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# Extension-related Miocene calc-alkaline magmatism in the Apuseni Mountains, Romania: Origin of magmas

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## Abstract

The Miocene magmatism of the Apuseni Mountains in the Carpatho-Pannonian area hosts some of Europe's largest porphyry epithermal Cu–Au ore systems associated with shallow subvolcanic intrusions. Detailed field observations combined with K–Ar ages, geochemical analyses, Sr–Nd isotopes and paleomagnetic data constrain a model for the geotectonic evolution and processes of melt generation that may account for the exceptional mineralizing potential of the magmatic activity in this region. The magmatic activity developed mainly between 14.7 and 7.4 Ma and after a gap ceased at around 1.6 Ma. Geotectonic conditions do not support contemporaneous subduction processes, but were represented by transtensional and rotational tectonics, which generated horst and graben structures and favoured the generation and ascent of magmas. The “subduction signature” of the magmas emphasizes the significant involvement of fluids (mantle lithosphere and/or lower crust) inherited during previous geodynamic events. The mechanism of magmatogenesis is considered to be related to decompressional melting (various degrees of) of a heterogeneous source situated at the crust–lithosphere mantle boundary. Mixing with asthenospheric melts generated during the extension-related attenuation of the lithosphere may also be implied. The evolution from normal to adakitic-like calc-alkaline and alkaline magmas generally is time-dependent as a consequence of variable fluid-present melting. Fractional crystallization-assimilation processes in shallow magma chambers are suggested for early magmatism but were almost absent from later magmatism, which related to an increasingly extensional regime. The youngest alkalic (shoshonitic) magmatism (1.6 Ma) is asthenosphere-derived, but in a different extensional event, being almost coeval with the OIB-like alkali-basaltic magmatism (2.5 Ma) occurring along the South Transylvanian fault. The fluid-present melting of the source seems to be the critical factor for the presence of the copper-gold-bearing mineralizing fluids.

**Keywords:** Romania, Apuseni Mountains, Miocene, calc-alkaline, alkaline, adakite-like, extension.

## Introduction

Neogene calc-alkaline and alkaline magmatic rocks in the Apuseni Mountains (Romania) crop out either in intra-mountain basins such as the ca. 100 km long Zarand-Brad-Zlatna Basin, or in a cluster of magmatic bodies that are orientated roughly NW–SE (Fig. 1). The isolated position of these magmatic products with respect to the Carpathian fold-and-thrust belt, some 200 km behind the East Carpathian main volcanic arc, is striking. Although a “subduction signature” has been already demonstrated for the volcanic rocks (e.g. Borcoș et al., 1972; Roșu et al., 1996), geotectonic

models, which invoke subduction-related processes (e.g. Rădulescu and Săndulescu; 1973; Bocaietti et al., 1973; Bleahu, 1974) have so far failed to explain the unusual position of this volcanism. In contrast, some authors (Roșu et al., 1996; Balintoni and Vlad, 1998; Seghedi et al., 1998) have suggested that these Neogene calc-alkaline to alkaline magmas were generated in response to extension of the lithosphere.

Neogene magmatism is accompanied by significant metallogenetic activity, particularly for Cu–Au (Mo), together with Au–Ag ± Te and base metal mineralization (Udubașa et al., 2001). However, not all the intrusions are accompanied by

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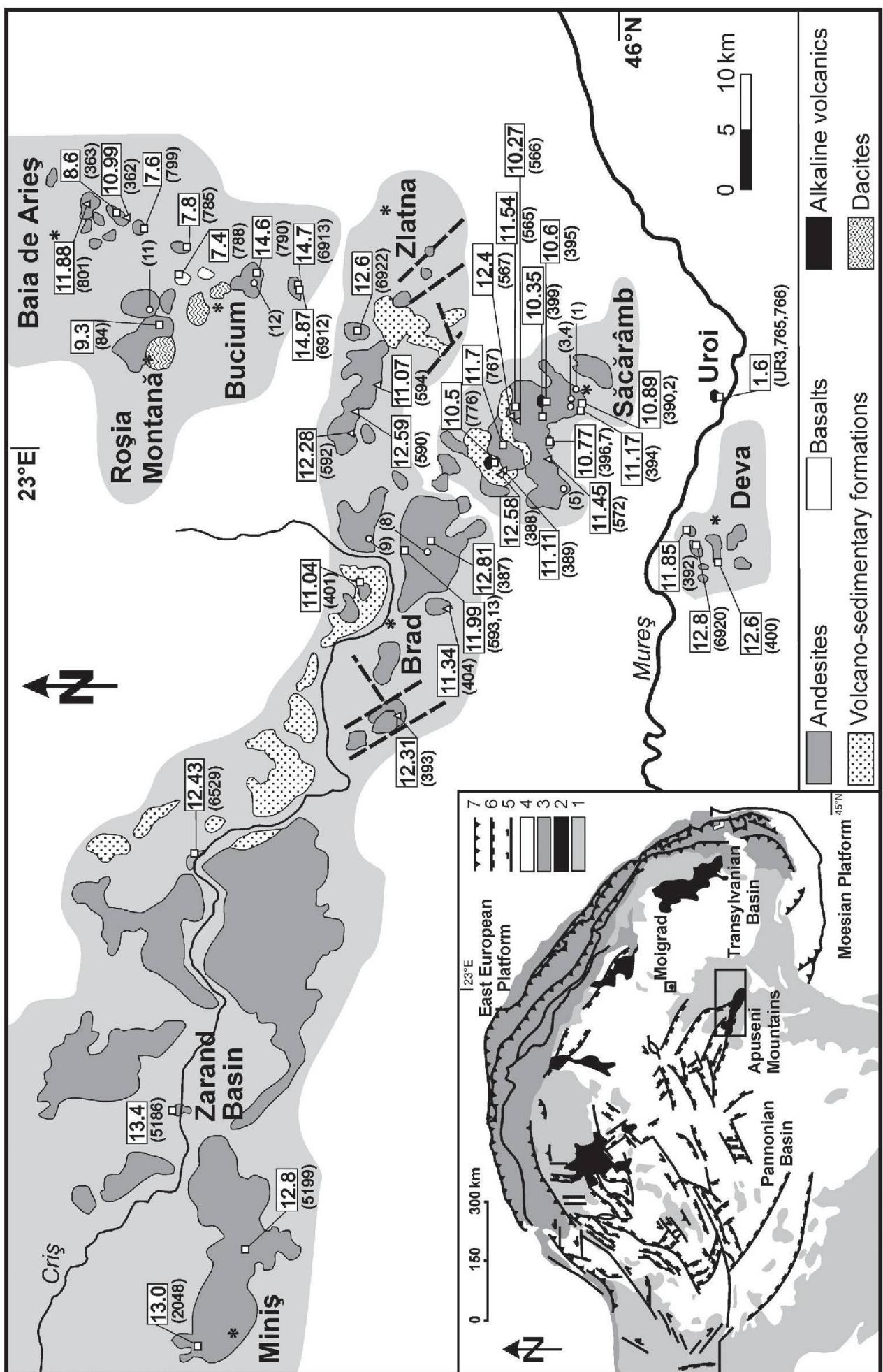


Fig. 1

mineralization. A variety of mineral deposit types are present including porphyry copper, low-sulfidation and rarer high-sulfidation epithermal veins, breccia pipes and replacement bodies.

The purpose of this paper is to describe and interpret the nature and origin of calc-alkaline and alkaline Neogene magmas in the Apuseni Mountains. The presence of adakite-like characteristics as pointed out by Roșu et al (2001) requires reconciling the “subduction signature” of the magmatic rocks with their unusual tectonic setting. This contribution provides a new insight into the generation of calc-alkaline magmatism in a tectonic setting, which was not subjected to subduction, on the basis of a review of existing and new geochemical and isotopic data.

### Geologic and tectonic setting

In recent geotectonic models of the Carpathian-Pannonian region (e.g. Csontos, 1995), the Apuseni Mountains are part of the Tisia (Tisza-Dacia) lithospheric block, whose eastward translation, along with the Alcapa block, constituted the active driving force of the Carpathian collision and consequent orogenesis during Cretaceous to Miocene times (Royden, 1988; Săndulescu, 1988; Royden and Burchfiel, 1989; Csontos et al., 1992; Csontos, 1995). Paleomagnetic data demonstrate that during Eocene-Early Miocene times, the Tisia block experienced a 20° clockwise rotation at the same time as the Alcapa block underwent counterclockwise rotation. The Apuseni Mountains underwent a further 60° clockwise rotation after cessation of the counterclockwise rotation of the Alcapa block. This rotation started around 14 Ma, diminished at 13 Ma to around 28° and ceased at around 12 Ma (Panaiotu, 1998, 1999).

The northern Apuseni Mountains have a basement of metamorphic rocks and associated anatetic Hercynian granites, belonging to the Inner Dacides (Ivanovici et al., 1969, 1976; Săndulescu, 1984; Balintoni, 1994, 1997), with a Permian post-tectonic sedimentary and volcanic cover (Stan, 1987). The southern Apuseni Mountains are dominated by the Tethyan ophiolitic suture zone (Săndulescu, 1984), including Lower-Jurassic pil-

low lava and sheeted dykes (Savu, 1996; Nicolae, 1995) and Jurassic calc-alkaline volcanics (Nicolae, 1995), as well as Lower to Upper Jurassic ocean-floor and continental shelf sediments, and Lower Cretaceous flysch and wildflysch (Lupu, 1976). Calc-alkaline intrusive rocks pierced the ophiolitic basement during Late Jurassic-Early Cretaceous times (Ştefan, 1986; Nicolae, 1995).

During the Late Cretaceous-Paleogene times, “banatitic” magmatism developed along a N-S alignment crossing through both the Apuseni Mountains and the western part of the South Carpathians and Dinarides (Ştefan et al., 1992; Berza et al., 1998), and was partly coeval with Maastrichtian-Paleocene molasse deposition. Post-Paleocene tectonic uplift of the whole area interrupted sedimentation until it resumed in Early-Middle Miocene times. The post-suture (i.e. post-Laramian) evolution of the Apuseni Mountains is related to brittle tectonics (Royden, 1988; Săndulescu, 1988) during the Paleogene-Neogene interval as a consequence of its behaviour as a single rigid block (Tisia block). This tectonic style, mainly marked by horst and graben structures, which are visible mostly along the northern and western edges of the Apuseni Mountains (Fig. 1), is a consequence of the Neogene extensional development of the neighbouring Transylvanian and Pannonian Basins (Fodor et al., 1999), as well as of the translational and rotational movements of the Tisia block (Pătrașcu et al., 1994; Csontos, 1995; Panaiotu, 1998; Seghedi et al., 1998; Roșu et al., 2000). From Badenian times the Apuseni Mountains formed the high-relief part of the Tisia block, left upstanding after the general subsidence of the Pannonian and early Transylvanian basins. Development of the Neogene volcanism was closely related to this extensional evolutionary stage of the Apuseni Mountains, as part of the Tisia block.

