbers correspond of the analyses from Table 1: 4-7, Kratovo-Zletovo volcanic and 10-11, plutonic rocks; 14-17, Bućim-Borov Dol (unpubl. data of Boev); from Table 2 - Kožuf area (Boev et al., 1997), 3-4, latites; 5-6, trachydacites.

Table 4 – Major (in %wt) and trace (in ppm) elements contents of the within-plate mafic volcanic rocks (according to Kononova et al., 1989).

	1	2	3	4	5	6	7	8	9
SiO_2	46.56	47.39	48.49	50.00	49.73	50.82	48.95	53.34	55.94
TiO_2	2.65	2.42	1.24	1.25	1.53	1.61	1.20	1.28	0.83
Al_2O_3	13.90	14.92	12.49	12.80	11.82	11.26	11.81	13.12	17.57
Fe_2O_3	5.22	7.54	7.91	4.12	1.33	3.87	4.59	3.71	4.42
FeO	3.38	1.24	0.21	4.06	6.66	4.17	3.91	3.90	2.48
MnO	0.12	0.13	0.09	0.14	0.13	0.13	0.14	0.14	0.08
MgO	7.16	7.40	9.67	9.44	9.30	9.55	10.60	8.09	4.64
CaO	8.60	7.98	7.38	7.47	8.99	8.16	8.56	6.04	6.74
Na_2O	3.07	3.36	2.53	2.28	1.95	1.67	2.10	3.33	2.12
K_2O	6.14	5.48	6.75	6.27	5.91	5.67	3.71	5.51	4.38
P_2O_5	1.67	1.62	1.66	1.57	0.03	1.37	1.47	1.04	0.12
L.O.I.	1.58	0.52	1.58	0.60	2.62	1.72	2.96	0.50	0.66
Total	99.30	100.00	100.00	100.00	99.60	100.00	99.46	101.30	99.98
$Mg^{\#}$	0.64	0.65	0.73	0.71	0.70	0.72	0.72	0.69	0.69
Li	5	7	8	12	12	13	21	8	n.d.
Rb	411	274	375	366	311	165	155	155	231
Cs	11	9	29	24	12	2	17	7	n.d.
Sr	1410	1512	1015	1015	1099	1010	1074	930	1350
Ba	4750	4310	2601	2344	2512	2160	1525	2512	2780
Y	43	44	41	38	40	35	33	34	43
Nb	28	25	17	17	14	16	12	18	10
Zr	385	374	293	299	409	443	326	307	513
Cr	170	137	1440	342	342	390	363	212	390
Ni	150	150	126	212	118	141	188	149	173
Co	29	30	23	29	35	35	35	24	25
V	213	224	n.d.	147	n.d.	196	282	180	151

Phonothephrites from: 1-2, Štip area; 3-4, Kumanovo area; 5-6, Demir Kapia area; 7, absarokite from Skopje; 8, shoshonite from St. Nikole; 9, shoshonite from Štip.