### Spatial distribution of Neogene volcanic rocks

The overall spatial distribution of Neogene magmatic rocks in the southern Apuseni Mountains (Fig. 1) strongly suggests a NW-SE oriented development of the igneous activity and a connection with coeval Miocene sedimentation in exten-

Fig. 1 Sketch map of Neogene volcanic rocks in the Apuseni Mountains and location of analyzed samples: open squares—K-Ar ages and geochemical data; open triangles—K-Ar ages only; open circles—geochemical data only. All ages are in Ma (boxed), samples numbers in brackets refer to Table 1. Light grey background shading: grouping of volcanic centres as in Fig. 2. Inset: geological sketch of Carpathian-Pannonian realm showing Apuseni Mountains and Moigrad occurrence (boxed). Regional map with schematic deformation structures during Middle Miocene after Mațenco (1997). Symbols: 1a—internal basement; 1b—ophiolite belt; 2—Neogene magmatic rocks; 3—thin-skinned belt; 4—autochthonous foreland and intra/inter-mountain depressions; 5—dextral/sinistral fault; 6—normal fault; 7—thrust fault.

sion-controlled small basins (Roșu et al., 1997; Ciulavu, 1999). However, closer examination of the spatial occurrence of the outcrops reveals a more complicated pattern (Fig. 1). Most magmatic rocks are distributed along a WNW–ESE trend, especially between Miniș in the west and Zlatna in the east, within a ca. 100 km long area. Many of the igneous rocks are concentrated in the eastern half of the area and apparently follow a NNE–SSW trend, ca. 60 km long, between Baia de Arieș in the north and Deva in the south (Fig. 1). These two “alignments” (we use the term “alignment” without any genetic or tectonic connotation) cross each other at the south-eastern end of the WNW–ESE “alignment”.

Along the NNE–SSW “alignment” magmatic rocks occur in several volcano-intrusive structures (Roșu et al., 1997). From north to south they are: (1) Baia de Arieș, Roșia Montană–Bucium; (2) Zarand, Brad, Zlatna; (3) Săcărâmb and (4) Deva, where we include also the youngest activity at Uroi (Fig. 2). There is an additional, isolated, small-scale occurrence at the northernmost edge of the Apuseni Mountains at Moigrad, located about halfway between the Southern Apuseni and the Oaș–Gutâi Neogene volcanic areas (Fig. 1) (Ştefan et al., 1986).

## Petrography

The Neogene magmatic rocks in the Apuseni Mountains range from basaltic-andesites to dacites, with subordinate occurrences of alkaline affinity. However, andesite is the most common and volumetrically the prevalent rock-type (Fig. 1). Basaltic andesites are present as two small-scale occurrences in the Detunata hills, but also occur in the Zarand area. These rocks are slightly porphyritic, with plagioclase, augite, olivine and resorbed amphibole microphenocrysts in a groundmass of plagioclase microlites and augite, magnetite, ilmenite, olivine, resorbed amphibole and glass (Savu et al., 1993).

Andesites display a large spectrum of varieties. Two-pyroxene andesites are abundant in the Zarand Mountains. They contain abundant clinopyroxene and orthopyroxene phenocrysts, generally showing corroded and opaque rims. Amphibole ± pyroxene andesites are present mostly in the eastern half of the region. They are largely porphyritic and have plagioclase, amphibole, clinopyroxene, and sometimes, corroded quartz and accessory Fe–Ti oxides, apatite, zircon, sulphides and Cr-spinels. Rare garnet-bearing varieties can be found at Zlatna, Bucium and in the Brad areas.

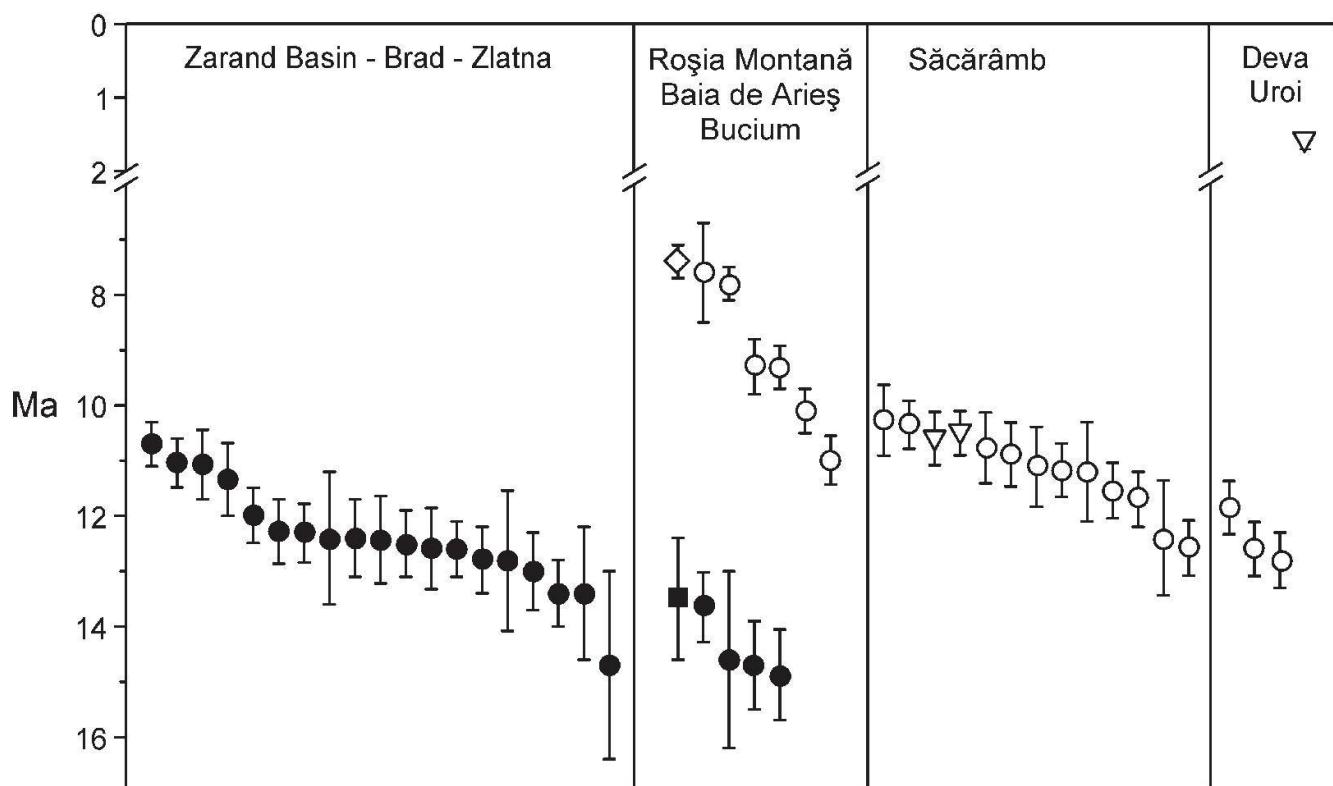


Fig. 2 Time evolution of volcanic activity in the Southern Apuseni Mountains. Symbols for rock types (black symbols are normal calc-alkaline rocks, open symbols are adakite-like calc-alkaline rocks): squares—dacites; circles—andesites; diamond—basalts andesites; triangle—alkaline rocks. (see Fig. 1 for area locations). K–Ar ages from Pécsay et al. (1995), Roșu et al. (1997) and this study.

Amphibole-biotite  $\pm$  pyroxene andesites occur in the Săcărâmb, Deva and Baia de Arieș areas. The main phenocrysts are plagioclase, amphibole, biotite, two pyroxenes and the same accessory minerals as for the amphibole  $\pm$  pyroxene andesites. These rock types also often contain quartz phenocrysts. The spatial distribution of different types of andesites seems to follow a systematic trend, with the more basic pyroxene-bearing varieties in the west, and more acidic, amphibole and biotite-bearing varieties in the east and towards the extremities of the NNE–SSW “alignment”. Intermediate-type, amphibole-pyroxene andesites occur in between. Andesites and porphyritic microdiorites also occur as a cluster of small intrusive bodies at Moigrad. Their phenocryst assemblage includes plagioclase, clinopyroxene and sporadic orthopyroxene and amphibole, and sometimes biotite (Ştefan et al., 1986).

Altered dacites are present as clasts in  $\sim$ 15 Ma intra-basinal volcaniclastic and sedimentary deposits (Cioflica et al., 1966; Roșu et al., 1997). They display a phenocryst assemblage of plagioclase, quartz, biotite and minor amphibole. Dacite intrusions are mostly found in the Roșia Montană area, showing porphyritic textures with plagioclase, quartz, amphibole and biotite as phenocrysts, and magnetite, ilmenite, apatite and zircon as accessory minerals in a microgranular groundmass.

Besides the above-mentioned lithologies, which are characterized by normal calc-alkaline compositions, there are a few small-scale occurrences of alkaline rocks in the south-eastern part of the region. Two of them occur in the Săcărâmb area (trachyandesite and microdiorite plotted in the basaltic trachyandesite field in Fig. 3) and the third, a trachyandesite body, at Uroi in the south-eastern extremity of the region (Fig. 3). Trachyandesite from Zâmbrița is slightly porphyritic, dark, and contains plagioclase, amphibole, two pyroxenes, biotite and quartz as microphenocrysts in a microgranular groundmass of the same composition, as well as apatite, zircon, magnetite, ilmenite and Cr-spinels as accessories. The microdiorites from Părâul lui Toader contain several generations of plagioclase, amphibole substituted either by clinopyroxene or an aggregate of clinopyroxene, plagioclase, quartz and biotite, fresh amphibole, clinopyroxene along apatite, zircon, magnetite, ilmenite and sulphides as accessories. The Uroi trachyandesites (according to a TAS diagram) or shoshonites (according to a  $K_2O$  vs.  $SiO_2$  diagram) closely resemble the shoshonites, which occur at the southeastern end of the East Carpathian volcanic range (Seghedi et al., 1986, 1987; Mason et al., 1996). They display a disequilibrium mineral assemblage containing Ti-augite, hypersthene, amphibole and biotite

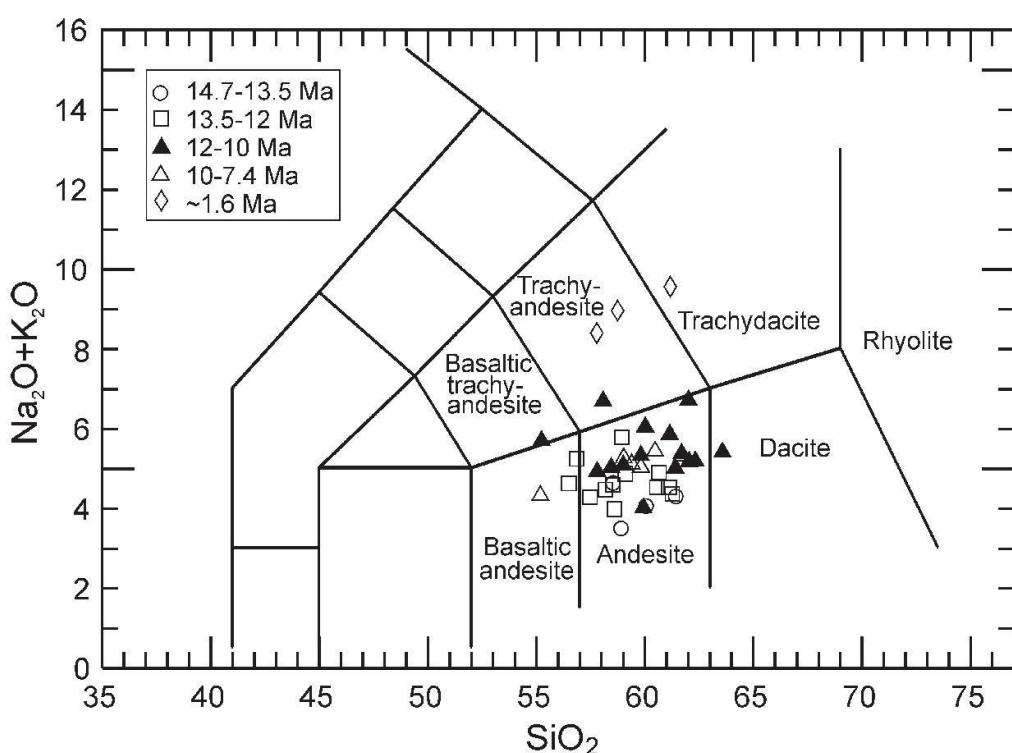


Fig. 3  $SiO_2$  vs.  $K_2O + Na_2O$  diagram for Neogene Apuseni magmatic rocks (classification after Le Bas et al., 1986). Legend: Rocks in the age interval: 14.7-13.5 Ma — open circles; 13.5-12 Ma — open squares; 12-10 Ma — filled triangles; 10-7.4 Ma — open triangles;  $\sim$ 1.6 Ma — open diamonds.

Table 1

Sample	Coordinates		Locality	Lithology	K (%)	$^{40}\text{Ar}_{\text{rad}}$ (%)	$^{40}\text{Ar}_{\text{rad}}$ (ccSTP/g)	K-Ar ages	Source
Roșia Montană - Bucium – Baia de Arieș									
6913	46.20979	23.17700	Bucium S	mδ am px	1.14	29.2	$6.618 \times 10^{-7}$	$14.7 \pm 0.8$	2, 5
6912	46.19866	23.17551	Bucium S	mδ am px	0.94	33.3	$5.401 \times 10^{-7}$	$14.87 \pm 0.82$	4
790	46.23337	23.21397	Citera	α am px	1.21	12.8	$6.916 \times 10^{-7}$	$14.6 \pm 1.6$	1, 5
801	46.36587	23.29209	Valea Lacului	α am bi	1.83	29.5	$8.481 \times 10^{-7}$	$11.88 \pm 0.59$	4
362	46.32415	23.28672	Poenița	α q am bi	1.25	61.0	$5.360 \times 10^{-7}$	$10.99 \pm 0.44$	4
363	46.32637	23.28811	Poenița	α q am bi	1.17	51.4	$4.245 \times 10^{-7}$	$9.31 \pm 0.39$	4, 5
84	46.31650	23.14720	Rotunda	α am px	1.41	33.7	$5.095 \times 10^{-7}$	$9.3 \pm 0.5$	1, 5
785	46.28560	23.24359	Geamăna W	α am px	1.27	55.8	$3.883 \times 10^{-7}$	$7.8 \pm 0.3$	2, 5
799	46.31692	23.24746	Şurligata	α am±px	1.20	12.3	$3.533 \times 10^{-7}$	$7.6 \pm 0.9$	2, 5
788	46.27841	23.19917	Detunata	αβ	1.30	37.3	$3.602 \times 10^{-7}$	$7.4 \pm 0.3$	1, 5
11	46.31279	23.15349	Varsii Mari	α am px					5
12	46.23293	23.18633	Bucium Tarnita	α am px					5
Zarand Basin – Brad - Zlatna									
117	46.22681	22.47834	Tălagiu	α am px	1.27	48.6	$6.623 \times 10^{-7}$	$13.4 \pm 0.6$	1, 5
5186	46.30802	22.35007	Dieci	α am px	1.01	16.2	$5.293 \times 10^{-7}$	$13.4 \pm 1.2$	1, 5
2048	46.32091	21.96474	Camna	α px	1.22	33.7	$6.195 \times 10^{-7}$	$13.0 \pm 0.7$	1, 5
387	46.08465	22.87728	Cireșata	α q am bi±px	1.05	14.6	$5.236 \times 10^{-7}$	$12.81 \pm 1.27$	4, 5
5199	46.27848	22.08141	Chișindia	α px	1.18	43.3	$5.897 \times 10^{-7}$	$12.8 \pm 0.6$	1, 5
6922	46.13984	23.14123	Trămpoiele	mδ am px	1.75	64.7	$8.621 \times 10^{-7}$	$12.6 \pm 0.5$	2, 5
590	46.16259	23.00294	Ciungi- Stanija	α am px	1.56	27.4	$7.664 \times 10^{-7}$	$12.59 \pm 0.73$	4
6529	46.28914	22.52739	Vârfuri	α px	0.94	24.0	$4.559 \times 10^{-7}$	$12.43 \pm 0.79$	4, 5
393	46.12654	22.67750	Caracel - Tebea	α px	1.11	47.0	$5.319 \times 10^{-7}$	$12.31 \pm 0.53$	4
592	46.14741	23.03215	Ciungi- Stanija	α am px	1.31	37.9	$6.275 \times 10^{-7}$	$12.28 \pm 0.58$	4
593	46.11360	22.86792	Valea Arsului	α am px	1.27	51.8	$5.491 \times 10^{-7}$	$11.99 \pm 0.50$	4
13	46.11360	22.86792	Valea Arsului	α am px					5
404	46.09054	22.79085	Plesa-Brad	α q am bi	1.65	26.7	$7.298 \times 10^{-7}$	$11.34 \pm 0.66$	4
594	46.12860	23.09202	Almasu Mare	α am px	1.61	27.6	$6.948 \times 10^{-7}$	$11.07 \pm 0.63$	4
401	46.13872	22.84443	Brad -v. Steau	α q am bi±px	1.35	62.7	$5.813 \times 10^{-7}$	$11.04 \pm 0.44$	4, 5
8	46.04439	22.86531	Valea Arsului	α am px					5
9	46.13367	22.90482	Valea Rovina	α am px					5
Săcărâmb									
5	45.98372	22.94086	Magura Certej	α am px					3
388	46.02055	22.96518	Valea Draica	α am bi±px	2.02	63.4	$9.913 \times 10^{-7}$	$12.58 \pm 0.50$	4
567	46.02928	22.98649	Dealul Paua-N	α q am bi±px	1.23	17.2	$5.969 \times 10^{-7}$	$12.40 \pm 1.04$	4
767	46.03355	22.97372	Cetraș	α q am bi±px	1.05	51.5	$4.793 \times 10^{-7}$	$11.7 \pm 0.5$	2, 5
565	46.02607	22.99025	Voia	α am	1.49	47.6	$6.707 \times 10^{-7}$	$11.54 \pm 0.5$	4, 5
572	46.00054	22.97061	Dealul Padina	α q am bi±px	1.91	30.1	$8.532 \times 10^{-7}$	$11.45 \pm 0.62$	4
394	45.97580	23.02167	Goruniste Sac.	α q am bi±px	1.40	47.6	$6.109 \times 10^{-7}$	$11.17 \pm 0.48$	4, 5
389	46.15384	22.84610	Valea Draica	α q am bi±px	1.09	23.6	$4.753 \times 10^{-7}$	$11.11 \pm 0.72$	4
390	45.97362	23.03062	Zuckerhut	α q am bi±px	1.49	31.0	$6.327 \times 10^{-7}$	$10.89 \pm 0.58$	4
2	45.97362	23.03062	Zuckerhut	α q am bi±px					3
1	45.97280	23.03473	Calvaria	α q am bi±px					3
3	45.97321	23.02225	Sarcau	α q am bi±px					3
4	45.98205	23.03633	Haitau	α q am bi±px					3
396	45.99087	22.99621	Hondol-Baiaga	α am	1.31	26.1	$5.449 \times 10^{-7}$	$10.77 \pm 0.64$	4, 5
7	45.99085	22.99698	Hondol-Baiaga	α am					5
395	45.99974	23.02868	Pârâul lui Toader	mδ am px±bi	1.72	42.1	$7.113 \times 10^{-7}$	$10.6 \pm 0.48$	4, 5
776	46.04559	22.95348	Zâmbrița	ta	2.46	61.7	$1.011 \times 10^{-7}$	$10.5 \pm 0.4$	1, 5
399	46.00322	23.01034	Macris-Hondol	α q am bi±px	2.04	51.7	$8.231 \times 10^{-7}$	$10.35 \pm 0.43$	4, 5
566	46.02696	22.99028	Dealul Paua S	α q am bi±px	1.72	24.6	$6.887 \times 10^{-7}$	$10.27 \pm 0.64$	4
Deva									
6920	45.88672	22.87769	Serhediu	α am	1.14	56.9	$5.671 \times 10^{-7}$	$12.8 \pm 0.5$	2, 5
400	45.86854	22.85582	Nocet	α am bi	1.40	71.0	$6.482 \times 10^{-7}$	$12.6 \pm 0.49$	4, 5
392	45.88859	22.90082	Cetate	α am bi	2.09	58.6	$9.645 \times 10^{-7}$	$11.85 \pm 0.48$	4, 5
Uroi									
765	45.85776	23.04474	Uroi	ta					5
766	45.85776	23.04474	Uroi	ta					5
UR3	45.85776	23.04474	Uroi	ta	3.38	21.0	$2.099 \times 10^{-7}$	$1.6 \pm 0.1$	1, 5
Moigrad - Northern Apuseni Mountains									
2479	47.60156	23.54186	Moigrad	α am px+bi	0.93	13.3	$4.484 \times 10^{-7}$	$12.4 \pm 1.3$	4, 5

**Table 1** Sample location, rock type, lithology and K–Ar ages (whole rocks) for Neogene magmatic rocks in Apuseni Mountains. Abbreviation:  $\alpha$ —andesite,  $\alpha\beta$ —basaltic andesite,  $\tau\alpha$ —trachyandesite,  $m\delta$ —microdiorite,  $\nu$ —dacite; am—amphibole, bi—biotite, px—pyroxene, q—quartz. Sources of K–Ar ages are from: (1) Pécsay et al., 1995; (2) Roșu et al., 1997; (4) this study. Geochemical data are from: (3) Alderton et al., 1998 and Alderton and Fallick, 2000; (5) this study.

phenocrysts and accidental quartz xenocrysts, many with reaction rims (Savu et al., 1994).

### Sampling and analytical techniques

We used 40 samples for the geochemical and isotopic study (Table 2), some of which are already published in Roșu et al. (2001), Alderton et al. (1998) and Alderton and Fallick (2000). Most of the samples have been dated by the K–Ar method (Roșu et al., 1997; Pécsay et al., 1995a); results for some are presented for the first time here (Table 1). Sample selection took account of freshness (limited amount of hydrothermal alteration) spatial coverage, and lithological representation of Neogene magmatism in the Apuseni Mountains, plus Moigrad at its northern periphery. Thus, the data set can be considered as being representative for the Miocene igneous activity in the Apuseni Mountains.

For K–Ar age determination approximately 500 mg was used for Ar analysis. An Ar extraction line and a mass spectrometer were used for the Ar measurement. Approximately, 100 mg of the pulverized material was digested in HF with the addition of some sulphuric and perchloric acids for the K measurements. 100 ppm of Na and Li were added as buffer and internal standard. K concentration was measured with a digitalized flame photometer of OE-85 type manufactured in Hungary. The inter-laboratory standards HD-B1, GL-O, LP-6 and Asia 1/65 were used for calibration. Details of the instruments, the applied methods and results of calibration have been described elsewhere (Balogh, 1985). The results of the K–Ar age determination are summarized in Table 1. Analytical ages were calculated according to the constants of Steiger and Jäger (1977). All analytical errors represent one standard deviation (i.e. 68% analytical confidence level).