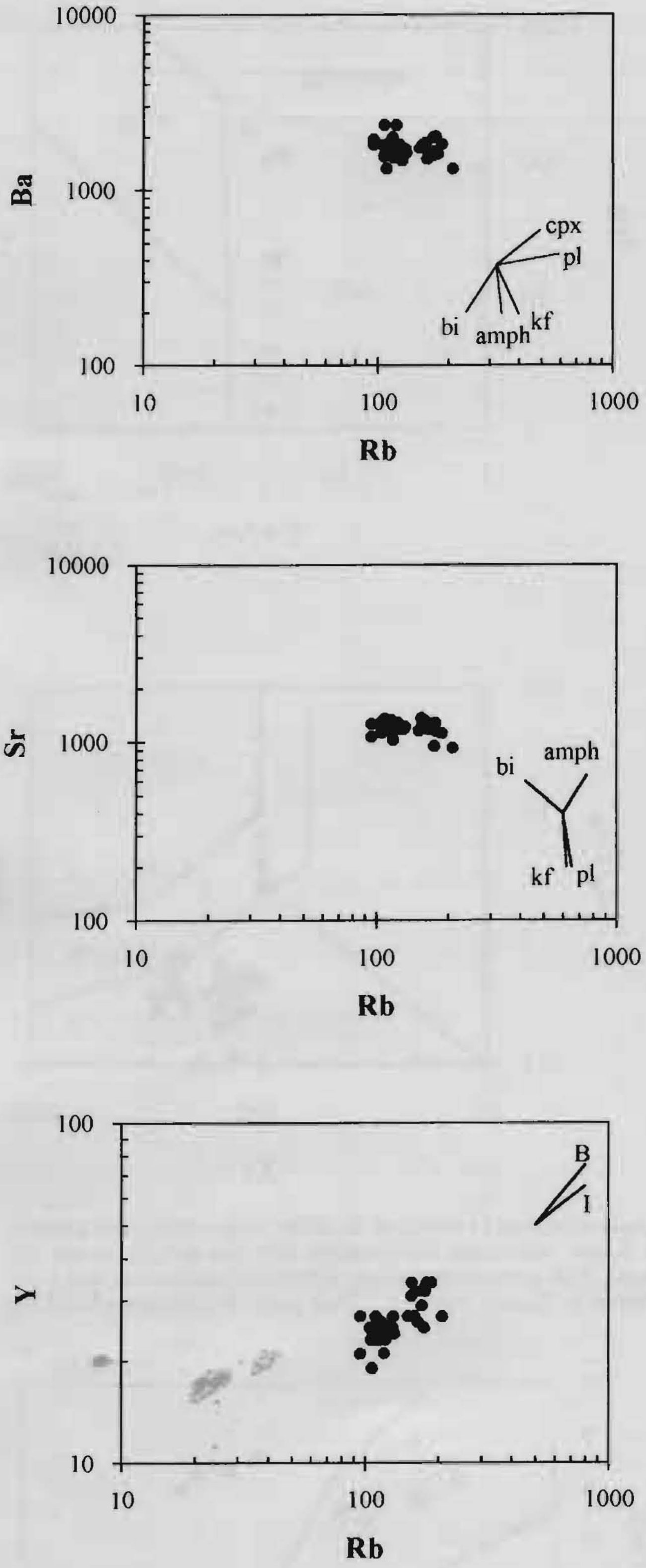

Fig. 11 – Rb vs. Ba, or Sr, or Y contents of Kozuf latites and trachydacites.

In the right corner: modeled fractionation vectors in the Rb vs. Ba and vs. Sr diagrams for biotite, clinopyroxene, plagioclase, K-feldspar and amphibole (after Harris et al., 1983); theoretical fractionation vectors in basic (B) and intermediate (I) magmas for 50% crystallization of plagioclase+clinopyroxene+olivine assemblages in the Rb vs. Y diagram (after Pearce et al., 1990). For the symbols of the rocks see Figure 6.

A similar hypothesis for the origin of magma in the transect Rila-Osogovo-Besna Kobila areas can be assumed with the following stages:

– magmas did not form in the nonvolcanic part (present day Serbo-Macedonian and Rhodopes massifs) of the Late Cretaceous island arc system during the northeast oriented subduction of the Tethyan ocean under Eurasia. Only metasomatization of the subcontinental mantle took place due to ascent of slab fluid (phlogopitized in the depth more then 100 km and amphibolitized in lesser depth according to the model of Pearce et al., 1990);

– during the complete closure of the Tethyan ocean at the end of the Cretaceous and the beginning of the Paleogene, continental collision precluded the subduction process and caused delamination and break-off of delaminated ocean crust. Because the ocean crust was heavier it sank into the mantle upwelling of a thermal diapir;

– the thermal diapir, reaching the phlogopitized mantle in a depth more then 100 km and located far from the ocean suture in the northeastern part of the studied transect (corresponding to the location of Rila pluton). The melts of higher K contents are formed. After that the thermal diapir rich the amphibolitized mantle in the central part of the studied transect (corresponding to the location of Osogovo-Besna Kobila area). The melts of lower K contents are formed. However, the thick crust (over 50 km according to Shanov & Kostadinov, 1992) served as a filter, which precluded their ascent to the surface. A better example of this process was described in the central part of the Andes where thick crust is stoped the access of mafic magma to the surface (Thorpe et al., 1982);

– in the crust itself these mantle-derived magmas provoke fusion and formation of granitoid magmas. In Rila area, where the crust was thicker (50 km) they did not reach the surface and the Rila plutons were formed, whereas the crust is thinner (40-45 km in Osogovo-Besna Kobila area) subvolcanic bodies intruded and volcanism occurred occasionally on the surface.

The $^{87}Sr/^{86}Sr$ ratio of these magmatic rocks is high (in Osogovo-Besna Kobila area up to 0.7102-0.7106, Boev et al., 1997), indicating that they are typical crustal product.