Major and trace element data (Table 2) were obtained on a Philips PW1480 X-ray fluorescence (XRF) spectrometer at the University of London XRF facility at Royal Holloway. Major element oxides were determined on fused glass discs and trace elements on pressed powder pellets, with matrix correction calculated from major element compositions. Analytical reproducibility for most trace elements is  $\pm 1$  ppm (2SD), but is about  $\pm 0.3$  ppm for Nb, Y and Rb. The REE elements La, Ce

and Nd have also been analyzed using this technique for all the samples.

REE concentrations (Table 3) were determined for selected samples by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) at Royal Holloway, using the method described by Walsh et al. (1981). Powdered samples (0.5 g) were dissolved in HF and HClO<sub>4</sub>. The residue was ignited and fused with NaOH, and the solution was passed through ion exchange columns, which separated and concentrated the REE before analysis. In-house standards were also analyzed to check the accuracy of the method.

Sr–Nd isotope ratios were determined using the VG-354 5-collector mass spectrometer at the University of London radiogenic isotope facility, with Nd analyzed as oxide (Thirlwall, 1991). Measured Sr and Nd isotope standard ratios for <sup>87</sup>Sr/<sup>86</sup>Sr on SRM987 and <sup>143</sup>Nd/<sup>144</sup>Nd on an in-house “Aldrich” standard were 0.710248 and 0.511420 (equivalent to La Jolla of 0.511857; Thirlwall, 1991). Results are shown in Table 2. No age corrections were made since they are not significant for <sup>87</sup>Sr/<sup>86</sup>Sr at  $\sim 10$  Ma.

### Age and time-space evolution

Neogene magmatic rocks in the Apuseni Mountains range in age from 14.7 to 7.4 Ma, except for one occurrence (Uroi), which is 1.6 Ma old (Pécsay et al., 1995a; Roșu et al., 1997, 1998, 2000). The general time-space evolution of volcanism is presented in Figs. 1 and 2. Compared with other Neogene volcanic areas of the Carpathians, the magmatic rocks in the Apuseni Mountains are roughly coeval with those from the Tokaj-Slanske area and Vihorlat-Beregovo-Oaș-Gutâi-Tibileș arc segment (Pécsay et al., 1995b, 2000; Roșu et al., 1997).

The earliest volcanic rocks are Lower Badeian (ca. 15 Ma old) dacitic tuffs, the age of which is inferred from stratigraphic relationships with paleontologically dated Miocene sediments (Roșu et al., 1997). Intermediate calc-alkaline magmatic activity has produced complex volcano-intrusive structures. Except for lava flows and associated volcaniclastic deposits in the Zarand Mountains, most of the porphyritic andesites occur as small intrusive bodies (Roșu et al., 1997).

Table 2 Chemical analyses of representative Neogene magmatic rocks from the Apuseni Mountains, Romania.

	ROSLA MONTANA-BUCIUM-BAIA DE ARIES										ZARAND-BRAD-ZLATNA									
	788	12	785	84	11	6913	799	790	363	6529	9	5186	2048	8	13	5199	6922	117	387	401
SiO <sub>2</sub>	55.20	58.91	59.04	59.4	59.83	60.06	60.47	61.43	61.7	57.48	58.61	58.52	58.19	59.10	59.94	60.56	60.67	61.13	61.27	63.58
TiO <sub>2</sub>	0.9	0.62	0.73	0.6	0.5	0.8	0.7	0.64	0.67	0.8	0.57	0.74	0.81	0.61	0.69	0.71	0.63	0.64	0.62	0.53
Al <sub>2</sub> O <sub>3</sub>	16.00	17.94	18.67	18.65	17.64	18.88	18.25	18.63	16.55	17.64	17.5	17.97	18.48	17.39	18.49	16.91	17.72	17.59	17.59	17.93
Fe <sub>2</sub> O <sub>3</sub>	6.83	6.81	5.59	5.59	4.84	6.78	5.20	6.05	5.62	7.92	6.92	7.12	6.86	7.48	6.86	7.14	7.19	6.36	6.18	4.53
MnO	0.15	0.15	0.12	0.14	0.15	0.17	0.14	0.13	0.14	0.16	0.14	0.13	0.12	0.06	0.1	0.11	0.18	0.14	0.14	0.24
MgO	6.36	3.08	3.55	2.69	2.04	2.19	2.95	2.45	2.93	3.63	3.55	3.18	2.61	3.23	1.83	3.19	2.54	2.8	2.62	1.50
CaO	9.71	7.74	7.19	7.31	6.62	7.44	6.33	7.06	5.87	7.8	7.21	7.29	7.72	5.72	5.89	7.09	6.73	6.73	7.43	5.43
Na <sub>2</sub> O	2.84	2.73	3.72	3.22	3.28	2.93	3.71	2.88	3.29	3.13	2.68	3.38	3.13	3.71	2.8	3.19	3.38	3.04	3.05	3.49
K <sub>2</sub> O	1.45	0.74	1.51	1.86	1.72	1.1	1.69	1.39	1.99	1.12	1.27	1.18	1.31	1.13	1.18	1.3	1.49	1.46	1.28	1.89
P <sub>2</sub> O <sub>5</sub>	0.26	0.12	0.36	0.25	0.26	0.19	0.25	0.14	0.28	0.23	0.14	0.19	0.21	0.17	0.22	0.18	0.14	0.19	0.21	0.18
Total	99.7	99.78	100.48	99.71	99.76	100.54	99.69	100.8	99.04	99.91	100.23	99.7	99.44	100.54	100.02	100.38	100.67	100.08	100.39	99.3
LOI	0.26	0.94	0.31	0.99	2.89	1.13	0.71	0.95	0.36	0.21	1.64	0.27	0.44	1.96	2.04	0.44	1.25	1.89	1.99	1.04
Mg#	0.649	0.473	0.557	0.488	0.455	0.39	0.529	0.445	0.508	0.476	0.504	0.47	0.43	0.461	0.346	0.469	0.412	0.466	0.456	0.397
Ni	26.1	7	18.5	5.8	5	8.4	8.7	3.9	7.3	10.7	9	10.9	15.8	6	8	14.5	6.2	8	7.2	5.8
Cr	224	9	55	12.4	5	22.4	13.2	15.9	25.3	30.8	27	21.9	18.7	5	11	24.8	9.8	18.4	16.3	12.8
V	205	169	154	169.5	122	96.2	187	140	150	203	154	149	209	167	128	176	146	139	163	106
Sc	38.4	25	19.1	23	16	14.8	27.5	20.2	20.4	26.5	22	17.7	22.3	22	24	23.9	18.4	19.9	23.2	15.6
Cu	61.3	332	49.3	39.1	8	8.3	27.6	11.5	31.8	40.4	145	38.8	95.3	990	34	57.3	70.8	33.1	39.3	14.1
Zn	59.1	88	59.3	58.5	64	91.4	63.8	59.8	64.3	68.6	95	71.1	66.7	78	70	61.2	54.8	67.1	71.9	67.9
Ga	23.4	18	19.3	18.2	19	20.7	18.3	17.4	17.8	17.2	17	18	17.7	17	19	16.4	17.2	16.6	17.4	17.5
Pb	11.6	3	15.2	15.1	12	6.9	19	7.7	19.7	5.3	10	3.9	6.6	7	10	6.6	34.3	7	7.3	10.8
Sr	596	207	1412	678	659	225	733	212	665	354	285	274	276	362	351	249	391	312	385	321
Rb	40.6	16	31.4	66.4	59	36.4	43.3	50.1	60	33.6	28	34.2	44.4	31	24	44.2	45.6	40.5	37	62.6
Ba	568	212	883	602	739	225	1245	262	1211	238	378	230	194	183	415	199	386	278	303	455
Zr	108	77	146	89	110	149	118	101	156	110	93	127	127	88	99	118	88.9	108	96.2	106
Nb	16.8	6	17.4	7.2	8	8.5	18.6	7.8	17.4	5.1	7	5.8	8	7	7	6.3	7	6.2	7.2	8.1
Th	4.6	4	7.9	5.1	6	4.1	10.9	4.6	8.8	3.9	4	4.6	4.7	3	4	4.5	3	4.5	4.3	5.3
Y	19.1	21	17.9	20.3	19	32.1	20.8	22.8	21.2	24.1	19	25.3	31	24	21	28.5	20.3	22.2	20.1	29.3
La	20.5	11	33	19.7	21	15.2	35.3	16.3	29.7	12.6	13	11.9	11.4	11	19	14.4	12	13.7	14	24.9
Ce	40	24	59.6	32.3	39	35.5	64.5	34.4	57	27.6	27	29.9	27.6	25	32	26.3	27.2	28	29.9	38.6
Nd	19.2	12	25.9	16.2	18	20.3	26.2	16.8	25	15.4	12	15.5	15.6	13	19	15.1	13	14.6	14.4	22.5
<sup>87</sup> Sr/ <sup>86</sup> Sr	0.7044	0	0	0	0	0	0.7083	0.12	0.7047	0.04	0	0	0	0	0	0.70464	0.706252	0	0	0.705608
<sup>143</sup> La/ <sup>144</sup> La	0.512689	0	0	0	0	0	0.512421	0.512667	0	0	0	0	0	0	0	0.512778	0.512426	0	0	0.512593
Age	7.4	14.2 <sup>a</sup>	7.8	9.3	9.4 <sup>a</sup>	14.7	7.6	14.6	9.31	12.43	13.4	13	12.6 <sup>a</sup>	11.9 <sup>a</sup>	12.8	12.6	13.4	12.81	11.04	

Table 2 (continued).

	SACARAMB										DEVA				MOIGRAD		UROI				
	395	7	396	776	1 <sup>c</sup>	5 <sup>c</sup>	3 <sup>c</sup>	4 <sup>c</sup>	767	2 <sup>c</sup>	399	565	394	6920	400	392	2479	765	766	UR-3	
SiO <sub>2</sub>	55.25	57.81	59.82	58.09	58.46	58.54	59.01	61.15	61.41	61.7	62.02	62.04	62.33	56.87	58.94	60.03	56.51	57.79	58.75	61.18	
TiO <sub>2</sub>	0.84	0.6	0.61	0.71	0.7	0.59	0.59	0.54	0.56	0.55	0.52	0.58	0.53	0.58	0.52	0.44	0.87	0.98	0.86	0.87	
Al <sub>2</sub> O <sub>3</sub>	17.6	18.46	19.39	15.79	17.18	18.36	17.02	17.23	17.49	17.19	16.81	17.84	16.97	18.3	19.18	17.59	18.3	15.57	17.15	15.64	
Fe <sub>2</sub> O <sub>3</sub>	7.84	5.74	5.62	5.82	5.99	5.16	5.72	5.27	5.23	4.31	5.35	5.2	5.69	5.51	4.2	7.62	5.5	4.44	4.44	4.04	
MnO	0.14	0.12	0.11	0.1	0.16	0.13	0.11	0.16	0.12	0.14	0.11	0.17	0.13	0.11	0.11	0.11	0.12	0.11	0.08	0.07	
MgO	3.7	1.56	2.19	4.85	2.43	2.37	2.8	2.49	3.69	2.46	3.12	2.42	3.32	2.94	2.76	2.02	3.18	3.87	3.41	2.96	
CaO	8.71	7.81	7.29	6.93	7.27	6.53	6.35	5.16	7.05	6.05	5.21	6.5	5.95	8.08	6.76	6.51	7.56	6.44	5.5	4.79	
Na <sub>2</sub> O	3.65	3.42	3.63	3.74	3.13	3.37	3.29	3.53	3.51	3.49	4.07	3.57	3.27	3.89	3.87	3.75	3.44	4.44	4.55	4.21	
K <sub>2</sub> O	2.01	1.47	1.66	2.92	1.86	1.24	1.76	2.29	1.47	1.85	2.61	1.57	1.88	1.32	1.88	2.26	1.15	3.93	4.38	5.32	
P <sub>2</sub> O <sub>5</sub>	0.2	0.17	0.17	0.34	0.35	0.19	0.21	0.25	0.18	0.21	0.31	0.23	0.2	0.22	0.21	0.17	0.29	0.71	0.57	0.61	
Total	99.94	99.81	100.49	99.29	99.63	99.34	98.03	99.29	100.71	99.7	99.09	100.27	99.78	98	99.74	97.08	99.04	99.34	99.69	99.69	
LOI	1.23	2.66	2.69	0.57	2.1	2.85	1.18	1.23	0.8	0.84	1.07	0.93	1.62	1.39	0.43	2.13	0.8	0.57	0.27	0.23	
Mg#	0.483	0.350	0.436	0.623	0.445	0.476	0.492	0.483	0.583	0.482	0.589	0.473	0.558	0.506	0.498	0.488	0.453	0.582	0.604	0.592	
Ni	7.2	8	7.2	5.6	9	6	10	9	22	11	34.6	6.4	14.3	3	4.2	3	4.9	42.6	44.9	41	
Cr	11.1	11	19.8	89.9	23	6	19	28	83.4	24	54.8	12.4	83.5	10	11.9	10	11.8	63.6	61	38.5	
V	290	142	142	149	147	141	169	152	131	141	111	138	133	360	190	150	146	116	75.7	84.3	
Sc	33.2	20	21.7	18.8	21	18	22	19	17.7	17	14	17.6	20	34	19	12	19.2	10.7	9.2	7	
Cu	15.8	24	15.1	25.3	33	5	57	46	33.4	46	19.9	25	29.1	90	30.2	14	8.9	46.7	25.8	20.4	
Zn	55.7	70	63.5	65.8	78	66	62	63	60.6	63	59.6	80.6	55.8	53	47.2	40	35.5	65.3	58.1	61.3	
Ga	18.9	18	17.9	19.7	18	18	17	17	17.8	17	19.4	18.3	17	16	17.8	19.5	18.9	22.7	22.3	23.4	
Pb	61.9	10	9.7	51.1	22	14	18	19	21.9	21	36.1	10.3	17	14	30	33	2.5	30.2	26.6	37.1	
Sr	2044	615	568	2770	909	533	799	756	1005	802	1737	654	635	2190	1742	1800	557	2355	2201	2353	
Rb	40.5	41	54.6	53.7	45	40	47	79	39.5	52	55.2	30.1	54.6	35	37.3	40	34.6	48	49.3	58.6	
Ba	2038	525	491	2165	1143	506	1356	1506	945	1441	1763	856	1274	2600	1897	2300	181	1780	1751	1878	
Zr	79.4	100	103	171	146	104	96	104	101	104	162	93.9	95.5	124	70.1	110	130	239	252	317	
Nb	7.3	7	7.5	16.3	11	8	10	11	9	11	15	9	10.3	12	7.9	16	6.9	29.8	23.6	24.9	
Th	9.9	3	2.6	21.6	10	5	8	9	5.7	9	19.7	6.5	7.5	10.8	11.7	20	3.2	19.3	17.2	20.1	
Y	21	20	21.2	22.2	29	21	20	19	19.6	17	25	27	76.3	23.3	25.5	47	48.8	63	20.1	121	106
La	42.7	15	15.1	92.7	37	16	28	25	27	27	141	44.1	45.8	80	84.2	120	42	205	177	184	
Ce	85.3	30	30.1	169.8	71	33	53	51	43	51	141	44.1	45.8	80	84.2	120	42	205	177	184	
Nd	37.9	15	15.2	67.2	29	16	22	21	20.1	21	55	17.8	19.9	42	30.2	61	20.7	66.7	57.8	63	
<sup>87</sup> Sr/86Sr	0	0.705143 <sup>b</sup>	0	0.70425	0	0.705316 <sup>b</sup>	0.704526 <sup>b</sup>	0.70467 <sup>b</sup>	0.704592	0.70466 <sup>b</sup>	0	0	0.704744	0	0.703982	0	0.705713	0	0	0.704441	
<sup>143</sup> La/ <sup>144</sup> Nd	0	0	0	0.512609	0	0	0	0	0	0	0	0	0.512656	0	0.512678	0	0.51258	0	0	0.51268	
Age	10.6	10.77 <sup>a</sup>	10.77	10.5	11 <sup>a</sup>	14.0 <sup>a</sup>	11 <sup>a</sup>	11 <sup>a</sup>	11.7	10.89	10.35	11.54	11.17	12.8	12.6	11.85	12.4	1.6	1.6	1.6	

Major element in weight percent and trace elements in parts per million; all iron as Fe<sub>2</sub>O<sub>3</sub>; LOI=loss on ignition.

a — inferred age based on the relationship with already known ages; b — <sup>87</sup>Sr/86Sr values from Alderton et al. (1988); c — Chemical analyses from Alderton and Fallick (2000). Errors quoted are the internal precision at 2 S.E. for Sr and Nd isotope analyses. Average for the Sr standard SRM 987 is 0.710239 ± 10 and for Nd Aldrich standard is 0.511414 ± 4.

Table 3 REE for selected samples.

Sample	790	5199	6922	400	2479	767	401	394	776	363	788	UR-3
La	15.70	11.50	13.90	46.50	20.70	23.40	23.30	25.20	91.70	30.60	20.4	111.30
Ce	35.10	27.80	30.60	85.20	45.50	45.80	41.10	47.37	178.8	62.59	43.00	205.70
Pr	4.10	3.56	3.57	9.17	5.27	5.46	6.02	5.58	20.44	7.34	5.06	21.50
Nd	15.80	14.60	13.80	27.50	20.70	19.10	21.70	15.40	66.40	21.00	19.40	67.3
Sm	3.28	3.38	2.94	4.20	3.98	3.29	4.16	3.33	9.43	4.62	3.76	8.76
Eu	0.97	0.94	0.87	1.21	1.28	1.05	1.25	1.02	2.51	1.27	1.22	2.32
Gd	3.48	4.13	3.23	3.30	4.00	3.40	4.66	3.05	6.50	3.95	3.79	5.53
Dy	3.68	4.59	3.32	2.65	3.85	2.99	4.41	2.80	4.02	3.42	3.40	3.26
Ho	0.73	0.93	0.65	0.55	0.76	0.59	0.88	0.56	0.75	0.67	0.66	0.57
Er	2.15	2.87	1.91	1.57	2.18	1.66	2.57	1.78	1.56	2.06	1.88	1.05
Yb	2.13	2.74	1.75	1.57	2.11	1.71	2.48	1.66	1.64	1.90	1.73	1.02
Lu	0.32	0.43	0.26	0.26	0.32	0.27	0.39	0.28	0.25	0.32	0.27	0.15
Eu/Eu*	0.88	0.77	0.87	0.99	0.98	0.97	0.86	0.98	0.98	0.91	1	1.02
(Ce/Yb)N	4.19	2.57	4.41	13.9	5.48	6.78	4.21	7.31	27.56	8.42	6.29	51.72
(Gd/Yb)N	1.31	1.2	1.46	1.69	1.51	1.58	1.5	1.48	3.15	1.67	1.75	4.37
(Yb/Lu)N	1.02	1	1.03	0.92	1.01	0.97	0.98	0.93	1	0.91	0.98	1.05

Basins development in the central part and western half of the region (in the Zarand Mountains) preserved the volcanic deposits in depressions. The oldest rocks (>14 Ma) dated by radiometric methods occur in the Roșia Montană-Bucium and Brad areas (Fig. 2). Volcanic activity then started in the west, in the Zarand basin, where K-Ar ages of 13.4–12.4 Ma have been obtained. From 13 to 11 Ma, volcanic activity shifted towards the east, occurring mostly in the eastern part of the WNW–ESE “alignment” and in the southern half of the NNE–SSW “alignment” (Fig. 1). At 11–9 Ma magmatic activity continued in the eastern part of the WNW–ESE “alignment”, but shifted again into the northern half of the NNE–SSW “alignment” where it remained focused for the next 2 Ma (9–7 Ma) (Fig. 1). The only magmatic occurrence younger than 7.4 Ma is at Uroi (1.6 Ma) at the south-eastern most edge of the Apuseni Mountains, after a gap in activity of about 6 Ma. The andesites at Moigrad are  $12.4 \pm 1.3$  Ma old, well within the range of the rest of the Apuseni Mountains volcanic rocks.

## Geochemistry

### Major elements

Major element compositions of the studied samples are given in Table 2. With few exceptions, most of them plot in the andesite field (Fig. 3). Besides the alkaline rocks of Uroi, which plot in the trachyandesitic and trachydacitic field, there are other three rocks displaying high-K calc-alkaline features. These are “Pârâul lui Toader microdiorite” (765), which plots in the basaltic-trachyande-

site field and the Zâmbrița (766) sample in the trachyandesite field, as do the Uroi trachyandesite and trachydacite (UR3). Two of them belong to the 7.4–10 Ma age group of the Săcărâmb area (Figs. 1, 2, Table 2). The Moigrad andesite plots within the medium-K andesite field together with most of the Apuseni Mountains Neogene rocks.

Major element variations are typical for calc-alkaline rocks, with MgO, FeO\*, CaO and TiO<sub>2</sub> decreasing with increasing SiO<sub>2</sub>. Na<sub>2</sub>O and P<sub>2</sub>O<sub>5</sub> remain roughly constant as no significant plagioclase and apatite fractionation occurred during magma evolution. When compared with Neogene volcanics in the East Carpathian arc, the Apuseni Mountains samples plot in a similar field to those in the Gutâi Mountains (Kovacs et al., 1998), except for the trachyandesites. Generally, they are very similar to Neogene volcanics of any of the East Carpathian arc segments, such as the Ukrainian Trans-Carpathian region (Seghedi et al., 2001), or the Călimani-Gurghiu-Harghita chain (Seghedi et al., 1995; Mason et al., 1996), but without large major element variations.

### Trace elements

Trace element contents are given in Table 2. Ni and Cr contents show a large variation. Ni ranges from 3.9 to 56 ppm, but most of the samples contain <20 ppm. Seven samples have >20 ppm, among them the Detunata basaltic andesite. Cr contents vary from 5 to 90 ppm with the exception of the Detunata basaltic andesite (224 ppm). Seven samples have >50 ppm Cr, among them most are trachyandesites. The relatively high Ni and Cr, along with high Mg# of the high-K rocks is striking, as compared with the rest of the rocks.

Mg# displays a wide spectrum, from 0.35 to 0.65, and is slightly higher among the younger rocks (Table 2).

High field strength elements (HFSE) such as Zr and Nb display a particular behaviour. Zr is in the range 70–317 ppm while Nb varies from 8 to 30 ppm. They do not vary significantly with SiO<sub>2</sub> content, but there is a slight increase of Zr with increasing Mg#. The most intriguing feature is, however, that the igneous rocks in the Apuseni Mountains very clearly cluster in three groups according to their Zr and, especially, Nb content (Fig. 4). The rocks can be grouped in “low-Nb” (5–12 ppm) to “medium-Nb” (15–20 ppm) and “high-Nb” (>23 ppm) rocks. Uroi trachyandesites form the high-Nb group. Detunata basaltic andesites, two of the three high-K rocks and other three samples belong to the medium-Nb group, while all the remaining samples constitute the low-Nb group. Zr content follows Nb as well, so that high-Nb rocks are also high-Zr rocks, although Zr-clustering is not as obvious as Nb-clustering. Data suggest that HFSE distribution is source-controlled and not related to differentiation processes.