In the southwestern part of the studied transect, the Kratovo-Zletovo, Bučim-Borov Dol and Kožuf volcanic areas have typical island-arc characteristic (Figs. 6, 13). Their $^{87}Sr/^{86}Sr$ ratio is higher that 0.705 what is a characteristic feature of island arc magmas, products from subducted sediments, ocean crust and metasomatized mantle: 0.7063-0.7077 for Kratovo-Zletovo, 0.7069 for Bučim-Borov Dol and 0.7085-0.709 for Kozuf (Boev et al., 1997).

We relate these occurrences to Late Tertiary Aegean subduction (Fig. 16) as active continental margin Andean type. The described Macedonian magmatic rocks are situated in the rear part of the supra-subduction area in which mildly alkaline magmas formed. The subduction process in the Miocene and Pliocene moved to the south and southwest probably due to the extension in the North Aegean region (Gautier et al., 1999) causing migration of volcanic activity in Macedonia in the same direction – from Kratovo-Zletovo to Kožuf area.

The within-plate K to ultra-K mafic volcanic rocks are a typical product of the asthenosphere, taking into account the very high Ni and Cr contents and high Mg# (64-72) of the rocks and of the olivine phenocrysts too (80.5-92 – Sveshnikova et al., 1986). The lack of plagioclase phenocrysts in the mafic rocks indicates high-pressure

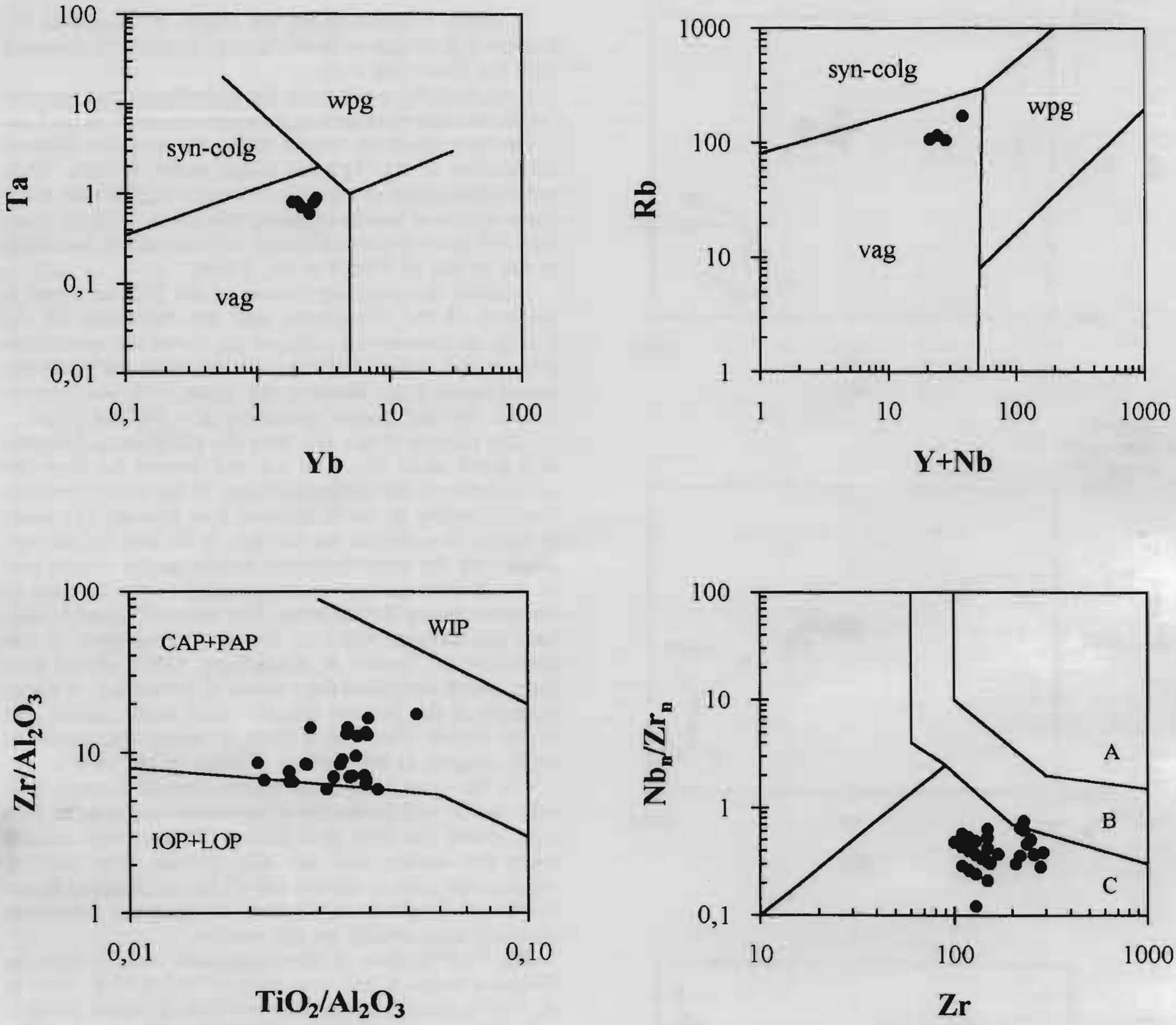

Fig. 12 – Discrimination diagrams for Kozuf trachydacites (the upper range) according to Pearce et al., 1984 (wpg-within plate granites, syn-colg-syncollision granites and vag-volcanic arc granites) and for Kozuf latites and trachydacites (the low range): *in the left*, according to Müller et al., 1992 (WIP – within plate, CAP-continental arc, PAP-postcollisional arc, IOP-initial oceanic arc and LOP-late oceanic arc magmatic rocks) and *in the right* according to Thieblmont & Tegyey, 1994 (A-within plate, B-collision related and C-subduction related magmatic rocks).

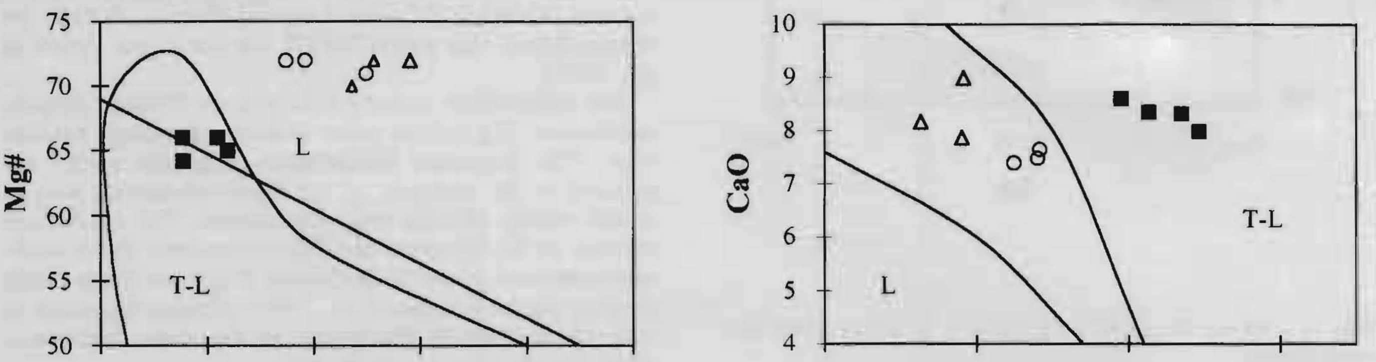

Fig. 13 – Parts of SiO_2 vs. Mg# diagram of Bogatikov et al. (1985) and of Al_2O_3 vs. CaO diagram of Foley et al. (1987) for ultra-K rocks with the points of Macedonian within plate mafic volcanic rocks: triangles, Demir Kapia area; circles, Kumanovo area; filled squares, Štip area. Series: L-lamproitic, T-L-tephrite-leucitic (after Kononova et al., 1989).