Large ion lithophile elements (LILE), especially Sr (212–2770 ppm) and Ba (181–2165 ppm), display a wide range of concentrations. They show no correlation with SiO<sub>2</sub> or Mg#, but correlate positively with each other (Fig. 5). Although the range in Sr and Ba concentrations is large, over one order of magnitude, there are no obvious gaps for high-Sr and/or high-Ba groups, as is the case of the Nb groups. However, generally, low-Sr rocks are older ones and high-Sr rocks are the younger ones (Roşu et al., 2000). High-Sr and high-Ba rocks are characteristic of the Apuseni Mountains, but they are uncommon in the East Carpathian arc (Kovacs, 1998; Seghedi et al., 2001), except for its southernmost segment where similar rocks are present (Seghedi et al., 1986, 1987; Mason et al., 1996). Boştinescu and Savu (1996) highlighted the andesites from the Deva area as being high-Sr and high-Ba, in contrast to the other Neogene volcanics in the Apuseni Mountains, but our data show that high-Sr and high-Ba rocks are not restricted to the Deva area. In addition to the Neogene magmatic rocks, Savu (2001, 2002) pointed out that Paleogene magmatic rocks in the Mureş Valley are also enriched in Sr and Ba, suggesting that the magmatic rocks have “adakitic features”, controlled by tectonic processes.

An important characteristic of the Apuseni Mountains samples is their high Sr/Y ratio (Fig. 6). This specific geochemical characteristic links part of the Apuseni Mountains magmatism with a particular association, i.e. the adakite-like rocks of Defant and Drummond (1990). The term

“adakite” was defined as an intermediate to acid calc-alkaline rock characterized by high Al<sub>2</sub>O<sub>3</sub> and Sr, low Y and HREE, and MORB-like Sr and Nd isotopic signature. Many of the younger calc-alkaline rocks and all the alkaline rocks of the Apuseni Mountains plot at the edge of the adakite field (Fig. 6) as defined by Defant and Drummond (1990). In the following we will use the term adakite-like as a descriptive term, without genetic connotation.

Incompatible trace element distribution, normalized to N-MORB (Sun and McDonough, 1989), is displayed in Fig. 7. The patterns are typical for subduction-related magmatic rocks in modern continental and island arcs, with obvious negative spikes of Nb, P and Ti, and a strong positive Pb spike (Wilson, 1989). Most of the Săcărâmb, Baia de Arieş (younger than 10 Ma), Deva and Uroi samples show higher incompatible element concentrations and a Sr spike (Fig. 7). With the exception of Pb, the Moigrad andesite is similar to the average incompatible trace element pattern of the Zarand-Brad-Zlatna samples. Compared to other Carpathian Neogene volcanic rocks, trace element patterns in samples from the Zarand-Brad-Zlatna are similar to those from the Gutâi Mountains (Kovacs, 1998), Ukraine (Seghedi et al., 2001) and the Călimani-Gurghiu-Harghita volcanic chain (Mason et al., 1996).

### REE geochemistry

Twelve samples of Apuseni Mountains Neogene volcanic rocks have been analyzed for REE. The results, along with some calculated parameters, are given in Table 3. Chondrite-normalized REE patterns, using the values of Sun and McDonough (1989), are relatively smooth and slightly concave for most of the analyzed rocks (Fig. 8). The most basic rocks (the Detunata basaltic andesites) have the smoothest patterns with no Eu anomaly. The Moigrad andesite is strikingly similar to the Detunata sample. Eu anomalies are absent or very low (>0.86), with one exception, 0.77 for one of the Zarand andesites (sample 5199, Table 3). Uroi and Zâmbriţa alkaline rocks are different from the other rocks, displaying strong enrichment in LREE and important depletion of HREE. Generally the younger volcanics with higher-K content show higher LREE and a slight depletion in HREE.

A comparison with the Neogene volcanics of the Gutâi Mountains reveals a significant difference, with overall REE values being higher in the Gutâi Mountains (Kovacs, 1998) than in the Apuseni Mountains REE patterns in the Ukraine volcanics (Seghedi et al., 2001) and in the Călimani-

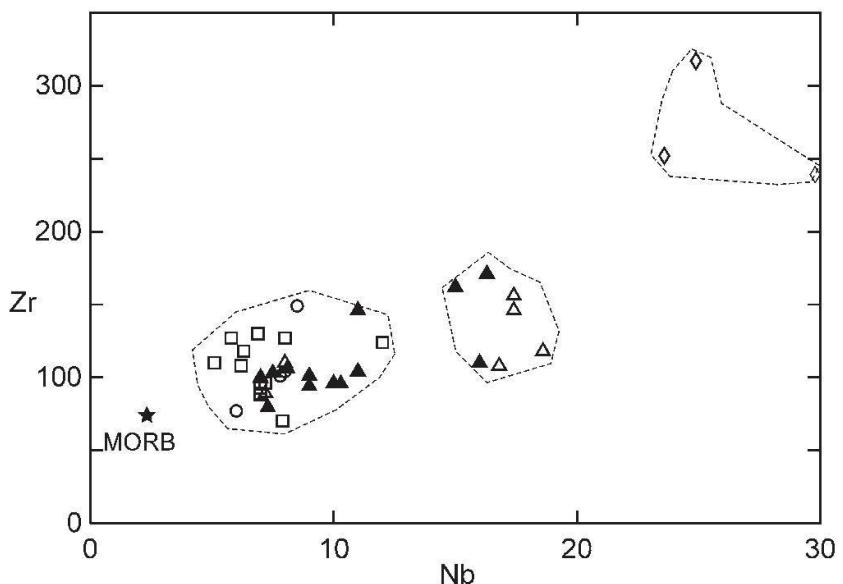


Fig. 4 Zr vs. Nb diagram for Neogene Apuseni magmatic rocks. Symbols as in legend of Fig. 3.

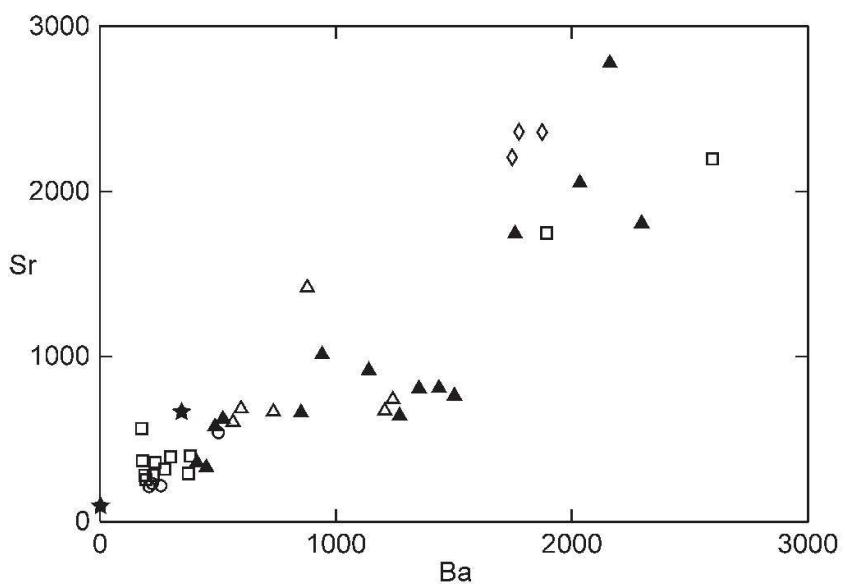


Fig. 5 Sr vs. Ba diagram for Apuseni magmatic rocks. Symbols as in legend of Fig. 3.

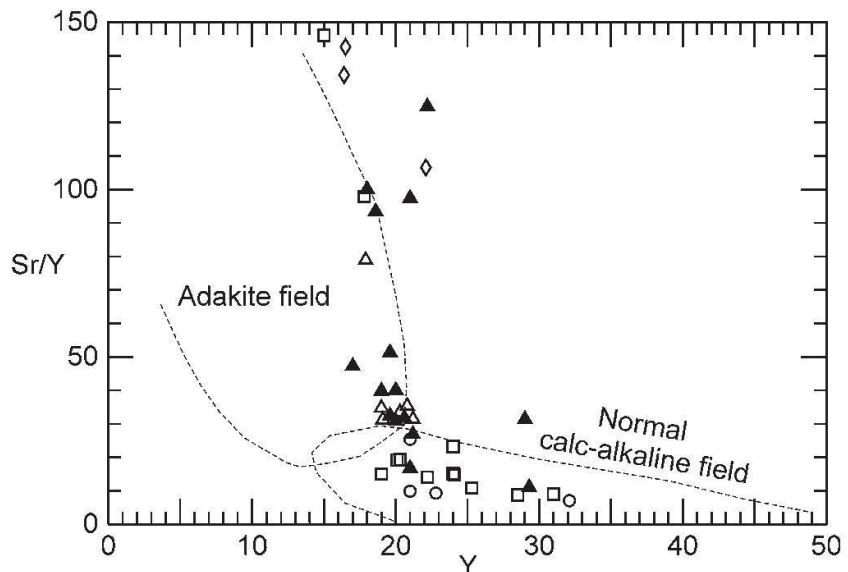


Fig. 6 Sr/Y vs. Y (Drummond and Defant, 1990) diagram discriminating between adakitic and normal calc-alkaline compositions. Symbols as in legend of Fig. 3.

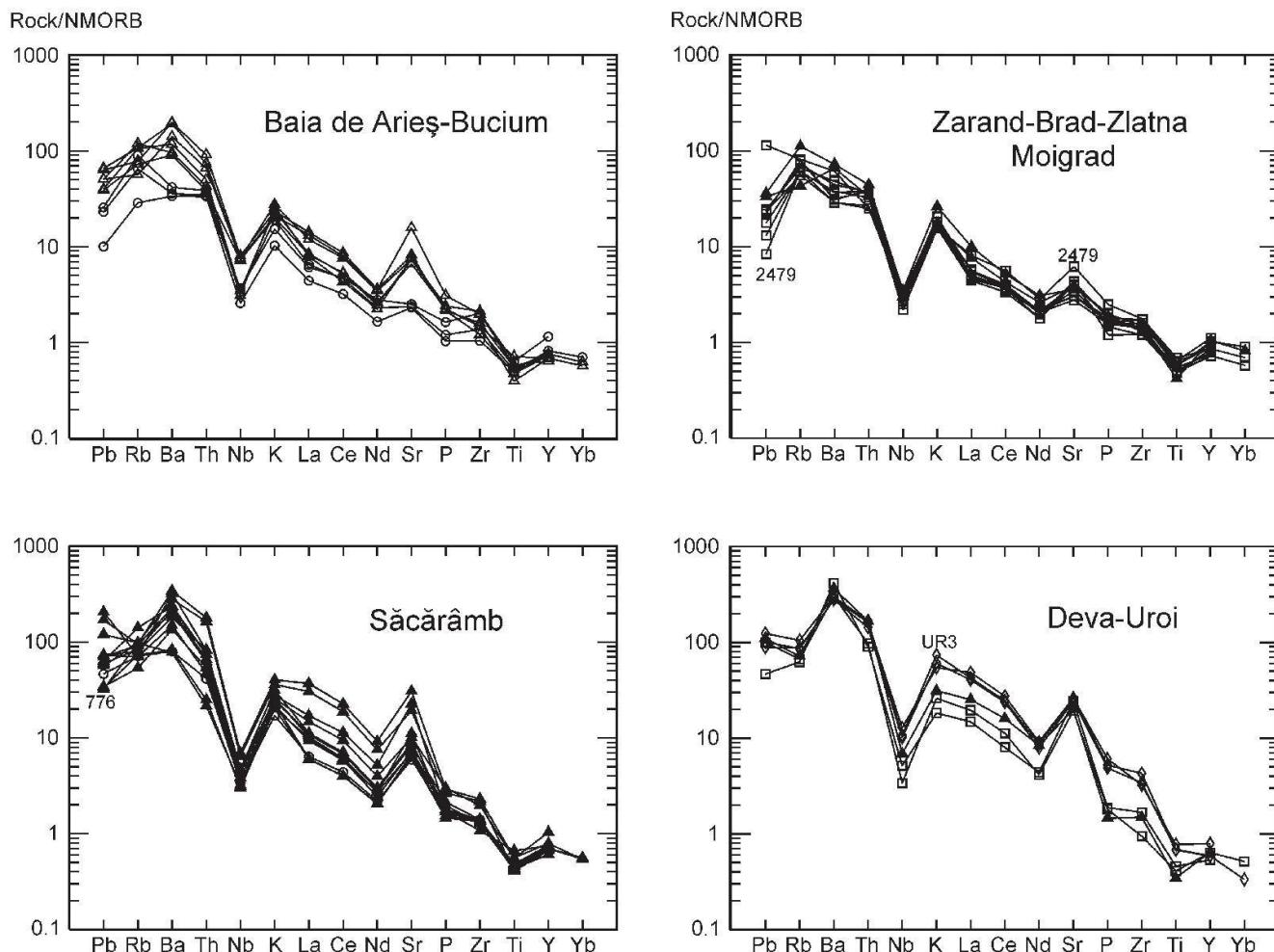


Fig. 7 N-MORB normalized trace element diagram for samples grouped in four areas: Baia de Arieș-Bucium; Zarand-Brad-Zlatna, Moigrad; Săcărâmb; Deva-Uroi. Normalizing coefficients from Sun and McDonough (1989). Symbols as in legend of Fig. 3.

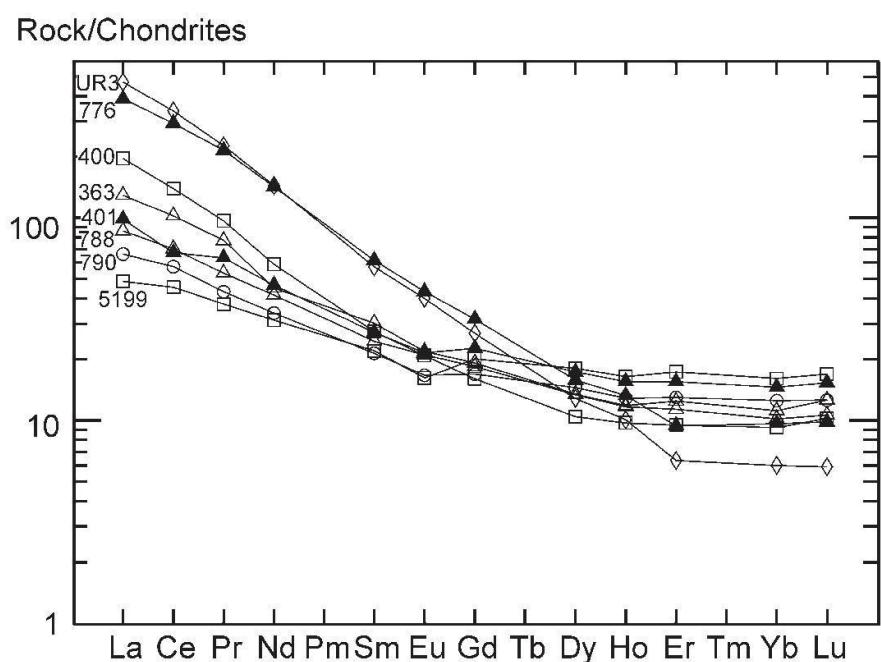


Fig. 8 Chondrite-normalized REE pattern for selected Apuseni Mountains magmatic rocks (Sun and McDonough, 1989). Symbols as in legend of Fig. 3.

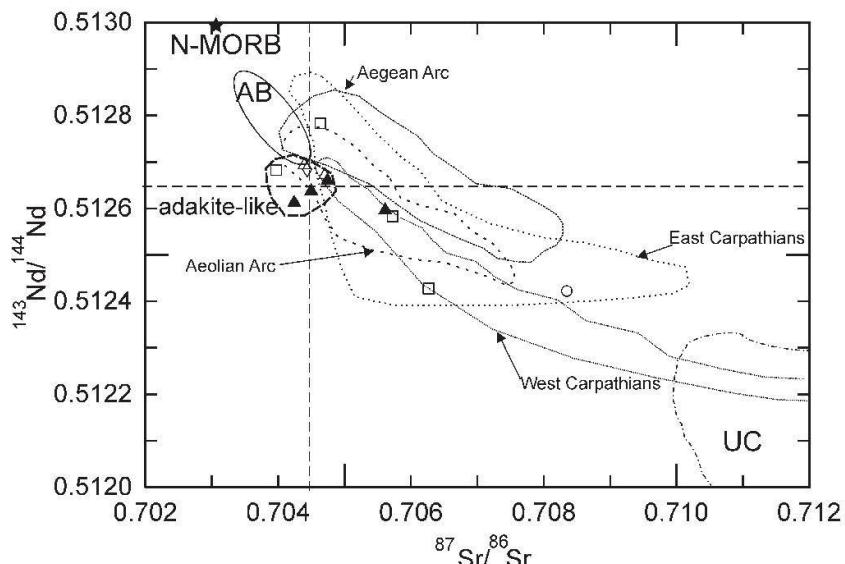


Fig. 9  $^{87}\text{Sr}/^{86}\text{Sr}$  vs.  $^{143}\text{Nd}/^{144}\text{Nd}$  variation of Apuseni Mountains magmatic rocks. Fields for alkali basalts (AB) from Downes et al. (1995); for Upper Crust (UC) from Mason et al. (1996); for Aegean arc from Brihuega et al. (1986); for East Carpathians arc from Mason et al. (1996); for West Carpathians from Salters et al. 1988; for Aeolian arc from Ellam et al. (1988). Symbols as in legend of Fig. 3.

Gurghiu-Harghita volcanic chain (Mason et al., 1996) are roughly similar to most of the Apuseni Mountains rocks. Shoshonites and high-K rocks in the South Harghita rocks (Seghedi et al., 1987; Mason et al., 1996) have almost identical REE patterns to those of the Uroi and Zâmbrița trach-yandesites.

### Sr and Nd isotopes

Sr and Nd isotopic compositions (Table 2) are plotted in Fig. 9 along with regions of known calc-

alkaline affinity (Aegean arc, Aeolian arc, East Carpathians, West Carpathians). The data suggest that adakite-like rocks plot at lower  $^{87}\text{Sr}/^{86}\text{Sr}$  and higher  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios, around the Bulk Silicate Earth reservoir (Zindler and Hart, 1986), compared with normal calc-alkaline samples, which are much more scattered. The oldest measured rock (790) has higher  $^{87}\text{Sr}/^{86}\text{Sr}$  and lower  $^{143}\text{Nd}/^{144}\text{Nd}$  (Fig. 9).

### Paleomagnetic data

Previous paleomagnetic data (Pătrașcu et al., 1992, 1994; Panaiotu, 1998) show that large areas from the Transylvanian basin, Apuseni Mountains and Southern Carpathians suffered a post-Eocene northward transport and a large clockwise rotation. Recent paleomagnetic studies on the Miocene volcanic rocks from the Apuseni Mountains (Panaiotu, 1998, 1999) combined with the new K-Ar data yield further constraints on the timing of the rotation.

Stable paleomagnetic directions from 65 sites (Fig. 10) are characterized by a mean inclination around  $65^\circ$  and a dispersion of declinations from the mean direction recorded in the Late Cretaceous magmatic rocks (around  $80^\circ$ ) toward the north. Figure 11 shows the declination of sites sampled both for paleomagnetism and K-Ar ages. A gradual change of declinations through time shows that most of the Tertiary clockwise rotations were very fast: around  $70^\circ$  between 14.5 Ma and 12 Ma. Calc-alkaline volcanism took place during this rotation. The adakite-like magnetism (with some exception in Deva and Săcă-

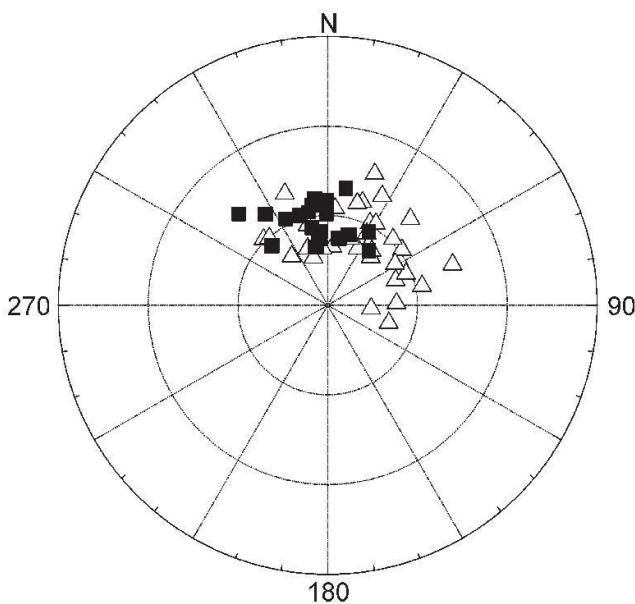


Fig. 10 Equal area projection of site-mean directions for all sampled Neogene volcanic rocks. Reversed magnetizations were inverted through the origin; so all directions are plotted in the lower hemisphere. Symbols: open triangles—common calc-alkaline; black squares—adakite-like rocks.

râmb areas) roughly came after the cessation of the large clockwise rotation.

Clockwise rotation in the Tisia block started after collision of the northern part of the Alcapa block with the European continental margin at about 16 Ma (Panaiotu, 1998). Collision provided the local causes for both a very rapid migration to the east of the slab's hinge and slab detachment, which then started to migrate eastward. This fast rotation corresponds to a period of strike-slip faulting with the accommodation of extension through pull-apart structures in the Pannonian Basin (Matenco, 1997; Fodor et al., 1999) and to extremely rapid Middle Miocene depocentre shift (around 500 km between 15.5 Ma and 11.5 Ma) coupled with a change in direction of the Carpathian foredeep depocentre migration (Meulenkamp et al., 1996). The paleomagnetic results show that the Miocene volcanism from the Apuseni Mountains took place in a very dynamic rotational setting.

### Discussions and conclusions

Magmatism in the Apuseni Mountains shows specific geochemical peculiarities which vary with age, e.g. a general increase of  $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ,  $\text{P}_2\text{O}_5$ ,  $\text{Mg}^{\#}$ ,  $\text{Nb}$ ,  $^{144}\text{Nd}/^{143}\text{Nd}$  and decrease of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio, suggesting more alkaline and at the same time more primitive sources (Szakács et al., 1999; Roșu et al., 2001). Petrographic and geochemical compositions of the Apuseni Mountains rocks do not satisfy the criteria for identifying significant fractionation processes (e.g. narrow  $\text{SiO}_2$  interval, lack of Eu anomaly, lower Sr and higher Nd isotope ratios for younger magmatic products). Most of the rocks plot in the andesitic field, with few basaltic andesites, and in addition they show (ac-

cording to their high Sr/Y ratio) an adakitic-like character for most of the rocks younger than 12.5 Ma.