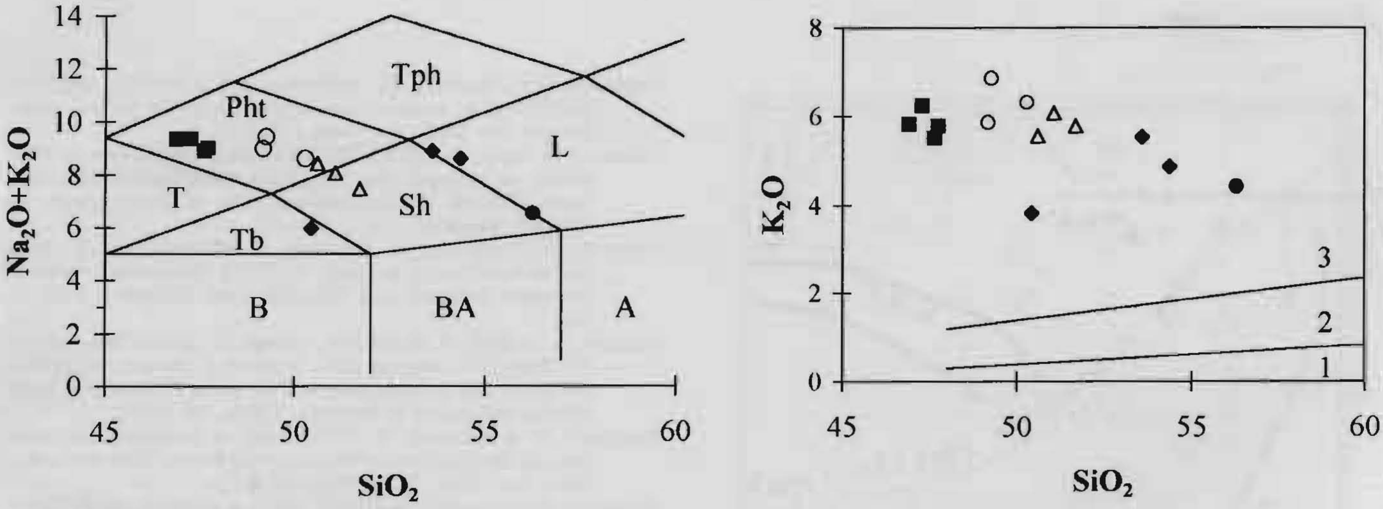

Fig. 14 – TAS (*left*) and SiO_2 vs. K_2O (*right*) diagrams (after Le Maitre, 1989) for the within plate volcanic rocks according to the data of Kononova et al. (1989); for the symbols see Figure 13, also filled zhombs, trachybasalts and shoshonites from Skopje - St. Nikole area; filled circle, shoshonite from Štip area. B-basalt, BA-basaltic andesite, A-andesite, Tb-trachybasalt (absarokite), Sh-shoshonite, L-latite, T-tephrite, Pht-phonotephrite and Tph-tephrophonolite.

(>30 km) crystallization of the phenocrysts (Green & Hibbson, 1970).

The high contents of LIL elements and the very high Ba/Nb ratio (107-180) witness an intermixing of asthenospheric melts and metasomatized magma (Downes et al., 1995). The origin from metasomatized magma is indicated also by the chondrite normalized pattern of the trace elements (Kononova et al., 1989) which is close to the theoretical curve of their affinity to an aqueous fluid, reflecting the mobility of elements in this fluid (Pearce et al., 1981).

The chemical and mineral composition of these mafic rocks is very close to the volcanic rocks of the ultra-K series in the Toscana magmatic province (Italy) and in particular to the volcanic rocks at Vico and Vulsini (Serri et al., 1993). That's why we can apply the genetic three-component model (mantle+two metasomatic components) of these authors, developed for the Toscana province for the Macedonian within-plate rocks.

According to Serri et al. (1993) model, the mantle component of these ultra-K magmatics is rich in phlogopite werhlite or olivine clinopyroxenite to phlogopite lherzolite. The two metasomatic components originate from sediments transported into the upper mantle during subduction of oceanic crust. The first component is rich in K and Si, and poor in Ca, Na and Sr (probably terrigenous sediments or acidic granulites). The second is rich in Ca and Sr, poor in Si and shows low Ce/Sr ratio (probably marine carbonate rocks). An asthenospheric diapir rises in the back part of the NE oriented Aegean subduction. Reaching the metasomatized subcontinental mantle it initiates partial melting. The complete melting of the phlogopite in it leads to formation of K-rich magmas (Wilson & Downes, 1995).

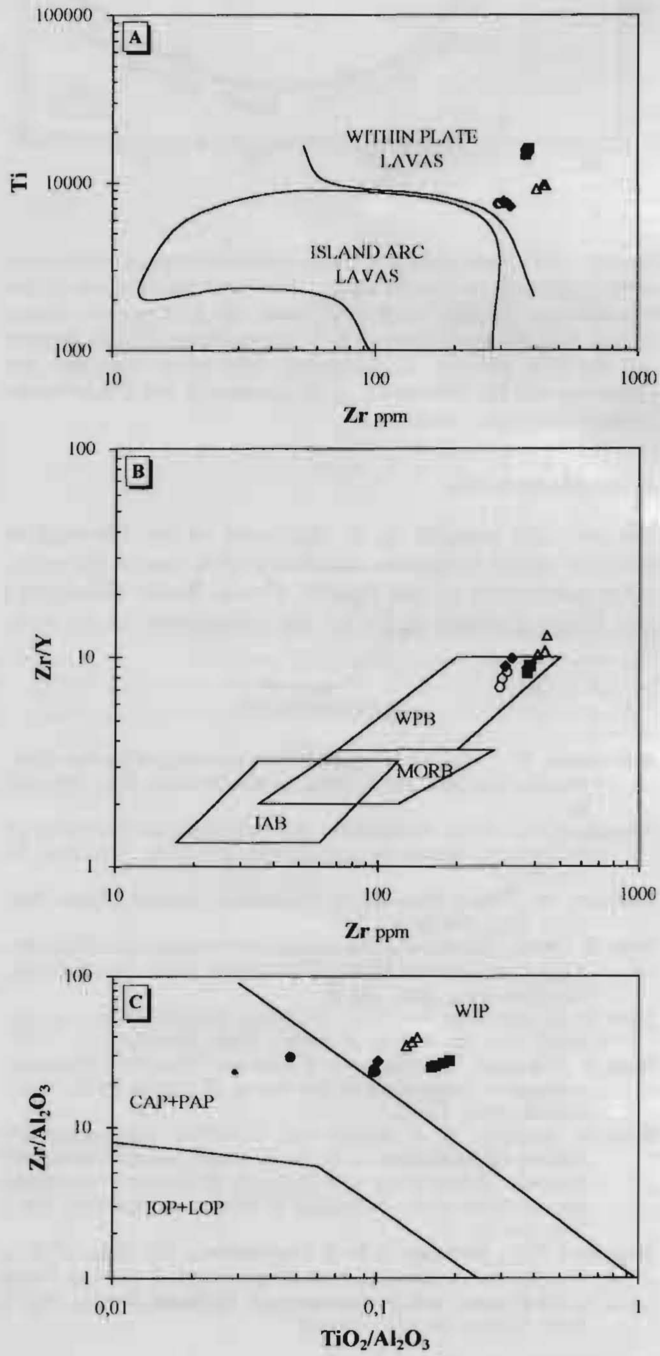

Fig. 15 – Discrimination diagrams (A, according to Pearce et al., 1981; B, according to Pearce & Norry, 1979 and C, according to Müller et al., 1992) with the points of Macedonian within plate mafic volcanic rocks (for the symbols see Figure 13). The data are from Kononova et al. (1989). WPB-within plate basalts, IAB-island arc basalts and MORB-mid-ocean ridge basalts; WIP-within plate, CAP-continental arc, PAP-postcollisional arc, IOP-initial oceanic arc and LOP-late oceanic arc magmatic rocks.

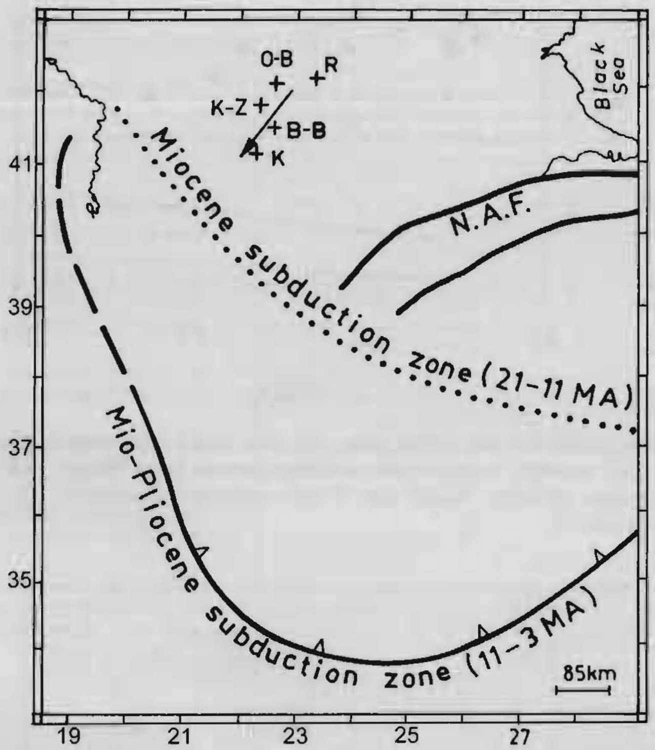

Fig. 16 – Schematic sketch of the positions of Aegean subduction zone (according to Gautier et al., 1999) and the location of the Macedonian Tertiary magmatic areas (O-B, Osogovo-Besna Kobila; K-Z, Kratovo-Zletovo; B-B, Bucim-Borov Dol; K, Kozuf) and the Rila plutons, R (Bulgaria). The arrow indicates the migration and the increasing of K contents in the Macedonian Tertiary magmatic rocks.

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