The common (non-adakitic) calc-alkaline rocks have high  $^{87}\text{Sr}/^{86}\text{Sr}$  for the older rocks, suggesting that in the initial stages of magma generation either a larger amount of assimilation-fractional crystallization (AFC) occurred in intermediary magma chambers or the source had a higher initial  $^{87}\text{Sr}/^{86}\text{Sr}$  (Fig. 12). This observation can be seen at a regional and at local scale in each volcano-intrusive structure, suggesting that there are specific relationships with age, which suggest a higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio for the Roșia Montană-Bucium and Zarand-Brad areas. On the other hand, high Ba/La ratios (Fig. 13), as well as high Ba/TiO<sub>2</sub> (Fig. 12), Ba/Nb and Th/Nb ratios suggest significant fluid involvement during magma generation, especially after 12 Ma, indicating the onset of the conditions required for the generation of adakitic-like magmas. The variable Ba/La ratio for the rocks of the same age interval (especially for the 12–10 Ma group) also suggests variable fluid involvement in magma generation (Fig. 12), which would characterize a heterogeneous, variably fluid-rich source, probably the lower crust or upper lithospheric mantle, and not asthenosphere, which normally is much more homogeneous. The observed temporal changes in source composition may have resulted from variability in the intrinsic composition of the source, compatible with a lower crust and/or mantle lithosphere origin. Also, the presence of garnet-bearing andesites in the Apuseni area (not analyzed here), along with variable depletion in HREE of some of the analyzed samples (suggesting some garnet and/or amphibole removal) requires a deep-seated hydrous source and rapid ascent to the surface along deep-seated faults in a tensional stress regime, as

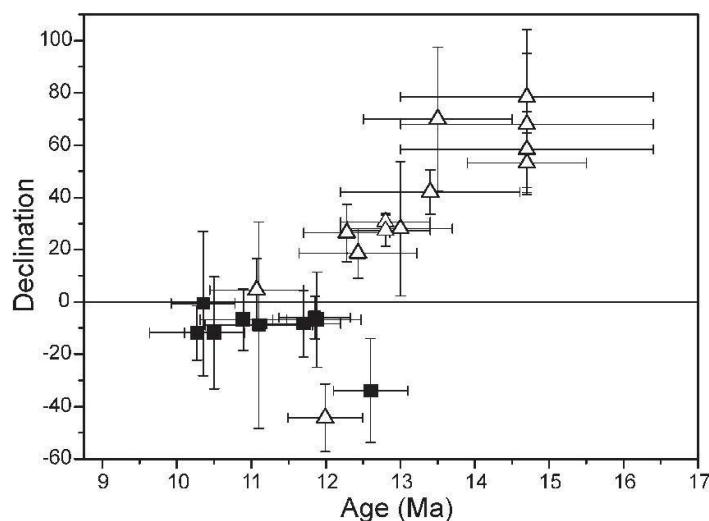


Fig. 11 Declination versus age for sites sampled both for paleomagnetism and K-Ar ages.

suggested by Harangi et al. (2001) for such rocks. However, variable and higher Nb and Nb/Y ratios in younger rocks (towards the OIB field) also suggest involvement of the asthenosphere in magma generation, probably via mixing with lower crust/mantle lithosphere-derived magmas (Fig. 13). This is most obvious for the magmas generated between 10–7.4 Ma and for the youngest alkaline rocks (1.6 Ma), suggesting a higher degree of involvement of asthenospheric (OIB-like) melts for the younger products, along with a still important hydrated source.

Based on K–Ar determinations and stratigraphic data, the Neogene magmatic activity in the Apuseni Mountains occurred between 14.7 and 7.4 Ma and (after a long gap) at around 1.6 Ma. The relationships between Miocene magmatism and extensional tectonics thereby, are of prime importance. Geodynamic conditions were not related to contemporaneous subduction, as the area was situated in a continental block affected by transtensional tectonics, which generated horsts and grabens and narrow sedimentary basins. As a consequence the magmas were derived initially from a source situated at the crust-lithospheric mantle boundary (low-Nb) and later from uprising asthenosphere (high-Nb, low  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio), as suggested by isotopic and trace element data. Higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios occur in the older rocks (>13 Ma), whereas younger ones have lower values. Thus upper crustal contamination was notable only in the initial stages and unimportant, as well as fractional crystallization processes, in later stages.

The lithospheric source must have been particularly rich in fluids. High LILE contents of the magmas favour a source-enrichment by subduc-

tion processes, in an old tectonic event and/or during the Eocene–Miocene subduction, prior to initiation of rollback processes in the front of the Carpathian subduction zone. The evolutionary trend from common calc-alkaline to adakite-like calc-alkaline and alkaline magmas is also largely variable in time and space and may have been a direct consequence of the extensional tectonic regime. Our preferred mechanism of magma generation is decompression melting facilitated by a transtensional tectonic regime (as suggested by Hooper et al., 1995; Hawkesworth et al., 1995; Wilson et al., 1997). Variable rotational movements between 40 and 15 Ma and mostly from 14 to 12 Ma may have accentuated the brittle tectonic and tensional stress field regime of the Apuseni part of the Tisia block and induced lower crust-lithosphere melting. Tensional fractures facilitated ascent of the early magmas, which experienced some fractionation and contamination in shallow magma chambers. Acceleration of the extensional regime favoured asthenospheric upwelling at the base of the lithosphere and as a consequence an increase in temperature, which created the necessary conditions for further adakite-like magma generation (fluid-present melting) of a fluid-rich upper lithospheric mantle or lower crustal source, but also mixing with asthenosphere-derived melts. This is one way to produce adakite-like magmas, the presence of which suggests derivation from high-pressure partial melting of either subducted oceanic crust or underplated or delaminated lower crustal basic material (Defant and Drummond, 1990; Martin, 1999; Xu et al., 2002).

The rotations between 14 Ma and 12 Ma facilitated the necessary pathway for emplacement of common calc-alkaline and then adakite-like calc-

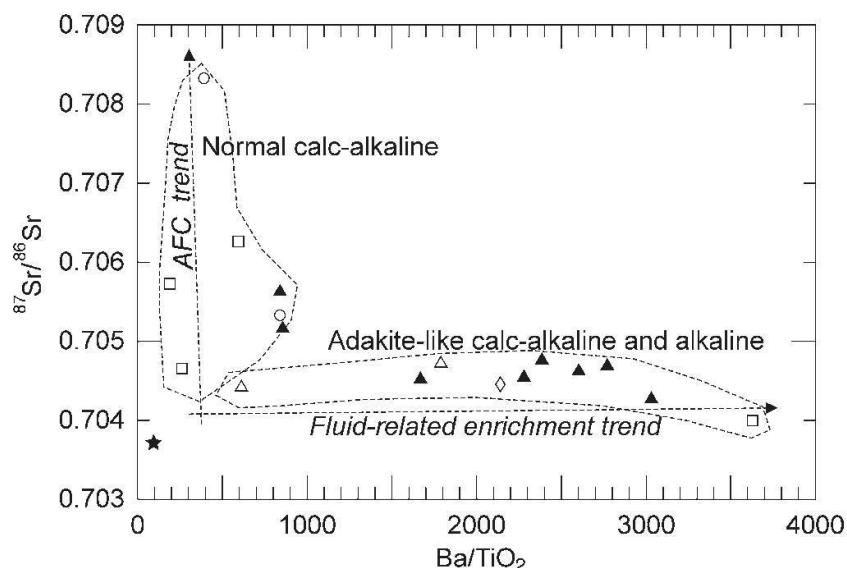


Fig. 12  $^{87}\text{Sr}/^{86}\text{Sr}$  vs.  $\text{Ba}/\text{TiO}_2$  diagram for Apuseni Mountains magmatic rocks. Symbols as in legend of Fig. 3.

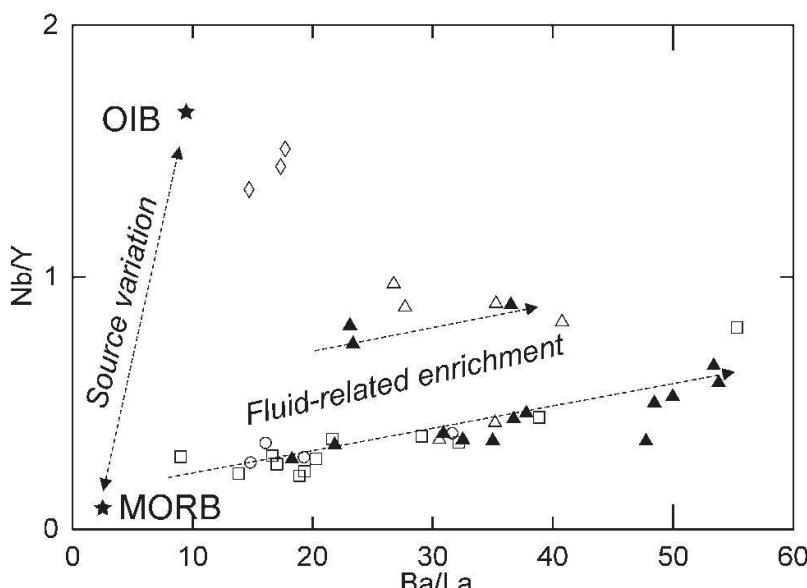


Fig. 13 Nb/Y vs. Ba/La diagram for Apuseni Mountains magmatic rocks. OIB and MORB from Sun and McDonough (1989). Symbols as in legend of Fig. 3.

alkaline and alkaline magmas after the end of clockwise rotation. The geochemical and isotopic features of the 12.5 Ma Moigrad andesite, situated much further to the north, suggest a similar magma genesis as for the common Apuseni calc-alkaline magmas, during important clockwise rotation. After ~12.5 Ma generation of the common calc-alkaline magmas declined in favour of adakite-like calc-alkaline and alkaline magmas. Trachyandesite generation after a gap of 6 Ma suggests a different tectonic setting, but also extensional, as it is contemporaneous with alkali-basaltic magmatism (2.5 Ma) (Downes et al., 1995) along the same South Transylvanian fault (Szakács and Seghedi, 1996). The geochemical and isotopic features of the trachyandesites suggest mixing of asthenospheric (low  $^{87}\text{Sr}/^{86}\text{Sr}$  and high Nb) and lithospheric components, as the trachyandesites fall close to the adakite-like field.

The most important petrogenetic process involved in magma generation in the Apuseni Mountains is probably variable degrees of partial melting of a heterogeneous source, which was isotopically depleted, but enriched in incompatible elements. The initial crustal contamination and insignificant fractional crystallization processes is characteristic for an initial storage of the magmas in the crust. Further rapid ascent of magmas was favoured by the increasingly extensional regime in which the magmatic province evolved. Although dominantly calc-alkaline, extension-related magmatism in the Apuseni Mountains is associated with minor alkaline magmas, which occur late in the magmatic sequence, suggesting an asthenospheric source beneath the attenuated

lithosphere or a decrease in the degree of melting at the end of extensional processes.

Our petrogenetic model, which suggests a fluid-rich upper lithospheric source, supports the unique metallogenetic features of the Apuseni area characterized by numerous porphyry copper-gold deposits (although many sub-economic class), base metal, world-class gold deposits and rich smaller deposits showing an uncommon density of ore bodies (Udubaşa et al., 2001).

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